CHAPTER ONE

1. Literature review

1.1. Wheat uses and importance in the world with emphasis on production and trade in Australia, the Middle East and Iraq

Wheat (*Triticum* spp.) is a very important food crop to human civilisation. Wheat was firstly domesticated in Mesopotamia (now known as Iraq), in the land between the two rivers Tigris and Euphrates. In addition, wheat was one of the first domesticated food crops, and was a basic staple food of the major civilizations of Europe, West Asia and North Africa. Across the ages, wheat has been linked to the Egyptian, Cretan, Grecian and Roman civilizations (Buller, 1919). Wheat was a key factor in moving from a hunter and gatherer existence to a sedentary based society (Diamond, 1997).

Wheat is unique among the cereals due to its gluten proteins that are necessary for making leavened and unleavened breads (Orth and Shellenberger, 1988). Wheat continues to be the most important food grain source for humans. Its production leads all the other crops, including rice, maize and potatoes. According to the information obtained from (FAOSTAT, 2005), wheat was planted all over the world, and the production was of about 600 million tonnes from an area of about 220 million hectares. A single hectare produces an average of about 2.5 tonnes World use and consumption of wheat crop can be classified as human food, animal feed, and seed for planting and processing for industry uses. Consumption of wheat all over the world has increased quickly since the early 1960s, with wheat production being around 655 Mt in 2012-13 and 713 Mt in 2013-14 (Grain Yearbook 2015 USDA, ABARE data). In developing countries, the use and consumption of wheat had increased by about 35% over the 13 years after 1963. Reasons for this include urbanisation, a preference for wheat over rice, and an increase in the consumption of wheat generally. From 1980 until now, the annual growth of wheat uses all over the developing countries has risen from about 2-5%. In high-income countries with mature food wheat markets, changes in consumption happen slowly over time and are driven by population growth and changing dietary preferences (CIMMYT, 1996).

A dramatic increase in wheat production was seen worldwide from 1951 to 1990 derived from expanding the areas that could be used for wheat planting (CIMMYT, 1996). From 1986 until 1990, there was an increase in production of about 63 million tonnes. The production per hectare was one tonne per hectare in 1951, and then rose to 2 tonnes per hectare in 1980 and to 2.5 in 1995. This has largely arisen because of improvements in agronomy, inputs such as fertilizers and organic matter and wheat breeding programmes developing higher yielding varieties (CIMMYT, 1996).

World trade of wheat gives an indication of the production and local consumption of wheat. China is the largest producer of wheat in the world. However, China is the seventh largest wheat importer, so they consume more than they produce. According to data from Spectrum Commodities, the United States of America is the largest exporter of wheat in the world, but only the third largest producer after China and India. Australia is marked as the seventh largest producer of wheat worldwide. The main crop grown in Australia by production is wheat followed by barley, canola, sorghum, oats, rice and then pulses and durum wheat.

According to the Australian bureau of statistics, (2014), production of wheat was 24 million tonnes. However, in 2011, the production was 27.4 million tonnes, and that was considered the highest production since records began in 1861. The production area in 2011 was 13.5 million hectares, and this is a little lower than 2010 (which was 13.9 million hectares), showing an increase in yield per hectare. Around 15 million tonnes were produced in the east coastal area of Victoria, NSW and Qld, (Fig 1) and nearly 10 million tonnes went for local consumption including for human, animal and industrial purposes. However, the rest of the production went for export. In addition, the most significant production of wheat in Australia was located in the western coastal area, with 80 to 90% of the production being exported. Australian wheat goes to different places all over the world. The biggest importing region is South East Asia countries at around 30% of exported grains. However, Europe is the smallest importer of wheat at just 2%. Middle East countries import about 10% of the exported Australian wheat (AGI, 2011).

In the Middle East, wheat forms more than 37% of a person's daily energy intake indicating a high level of consumption (Ahmed et al., 2013). According to the USDA, wheat production in Iraq was 2.18 million metric tonnes in 2012-2013 (Table 1), with all that production going for local consumption in addition to imported wheat. The majority of wheat production provinces depend on winter rainfall especially during March and April, and the rest of production relies on irrigation. The northern part of the country gets most of the rainfall (Fig 2). The biggest production of wheat was 2.32 mt/ha in the Diyala province (Table 1). However, the lowest production was 0.80 mt/ha in both Arbil and Sulymaniyah provinces. The country’s production of wheat for local consumption was 2.7 million metric tonnes (FAO, 2015), with the annual local consumption being about 7 million tonnes. When the local production of wheat is not sufficient for local consumption, Iraq imports wheat from different countries. To meet local consumption needs, 4 million metric tonnes of wheat was imported in 2013. In Iraq, the Public Distribution System (PDS) is a food program run by the government. This program provides 9 kg of flour for each Iraqi person monthly. Australia is one of the main exporters of wheat to Iraq (AEGIC, 2014). Between 2009 and 2013, Iraq imported around 1 million metric tonne of Australian wheat. Table 2 shows the exported wheat from Australia to Iraq. According to the table, WA was the largest exporter to Iraq. The largest amount of wheat imported by Iraq from Australia was in 2013, and that amount formed around half of the total imported wheat that year.

Fig.1.1. Production map of Australian wheat: Source, Australian Centre for International Agriculture Research.

Fig.1.2. Wheat production in Iraq by province. Source: USDA.

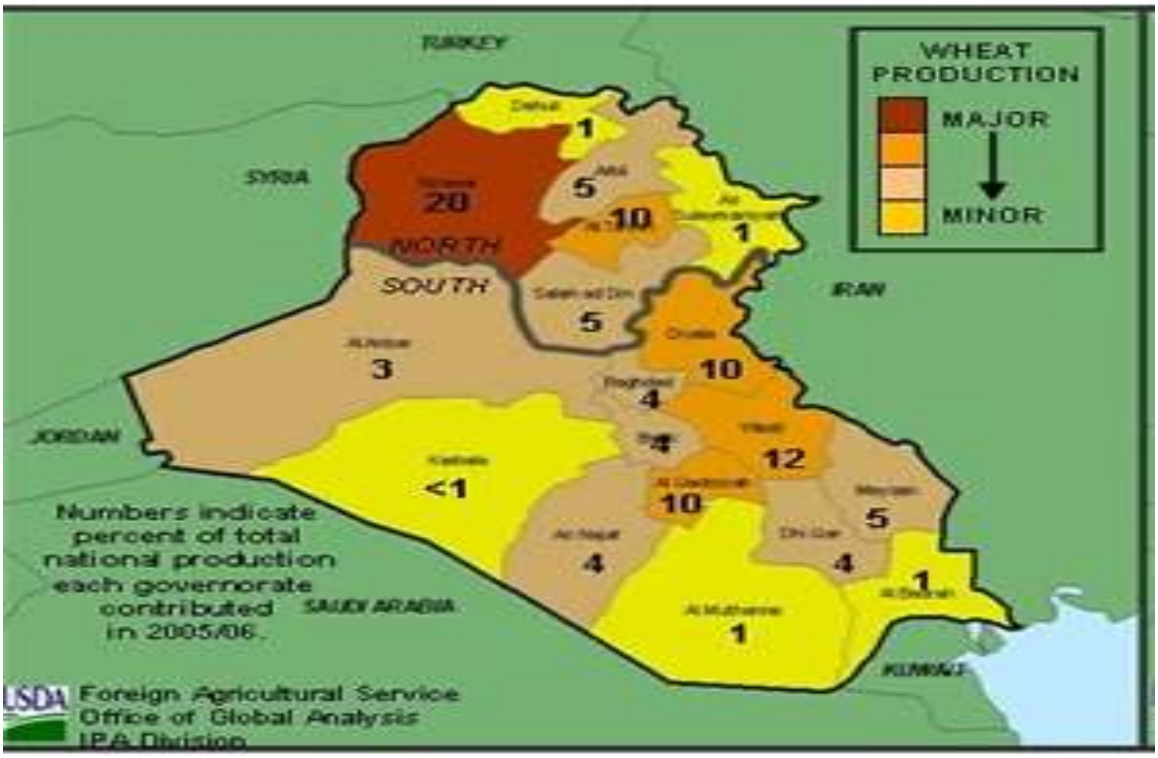


Table.1.1. Wheat production in Iraq by province shows planted area, yield and production per hectare for the agricultural season 2011-2012.

Province	Planting Intentions (Donum*)	Percent Planted	Planting Actual (Donum*)	Yield (Metric Tons/ hectare)	Production (Metric Tons)
Kurdistan					
Dahuk	750,000	75%	562,500	1	168,750
Arbil	900,000	50%	450,000	0.80	90,000
Sulymaniyah	475,000	65%	308,000	0.80	77,188
Total	2,125,000	62%	1,321,250	1	335,938
Rest of Iraq					
Ninewa	1,639,285	50%	819,643	0.36	122,946
Kirkuk	735,189	80%	588,151	1.75	249,964
Salah ad Din	712,000	77%	548,240	1.32	246,708
Diyala	429,300	75%	321,975	2.32	177,086
Anbar	281,115	95%	267,059	1.32	133,530
Baghdad	245,625	92%	225,975	1.60	90,390
Wasit	751,000	90%	675,900	1.60	270,360
Babil	312,345	98%	306,098	1.36	104,992
Karbala	15,205	100%	15,205	1.52	6,082
Najaf	201,098	106%	213,164	1.8	95,924
Muthanna	69,950	63%	44,069	1	11,017
Qadisiyah	380,000	92%	349,600	1.4	174,800
DhiQar	277,600	70%	194,320	1.32	87,444
Maysan	219,100	100%	219,100	1.12	60,800
Basrah	70,400	70%	49,280	1.08	12,320
Total	6,6,339,212	76%	4,837,778	1.52	1,844,363
Grand Total	8,464,212	73%	6,159,028	1.4	2,180,301

Source: USDA

*One Donum equals 0.25 hectare.

Table.1.2. Exported Australian wheat in metric tonnes to Iraq from 2009 to 2013.

State	2009	2010	2011	2012	2013
NSW	326,571	51,182		115,719	132,000
QLD	255,007			52,500	25,500
SA	203,859	145,400		289,846	511,687
VIC				82,400	87,500
WA	127,434	101,346	970,846	20,241	966,822
Total	912,871	246,746	1,022,028	560,706	1,723,509

Source: AEGIC

1.2. Wheat types and technological uses

Before talking about the uses of different types of wheat, understanding the Australian grading system for wheat is important. In Australia, wheat is graded into different categories depending on usage and determined by specific technological properties. The categories are hard, prime hard, durum, premium white, standard white, noodle, soft, general purpose and feed wheat (Blakeney et al., 2009). The Australian prime hard should contain a minimum protein percentage of 13%. It is made up from selected hard white wheat varieties of exceptional milling quality. Flour milled from it is used to produce Chinese style yellow alkaline noodles and Japanese ramen noodles and for the production of high protein, high volume breads. It is blended with lower protein wheats to produce flour suitable for a wide range of baked products and noodles. Australian durum wheat grade 1 has a minimum protrin content of 13%. is highly vitreous, of high water absorption ideal for making good quality pasta (Sissons et al., 2012). Hard wheat has approximately 11.5% protein and is used for making different types of bread and high quality alkaline noodles. Premium white has protein around 10% and is prepared by mixing a number of hard white wheat varieties and is ideal for making Asian noodles and breads. Standard White wheat has medium to low protein and is the main source of Middle Eastern and Indian style flat breads, European style breads and rolls, and Chinese steamed bread. Australian soft wheat has approximately 9.5% protein, and is appropriate for making confectionery and baked products including sweet biscuits, cookies, pastries, cakes, steamed buns and snack foods. Australian General Purpose is wheat that is not graded and is lower in quality used for general purposes. Feed wheat is used for animal feeding.

1.2.1. Bread

Bread is one of the main foods for many people. Humans consume bread for its nutritional value and taste (Dendy et al., 2001). Bread making procedures are different across the world.

The ingredients of bread are driven by a number of factors such as traditional methods, cost, energy content, quality of flour, the type of bread required and the time between preparing and using bread (Delcour and Hoseney, 2010). As bread is one of the yeast-leavened products, the general ingredients of bread making are flour, yeast, salt and water (Delcour and Hoseney, 2010). There are a number of bread types that can be made from dough. For example, Pan breads, Hearth breads, Flatbreads, Tortillas and Steamed breads and buns (Blakeney et al., 2009).

1.2.2. Pasta

The history of pasta goes back a long way, and it is believed that pasta was first made in China and then was brought to Italy by Marco Polo in 1292 A.D (Sissons, 2004). There are a number of characteristics such as versatility, ease of transportation, handling, cooking, and storage properties, availability in numerous shapes and sizes, high digestibility, good nutritional qualities and relatively low cost that has made pasta popular with a significant number of the world's population. Durum semolina is the preferred raw material of manufacturers for making pasta. There are about 600 shapes of pasta produced all over the world, and the most popular is spaghetti. In Italy, pasta shapes can be classified as long goods (spaghetti, vermicelli and linguine. short goods (elbow, macaroni, rigatoni and ziti); pasta containing egg; and speciality items (lasagne, manicotti, jumbo shells and stuffed pasta) (Dick and Matsuo, 1988). After manufacturing pasta, there are a number of tests that are done to evaluate the quality of the product (Sissons et al. 2012). For consumers cooking quality and colour are the most attractive characteristics of pasta.

1.2.2.1. Pasta quality

There are a number of characteristics that made pasta popular such as immaculate storability, exportability, handling in addition to the easiness of preparation and nutritional benefits (Nisha et al, 2014). Pasta normally is made from durum wheat after being milled into semolina (Pollini, et al 2012), and pasta has a low glycaemic index source of food (Granfeldt and Björck, 1991).

Pasta is the product of mixing semolina with appropriate and recommended amount of water (Pollini et al, 2012). Produced pasta is subject to a series of assessment tests for determining the quality (Sissons et al 2012). Consumers are interested in cooking quality and colour as the most attractive characteristics of pasta. Therefore, when pasta is cooked it should not have a thick appearance or be converted to a sticky material. Firmness and stickiness tests are used for determining the quality of cooked pasta, and proteins affect both. In addition, the tri combination among drying temperature, protein and gluten quality was found by Cubadda et al, (2007) to have an impact on cooking quality. Yellow colour of pasta is preferred. However, brown or red colours due to extreme drying conditions are not desired (Feillet et al 2000). Carotenoids content in grain influences semolina and then pasta colour in addition to decrease the pigments by lipoxygenease through the oxidation affect during processing. (Irvine and Winkler1950, Irvine and Anderson 1953, Borrelli et al 1999). The acceptability of the produced pasta by the consumer depends on the cooking quality of the product. Pasta can be tested instrumentally, chemically and sensorially such as firmness, stickiness and bulkiness in addition to aroma and taste (Matsuo, 1988). The instrumental tests such as firmness, stickiness and bulkiness for assessing cooked pasta are normally done after cooking pasta to optimum cooking time. Optimum cooking time is the disappearance of the white starchy-centred thread along the pasta strand (Cubadda 1988). In addition, testing cooked weight and cooking loss give an indication about the cooked pasta quality as they give information regarding the absorption of water by pasta strands and the resulting leaching of starch into the cooking water that may increase the weight and volume of cooked pasta (Sissons et al, 2012). The impact of SF fumigation on durum wheat, semolina and pasta technological characteristics has not yet been reported.

1.3. Grain quality

Grain quality is subject to the effect of field conditions before and during harvest, and is also due to the impact of storage conditions. In the field, the environmental effects happen at the time of grain maturity. Therefore, the determination of the suitability of the grain for harvest, and the manner of harvest are important. After harvest and even before handling grain for storage, there can be insect or fungal infestation affecting grain quality (Rajendran, 2003). For example, some insects infest grain even before storage. In addition, a number of crops are harvested with 15% moisture content or higher, increasing the probability of fungal infection unless the grain is dried artificially or naturally (Sample, 1992). Storability is a crucial aspect in grain storage, for that reason, a number of technological, physical and chemical examinations of the grain are performed before storage (Multon, 1988). A critical factor is grain moisture content. If moisture is too high, a rapid infestation and multiplication of insects can occur. In addition, handling high moisture grain for drying or storage may cause damage to the grain to some extent.

Not only do field conditions affect the quality of grain, but also physical factors during storage can have a significant impact on the grain quality. Quality and quantity losses of grain were reported to be of about 50% in some countries (Parpia, 1976). Losses of grain during storage happen due to infestation by insects, rodents, birds, microorganisms and respiration. Importantly, insects are reported to be the first enemy of stored grains (Rajendran, 2003). About 100 species of insects are known to infest stored grain (Rajendran, 2003). Moreover, they develop in a rapid manner and effective ways for controlling them are limited. Losses of grain value during storage are strongly affected by temperature, moisture and grain respiration. The best temperature for insect growth is 25 to 32°C (Rajendran, 2003). Respiration has a significant influence in grain quality. High respiration rate degrades the nutritive quality, viability and the processing quality of grain products (Pomeranz, 1992).

Test weight is broadly used as one of the factors for assessing grain grade defined as the bulk density of the grain or the mass of the grain in a given volume. The results are in kilogram per hectolitre as in the Schopper chondrometer that is equipped with a 1 L container (Sissons et al., 2012). Grain hardness is ranked as an important characteristic of grain. The hardness of the grain reflects the resistance of the grain against any physical action of breaking or converting the whole grain to smaller particles. Hardness varies between wheat types and even from grain to grain. The two main factors involved in controlling hardness are genetic and growing environment. The importance of hardness comes from the resistance against insect, the resistance to breakage through handling and the damage to starch during dry milling (Hoseney, 1986). One thousand grain weight is considered as an indicator for grain size. Sometimes the variation of 1000-grain weight is recorded within the same variety due to the effect of growing environment and filling of grain or the maturity. For transportation and handling purposes, this weight helps provide an idea of required equipment and size. This weight gives an idea about grain size as well (Sablani and Ramaswamy, 2003). Protein content of the grain is considered internationally as important characteristic of grain. Its importance comes from its impact on the particle size during milling process. Therefore, high protein content grain is in the interest for producing uniform pasta product (Sissons et al., 2012). In addition, ash content is an indicator of the mineral levels in grain, and it is evaluated as a factor for assessing the refining process of semolina as bran has much higher ash content that semolina.

1.4. Grain storage and protection

Food security or protection of the viability and safety of food is very important. Wheat grain is typically stored in large silos until processed and this period can vary considerably. Storage is a crucial process after harvest as it ensures a stable supply of grain (World Bank, 1986). Farmers also preserve cereals for their own use and for replanting; the grain industry stores grain to market and governments to maintain a reliable supply of food for their people. Storage procedures and methods are extremely important due to the threat of lost quality and viability (Jayas et al., 1995).

Optimum storage conditions are crucial for grain, and should be achieved before storing grains. After harvest, cleaning and drying are done to prepare grains for storage. Facilities for storage need to be provided and should be suitable for sorting, cleaning, sizing, drying and fumigation. In addition, optimum or safe storage should include maintaining grain quality characteristics, and this includes keeping grain quality safe from the impact of environmental conditions, moisture and temperature, moulds, microorganisms, insects, rodents, birds, unwanted odors or contaminants (Bailey, 1992).

Grain protection is important to maintaining grain quality. Protecting grain in storage facilities is achievable and can be done physically, chemically, biologically and using expert systems (Rajendran, 2003). Physical protection for the grain does not include chemical treatment for the grain. Therefore, it is free of chemical residues. Physical protection mechanisms include:

1. Sanitation of storage facility from insect infestation (Longstaff, 1994),

2. Drying grain before storage to achieve safe storage moisture limit (Ren et al., 1996),

3. Aeration cooling helps keep storage temperature to within the safe limits to prevent insect activity, as temperature is an important factor for insect population growth (Maier, 1996)

4. Monitoring the grain storage facility and inspecting it for infestation helps discover the infestation early making it treatable.

5. Inert dusts such as wood ash, paddy husk ash, kaolins, lime and clay are traditional treatments applied to storage systems and were considered as grain preservation materials (Golob, 1997).

6. Temperatures between 25-32°C are optimum for insect population growth. Therefore, temperatures higher or lower cause delays in development of the insects, prevent reproduction and may even kill the insects (Evans et al., 1983).

7. Irradiation is one of the physical methods for grain protection. This method was approved for use by FAO/IAEO/WHO with doses up to 10 kGy. This radiation can be obtained from cobalt-60, cesium-173 and accelerating electrons of less than 10-V (Ahmed, 1990).

8. Controlled atmosphere is a method to control the storage system using atmospheric gases with extremely low oxygen concentration. For example, to be effective this would be 0.5% oxygen and 99.5% nitrogen, instead of the normal gaseous composition of 78% nitrogen, 21% oxygen and 0.03% carbon dioxide (Paster et al., 1991).

Chemical methods can be used in stored grain to control insect infestation and these include fumigants, contact insecticides and insect growth regulators.

1. Fumigants are gases that are applied to the storage system for rapid disinfestation, and fumigants are considered the fastest way to control insect infestation. The effective impact of the fumigant depends on the concentration applied, exposure time, temperature, moisture, the density of infestation, fumigant type and the availability of a gastight storage system. Sorption of the fumigant by the commodity during fumigation may leave unwanted chemical residues (EPPO, 1984).

2. Contact insecticides are another useful method for preventing insect infestations in stored grain. These substances are applied to the grain before storage (Snelson, 1987) but the presence of residues may make markets reluctant to accept protectant treated grain.
3. Insect growth regulators work physiologically inside the insect body after penetrating it. They target the metabolic pathways preventing the early life stages from becoming an adult (Bengston and Strange, 1994).

Another option for protecting grain is biological control. Natural enemies of stored product insects (such as predators, parasitoids and pathogens) may be used for controlling insects that affect grain storage. This protection is considered as a residue free procedure (Harein and Davis, 1992). In addition, expert systems can be counted as one of the important elements in grain protection, as grain storage and protection is a complex procedure. Having enough experience on specific details for storage factors such as temperature, moisture, gas degradation and degree of infestation level is important for appropriate advice. For this, computer programs are available and they are called expert systems or decision support systems (Wilkin et al., 1990).

1.5. Importance and management of insect pests of cereal grains in Australia and Iraq

The importance of managing insect pests that infest cereal grains and other stored products comes from the value of the stored products and the subsequent damage to the grain that occurs due to the infestation. Insects are the main predator for stored grains, (Pedersen, 1992), and there are more than 100 species that infest stored products. However, the important species are listed in Table 3 (Rajendran, 2003). Insects consume significant amounts of the stored grains and convert them to un-acceptable waste products. This damage can be classified as direct and indirect.

Direct damage is where the insect consumes the grain entirely or some of its components. For example, the rice and maize weevils, lesser grain borer, and Angoumois grain moth target the endosperm of the grain (Table 3). The larval stage of Angoumois grain moth feeds on the embryo for its growth and development (Bell, 1971). In addition, the digging of some species of insects into the wooden structure or storage system is considered as a direct damage to the storage structure. For example, the cadelle larvae have the ability to make holes in wooden structures and sieve frames and other wooden tools that are used in storage systems or flourmills resulting in the build-up of dust (Wilbur and Halazon, 1965).

Indirect damage occurs when cereals become contaminated with eggs, larvae, pupae, or even adult insects inside the grain. Once inside the grain, it is impossible to remove them, and they will appear in processed products made from the contaminated grain (FDA, 1988). The indirect damage happens through the storage atmosphere heating up, facilitating mould infestation, which then makes the product unacceptable to the consumer. Consumption of the grain by insects causes heating up of the storage systems. The insect's metabolism produces the build-up of carbon dioxide, water and heat in the storage system. (Cotton et al., 1960). Insect and mite infestations create mould development by increasing the moisture through production of metabolic water. This makes the environment more conducive to fungal growth and spread of spores through the grain bulk (Christensen and Kaufmann, 1969). If a potential buyer finds an infestation, the grain or its product is then rejected. Sometimes a full load is rejected by finding one insect (Pedersen, 1992).

Table.1.3. Common and scientific names of the important insect pests of stored cereal grains and the major commodity that can be infested by each insect.

Common name	Scientific name	Major commodity
Beetles		
Lesser grain borer	*Rhyzopertha dominica*	Wheat and paddy
Rice weevil	*Sitophilus oryzae*	Wheat and rice
Maize weevil	*Sitophilus zeamais*	Maize
Granary weevil	*Sitophilus granarius*	Wheat
Khapra beetle	*Trogoderma granarium*	Wheat and rice
Larger grain borer	*Prostephanus truncatus*	Maize
Red flour beetle	*Tribolium castaneum*	Rice
Confused flour beetle	*Tribolium confusum*	Rice
Long headed flour beetle	*Latheticus oryzae*	Rice
Sawtoothed grain beetle	*Oryzaephilus surinamensis*	Rice
Rusty grain beetle	*Cryptolestes ferruigeus.*	Rice, wheat
Moths		
Angoumois grain moth	*Sitotroga cerealella*	Paddy
Almond moth	*Ephestia cautella*	Wheat and rice
Rice moth	*Corcyra cephalonica*	Rice
Indian meal moth	*Plodia interpunctella*	Wheat
Psocids		
Booklice	*Liposcelis* spp.	Rice

The lesser grain borer, *Rhyzopertha dominica* (F.) is one of the most important enemies to the stored grains. Mahroof et al. (2010) mentioned that the insect has the ability to fly and travel from agricultural areas to a non-agricultural areas and then return. Temperatures of 18.2 to 39°C, moisture up to 14% and 70% relative humidity represent the developmental range for *R. dominica* (Longstaff, 1999). The lifecycle of *R. dominica* has four stages: egg, larva, pupa, and adult. Although all the different life stages of the insect can be controlled with phosphine (Collins, 2006; Daglish et al., 2010), which is a cost-effective fumigant, *R. dominica* has shown strong resistance against phosphine and other grain protectants more recently (Collins, 2006). Resistance to phosphine is a concern all over the world, and finding an alternative is difficult (Lorini and Collins, 2006). The lesser grain borer has two genes endowing strong resistance to phosphine (Collins et al., 2002). To be effective, a longer exposure time or an increased concentration of phosphine is required to obtain complete control (Collins, 2006).

1.6. Characteristics of fumigants used for grain fumigation

Fumigants are gases at room temperature and toxic to many life forms (Bond, 2007). There are significant numbers of gases that can be used as fumigants against infestation in stored grain facilities. Toxicity of fumigant against insects should be high or achieve complete control of infestation. However, the efficacy of the fumigant against insects is affected by storage factors. These factors are temperature (Sun, 1946, Moore, 1936;), humidity (Cotton and Young, 1929; Jones, 1938; Kashi and Bond, 1975; Bond and Buckland, 1978), insect resistance to the fumigant (Champ and Dyte, 1976), and potential sorption of the fumigant by the commodity (Banks, 1990). Sorption is the loss of the fumigant from the fumigation system into the fumigated commodity (e.g. wheat grain) stored in that system. The choice of fumigant is crucial as sorption may decrease the concentration of the fumigant to under the lethal level, and decrease the efficacy of the fumigant against the pest (Daglish and Pavic, 2009). In addition, sorption can result in unwanted residues in the commodity, as the gas may not desorb completely from the commodity (Hilton and Banks, 1997a). Sorption has two phases: physical and chemical. Physical sorption is sorption of the gas by the grain but is reversible. However, when the sorbed gas is bound into the grain due to metabolic conversion of the gas into reactive ions with commodity compositions. The process is called chemical sorption and this is not reversible althogh some chemical sorption is reversable due to hydrogen bonding (Bond, 1984; Berk, 1964). Physical and chemical sorption is affected by conditions such as temperature, relative humidity, moisture content of the commodity, filling ratio (the extent of the fumigation storage vessel occupied by the commodity) and the applied dose (Lindgren and Vincent, 1962).

Residues are the amounts of the fumigant that are bound inside the commodity chemically and are not desorbed from the commodity (Bond, 2009; Hilton and Banks, 1997a; Lindgren et al., 1968). Recently, the issue of pesticide residues in foodstuffs has been raised by (FAO) Food and Agriculture Organisation and World Health Organisation (WHO) (Bond, 2007). Chemical residues left after fumigation may cause serious health problems. In addition, residues may affect animals and living plants physiologically due to inhibiting growth. Residues may affect the germination and seedling growth of fumigated seed. Some possible chemical impacts of fumigant residues could be the creation of unpleasant odours from the fumigated commodity. In addition, chemical reactions with metal (e.g. phosphine with copper) that causes corrosion (Lindgren et al., 1968).

The impact of the fumigant can be seen in obvious damage such as a heavy corrosion to the storage, shipping and fumigation containers. Fumigated structures, containers and related fittings should be free of spoilage due to fumigant adhesion. Leaving chemical compounds in food products may be harmful to health due to poisoning. They could leave undesired coloured spots or create unpleasant odours. Therefore, a suitable fumigant should be unreactive and easily and quickly removed from the commodity after fumigation. Sorption of the fumigant by grain does not just leave undesired residues, but it may also impact the tissue of the grain negatively, affecting the germination outcomes of the grain (Bell, 2006).

Public health and worker safety is crucial in fumigation procedures (Calvert et al, 1998). Some of the fumigants were reported to cause long term damage showing that some gases are poisonous to humans. A fumigant such as methyl bromide has been phased out of use because it causes depletion of the ozone layer. Therefore, the use of some fumigants has been banned or restricted (Heuser, 1975).

1.7. Principles of fumigation

Fumigation is releasing a gas substance into the storage system to control insects. Fumigation is a procedure that includes injecting a toxic chemical (in a gaseous state) into the storage system to reach any target pest (USDA, 2014). Fumigants are restricted gases in terms of their application because in addition to insects they are toxic to humans and animals. Therefore, only qualified people should deal with them (Baker and Carlo, 2003). The label on the fumigant container is very important as it relates information about precautions against the toxicity of the stored fumigant, and about methods of handling and dealing with the fumigant. During fumigation, signs need to be displayed clearly. (Baker and Carlo, 2003).

Identifying the pest involved before choosing the type of fumigant is important, as is calculating the required dose. Identifying the type of the pest gives information about the biological level of resistance in order to calculate the appropriate dose of the fumigant (Bond, 2007). Many species of insects have developed resistance against the applied fumigant phosphine (Champ and Dyte, 1976). The fumigant sulfuryl fluoride was used in Australia as a resistance breaker for the strongly phosphine resistant rusty grain beetle, *Cryptolestes ferrugineus* (Australian Grain, 2013).

Dosage is the amount that is calculated by the weight of the fumigant as related to the volume of the fumigation system. For example, milligrams per litre (mg/L). The method of fumigant application depends on the type of the structure and fumigant. For example, aluminium phosphide to produce phosphine gas is applied to wheat storage silos as tablets or as bag chains (Warrick, 2011). However, SF is applied to flourmills or the other structures as a gas from high pressure steel cylinders (Cox, 1997), and methyl bromide was applied to soil as liquid (Blecker and Thomas, 2012). The ability to detect fumigant is crucial as unqualified people may not be aware of the presence of the fumigant in a fumigated atmosphere.

Therefore, gas detection equipment is necessary for detecting the gas (Thain, 1980).

The pressure test is one of the fumigation principles. It is broadly practiced in Australia and recommended by the Australian standard (AS2628). This test has a relationship with both storage and fumigation processes. It involves testing the pressure of the silo before storage. It is beneficial for insuring a gas tight silo, so that there will be no chance of a gas leak. Gas leaks help the development of insect resistance by leaving the resistant individual alive. In addition, gas leakage threatens personal safety as fumigant leaks into the atmosphere (Warrick, 2011). Therefore, first aid and medical supervision are necessary as a part of the fumigation procedure (Bond, 2007).

Table.1.4. Common fumigants.

Common name	Chemical formula
Phosphine	PH_3
Sulphuryl fluoride	SO_2F_2
Methyl Bromide	CH_3Br
Hydrogen cyanide	HCN
Ethylene dibromide	$(CH_2BR)_2$
Ethylene oxide	$(CH_2)_2O$
Ethylene dichloride	$CH_2Cl\ CH_2Cl$
Carbon disulphide	CS_2
Carbon tetrachloride	CCl_4
Chloropicrin	$CCl_3\ NO_2$
Dichlorvos (DDVP)	$CCl_2 = CHO.P0.(OCH3)_2$
Acrylonitrile	CH_2: CH.CN

Source: Bond, 2007.

1.8. Principles of sorption and desorption

Sorption is a term that refers to the movement of the fumigant gas into the commodity while desorption is the movement of fumigant from the commodity back into the vessel (Darby, 2008; Dumus, 1980). Sorption has two phases, chemical and physical (Banks, 1986), and it is usual for both phases to occur together resulting in a nonlinear loss of the fumigant into the commodity (Hilton and Banks, 1997a, b; Daglish and Pavic 2008 and 2009, Darby, 2008 and Darby et al 2009). Gas diffuses into the commodity in two stages referred to as adsorption and absorption. Adsorption happens when the gas molecules are attracted by the commodity surface and linked to it by van der Waals forces (Ruthven, 1984). Therefore, adsorption needs two phases to be completed. First the gas molecules (known as the adsorbate) gather over the surface of a commodity (known as the adsorbent). Second, the adsorbent then attracts and adsorbs the molecules of the adsorbate (Crini and Badot, 2010). The commodity surface molecules are relatively unstable because of the positive surface free energy. For the grain or the particle, there are unbalanced attraction forces on the surface and balanced attraction forces inside the grain or the particle. These unbalanced attraction forces are actually attracted to those that are balanced (Weber et al, 1991). Due to the positive free energy of the surface molecules and instability of the grain or the particle, these forces on the surface of the grain or the particle have a tendency to attract the molecules of the fumigant to minimize the surface energy. The unstable attraction forces for the grain and the particle happen repeatedly as the balanced forces inside the grain or the particle are attracting the unbalanced surface forces. In this way, the gas molecules move from the surface of the grain or particle to inside and may be retained inside by chemical bonds.

Absorption occurs when the fumigant molecules move from the surface to inside the grain or the flour particle and bond with compounds inside the grain (Sriranjini and Rajendran, 2008). Therefore, absorption is a bulk phenomenon while adsorption is a surface phenomenon.

Sorption and desorption are extremely important issues in the process of fumigation of stored products for a number of reasons. Firstly, they control the toxicity of any fumigant against insects. When sorption occurs during fumigation, the targeted insects may not be exposed to a lethal gas concentration. As is well known, several stored product insects have different levels of tolerance to fumigants. This makes sorption an important factor that should be considered during the fumigation process. When desorption occurs (after cessation of exposure to the fumigant), this also exposes insects to the fumigant. Secondly, sorption of the fumigant by the commodity may affect the chemical composition of the commodity due to the chemical sorption, and this may lead to changes in food processing characteristics and food safety. Sulfuryl fluoride (SF) contains in its chemical structure the ions of sulphate and fluoride, and these ions may have negative effects on health (APVMA, 2007) or commodity quality.

1.9. Residues in wheat

A residue according to FAO (Bond, 2007) is "a pesticide chemical, its derivatives and adjuvant in or on a plant or animal". The residues are expressed as mg/kg/day that represents the allowed daily intake. The mg refers to the residue amount, kg is the body weight of a person and the day is 24 h. Sorption of the fumigant by commodity may leave different levels and types of residues of each fumigant. These residues depend on the reaction between the fumigant molecules and the stored reserves in the grain or the products of the grains. It is recommended that a fumigant should be desorbed easily from the commodity with time. However, some of the fumigants that have high sorption rates with the commodity leave high levels of residues even after aeration. For example, ethylene dibromide and hydrogen cyanide (Amuh, 1975; Jagielski et al., 1978; Lindgren et al., 1968). Fumigants also react with the commodity compounds forming toxic substances. For example, chlorohydrins and bromohydrins are produced when the fumigant ethylene oxide penetrates the commodity (Scudamore and Heuser, 1971). However, Winteringham et al. (1955) mentioned that methyl bromide reacts with wheat without forming any toxic compound, as it is desorbed from wheat grains after only a few hours (Dumas and Bond, 1977).

The fumigant residues formed depend on the nature of the fumigant. Treating wheat with acrylonitrile leaves fumigant residues for many days. Moreover, the temperature of fumigation has a crucial impact on the residues that can be left after fumigating wheat grains or their products. The residues of the fumigant methyl bromide increased with increasing fumigation temperature even when the applied dose was reduced (Lindgren et al., 1962; Vardell, 1975). The moisture content of grain or the humidity during fumigation has an impact on the fumigant residues. Increasing the moisture content of the commodity may lead to an increase in residues (Sinclair et al., 1964). Humidity of the fumigation system was described to have an important impact on fumigation sorption and residues. Dumas and Bond (1979) found that wheat grains desorbed ethylene dibromide at high atmospheric humidity more than at lower humidity. Multiple fumigations were found to influence the residues of the fumigant methyl bromide. The cereals that were subjected to successive fumigation gave higher residue levels that increased with increasing number of fumigations. This does not just apply to the whole grain, but also affects the flour that was produced from milled grains (Kawamoto et al., 1973; Vardell, 1975; Banks et al., 1976).

1.10. Sulfuryl fluoride

Sulfuryl fluoride (SF) or (SO_2F_2) is known as ProFume or Vikane. It is nominated as a finite use gas, and it has broad spectrum as a fumigant. It can be created from reacting barium difluorosulfate or silver fluoride with sulfur dioxide under heated conditions. When it is sorbed into the grain and an insect, it is broken down into sulfate and fluoride anions. SF is compressed in steel cylinders under high pressure so when it is released from the storage container, it becomes a gas at atmospheric pressure. The label Vikane means the fumigant is prepared for use in storage and handling facilities, structures, buildings, shipping containers and vehicles etc. (Cox, 1997). Vikane has chloropicrin added as a warning agent which has a pungent odour. The ProFume label means the gas does not have chloropicrin agent, and is for commodity fumigation uses (INTERIM, 2008).

1.10.1. Chemical and physical properties

Table.1.5. Chemical and physical properties of sulfuryl fluoride

Chemical formula	SO_2F_2
Molecular weight	102.1
Boiling point	-55.2°C
Melting point	-120°C
Physical state	Colourless gas
Solubility in water	750 mg/kg @ 25ºC
Vapour pressure	12,750 mmHg @ 21.1ºC
Flammability	None
Conversion factors	1 ppm = 4.17 mg/m3 1 mg/m3 = 0.2392 ppm
gas (air = 1)	2.88
liquid (water at 4°C = 1)	1.342 at 4°C
Vapour density (air =1)	3.5
Commercial purity	98%

Sources: EPA, 1993, Cox, 1997 and INTERIM, 2008

1.10.2. Historical use

SF was produced by Dow AgroSciences as a fumigant gas for use against insects and rodents in enclosed structures. SF gas is 99.8% pure and the remaining 0.2% includes other ingredients not specified (Prabhakaran, 2006). The first registration for SF was 1959 in the United States of America, but there were reported negative impacts of it on public health. The EPA (US Environmental Protection Agency) reissued the registration in 1993 after testing the product for acute and chronic symptoms. (EPA, 1993).

The use of the of SF continued until 2004/2005 until it was registered by EPA for use in the fumigation of cereal grains, dried fruits, tree nuts, cocoa beans, coffee beans, and also in food handling and processing facilities. (http://www.epa.gov/pesticides/sulfuryl-fluoride/evaluations.html) In 2011, SF was re-evaluated and was found to exceed the safety standards for fluoride exposure by the American Federal Food, Drug, and Cosmetic Act (FFDCA). It was recommended that the fumigant should be withdrawn from use within three years (while an alternative fumigant was found to replace methyl bromide). In Australia SF was registered in 2007 with a maximum CT (Concentration x Time) of 1500 mg h/L (http://mbao.org/2010/71Barnekow.pdf). It was introduced in Australia as an alternative to methyl bromide.

1.10.3. Use as a fumigant in stored products

The first usage of SF was 45 years ago in order to fumigate dry wood termite, (Bell et al., 1999). The information regarding using SF in stored products is still under testing, and the impact on the quality of fumigated products is not known (Navarro, 2006 and Bell, 2006). The trade name Vikane is available for use in Germany, USA, and Sweden for stored products fumigation. However, the trade name ProFume is used for stored products protection (Schneider, et al., 2003).

1.10.4. Toxicity against insects

Results of experiments on the toxicity of SF on different species of insects indicated that the egg life stage was the most tolerant to SF compared to the other life stages (Su and Scheffrhan, 1990; Baltaci et al., 2009). Bell et al. (2003) showed that a CT product of 500 g h/m^3 at 30 °C or 1000 g h/m^3 at 25 °C was sufficient to kill all the eggs of most species. The authors also showed that strains of *Tribolium castaneum* resistant to phosphine did not show cross-resistance to SF. Bell et al. (2003) concluded that high concentrations and short exposure periods were sufficient to control the egg, larval and pupal stages of insect populations. Moreover, Reichmuth et al. (2004) assessed the efficacy of SF CTPs from 1860 to 2255 g h/m^3 against all life stages of stored product insects by introducing them into airtight sealed mills at temperatures ranging from 20 to 40°C, and at relative humidity's from 20 to 80% for 36 to 48 h. The study revealed that very high mortalities of all tested species and for all life stages were achieved. SF was effective not only at the laboratory scale but also at the field scale.

1.10.5. Sorption

Information regarding sorption of SF into different commodities is scant. Furthermore, sorption of the other fumigants such as phosphine, methyl bromide and other fumigants was described with only a few studies. Sorption of sulfuryl fluoride into whole meal flour was described by Bell et al. (2004). Sorption of SF by flour up to 10°C did not impact sorption rate, but generally sorption and SF concentration was much higher with increasing fumigation temperature at the surface of flour (or the headspace). This shows the high sorption of the fumigant by the commodity compared to feed wheat used in the same experiment.

1.10.6. Effects on grain quality

No information is available about the impact of SF on grain quality.

1.10.7. Residues

The residues of concern with SF are SF itself and its metabolite, inorganic fluoride ion (Australian Pesticide Veterinary Medicines Authority, APVMA, 2007). A full assessment was done by (APVMA, 2007) where all the types of the Australian wheat were fumigated with SF to determine the residues left, in order to determine the suitability of the fumigant to be released in Australia. The maximum residue limits in Australia for both SF and fluoride were mentioned in the report. Currently the maximum residue limit in cereal grains of SF is 0.028 mg/kg and 7 mg/kg for fluoride ion. Results (APVMA, 2007) indicated that residues of SF and fluoride in Australian wheat were under the maximum residue limit. Meikle (1964) reported results where wholemeal flour contained residues. One third of the residues were insoluble, but two thirds of the residues were quantifiable chemically.

The residues of SF were found to both react to and bind to the amino groups of proteins. Furthermore, Osbrink et al. (1988) evaluated the residual amounts of SF in eight different foods protected by polyethylene bags. Results confirmed the significant effect of aeration on SF residues as SF was desorbed completely after five days of aeration. In addition, Scheffrahn et al. (1989) quantified fluoride and sulfate ions in food commodities after fumigation. Unpacked, eight food items were exposed to 36 and 360 mg/L SF for 20 h and aerated for 1, 8 and 15 h. SF was sorbed by the food items with different rates depending on commodity type, and sorption percentage increased with increasing dosage of SF. The highest level of fluoride ion was found in dried beef and the highest residue of the sulfate ion was discovered in dry milk. There was no significant effect of aeration time on the residue levels of fluoride or sulfate ions. Meikle and Stewart, (1962) recorded an increase in SF residues with increasing temperature.

1.10.8. Toxicity against human and animal

A number of deaths have been reported due to direct and indirect inhalation and skin sorption of SF. Scheuerman (1986) said three people at different ages were reported dead after fumigating their properties with SF. The same year a fumigator allowed a husband and a wife to enter their home eight hours post ventilation, and their deaths were attributed to SF (Nuckolls et al., 1987). In addition, some cases were reported as toxic but not lethal, for example Taxay (1966) reported that a fumigator had experienced SF and chloropicrin (which has warnings about irritation on the label) inhalation for four hours. That person then reported to the hospital with symptoms and was discharged after half a week.

Torkelson, (1959) exposed rats to SF inhalation (1000 to 15000 mg/L) ranging from 6 min to 6 hours. Rats exposed to 15000 mg/L for twelve minutes died after 3 h, and exposing rats to 800 mg/L for 30 minutes caused death as well. However, ten percent of the rats died after exposure to 1000 mg/L for two hours. In addition, Eisenbrandt and Nitschke, (1989) reported 90% of exposed rabbits died between 2 to 6 weeks of exposure to 600 mg/L of SF.

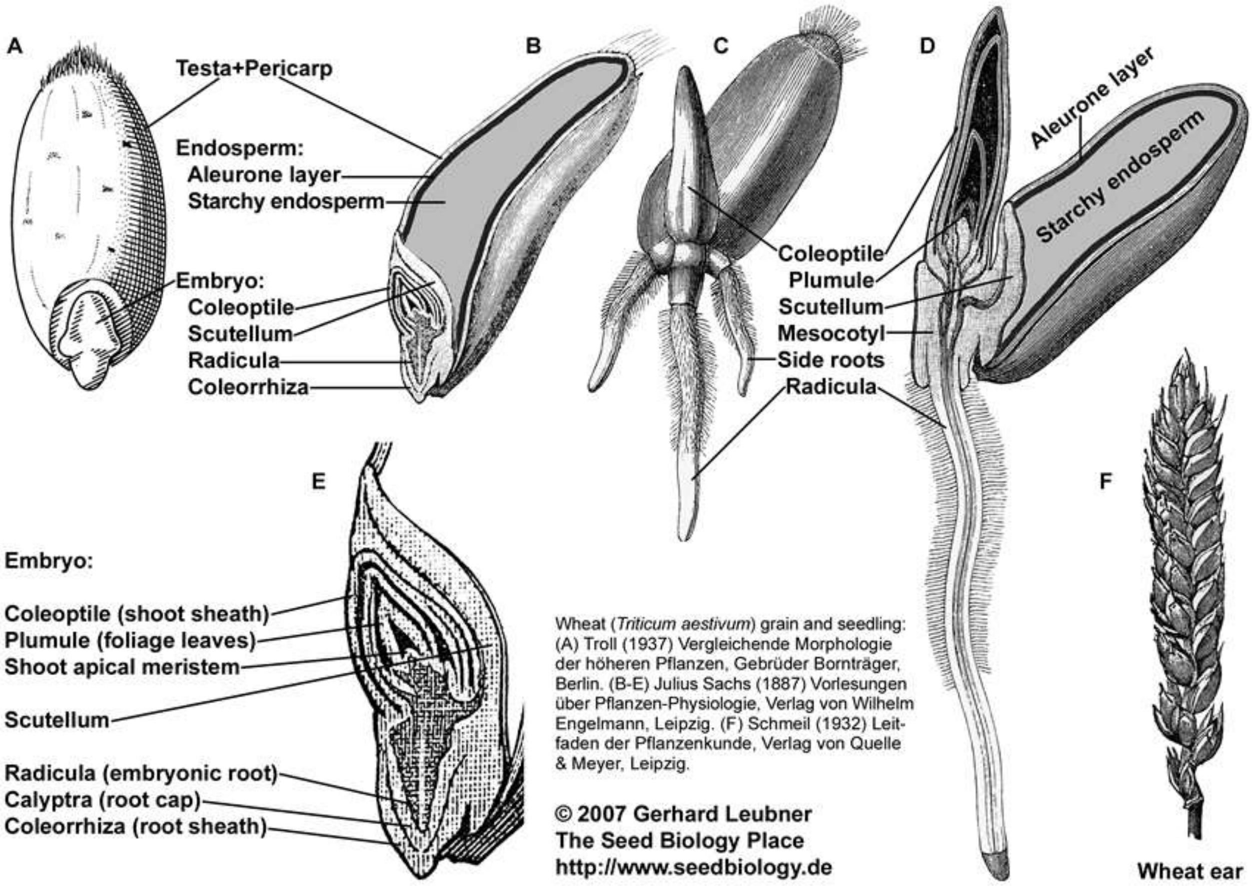
1.11. Germination

Germination is defined as seeing the first sign of growth from a kernel, for example the appearance of protrusion of the radical (Black, 1970). Understanding the germination process and the changes that happen in grain during germination helps explain issues that relate to any change in germination percentage with fumigation. The germination process has three phases, imbibition (water absorption by seed), activation and visible germination. The imbibition itself happens within three phases as well. These phases show rapid imbibition, then a gradual phase, and finally another increase in water absorption by seed after the germination when it starts the seedling stage (Bewley, 1997). After adding water to seeds for germination, normally the seed imbibes the water. During that, there is no change in carbohydrate levels until after 10 to 12 h of germination. Enzymes appear in the scutellum and coleorhiza, and starch in the root cap (Choate, 1921). When the water penetrates the cell, it causes some disruption of the cell membrane. This leads to a leak of solutes and light molecular weight substances from the cell out to the liquid around the cell. This happens due to changing membrane structure. However, shortly after rehydration, the membrane goes back to its previous structure and stops leaking solutes (Crowe and Crowe, 1992). With the start of imbibition, a clear seed respiration is starting which is indicated by the drop in the high consumption of oxygen (Bewley and Black, 1994; Botha et al., 1992). Glycolytic and oxidative pentose phosphate pathways are both present during the first phase of rapid sorption of water. Moreover, Kreb's cycle enzymes are activated (Necolas and Aldasoro, 1979). Some of the enzymes that are responsible for activating the germination process are present in the dry seed originally. Therefore, a number of metabolic processes happen immediately with the water imbibition. Reducing the availability of oxygen around cells decreases the diffusion of gases through seed structures, leading to the structures becoming dense and producing ethanol (Morohashi and Shimokoriyama, 1972).

Mitochondrial organelles already exist in the tissue of the dry seed, and have Kreb's cycle enzymes and oxidising agents. That enables mitochondria to provide ATP (adenosine triphosphate) energy for a number of hours to aid the metabolic process (Ehrenshaft and Brambl, 1990). In addition, more mitochondria are synthesized to continue the germination process. The development of mitochondria during embryo germination stage depends on the type of the stored reserves. If the reserves are wheat starch, mitochondria that are already in the dry seed are repaired. However, if the stored reserves are oils, new mitochondria are produced (Morohashi and Bewley, 1980; Morohashi, 1986).

The next step of activation is protein synthesis. During this process, proteins are created from stored mRNAs in embryo. These messages are used by the DNA for the seed development in order to complete maturity of cells and tissues of different seed parts. Then used again at the beginning of germination during the imbibition (Comai and Harada, 1990; Lane, 1991), and then proteins are synthesised by the newly created mRNAs continuously. Once the radical appears (visible germination), the germination process ends. The radical extends through the structure that surrounds the embryo. The radical starts growing during the imbibition phase. DNA in these radical cells are repaired because it has been damaged through the seed maturation process, by the hydration process during the imbibition, and even during the repairing the mitochondrial DNA (Zlatnova et al, 1987). The effect of fumigation on germination is explained chapter 6.

Fig.1.3. Structure and germination of wheat grain.



Chapter 2 of this thesis has been published as:

Hwaidi, M., Collins, P., Sissons, M., Pavic, H., & Nayak, M. (2015). Sorption and desorption of sulfuryl fluoride by wheat, flour and semolina. *Journal of Stored Products Research, 62*, 65-73. http://dx.doi.org/10.1016/j.jspr.2015.04.005

CHAPTER THREE

Does sorption of sulfuryl fluoride by wheat affect the efficacy of the fumigant against the adult and egg of *Rhyzopertha dominica*?

ABSTRACT

Despite its growing importance as a fumigant for grain, there is no information on the impact of sorption on the efficacy of sulfuryl fluoride (SF) against target insect pests. Eggs and adults of the major grain pest, *R. dominica*, living in wheat (12% m.c, 25°C, 60% rh) were fumigated with SF at 0.5, 1, and 2 mg/L for 168 h. Sorption of the fumigant by the grain was exponential with a rapid initial physical and then gradual chemical phase. Physical sorption decreased the mortality rate of both adults and eggs. There was a quadratic relationship between the mortality rate constants of adults and eggs with physical sorption. The impact of chemical sorption on adult and egg mortalities was less than physical sorption, and the relationship between chemical sorption and both the mortalities was linear showing that the mortality of both adults and eggs continued despite chemical sorption. Sorption of SF into the commodity has the potential to reduce the efficacy of a fumigation resulting in control failures. Concentration x time protocols may need to be revised to account for this phenomenon.

3.1. Introduction

Sulfuryl fluoride (SF) has been registered in USA for more than 50 years and was originally used primarily as a treatment for pests of buildings, particularly termites (Osbrink et al. 1987). Its use was expanded to replace methyl bromide in several applications when the latter began to be phased out of broad use under the Montreal Protocol Agreement

(Anonymous, 1994; Bell et al., 2002; Drinkall et al., 2004). These applications included use in flour mills and food handling facilities. This expansion also included disinfestation of food commodities including cereal grains (Australian grain, 2013), and dried fruit and nuts (Drinkall et al., 2004) in several countries.

Information on the efficacy of SF against insect pests is generally limited to fumigation times of 48 h or less, matching the usual application protocol for methyl bromide, which it was developed to replace. Laboratory bioassays indicate that insect eggs are generally 5-30 fold more tolerant to SF than other life stages. This has been demonstrated in beetle pests of museums (Su and Scheffrahn 1990), wood boring beetles (Williams and Sprenkel, 1990), as well as beetle (Ciesla and Ducom, 2009) and moth (Baltaci et al., 2009; Bell and Savvidou, 1999) pests of stored products. Older eggs appear to be more tolerant than early stage eggs (Baltaci et al., 2009; Williams and Sprenkel, 1990). Published information is limited, but at 25^{o}C, for example, some dosages that result in >99% or complete mortality of eggs of mixed age have been reported: 1669 mg.h/L for *Tribolium castaneum* (Bell et al., 2003); 1000 mg.h/L for flour moth *Ephestia kuehniella* (Bell and Savvidou, 1999); 960 mg.h/L for rusty grain beetle, *Cryptolestes ferrugineus*, (Baltaci et al., 2009), and 1920 mgh/L (48 h x 40 mg/L) for *R. dominica*.

The lesser grain borer, *Rhyzopertha dominica* (Fab.), is a major pest of stored cereals in warm temperate to tropical regions of the world (Edde, 2012). If left uncontrolled, it is capable of completely destroying grain stocks. In Australia and internationally, the fumigant phosphine is by far the preferred treatment for controlling infestations of *R. dominica*. However, serious levels of resistance in this pest to phosphine have been documented in several regions (Edde, 2012) threatening the viability of phosphine. In Australia, SF has been adopted as an alternative treatment for control of phosphine-resistant *R. dominica* and other pest species (Nayak et al. 2010).

An important factor to consider in the practical application of fumigants is the impact of sorption of the gas into the commodity during fumigation. Sorption may reduce the biological activity of a fumigant by reducing the concentration of gas available to target insects (Banks 1993). As outlined in Chapter 2, sorption of SF into commodities is significant, however, no information is available on the impact of sorption on the biological efficacy of this fumigant against target pests. The aim of the research described here was to determine the impact of sorption on the biological activity of SF against target pests during fumigations.

3.2. Materials and methods

3.2.1. Experimental design

Eggs and adults of a phosphine resistant strain of *R. dominica* were fumigated with three different concentrations of SF: 0.5, 1.0 and 2.0 mg/L at exposure times ranging from 24 to 168 h. The design was a randomized complete block with three replicates. Each replicate consisted of a glass Erlenmeyer flask of approximately 2.4 L capacity containing hard wheat grains *Triticum aestivum* (12% m.c.) filled to 50% flask volume (filling ratio of 0.50) and equilibrated for two days at 25°C. Flasks were sealed with a glass stopper containing a silicone rubber septum to facilitate addition of fumigant and gas sampling.

3.2.2. R. dominica culturing

QRD 569 strain was obtained from the Postharvest Grain Protection Laboratories, Department of Agriculture and Fisheries, Brisbane, Queensland. This strain is classified as strongly resistant to phosphine (Collins et al. 2002), and was cultured fortnightly at 30°C and

60% r.h. A culture was initiated at week 0 by adding 600 insects into a bottle containing certified organic wheat at 12% m.c. Adult insects were transferred at weeks 2, 4 and 6 to new grain samples. At week 8, progeny from week 0 and parents from week 6 were mixed and added to a bottle containing fresh medium to set up a new culture. The main purpose of mixing old and new adults was to maintain genetic diversity within the culture. All glassware and bottles were sterilised in the oven ($100^{o}C$) for at least 1 hour prior to use. All brass and plastic equipment was stored in an oven at $60^{o}C$.

3.2.3. Commodity moisture

Details are in chapter two, section 2.2.2

3.2.4. Calculating commodity volume V_g

Details are in chapter two, section 2.2.7

3.2.4. Fumigation

Details in chapter two, section 2.2.4

3.2.4.4. Bioassay fumigation

Bioassays were undertaken in glass Erlenmeyer flasks. The volume of each experimental flask was pre-determined by filling with water to the end of the stopper and weighing. Flasks (fumigated and control or nonfumigated) were placed in a glassware cabinet at 60°C for 24 h for complete drying. Silicone grease was added to the stoppers before closing the flasks to prevent gas leakage.

All the flasks (fumigated and control) were filled to 50% volume with wheat of 12% m.c., and 100 adult *R. dominica* (a mixture of males and females) were placed in each flask. The insects were left for 4 days without fumigation to give the adults enough time to lay eggs. Flasks (except control) were then fumigated by adding the desired volume of SF gas using syringes. The flasks were then held in a controlled environment room for up to 168 h at 25°C and 60% relative humidity. Flasks for each replicate (fumigated and control) were removed from the room periodically starting from 24 h and thereafter each 24 h for sampling the headspace in the flask to measure the SF concentration.

After completion of the fumigation, flasks were removed from the controlled environment room. The flaks were opened and aerated under a fume hood for 2 h to disperse the gas. The wheat and insects were gently sieved using a 2 mm mesh sieve to separate all adults from the grain. The grain was carefully inspected to ensure that all adults had been removed. After making sure that 100 adults were found, the adults were scored for mortality and destroyed.

The grain from fumigated and control flasks, which now contained eggs laid by the test adults before the fumigation, was then transferred to 2 L plastic jars. The lids of plastic jars had been drilled and supplied with a filter paper and piece of gauze to ensure gaseous exchange. The jars were stored at 25°C and 60% relative humidity for 8 weeks to allow all eggs sufficient time to develop into adults. After the 8 weeks, the 2 L plastic jars were opened to count the number of adults remaining.

3.2.4.7. Statistical analysis

Data were analysed using SAS software. The decay of gas concentrations and egg and adult numbers were fitted to mathematical models described below:

The decrease in gas concentration was best described as the exponential relationship:

$$C_t=C_0e^{-k*t} \quad (1)$$

Where C_t is the gas concentration C at time t. C_0 is the gas concentration C at time zero ($t=t_0=0$). At t=0 the sorption rate constant k=0 because when substituting zero into equation number (4,1) and multiplying the zero by k, gives zero sorption. In this case $C_{t=}$ C_0.

Egg survival was described as:

$$N_t=N_0e^{-E*t} \quad (2)$$

where (N_t) is the number of adult progeny (N) at a given time (t). (N_0) is the number of progeny at time zero ($t=t_0=0$). E is the rate constant of adult progeny hatched from fumigated eggs. When substituting (t) for zero in equation 2 and multiplying by (E), the result would be zero. In this case $N_t=N_0$.

Adult (parent) mortality was described as:

$$M_t=M_0e^{-R*t} \quad (3)$$

Where M_t was the number of adults M at any time t and R is the rate constant for adult mortality. M_0 was adult number M at time zero ($t=t_0=0$).

The sorption rate for a full container, k_f, and the tendency to take up fumigant by physical sorption, known as the partition rate (or co-efficient) of physical sorption, K, were each calculated following the methods of Hilton and Banks (1997a).

$$k_f=k/f, \quad (4)$$

where k was sorption rate constant from equation 1, and f was the experimental filling ratio.

$$K=(C_a-C_i)*V_g / C_i*V_f \quad (5)$$

where C_a was the applied concentration, C_i was the predicted concentration, V_g was the gas volume and V_f was the volume occupied by the commodity.

The relationships between the partition ratio, K, and the egg survival rate constant, E, and the adult mortality rate constant, R, were fitted using a polynomial quadratic regression.

$$E= B_0 + B_1K + B_2K^2 \quad (6)$$

$R = B_0 + B_1K + B_2K^2$ (7)

Where B_0 was the intercept, B_1 was the linear coefficient and B_2 was the quadratic coefficient. The turning point of the standard polynomial quadratic regression line was calculated using the following equation.

$K = -B_1 / 2B_2$ (8)

The turning point of the fitted line is the point at which the line turns back toward zero. Therefore, it is considered to be the point of maximum partition of physical sorption (*K*). This gives the maximum egg survival (*E*) or adult mortality (*R*) rate constant. After calculating *K*, equations 6 and 7 were used to calculate the maximum rate of mortality by substituting the derived values of *K* into each equation.

A linear regression formula was used to describe the relationship between both the egg survival rate (*E*) and the adult mortality rate (*R*) with the sorption rate (*k*).

$E = B_0 + B_1(k)$ (9)

$R = B_0 + B_1(k)$ (10)

In addition, the same linear equation was used to describe the relationship between egg survival (E_g) and exposure time (*t*) when the fumigant was not present.

$E_g = B_0 + B_1(t)$ (11)

Moreover, the percentage of gas loss, egg survival and adult mortality per h were calculated using the following equation:

$\text{Percentage} = 100\,(1-e^{i})$ (12)

Where (*i*) was (*k*, *E* or *R*).

Finally, time to half (equation 13) and 99% (equation 14) loss of the gas, and egg survival and adult mortality were calculated according to the following.

$1/2L_0 = L_0\,e^{-i*t}/L_0,\ 1/2 = e^{-i*t},\ t = \ln(1/2)/-i.$ (13)

$0.01L_0 = L_0\,e^{-i*t}/L_0,\ 0.01 = e^{-i*t},\ t = \ln(0.01)/-i.$ (14)

Where, L_0 was C_0, N_0 or M_0, i was k, E or R and ln was the natural logarithm.

3.3. Results

The concentration of SF in the headspace of fumigated flasks containing wheat with *R. dominica* adults and eggs present declined with time in each treatment (Fig. 1).

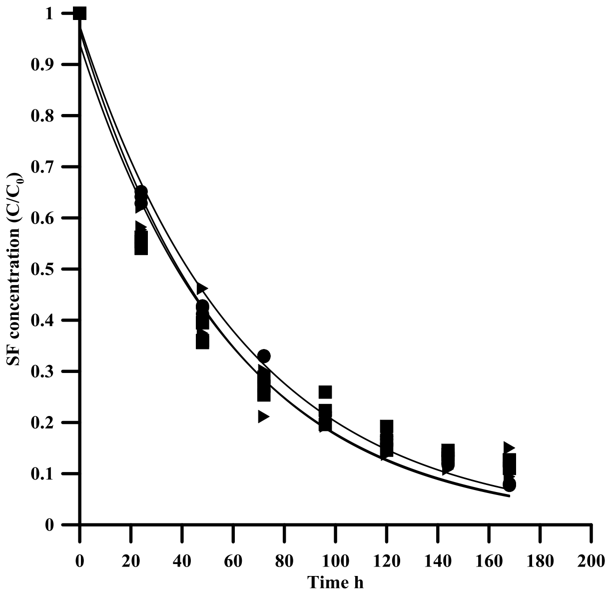


Fig.3.1. Headspace sulfuryl fluoride concentration over time in flasks containing wheat (12% mc) and *R. dominica* adults and eggs fumigated with 0.5 ●, 1.0 ■ and 2.0 ►mg/L at 25°C, 60% rh for 168h.

Table.3.1. Modelling the change in headspace concentration of sulfuryl fluoride (SF) over time in flasks containing wheat (12% mc) and *Rhyzopertha dominica* adults and eggs fumigated at different SF doses (0.5, 1 and 2 mg/L at 25°C, 60% rh). Constant rate of reaction of gas (k_f) with commodity, percentage loss per h, partition coefficient (K) and time to half and complete loss of the fumigant in half filled and full containers of wheat.

SF mg/L	Regression	Significance	R^2	% loss per h	Time (h) to 50% loss of SF at 0.5 filling ratio	Time (h) to 99% loss of SF at 0.5 filling ratio	k_f	Time (h) to 50% loss of SF at 1.0 filling ratio	Time (h) to 99% loss of SF at 1.0 filling ratio	K
0.5	C_t=0.4547$e^{-0.016t}$	$F_{2,22}$= 366.91; P<.0001	0.92	1.610	43.04	286.03	0.0322	21.52	118.53	0.339
1	C_t=0.9584$e^{-0.007t}$	$F_{2,22}$= 994.11; P<.0001	0.93	0.749	92.52	614.16	0.0149	46.26	304.58	0.147
2	C_t=1.9823$e^{-0.005t}$	$F_{2,22}$=1191.65; P<.0001	0.90	0.561	123.52	820.08	0.0112	61.76	471.42	0.03

This decline indicates that the fumigant was sorbed into the wheat, otherwise the concentration would have remained stable. Sorption was initially rapid followed by a slowing of rate. The rate of decline differed between the three SF concentrations but each was best described using an exponential model, which provided a highly significant fit to the data with all treatments (Table 1). The decline in SF concentration over time was fastest at 0.5 and slowest at 2.0 mg/L. The calculated (from equations 13 and 14) time to 50% and complete loss of SF increased with increasing applied SF concentration (Table 1).

3.3.1. Adult mortality

Fumigating with SF resulted in an exponential decline in numbers of adult *R. dominica* over time (Fig. 2). The mortality rate constant for adults *R* increased with increasing SF concentration. However, complete control of adults was not achieved in the presence of wheat even at the highest concentration (2 mg/L) of SF tested (Fig 3). The exposure period required at 2 mg/L for complete control can be predicted from the regression equation (Table 2). Thus, the exposure time would need to be extended to 422 h to achieve 100% mortality of adults, as 99% of the fumigant would be lost after 820 h (Table 1).

The partition ratio, *K*, had a strong impact on the adult mortality rate, *R*, and this effect was stronger at the lower SF concentrations (Figs 4, 5 and 6) i.e., *K* and *R* were larger in magnitude at 0.5 than at 2 mg/L. The quadratic model provided the best fit for this relationship with high R^2 values of 0.94 to 0.97 (Table 3). The relationship between *K* and *R* showed turning points at which the rate of adult mortality *R* began to decrease as *K* values increased (Figs 4, 5 and 6).

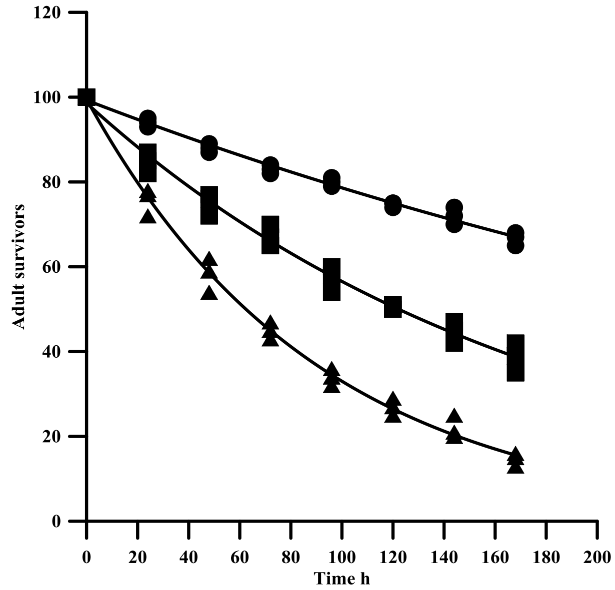


Fig.3.2. Effect of sulfuryl fluoride fumigation (0.5 ●, 1 ■ and 2 ▲mg/L) on % survival of *R. dominica* adults in the presence of wheat (12% moisture at 25°C and 60% rh).

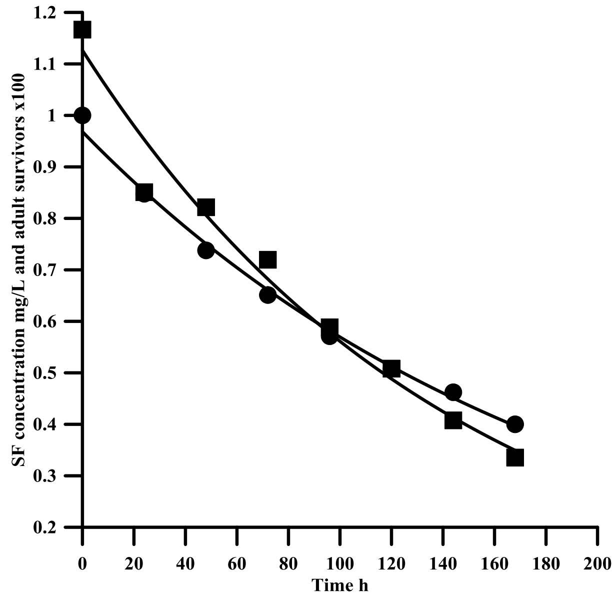


Fig.3.3. Effect of sulfuryl fluoride fumigation (■) of wheat for 168 h (12% moisture at 25°C and 60% rh) on survival of *R. dominica* adult (●).

Table.3.2. Effect of sulfuryl fluoride fumigation (0.5, 1 and 2 mg/L) of wheat (12% moisture at 25°C and 60% rh) on the adult mortality rate constant (*R*), percentage kill per h, and time to half and complete mortality of the fumigant at 0.5 and 1.0 filling ratios.

SF mg/L	Regression	Significance	R^2	% kill per (h) at filling ratio 0.5	Time (h) to 50% kill at filling ratio 0.5	Time (h) to kill 99% at filling ratio 0.5	Time (h) to 50% kill at filling ratio 1.0	Time (h) to kill 99% at filling ratio 1.0
0.5	M_t=98.87$e^{-0.002t}$	$F_{2,22}$=595; P<.0001	0.96	0.229	301.30	2002.24	602.60	4004.48
1	M_t=98.19$e^{-0.005t}$	$F_{2,22}$=109; P<.0001	0.93	0.550	125.54	834.26	251.08	1668.53
2	M_t=98.74$e^{-0.010t}$	$F_{2,22}$=715; P<.0001	0.98	1.084	63.57	422.49	127.15	844.98

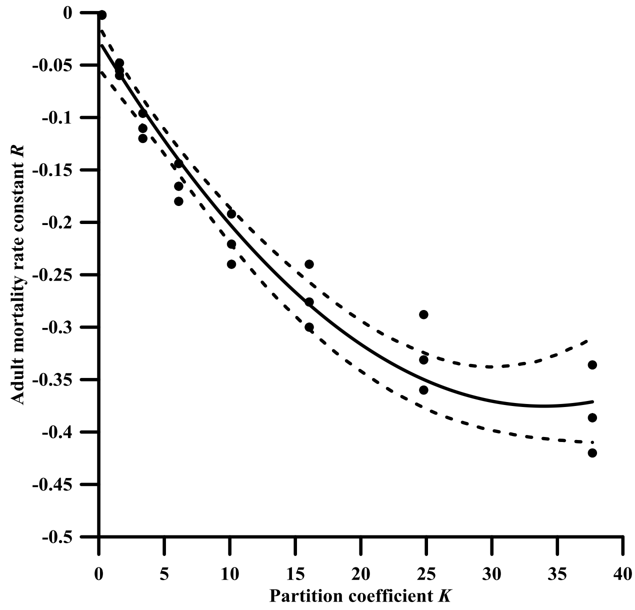


Fig.3.4. Relationship between the partition coefficient of physical sorption (*K*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 0.5 mg/L sulfuryl fluoride for 168 h versus *R. dominica* adult mortality rate constant (*R*), (---) upper and lower confidence limits at 95%.

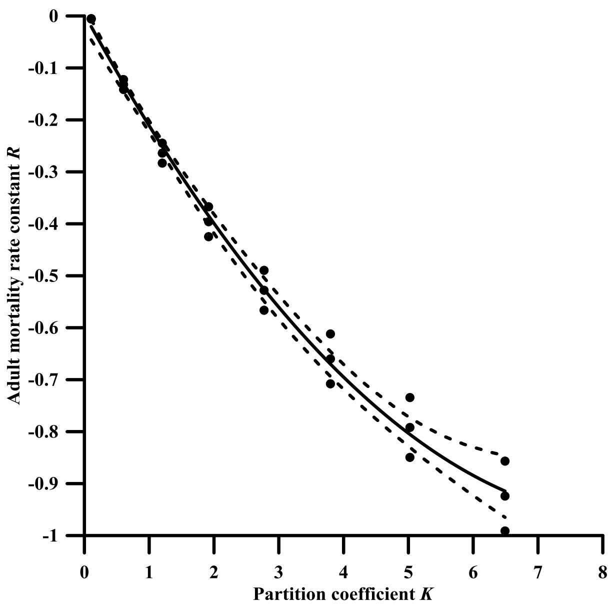


Fig.3.5. Relationship between the partition coefficient of physical sorption (*K*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 1 mg/L sulfuryl fluoride for 168 h versus adult mortality rate constant (*R*) of *R. dominica*, (---) upper and lower confidence limits at 95%.

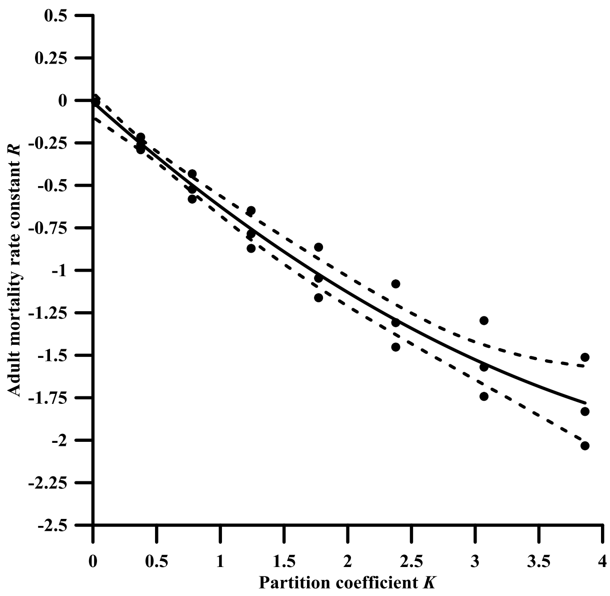


Fig.3.6. Relationship between the partition coefficient of physical sorption (*K*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 2 mg/L sulfuryl fluoride for 168 h versus adult mortality rate constant (*R*) of *R. dominica* adult, (---) upper and lower confidence limits at 95%.

Adult survival rate, *R*, increased as sorption rate, *k*, increased in absolute value (Fig. 7-9). A linear model, with excellent fit of the data (Table 4), best described the relationship between *R* and *k*. At each concentration, *R* continued to increase as *k* increased despite sorption continuing indicating that the chemical sorption rate had little impact on adult mortality and that the greatest impact was due to loss of fumigant through physical sorption. For example, increasing *k* by one unit leads to an increase in *R* by 0.142, 0.731 and 1.930 at 0.5, 1 and 2 mg/L, respectively (Table 4).

Table.3.3. Model fit for the relationship between partition ratio of physical sorption (*K*) at three sulfuryl fluoride concentrations, 0.5, 1 and 2 mg/L with the mortality rate constant (*R*) of adults fumigated at 25°C and 60% r.h in presence of wheat (12% m.c) at 0.5 filling ratio. Turning points of the fitted line are at Maximum *R* and *K*.

SF mg/L	Orthogonal polynomial regression	significance	R^2	Maximum *R*	Maximum *K*
0.5	R= -0.02692+ -0.02084*K+0.00030670*K^2	$F_{2,21}$=499; P<.0001	0.97	-0.381	33.970
1	R= 0.00359+-0.22799*K+0.01334*K^2	$F_{2,21}$=855; P<.0001	0.99	-0.972	8.545
2	R= -0.01067+-0.68295*K+0.05554*K^2	$F_{2,21}$=292; P<.0001	0.94	-3.867	6.148

Table.3.4. Model fit for the relationship between constant rate of reaction (*k*) at sulfuryl fluoride concentrations 0, 0.5, 1 and 2 mg/L on the mortality rate constant of eggs (*R*) fumigated at 25°C and 60% r.h in presence of wheat (12% m.c) at 0.5 filling ratio.

SF mg/L	Linear regression	significance	R^2
0.5	R= -0.00171+0.14278*k	$F_{1,22}$=247; P<.0001	0.98
1	R= -0.00359+0.73187*k	$F_{1,22}$=281; P<.0001	0.96
2	R= -0.00583+1.93060*k	$F_{1,22}$=243; P<.0001	0.95

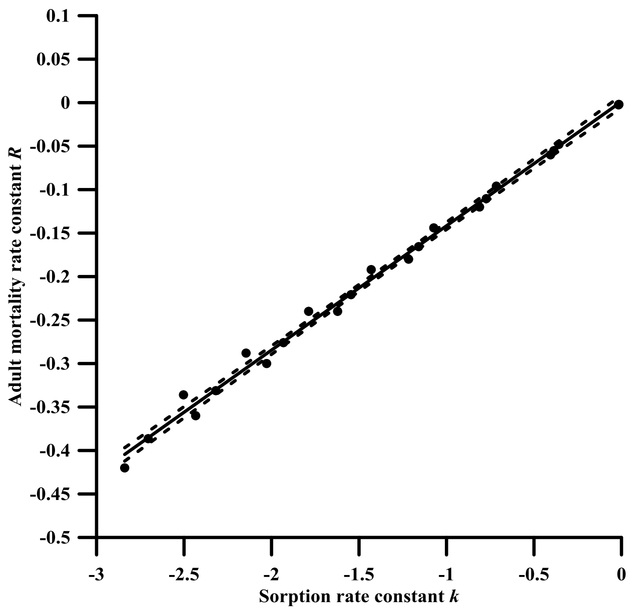


Fig.3.7. Relationship between the adult mortality rate constant (*R*) and the sorption rate constant (*k*) of bread wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 0.5 mg/L sulfuryl fluoride for 168 h, (---) upper and lower confidence limits at 95%.

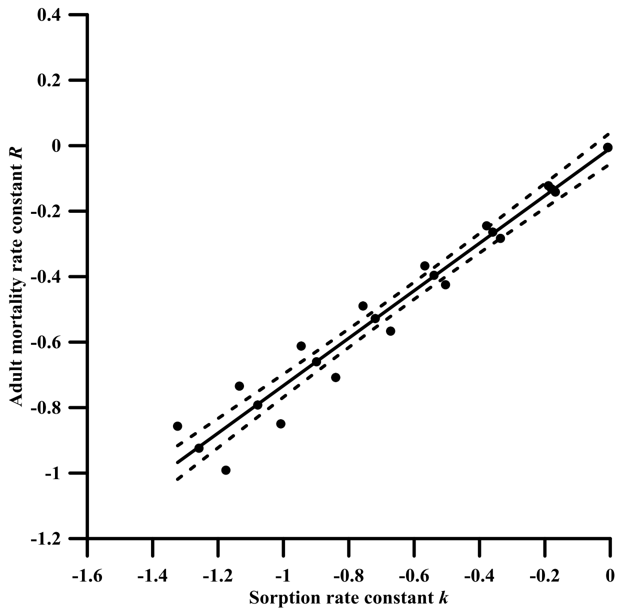


Fig.3.8. Relationship between the adult mortality rate constant (*R*) and the sorption rate constant (*k*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 1 mg/L sulfuryl fluoride for 168 h, (---) upper and lower confidence limits at 95%.

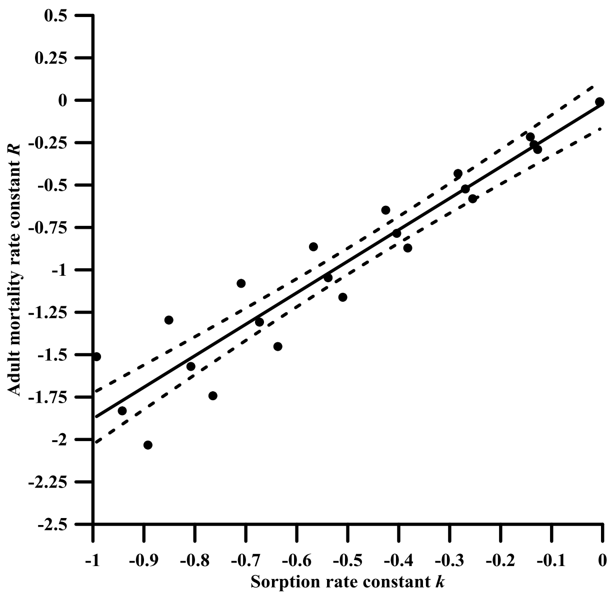


Fig.3.9. Relationship between the adult mortality rate (*R*) and the sorption rate (*k*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 2 mg/L sulfuryl fluoride for 168 h, (---) upper and lower confidence limits at 95%.

3.3.2. Egg survival

Number of adult progeny and therefore egg survival, decreased as SF concentration increased and over time. Egg survival occurred in all treatments, however. Even at the highest concentration tested, 2 mg/L for 160 h,. The rate constant of egg mortality *E* closely fitted the exponential model, explaining 98-99% variation in the data (Table 5). Egg mortality was, therefore, strongly affected by sorption of the fumigant by the commodity. In contrast, the numbers of adult progeny emerging from the grain in the control (non-fumigated) treatment increased rapidly with time (Fig 10). In contrast to the situation with adults, these curves predict that, at 2 mg/L, 99% of the fumigant would be lost before 99% control of eggs could be achieved.

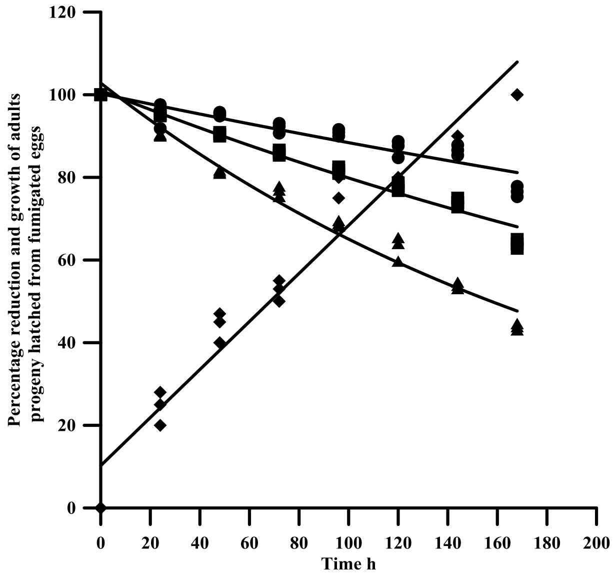


Fig.3.10. Percentage reduction of adult progeny hatched from eggs fumigated with sulfuryl fluoride (0.5 ●, 1 ■ and 2 ▲mg/L) and percentage growth of adult progeny hatched from eggs of control ♦ (not fumigated) of *R. dominica* in presence of wheat (12% moisture at 25°C and 60% rh).

The relationship between egg mortality rate *E* and the partition ratio of physical sorption, *K*, was curvilinear at the three concentrations tested (Figs. 11, 12, 13). This relationship was best explained by the quadratic model, which was an excellent fit for the data (R^2=0.95-0.99) (Table 6). The turning points of the fitted lines (Fig. 11, 12, 13) reveal an increase of *E* values with increasing applied SF concentration, but showed a decrease in *K* values. Thus, although egg mortality rate initially increased as concentration increased, physical sorption removed fumigant resulting in a decrease in the rate of mortality until it reached zero, and then the numbers of eggs surviving begin to increase. As expected, maximum or turning point *K* values obtained for adult and eggs were identical are they are derived from the same fumigation (Tables 3, 6). However, *E* values were lower than *R* values reflecting the higher sensitivity of adults to the fumigant (Tables 3, 6).

Table.3.5. Effect of sulfuryl fluoride fumigation (0.5, 1 and 2 mg/L) of wheat (12% moisture at 25°C and 60% rh) on survival of *R. dominica* egg determined by egg mortality rate (*E*), percentage kill per h, and time to half and complete mortality of the fumigant in half filled and full containers.

SF mg/L	Regression	Significance	R^2	% kill per (h) at 50% filling ratio	Time (h) to 50% kill at 50% filling ratio	Time (h) to kill 99% at 50% filling ratio	Time (h) to half kill at 100% filling ratio	Time (h) to kill 99% at 100% filling ratio
0.5	N_t=2.543$e^{-0.001t}$	$F_{2,19}$=223.92; P<.0001	0.99	0.099	693.00	4605.17	1386.00	9210.34
1	N_t=2.392$e^{-0.002t}$	$F_{2,19}$=343.18; P<.0001	0.98	0.209	330.01	2192.93	660.00	4386.27
2	N_t=2.3475$e^{-0.004t}$	$F_{2,19}$=151.20; P<.0001	0.99	0.439	157.50	1047.02	315.00	2093.25

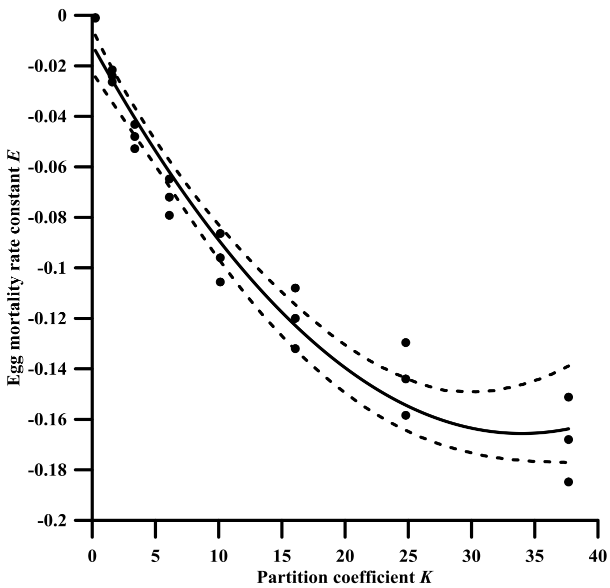


Fig.3.11. Relationship between the egg mortality rate constant (*E*) of *R. dominica*, and the partition coefficient of physical sorption (*K*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 0.5 mg/L sulfuryl fluoride for 168 h versus (---) upper and lower confidence limits at 95%.

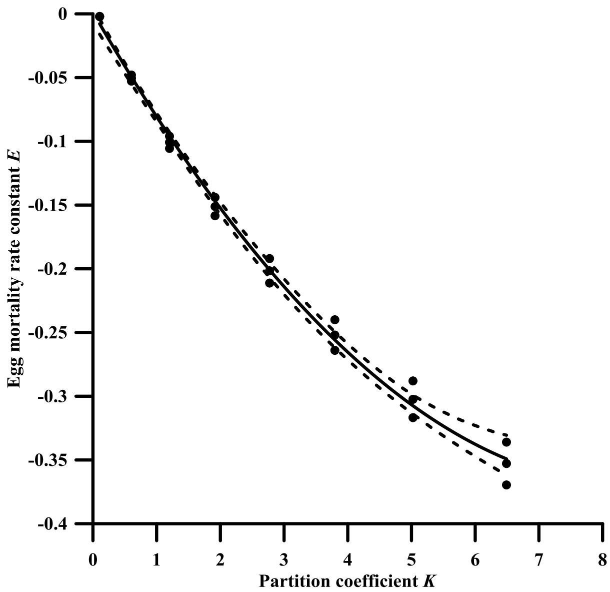


Fig. 3.12. Relationship between the egg mortality rate constant (*E*) of *R. dominica*, and the partition coefficient of physical sorption (*K*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 1 mg/L sulfuryl fluoride for 168 h versus egg mortality rate constant (*E*) of *R. dominica*, (---) upper and lower confidence limits at 95%.

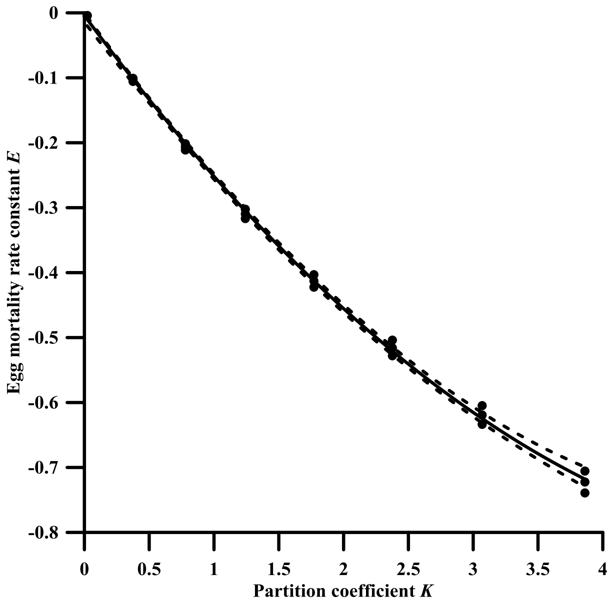


Fig.3.13. Relationship between the egg mortality rate constant (*E*) of *R. dominica*, and the partition coefficient of physical sorption (*K*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 2 mg/L sulfuryl fluoride for 168 h versus egg mortality rate constant (*E*) of *R. dominica,* (---) upper and lower confidence limits at 95%.

The relationship between sorption rate *k* and egg mortality rate *E* was linear at each concentration tested (Figs 14, 15, 16; Table 7). For example, at 0.5 mg/L SF (Fig. 14), when SF was 0.5 mg/L, the mortality increased to about 0.061 with an increase in *k* of one unit, and when SF was 2 mg/L, the mortality increased by about 0.76 with each increase in *k* of one unit.

The egg mortality rate constant *E* was much lower than the adult mortality rate *R* at all concentrations tested, demonstrating that eggs were more tolerant than adults to SF (Tables 2 and 5). Thus, the time required to control adults was much shorter than needed to control eggs. For example, at 2 mg/L, *R* was 0.01 per h, whereas *E* was 0.004 per h. The adult mortality rate constant, *R*, increased significantly with increasing fumigant concentration (Table 2) and time to 50 and 99% kill decreased with increasing SF concentration. In addition, increase in *E* with *k* was much slower than the increase in *R* with *k* (Table 7), and *R* continued to increase under the impact of *k*. However, both *R* and *E* increased to a maximum at which point they began to decrease due the impact of *K*.

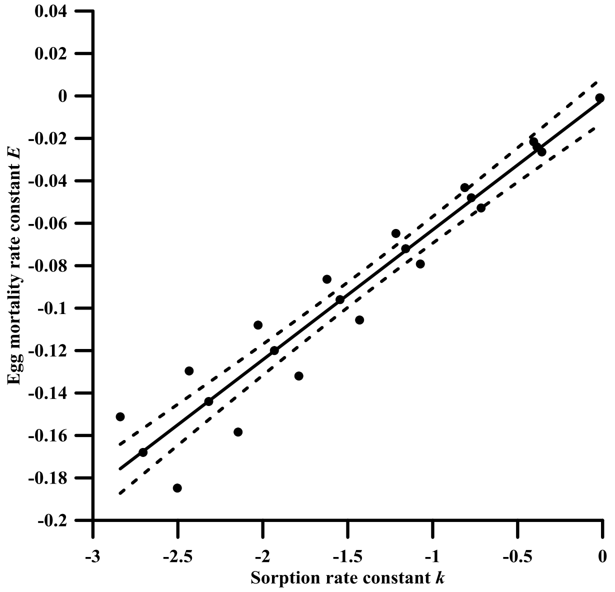


Fig.3.14. Relationship between the egg mortality rate constant (*E*) and the sorption rate constant (*k*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 0.5 mg/L sulfuryl fluoride for 168 h, (---) upper and lower confidence limits at 95%.

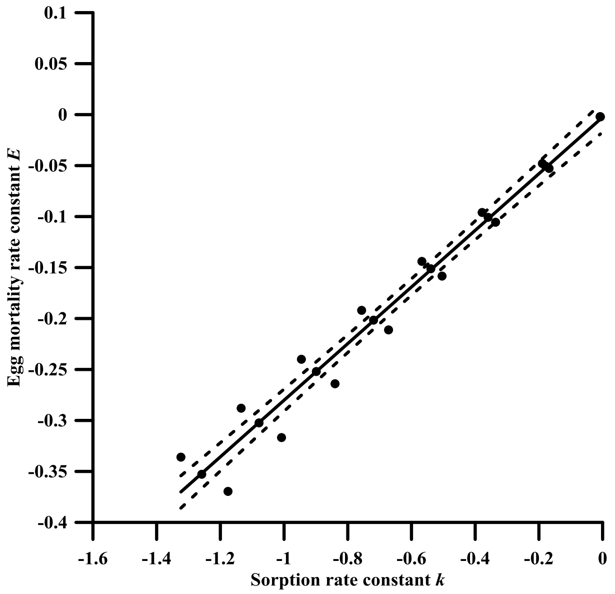


Fig.3.15. Relationship between the egg mortality rate constant (*E*) and the sorption rate constant (*k*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 1 mg/L sulfuryl fluoride for 168 h, (---) upper and lower confidence limits at 95%.

Table.3.6. Model fit for the relationship between partition coefficient of physical sorption (*K*) at three SF concentrations, 0.5, 1 and 2 mg/L on the response of egg mortality rate constant (*E*) for egg fumigated at 25°C and 60% r.h in presence of wheat (12% m.c) at 50% filling ratio and adult insects. Turning points of the fitting line (maximum *E* and *K*).

SF mg/L	Orthogonal polynomial regression	significance	R^2	Maximum *E*	Maximum *K*
0.5	E= -0.01171+-0.00906*K+0.00013335*K^2	$F_{2,21}$=239.29; *P*<.0001	0.95	-0.167	33.971
1	E= 0.00132+-0.08705*K+0.00509*K^2	$F_{2,21}$=1664; *P*<.0001	0.99	-0.371	8.551
2	E= -0.00421+-0.26942*K+0.022191*K^2	$F_{2,21}$=6688; *P*<.0001	0.99	-0.832	6.148

Table.3.7. Model fit for the relation between sorption rate constant (*k*) for SF concentrations, 0, 0.5, 1 and 2 mg/L on the egg mortality rate constant (*E*) of eggs fumigated at 25°C and 60% r.h in presence of wheat (12% m.c) at 50% filling ratio and adult insects and adult progeny population growth rate constant from not fumigated egg (*Eg*).

SF mg/L	Linear regression	significance	R^2
0.5	E= -0.00191+0.06121*k	$F_{1,22}$=360.64; *P*<.0001	0.94
1	E= -0.00247+0.27768*k	$F_{1,22}$=852.77; *P*<.0001	0.97
2	E= -0.00180+0.76267*k	$F_{1,22}$=4565; *P*<.0001	0.99
0	Eg= 0.011+2.307*t	$F_{1,22}$=815.33; *P*<.0001	0.97

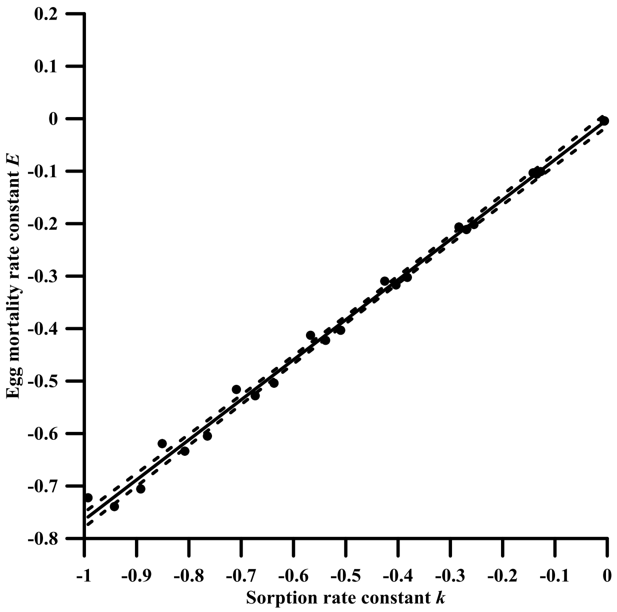


Fig.3.16. Relationship between the egg mortality rate constant (*E*) and the sorption rate constant (*k*) of wheat (12% mc, 25°C, 60% rh with adult and egg) fumigated with 2 mg/L sulfuryl fluoride for 168 h, (---) upper and lower confidence limits at 95%.

3.4. Discussion

The aim of this study was to determine the impact of sorption on the efficacy of sulfuryl fluoride against the lesser grain borer, a major pest of stored grain. Sorption was characterised using the partition ratio *K* and the sorption rate constant *k* of the chemical reaction between SF and grain. These factors were compared with the mortality rate constants for egg *E* and adult *R* life stages.

The results of these experiments indicate that sorption into wheat led to a decrease in the efficacy of SF against eggs and adults of *R. dominica.* SF was sorbed by wheat grains in an exponential manner and the data closely fitted first order exponential decay equations describing the relationship between fumigant sorption and exposure time as discussed in detail in Chapter 2. There was a rapid initial (first 24 h) decline in headspace concentration of SF (Fig. 1), which is typical of fumigant sorption behaviour (Hilton and Banks, 1997a; 1997b; Banks, 1990; Daglish and Pavic, 2008). The sorption rate constant, *k*, was independent of the applied concentration, consistent with results described in Chapter 2.

The relationship between concentration and adult and egg mortality rates, *E* and *R*, respectively, were described by first order equations (Tables 2 and 5) indicating the potential impact of sorption on the toxicity of SF during commodity fumigation. It is well known that increasing fumigant concentration (Baltaci et al., 2009; Aung et al., 2001; Daglish et al., 2002) increases mortality in the egg and other life stages. However, a decline in the efficacy of the applied concentration against eggs and adults over time due to sorption was demonstrated in this study (Tables 1, 2 and 5).

Sorption caused a decline in the effective SF concentration during the fumigations, resulting in an increase in the estimated time required for control of *R. dominica*. For example, the time estimated for 99% control of eggs at 1.0 filling ratio fumigated with 2 mg/L SF at 25°C, was ~87 d. However, under these conditions, 99% of the fumigant would be sorbed after only ~20 d making it impossible to obtain complete kill. The results of this study indicate the effect of sorption on the exposure time, that is, a longer time of exposure combined with a higher concentration would be required for complete control of the insect eggs.

Both the egg and adult mortality rates *E* and *R* were strongly influenced by the partition ratio of physical sorption *K* (Figs 4, 5, 6, 11, 12 and 13). *K* had an inverse relationship with *E* and this relationship was quadratic. An increase in *E* is expected to occur due to the toxic effect of the gas over time (Figs 11, 12 and 13). However, the continued physical sorption with time resulted in a reversal of the toxic effect and after reaching a turning point, *E* began to decrease resulting in an increase in survival of insects when the fumigant was sorbed completely. Dumas (1980) reported that physical sorption of phosphine increased with increasing exposure time, and these effects were very clear in the results of the present study. Time had a strong impact on the toxicity of SF to eggs and adults, as sorption resulted in a decrease in the gas concentration to below the toxic level.

Unlike the relationship between *K* and both *R* and *E*, the relationship between the rate of sorption of the fumigant by the commodity *k* and the mortality rate for eggs, *E*, was linear (Figs 7, 8, 9, 14, 15 and 16). The reaction of the gas with the commodity is a one directional, irreversible chemical reaction, but the rate of that reaction did not impact on *E* under the conditions of this study. The linear relationship indicated an increase in the mortality rate, even though the reaction rate for the fumigant and the commodity increased (Table 7). It appears that although rapid physical sorption occurred in the first 24 h (Fig 1), much of this SF was still biologically active as it had not undergone chemical reaction. The latter occurred gradually with time. These results also predict that the relationship between the sorption rate of the fumigant by the commodity, *k*, and the mortality rate for eggs *E*, would become nonlinear as continuing chemical sorption with time decreases the available gas then *E* decreases and a quadratic directional relationship with *k* follows. That is, the chemical reaction between the gas and the commodity would continue until all the gas is sorbed completely, and the relationship would become quadratic. Further experiments are required to assess to what extent the relationship will continue to be linear, and how sorption affects the relationship between the reaction rate and the toxicity of the fumigant.

These results demonstrate that sorption is a key process that should be taken into account when developing fumigation protocols for SF. This is particularly important as under-dosing due to loss of fumigant is an important factor in the development of resistance to fumigants in insect pests of stored products (Opit et al. 2012). In addition, when developing higher rates to overcome resistance, extending exposure times with assumed sufficient concentrations without considering sorption may not be effective.

3.5. Conclusion

These experiments demonstrate that sorption reduces the efficacy of SF against eggs and adults of *R. dominica* by removing biologically active fumigant from the system. The major effect of sorption on toxicity occurs during physical sorption phase, and the relationship between mortality rate constants for both eggs and adults with physical sorption is quadratic. The sorption rate constant of the fumigant by the commodity (k), has a linear relationship with both egg and adult mortality rate constants.

Extending exposure time without ensuring adequate dosage or increasing dose with an inadequate exposure time, may not be sufficient to control infestations due to the phenomenon of sorption of the fumigant by the commodity. Therefore, sorption should be considered as a fundamental factor affecting the fumigation process.

CHAPTER FOUR

The effect of varying filling ratio on the sorption of sulfuryl fluoride by wheat

ABSTRACT

Filling ratio is an important aspect in grain storage as the storage capacity has an impact on sorption of the fumigant that may affect the toxicity of the fumigant and residues. The level to which the storage facility is filled with wheat (filling ratio) is an important aspect of fumigation. The degree to which the fumigant is sorbed by the commodity is affected by the filling ratio. Durum wheat of 12% moisture content was added to a fumigation vessel at 0.95, 0.75 and 0.50 filling ratios and fumigated with 1 mg/L of SF for 168 h at 25°C. The highest sorption rate of SF was at 0.95 and the time to sorb the fumigant decreased with increasing filling ratio. However, the lowest sorption rate was at the lowest filling ratio, and time to loss of the fumigant was longer than at highest filling ratios. Physical sorption and accumulation of SF on grain was the same rate at different densities of grain and increased with increasing filling ratio. However, chemical sorption increased with increasing the density of the grain and filling ratio. The relationship between the physical sorption and time was quadratic, and the relationship between chemical sorption and time was linear. The relationship between the physical sorption and chemical sorption was quadratic as the chemically sorbed amounts depend on physically available amount of the gas. Storing grain in a full system is economically beneficial; however, sorption, residues and infestation need to be considered. When the appropriate level of grain in a storage system (a 0.50 filling ratio is recommended) is used, an accurate and effective fumigation procedure against insects with minimal sorbed gas by the grain will be achieved.

4.1. Introduction

Applying fumigants to prevent or control infestation in presence of a commodity results in an initial increase in the concentration of the fumigant in the storage vessel if the volume occupied by the commodity is not taken into account. However, depending on the properties of the commodity, sorption of the fumigant by the commodity may act to decrease its concentration. Sorption is greater with high filling ratios compared to low filling ratios. Hilton and Banks (1997a) showed that the sorption behaviour of sultanas (dried yellow grapes) and wheat (Banks, 1990) was the same. That is, the rate of sorption was represented as a log of the fumigant concentration over the initial value and was dependent on the filling ratio of the commodity, and independent of the initial fumigant concentration. Different filling ratios show different sorption rates, and the rate of sorption increased with increasing filling ratio. Daglish and Pavic (2008) fumigated wheat grains with different doses of phosphine at different filling ratios. Phosphine concentration decreased exponentially in all experimental flasks, and percentage phosphine sorbed daily correlated with the filling ratio. Damcevski and Annis (2006) studied the efficacy of ethyl formate against *Sitophilus oryzae* (L.) at different wheat filling ratios. The target insect was exposed to the fumigant for 24 h at filling ratios ranging from 0% to 60%. The predicted concentration for 99% mortality in an empty flask was 11.2 mg/L. However, 81.2 mg/L was required for controlling the same percentage in flasks that were filled to 60% capacity with wheat grains. In addition, Lindgren and Vincent (1951) used different types of fumigants to assess the mortality of *Tribolium confusum* and *Sitophilus granarius* adults in the presence and absence of wheat. Fumigation without commodity gave mortality higher than with the commodity.

The purpose of this study was to show the effect of different filling ratios and densities of wheat, on the sorption behaviour of SF. Also, to understand the impact of filling ratio on both physical and chemical sorption behaviour of SF.

4.2. Materials and methods

4.2.1. Experimental design

In this experiment, nine flasks were filled with durum wheat (*Triticum durum)* of 12% moisture content at different filling ratios. This experiment was replicated three times. Three flasks were filled to 0.95 of the total volume of the flask, three flasks were filled to 0.75 of the total volume of the flask and three flasks were filled to 0.50 of the total volume of the flask. All the experimental flasks were injected with 1 mg/L SF for 168 h.

Three standard flasks (control flasks) were prepared for making calibration curves (no wheat) by injecting one flask with the desired and applied concentration, 1 mg/L, one flask had a concentration lower than the desired concentration and one flask had a concentration higher than the desired concentration.

4.2.2. Commodity

Details are in chapter 2 section 2.2.2

4.2.2.1 Moisture content

Details in chapter 2 section 2.2.2

4.2.3. Determining bulk, true density and commodity volume V_g

Details are in chapter 2 section 2.2.7 and Table2.6.

4.2.4. Fumigation

Details are in chapter 2 section 2.2.4

4.2.4.8. Statistical analysis

Data was analysed using SAS software, and for graphing the Grapher program (Golden Software) was used. The decay of gas concentrations and the growth of the partition coefficient of physical sorption with time were analysed using first order equations

$$C_t=C_0e^{-k*t} \quad (1)$$

$$K_t=K_0e^{g*t} \quad (2)$$

where C_t was the gas concentration C at time t, C_0 was gas concentration C at time zero ($t=t_0=0$), $-k$ was the constant for rate of reaction, K_t was the partition coefficient of physical sorption as units K at time t, K_0 was the partition coefficient of physical sorption as units K at time zero ($t=t_0=0$), g was the rate of increase of K. t was time (Hilton and Banks, 1997a; Daglish and Pavic, 2008).

Hilton and Banks (1997a) also used another set of models for calculating k_f, the constant rate of reaction for a full container, and K as a partition ratio of physical sorption measured by units.

$$k_f=k/f, \quad (3)$$

where k was the observed rate of reaction for a particular filling ratio, f .

$$K=(C_a-C_i)*V_g / C_i*V_f \quad (4)$$

Where C_a was the applied concentration, C_i was the predicted concentration through the equation line or curve, V_g was the gas volume and V_f was the volume occupied by the commodity.

The relationships between the partition coefficient K and the constant rate of reaction between SF and commodity k was investigated by a polynomial quadratic regression.

$$k= B_0 + B_1(K) + B_2(K)^2 \quad (5)$$

Where B_0 was the intercept, B_1 was the linear coefficient and B_2 was the quadratic coefficient.

The turning point of the standard polynomial quadratic regression line was calculated using the following equation:

$$K= - B_1 / 2 B_2 \quad (6)$$

The turning point of the fitted line is the point at which it turns back toward zero. This point was the point of maximum partition ratio of physical sorption K that gives the maximum rate of reaction constant k. After calculating K using the equation 6, equation 5 was used to calculate the maximum k by substituting the obtained values of K into each equation.

A linear regression formula was used to describe the relationship between k and time, t, where

$$k = B_0 + B_1(t) \tag{7}$$

In addition, the percentage gas loss per h was calculated using the following equation:

$$\text{Percentage} = 100\ (1-e^{i}) \tag{8}$$

Where i was k.

Time to half (equation 9) and 99% (equation 10) loss of the gas was calculated as follows:

$$1/2L_0 = L_0\, e^{-i*t}/L_0,\ 1/2 = e^{-i*t},\ t = \ln(1/2)/-i. \tag{9}$$

$$0.01L_0 = L_0\, e^{-i*t}/L_0,\ 0.01 = e^{-i*t},\ t = \ln(0.01)/-i. \tag{10}$$

Where, L_0 was C_0, i was k and ln was the natural logarithm.

Time to double and ten times physical sorption was calculated as follows:

$$2L_0 = L_0\, e^{i*t}/L_0,\ 2 = e^{i*t},\ t = \ln(2)/i. \tag{11}$$

$$10L_0 = L_0^{\ i*t}/L_0,\ 10 = e^{i*t},\ t = \ln(10)/i. \tag{12}$$

Where, L_0 was K_0 (from equation 2), i was g (from equation 2) and ln was the natural logarithm.

The tota1 number of durum grains was calculated as follows:

Total number of grain= total weight of sample*1000 grain / weight of 1000 grain. (13)

Where k and K for bulk density were taken directly from equations 1 and 4 at each time interval, and for individual were calculated according to the following equations:

k for grain density of 1.436 g/mL = k (for the bulk) from equation 1/ total number of grains.(14)

K for grain density of 1.436 g/mL = K (for the bulk) from equation 4/ total number of grains.(15)

Where, bulk is the amount of the commodity in the fumigation system.

Both of bulk density and grain density were taken from table 6 in chapter 2 for durum wheat at 12% moisture.

4.3. Results

Rate of sorption of SF increased as filling ratio increased (Table 1, Fig 1). The high goodness of fit represented by R^2 and F values from the first order exponential equations indicate appropriate models for fitting the raw data. Differences in sorption rates at the different filling ratios were recorded. For example, the sorption rate constant k of SF by durum wheat increased from 0.00611 per h at 0.50 filling ratio to 0.0121 per h at 0.95 filling ratio with difference of 0.00938 per h (Table 1). The difference in sorption rate k between 0.75 and 0.50 filling ratios was 0.00327 showing sorption rates higher at 0.75 filling ratio than at 0.50 filling ratio. Time to sorb the fumigant decreased with increasing filling ratio. Thus, times to half and 99% sorption were 113 and 757 h, respectively, at 0.50 filling ratio and only 57.3 and 380 h, respectively, at 0.95 filling ratio. The filling ratio of grain in the storage

Table.4.1. Exponential decay of SF with time due to sorption at different filling ratios of durum wheat grain, 12% moisture content at 25°C. Percentage loss per h, time to half and 99% loss of the applied fumigant.

Filling ratio	Regression	Significance	R^2	% loss per h	Time h to half loss	Time h to loss 99%
0.95	$C_t=0.994e^{-0.0121*t}$	$F_{2,19}$=196.41; P<.0001	0.90	1.202	57.272	380.578
0.75	$C_t=0.5218e^{-0.00938*t}$	$F_{2,19}$= 2101.1; P<.0001	0.95	0.938	73.880	490.938
0.50	$C_t=0.4711e^{-0.00611*t}$	$F_{2,19}$= 3775.2; P<.0001	0.97	0.609	113.420	756.682

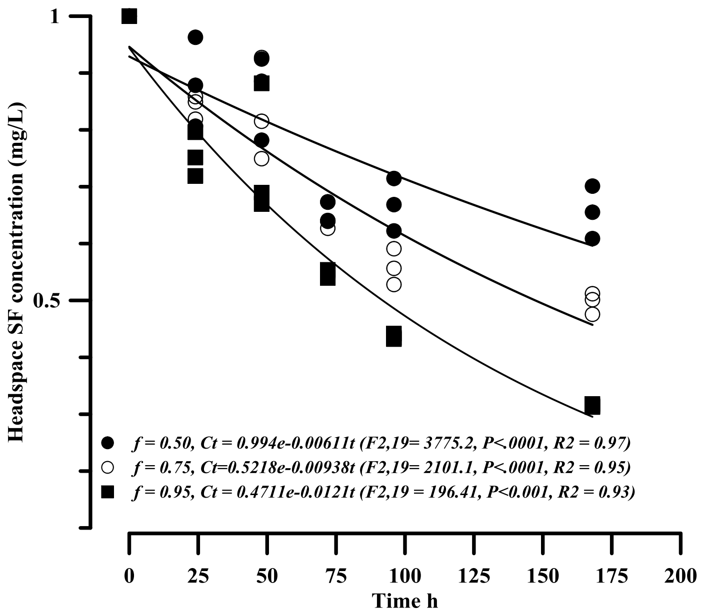


Fig.4.1. Exponential decay curves of sulfuryl fluoride 1 mg/L sorbed by durum wheat 12% moisture content at 25°C and 0.50 ●, 0.75 ○ and 0.95 ■ filling ratios.

system also showed significant impacts on both physical and chemical sorption into the bulk and the grain. Unlike *k*, *K* decreased as filling ratio increased (Table 2). At 0.95, 0.75 and 0.50 filling ratios, *K* was 0.863, 1.337 and 2.045, respectively, and for the grain density, at 0.95, 0.75 and 0.50 filling ratios, *K* was 0.0187, 0.0388 and 0.0887, respectively (Table 2).

As the fumigant was sorbed exponentially by durum grain, changes in the partition ratio, *K*, indicated that fumigant was lost from the system by accumulation in the grain. The rate of accumulation was analysed and found to be exponential at all filling ratios and densities (Figs 2 - 7). The accumulation rate constant *g* was the same for the different densities at each filling ratio (Table 3). For example, at 0.95 filling ratio, *g* was 0.0164 per h for the two densities. In addition, the times taken to accumulate two and 10-fold of the physically sorbed amounts at each filling ratio were the same despite variation in densities (Table 3). The accumulation rate constant *g* (*K* as units*)* was 0.0164, 0.0108 and 0.00864 per h at 0.95, 0.75 and 0.50 filling ratios, respectively.

In contrast, the relationship between time and *k* (sorption rate constant) was linear at all filling ratios and densities (Figs 8-13). My results showed that *k* increased linearly with time. For example, based on bulk density, *k* values at 0.95, 0.75 and 0.50 filling ratios increased by 0.0142, 0.0096 and 0.0054, respectively, for each hour of time (Table 4). Similar trends were evident with the other density measurements. Unlike *K* (Table 3), however, the increases in *k* values with time (Table 4) varied at the different densities for each filling ratio. For example, *k* values at 0.95 filling ratio were 0.00016 and 0.00030 at bulk density and grain density. Furthermore, *k* responded in a quadratic manner to the impact of *K* despite the differences among filling ratios and densities in the storage systems (Figs 14-19). Maximum rate of reaction *k* increased as the level of grain in the storage system and the density increased (Table 5).

Table.4.2. Sorption rate constant (*k*) of SF and partition coefficient of physically sorbed amounts (*K*, as units) by durum wheat, 12% moisture content at 25°C, for different storage densities (bulk and grain) at different filling ratios.

Filling ratio	Density g/ml			
	Bulk		grain	
	k	*K*	*k*	*K*
0.95	-0.0121	0.863	-0.00031	0.0187
0.75	-0.00938	1.337	-0.00027	0.0388
0.50	-0.00611	2.045	-0.00023	0.0887

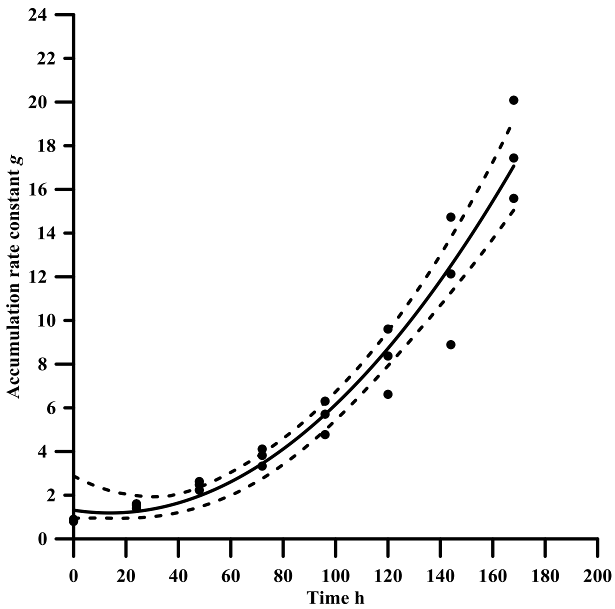


Fig.4.2. Accumulation rate constant *g* of physically sorbed sulfuryl fluoride (*K*, as units) by durum wheat, bulk density of 0.843 g/ml at 12% moisture content, 25°C and 0.95 filling ratio.

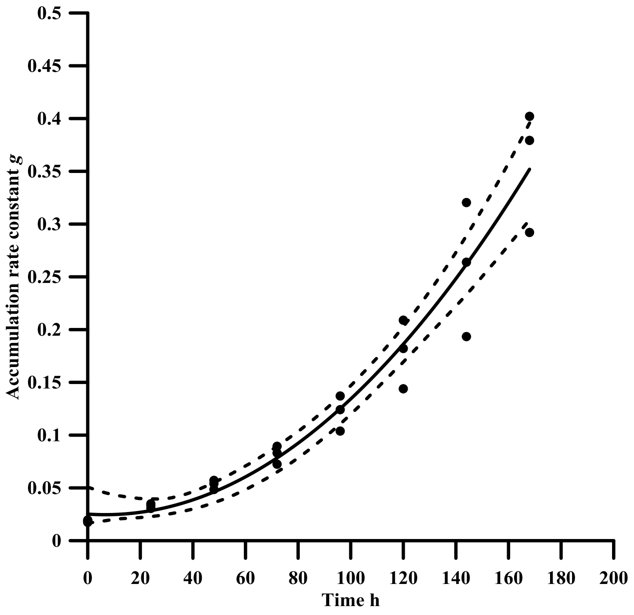


Fig.4.3. Accumulation rate constant *g* of physically sorbed sulfuryl fluoride (*K*, as units) by durum wheat, grain density of 1.436 g/mL at 12% moisture content, 25°C and 0.95 filling ratio.

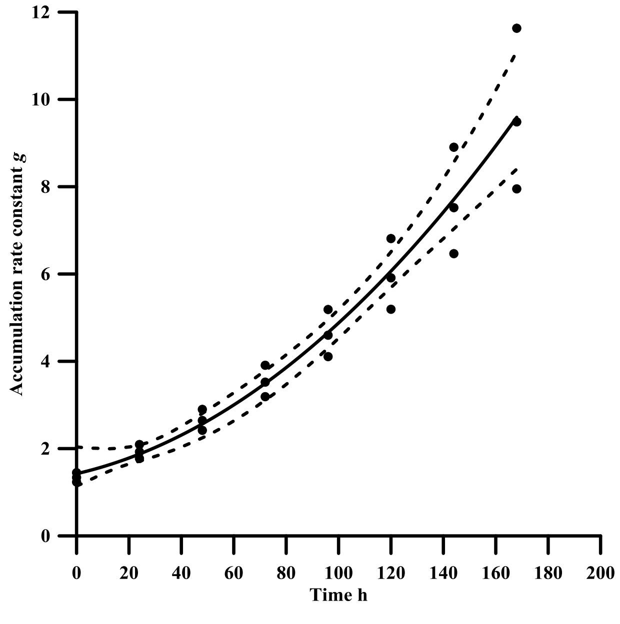


Fig.4.4. Accumulation rate constant *g* of physically sorbed sulfuryl fluoride (*K*, as units) by durum wheat, bulk density of 0.843 g/ml at 12% moisture content, 25°C and 0.75 filling ratio.

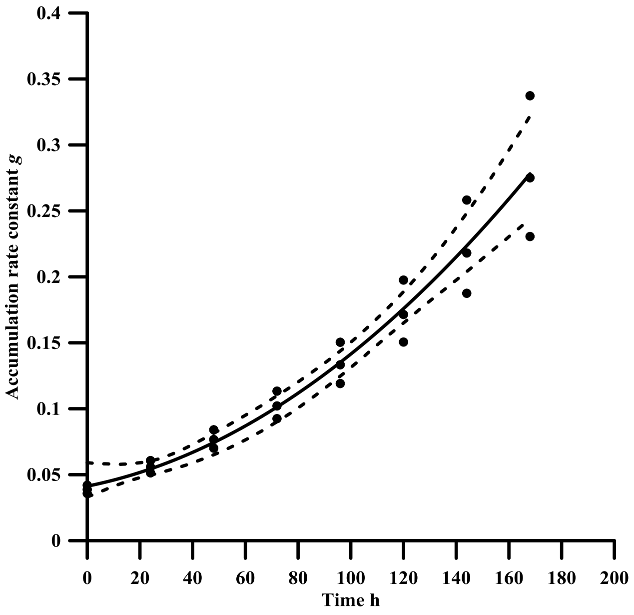


Fig.4.5. Accumulation rate constant *g* of physically sorbed sulfuryl fluoride (*K*, as units) by durum wheat grain density of 1.436 g/ml at 12% moisture content, 25°C and 0.75 filling ratio.

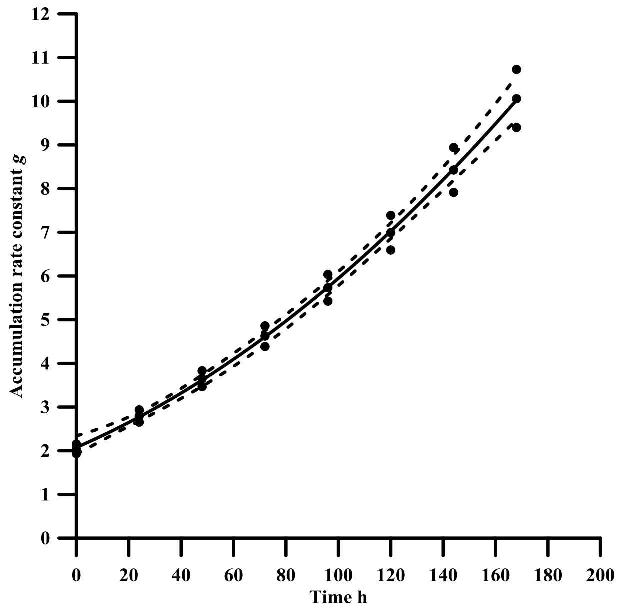


Fig.4.6. Accumulation rate constant *g* of physically sorbed sulfuryl fluoride (*K*, as units) by durum wheat bulk density of 0.843 g/ml at 12% moisture content, 25°C and 0.50 filling ratio.

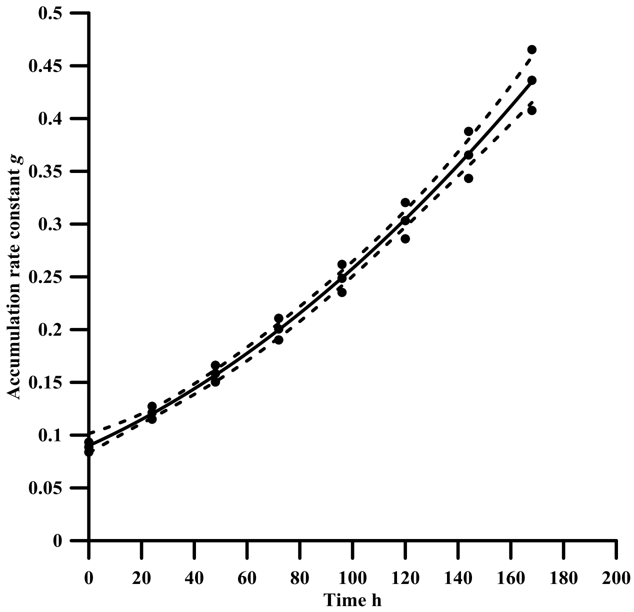


Fig.4.7. Accumulation rate constant *g* of physically sorbed sulfuryl fluoride (*K*, as units) by durum wheat grain density of 1.436 g/ml at 12% moisture content, 25°C and 0.50 filling ratio.

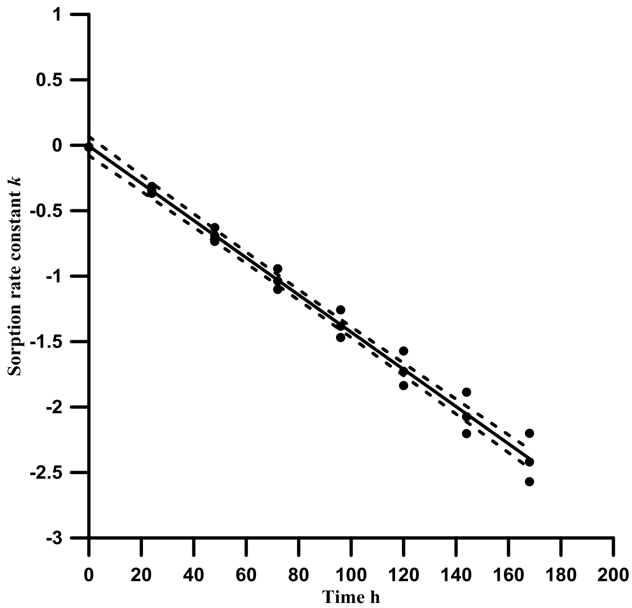


Fig.4.8. Linear relationship between sorption rate constant (*k*) of sulfuryl fluoride by durum wheat bulk density of 0.843 g/ml at 12% moisture content, 25°C and 0.95 filling ratio.

Table.4.3. Exponential growth and accumulation rate constant *g* of partition ratio of physical sorption (*K* as units) with time at three different filling ratios of durum wheat, 12% moisture content at 25°C and the percentage accumulation per h, time to accumulate double and ten times K_0.

Filling ratio	Density g/ml	Model	significance	R^2	% accumulation per h	Time h to accumulate double K_0	Time h to accumulate ten times of K_0
0.95	Bulk	$K_t=1.1343e^{0.0164*t}$	$F_{2,21}$=239.29; P<.0001	0.95	1.640	42.25	140.36
	Grain	$K_t=0.0271e^{0.0164*t}$	$F_{2,21}$=239.29; P<.0001	0.94	1.640	42.25	140.36
0.75	Bulk	$K_t=1.5901e^{0.0108*t}$	$F_{2,21}$=239.29; P<.0001	0.93	1.080	64.16	213.14
	Grain	$K_t=0.0461e^{0.0108*t}$	$F_{2,21}$=239.29; P<.0001	0.93	1.080	64.16	213.14
0.50	Bulk	$K_t=2.4092e^{0.00864*t}$	$F_{2,21}$=239.29; P<.0001	0.98	0.864	80.20	266.43
	Grain	$K_t=0.1045e^{0.00864*t}$	$F_{2,21}$=239.29; P<.0001	0.97	0.864	80.20	266.43

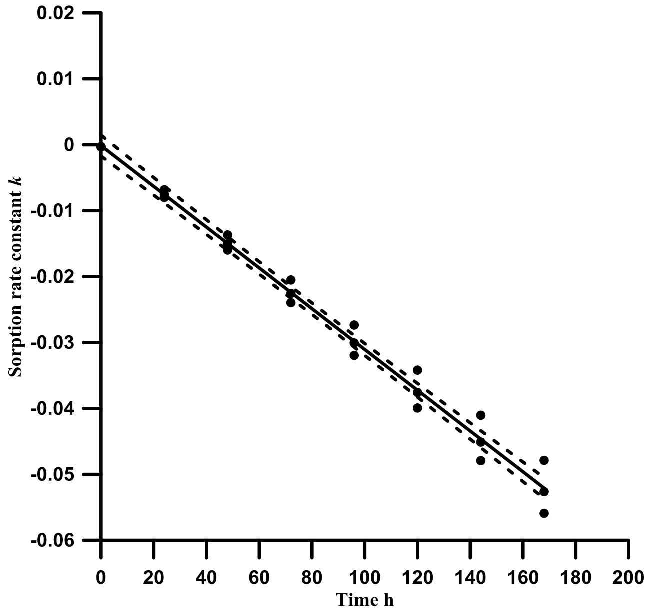


Fig.4.9. Linear relationship between sorption rate constant (*k*) of sulfuryl fluoride by durum wheat grain density of 1.436 g/ml at 12% moisture content, 25°C and 0.95 filling ratio.

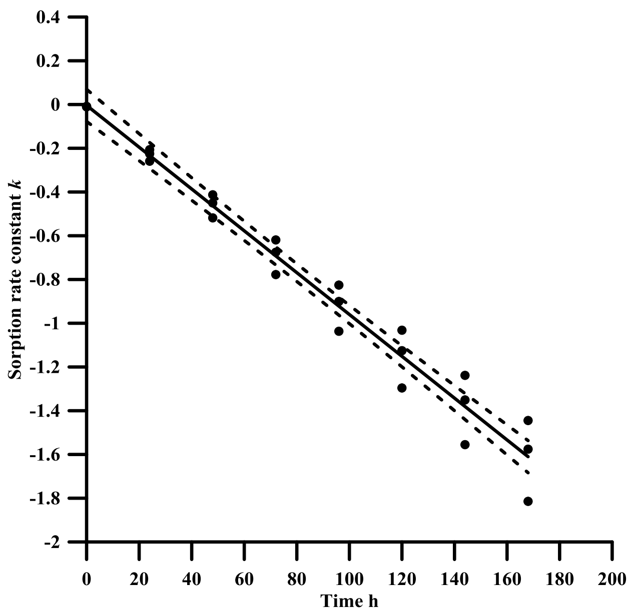


Fig.4.10. Linear relationship between sorption rate constant (*k*) of sulfuryl fluoride by durum wheat bulk density of 0.843 g/ml at 12% moisture content, 25°C and 0.75 filling ratio.

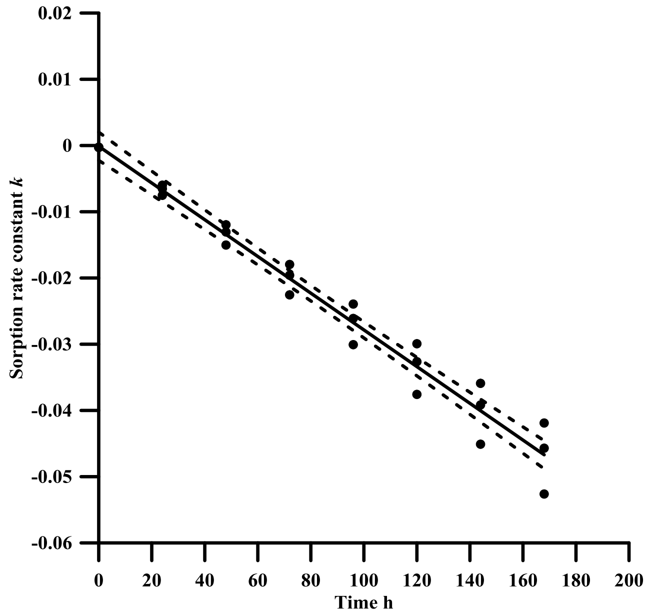


Fig.4.11. Linear relationship between sorption rate constant (*k*) of sulfuryl fluoride by durum wheat grain density of 1.436 g/ml at 12% moisture content, 25°C and 0.75 filling ratio.

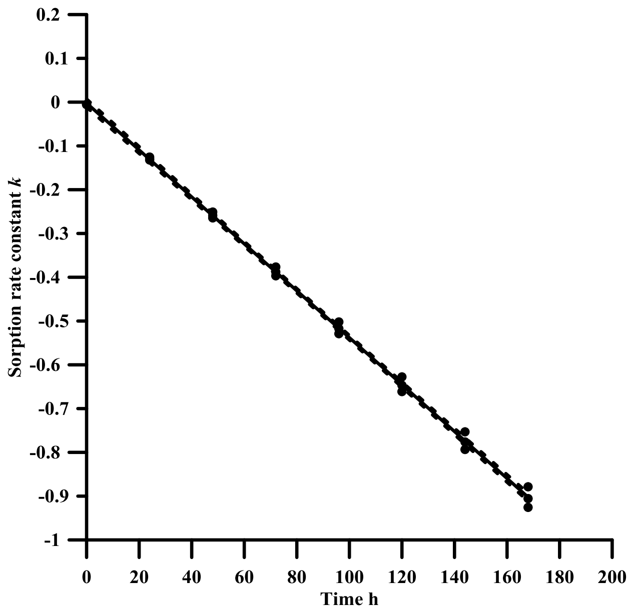


Fig.4.12. Linear relationship between sorption rate constant (*k*) of sulfuryl fluoride by durum wheat bulk density of 0.843 g/ml at 12% moisture content, 25°C and 0.50 filling ratio.

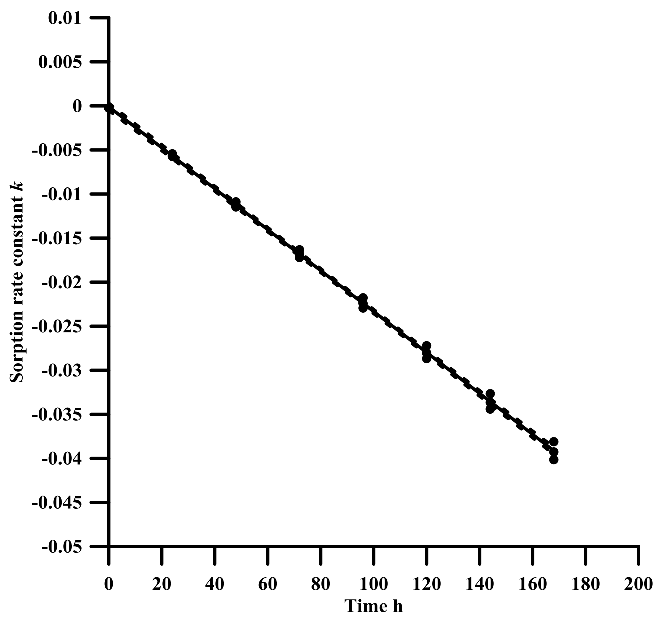


Fig.4.13. Linear relationship between sorption rate constant (*k*) of sulfuryl fluoride by durum wheat grain density of 1.436 g/ml at 12% moisture content, 25°C and 0.50 filling ratio.

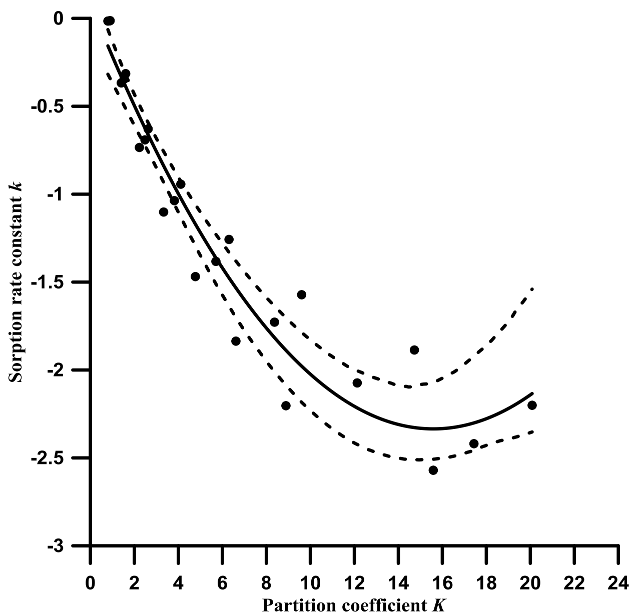


Fig.4.14. Quadratic response of sorption rate constant *k* to the impact of partition coefficient of physical sorption (*K*, as units) of sulfuryl fluoride sorbed by durum wheat bulk, density of 0.843 g/ml at12% moisture content at, 25°C and 0.95 filling ratio.

Table.4.4. Linear relationship between time and sorption rate constant (*k*) of SF by durum grain 12% moisture content at 25°C and three different filling ratios for bulk, individual and 50% less individual density.

Filling ratio	Density g/ml	Linear regression	significance	R^2
0.95	Bulk	*k* = -0.00594+-0.01422**t*	$F_{1,22}$=1632; *P*<.0001	0.98
	Grain	*k* = -0.00013+-0.00030**t*	$F_{1,22}$=1530; *P*<.0001	0.96
0.75	Bulk	*k* = -0.004+-0.0096**t*	$F_{1,22}$=726.66; *P*<.0001	0.95
	Grain	*k* = -0.00012+-0.00028**t*	$F_{1,22}$=830.21; *P*<.0001	0.97
0.50	Bulk	*k* = -0.00224+-0.0054**t*	$F_{1,22}$=1417; *P*<.0001	0.97
	Grain	*k* = -0.000098+-0.00024**t*	$F_{1,22}$=1310; *P*<.0001	0.96

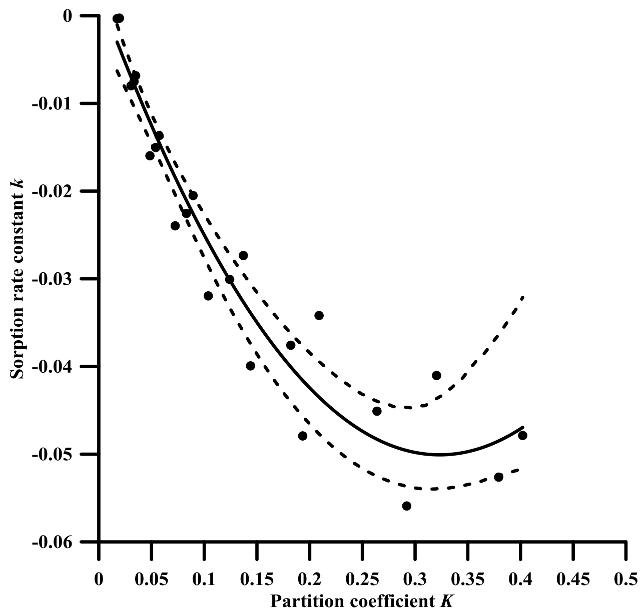


Fig.4.15. Quadratic response of sorption rate constant *k* to the impact of partition coefficient of physical sorption (*K*, as units) of sulfuryl fluoride sorbed by durum wheat grain, density of 1.436 g/ml at12% moisture cont

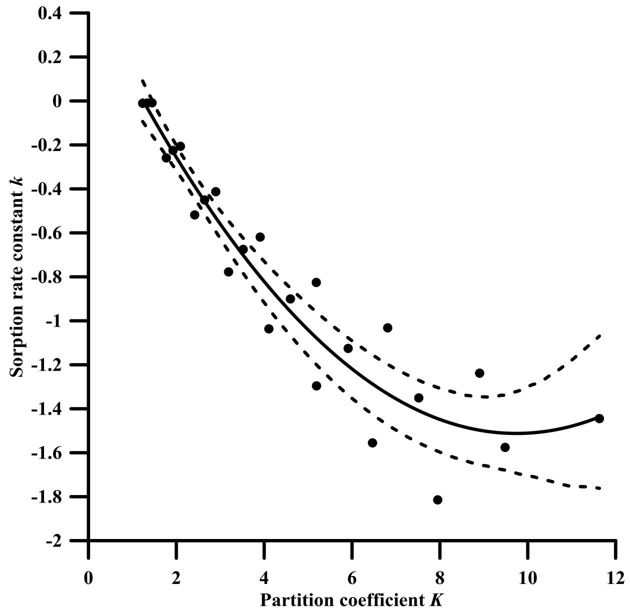


Fig.4.16. Quadratic response of sorption rate constant *k* to the impact of partition coefficient of physical sorption (*K*, as units) of sulfuryl fluoride sorbed by durum wheat bulk density of 0.843 g/ml at12% moisture content at, 25°C and 0.75 filling ratio.

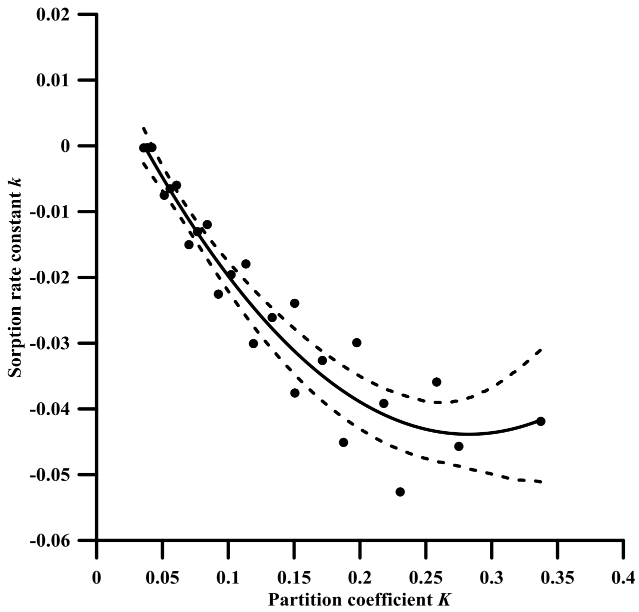


Fig.4.17. Quadratic response of sorption rate constant *k* to the impact of partition coefficient of physical sorption (*K*, as units) of sulfuryl fluoride sorbed by durum wheat grain density of 1.436 g/ml at12% moisture content at, 25°C and 0.75 filling ratio.

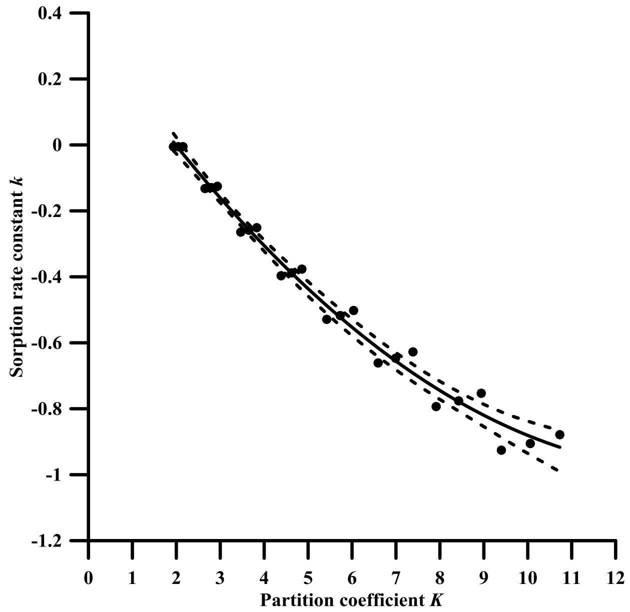


Fig.4.18. Quadratic response of sorption rate constant *k* to the impact of partition coefficient of physical sorption (*K*, as units) of sulfuryl fluoride sorbed by durum wheat bulk density of 0.843 g/ml at12% moisture content at 25°C and 0.50 filling ratio.

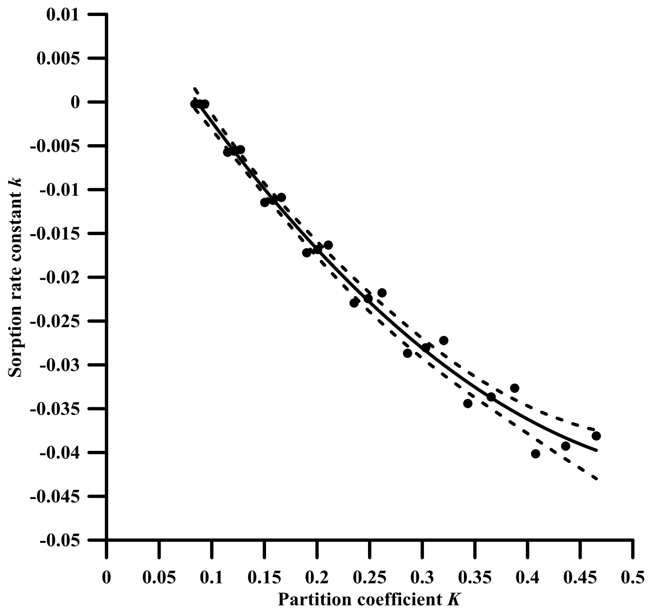


Fig.4.19. Quadratic response of sorption rate constant *k* to the impact of partition coefficient of physical sorption (*K*, as units) of sulfuryl fluoride sorbed by durum wheat grain density of 1.436 g/ml at12% moisture content at, 25°C and 0.50 filling ratio.

For bulk density of the commodity, maximum *k* values for 0.95, 0.75 and 0.50 filling ratio were -2.336, -1.509 and -0.973, respectively. Similar results were recorded for grain density. In the same way, at the highest filling ratio, maximum *k* for bulk density and grain density were -2.336 and -0.0501 respectively, and the same trends were noted with the other filling ratios. However, the maximum *K* values were independent of filling ratio (Table 2). For example, maximum *K* values for bulk density at 0.95, 0.75 and 0.50 were 15.598, 9.725 and 13.79, respectively (Table 5). The dependency of maximum *k* on filling ratio and density, the dependency of maximum *K* on density, and the independence of maximum *K* on filling ratio influenced the turning points of the fitted line toward zero. For instance, the turning point of *K* at 0.50 filling ratio and bulk density was 0.973 and 13.79, respectively (Table 5), but the turning point of 0.75 filling ratio and bulk was 1.509 and 9.725, respectively.

Table.4.5. Quadratic impact of partition coefficient of physical sorption (*K*) on sorption rate constant (*k)* of SF by Durum wheat 12% moisture content at 25°C and three different filling ratios and maximum values of both rates showing the turning points of (*k)* due to the impact of (*K*) at each density.

Filling ratio	Density g/ml	Orthogonal polynomial regression	Significance	R^2	Maximum *k*	Maximum *K*
0.95	Bulk	$k = 0.085+-0.3104* K+0.00995*K^2$	$F_{2,21}$=146.90; *P*<.0001	0.93	-2.336	15.598
	Grain	$k = 0.0025+-0.323*K+0.50306*K^2$	$F_{2,21}$=146.50; *P*<.0001	0.94	-0.0501	0.321
0.75	Bulk	$k = 0.477+-0.40844*K+0.021*K^2$	$F_{2,21}$=101.97; *P*<.0001	0.90	-1.509	9.725
	Grain	$k = 0.014+-0.41*K+0.723*K^2$	$F_{2,21}$=111.32; *P*<.0001	0.92	-0.044	0.284
0.50	Bulk	$k = 0.357+-0.193*K+0.007*K^2$	$F_{2,21}$=638.33; *P*<.0001	0.98	-0.973	13.79
	Grain	$k = 0.0155+-0.19334* K+0.1605*K^2$	$F_{2,21}$=601.57; *P*<.0001	0.97	-0.0427	0.602

4.4. Discussion

The results of this study indicated an increase in sorption and percentage loss of SF from the headspace per h with increase in filling ratio (Table 1). However, time to lose the fumigant from the storage system to and into commodity decreased with increase in the filling ratio (Table 1) as the increasing sorption rate decreased the time required to sorb half or all the fumigant by the commodity. Results in Table 1 showed that sorption rate constant, time to loss 50 and 99% of the fumigant at 0.50 filling ratio were 0.00611 per h, 113 h and 757 h, respectively. However, at 0.95 filling ratio, sorption rate constant, time to loss 50 and 99% were 0.0121 per h, 57 h and 380 h. Sorption rates, *k*, observed in this study were very close to those reported by Hilton and Banks, (1997a) for methyl bromide fumigation for 7 days and were for the full container 0.0106, 0.0112 and 0.0117 calculated from 0.25, 0.50 and 0.95 filling ratios, respectively. However, the sorption rates reported by Daglish and Pavic (2008) after fumigating bread wheat with 1 mg/L of phosphine were lower than the sorption rates for SF in the study. Hourly sorption rates for phosphine were 0.0006, 0.0008, 0.0017 and 0.0023 at 0.25, 0.50, 0.75 and 0.95 filling ratios.

The reaction constant between the fumigant and the grain, *k*, was dependent on the filling ratio., whereas, the partition ratio of physical sorption, *K*, was independent of the filling ratio (Table 2). However, both K and *k* were dependent on the density of the commodity at each filling ratio. Hilton and Banks (1997b) also reported that sorption rate constant increased as the filling ratio increased and that partition ratio was independent of filling ratio. My results showed that a first order, nonlinear equation fitted the data of the experiment very closely (R^2 = 0.90 – 0.97). The nonlinearity of fumigant sorption was also reported by Darby (2008) who indicated that the fumigant mass is transferred nonlinearly from the headspace into grains and the model does not discriminate between the reversible physically sorbed amount and the irreversible chemically sorbed amount. In addition, the first order nonlinear model was used by Daglish and Pavic, (2009) with a high goodness of fit (R^2) ranging from 0.9983 to 0.9998. My results indicate that a high ratio of grain volume to storage volume (that is, filling ratio) results in sorption of high amounts of fumigant whereas sorption was less with low filling ratios. For example, sorption rate at 0.95 filling ratio was 0.0121 per h, and at 0.50 filling ratio sorption rate was 0.00611 per h (Table 1). Therefore, adding large amounts of grains into a storage system increases sorption. When an insect infestation occurs in a full silo, control of the infestation is more difficult than would be the case if the silo was half-filled or less with grain as the concentration of fumigant available to act against target pests may be considerably reduced at the high filling ratio. In addition, the results of this study showed that the fumigant was sorbed by the commodity exponentially, the physically sorbed amounts were found to accumulate exponentially as well (Figs 2 - 7). The accumulation rate constant *g* of the fumigant in this study was reported before by Cofie-Agblor et al., (1998, 1995 and 1993) and Navarro (1997) for fumigation of wheat with CO_2. They noticed that the applied CO_2 accumulated exponentially similar to results of this study and the first order model with different formula from the one used here showed the best fit. However, the rate of accumulation of CO_2 was not close to the accumulation rate of SF due the difference in chemical and physical properties of the two gases in addition to the difference in the fumigation conditions of the two experiments. The accumulation rates increased with increasing filling ratio (Table 3). Results indicated an increase in the accumulation rate constant *g* with increasing filling ratio. At 0.50 filling ratio, the rate was 000864 units per h. However, it became 0.0164 units per h at 0.95 filling ratio. This indicates that the physically sorbed amount of the fumigant increases with increasing filling ratio.

The relationship between time and sorption rate constant was linear (Figs 8-13). Thus, *k* increased by 0.01422 with each hour at 0.95 filling ratio (Table 4). In addition, the density of grain at each particular filling ratio did not affect the accumulation rate constant *g* of the partition ratio of physical sorption *K*. However, it was affected by filling ratio. For example, *g* at 0.95 filling ratio and bulk density of 0.843 g/ml and grain density of 1.436 g/ml was 0.0164. However, it was 0.00864 at 0.50 filling ratio for bulk density and grain density (Table 3). Hence, *k* decreased at each particular filling ratio due to the impact of density. Results in Table 4 showed that the increase in the rate of *k* per one unit of ,*t*, was 0.01422 for bulk density at 0.95 filling ratio and 0.0003 for grain density. The dependency of physical sorption on the density and the accumulation of the fumigant physically at the same rate shows that the desorption of the fumigant will be the same at each density. However, the variation in chemical sorption due to different densities indicates the dependency of the chemically sorbed amount as it increases with density. Therefore, the predicted residues at low density grain will be less than in higher density grain.

Our results indicate that the rate of chemical sorption, *k*, was related to the physical sorption *K* (Figs 14-19). Darby (2008) developed a model of physical and chemical sorption showing that sorption occurred in a nonlinear manner with time. However, the impact of *K* on *k* was not mentioned in that study. The results of my work demonstrate the importance of investigating this relationship. The results of these experiments show that when *K* was high, the chemical reaction rate, *k*, increased (Table 5). That is, when rate of physical sorption increased, the rate of chemical reaction (sorption) increased as well. The high amounts of *K* are not a source of concern, as this is reversible (Hilton and Banks, 1997a). Turning points of the fitted line at each filling ratio and density provide crucial information and make the relationship between both rates easily understandable. The importance of the turning point is that it gives an indication about at which point of K the chemical sorption rate constant *k* will be reduced. The reaction constant *k* increases with filling ratio and density of commodity, but *K* is independent of filling ratio. As the results of this study indicate, the physical sorption happens with rates much greater than the chemical sorption (Tables 1 and 3). Therefore, physical sorption has the majority of control of the totally sorbed amount of the fumigant (physically and chemically). This gives the physical sorption an importance of controlling the lost amount of the fumigant into grain. In addition, due to the large amounts of physical sorption comparing to the chemical sorption, the factor of physical sorption affects the sorption process but not the chemical sorption.

The accumulation of physically sorbed fumigant into the wheat decreases the rate at which gas is chemically bound into the grain. The difference in *K* values between 0.95 and 0.50 filling ratio were not significant when compared with the rate at 0.75 filling ratio. Accordingly, the filling ratio of 0.50 seems to be the best filling ratio, as it a moderate filling ratio that controls the fumigation process by minimizing sorption and any potential residues. The independency of physical sorption from filling ratio was also observed by Hilton and Banks (1997a and b) with methyl bromide and ethyl formate applied to vine fruits.

4.5. Conclusion

Sorption of sulfuryl fluoride by wheat occurred at all filling ratios, and was higher in full containers than in half-filled ones. Sorption and the percentage loss per h of SF increased with increasing filling ratio and the time to lose half or 99% of the fumigant from the storage system into grain decreased as filling ratio increased. Sorption rate constant, *k*, depends on the filling ratio while physical sorption rate. *K* is independent of filling ratio. Both K and *k* depend on the density of the grain and both increase with increasing grain density. In addition, as the fumigant is sorbed exponentially, the accumulation rate constant, *g*, of physically sorbed SF increases exponentially as well. The accumulation rates increases with increasing filling ratio but not with increasing density. Moreover, the relationship between time and *k* is linear at all filling ratios and densities. The rate of *k* increases with increasing filling ratio and density. While the relationship between *k* and *K* is quadratic, and maximum *k* increases with increasing proportion of grain in the storage system and with the density of the wheat. The maximum *K* values are independent of filling ratio, but they are dependent on density. The dependency of maximum *k* on filling ratio and density, the dependency of maximum *K* on density, and the independency of maximum *K* on the filling ratio affect the turning points of the fitted line toward zero and gives *K* the major effect over *k*.

CHAPTER FIVE

Changes in sorption rates and fluoride residues during repeated fumigation of wheat with sulfuryl fluoride

ABSTRACT

Repeated fumigation is a normal procedure in the grain industry due to the failure of a first application to control infestation by insects. Repeated fumigation could lead to significant amounts of sorbed fumigant and possible residues. In this study, hard and soft wheat of 12.5% moisture content was fumigated five times with sulfuryl fluoride at 8.928 mg/L for 168 h at 25 C. Fumigant was sorbed into bread wheat more than into soft wheat. Sorption rate for bread wheat at the first fumigation was 0.0107 per h. However, it was 0.00857 per h for soft wheat at the same fumigation. Bread wheat continued to sorb the fumigant in rates more than soft wheat. At the last fumigation sorption rate for bread wheat was 0.00270 per h. However. it was 0.00234 for soft wheat. For both commodities, sorption rate decreased with each successive fumigation. However, fluoride residues increased with each fumigation, with the highest residue at the fifth fumigation. Repeating fumigation with tested concentration and fumigation conditions for four times or more makes fluoride residues higher than the current maximum residue in Australia.

5.1. Introduction

Repeated fumigation of the same parcel of grain often occurs in tropical and sub-tropical regions where insect pests will readily re-colonise fumigated grain. It is also common practice when initial fumigations result in poor control due to application problems. A serious issue with repeat fumigation is the risk of chemical residues reaching unacceptable levels, making the grain unfit for consumption and sale. Sulfuryl fluoride fumigation leaves sulphur and fluoride residues in grain with the latter being of particular concern. Despite this, there is no published information on the potential risks to residue levels resulting from repeat fumigation of grain with SF.

Successive decreases in the rate of sorption with sequential or repeated fumigations has been observed with phosphine (Daglish and Pavic, 2008) and methyl bromide (Banks, 1993), possibly due to the unavailability of “sites” on the commodity to react with the fumigant (Reed and Pan, 2000). This might lead to a reduction in the rate of residue uptake. Bromide residues in sultanas and raisins fumigated with methyl bromide at 5 mg/L (Hilton and Banks, 1997a) were 1.07 mg/kg for control, 2.15 mg/kg after one fumigation and 4.28 mg/kg after the fourth fumigation. Fumigating with methyl bromide at 60 mg/L left residues of 9.55 mg/kg after the first fumigation and 41.60 mg/kg after the fourth fumigation. In a survey of fluoride residues in Australian wheat fumigated once at 1500 CTP (fumigation time not given), fluoride residues ranged from <0.5 to 2.1 mg/kg (APVMA, 2007). APVMA (2007) also reported sulfuryl fluoride residues ranging <0.008 – 0.028 mg/kg in wheat fumigated once or twice for 24 h exposure periods, and fluoride residues in wheat ranging 1.42 – 14.3 mg/kg in trials conducted in Europe and USA.

The aim of this work was to quantify any changes in sorption rates associated with sequential fumigations in two commonly fumigated and traded commodities, bread (hard) wheat and soft wheat, and to determine the impact of this practice on fluoride residues in these commodities.

5.2. Materials and methods

5.2.1. Experimental design

This experiment involved fumigating bread wheat (hard) and soft wheat (*Triticum aestivum*) of 12.5% moisture content with 8.928 mg/L SF for 168 h, and the experiment was replicated three times. There are a number of differences between hard wheat and soft wheat. The most important of these is protein content, which is 11-13% for hard wheat. This type of wheat includes hard grained varieties that show excellent milling quality and are suitable for baking bread. Soft wheat protein is in the range 7-9%, and the flour makes a weak dough suitable for making cakes, biscuits and pastry. The words hard and soft refer to the force required for crushing the grain (Blakeney, 2009).

At each replicate, 12 flasks were prepared, six flasks (five were fumigated and one as control or nonfumigated) were filled to 0.5 total flask volume with bread wheat, and six flasks (five were fumigated and one as control or nonfumigated) were filled with soft wheat at the same filling ratio. The total number of flasks for three replicates was 36. Six of these flasks were controls, which contained the commodity but were not fumigated, and 30 were experimental flasks, which contained the commodity and were fumigated with SF. All flasks were stored at 25°C and 60% r h.

5.2.2. Commodity

Details are in chapter two, section 2.2.2.

5.2.2.1 Moisture content

Details are in chapter two, section 2.2.2.

5.2.3. Determining bulk, true density and commodity volume V_g

Details are in chapter two, section 2.2.7 and table 5.1.

Table.5.1. Moisture content, test temperature and cell volume used in determining bulk and true density for bread wheat and soft wheat.

Commodity	Moisture content%	Bulk density g/ml	True density g/ml	Standard deviation g/ml	Sample mass g	Test temperature °C	Sample volume ml	Standard deviation ml
Calibration ball	----------	0.0597	0.0597	0.000	1.0000	26.96	16.7566	0.0035
Bread Wheat	12.5	0.835	1.434	0.0050	23.8261	26.45	16.6145	0.0050
Soft wheat	12.5	0.835	1.421	0.0011	24.4951	26.52	17.2314	0.0135

5.2.4. Fumigation

Details are in chapter two, section 2.2.4

The procedure for multiple fumigation was as follows: A set of flasks were fumigated and after the first fumigation, all flasks were opened under the fumehood for aeration. Then two flasks, one for the bread wheat and one for the soft wheat sample were taken aside (first fumigation). The remainder of the flasks were fumigated for the second time, after checking moisture content, aerated and another set put aside and so on until all 5 fumigations were completed

5.2.5. Statistical analysis

Data was analysed using SAS software, and for graphing the Grapher program (Golden Software) was used. The decay of gas concentrations and the growth of the partition coefficient of physical sorption with time were analysed using first order equations

$$C_t=C_0e^{-k*t} \qquad (1)$$

Where, C_t was the gas concentration C at time t, C_0 was gas concentration C at time zero ($t=t_0=0$), $-k$ was the constant for rate of reaction, t was time

Hilton and Banks (1997a) also used another different set of models for calculating k_f, the constant rate of reaction for a full container

$$k_f=k/f, \quad (2)$$

where k was the observed rate of reaction for a particular filling ratio, f.

In addition, the percentage gas loss per h was calculated using the following equation:

$$\text{Percentage}= 100\ (1-e^{i}) \quad (3)$$

Where i was k.

Time to half (equation 4) and 99% (equation 5) loss of the gas was calculated as follows:

$$1/2L_0 = L_0\, e^{-i*t}/L_0,\ 1/2= e^{-i*t},\ t= \ln (1/2)/-i. \quad (4)$$

$$0.01L_0 = L_0\, e^{-i*t}/L_0,\ 0.01= e^{-i*t},\ t= \ln (0.01)/-i. \quad (5)$$

Where, L_0 was C_0, i was k and ln was the natural logarithm.

5.3. Results

5.3.1 Sorption

Sorption of the fumigant by both bread wheat and soft wheat was initially rapid followed by a more gradual phase in all fumigations (Figs 1-3) and rate of sorption, k, was higher in bread wheat than in soft wheat for all fumigations (Table 2). With each successive fumigation there was a decrease in %loss SF/h from the headspace of the experimental flasks (Fig 4) and the rate of reaction, *k*, decreased concomitantly (Table 2) in both commodities. Times to 50 and 99% loss of the fumigant increased with successive fumigations and were longer with soft wheat than with bread wheat (Fig 5). For example, for bread wheat and soft wheat, time to 50% sorption at the first fumigation were 65 and 81 h, respectively, while time to 99% sorption was estimated to be 537 h for soft wheat and 430 h for bread wheat. By the final fumigation, time to 50% loss of SF had increased from 65 h after the first fumigation to 257 h after the final fumigation for bread wheat and from 81 h to 296 h for soft wheat, respectively.

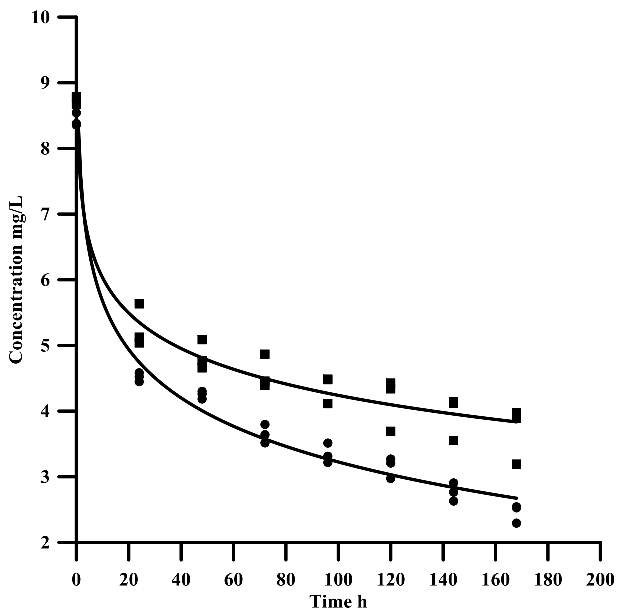


Fig.5.1. Sorption of SF into hard wheat ● and soft wheat ■ at 12.5% m.c., 25^{o}C fumigated at 8.928 mg/L for 168 h (average over five fumigations).

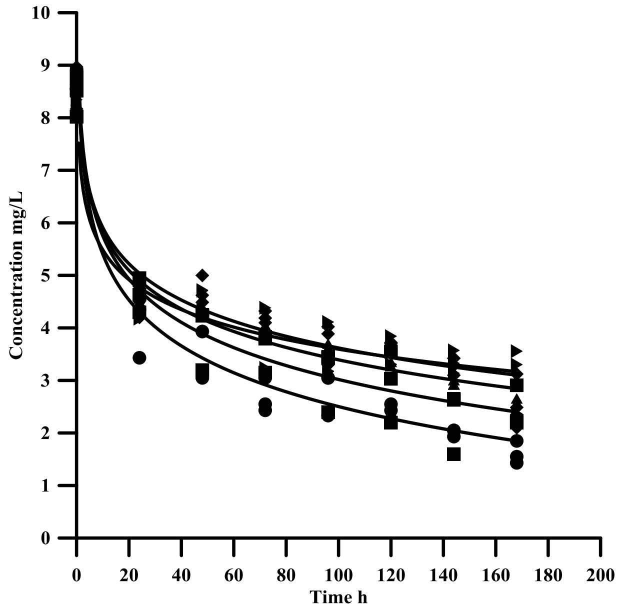


Fig.5.2. Sorption of SF into bread wheat 12.5% m.c. over five successive fumigations. Symbols indicate first ●, second ■, third ▲, fourth ♦ and fifth ► fumigation. SF was applied at 8.928 mg/L flask volume for 168 h at 25 C.

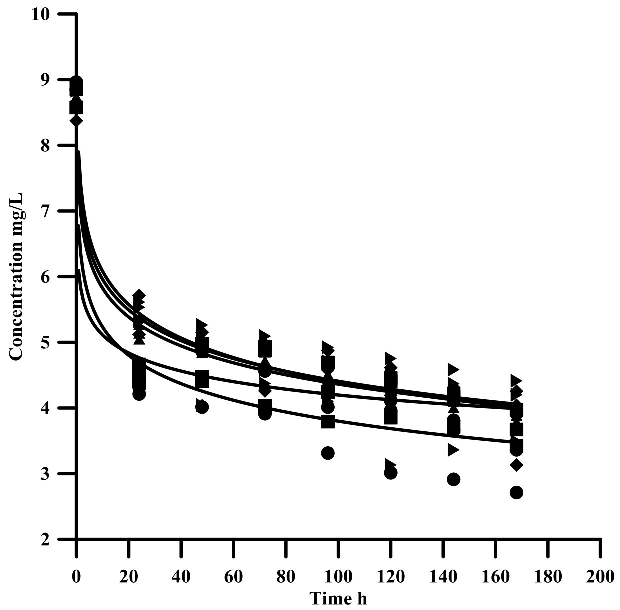


Fig.5.3. Sorption of SF into soft wheat 12.5% m.c. over five successive fumigations. Symbols indicate first ●, second ■, third ▲, fourth ♦ and fifth ► fumigation. SF was applied at 8.928 mg/L flask volume for 168 h at 25 C.

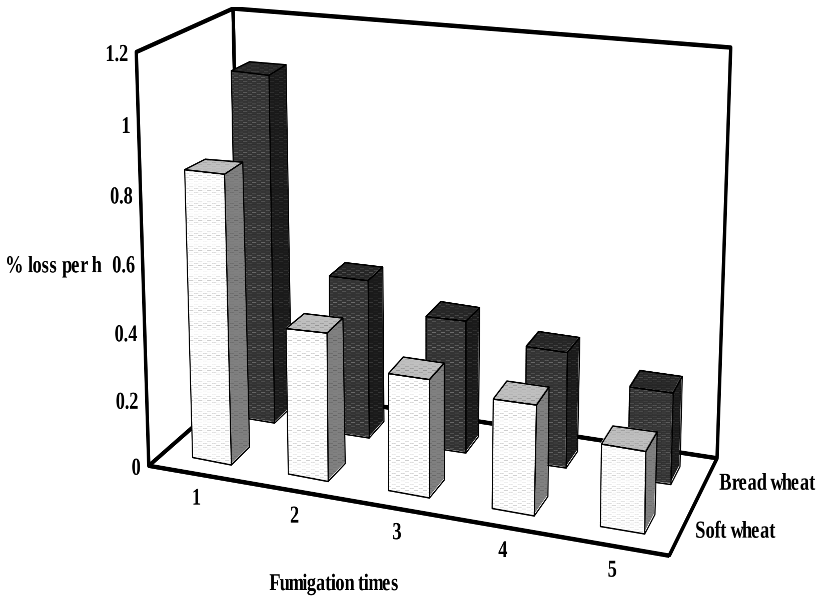


Fig.5.4. Change in % loss SF/h with successive fumigations of bread wheat and soft wheat at 12.5% m.c. fumigated with SF 8.928 mg/L flask volume for 168 h at 25 C.

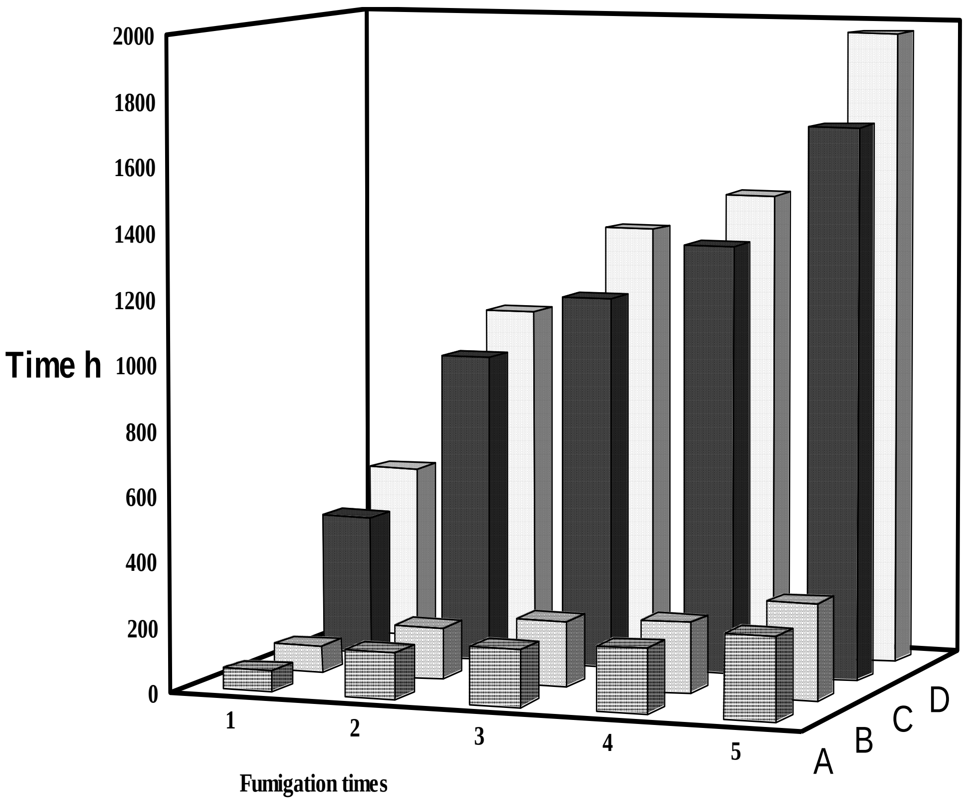


Fig.5.5. Change in time to sorb 50 and 99% of SF by bread wheat (A and C) and soft wheat (B and D) with successive fumigations with sulfuryl fluoride applied at 8.928 mg/L flask volume for 168 h at 25^{o}C. Wheat moisture content = 12.5%.

Table.5.2. Sorption of sulfuryl fluoride into hard wheat and soft wheat over 5 successive fumigations. SF was added to each fumigation flask at 8.928 mg/L flask volume for 168 h at 25°C, wheat filling ratio = 0.5, wheat m.c. = 12.5%. *kf* = sorption rate at 1.0 filling ratio.

Wheat	Fumigation	Regression	Significance	R^2	*kf*	% loss per h	Time to 50% loss (h)	Time to loss 99% (h)
Bread	1	$C_t=7.6087e^{-0.0107*t}$	$F_{2,22}=740.12$; $P<.0001$	0.96	0.021	1.064	65.26	430.37
	2	$C_t=7.2170e^{-0.00482*t}$	$F_{2,22}=635.33$; $P<.0001$	0.93	0.0096	0.480	143.77	955.39
	3	$C_t=7.1485e^{-0.00399*t}$	$F_{2,22}=670.31$; $P<.0001$	0.94	0.0079	0.398	174.28	1154.38
	4	$C_t=6.9524e^{-0.00347*t}$	$F_{2,22}=683.98$; $P<.0001$	0.95	0.0069	0.346	199.51	1327.08
	5	$C_t=6.8667e^{-0.00270*t}$	$F_{2,22}=417.75$; $P<.0001$	0.91	0.0054	0.270	257.16	1705.55
Soft	1	$C_t=7.4337e^{-0.00857*t}$	$F_{2,22}=408.51$; $P<.0001$	0.90	0.017	0.853	81.26	537.33
	2	$C_t=7.2672e^{-0.00434*t}$	$F_{2,22}=620.32$; $P<.0001$	0.93	0.0086	0.433	160.07	1061.05
	3	$C_t=7.1013e^{-0.00345*t}$	$F_{2,22}=581.02$; $P<.0001$	0.92	0.0069	0.344	201.27	1335.18
	4	$C_t=7.1626e^{-0.00317*t}$	$F_{2,22}=705.9$; $P<.0001$	0.95	0.0063	0.316	219.01	1453.08
	5	$C_t=7.2087e^{-0.00234*t}$	$F_{2,22}=680.51$; $P<.0001$	0.94	0.0046	0.233	296.15	1970.34

5.3.2 Residues

Fluoride residues increased with increasing number of fumigations (Table 3). The results showed that fluoride residues in un-fumigated wheat had a fluoride residue of <0.2 mg/kg and the same as recorded after the first fumigation. However, the residues at second, third, fourth and fifth fumigations were increased. There was no increase in SF residue detected after the first fumigation. Two possible reasons why no detectable F was found after one fumigation are 1) error in the measurement at these low concentrations as we only did single estimations due to cost contraints; 2) it takes more than one fumigation before detectable levels of F are found with this assay.

Table.5.3. Effect of repeated fumigation by SF 8.928 mg/L at 25°C for 168 h on fluoride residues of bread wheat 12% moisture.

Number of fumigations	Fluoride residue (mg/kg)
0	<0.2
1	<0.2
2	3.3
3	5.1
4	8.5
5	13.0

5.4. Discussion

In these experiments, SF continued to be sorbed through successive fumigations but the extent (%) and rate of sorption decreased with successive fumigations (Fig 4) in both commodities. However, fluoride residues increased with each fumigation (Table, 3). Continued sorption may lead to the accumulation of significant amounts of chemical residues as has been demonstrated with methyl bromide fumigation of vine fruit (Hilton and Banks, 1997a). Bromide residue increased from 9.55 mg/kg at the first fumigation to 41.60 mg/kg at the fourth fumigation. However, there was a successive decrease in sorption with each fumigation. Therefore, we predict that multiple fumigation of wheat with SF would lead to an increase in residues as observed. In these experiments, fluoride residue was 0.2 mg/kg at control (Table 3). However, the residues became 13 mg/kg at the fifth fumigation. These results are similar to those of Hilton and Banks, (1997a) with methyl bromide, but with higher residue level than we obtained using SF. This may be because SF is sorbed relatively slowly compared to methyl bromide. The results of this study when fumigating hard wheat of 12% moisture with 8.298 mg/L SF for 168 h at 25°C for four times or more indicate that repeating fumigation increases fluoride residues, and the residues becomes higher than the Australian maximum residue limit of fluoride after 4 or more fumigations. The current maximum residue limit of fluoride for wheat in Australia is 7 mg/kg (APVMA, 2007). Their studies did not look at residue levels for repeated fumigations with SF under varying conditions of wheat moisture content, fumigation temperature, and dose of SF.

Our results indicated that SF was sorbed at a greater rate into bread wheat than into soft wheat (Table 2, Fig. 1). The results indicate sorption rates of bread wheat at the fumigation from one to five were 0.0107, 0.00482, 0.00399, 0.00347 and 0.00270 compared to soft wheat of:0.00857, 0.00434, 0.00345, 0.00317 and 0.00234. This decrease in k with more fumigations is similar to that reported by Reed and Pan, (2000) for bread wheat fumigated with phosphine. Comparing sorption rates for SF with the rates calculated by Reed and Pan (2000) for phosphine shows a difference between SF and phosphine when fumigating 1.4 tonne of bread wheat in sealed bins with moisture content of 13.5% and using higher concentrations of phosphine than with this study. However, the difference of sorption rate, k, between the first and second fumigation for both phosphine and SF was very close at about 25% although SF was sorbed more than phosphine. The difference in sorption rates between the fourth and fifth fumigations was around 7% for both fumigants.

Bread wheat contains higher protein (11-13%) than soft wheat (7-9%) grains (Blakeney, 2009), and this may be the reason for the significant difference in sorption rates between the commodities. SF is believed to bind with proteins and amino acids in the commodity (Meikle, 1964) and is converted to sulphate and fluoride ions. Therefore, the results of this study indicated a higher sorption in bread wheat compared with soft wheat because of a higher protein content.

Although Pepper et al., (1974) concluded that the amount of CCl_4 sorbed by wheat was a function of lipid content; most studies indicate that protein content is the most important factor. Mitsuda et al., (1975) demonstrated that the sorption of CO_2 was correlated with protein content. Similarly, Lewis and Eccleston, (1946); Berck, (1968) and Banks, (1986) demonstrated the relationship between the accumulated amount of the fumigant onto and into grain and protein content.

5.5. Conclusion

The rate of sorption of the fumigant declines with increasing number of fumigations. However, the accumulated amounts of the fumigant by both bread wheat and soft wheat increase with repeating fumigation. Fluoride residues increase with increasing the number of fumigations. Protein content could be a key component that affects the amount sorbed by the commodity as bread wheat sorbed the fumigant with rates more than soft wheat and is of higher protein content. Flouride residues of this chapter were based on single replicate and to confirm the trend these experiments should be repeated with a minimum of two replicate estimations.

CHAPTER SIX

Effects of fumigation with sulfuryl fluoride on some technological characteristics of wheat and durum

ABSTRACT

A selection of samples of wheat, durum grains. flour and semolina fumigated under the conditions discussed in chapter 2 were analysed for several technological properties. In addition, fluoride residue was tracked in a selection of samples to help understand where this residue is being deposited. SF affected germination of wheat significantly greatly reducing the % germination. The lowest germination percentage was 1.5% at SF 31.25 mg/L, 15% moisture and 35°C compared to the highest of 90.25 percent at 0 SF, 15°C and 15% moisture. Applying SF decreases the hectolitre weight and ash content but increases the hardness of the grain. Fumigation with SF increases the yellowness of semolina, cooking loss, over cooking tolerance, firmness and stickiness. However, Fumigation with SF at different conditions did not show any impact on tested bread. A fluoride residue in cereal grain 13 mg/kg was higher than the maximum residue limit of 7 mg/kg. However, after milling fluoride residue was lower than the maximum residue limit in the semolina as the majority of fluoride residues were concentrated in bran. Therefore, milling whole grain for flour or semolina reduces fluoride residue of fumigated wheat. Fluoride residues decreased after making pasta, falling from 6.1 mg/kg in the fumigated semolina to 2.1 mg/kg after processing into pasta. However, high levels of fluoride (53 mg/kg) were measured in cooking water but in cooked pasta the residue level was 6.7 mg/kg.

6.1. Introduction

During storage both durum wheat (*Triticum durum*) and bread wheat (*Triticum aestivum*) are at risk of insect infestation which can lead to significant physical damage, mould contamination and reduction in nutritional and other qualities of the grain making it unsuitable for its intended uses (Bailey, 1992; Rajendran, 2003). Grain storage managers often resort to use of fumigants and other chemicals to protect and disinfest stored wheat. These chemicals are known to be sorbed by the grain and to react with various components of the seed potentially leaving unwanted residues and perhaps changing the inherent characteristics of the grain (Bond, 2007; Hwaidi et al., 2015). Applying fumigants during storage may affect the germination ability of the fumigated seed. Fumigation with methyl bromide Powell, 1975; Minett et al 1976; Strong and Lindgren, 1959 decreased the germination of onion seed and wheat significantly. However, Orth et al, 1977; Lubatti and Blackith, 1957; Whitney et al, 1958 reported no reduction in germination percentage using methyl bromide on wheat. Fumigating rice and wheat with phosphine did not affect germination even after storage (Krishnasamy and Seshu, 1990; Bakheit et al 1985). In addition, fumigating wheat and corn with phosphine did not affect germination (Cogburn and Tilton, 1963; Lindgren et al, 1958). Applying commercial dose of methyl bromide to wheat, wheat flour or milled flour from fumigated wheat did not change bread baking characteristics (Orth et al., 1977; Matthews et al., 1970; Shepard and Buzicky, 1939; Hermitte and Shellenb, 1947). However, applying methyl bromide with doses higher than the commercial dosage increased the resistance of the dough and decreased loaf volume (Minett et al, 1976). Australian Pesticide and Veterinary Medicines Authority (APVMA, 2007) investigated the fluoride residues in different types of Australian wheat after fumigation with CT of 1500 mg.h/L and results indicated fluoride residues in different wheat types were between 0.5-2.1

mg/kg which was lower than the maximum residue limit in Australia of 7 mg/kg. The effect of fumigation with SF on grain and pasta quality is unknown.

There is no information regarding the influence of fumigation with SF on technological characteristics of durum wheat and the residues that can accumulate. Therefore, this work was designed to investigate the effect of fumigation with SF at different conditions on grain germination and the technological quality of grain, semolina, pasta, bread making quality and quantifying any residual fluoride in these fractions.

Investigating the grain and its products technologically gives an indication of the suitability of using the grain as seed for planting and the product for human consumption

6.2. Materials and methods

6.2.1. *Germination*

This experiment included durum wheat seeds for the germination test. Seeds had been stored at -20°C for 2.5 years before commencing the experiments. Samples were taken from the freezer and pre-chilled between 5 to 10°C for 7 days before carrying out the tests. Petri dishes, filter paper (top paper germination medium) were used for the purpose. Lots of 100 seeds were taken for each treatment in the design and placed into petri dishes. Water was added as needed, and germination temperature was 20°C. Germination was monitored and recorded daily after the fourth day until 8 days. Germination percentage was calculated (ISTA 2011).

6.2.2. *Thousand grain weight*

Grain weight varies with cultivar and the thousand grain weight (TGW) can provide additional information about the size and density of the grain. Uniform grain weight is important for consistent grain quality. A NUMIGRAL seed counter model C 3501 was used for counting for hard and durum wheat grains. The appropriate disc (for bread wheat and durum wheat) was used and aligned with chute that leading to the collecting container. Counting button was set to (0) before grain collection. When a count of 250 kernels was completed, the collection container was removed, and the grain sample was weighed. This weight was multiplied by four to give the 1000 grain weight.

6.2.3. *Test weight or hectolitre weight (HLW) and grain hardness*

A Franklin chondrometer I pint mark II type (choke hopper) was used for measuring hectolitre weight. The choke hopper was placed with the blade closed in the top of the bucket so that the pin protruding inside the rim locked in the slot on the outside of the hopper bottom. A sampling scoop was filled to about 25-50 mm from the top with the lip of the scoop resting on the edge of the hopper inlet tube. Wheat grain was poured in smoothly and steadily until it spilled over into the overflow well then the hopper blade was opened. With blade closed, the excess amount of grains from the top of the bucket was poured into the scoop. The grain weight and the determination of the hectolitre weight (HLW) was calculated using the equation ($x = y / 5.69$). where y = the grain weight and x = Kg/hL (1)

Grain hardness was measured by NIR.

6.2.4. *Ash content*

Approximately 2.5 g accurately weighed sample of well mixed semolina or flour was placed into metal moisture tins with lid off. A check sample (RACI) with known ash content, was included with the samples. Samples and crucibles were placed in an oven (Thermoline Scientific Equipment Pty Ltd, model 045F-421-D-MOD) at 85°C for 48 h then at 40°C for about 30 minutes, to equilibrate. Two moisture tins were taken out of the oven each time in addition to the same number of crucibles and weighed. Samples were incinerated in a. muffle furnace, (S.E.M Pty, Ltd, Australia with a pyrometric controller, model N759) and programmed for 2 hours at 190°C, 4 hours at 600°C, 24 hours at 190°C . Crucibles were left in a desiccator for 5 minutes to cool down. Crucibles were weighed and the ash percentage calculated:

%Ash (dry matter basis) = (weight of residue) / sample weight x 100 (2)

Where:

Weight of residue = crucible + ashed sample weight - crucible weight.

Sample weight = crucible + sample weight - crucible weight.

6.2.5. Fluoride residue analysis

Different grain fractions were prepared for fluoride (F) residue analysis such as fumigated whole bread and durum grains, and fumigated grain that was milled into semolina or wholemeal, and bran fractions. In addition, commercial semolina and flour that were fumigated were also tested (samples were taken from chapters 2, 4 and 5). Pasta was made from fumigated semolina and cooked and the cooking solids and cooked pasta were also examined for F residue. Whole grains were milled at the laboratory using a Quadramat Junior Mill and the bran and semolina fractions were analysed for residue to determine where the residues. Pasta made from fumigated semolina was cooked and the cooking water was dried in a (DYNAVAC) freeze dryer model (FD1) at -45°C for 7 days to follow the fate of fluoride if it was desorbed from pasta in the cooking water or still retained in the pasta. Fluoride analysis was contracted to a laboratory in the George Weston. Foods Pty Ltd, Sydney, Australia group.

6.2.6.1. Milling grain into semolina using the Quadrumat Junior Mill

The Brabender Quadrumat® Junior Mill with sieve (6xxx 212 µm) is a precision laboratory roller mill with high throughput capacity. The multi-step grinding process allows the production of flour/semolina from small quantities of grain enabling further small scale testing of flour/semolina quality. Grains were conditioned before milling by measuring moisture content using NIR. Then, all samples were conditioned to 15% m.c by using the following equation:

[(100-original moisture% / 100-desired moisture%)-1] x weight of sample, g. (3)

The required amounts of water were added to the grains which were then tumbled for one hour and left overnight. Each conditioned grain sample was placed in the hopper of the mill and the feed gate was turned on until the grain just started to go through the mill until the last grain milled. A small bottle brush was used for cleaning the spout. The flour tray was removed and the weight of the contents was recorded as Total Flour (g). The bran tray was removed and the content weight was recorded as the Total Bran (g). The reel sifter was removed and the content was poured into a plastic bag as (reel sifter contents). Through the opening for the reel sifter, all flour adhering to the walls of the sifting compartment into the flour draw was brushed. The draw was removed and the weight of this material with the reel sifter contents was recorded as the Total Reel weight (g).

6.2.6.2. Milling of wheat for pasta making

A Buhler mill (model ML202 laboratory mill, Buhler AG, Uzwil, Switzerland) was used for milling durum into semolina for pasta making. Approximately 18 h before milling, samples were conditioned to 15% moisture by adding the required amount of water depending on NIR moisture determination. The required amount of water was calculated according to the following equation:

[(100-original moisture% / 100-desired moisture%)-1] x weight of sample g. (4)

Then samples were tumbled for 15 to 20 min and left overnight. About 30 min prior to milling, 1% of water, depending on sample weight, was added to each sample as a temper. Samples were placed subsequently into the mill with a feed rate of 100 g per min. Then the next sample was tempered while the previous one was being milled. The mill was cleaned up between samples following the recommended instructions. After milling each sample, semolina, bran and pollard were collected. Fractions were weighed and the mill yield or extraction percentage was calculated as:

Extraction %= total flour weight / total products weight * 100. (5)

Semolina yield %= semolina weight / total products weight * 100. (6)

6.2.6.3. Experimental pasta making and evaluation

Semolina prepared from Buhler milled wheat was purified on a small-scale purifier (model GW inox ZAC DES CADESREAUX, 2001) whereas commercial semolina was used as received. Purified semolina was used to prepare long pasta (spaghetti) using a Namad pasta extruder (Appar Laboratorio, Rome, Italy). Semolina was mixed with distilled water (30% by weight) in a pre-mixing chamber for 10 min. The mixture was then extruded under partial vacuum (8 kPa) at 50°C through a teflon coated die piece into spaghetti, cut and looped over metal rods and hung in a drying cabinet maintained at 25°C and 85% rh (diameter of dried pasta, 1.82 ± 0.025 mm). After the last sample was processed, the drying cycle commenced and pasta was dried at 65°C at 70% rh for 45 min then for 13 h at 50°C and 80-70% rh followed by cooling to 25°C at 55% rh for 4 h. The pasta was kept at room temperature for a minimum of a week to stabilise moisture movement before further analysis. All pasta samples were cooked to their optimum cooking time (OCT) and texture, water absorption, and cooking loss were assessed using methods as described below.

6.2.6.4. Pasta optimum cooking time (OCT)

The optimum cooking time can be defined as the time at which the white starchy thread disappears used for all tested pasta. Broken pasta strands of 8 to 7 cm length were cooked in 250 ml of boiling water in a beaker. After ten minutes, three strands were taken after each 30 seconds and squashed between hinged perspex plates and viewed against black background to search for starchy white threads because a clear starchy thread means inadequate cooking time. The process was continued until the starch white threads disappeared in all three test strands and this time was called the optimum cooking tome. At each time test, the number of cooked and uncooked pasta strands was recorded.

6.2.6.5. Pasta cooking loss and water absorption

Cooking loss was measured by cooking pasta to its optimum cooking time then weighing the residue left in the beaker after the evaporation of the cooking/rinse water. Cooking loss is expressed as a percentage of the uncooked pasta weight.

Water absorption is measured by cooking pasta to its optimum cooking time, then weighing the cooked pasta sample after cooling and draining. The amount of water absorbed by the pasta is expressed as a percentage of uncooked pasta weight.

Samples of approximately 5 g of 3.5 cm length pasta strands were weighed accurately. 125 ml of RO (Reverse Osmosis) water were placed into a 250 ml tall form beaker and brought to a rolling boil on a hotplate (a wooden skewer was added to the beaker so it was easier to tell if the water was boiling). A 5 g pasta sample was added, stirred with the wooden skewer to separate strands. The skewer was removed, and pasta strands were cooked to their OCT. The pasta strands were stirred with the skewer periodically during cooking. When cooking of pasta strands reached OCT, pasta cooking water was collected into pre weighed and numbered beakers while capturing the pasta on a nylon sieve. Pasta strands were washed briefly with a stream of distilled water and this was added to the cooking water. The pasta was placed into a beaker of 250 ml water at room temperature water for two minutes to arrest cooking, drained through the nylon sieve and excess water was blotted on a tea towel. The cooked pasta was weighed to obtain a measure of water absorption. Pre weighed beakers containing the cooking water were evaporated to dryness (constant weight) in an air oven at 100°C ± 1° C. The beakers were cooled in a desiccator for 20 min and weighed to 0.01g.

% cooking loss and % water absorption were calculated as following

% cooking loss = B-A/Y*100/1 (7)

% water absorption= X-Y/Y*100/1 (8)

A= Empty beaker weight

B= Weight of evaporated beaker

X = Cooked pasta weight

Y = Uncooked pasta weight

6.2.6.6. Pasta colour: cooked and uncooked

Uncooked spaghetti colour (DP-) was measured on the HunterLab scale for L*, a*, b* and whiteness index using 7 cm length pieces of spaghetti aligned together and measured in triplicate. Cooked pasta colour (CP-) was measured on optimally cooked pasta that had been drained and blotted dry and the strands aligned.

Colour stability was calculated as:

$$\sqrt{[(\text{Dry } L^*-\text{Cooked } L^*)2 + (\text{Dry } a^*-\text{Cooked } a^*)2 + (\text{Dry } b^*-\text{Cooked } b^*)2]} \quad (9)$$

6.2.6.7. Cooked pasta Firmness

Cooked spaghetti firmness is an important quality parameter for pasta. Good quality pasta should have a firm bite after cooking ('al dente') and should maintain this firmness after some overcooking. Cooked pasta diameter is a useful indicator of pasta absorption during cooking. Five spaghetti strands which had been cooked to their OCT were placed on the platform of the Stable Microsystems TAXT2i Texture Analyser fitted with the firmness probe (A/LKB). In firmness testing the probe cuts at a right angle through the spaghetti to a distance of 0.3 cm from the platform base. A force vs. time graph was generated, and the peak height and area under each curve calculated. The peak height was taken as the firmness value. The diameter of the cooked spaghetti strands can be calculated and an indication of the spaghetti water absorption deduced.

Cooking Stock Solution, 10ml [175g of salt (NaCl) and approximately 0.125g of Sodium Hydrogen Carbonate ($NaHCO_3$) in 1,000ml of RO water] and 240 ml of water were boiled in a beaker using a hotplate capable of heating to 400°C. Pasta strands were broken into 15 strands of approximately 7 cm in length and five strands of pasta were placed into rapidly boiling cooking water and cooked to OCT, drained and cooled in 250 ml of RO water at room temperature for 2 minutes. Pasta was drained using a small nylon sieve, and the sieve was placed onto the absorbent paper in a plastic bag. After one minute, 5 strands of pasta were removed and placed on the base plate of the Texture Analyser, parallel to but not touching one another so that the knife cut each strand about in half. After completing the test, the next five strands were tested using the same procedure.

6.2.6.8. Cooked pasta stickiness

The surface stickiness of pasta is an important quality parameter. High surface stickiness is undesirable. Ten mls of Cooking Stock Solution [(175 g of salt (NaCl) and 0.125 g of sodium hydrogen carbonate ($NaHCO_3$)] and 250 ml of RO filtered water was added to a 600 ml beaker and placed on a hotplate to obtain a rolling boil. Five strands of pasta were placed into the boiling water and cooked to their OCT. Strands were drained and placed in 250 ml of RO filtered water at room temperature for 2 minutes, and a timer was set at 3 minutes counting down to allow for the generation of some stickiness. Strands were then placed on TAXT2i Texture Analyser heavy duty platform fitted with the stickiness probe. After finishing each test, values of force, area 1:2 and area 3:4 were recorded. The texture analyser was repositioned and recalibrated before the next test, and the probe was recalibrated for distance between probe and platform for every test. Test was repeated for the remaining two replicates.

6.2.6.9. Semolina colour

In 1976, the Commission International de I'Eclairage (CIE) recommended the CIE L*a*b* or CIELAB colour scale for use. It was intended to provide a standard, approximately uniform colour scale which could be used by everybody.

Semolina samples were mixed in a paddle tumbler for 10 minutes and placed in a glass tray and made flat with a ruler and a Minolta Chroma Meter CR-410 (calibrated against a white tile) was used to collect three measurements moving the camera to different parts of the sample. The average of three measurements was displayed by the camera and recorded. The measurements collected were L*, a*, b*, WI and YI. Briefly, L* is a measure of brightness from black (0) to white (100). a* is a function of the red-green difference: positive a* is redness, and negative a* is greenness. Positive b* indicates yellowness; negative b* indicates blueness. Other colour parameters collected are, Y1 (yellowness index or degree of yellowness) and W1 (whiteness index or degree of whiteness).

6.2.6.10. Mixograph

The 10 g mixograph is an ideal instrument for measuring the important mixing properties of wheat flour and durum semolina. The mixing curve (mixogram) measures and records the resistance of a dough to mixing, tolerance to over mixing, and the optimum development time (which is the point of minimum mobility) and other dough characteristics.

Semolina (10±0.01 g) was weighed into the mixing bowl to which 7 mL of RO filtered water was added. The mixing continued for 9 minutes using a procedure described elsewhere (AACC 2001). The mixograph manual trace and a computer analysed graph using mixsmart was collected. Measurements collected include MPT (mixograph peak time), MPH (mixograph peak height), WAP (width of mixogram at peak time), WA8 (width of mixogram 8 minutes past peak mixing time), RBD (resistance breakdown calculated as:

$$RBD = (WAP - WA8) / WAP * 100 \quad (10)$$

6.2.7. Bread baking

6.2.7.1. Milling of durum wheat for bread making

As a part of the preparation for bread making the milling of durum wheat was done at Tamworth Agricultural Institute (TAI) using a Perten falling number mill 3100 with a 0.8 mm sieve. Each sample was put in a plastic bag and marked with the sample number. Wholemeal samples were sent to the Wagga Wagga Agricultural Research Institute for bread making and evaluation.

6.2.7.2. Test bake - rapid process

Durum wheat samples were milled into wholemeal flour as described in section 6.2.7.1 In addition, a base hard wheat commercial flour was used as a control and mixed with the wholemeal durum sample as a 30% substitution. Baking was carried out according to CCD 07-03 method. 200 g of mixed durum wholemeal - commercial hard wheat flour was mixed with 1g improver, 80 ml of 5% salt solution, 6 g yeast, 2 g fat and 4 ml of acetic acid solution. Dough was mixed at 180 rpm until development. Loaves were scored in two replicates next day, with a full replicate (randomised order) occurring each day including check bakers flour loaves. Dough was mixed to optimum development in a pin mixer (National Manufacturing, Nebraska, USA), then fermented for 3 h at 30°C. Dough was run through a Mono Universal moulder (Mono Equipment, Swansea, UK) at a roll gap of 4.5 mm and pressure board height of 35 mm. Dough was scaled two X 150 g each after 7 min rest time and moulded at 10 min after mixing then moved to tins after 8 min rest time. Dough was proofed for 70 min at 34°C and 80% rh before baking at 215°C for 20 min in a Rotel II bakery oven (Moffat, Australia). Loaves were cooled overnight and judged the following day. Loaf volume was determined using rape seed displacement in a pup volumeter (National Manufacturing, Nebraska, USA); crumb colour was measured in CIE L*, a*, b* colour space using a Minolta Chromameter (Konica Minolta Sensing Inc, Japan) fitted with a 50 mm glass fronted head. External loaf appearance, crumb texture (softness and resilience) and crumb cell structure were judged subjectively against the baker's flour comparison loaves. Bake scores were awarded on the basis of a maximum 20 points for volume, and 10 each for external appearance, crumb texture and crumb structure.

6.2.8. Experimental design

6.2.8.1. Germination and 1000 grain weight

Samples in this study were taken from chapter 2 section 2.2 of durum wheat (*Triticum durum*) exposed to SF at four concentrations (0, 4.167, 8.928, 31.25mg/L) at either 15, 25, or 35°C and either 12 or 15% grain moisture content, in triplicate (total samples=24). These samples were subject to a germination test that was performed in quadruplicate and 1000 grain weight determination.

6.2.8.2. Protein, grain hardness, hectolitre weight and milling study

The same pool of samples in section 6.2.8.1 used for the germination test also had protein, grain hardness and test weight measurements preformed.

6.2.8.3. Ash study

Durum semolina and common wheat flour samples of 12 and 15% m.c that had been fumigated with SF 0-31 mg/L at 15, 25 and 35°C as described in section 6.2.4 were analysed for %ash.

6.2.9. Fluoride residues

Fluoride residue was determined in fumigated durum wheat grains, milled semolina and bran from durum, commercially supplied semolina, freeze-dried cooking water residue and uncooked and cooked freeze-dried pasta. Bran was obtained from the durum wheat grain milling process, and cooking water was obtained from cooking pasta to its optimum cooking time, draining the pasta and drying the cooking water to dryness in an oven. The resulting residue was analysed for fluoride.

Table.6.1. Experimental design for fluoride residue.

Sample	SF mg/L	Exposure time h	Temperature °C	Replicates	Moisture content %
Fumigated durum wheat	8.928	168	15	1	12
Fumigated durum wheat	8.928	168	25	1	12
Fumigated durum wheat	8.928	168	35	1	12
Milled Semolina	4.167	360	25	1	12
Milled Semolina	8.298	168	25	1	12
Milled Semolina	31.25	48	25	1	12
Bran from milling	4.167	360	25	1	12
Bran from milling	8.298	168	25	1	12
Bran from milling	31.25	48	25	1	12
Commercial semolina	0	168	15	1	12
Commercial semolina	0	168	25	1	12
Commercial semolina	0	168	35	1	12
Commercial semolina	8.928	168	15	1	12
Commercial semolina	8.928	168	25	1	12
Commercial semolina	8.928	168	35	1	12
Pasta control-uncooked and cooked	0	168	25	1	12
Fumigated pasta-uncooked and cooked	8.928	168	25	1	12
Freezedried cooking water	8.928	168	25	1	12

6.2.9.2. Effect of different filling ratio on fluoride residues in durum wheat.

Sample for this study were taken from chapter 4 section 2.1.

6.2.10. Pasta

6.2.10.1. Pasta testing

Durum wheat samples (3 replicates per treatment) from fumigation experiments SF 0-31 mg/L, temperature 15-35°C at 12 % m.c.; as described in section 6.2.6.3 were tested milled and the semolina made in to pasta. Pasta was evaluated for cooking time, cooking loss, Cooked and uncooked pasta colour, cooked firmness, stickiness, over cooking tolerance and fluoride residue.

6.2.10.2. Mixograph and semolina colour study

Durum wheat samples fumigated from the study described in sections 6.2.6.10 and 6.2.6.9 respectively, (SF 0-31 mg/L; temperature 15, 25 and 35°C; 12 and 15% m.c), 3 replicates per treatment were milled into semolina and examined for colour space parameters and mixograph test was performed.

6.2.11. Bread

The grain used for the bread making study was fumigated with SF 0-31 mg/L; temperature 25°C; m.c 12%, in triplicate) and the samples milled into wholemeal then mixed at 30% replacement with a commercial hard wheat and made into 100g pup loaves and assessed for External assessment (Oven spring, Loaf volume, External colour, Blister) and Internal assessment (Distribution, Structure) and Softness and resilience

6.2.12. Statistical analysis

Data was analysed for F-test using the statistical programme Gen-Stat version 9, and the means were tested for the significant differences by the Least Significant Difference (LSD).

6.3. Results

6.3.1. Germination

Results of this study indicated a highly significant decrease in germination percentage with increasing SF concentration mg/L (Table 2). Results in Table 3 show the highest germination percentage of 89 in control (not fumigated grain). Germination percentage decreased significantly with increasing SF concentration, temperature and moisture. Germination percentage was 30, 24 and 19 at SF 4.167, 8.928 and 31.25 mg/L, respectively

(Table 3). Storage temperature showed a significant negative affect on the germination in the presence of the fumigant (Table 2). For instance, at the storage temperatures of 15, 25 and 35°C the reductions of germination were about 44, 61 and 72%, respectively. Moreover, the increase in grain moisture content reduced the germination percentage also in the presence of SF. At 12% moisture content, the overall mean %G was 51% but at 15% moisture content, germination mean fell further to 31% (Table 3). In addition, the combination effect of SF x temperature, SF x moisture, temperature x moisture and SF x temperature x moisture decreased the germination percentage significantly (Table 2). Results in Table 3 showed that the highest percentages of germination were 90 at (15°C x control), 90 at (12%M x control), 74 at (15°C and 12% moisture content) and 90 at (0 SF x 15% x 15°C). However, the lowest percentages of germination were 1 at (35°C x 31.25 mg/L of SF), 7 at (15% x 31.25 mg/L), 25 at (35°C and 15% moisture content) and 1 at (31.25 mg/L x 15% x 35°C).

Table.6.2. Germination response of durum wheat 12 and 15% moisture fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	3	5.69E+00	1.89E+00	3.44	ns
SF	3	7.71E+04	2.57E+04	46621**	<.001
Temperature	2	1.26E+04	6.33E+03	11485**	<.001
Moisture	1	9.94E+03	9.94E+03	18029**	<.001
SF x Temperature	6	1.06E+04	1.78E+03	3230**	<.001
SF x Moisture	3	3.48E+03	1.16E+03	2108**	<.001
Temperature x Moisture	2	3.69E+03	1.84E+03	3353**	<.001
SF x Temperature x Moisture	6	3.17E+03	5.29E+02	959.31**	<.001
Error	69	3.80E+01	5.51E-01		
Total	95	1.20E+05			

** Significant at 0.01%
* Significant at 0.05

Table.6.3. Germination response of durum wheat 12 and 15% moisture fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
Moisture content	12%	90a	86c	78d	42f	90a	50e	36g	18.i	90a	24h	8m	1o	1.04
	15%	90a	41f	10k	9.l	90a	12j	12j	6n	88b	8lm	1o	1o	
Temperature X SF		90a	63b	44c	26e	90a	31d	24f	12h	89a	16g	5i	1j	0.74
Temperature means		56a				39b				28c				0.37

SF mg/L X Moisture	0		4.167		8.928		31.25		LSD 5%
	12%	15%	12%	15%	12%	15%	12%	15%	
	90a	89a	43b	18e	41c	8f	31d	7g	0.60

SF mg/L means	0	4.167	8.928	31.25	LSD 5%
	89a	30b	24c	19d	0.42

Moisture X Temperature	12%			15%			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	74a	48	31d	37c	30e	25f	0.52
Moisture means	51a			31b			0.30

Same letter refers to non-significant differences.
Different letters refer to significant differences.

6.3.2. Effects of SF fumigation on grain quality traits of durum wheat

Hectolitre weight was affected significantly by SF, moisture and the combined effect of SF x moisture (Table 4). The highest weight was 82.3 Kg/hL for the control (not fumigated), and this decreased slightly with SF although there were no significant difference between the various SF concentrations. Hectolitre weight was 81.1, 80.9 and 80.9 Kg/hL at 4.167, 8.928 and 31.25 mg/L, respectively (Table 5). The difference between the two moistures was significant and the highest weight was 82.2 Kg/hL at 12%. However, it was 80.40 Kg/hL at 15% moisture. The weight of 82.3 Kg/hL was at the combination of (12% moisture content x 8.928 mg/L).

Grain hardness was affected significantly by SF fumigation only but not the other storage conditions (Table 6). The hardness of the grain increased with fumigation but there were no significant differences among the three concentrations of SF (Table 7).

The impacts of SF, temperature, moisture and their combinations on 1000-grain weight (Tables 8; 9) and protein content (Tables 10; and 11) were not significant.

Ash content was significantly lower for durum semolina compared to bread wheat flour (Table 12). The overall mean ash percentage in wholemeal flour was 1.78 and in semolina, 0.94 percent. Fumigation with SF decreased the ash content compared to the control. The highest ash content was 1.93 percent at the combination of (control x flour) (Table 13). The impact of temperature, moisture and their combinations with the other factors were not significant.

Table.6.4. Analysis of variance of durum wheat 12 and 15% moisture hectolitre weight Kg/hL response to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	1.398	0.699	4.50	
SF	3	21.665	7.222	46.47**	<.001
Temperature	2	0.095	0.048	0.31	0.736
Moisture	1	63.131	63.131	406.20**	<.001
SF x Temperature	6	0.035	0.006	0.04	1.000
SF x Moisture	3	23.747	7.916	50.93**	<.001
Temperature x Moisture	2	0.083	0.041	0.27	0.766
SF x Temperature x Moisture	6	0.282	0.047	0.30	0.933
Error	46	7.149	0.155		
Total	71	117.588			

** Significant at 0.01%
* Significant at 0.05

Table 6.5. on page 146

Table.6.6. Analysis of variance table durum wheat 12 and 15% moisture single kernel hardness index response to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	259.307	129.654	34.24	
SF	3	92.737	30.912	8.16**	<.001
Temperature	2	2.350	1.175	0.31	0.736
SF x Temperature	6	7.842	1.307	0.35	0.905
Error	22	83.299	3.786		
Total	35	445.535			

** Significant at 0.01%
* Significant at 0.05

Table 6.7. on page 147

Table.6.8. Analysis of variance table of durum wheat 12 and 15% moisture 1000 grain weight response to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.765	0.382	0.15	
SF	3	4.719	1.79	0.162	14.157
Temperature	2	5.027	2.514	0.95	0.392
Moisture	1	2.694	2.694	1.14	0.32
SF x Temperature	6	6	16.190	2.698	1.02
SF x Moisture	3	3	7.960	2.653	1.01
Temperature x Moisture	2	1.777	0.889	0.34	0.715
SF x Temperature x Moisture	6	8.450	1.408	0.53	0.779
Error	46	121.110	2.633		
Total	71	202.130			

** Significant at 0.01%
* Significant at 0.05

Table 6.9. on page 148

Table.6.10. Analysis of variance of durum wheat 12 and 15% moisture for protein content % response to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.1506	0.0753	2.13	
SF	3	0.3031	0.1010	2.86	0.080
Temperature	2	0.0939	0.0470	1.33	0.285
SF x Temperature	6	0.1061	0.0177	0.50	0.801
Error	22	0.7761	0.0353		
Total	35	1.4297			

** Significant at 0.01%
* Significant at 0.05

Table 6.11. on page 149

Table.6.12. Analysis of variance table showing the response of ash content percentage to the fumigation of durum semolina and wheat flour 12 and 15% moisture by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on the percentage of ash content.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.0006	0.0003	0.08	
Commodity	1	26.053	26.053	663**	<.001
SF	3	0.389	0.129	33.11**	<.001
Temperature	2	0.005	0.003	0.73	0.484
Moisture	1	0.00009	0.00009	0.02	0.882
Commodity x SF	3	0.217	0.072	18.43**	<.001
SF x Temperature	2	0.0008	0.0004	0.11	0.899
Commodity x Temperature	6	0.009	0.0015	0.38	0.891
Commodity x Moisture	1	0.004	0.004	0.97	0.326
Commodity x SF x Temperature	3	0.025	0.008	2.14	0.100
Temperature x Moisture	2	0.011	0.0056	1.42	0.247
SF x Moisture	6	0.011	0.002	0.46	0.836
Commodity x SF x Moisture	3	0.015	0.005	1.29	0.283
Commodity x Temperature x Moisture	2	0.004	0.002	0.52	0.597
SF x Temperature x Moisture	6	0.019	0.003	0.81	0.567
Commodity x SF x Temperature x Moisture	6	0.020	0.003	0.86	0.529
Error	94	0.369	0.004		
Total	143	27.155			

** Significant at 0.01%
* Significant at 0.05

Table 6.13. on page 150

Table.6.5. Hectolitre weight Kg/hL response of durum wheat 12 and 15% moisture to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Same letter refers to non-significant differences.

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
Moisture content	12%	82.1a	82.1a	82.3a	82.1a	82.3a	82.2a	82.4a	82.1a	82.2a	82.4a	82.4a	82.2a	2.6
	15%	82.3a	80.0a	79.4a	79.7a	82.2a	80.0a	79.5a	79.7a	82.3a	79.7a	79.7a	79.7a	
Temperature X SF		82.2a	81.1a	80.9a	80.9a	82.2a	81.1a	80.9a	80.9a	82.3a	81.1a	81.0a	81.0a	1.5
Temperature means		81.2a				81.3a				81.3a				0.2

SF mg/L X Moisture	0		4.167		8.928		31.25		LSD 5%
	12%	15%	12%	15%	12%	15%	12%	15%	
	82.2a	82.3a	82.2a	79.9b	82.3a	79.5c	82.1a	79.7bc	0.3
SF mg/L means	82.2a		81.1b		80.9b		80.9b		0.2

Moisture X Temperature	12%			15%			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	82.1a	82.2a	82.3a	80.4b	80.4b	80.4b	0.3

Moisture means	12%	15%	LSD 5%
	82.2a	80.40b	0.1

Different letters refer to significant differences.

Table.6.7. Single kernel hardness index of durum wheat 12% moisture response to the fumigation SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	95.25a	95.07a	94.46a	94.93b
4.167	99.19a	99.29a	97.71a	98.73a
8.928	98.81a	97.95a	99.32a	98.69a
31.25	98.54a	98.92a	97.90a	98.45a
Mean	97.94a	97.81a	97.35a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 1.90.
LSD value for temperature was 1.64.
LSD value for SF x temperature was 3.29.
Moisture content did not include in the test as the kernel hardness index was obtained based on 14% moisture.

Table.6.9. 1000 grain weight of durum wheat 12 and 15% moisture response to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
Moisture content	12%	42.41	41.53	41.41	42.17	42.28	41.56	41.83	41.81	42.96	41.51	40.99	41.93	2.66
	15%	43.73	41.01	42.53	43.40	41.84	41.92	42.91	42.92	43.09	43.20	42.76	41.69	
Temperature X SF		42.07a	41.27a	41.97a	42.79a	42.06a	41.24a	41.87a	41.37a	42.03a	42.35a	41.87a	40.81a	1.88
Temperature means		42.28a				41.63a				42.02a				0.94

SF mg/L X Moisture	0		4.167		8.928		31.25		LSD 5%
	12%	15%	12%	15%	12%	15%	12%	15%	
	42.55a	42.89a	41.20a	42.04a	41.08a	42.73a	41.64a	42.67a	1.54
SF mg/L means	42.72a		41.62a		41.90a		41.66a		1.18

Moisture X Temperature	12%			15%			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	41.88a	41.87a	41.35a	42.37a	42.40a	42.69a	1.33
Moisture means	41.37a			41.58a			0.77

Different letters refer to significant differences
Same letter refers to non-significant differences.

Table.6.11. Protein content% of durum wheat response to the fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	13.10a	13.10a	13.26a	13.15a
4.167	13.30a	13.06a	13.16a	13.17a
8.928	13.23a	13.13a	13.23a	13.20a
31.25	13.43a	13.40a	13.20a	13.34a
Mean	13.26a	13.17a	13.21a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 0.18.
LSD value for temperature was 0.15.
LSD value for SF x temperature was 0.31.
Moisture content did not include in the test as the protein content was obtained based on 14% moisture.

Table.6.13. Influence of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on the percentage of ash content in durum semolina (S) and wheat flour (F) at 12 and 15% moisture contents.

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
commodity	F	1.91a	1.73a	1.81a	1.70a	1.95a	1.71a	1.79a	1.72a	1.92a	1.69a	1.80a	1.69a	0.07
	S	0.97a	0.93a	0.93a	0.94a	0.97a	0.94a	0.91a	0.91a	0.93a	0.92a	0.92a	0.93a	
Temperature X SF		1.44a	1.33a	1.37a	1.32a	1.46a	1.32a	1.35a	1.31a	1.43a	1.30a	1.36a	1.31a	0.05
Temperature means		1.37a				1.36a				1.35a				0.02

SF mg/L X Commodity	0		4.167		8.928		31.25		LSD 5%
	F	S	F	S	F	S	F	S	
	1.93a	0.96e	1.71d	0.9304e	1.80b	0.92e	1.70d	0.93e	0.04
SF mg/L means	1.44a		1.32c		1.36b		1.31c		0.02

Commodity X Temperature	F			S			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	1.79b	1.79b	1.77b	0.94a	0.93a	0.93a	0.03
Commodity means	1.78a			0.93b			0.02

Different letters refer to significant differences.
Same letter refers to non-significant differences.

6.3.3. Effects of SF fumigation on semolina quality traits of durum wheat

The impact of fumigation under different conditions on pasta brightness was tested. Semolina milled from durum wheat was brighter than commercially supplied semolina and this accounted for the significant semolina affect in the ANOVA (Table 14). No other factors associated with the fumigation or SF itself affected semolina brightness (Table 14). The value of the brightness of milled semolina was 86.50 and 81.83 for commercially supplied semolina (Table 15). In addition, the red-greenness colour for semolina was affected significantly by the commodity and the interaction between the commodity and SF (Tables 16; 17). However, the effect of the other factors and their combinations were not significant. The greenness of milled semolina was less than in commercially supplied, and the highest greenness was at the combination of (control x commercially supplied semolina). Moreover, fumigation with SF and semolina type and the interaction between them affected the yellowness of semolina significantly (Tables, 18). The yellowness of commercially supplied semolina was higher than that milled from fumigated durum. Results indicated a yellowness value for commercial semolina of 28.31, and this recorded a significant difference with milled durum 21.48 (Table 19). In addition, the yellowness of semolina was the highest with fumigation of 4.167 mg/L. However, the other SF concentrations did not show any significant differences from the control and between each other.

Mixograph: Maximum peak time was affected significantly by semolina type, SF, SF x semolina and SF x temperature (Table 20). Commercial semolina took longer than that laboratory milled from durum to reach maximum peak. In addition, fumigation by SF decreased time for maximum peak, and the lowest time was at SF 31.25 mg/L. However, control did not show a significant difference in MPT with the other SF concentrations. The highest maximum peak time was 5.1 min (Table 21) at the combination of (control x commercial semolina). However, the lowest was 3.9 at the combination of (control x milled semolina). The interaction between temperature and semolina type recorded 4.9 min at (25°C x commercial semolina) and 4.210 min at (35°C x milled semolina).

Both resistance breakdown and width of mixogram at peak time did not show any significant responses to the impacts of the semolina type, SF, temperature and their combinations (Tables 22 - 25).

Table.6.14. Analysis of variance table for influencing of fumigation on semolina brightness (L*) for commercially supplied semolina and milled from durum semolina by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.9711	0.4856	0.52	
Semolina	1	393.120	393.120	421.89**	<.001
SF	3	6.084	2.028	2.18	0.104
temperature	2	1.303	0.652	0.70	0.502
Semolina x SF	3	6.139	2.046	2.20	0.101
Semolina x Temperature	2	1.676	0.838	0.90	0.414
SF x Temperature	6	7.484	1.247	1.34	0.260
SF x Temperature x Semolina	6	.9368	0.823	0.88	0.515
Error	46	42.863	0.932		
Total	71	464.579			

** Significant at 0.01%
* Significant at 0.05

Table 6.15. on page 156

Table.6.16. Analysis of variance table for influence of fumigation on semolina redness (a*) for commercially supplied semolina and milled from durum semolina by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.0194	0.009	1.10	
Semolina	1	1.564	1.563	176.81**	<.001
SF	3	0.022	0.007	0.84	0.476
temperature	2	0.012	0.006	0.69	0.506
Semolina x SF	3	0.147	0.049	5.54**	0.002
Semolina x Temperature	2	0.035	0.017	1.96	0.152
SF x Temperature	6	0.090	0.0151	1.71	0.139
SF x Temperature x Semolina	6	0.0516	0.008	0.97	0.453
Error	46	0.407	0.008		
Total	71	2.348			

** Significant at 0.01%
* Significant at 0.05
Data multiplied by -1 before the analysis.

Table 6.17. on page 157

Table.6.18. Analysis of variance table for influencing of fumigation on semolina yellowness (b*) for commercially supplied semolina and milled from durum semolina by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.725	0.362	1.70	
Semolina	1	840.705	840.705	3947.35*	<.001
SF	3	8.521	2.840	13.34**	<.001
temperature	2	0.337	0.167	0.79	0.459
Semolina x SF	3	7.002	2.334	10.96**	<.001
Semolina x Temperature	2	0.732	0.366	1.72	0.191
SF x Temperature	6	0.567	0.094	0.44	0.846
SF x Temperature x Semolina	6	0.353	0.058	0.28	0.945
Error	46	9.797	0.213		
Total	71	868.739			

** Significant at 0.01%
* Significant at 0.05

Table.6.20. Analysis of variance table for influence of fumigation on mixograph maximum peak time for commercially supplied semolina and milled from durum semolina by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	5	1.292	0.258	0.59	
Semolina	1	11.514	11.515	26.07**	<.001
SF	3	4.215	1.405	3.18*	0.027
temperature	2	2.033	1.017	2.30	0.105
Semolina x SF	3	7.258	2.419	5.48**	0.001
Semolina x Temperature	2	5.286	2.643	5.98**	0.003
SF x Temperature	6	2.999	0.499	1.13	0.349
SF x Temperature x Semolina	6	6		3.2081	0.5347
Error	114	50.357	0.442		
Total	142	87.830			

** Significant at 0.01%
* Significant at 0.05

Table 6.19. on page 158

Table.6.22. Analysis of variance table for influence of fumigation on resistance breakdown for commercially supplied semolina and milled from durum semolina by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	5	1027.9	205.6	0.60	
Semolina	1	280.0	280.0	0.82	0.366
SF	3	963.2	321.1	0.94	0.422
temperature	2	64.4	32.2	0.09	0.910
Commodity x SF	3	1856.9	619.0	1.82	0.148
Semolina x Temperature	2	1741.6	870.8	2.56	0.082
SF x Temperature	6	2669.3	444.9	1.31	0.259
SF x Temperature x Semolina	6	814.6	135.8	0.40	0.878
Error	114	38791.0	340.3		
Total	142	48158.5			

** Significant at 0.01%
* Significant at 0.05

Table 6.23. on page 160

Table.6.24. Analysis of variance table for influence of fumigation on width of mixogram at peak time for commercially supplied semolina and milled from durum semolina by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Source of variation	DF	SS	MS	F	P
Replicates	5	90.99	18.20	0.45	
Semolina	1	26.17	26.17	0.65	0.422
SF	3	26.71	8.90	0.22	0.881
temperature	2	11.73	5.87	0.15	0.865
Semolina x SF	3	25.96	8.65	0.22	0.886
Semolina x Temperature	2	195.75	97.87	2.43	0.092
SF x Temperature	6	330.26	55.04	1.37	0.234
SF x Temperature x Semolina	6	487.14	81.19	2.02	0.069
Error	114	4587.68	40.24		
Total	142	5775.82			

** Significant at 0.01%
* Significant at 0.05

Table 6.25. on page 161

Table.6.15. Influence of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on semolina brightness (L*) for the commercially supplied semolina (C) and milled from durum wheat (M).

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
Commodity	C	81.97a	81.50a	81.83a	82.03a	82.12a	81.62a	81.87a	81.81a	81.90a	81.52a	81.86a	81.92a	1.58
	M	87.00a	86.46a	86.62a	86.59a	87.34a	86.74a	86.53a	83.80a	87.10a	86.67a	86.75a	86.44a	
Temperature X SF		84.49a	83.98a	84.23a	84.31a	84.73a	84.18a	84.20a	82.80a	84.50a	84.10a	84.31a	84.18a	1.12
Temperature means		84.25				83.98				84.27				0.56

SF mg/L X Commodity	0		4.167		8.928		31.25		LSD 5%
	C	M	C	M	C	M	C	M	
	82.00a	87.15a	81.55a	86.62a	81.85a	86.63a	81.92a	85.61a	0.91
SF mg/L means	84.57a		84.08a		84.24a		83.77a		0.64

Commodity X Temperature	C			M			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	81.83a	81.85a	81.80a	86.67a	86.10a	86.74a	0.79

Commodity means	C	M	LSD
	81.83b	86.50a	0.45

Different letters refer to significant differences
Same letter refers to non-significant differences.

Table.6.17. Influencing of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on semolina redness (a*) for the commercially supplied semolina (C) and milled from durum semolina (M).

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
commodity	C	2.18a	2.36a	2.24a	2.34a	2.28a	2.29a	2.26a	2.31a	2.26a	2.25a	2.22a	2.31a	0.15
	M	2.62a	2.54a	2.56a	2.61a	2.70a	2.53a	2.51a	2.35a	2.66a	2.59a	2.65a	2.52a	
Temperature X SF		2.40a	2.45a	2.40a	2.47a	2.49a	2.41a	2.38a	2.33a	2.46a	2.42a	2.44a	2.41a	0.10
Temperature means		2.43a				2.40a				2.43a				0.05

SF mg/L X Commodity	0		4.167		8.928		31.25		LSD 5%
	C	M	C	M	C	M	C	M	
	2.24c	2.66a	2.30c	2.55b	2.24c	2.57b	2.32c	2.49b	0.09
SF mg/L means	2.45a		2.43a		2.41a		2.41a		0.06

Commodity X Temperature	C			M			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	2.28a	2.28a	2.26a	2.58a	2.52a	2.60a	0.08

Commodity means	C	M	LSD 5%
	2.28b	2.57a	0.04

Different letters refer to significant differences.
Same letter refers to non-significant differences.
Data multiplied by -1 before the analysis.

Table.6.19. Influencing of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on semolina yellowness (b*) for the commercially supplied semolina (C) and milled from durum semolina (M).

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
Semolina	C	27.53a	29.55a	28.00a	28.34a	27.77a	29.30a	28.02a	28.37a	27.58a	29.24a	27.88a	28.16a	0.76
	M	21.44a	21.74a	21.47a	21.74a	21.23a	21.32a	21.55a	20.92a	21.64a	21.61a	21.68a	21.40a	
Temperature X SF		24.48a	25.65a	24.74a	25.04a	24.50a	25.31a	24.78a	24.64a	24.61a	25.43a	24.78a	24.78a	0.54
Temperature means		24.98a				24.81a				24.90a				0.27

SF mg/L X Semolina	0		4.167		8.928		31.25		LSD 5%
	C	M	C	M	C	M	C	M	
	27.63c	21.44d	29.36a	21.56d	27.97bc	21.56d	28.29b	21.35d	0.43
SF mg/L means	24.53b		25.46a		24.77b		24.82b		0.31

Semolina type X Temperature	C			M			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	28.35a	28.36a	28.22a	21.60a	21.26a	21.6a	0.38
Semolina	28.31a			21.48b			0.22

Different letters refer to significant differences
Same letter refers to non-significant differences

Table.6.21. Influence of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on mixograph peak time (min) for the commercially supplied semolina (C) and milled from durum wheat (M).

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
semolina	C	4.3a	4.3a	4.4a	4.0a	5.6a	4.4a	5.3a	4.5a	5.5a	4.3a	4.9a	4.4a	1.7
	M	4.1a	4.4a	3.8a	4.3a	3.5a	4.3a	4.3a	3.7a	4.0a	4.3a	4.5a	3.9a	
Temperature X SF		4.2a	4.3a	4.1a	4.1a	4.6a	4.3a	4.8a	4.1a	4.7a	4.3a	4.8a	4.2a	0.5
Temperature means		4.2a				4.5a				4.5a				0.27

SF mg/L X semolina	0		4.167		8.928		31.25		LSD 5%
	C	M	C	M	C	M	C	M	
	5.1a	3.9d	4.4c	4.3c	4.9b	4.3c	4.3c	4.0cd	0.4
SF mg/L means	4.5a		4.3a		4.6a		4.1b		0.3

semolina X Temperature	C			M			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	4.3b	4.9a	4.8a	4.2b	3.9b	4.2b	0.4
semolina means	4.7a			4.1b			0.2

Different letters refer to significant differences
Same letter refers to non-significant differences.

Table.6.23. Influence of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on resistance breakdown for the commercially supplied semolina (C) and milled from durum semolina (M).

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
semolina	C	33a	35a	48a	44a	28a	38a	26a	43a	28a	41a	36a	54a	21
	M	33a	35a	39a	22a	45a	38a	31a	49a	33a	35a	31a	30a	
Temperature X SF		33a	35a	43a	33a	36a	38a	29a	46a	30a	38a	33a	42a	15
Temperature means		36a				37a				36a				7

SF mg/L X semolina	0		4.167		8.928		31.25		LSD 5%
	C	M	C	M	C	M	C	M	
	30a	37a	38a	36a	37a	34a	47a	34a	12
SF mg/L means	33a		37a		36a		40a		9

semolina X Temperature	C			M			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	40a	34a	40a	32a	41a	32a	11
semolina means	38a			35a			6

Different letters refer to significant differences
Same letter refers to non-significant differences

Table.6.25. Influence of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C on width of mixogram at peak time for the commercially supplied semolina (C) and milled from durum semolina (M).

Temperature		15°C				25°C				35°C				LSD 5%
SF mg/L		0	4.167	8.928	31.25	0	4.167	8.928	31.25	0	4.167	8.928	31.25	
semolina	C	19.2a	16.3a	16.7a	15.0a	14.1a	13.3a	18.0a	15.4a	13.2a	18.4a	15.6a	21.9a	7.3
	M	16.2a	16.9a	20.3a	13.3a	20.4a	18.1a	14.1a	24.5a	16.5a	15.7a	15.9a	15.4a	
Temperature X SF		17.7a	16.6a	18.4a	14.1a	17.3a	15.7a	16.0a	19.9a	14.8a	17.0a	15.7a	18.6a	5.1
Temperature means		16.7a				17.2a				16.5a				2.5

SF mg/L X semolina	0		4.167		8.928		31.25		LSD 5%
	C	M	C	M	C	M	C	M	
	15.5aa	17.7a	16.0a	16.9a	16.7a	16.7a	17.4a	17.7a	4.2
SF mg/L means	16.6a		16.5a		16.7a		17.5a		3.0

semolina X Temperature	C			M			LSD 5%
	15°C	25°C	35°C	15°C	25°C	35°C	
	16.8a	15.2a	17.3a	16.7a	19.3a	15.9a	3.6
Commodity means	16.4a			17.3a			2.1

Different letters refer to significant differences
Same letter refers to non-significant differences.

6.3.4. Effects of SF fumigation on pasta quality traits of durum wheat

Pasta cooking properties: Optimum cooking time and water absorption did not show any significant response to fumigation by SF, storage temperature and the combination between SF and temperature (Tables 26 - 29). However, cooking loss was affected significantly by SF and the interaction between SF and temperature. The highest loss (8.6 g) was recorded at (35°C x 31.25 mg/L) (Tables 30; 31). CL seemed to increase with dose of SF but this was not consistent so at 15°C CL was lower at SF 31.25 mg/L than in control but higher at SF 4.167 mg/L than control whereas at 35°C an opposite trend was noted.

Firmness force 1 was affected significantly by storage temperature and the interaction between temperature and SF (Table 32). Storage temperature of 35°C did not show significant difference from 25°C. However, 15°C showed significantly lower firmness than the other temperatures. The combination of 35°C x 4.167 mg/L showed the highest firmness force (Table 33). The changes in firmness with SF dose were inconsistent because of the SF x T interaction. For the firmness area 1:2 g/sec only the combination effect of SF and temperature impacted the area significantly, and the highest area was recorded at the combination of 35°C x 4.167 mg/L (Tables 34; 35). There was no difference in OC tolerance across temperature for the control but this increased for the SF 4.167 mg/L but not for SF 31.25 mg/L, so no clear trend (Tables 36; 37). SF and increasing the dose again did not show a consistent trend due to the SF x T interaction but the mean values with fumigation were significantly higher than the control. This showed that the pasta was less able to resist loss in firmness following a period (10min) of overcooking, which is undesirable. A lower value is better as the closer the firmness is after 10min over cooking to the OCT firmness, the more tolerance.

Stickiness force was affected significantly by the fumigant SF but not temperature and the combination of the two factors (Table 38). The concentration 4.167 mg/L caused the highest stickiness force and did not show significant difference from 31.25 mg/L. However, the control showed significantly lower stickiness compared to SF concentrations (Table 39). Temperature had no effect on stickiness force. Stickiness area 1:2 was affected significantly by SF and temperature but not the combination effect of the two factors (Table 40). Stickiness area 1:2 was 13.2 g/sec at the concentration 4.167 mg/L and showed significant differences comparing to control and 31.25 mg/L. However, the difference between the control and the 31.25 mg/L concentration was not significant (Table 41).

Cooking pasta affected the brightness significantly, for example, at 4.167 mg/L and 25°C, L* for uncooked pasta was 67.2 and was 76.4 for cooked. The same results were noticed for a* and b* (Table 42).

Table.6.26. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta optimum cooking time.

Source of variation	DF	SS	MS	F	P
Replicates	1	0	0	0	0
SF	2	0.218	0.109	0	0
Temperature	2	0.871	0.435	0	0
SF x Temperature	4	0.436	0.109	0	0
Error	8	0.000	0.000		0
Total	17	1.524			0

** Significant at 0.01%
* Significant at 0.05

Table.6.27. Optimum cooking time of pasta made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	12.0a	11.3a	12.0a	11.8a
4.167	12.0a	12.0a	12.0a	12.0a
31.25	12.0a	11.3a	12.0a	11.8a
Mean	12.0a	11.5a	12.0a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was not calculated able.
LSD value for temperature was not calculated able.
LSD value for SF x temperature was not calculate able.

Table.6.28. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta water absorption.

Source of variation	DF	SS	MS	F	P
Replicates	1	6.42	6.42	0.59	
SF	2	90.16	45.08	4.12	0.059
Temperature	2	3.92	1.96	0.18	0.839
SF x Temperature	4	53.99	13.50	1.23	0.369
Error	8	87.45	10.93		
Total	17	241.94			

** Significant at 0.01%
* Significant at 0.05

Table.6.29. Water absorption of pasta made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	157.7a	158.1a	156.9a	157.5a
4.167	153.2a	157.2a	155.6a	155.4a
31.25	155.7a	149.9a	150.7a	152.1a
Mean	155.5a	155.1a	154.4a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 4.402.
LSD value for temperature was 4.402.
LSD value for SF x temperature was 7.624.

Table.6.30. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta cooking loss.

Source of variation	DF	SS	MS	F	P
Replicates	1	0.062	0.062	0.19	
SF	2	4.095	2.048	6.47*	0.021
Temperature	2	0.052	0.026	0.08	0.922
SF x Temperature	4	20.472	5.119	16.18**	<.001
Error	8	2.531	0.316		
Total	17	27.213			

** Significant at 0.01%
* Significant at 0.05

Table.6.31. Cooking loss g of pasta made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	6.8bc	5.9cd	6.0c	6.2b
4.167	8.0ab	6.2c	5.6cd	6.6b
31.25	5.3d	8.3a	8.6a	7.4a
Mean	6.7a	6.8a	6.7a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 0.749.
LSD value for temperature was 0.749.
LSD value for SF x temperature was 1.297.

Table.6.32. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta firmness force 1 g.

Source of variation	DF	SS	MS	F	P
Replicates	3	901.1	300.4	1.21	
SF	2	1534.5	767.3	3.09	0.064
Temperature	2	4949.4	2474.7	9.96**	<.001
SF x Temperature	4	9916.6	2479.2	9.97**	<.001
Error	24	5965.8	248.6		
Total	35	23267.4			

** Significant at 0.01%
* Significant at 0.05

Table.6.33. Firmness force 1 g of pasta made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	572b	577b	561bc	570a
4.167	548c	567bc	610a	575a
31.25	534c	590ab	553c	559a
Mean	552b	578a	575a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 14.
LSD value for temperature was 14.
LSD value for SF x temperature was 23.

Table.6.34. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta firmness area 1:2 g/sec.

Source of variation	DF	SS	MS	F	P
Replicates	3	250.98	83.66	1.67	
SF	2	62.52	31.26	0.62	0.544
Temperature	2	196.86	98.43	1.97	0.162
SF x Temperature	4	1332.13	333.03	6.65**	<.001
Error	24	1201.37	50.06		
Total	35	3043.86			

** Significant at 0.01%
* Significant at 0.05

Table.6.35. Firmness area 1:2 g/sec of pasta made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	191ab	183b	181b	185a
4.167	177b	185b	197a	186a
31.25	176b	192ab	181b	183a
Mean	182a	187a	186a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 6.
LSD value for temperature was 6.
LSD value for SF x temperature was 11.

Table.6.36. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta over cooking tolerance.

Source of variation	DF	SS	MS	F	P
Replicates	3	7.131	2.377	0.38	
SF	2	368.550	184.275	29.72**	<.001
Temperature	2	56.805	28.403	4.58*	0.021
SF x Temperature	4	446.983	111.746	18.02**	<.001
Error	24	148.823	6.201		
Total	35	1028.293			

** Significant at 0.01%
* Significant at 0.05

Table.6.37. Over cooking tolerance for pasta made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	35.8d	39.1c	35.0d	36.7c
4.167	37.0c	44.7b	51.4a	44.4a
31.25	44.6b	40.5c	39.7c	41.6b
Mean	39.1b	41.4a	42.0a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 2.1.
LSD value for temperature was 2.1.
LSD value for SF x temperature was 3.6.

Table.6.38. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta stickiness force.

Source of variation	DF	SS	MS	F	P
Replicates	2	0.347	0.174	0.08	
SF	2	27.461	13.730	6.08*	0.011
Temperature	2	2.494	1.247	0.55	0.586
SF x Temperature	4	25.995	6.499	2.88	0.057
Error	16	36.113	2.257		
Total	26	92.410			

** Significant at 0.01%
* Significant at 0.05

Table.6.39. Pasta stickiness force made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	20.4a	22.2a	20.9a	21.2b
4.167	22.1a	24.8a	23.5a	23.5a
31.25	24.7a	22.0a	22.5a	23.1a
Mean	22.4a	23.0a	22.3a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 1.5.
LSD value for temperature was 1.5.
LSD value for SF x temperature was 2.6.

Table.6.40. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360 and 31.25 mg/L x 48 h) at 15, 25 and 35°C on pasta stickiness area 1:2 g/sec.

Source of variation	DF	SS	MS	F	P
Replicates	2	1.434	0.717	0.54	
SF	2	19.146	9.5573	7.25**	0.006
Temperature	2	9.702	4.8851	3.68*	0.049
SF x Temperature	4	11.534	2.884	2.18	0.117
Error	16	21.117	1.320		
Total	26	62.933			

** Significant at 0.01%
* Significant at 0.05

Table.6.41. Area 1:2 g/sec of pasta stickiness made from semolina fumigated by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 15, 25 and 35°C.

Temperature°C / SF mg/L	15	25	35	Mean
0	11.4a	10.6a	11.6a	11.2b
4.167	13.3a	13.2a	12.9a	13.2a
31.25	13.7a	10.5a	10.9a	11.7b
Mean	12.8a	11.4a	11.8a	

Different letters refer to significant differences.
Same letter refers to non-significant differences.
LSD value for SF was 1.2.
LSD value for temperature was 1.2.
LSD value for SF x temperature was 2.0.

Table.6.42. Uncooked and cooked pasta colour fumigated at different conditions.

SF mg/L	Temperature°C	Moisture%	Uncooked pasta				Cooked pasta			
			L*	a*	b*	WI	L*	a*	b*	WI
0	25	12	66.5	2.3	46.2	-63.4	75.8	-2.3	32.0	-47.9
0	25	12	66.7	2.4	45.2	-62.4	75.6	-2.1	31.0	-45.2
0	35	12	65.9	2.8	44.8	-60.8	75.2	-1.9	31.1	-45.3
0	35	12	66.1	2.8	45.0	-61.3	75.5	-1.8	30.6	-44.2
0	15	12	66.7	2.4	45.5	-62.9	75.9	-2.1	31.3	-46.4
0	15	12	66.6	2.5	44.3	-61.0	75.5	-2.1	30.5	-43.9
4.167	15	12	67.8	1.8	45.9	-65.2	75.9	-2.7	32.2	-48.4
4.167	25	12	67.2	2.0	48.1	-67.3	76.4	-2.9	32.3	-49.1
4.167	35	12	66.4	2.1	46.8	-64.2	75.7	-2.8	32.0	-47.9
31.25	15	12	67.5	2.0	46.3	-65.2	75.8	-2.4	32.8	-49.8
31.25	25	12	66.6	2.1	45.8	-63.2	75.7	-2.5	32.1	-48.1
31.25	35	12	67.7	1.8	45.0	-63.7	75.8	-2.6	32.7	-49.6

6.3.5. Bread

Bread tested characteristics such as loaf volume, appearance, external volume score of bread loaf, internal structure of bread loaf, total score of bread loaf, loaf colour did not show any significant affects from fumigation with SF (Tables 43). The highest loaf volume 430.8 and external volume score 17.2 were at the concentration 31.25 mg/L. However, there was no significant differences from the other concentrations or the control. The lowest internal structure was 4.7 at the concentration 8.928 mg/L and did not show significant differences with other concentrations. The appearance was the same at all the treatments and all the other characteristics showed some variation, but that was not significant (Table 44).

Table.6.44. Effect of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 25°C and 12% moisture content on some characteristics of bread made from durum wheat.

SF mg/L	Loaf volume	appearance	External volume score	External oven spring score	Internal structure	Total score	Crumb colour		
							L*	a*	b*
0	419.2a	12	16.8a	4.2a	5.0a	56.6a	71.8a	3.6a	19.1a
4.167	419.2a	12	16.8a	4.0a	5.0a	56.4a	71.9a	3.5a	18.9a
8.928	421.7a	12	16.9a	4.8a	4.7a	57.1a	71.9a	3.5a	19.0a
31.25	430.8a	12	17.2a	4.1a	5.0a	57.1a	72.0a	3.5a	19.0a
LSD	19.40	0	0.8	2.2	2.4	4.5	0.8	0.2	0.2

Different letters refer to significant differences.
Same letter refers to non-significant differences.

Table.6.43. Analysis of variance table for influence of fumigation by SF (control, 4.167 mg/L x 360, 8.928 mg/L x 168 h and 31.25 mg/L x 48 h) at 25°C and 12% moisture content on durum bread loaf.

Volume					
Source of variation	DF	SS	MS	F	P
	2	201.04	100.52	1.07	
SF	3	276.56	92.19	0.98	0.463
Error	6	565.62	94.27		
Total	11	1043.23			
Appearance					
Replicates	2	0	0	0	
SF	3	0	0	0	0
Error	6	0	0		
Total	11	0	0		
External volume score					
Replicate	2	0.322	0.160	1.07	
SF	3	0.443	0.147	0.98ns	0.463
Error	6	0.905	0.150		
Total	11	1.669			
Internal structure					
Replicates	2	0.167	0.083	0.06	
SF	3	0.250	0.083	0.06	0.980
Error	6	8.500	1.417		
Total	11	8.917			
Total score					
Replicates	2	6.816	3.408	0.67	
SF	3	1.060	0.353	0.07ns	0.974
Error	6	30.348	5.058		
Total	11	38.224			
Brightness					
Replicates	2	0.156	0.078	0.53	
SF	3	0.063	0.021	0.14	0.930
Error	6	0.880	0.147		
Total	11	1.099			
Redness					
Replicates	2	0.012	0.006	0.70	
SF	3	0.019	0.006	0.72	0.576
Error	6	0.053	0.009		
Total	11	0.084			
Yellowness					
Replicates	2	0.077	0.038	3.03	
SF	3	0.036	0.012	0.96	0.469
Error	6	0.076	0.013		
Total	11	0.188			

6.3.6. Fluoride residues

Fluoride residues in durum grain of 12% moisture content fumigated with 1 mg/L for 168 h at 25°C increased with increasing filling ratio (Table 45). Moreover, the quantified fluoride residues from fumigating durum wheat of 12% moisture content with 31.25 mg/L SF for 48 h at 15, 25 and 35°C showed differences due to the impact of storage temperature on increasing the sorption and then the residues (Table 46). For example, the residues at 15, 25 and 35°C were 7.4, 9.9 and 13 for the fumigated wheat. These are the background F content of the wheat and are not affected by storage T but do increase when SF is applied above 15°C.

Fluoride residues in milled semolina and the bran derived from milling the durum wheat of 12% moisture, fumigated at 25°C with 4.167 mg/L of SF for 360 h, 8.928 mg/L for 168 h and 31.25 mg/L x 48 h increased with increasing concentration of SF. The majority of the quantified amounts of residues were found in bran (Table 47) representing about 75-89% of total residue. For instance, the total fluoride residues in wheat fumigated with 4.167 mg/L was 11.6 mg/kg. However, 9.4 mg/kg was found in the bran, and only 2.2 mg/kg was in the grain. Fumigated semolina of 12% moisture with 8.928 mg/L SF for 168 h at 15, 25 and 35°C showed an increase in fluoride residues with increasing storage temperature. Results in Table 54, 48 showed that fluoride residue at 15°C were 4.4 mg/kg and 7.2 mg/kg at 35°C. The residue level in fumigated semolina was significantly higher than the level in the control (not fumigated). For pasta, the cooking water was investigated for fluoride residues. Results in Table 49 showed the uncooked pasta recorded residues of 2.3 mg/kg, and the residue level increased after cooking and became 6.7 mg/kg. Cooking water from the fumigated pasta had a much higher level of residue (53 mg/kg) than the water from not fumigated pasta (5.3 mg/kg).

Table.6.45. Effect of fumigating different filling ratios of durum wheat 12% moisture content at 25°C by SF 1 mg/L for 168 h on fluoride residues.

Filling ratio	Fluoride residue mg/kg
0.50	6.8
0.75	7.5
0.95	9.2

Table.6.46. Fluoride residues from fumigating durum wheat of 12% moisture content with 31.25 mg/L SF for 48 h at 15, 25 and 35°C at the same conditions.

Fumigation conditions (8.928 SF mg/L for 168 h)	Fluoride residue mg/kg in fumigated grain	Conditions
35°C	13	Storage for 48 h 35°C 12% moisture content
25°C	9.9	Storage for 48 h at 25°C 12% moisture content
15°C	7.4	Storage for 48 h at 15°C 12% moisture content

Table.6.47. Fluoride residues in milled semolina and the bran from durum wheat fumigated with 4.167 mg/L for 360 h at 25°C and 12% moisture content, 8.298 mg/L for 168 h 25°C and 12% moisture content and 31.25 mg/L x 48 h at 35°C and 12% moisture.

Fumigation conditions	Fluoride residue mg/kg in milled semolina	Fluoride residue mg/kg in bran
4.167 SF mg/L for 360 h at 25°C 12% moisture content	2.2	9.4
8.928 SF mg/L for 168 h at 25°C 12% moisture content	3.3	10
31.25 SF mg/L for 168 h at 35°C 12% moisture content	3.9	33

Table.6.48. Fluoride residues in commercially supplied semolina of 12% moisture content fumigated with 8.298 mg/L for 168 h at 15, 25 and 35°C and control non-fumigated commercially supplied semolina at the same conditions.

Fumigation conditions	Fluoride residue mg/kg in fumigated semolina	Fluoride residue mg/kg in control non-fumigated semolina	Difference of fluoride residues mg/kg
8.298 SF mg/L for 168 h at 15°C 12% moisture content	4.4	1.1	3.3
8.928 SF mg/L for 168 h at 25°C 12% moisture content	6.1	0.2	5.9
8.298 SF mg/L for 168 h at 35°C 12% moisture content	7.2	0.4	6.8

Table.6.49. Fluoride residues in uncooked pasta, cooked pasta and water from cooked pasta made semolina milled from durum wheat fumigated with 8.298 mg/L for 168 h at 25°C and 12% moisture, and control of cooked pasta and its water at the same conditions but not fumigated.

sample	Fumigation conditions	Fluoride residues mg/kg
Fumigated Uncooked pasta	8.928 mg/L for 168 h at 25°C and 12% moisture content	2.3
Fumigated cooked pasta	8.928 mg/L for 168 h at 25°C and 12% moisture content	6.7
Water from fumigated and cooked pasta	8.928 mg/L for 168 h at 25°C and 12% moisture content	53
Control not fumigated and cooked pasta	stored for 168 h at 25°C and 12% moisture content	0.4
Control water from not fumigated and cooked pasta	stored for 168 h at 25°C and 12% moisture content	5.3

6.4. Discussion

The aim of this study was to investigate the impact of fumigation with SF under various conditions (dose, temperature, commodity moisture content) on potential changes in grain germination ability, grain, semolina and pasta and bread quality. In addition, the amount of the main residue from SF fumigation, fluoride (F) has been measured in various fractions to understand where F is mainly located and how SF conditions affect the residue content.

6.4.1. Germination

Germination was defined by Black, (1970) as seeing the first sign of growth for example the appearance of a protruding radical. High quality seed for sowing is important for reliable plant growth and yield. Fumigation should not decrease germination ability and vigour of seedlings. Therefore, fumigation should not affect the germination ability of seeds. The results of this study (Tables 2 and 3) showed that the reduction of the germination capability of durum wheat was greatly affected by the SF treatment and interactions between SF and fumigation conditions of temperature and moisture. Onion seed germination was affected by fumigation with different concentrations of methyl bromide (Powell, 1975) and a significant reduction up to 90% was recorded in germination. Howe, (1934) noticed that adding superphosphate fertilizer reduced the germination of corn significantly up to 90% as well, This affect was thought to be due to the presence of fluorine since germination was unaffected using the same fertiliser without fluorine. Applying higher dosages of methyl bromide than the commercial dose decreased the germination of wheat significantly Minett et al., (1976). The results of this study showed increasing SF concentration reduced germination by 99% even with the hardly extreme storage and fumigation conditions of 15% moisture and 35°C.

Results of this study with SF are in agreement with the results found by Powell, (1975) and Howe, (1934) that recorded a reduction of germination up to 90% for onion and corn due to applying methyl bromide and the fertilizer that has soluble fluorine, respectively. Applying fertiliser that has no fluorine in it did not change corn percentage of germination. Therefore, it was proved that fluorine was the main reason for germination reduction. Presumably, the main reason for the reduction in germination is that fluoride ion is toxic and the fluoride residue that was quantified proved the presence of fluoride ion in the grain affecting germination of such grain. In addition, Meikle (1964) confirmed that sulfuryl fluoride is sorbed by the grain, and it is broken down into sulfate and fluoride that reacts with amino acids . This is likely to inhibit germination.

The germination process starts with imbibition and the required energy for germination comes from the mitochondria in the cell, (Bewley et al., 2013). Possibly fluoride ion causes reduction of germination by interrupting the Kreb's glycolysis cycle interfering with energy production. Thus, the required energy from the seed cell's mitochondria is interrupted. In addition, repairing DNA, proteins and growth hormone synthesis can also be interrupted. The reduction in ATP energy supply leads to failure of seed germination. The significant impact of temperature and moisture on germination percentage related to increased sorption of the fumigant at higher temperature and moisture conditions leading to more residues.

Unlike sulfuryl fluoride and methyl bromide, phosphine was reported to not affect wheat germination significantly. Fumigating wheat grains 12% moisture content with phosphine concentration of 32 g/ton and storage for one year did not affect germination percentage significantly (Bakheit et al 1985). In addition, Krishnasamy and Seshu, (1990) noticed that fumigating rice with a recommended concentration of 3 mg/L of phosphine did not influence germination after fumigation directly or after nine months of storage. The results of this study showed that storage time did not affect the germination percentage, as the reduction of control (not fumigated) was around 10% even though the storage temperature and moisture content were high. Therefore, the main reason of the germination reduction in my study with SF is its toxicity to the grain. Storage conditions have different effects on germination. AL-Yahya, 2001 found that storage of wheat grains at different temperatures and moistures for short to long storage times did not affect the germination significantly except after 172 days. In another study done by Sawant et al., (2012), a reduction in germination of about 8% to 40% was recorded at the end of storage time and the reason was reported to be using different storage systems. However, moisture and temperature of storage did not affect the germination for each storage system. Using different storage systems may affect the germination percentages due to the different conditions of each system. However, using the same system does not change the germination significantly as it is proved by this study and the other studies.

6.4.2. Effect of SF on grain quality traits

The effect of SF on grain quality characteristics showed a significant decrease in hectolitre weight but the change was only minor and is most likely related to the moisture difference between control and SF treated wheat as there was a significant SF x M interaction. Fumigation increased grain hardness compared to control but no difference in SKHI between the 3 SF doses. There is no clear explanation for this phenomena.

Ash content has a relationship with both grain hectolitre weight and hardness. Smaller grain can increase hardness and proportionally more bran/endosperm therefore ash in semolina milled from such grain will increase. However, grain size was not affected although density was (hectolitre weight). No change in thousand-grain weight and a very small decrease in HLW did not affect protein content of grain.

Ash content of semolina and flour showed a significant response to commodity, SF and the interaction between the commodity and SF. The differentiation between semolina and flour in ash content may be related to the differentiation in wheat type that has been milled. In addition, differences in milling procedure between laboratory and commercial milling will affect ash content. Hard wheat is milled intensely to create flour and this results in more fine bran which contaminates the flour and increases ash, whereas the coarser semolina particles result in proportionately less ash in the semolina. Ash content was higher in control compared to the other SF concentrations. The effect of SF on ash content could be due to reacting the inorganic ions of fluoride and sulfate with the proteins and the minerals of the grain and converting them to more volatile elements that reduce the ash percentage in the grain.

6.4.3. Effect of SF on semolina and pasta technological properties

The brightness, redness and yellowness in milled semolina were higher than in commercially supplied semolina (Tables 14 - 19). This may be because of the way milling or because commercial milling semolina can more effectively remove bran from the semolina compared to laboratory milling and this would change the colour characteristics of semolina. Fumigation had no effect on semolina L* and a* and the effect on b* was inconsistent showing a slight increase in commercial semolina but not laboratory milled (M) semolina with SF dose. There were no differences between the SF treated semolina so a clear explanation for what is happen is not unknown.

The SF concentration only had a minor impact on MPT (mixograph peak time) which only decreased at the highest dose compared to the control. The interaction with temperature affected MPT but the changes in practice are insignificant suggesting no change in dough strength. This is supported by no effect of SF on RBD (resistance breakdown) and MPH (mixogram peak height).

For pasta, cooking loss (Tables 30; 31) and over cooking tolerance (Tables 36; 37) increased due to sorption of SF with an interaction with temperature Inconsistent results were obtained with a decrease in CL at 15°C with SF 31.25 vs. control but increases at the other higher temperatures (Table 31). In practice, these increases were relatively large and represent a negative impact on pasta eating quality although it has been found that CL needs to exceed 8% to be detrimental to eating quality. The increased CL could be coming from the additional fluoride residue with higher doses of SF leaching from the pasta into the cooking water.

SF had no effect on cooked pasta firmness (force and area 1:2) (Tables 32 - 35) although there were small but significant increases in the overcooking tolerance values which means that the fumigated grain made pasta that had a tendency to lose its firmness more with overcooking than the control. Firmness means were higher at 25/35°C than at 15°C. This could be related to changes in the protein structure of the glutenins forming when dough is made affecting pasta firmness. Dough strength affects pasta firmness and storage temperature and could alter protein structure.

Stickiness force and area 1:2 increased due to the fumigation with SF (Tables 38 - 41) but the differences failed to be significant at individual temperatures and only overall comparison between SF and control for peak height. Stickiness area seemed to be more affected by SF showing higher significance but only on the overall mean and at SF 4.167 mg/L and not SF, 31.25 mg/L. The increased CL could contribute to an increase in force of adhesion during the compression testing with the texture analyser used to measure stickiness peak area. The loss of starch during cooking would lead to accumulation of amylose on the pasta surface.

Optimum cooking time, water absorption and cooked and uncooked pasta colour did not show any significant differences.

6.4.3. Bread

Fumigation with SF at different concentrations did not affect any of the measured bread characteristics. The lack of any significant changes in semolina dough characteristics would help explain why key features of bread quality, like loaf volume were not affected. Clearly, the F residue also had no impact on yeast fermentation or appearance of the flour so that bread visual score was not affected (Tables 43; 44). This is good news for bakers using fumigated durum wheat, and same baking results could be obtained with bread wheat. The results of this study came in conformity with the results by Orth et al.,(1977); Matthews et al., (1970); Shepard and Buzicky, (1939); Hermitte and Shellenb, (1947). Applying the commercial dosages of methyl bromide directly as a fumigant to wheat and flour did not change the baked bread characteristics either when the flour was milled from grain or the flour fumigated directly was used. Water absorption, dough breakdown, extensibility and loaf volume for wheat and flour were not affected significantly by the fumigation. Fumigated flour samples showed a slight difference compared with control in extensogragh maximum resistance measurement. Hence, fumigating wheat grains with dosages higher than the commercial application by four to twenty times increased maximum resistance to extension and decreased loaf volume for both soft and hard wheat samples significantly (Minett et al, 1976). Unlike methyl bromide, when the concentration of the applied SF was increased up to 31.25 mg/L the tested characteristics of baked bread did not show any significant differences and that suggests that that SF has an advantage which is not affecting the baked bread even when the concentration is increased.

6.4.4. Fluoride residues

Fluoride residues increased significantly in durum wheat grain with increasing filling ratio (Table 45). Fluoride residue at 0.50 filling ratio was 6.5 and became 9.2 mg/L at 0.95 filling ratio. This increase in fluoride residue with filling ratio is due to the increase of SF sorption rate with increasing the level of grain in the storage system (chapter 4). Australian Pesticide and Veterinary Medicines Authority (APVMA, 2007) published an evaluation for the suitability of sulfuryl fluoride to be used as a fumigant in Australia. That evaluation included an assessment of fluoride residues in Australian wheat after single and repeated fumigation. Different batches of different types of Australian wheat were fumigated with SF 1500 mg.h/L. There was a variation in the quantified amounts of fluoride residues, and the range was between 0.5 - 2.1 mg/kg for single fumigation. That means the maximum fluoride residue was 2.1 mg/kg after fumigation with 1500 mg/h/L according to the (APVMA, 2007). The problem with APVMA study was no indication of the filling ratio used, and as the results of this study indicated, filling ratio affects the sorption rate of the fumigant. SF rates increased significantly with increasing the filling ratio (chapter 4). Fluoride residues results in this chapter explained clearly how increasing filling ration increases fluoride residues. Only 0.50 filling ratio showed residues less than the maximum residue limit in Australia.

Moreover, fluoride residues increased in durum grain with increasing storage temperature (Table 46). Temperature during fumigation is an important factor that increases sorption rate of the fumigant by the grain (chapter 2; Hwaidi et al., 2015). Therefore, fluoride residue was 7.4

mg/kg at 15°C and became 13 mg/kg at 35°C. According to the (APVMA) studies, all types of Australian wheat did not hold fluoride residues more than the maximum residue limit and the maximum fluoride residue found was 2.1 mg/kg in cereal grains and 9.66 mg/kg in germ. In my study, SF was applied within an experimental design that contained different factors such as fumigant concentration, exposure time, moisture content, fumigation temperature, different filling ratios, and repeated fumigations. However, the conditions of APVMA study were not clear.

Fluoride residue in milled products (semolina and bran) increased with increasing SF concentration (Table 47) and (chapter 2). However, a significant amount of fluoride residue went to the bran rather than to milled semolina. For example, at the highest concentration SF 31.25 mg/L, the residue was the highest in semolina and bran compared to the other concentrations, and the residue level in semolina was 3.9 mg/kg and in bran, 33 mg/kg, almost 10 fold higher. This gives an indication about the part of the kernel that sorbs most of the SF and makes first contact in the fumigation vessel. Therefore, it helps in understanding and explaining the sorption behaviour of this fumigant. It appears that SF is accumulated extensively in the outer layer of the grain although it can accumulate inside the grain and be bound with protein amino groups (Meikle, 1964). Despite the rapid desorption of the fumigant from the grain, the accumulated residues in outer and inner parts of the grain gives an idea about the strong penetration of the fumigant. The (APVMA, 2007) experiment did not contain milled products such as flour and semolina. The results of this study (chapter 2) showed that the milled products sorbed SF more rapidly than the whole grains. Sorbing high amounts of the fumigant by flour and semolina may leave high levels of residues compared to the whole grains.

Fluoride residues in commercially supplied and fumigated semolina increased with increasing fumigation temperature (Table 48) and (chapter 2). This was due to increasing the sorption rate with increasing temperature. The results of this study showed that making pasta helps to reduce some of the fluoride residue (Table 49) as the pasta was found to have 2.3 mg/kg fluoride compared to semolina fumigated under the same conditions that had 6.1 mg/kg (Table 48. However, since the cooking water contains significant amounts of fluoride it causes an increase in fluoride residue in the cooked pasta. Cooking left high residues of fluoride in the cooking water that normally is drained after finishing the cooking process. This means less residue would be consumed by humans eating the pasta. This study confirms the impact of storage conditions on sorption of the fumigant and then the residues. However, the APVMA study did not indicate detailed conditions of the fumigation such as temperature, moisture exposure time and filling ratio. In addition measuring sorption and the procedure of quantifying the residues was not clear. The fumigated grains were aerated after fumigation in this study, and this study showed there were residues of fluoride with significant amounts due to the fumigation with SF although all the used commodities were aerated. That means aeration helps releasing the residues of SF itself but not fluoride resides as they are probably bound with amino acid groups. These results confirm the results reported by Osbrink et al., (1988) who showed the significant effect of aeration on SF residues but not fluoride residues as SF was desorbed completely after five days of aeration. The strongly bound fluoride residues in food stuffs was reported by Scheffrahn et al., (1989) who showed significant amounts of fluoride residues in different food stuffs although the aeration after fumigation and the left residues depended on the innate characteristics of the commodity.

6.5. Conclusion

The aim of this study was to investigate the impact of fumigation with SF on fluoride residues and the quality of grain, pasta and bread. SF decreases germination significantly especially with increasing fumigation temperature and moisture as germination percentage decreases significantly with the main factors and all of their combinations. Fumigation with SF decreased the hectolitre weight but in practice this is insignificant and is probably most likely due to moisture affects. Ash decreased in both flour and semolina with SF treatment which is a desirable feature. Applying SF at different fumigation conditions did not affect either 1000 grain weight or protein content of the grain.

Fumigation with SF had no impact on semolina colour and dough properties assessed by a mixograph. Pasta cooking loss increased only at the highest SF dose and pasta over cooking tolerance was made worse with SF treatments which are negative aspects of SF fumigation. Some affects were noted in pasta stickiness but these changes were not clear cut and more investigation is warranted. Increasing fumigation temperature increased firmness force 1, but there was no difference between 25 and 35°C. Fumigation at 35°C increased the firmness area 1:2 g/sec that may not be related to SF fumigation. Stickiness force, stickiness area 1:2 increased when SF was applied compared to the control, and increasing fumigation temperature increases both stickiness area 1:2 and stickiness but results are not consistent. Fumigation with SF had no impact on other pasta quality traits (optimum cooking time, water absorption and colour space parameters: L*, a* and b* for uncooked and cooked pasta). Fumigation with SF does not affect bread quality characteristics.

Fluoride residue increased with increasing filling ratio of the grain in the fumigated storage system. Increasing fumigation concentration and temperature increases fluoride residues into the fumigated commodity. The majority of fluoride residues are concentrated in the bran layer and this would be removed on milling. Cooking pasta leads to increasing fluoride residues in the cooking water and is a way to further reduce F residues as only the cooked pasta is consumed.

SF affects the seed germination significantly and impacts on some technological properties of durum wheat, semolina and derived pasta leaving significantly high fluoride residue. However, the fumigant did not affect bread making quality. Therefore, SF is not recommended if the grains are to be used as seeds for agricultural production. Wheat fumigated with SF retains most of the F residue in the bran which is removed during milling, reducing residues in end products derived solely from semolina/flour. However, 100% wholemeal products should be avoided. Pasta processed from fumigated semolina is relatively unaffected with further loss of residue occurring during its cooking making it safe for human consumption.

CHAPTER SEVEN

7. Aims, General discussion, significance, recommendations, future work and conclusion

Sulfuryl fluoride is a fumigant that has been used over the past 60 years against termite infestation of structures. The international phasing out of methyl bromide from use, and more recently, the development of resistance to phosphine requires a successful replacement. Although SF is already registered in several countries for the protection of grain, many of its properties in relation to grain are little known and any alternative treatment should be carefully tested. An important property of any fumigant is its sorption and desorption characteristics on various commodities under a range of conditions. This has significant effects on application, efficacy and management of the fumigant.

7.1. Aims

This project contains a number of aims. Firstly, quantifying the process of sorption and desorption of SF in wheat grains and their derived semolina/flour under a range of storage conditions that might typically be encountered in bulk storage facilities, flourmills and other processing facilities used in many countries. In addition, fumigation with SF at the tested range of storage conditions used in this study should reflect typical conditions experienced in many wheat growing regions of the world. Secondly, it should investigate the impact of sorption of the fumigant by the commodity on the efficacy of the fumigant against an important phosphine resistant insect, *Rhyzopertha dominica*, and highlight the impact of both chemical and physical sorption on the mortality of both adult and egg of this insect species. Thirdly, it should evaluate the impact of the ratio of grain to storage area (filling ratio) on the sorption of the fumigant by the grain and its efficacy and potential residues. Fourthly, it should, investigate to what extent repeating fumigation could have on residues. Finally, the impact of fumigation with sulfuryl fluoride on the technological characteristics of wheat grains and their products such as pasta and bread and on fluoride residues were evaluated under different conditions of fumigation with SF.

7.2. General discussion

Protecting cereal grain is a valuable procedure as grain nutrition value is crucial for human life. Without protection against pests, the value of the grain will deteriorate due to contamination from insect infestation. Fumigation is the most important element in grain protection as it gives rapid results for controlling insect infestations at different life stages.

The largest natural threat to the safe storage and distribution of grains is insect infestation. This is particularly the case in warmer climates that favour insect population growth resulting in very high infestation pressure. There are more than 100 species that attack the stored grain. The lesser grain borer, *Rhyzopertha dominica* is one of the most serious pests of stored grain because it has a strong resistance to the currently used fumigant phosphine, and it is a common wheat grain attacker worldwide. Several pest controlling procedures are used in Australia by stored grain holders. These procedures include cooling, drying and sanitation. However, chemical treatments, especially fumigation, are still the most effective tools for controlling insect infestations. Fumigation is therefore important for food security and continuity of the supply chain.

The fumigant phosphine is still the main fumigant used by the grain industry. Phosphine provides a number of benefits such as low cost, does not leave grain residues and provides a good level of toxicity to many insect species. An insect infestation is a serious threat for the Australian grain industry (worth ~ AUD 9 billion). SF has been used for many years as a treatment for dry wood termites in the USA. With the international phase-out of methyl bromide, Dow AgroSciences developed SF as a replacement for methyl bromide for disinfestations of many commodities including stored products and storage facilities. The Dow product, ProFume®, was developed principally to fumigate mills and other structures over 2-day periods. ProFume® was also registered for the disinfestation of stored grain and grain storage facilities in Australia in 2007.

Australian grain handling companies, for example Grain Corp, are now working cooperatively with scientists from DAF Queensland and NSW DPI to develop more effective fumigation procedures by testing modified fumigation protocols such as extending exposure time and increasing the applied concentration to overcome low mortality and the resistance of different species to phosphine.

Gases such as fumigants diffuse into and out (sorb and desorb) of grains and other materials during fumigation and continue to desorb after the fumigant has been removed unless the fumigant is bound into the grain and not available for desorption. Sorption can be purely physical but often involves chemical reaction with components of seeds and can lead to the retention of chemical residues in the seed that may impact the technological quality of the grain and derived products. The rate of sorption and desorption is influenced by many variables with the most important being commodity type, fumigant concentration and exposure period, moisture content of the seed and temperature (Hwaidi et al., 2015).

Knowledge of the sorption and desorption properties of a fumigant is essential for its effective management and use, and for workplace health and safety procedures, grain handling logistics and toxicity to target pests (effectiveness as a disinfectant). In addition, the consequent chemical reactions may impact on properties of the fumigated commodity and its downstream products. Health, safety, environmental and economic considerations severely limit the range of chemicals that can be applied to grain, and in recent years we have seen various authorities around the world reduce the number of chemicals available. Chemicals that can be applied to grain are rare and very costly to develop. By far the most important fumigant used in grain protection world-wide is phosphine (PH_3). However, the efficacy of it against stored product insect decreases because of the resistance.

The Australian Pesticide Veterinary Medicines Authority (APVMA) publically released their evaluation of sulfuryl fluoride in the product ProFume gas fumigant in 2007 (APVMA, 2007). They reported results of a survey of samples of various Australian wheat types exposed to 1500 mg.h/L SF with single and repeated fumigation. Their results showed residues of fluoride less than the maximum residue limit (MRL_s) of fluoride, which is 7 mg/kg for cereal grain in Australia and around 60-70 mg/kg in the US and EU. However, the fumigation conditions were not mentioned in their study as it is generally known that conditions such as temperature, moisture content, exposure time, concentration and filling ratio affect the chemical sorption of the fumigant by the commodity and level of chemical residue (Hilton and Banks, 1997 a and b). The results of my study indicated fluoride residues higher than the maximum residue limit in Australia for cereal grain fumigated when the filling ratio is more than 0.50 and fumigation with 8.298 mg/L of SF is repeated for four times or more (chapters 4 and 5). However, the residues become less than the MRL after milling the grains. The results indicate SF was sorbed into the different commodities in an exponential way and the data describing the decay of the fumigant concentration into the fumigated system was fitted with a first order equation, the same as phosphine (Daglish and Pavic, 2008 and 2009), methyl bromide and ethyl formate (Hilton and

Banks, 1997 a and b). SF sorption by the commodities was affected by fumigation and storage conditions as the rate of sorption was increased with increasing temperature, commodity moisture content and fumigant concentration. In addition, the smaller the particle size of the fumigated commodity which determines the amount of surface area exposed to the fumigant, the higher the sorption rate. Flour sorbs fumigant more rapidly than semolina which has coarser particles In the same context, a number of other studies indicated a relationship between protein and sorption of the fumigants.

In grain industry principles, a silo is fumigated with the same dosage whether the silo is full or empty of grain. The initial concentration of the fumigant has a weak relationship with sorption rate. However, it affects strongly the final residues and efficacy against insects. Sorption of SF is affected by fumigation conditions. When both temperature and moisture content of the commodity are increased, the sorption rate increases as well. Temperature increases the transfer energy of fumigant molecules due the friction between the fumigant molecules and the surface of the particles. Therefore, more gas molecules penetrate the fumigated commodity and more residues could be left if the fumigant is not desorbed completely and even partially and/or does not chemically react or bind to commodity components. Moisture content of the commodity has an impact on the sorbed amounts of the fumigant also.

Exposure time of commodity to the fumigant is one of the key factors that strongly affect the sorption of the fumigant and efficacy against the fumigated pest and may affect the required concentration to form an effective CT against the target insect. Long exposure time means low concentration, and short exposure time means a high concentration is needed for the same CT product. The presence of the commodity and the filling ratio used has been shown to impact the efficacy of the fumigant (against *R. dominica*) and the sorption rate. A low exposure time (of 48

h) was required to achieve high mortality of the different life stages of different species when the concentration of SF was high and the commodity was absent during the fumigation process. Less than 1500 mg.h/L SF was required for complete mortality of eggs and adults (Baltaci et al., 2009; Bell et al., 2002; Reichmuth et al., 2003; Su and Scheffrhan, 1990). The work described in this thesis proved that the presence of the commodity has a very strong impact on its efficacy. Presence of the commodity during the fumigation process decreases the fumigant concentration below toxic levels when the fumigated system has 0.50 of its volume as grain. Similarly, increasing the commodity level increases the sorption rate, and this is proved by this work and supported by other research on other fumigants. Thus, insect infestation using high filling ratios leads to significantly high sorption rates and a rapid decrease of the effective fumigant concentration into the fumigated system. Applying high SF concentrations may achieve the required mortalities although increasing sorption rate with filling ratio.

The effectiveness of the fumigant against the targeted insect was investigated by evaluating the mortality (Warrick, 2011). When the conditions of the fumigation are extreme (for example in this study, 15% m.c and 35°C) and the silo is full, obtaining the desired mortality will be difficult. In the grain industry in Australia, fumigations may need to be repeated due to the failure of a previous fumigation to achieve a complete control of the insect pests (Shi et al., 2012). The failure of complete control may happen due to leaks in the storage system and a decline in the concentration inside the fumigation system to be lower than that required to control insect pests. Sorption of the fumigant by the commodity is considered as another type of loss during the fumigation process in gastight storage systems as discovered by this study. This loss of the fumigant into stored commodity increases when temperature, commodity moisture content, exposure time, filling ratio increase, and this may lead to reduction of the efficacy of the fumigant with applied concentration here in this study. However, increasing the fumigant dose may compensate the sorbed amounts by the commodity and may increase the efficacy of the fumigant against insects. Results of this study indicated an increased sorption of SF into wheat with increasing the number of the fumigations. Similar findings were reported by Reed and Pan, (2003) who found phosphine sorption by wheat increased with increasing the number of fumigations. Accumulation of SF into grains led to an increase of fluoride residues, and the maximum quantified residues was 13 mg/kg after the fifth fumigation cycle. Repeating fumigation for four times or more with conditions mentioned in chapter five leaves residues exceeding the maximum residue limit for Australia, which is 7 mg/kg as fluoride residue for cereal grains (APVMA, 2007).

The impact of the fumigant SF on the technological characteristics of wheat was very clear from the results of chapter (6). SF affects the ability of wheat grain to germinate with a marked reduction in % germination with increasing SF concentration, wheat moisture and higher ambient temperature. Similar results were reported by Howe (1934) who discovered that adding fluorine to the superphosphate fertiliser decreased the germination of corn up to 90%.The most likely reason for decreasing germination with SF fumigation in this study could be due to the toxicity of the gas itself. Sulfuryl fluoride interrupts the glycolytic cycle of living tissues and that stops the mitochondria from producing and supplying the required energy for growth and germination. The fluoride residues detected in this study reflect the extent of SF penetration into the living tissue of the grain. The quantified fluoride residues in this study were shown to increase with an increase in the filling ratio (chapter 6). This increase of fluoride residue actually comes from increasing the sorption rate of the fumigant SF by the grain with increasing filling ratio (chapter 4). The increase of fumigant sorption with increasing filling ratio was recorded before with phosphine by

Daglish and Pavic, (2008) and for methyl bromide and ethyl formate by Hilton and Banks, (1997a and b, respectively). Fluoride residues quantified from durum wheat samples were found to increase with increasing fumigant concentration and temperature. However, after milling, the majority of the residue was in the bran fraction and the residues in the semolina were less than the maximum residue limit. Manufacturing pasta from fumigated semolina reduces the residue levels of fluoride even further. This helps in decreasing consumer concerns regarding the residues of fluoride due to fumigation with SF. Cooking water of the cooked pasta has a significant amounts of fluoride residues. However, the cooking water normally is drained and not used for any other cooking purposes.

Fumigation with SF increases the yellowness of semolina but this was not translated into pasta yellowness changes so is considered practically not important (Chapter 6). Moreover, fumigation with SF in the presence of higher temperatures and grain moisture content decreased the mixograph peak time without affecting dough strength, which saves time for blending semolina with water for making pasta or making the required time for mixing shorter. Cooking loss and over cooking tolerance increased due to sorption of SF and temperature by semolina, and this may occur because the presence of sorbed SF with high temperature increased starch leakage from pasta to the cooking water. These changes are slight negative affects on perceived quality but a sensory analysis would be needed to see if there are any real impacts on consumer acceptance which is unlikely. SF increased cooked pasta firmness significantly, and this causes increasing resistance of pasta incision with the teeth which would be desirable for some consumers. Stickiness force and area 1:2 increased as well. The loss of starch and protein during cooking because of the effect of SF may lead to accumulation of sticky amylose on the pasta surface. Fumigation with SF does not affect the baked bread making quality parameters.

7.3. Significance

1. There is very little information regarding the sorption of the fumigant SF into commodities at different fumigation and storage conditions. Knowledge of the sorption characteristics and consequences of SF use under Australian conditions is essential for the safe and effective application of this fumigant for the protection of grain. Knowledge of sorption and desorption phenomena are required to ensure adequate dosages are applied to control insect pests and the appropriate workplace health and safety procedures are in place. In addition, it is important for facilitating grain handling logistics (grain cannot be moved if it is de-sorbing toxic gas) and acceptability to markets. It is important that this information be made available to the Australian grain industry.

2. The impact of sorption of SF by the commodity during fumigation of the target insect has not been investigated. The significance of this work is that the presence of the commodity during fumigation decreases the amount of gas available to control the target insect at different life stages due to sorption. This research develops new knowledge and recommends new procedures regarding the fumigation process in the presence of the commodity by indicating that sorption is the most important factor that causes the loss of the fumigant from the fumigation system into the commodity and reducing the concentration below the toxic level.

3. Identifying the behaviour of SF sorption with different filling ratios provides information about how the fumigant will be sorbed at different filling ratios. This provides important information for the grain industry and helps in understanding the appropriate storage level when using SF. Sorption of the fumigant chemically at the different densities provides information regarding the importance of grading of the grain before storage. As the bulk sorbs more fumigant than the grain, this is an indication that shows increasing chemical sorption with increasing grain density.

In addition, it explains the relationship between both physical and chemical sorption and the sorption behaviour of the fumigant and the predicted residues. Repeated fumigation using SF has not been mentioned before, and quantifying residues for repeated fumigation with a range of fumigation and storage conditions is not available.

4. Repeating fumigation using SF increases sorbed amounts of the fumigant although the sorption rate decreases when the fumigation is repeated. Fluoride residues increase with increasing the number of fumigations and makes the residues become higher than the maximum residue limit in Australia. The impact of fumigation with SF on the technological characteristics of the grain, pasta, bread and residues of fluoride due to applying SF at different storage conditions to wheat and its products has not been mentioned before.

5. There is very little information regarding the effect of SF on the germination and the technological characteristics of wheat grain, pasta and bread due to leaving fluoride resides. This study demonstrates that applying SF to the grain as a fumigant at different conditions provides new information regarding the impact on germination It also provides information on the affect on the technological characteristics of the grain, including hectolitre weight and grain hardness. However, it did not change 1000 grain weight and protein content. Bread characteristics did not change in grain fumigated with SF

7.4. Recommendations

1. SF is recommended for stored grains and their products, as it is sorbed by wheat with rates less than other fumigants but higher than phosphine. It is considered as a phosphine resistance breaker in Australia. The quick desorption of SF serves work place health and safety requirements. Fumigation of SF at high temperatures (>35°C) and moistures (>12% m.c) should be avoided as

high sorption of the fumigant occurs especially for long exposure times or using SF high concentrations. However, in some cold climates higher than 25°C is required to increase the efficacy of the fumigant against some insect species.

2. Due to the rapid sorption of SF by the commodity especially at high temperature and moisture content, and high SF concentration, exposing wheat and flour for long exposure times is not recommended.

3. Fumigation with SF in the presence of the commodity or any material absorbing the fumigant should be done on the basis considering sorption as a key factor of the process. It was found by this study that the fumigant is sorbed physically and chemically by wheat and both types of sorption have different relationships with mortality of insect pests. The major effect relates to physical sorption as it is available for desorption.

4. SF sorption increased with increasing the filling ratio of the durum grain, and a recommended filling ratio to be used is 0.5 because there was lower sorption and less F residue while still being effective against *R. dominica*. However, in commercial practice where optimisation of storage capacity is important for as short a period as practical, a higher filling ratio is desirable and this would require a higher SF concentration than 1mg/L to shorten exposure times. Leaking of gas would also play a role in achieving target CT products under these situations. Chemical sorption of the fumigant increases with increasing grain density and filling ratio. However, physical sorption was not dependent on either bulk density or true density or on filling ratio. The physically sorbed amount of the fumigant by the grain increases with time in an exponential manner. The exponential sorption of phosphine, methyl bromide, ethyl formate, and other fumigants with time was reported before (Daglish and Pavic, 2008; 2009; Hilton and Banks, 1997 a; b). Chemical sorption also was demonstrated by this study to be linear with time.

5. Fumigating wheat with multiple fumigations (repeated for four times or more) is not recommended as it leaves fluoride residue that may exceed the Australian maximum residue limit of fluoride in cereal grains.

6. Only minor changes in pasta quality were noted that are unlikely to be significant to the consumer. Some increases in cooking loss and the pasta being less tolerant to overcooking and minor increases in stickiness were found but these changes were only small. SF fumigant under all the conditions evaluated had no impact on bread baking quality.

7.5. Future work

1. Sorption rate of wheat and its products increases when the temperature and commodity moisture content increase. In addition to that, the exposed surface of the commodity to the fumigant is important. In this study and other studies on phosphine, methyl bromide and ethyl formate no impact of applied initial concentration on sorption rate was found. Therefore, shorter exposure times and higher concentrations than those used here need to be tested under various temperature conditions. In addition, analysis of fluoride residues is needed to ensure limits are not exceeded.

2. Sorption of the fumigant by commodity decreases the efficacy of SF against insect pests. The investigated concentrations did not achieve complete mortality for either life stage tested. However, there was a probability of achieving complete control within the adult life stage. Therefore, more studies are required to test using the same concentrations but extending the exposure times for longer than the one tested here on the survival of the eggs. In addition, higher concentrations with shorter exposure times need to be tested for a complete control of egg life stage.

There was a quadratic relationship between the physical sorption and the mortality for both adult and egg. Therefore, more studies are required to investigate to what extent this relationship will continue and whether there is any probability for the relationship to be affected by desorption or resorption with longer exposure time.

Moreover, it was demonstrated by this study that the relationship between the chemical reaction of the fumigant with grain and the mortality of both adult and egg is linear. Therefore, the linear relationship needs to be investigated to what extent it will continue linear, and is there any probability of becoming quadratic especially with extending the exposure time and increasing the chemically accumulated amounts of the fumigant into the grain. Physically sorbed amounts may be desorbed. However, chemically sorbed amounts are not.

3. Examining sorption of the fumigant SF with other types of Australian grain is required as only durum was tested with different filling ratios. Also, a wide range of protein content within grain species should be exposed to SF as it is demonstrated in chapter 5 the possible difference in sorption due to the different protein content. Grading grain appears to be another important factor in fumigation with SF. Therefore, it is suggested by this work to expose different grain densities within the same bulk and different grain densities within different bulk densities to SF.

4. A range of types of wheat with different content of protein need to be tested again to investigate the impact of protein on sorption and residues. Moreover, another type of grain such as malting barley, pulses and oilseeds need to be tested at the same or different conditions to give more information regarding the impact of grain composition on the sorption and residues of SF.

5. This research suggests investigating the impact of SF on germination at lower concentrations than in chapter 2 as all the tested conditions in that chapter decreased the germination percentage significantly. Testing concentrations less than 1 mg/L and 8.928 mg/L in chapters 4, 5,

respectively, for quantifying the residues or changing the fumigation conditions will give an idea about suitability of the fumigant for cereal grains. As this fumigant is nominated as resistance breaker and phosphine replacement, more work will be required to achieve the concentrations that enable the minimum impact on the technological characteristics of wheat grain products. Additional work will be required to investigate the impact of SF sorption on the amino acids and glutens.

6. More quality studies will be required to test the baking quality on the fumigated grains after storage, especially low germination grains. Dead germs grains are subject to degradation of quality as they not protected physiologically.

7.6. Conclusion

SF is sorbed significantly by commodity when the temperature of the fumigation and the moisture content of the commodity are high and exposure time is long. However, rate of sorption of SF by whole wheat grain and its products, flour and semolina, is less than other fumigants such as methyl bromide and ethyl formate but higher than phosphine. SF desorbs rapidly from the fumigated commodity after aeration making it lower risk than other fumigants in the workplace. Sorption of the SF by the commodity impacts the efficacy of SF against adults more than eggs. At low physical sorption is a major determinant of SF efficacy as the relationship between mortality and physical sorption was quadratic and this was revealed as a decrease in kill when all the fumigant was sorbed physically. There was no such relationship with chemical sorption. Furthermore, sorption of SF increases with increasing filling ratio of the commodity into the storage system. The physically sorbed amount of the fumigant by the grain does not depend on the density of either the bulk or the true grain density, however, the chemical sorption does.

Moreover, repeating fumigation increases the physically sorbed amounts of the fumigant and fluoride residue content in the grain. Fluoride residues may be a concern as they can exceed the maximum residue limit but only if grain is fumigated with SF four or more times and with a filling ratio above 0.5. However, consuming pasta is not a concern even if the semolina is fumigated directly by SF as the fumigant is predicted to be desorbed rapidly. Fumigation with SF increases the yellowness, cooking loss, over cooking tolerance, stickiness and firmness. However, this increase does not affect the technological characteristics of pasta and is likely to be accepted by the consumer as the changes in pasta quality did not go over acceptable limits. Baked bread is not affected in any way by SF fumigation.

REFERENCES

AACC, 2010. Approved Methods of the AACC. American Association of Cereal Chemists, St. Paul, MN, USA.

AGI, 2011. The Australian Grain Industry, the basics, 1-18. http://www.pwc.com.au/industry/agribusiness/assets/Australian-Grains-Industry-Nov11.pdf

Ahmed, G., Hamrick, D., Guinn, A., Abdulsamad, A., Gereffi, G., 2013. Wheat Value Chains and Food Security in the Middle East and North Africa Region, Duke University 1-51.

Ahmed, M., 1990. Irradiation disinfestation of stored foods. In: Fleurat-Lessard, F.; Ducom, P. (Eds), Environmental and Physical Control Methods. Proceedings of the 5th International Working Conference on Stored Product Protection, 9-14 September 1990, Bordeaux, France. Imprimerie du Médoc, Bordeaux, France, pp. 1105-1116.

AEGIC, 2014. Global Grain Market Series. Australian Export Grains Innovation Centre 1-2.

Al- Yahya, S.A., 2001. Effect of storage conditions on germination in wheat. Journal of Agronomy and Crop Science, 186, 273- 279.

Amuh, I.K.A., 1975. Persistence of ethylene dibromide residues in fumigated maize. Ghana Journal of Science 15, 59-63.

Anonymous,1994. Monteral protocol on substances that deplete ozone layer: Report of the methyl bromide technical options committee, 1995 assessment. UNEP, ozone secretariat,Nairobi, enya,http://ozone.unep.org/teap/reports/MBTOC/MBTOC94.pdf.

APVMA, 2007. Public release summary. An evaluation of the new active sulfuryl fluoride in the product Profume gas fumigant. September 2007. Australian Pesticides and Veterinary Medicines Authority.

Aung, L.A., Leesch, J.G., Jenner J.F., Cardwell, G.E.E., 2001. Effects of carbonyl sulphied, methyl bromide and sulfuryl fluoride on fruit phytotoxicity and insect mortality. Annals of Applied Biology 139, 93-100.

Australian bureau of statistics, 2011. http://www.abs.gov.au/ausstats/abs@.nsf/Lookup/7124.0Chapter72010-11. Accessed on 14/03/2015.

Australian grain, 2013. New grain storage products and equipment. Vol 23, No 2, 24.

Bailey, J.E., 1992. Whole grain storage. In Sauer, D.B., (Ed). Storage of cereal grains and their products. Fourth edition. American Association of Cereal Chemists, Inc, St Paul, MN 55121-2097, USA, pp. 157-182.

Baker, P.B.,Carlo, L., 2003. Agricultural fumigation safety guide for the Arizona pesticide application certificate, training manual, The University of Arizona, College of Agriculture and Life Sciences 1-47.

Bakheit, B.R., Abdou, R.F., Abdalla, F.H., 1985. Germination, seedling vigour and induction of chromosomal aberrations in wheat grains fumigated with phosphine. Seed Science and Technology, 13, pp, 725-739.

Baltaci, D., Klementz, D., Gerowitt, B., Drinkall, M.J., Reichmuth C., 2009. Lethal effects of sulfuryl fluoride on eggs of different ages and other stages of the warehouse moth *Ephestia elutella* (Hübner). Journal of Stored Products Research, 45, 19–23.

Banks, H.J., 1986. Sorption and desorption of fumigants on grains: mathematical descriptions. In: Champ,B.R., and Highley,E. (Eds), Pesticides and Humid Tropical Grain Storage Systems. Proceedings International Seminar, Manila, Philippines. Australian Centre for International Agricultural Research, Canberra, Australia, Proceedings No. 14, pp. 179–193.

Banks, H.J., 1989. Behaviour of gases in grain storage. In: Champ B.R., Highley E., Banks H.J. (Eds), Fumigation and Controlled Atmosphere Storage of Grain. Proceedings of an International Conference, 14-18 February 1989, Singapore, pp. 96-107.

Banks, H.J., 1990. Behaviour of gases in grain storages. In: Champ, B.R., Highley, E., and Banks, H.J. (Eds), Fumigation and Controlled Atmosphere storage of Grain. Proceedings of the International Conference on Controlled Atmosphere and Fumigation in Stored Products, 14-18 February, 1989, Singapore, Australian Centre for International Agricultural Research Proceedings, 25, pp. 96-107.

Banks, H.J., 1993. Uptake and release of fumigants by grain: sorption/desorption phenomena. In: Navarro,S., and Donahaye, E. (Eds). Physical and Chemical Processes in the Application of Controlled Atmosphere and Fumigation. Proceedings of International Conference on Controlled Atmosphere and Fumigation in Grain Storages, June 1992, Winnipeg, Canada, pp. 241-260.

Banks, H.J., Desmarchelier, J.M., Elek, J.A., 1976. Determination of bromide ion content of cereals and other foodstuffs by specific ion electrode. Pesticide Science 7, 595-603.

Bell, C.H., Savvidou, Smith, T.J.W., 1999. The toxicity of sulfuryl fluoride (Vikane) to eggs of insect pests of flourmills. In: Jin, Z., Liang, Q., Liang, Y., Tan, X., Guan, L. (Eds.), Fumigation and Controlled Atmosphere. Proceedings of the 7th International Working Conference on Stored Product Protection, 14-19 October 1998, Beijing, China. Sichuan Publishing House of Science and Technology, Chengdu, China, pp. 345-350.

Bell, C.H., Wonter-smith, T.J., Savvidou, N., 2002. Some properties of sulfuryl fluoride in relation to its use as a fumigant in the cereals industry. In: Credland, P.F., Armitage, D.M., Bell, C.H., Cogan, P.M., Highley, E. (Eds), Advances in Stored Product Protection, Proceedings of the 8th International Working Conference on Stored Product Protection, 22–26 July 2002, New York, UK. CAB International, Oxon, UK, pp. 910-915.

Bell, C.H., Savvidou, N., Smith, T.J.W., Cardwell, S.K., Bodle, C., 2004. Development of sulfuryl fluoride as a fumigant for the milling industry. HGCA, Caledonia House, 223 Pentonville Road, London. N1 9HY, 1-67.

Bell, C.H., 2006. Factors affecting the efficacy of sulfuryl fluoride as a fumigant. In: Lorini.I., Bacaltchuk. B., Beckel. H., Deckers. D., Sundfeld. E., dos Santos. J.P., Biagi. J.D., Celaro. J.C., Faroni. L. R. D'A., Bortolini. L.de O. F., Sartori. M. R., Elias. M. C., Guedes. R. N. C., da Fonseca R. G., Scussel., V. M. (Eds.), Fumigation and Controlled Atmospher. Proceedings of the 9th International Working Conference on Stored Product Protection, 15 to 18 October 2006, Campinas, São Paulo, Brazil. Brazilian Post-harvest Association - ABRAPOS, Passo Fundo, RS, Brazil, pp. 519-526.

Bell, K.O., 1971. Angoumois grain moth, *Sitotroga cerealella* (Olivier), reared on pellets and meals composed of various combinations of endosperm, germ and bran of wheat. Ph.D dissertation. Kansas State University, Manhattan, 1-188.

Bengston, M., Strange, A.C., 1994. Recent developments in grain protestants for use in Australia. In: Highley, E., Wright, E.J., Banks, H.J., Champ, B.R. (Eds.), Grain Protectants. Stored Product Protection, Proceedings of the 6th International Working Conference on Stored-Product Protection, 17-23 April 1994, Canberra, Australia. CAB International, Wallingford, United Kingdom, pp. 751-754.

Berck, B., 1964. Some parameters in the use of fumigants. World Review of Pest Control 3, 156-174.

Berck, B., 1968. Sorption of phosphine by cereal products. Journal of Agricultural and Food Chemistry 3, 419-425.

Bewley, J.D., 1997. Seed germination and dormancy. American Society of Plant Physiology.The Plant Cell 9, 1055-1066.

Bewley, J.D., Black, M., 1994. Seeds: physiology of Developement and Germination. New Yourk Press.

Bewley, J.D., Bradford, K.J., Hilhorst, H.W.M., Nonogaki, H., 2013. Seeds physiology of development, germination and dormancy. Springer New York Heidelberg Dordrech London.

Black M., 1970. Seed germination and dormancy. Science Progress 58, 379- 393.

Blakeney, A.B., Cracknell, R.L., Crosbie, G.B., Jefferies, S.P., Miskelly, D.M., O'Brien, L., Panozzo, J.F., Suter, D.A.I., Solah, V., Watts, T., Westcott, T., Williams, R.M., 2009. Understanding Australian wheat quality. A basic introduction to Australian wheat quality. Grain Research and Development Corporation. Wheat Quality Objective Group, 1-40.

Blecker, L.A., Thomas, J.M., 2012. A national pesticide applicator certification study guide, soil manual fumigation guide, Published by the National Association of State Departments of Agriculture Research Foundation 1-128.

Bond, E.J., Buckland. C.T., 1978. Control of insects with fumigants at low temperatures: toxicity of fumigants in atmospheres of carbon dioxide. Journal of Economic Entomology 71, 307-309.

Bond, E. J., 1984. Manual of Fumigation for Insect Control. Plant Production and Protection. FAO, Rome. Paper 54, pp. 432.

Bond, E.J., 2007. Manual of fumigation for insect control, FAO 1-364.

Borrelli, G.M., Troccoli, A.D.F., Fares, C., 1999. Durum wheat lipoxegenase activity and other quality parameters that affect pasta colour. Cereal Chemistry Journal, 76, 335-340.

Botha, F.C., Potgieter G.P., Bothat, A.M., 1992. Respiratory metabolism and gene expression during seed germination. Journal of Plant Growth Regulators 11, 211-224.

Borrelli, G.M., Troccoli, A.D.F., Fares, C., 1999. Durum wheat lipoxegenase activity and other quality parameters that affect pasta colour. Cereal Chemistry 76, 335-340.

Braby, M.F., 1992. Sorption of methyl chloride on wheat. Journal of Stored Products Research 28, 183-186.

Buller, A.H.R., 1919. Essay on Wheat. The Macmillan Company, New York.

Calvert, G.M., Mueller, C.A., Fajen, J.M., Chrislip, D.w., 1998. Health effects associated with sulfuryl fluoride and methyl bromide exposure among structural fumigation workers. American Journal of Public Health, 88, 1774-1780.

Champ, B.R., Dyte, C.E., 1976. Report of the FAO global survey of pesticide susceptibility of stored grain pests. FAO Plant Production and Protection Series No. 5, FAO, Rome 1-297.

Choate, H.A., 1921. Chemical changes in wheat during germination. Botanical Gazette 7, 409-425.

Christensen, C.M., Kaufmann, H.H., 1969. Grain storage: The role of fungi in quality loss. University of Minnesota Press, Minneapolis 1-153.

Ciesla. Y., Ducom. P., 2009. Ministère de l'agriculture – Laboratoire National de la Protection des Végétaux – LNDS-QUALIS – 71, avenue Edouard Bourlaux – BP 81 – 33883 Villenave d'Ornon Cedex, France. [yann.ciesla@laposte.net] 62-1 - 62-5.

CIMMYT, 1996. CIMMYT 1995-96 world wheat facts and trends: understanding global trends in the use of wheat diversity and international flows of wheat genetic resources. Mexico, DF, 1-80.

Cofie-Agbolr, R., Muir, W.E., Cenkowski, S., Jayas, D.S., 1993. Carbon dioxide gas sorption in stored wheat. In: Navarro, s., Donahaye, E. (Eds), Physical and Chemical Processes in the Application of Controlled Atmosphere and Fumigation. Proceedings of the International Conference on Controlled Atmosphere and Fumigation in Stored Products, June 1992, Winnipeg, Canada 1992, CAF home, pp. 261-269.

Cofie-Agbolr, R., Muir, W.E., Sinicio, R., Cenkowski, S., 1995. Characteristics of carbon dioxide sorption by stored wheat. Journal of Stored Products Research 31, 317-324.

Cofie-Agbolr, R., Muir, W.E., Jayas, D.S., White N.D.G., 1998. Carbon dioxide sorption by grains and canola at two CO_2 concentrations. Journal of Stored Products Research 34, 159-170.

Cogburn, R.R., Tilton, E.W., 1963. Studies of phosphine as a fumigant for sacked rice under gas-tight trapaulins. Journal of Economic Entomology, 56, 706-708.

Collins, P.J., Daglish, G.J., Bengston, M., Lambkin, T.M., Pavic, H., 2002. Genetics of resistance to phosphine in *Rhyzopertha dominica* (Coleoptera: Bostrichidae). Journal of Economic Entomology 95, 862-869.

Collins, P.J., 2006. Resistance to chemical treatments in insect pests of stored grain and its management. In: Lorini, I., Bacaltchuk, B., Beckel, H., Deckers, D., Sundfeld, E., dos Santos, J.P., Biagi, J.D., Celaro, J.C., Faroni, L.R.D.'A., Bortolini, L.de O.F., Sartori, M.R., Elias, M.C., Guedes, R.N.C., da Fonseca, R.G., Scussel, V.M. (Eds.), Proceedings of the 9th International Working Conference on Stored Product Protection, 15-18 October 2006, Sao Paulo, Brazil, pp. 277-282.

Comai, L., and Harada, J.J., 1990. Transcriptional activity in dry seed nuclei indicate the timing of the transition from embryogeny to germination. Proceedings of the National Academy of Sciences 87, 2671-2674.

Cotton, R.T., Walkden, H.H., White, G.D., 1960. Causes of outbreaks of stored grain insects. Kansas Agricultural Experiment Station Bulletin 416, 1-35.

Cotton, RATE, Young, H.D., 1929. The use of carbon dioxide to increase the insecticidal efficacy of fumigants. Proceedings of the Entomological Society of Washington 31, 97-102.

Cox, C., 1997. Sulfuryl fluoride, fumigation fact sheet. Journal of Pesticide Reform 17, 17-20.

Crini, G., Badot, P.M., 2010. Sorption processes and pollution: Conventional and non-conventional sorbents for pollutant removal from wastewaters. Besançon: Presses Universitaires de Franche-Comté, 1-43.

Crowe, J.H., Crowe, L.M., 1992. Membrane integrity in anhydrobic organisms: Toward a mechanism for stabilizing dry seed. In: Somero, G.N., Osmond, C.B., Bolis, C.L. (Eds.), Water and Life. Berlin: Springer-Verlag, pp. 87-103.

Cubadda, R., 1988. Evaluation of durum wheat semolina and pasta in Europe. Pages 224-226 in: Chemistry and Technology, 1st ed. G. Fabriani and C. Lintas, Eds. American Association of Cereal Chemists, St. Paul, MN. USA.

Cubadda, R., Carcea, M., Marconi, E., Trivisonno, M.C., 2007. Influence of gluten proteins and drying temperature on the cooking quality of durum wheat pasta. Cereal Chemistry Journal 84, 48-55.

Curtis, B.C., 1982. Potential for a yield increase in wheat. In: Proceeding of the National Wheat Research Conference., Beltsville, MD, USA, 26-28 Oct, Washington, DC, National Association of Wheat Growers Foundation, pp. 5-19.

Daglish, G.J., Collins, P.J., and Pavic, H., Kopittke, R.A., 2002. Effects of time and concentration on mortality of phosphine -resistant *Sitophilus oryzae* (L) fumigated with phosphine. Pest Management Science 58, 1015-1021.

Daglish, G. J., Pavic, H., 2008. Effect of sorption dose on sorption in wheat. Pest Management Science 64, 513-518.

Daglish, G.J., Pavic, H., 2009. Changes in phosphine sorption in wheat after storage at two temperatures. Pest Management Science 65, 1228-1232.

Daglish, G.J., Ridley, A.W., Walter, G.H., 2010. Resistance management and the ecology of *Rhyzopertha dominica* (F.) and *Tribolium castaneum* (Herbst) in subtropical Australia. In: Carvalho, M.O., Fields, P.G., Adler, C.S., Arthur, F.H., Athanassiou, C.G., Campbell, J.F., Fleurat-Lessard, F., Flinn, P.W., Hodges, R.J., Isikber, A.A., Navarro, S., Noyes, R.T., Riudavets, J., Sinha, K.K., Thorpe, G.R., Timlick, B.H., Trematerra, P., White, N.D.G. (Eds.), Proceedings of the 10th International Working Conference on Stored-product Protection, Estoril, Portugal, pp. 104-109.

Damcevski, K. A., Annis, P. C., 2006. Influence of grain and relative humidity on the mortality of *Sitophilus oryzae* (L.) adults exposed to ethyl formate vapour. Journal of Stored Products Research 42, 61-74.

Darby, J. A., 2008. A kinetic model of fumigant sorption by grain using batch experimental data. Pest Management Science 64, 519–526.

Darby, J.A., Trasy, W., Katherine, D., 2009. Modelling the kinetics of ethyl formate sorption by wheat using batch experiments. Pest Management Science 65, 982-990.

Delcour, J.A., Hoseney, R.C., 2010. Principles of cereal science and technology. AACC International, Inc. American Association of Cereal Chemists, St. Paul, Minnesota 55121, USA.

Dendy, D.A.V., Dobraszczyk., 2001. Cereals and cereal products chemistry and technology. Aspen Publishers, Inc. Gaithersburg, Maryland.

Diamond, J., 1997. Guns, germs and steel. The fates of human societies. W.W. Norton and Co, New York.

Dick, J. W., Matsuo, R. R., 1988. Durum wheat and pasta products. In: Pomeranz, Y. (Ed), Wheat Chemistry and Technology, American Association of Cereal Chemists. St. Paul, MN, pp 507-547.

Drinkall, M.J., Pye, C. D., Bell, C. H., braithwaite, M., Clack, S. R., Ive, J., Kershaw S. 2004. The practical use of the fumigant sulfuryl fluoride to replace methyl bromide in UK flour mills. Proceedings of the 12th International ICC cereal and bread congress 24-26 May, pp 245-249.

Dumas, T., 1980. Phosphine sorption and desorption by stored wheat and corn. Journal of Agricultural and Food Chemistry 28, 337-339.

Dumas, T., Bond, E.J., 1977. Penetration, sorption and desorption of fumigant in the treatment of food materials with a methyl bromide acrylonitrile mixture. Journal of Agriculture and Food Chemistry, 25, 677-680.

Dumas, T., Bond, E.J., 1979. Relation of temperature to ethylene dibromide desorption from fumigated wheat. Journal of Agriculture and Food Chemistry 27, 1206-1209.

Edde, P.A., 2012. A review of the biology and control of *Ryzopertha dominica* (F.) the lesser grain borer. Journal of Stored Products Research 48, 1-18.

Ehrenshaft, M., Brambl, R., 1990. Respiration and mitochondria biogenesis in germinating embryos of maiz. Plant Physiology 4, 1251-1261.

Eisenbrandt, D.L., Nitschke, K.D., 1989. Inhalation toxicity of sulfuryl fluoride in rats and rabbits. Fundamental and Applied Toxicology 12, 540-557.

EPA, 1993. Registration Eligibility Decision of Sulfuryl Fluoride, document number EPA-738-R-93-016, office of prevention pesticides and toxic substances (H7508W), USA, 1-28.

EPA, (2011). http://iaspub.epa.gov/apex/pesticides/. Accessed 24 February 2015.

Freund, R. J., Littell, R. C., 2000. SAS System for Regression, SAS institute. The United States.

EPPO, 1984. EPPO recommendation on fumigation standards. EPPO Bulletin, 589-609.

Evans, D.E., Thorpe, G.R., Dermott, T., 1983. The disinfestation of wheat in continuous-flow fluidized bed. Journal of Stored Products Research 19, 125-137.

FAO, 2015. Country brief of Iraq. Global Information and Early Warning System on Food and Agriculture (GIEWS), 1-4.

FAOSTAT, 2005. web www.fao.org. Accessed on 11/12/2014.

FAO/WHO, 1965. Evaluation of the hazards to consumers resulting from the use of fumigants in the protection of food. Rome. FAO Meeting Report No. PL/1965/10/2; WHO/Food Add./28.65.

Feillet, P., Autran, J.C., Icard-vernière, C., 2000. Pasta brownness :An assessment . Journa of Cereal Sciences, 32, 215-233.

Food and Drug Administration (FDA), 1988. Wheat flour- Adulterated with insect fragments and rodent hairs. Compliance Policy Guides, Chapter 4, Processed grain Guide 7104.06.

Freund, R. J., Littell, R. C., 2000. SAS System for Regression, SAS institute. The United States.

Golden Software Inc, 2005 . Grapher Version 6, Graphic System, Golden Software Inc. Golden Software, LLC, 809 14th street, Golde, CO 80401-1866 USA.

Golob, P., 1997. Current status and future perspectives of inert dusts for control of stored product insects. Journal of Stored Products Research 33, 69-79.

Gruber, W. Sarkar, A., 2012. Durum wheat milling. In: Sissons, M., Abecassis, J., Marchylo, B., Carcea, M (Eds). Durum wheat chemistry and technology, 2nd edition. American Association of Cereal Chemists International, pp. 139-160.

Hanson, H., Borlaug, N.E. & Anderson, R.G. 1982. Wheat in the third world. Boulder, CO, USA, West view Press.

Harein, P.K., Davis, R., 1992. Control of stored grain insects. In: Sauer, D.B. (Ed), Storage of cereal grains and their products., American Association of Cereal Chemists, Inc. St. Paul, MN, USA, pp. 491-534.

Hermitte, J.J, Shellenberger, J.A., 1947. Effect of excessive methyl bromide fumigation on flour. Cereal Chemistry 24, p 449.

Heuser, S.G.,1975. Factors influencing dosage and choice of toxicant. in: Brady, E.U.; Brower, J.H.; Hunter, P.E.; Jay, E.G.; Lum, P.T.M.; Lund, H.O.; Mullen, M.A.; Davis, R. (Eds.). Stored-Product Fumigation. Proceedings of the 1st International Working Conference on Stored-Product Entomology, 7-11 October 1974, Savannah, Georgia, USA. Savannah, Georgia, USA, 1975. pp. 246 - 253.

Hilton, S. J., Banks, H.J., 1997a. Methyl bromide sorption and residue on sultanas and raisins. Journal of Stored Products Research 33, 249-1997.

Hilton, S.J., Banks, H.J., 1997b. Ethyl formate as a fumigant of sultanas: sorption and efficacy against six pest species. In: Donahaye, E.J., Navarro, S., Varnava, A., (Eds). Integrated commodity management methods with controlled atmosphere. Proceedings of the International Conference on Controlled Atmosphere and Fumigation in Stored Products, 21-26 April 1996, Printco Ltd, Nicosia, Cyprus, pp. 409-422.

Hoseney, R.C., 1986. Principles of cereal science and technology. American Association of Cereal Chemistry, St. Paul, MN, P 327.

Hoseney, R. C., Faubion, J. M., 1992. Physical properties of cereal grains. In: Sauer D. B. (Ed.) Storage of Cereal Grains and Their Products. American Association of Cereal Chemists, p. 30.

Howe, H.M., 1934. The toxic influence of fluorine in phosphatic fertilizers on the germination of corn. PhD thesis, Michigan Agricultural Experiment Station, 177-195.

Hwaidi, M.I., Collins, P.J., Sissons, M., Pavic, H., Nayak, M.K., 2015. Sorption and desorption of sulfuryl fluoride by wheat, flour and semolina, Journal of Stored Products Research 62, 65-73.

INTERIM, 2008. Acute exposure guideline levels for sulfuryl fluoride, 1-43.

Irani, R.R. and Fong, W.S., 1961. Measurements of the particle size distribution of flour. Cereal Chemistry 38, 67-75.

Irvine, G.N., Anderson, J.A., 1953. Variation in principle quality factors of durum wheat with a quality prediction test for wheat of semolina. Cereal Chemistry 30, 334-342.

Irvine, G.N., Winkler, C.A., 1950. Factors affecting the colour of macaroni. II. Kinetic studies of pigment destruction during pasta making. Cereal Chemistry 27, 205-218.

ISTA, 2011. International Rules for Seed Testing. The International Seed Testing Association, Zurichestr, CH-8303 Bassersdorf, Switzerland.

Jacob, H.E., 1944. Six thousand years of bread: Its holy and unholy history. New York. Doubleday.

Jagielaski, J., Scudamore, K.A., Heuser, S.G., 1978. Residues of carbon tetrachloride and 1, 2-dibromoethane in cereals and processed foods after liquid fumigant grain treatment for pest control. Pesticide Science 9, 117-126.

Jayas, D.S., White N.D.G, Muir W.E, 1995. Stored grain ecosystem. Marcel Dekker, New Yourk.

Johnson, V.A., Briggle, L.W., Axtel, J.D., Bauman, L.F., Leng, E.R. & Johnston, T.H. 1978. Grain crops. In: Milner, M., Scrimshaw, N.S., Wang, D.I.C. (Eds). Protein resources and technology, Westport, CT, USA, AVI Publishing, pp. 239-255.

Jones, R.M., 1938. Toxicity of fumigant-CO_2 mixtures to the red flour beetle. Journal of Economic Entomology 3, 298-309.

Kashi, K.P., Bond, E.J., 1975. The toxic action of phosphine: role of carbon dioxide on the toxicity of phosphine to *Sitophilus qranarius* (L.) and *Tribolium confusum* Du Yal. Journal of Stored Products Research 11, 9-15.

Kawamoto, N., Tateya, A., Hoshino, Y., Hiroaki, S., Sakai, T., Kimura, S., 1973. Determination of bromine residues in several imported cereals fumigated with methyl bromide. Research Bulletin of the Plant Protection Service Japan, 11, 20-25.

Krishnasamy, V., Seshu, D.V., 1990. Phosphine fumigation influence on rice seed germination and vigour. Crop Science, 30, 82-85.

Lane, B.G.,1991. Cellular desiccation and hydration: Developmentally regulated proteins and the maturation and germination of seed embryo. The Journal of the Federation of American Sciences for Experimental Biology 5, 2893-2901.

Lewis, S.E., Eccleston, K., 1946. Residues in wheat flour fumigated with methyl bromide, Journal of the Society of Chemical Industry 65, 149-152.

Lindgren, D.L., Vincent, L.E., 1951. Relative Toxicity of Fumigants to *Tribolium confusum* and *Sitophilus granarius* in Wheat. Journal of Economic Entomology 44, 975-979..

Lindgren, D.L., Vincent, L.E., 1962. Fumigation of food commodities for insect control. Advances in Pest Control Research 5, 85-152.

Lindgren, D.L.,Vincent, L.E., Strong, R.G., 1958. Studies on hydrogen phosphide as a fumigant. Journal of Economic Entomology, 51, 900-903.

Lindgren, D.L., Gunther, F.A., Vincent, L.E., 1962. Bromine residues in wheat fumigated with methyl bromide. Journal of Economic Entomology 55, 773-776.

Lindgren, D.L., Sinclair, W.B., Vincent, L.E., 1968. Residues in raw and processed foods resulting from post-harvest insecticidal treatments. Residue Reviews 21,1-121.

Longstaff, B.C., 1999. An experimental and modeling study of the demographic performance of *Rhyzopertha dominica* (F.). I. development rate. Journal of Stored Products Research 35, 89-98.

Longstaff, B.C., 1994. The management of stored product pests by non-chemical means: an Australian perspective, Journal of Stored Product Research 30, 179-185.

Lorini, I., Collins, P.J., 2006. Resistance to phosphine in *Rhyzopertha dominica* (F.) (Coleoptera: Bostrychidae) collected from wheat storages in Brazil. In: Lorini, I., Bacaltchuk, B., Beckel, H., Deckers, D., Sundfeld, E., dos Santos, J.P., Biagi, J.D., Celaro, J.C., Faroni', L.R.D.A., Bortolini, L.de O.F., Sartori, M.R., Elias, M.C., Guedes, R.N.C., da Fonseca, R.G., Scussel, V.M. (Eds.), Proceedings of the 9th International Working Conference on Stored-product Protection, 15-18 October 2006, Campinas, São Paulo, Brazil, pp. 319-323.

Lubatti, O.F., Blackith, R.E., 1957. Germination and growth of cereals fumigated with methyl bromide. Journal of the Science of Food and Agriculture, 8, p 691.

Lubatti, O.F., Harrison, A., 1944. Determination of fumigants. XVII. Comparison of the sorption of hydrogen cyanide, ethylene oxide, trichloroacetonitrile and methyl bromide by wheat. Journal of the Society of Chemical Industry 63, 353–359.

Mahroof, R.M., Edde, P.A., Robertson, B., Puckette, T., Phillips, T.W., 2010. Dispersal of *Rhyzopertha dominica* F. in different habitats. Environmental Entomology 39, 30-938.

Maier, D.E., 1996. Preservation of grain with aeration. In: Champ, B.R., Highley, E., Johnson, G.I., (Eds). Grain Drying in Asia. Proceedings of an International Conference held at the

FAO Regional Office for Asia and the Pacific, 17-20 October 1995, Bangkok, Thailand, Australian Centre for International Agricultural Research, (ACIAR), Canberra, pp 379-397.

Matsuo, R.R., 1988. Evaluation of durum wheat, semolina and pasta in Canada. Pages 249-262 in: Durum Wheat: Chemistry and Technology, 1st ed. G. Fabriani and C. Lintas, Eds. Am. Assoc. Cereal Chem., St. Paul, MN. USA.

Matthews, R.H., Fifield, C.C., Hartsing, T.F., Storey, C.L., Dennis, N.M., 1970. Effect of fumigation on wheat in storage. I. Physical measurements of flour. Cereal Chemistry 47, 579.

Meikle, W.R., Stewart, D., 1962. The residues potential of sulfuryl fluoride, methyl bromide, and methane sulfonyl fluoride in structural fumigation. Journal of Agriculture and Food Chemistry 10, 393-397.

Meikle, W.R., 1964. The fate of sulfuryl fluoride in wheat flour. Agricultural and Food Chemistry 12, 464-467.

Mitsuda, H., Kawai, F., Yamamoto, A., Nakajima, K., 1975. Carbon dioxide protein interaction in a gas-solid phase. Journal of Nutritional Science and Vitaminology 21, 151-162.

Minett, W., Orth, R.A., Cook, L.J., 1976. Methyl bromide fumigation. I. effect of high dosages on bread making quality and germination of wheat. Cereal Chemistry Journal 53, 41-50.

Moore, W., 1936. Differences between resistant and non-resistant red scale in California. Journal of Economic Entomology 29, 65-78.

Morohashi, Y., Shimokoriyama, J.D., 1972. Physiological studies on germination of *Phaseolus mungo* seeds. II. Glucose and organic-acid metabolism in the early phases of germination. Journal of Experimental Botany 23, 54-61.

Morohashi, Y., Bewley, J.D., 1980. Development of mitochondrial activities in pea cotyledons during and following germination of the axis. Plant physiology, 66, 70-73.

Morohashi, Y., 1986. Patterns of mitochondria development in reserve tissue of germinated seeds: A survey. Physiologia Plantarum 66, 653-658.

Multon, J.L., 1988. Spoilage mechanisms of grains and seeds in the post harvest ecosystem. The resulting losses and strategies for the defence of stocks. In: Multon, J.L. (Ed), Preservation and storage of grains, seeds and their by products: Cereals, Oilseeds, Pulses and Animal Feed. Lavoisier, New York, pp. 3-63..

Navarro, S., 1997. Sorption of carbon dioxide by wheat. In: Donahaye, E.J., Navarro, S., and Varnava, A. (Eds), Fumigation and Controlled Atmosphere storage of Grain. Proceedings of The International Conference on Controlled Atmosphere and Fumigation in Stored Products.21-26 April 1996, Printco Ltd, Nicosia, Cyprus, pp. 193-202.

Navaro, S., 2006. New global challenges to the use of gaseous treatments in stored products. In: Lorini, I., Bacaltchuk, B., Beckel, H., Deckers, D., Sundfeld, E., dos Santos, J. P., Biagi, J. D., Celaro, J. C., Faroni, L. R. D'A., Bortolini, L.de O. F., Sartori, M. R., Elias, M. C., Guedes, R. N. C., da Fonseca, R. G., Scussel, V. M. (Eds), Stored Products Protection. Proceedings of the 9th International Working Conference on Stored Product Protection, 15 to 18 October, Campinas, São Paulo, Brazil. Brazilian Post-Harvest Association - ABRAPOS, Passo Fundo, RS, Brazil, pp. 495-509.

Nayak, M., Holloway, J., Pavic, H., Head, M., Reid, R., Collins, P.J., 2010. Developing strategies to manage highly phosphine resistant populations of flat grain beetles in large bulk storages in Australia. In: Carvalho, M.O.; Fields, P.G.; Adler, C.S.; Arthur, F.H.; Athanassiou, C.G.; Campbell, J.F.; Fleurat-Lessard, F.; Flinn, P.W.; Hodges, R.J.; Isikber, A.A.; Navarro, S.; Noyes, R.T.; Riudavets, J.; Sinha, K.K.; Thorpe, G.R.; Timlick, B.H.; Trematerra, P.; White, N.D.G. (Eds.), Fumigation, Modified Atmospheres and Hermetic Storage. Proceedings of the 10th International Working Conference on Stored Product Protection, 27 June to 2 July 2010, Estoril, Portugal. Julius Kühn-Institut, Berlin, Germany, pp 396- 401.

Necolas, G., Aldasoro, J.J., 1979. Activity of the pentose phosphate pathways and changes in nicotinamide nucleotide content during germination of seeds of *Cicer arietinum* L. Journal of Experimental Botany 30, 1163-1170.

Nisha, R., Fellows, C.M., Sissons, M., 2014. Evaluation of the technological and sensory properties of durum wheat spaghetti enriched with different dietary fibers. Journal of the Science of Food and Agriculture, Wiley Online Library, DOI 10.1002/jsfa.6723.

Nuckolls, J.G., Galax, D.C., Smith, D.C., 1987. Leads from the MMWR. Fatalities resulting from sulfuryl fluoride exposure after home fumigation-Virginia. The Journal of the American Medical Association, 25, 2041-2044.

Opit, G.P., Phillips, T.W., Aikins, M.J., Hasan M.M., 2012. Phosphine resistance in *Tribolium castaneum* and *Rhyzopertha dominica* from stored wheat in Oklahoma. Journal of Economic Entomology 105, 1107-1114.

Orth, R.A., Minett, W., Cook, L.J., 1977. Methyl bromide fumigation. II. Effect of normal dosages on flour and wheat-bread making quality and wheat germination. Cereal Chemistry 54, 713-717.

Orth, R.A., Shellenberger, J.A., 1988. Origin, production, and utilisation of wheat. In Y. Pomeranz, ed. Wheat chemistry and technology. St Paul, MN, USA, American Association of Cereal Chemists.

Osbrink, W.L.A., Scheffrhan, R.H., Hsu, R.C., Su, N.Y., 1988. Sulfuryl fluoride residue of fumigated foods protected by polyethylene film. Journal of Agriculture and Food Chemistry 36, 853-855.

Osbrink, W. L. A., Scheffrahn, R. H., Su, N. Y., Rust, M. K., 1987. Laboratory comparison of sulfuryl fluoride toxicity and mean time of mortality among ten termite species (Isoptera: Hodotermitidae, Kalotermitidae, Rhinotermitidae). Journal of Economic Entomology 80, 1044-1047.

Parpia, H.A.B. 1976. Post harvest losses - Impact of their prevention on food supplies nutrition and development. In: Scrimshaw,N.S. and Behar, M, (Eds.), Nutrition and Agriculture Development. Plenum press, New York, pp. 195-206.

Paster, N., Calderon, M., Meneshervo, M., Barak, V., Mora, M., 1991. Application of bio generated modified atmospheres for insect control in small grain bins. Tropical Sciences 32, 355-358.

Pedersen, J.R., 1992. Insects: identification, damage and detection. In: Sauer, D.B., (Ed), Storage of cereal grain and their products. American Association of Cereal Chemists. St Paul, MN, USA, pp 435-489.

Pepper, J.H., Hastings, E., Douglas, T.A., 1947. The mode of absorption of carbon tetrachloride by wheat. Journal of Economic entomology 40, 67-73.

Pollini, C.M., Panto, F., Nespoli, A., Sissons, M., Abecassis, J., 2012. Manufacture of pasta products, in Durum Wheat: Chemistry and Technology (2nd edn), ed. By Sissons, M., Abecassis, J., Marchylo, B., Carcea, M., AACC International, St Paul, MN, pp. 161-176.

Pomeranz, y., 1992. Biochemical, functional and nutritional changes during storage. In: Sauer, D.B, (Ed). Storage of Cereal Grains and Their products. American Association of Cereal chemists, Inc. St. Paul, MN, pp 55-141.

Powell, D.F., 1975. The effect of methyl bromide fumigation on the germination of onion seed and vigour of the seedlings. Plant Pathology, 24, 237-241.

Prabhakaran, S., 2006. Commercial performance and global development status of profume gas fumigant. In: Lorini, I., Bacaltchuk, B., Beckel, H., Deckers, D., Sundfeld, E., dos Santos, J. P., Biagi, J. D., Celaro, J. C., Faroni, L. R. D'A., Bortolini, L.de O. F., Sartori, M. R., Elias, M. C., Guedes, R. N. C., da Fonseca, R. G., Scussel, V. M. (Eds), Stored Products Protection. Proceedings of the 9th International Working Conference on Stored Product Protection, 15 to 18 October, Campinas, São Paulo, Brazil. Brazilian Post-harvest Association - ABRAPOS, Passo Fundo, RS, Brazil, pp. 635-641.

Rajendran, S, 2003. Grain storage: perspectives and problems. In: Chakraverty, A. Mujumdar, A.S. Vijaya Raghavan, G.S and Ramaswamy, H.S (Eds), Handbook of postharvest technology. Cereals, fruits, vegetables, tea and spices. Marcel Dekker AG, Hutgasse 4, Postfach 812, CH-4001 Basel, Switzerland, pp.183-214.

Reed, C., and Pan, H., 2000. Loss of phosphine from unsealed bins of wheat at six combinations of grain temperatures and grain moisture content. Journal of Stored Product Research 36, 263-279.

Reichmuth, Ch., Rassmann,W., Binker, G., Froba, G., Drinkall, M.J., 2003. Disinfestation of rust-red flour beetle (*Tribolium castaneum*), saw-toothed grain beetle (*Oryzaephilus surinamensis*), yellow meal worm (*Tenebrio molitor*), Mediterranean flourmoth (*Ephestia kuehniella*) and Indian meal moth (*Plodia interpunctella*) with sulfuryl fluoride. In: Credland, P.F., Armitage, D.M., Bell, C.H., Cogan, P.M., Highley, E. (Eds), Advances in Stored Product Protection. Proceedings of the 8th International Working Conference on Stored Product Protection, 22–26 July 2002, New York, UK, CAB International, Oxon, UK, pp. 736–738.

Ren, Y., Graver, J., Van, S., 1996. Grain drying in China: problems and priorities. In: Champ, B.R., Highley, E., Johnson, G.I., (Eds). Grain Drying in Asia. Proceedings of an International Conference held at the FAO Regional Office for Asia and the Pacific, 17-20 October 1995, Bangkok, Thailand, Australian Centre for International Agricultural Research, (ACIAR), Canberra, pp. 23-29.

Ruthven, D.M., 1984. Principles of adsorption and adsorption process. John Wiley and Sons.

Sample, R.L., 1992. Post harvest technology: Storage development and application in developing Asian countries. In: Sample, R.L., Hicks, P.A., Lozare, J.V., and Castermans, A. (Eds.), Grain Storage Systems in Selected Asian Countries, REAPASIA, pp. 9-59.

Sablani, S.S., Ramaswamy, H.S., 2003. Physical and thermal properties of cereal grains. In: Chakraverty, A. Mujumdar, A.S. Vijaya Raghavan, G.S and Ramaswamy, H.S (Eds), Handbook of postharvest technology. Cereals, fruits, vegetables, tea and spices. Marcel Dekker AG, Hutgasse 4, Postfach 812, CH-4001 Basel, Switzerland, pp17-40.

SAS Institute Inc, 1999. version 8, Cary, NC, USA.

Sato, K., Suwanai, M., 1974. Adsorption of hydrogen phosphide to cereal products. Applied Entomology Zoology 9,127-132.

Sawant, A.A., Patil, S.C., Kalse, S.B., Thakor, N.J., 2012. Effect of temperature, relative humidity and moisture content on germination percentage of wheat stored in different storage structures. Agriculture Engineering International: CIGR Journal, 14, 110-118.

Schefferahn, R. H., Hus, R.C., Osbrink, W.L.A., Su, N.Y., 1989. Fluoride and sulfate residues in foods fumigated with sulfuryl fluoride. Journal of Agriculture and Food Chemistry 37, 853-855.

Scheuerman, E.H., 1986. Suicide by exposure to sulfuryl fluoride. Journal of Forensic Sciences 31, 1154-1158.

Schneider, B., Voglewede, C., Houtman, B., 2003. The technical foundation for precision stored product pest fumigation with ProFume gas fumigant. In: Credland, P.F., Armitage, D.M., Bell, C.H., Cogan, P.M., Highley, E. (Eds.), Chemical and Physical Control. Proceedings of the 8th International Working Conference on Stored Product Protection, 22-26 July 2002, York, UK. CAB International, Wallingford, United Kingdom, pp. 561-564.

Scudamore, K.A., Heuser, S.G., 1971. Ethylene oxide and its persistent reaction products in wheat flour and other commodities: Residues from fumigation or sterilization, and effects of processing. Pest Management Science 2, 80-91.

SF registration in Australia, 2007. http://mbao.org/2010/71Barnekow.pdf. Accessed on 25/11/2014.

Shepard, H.H., Buzicky, A.W., 1939. Further studies on methyl bromide as an insect fumigant. Journal of Economic Entomology, 42, 854.

Shi, M., Collins, P.J., Smith, J.R., Renton, M., 2012. Individual-based modelling of the efficacy of fumigation tactics to control lesser grain borer (*Rhyzopertha dominica*) in stored grain. Journal of Stored Products Research 51, 23-32.

Sinclair, W.B., Lindgren, D.L., Forbes, R., 1964. Recovery of ethylene chlorobromide from agricultural products. Journal of Economic Entomology 57, 346-348.

Sissons, M.J., 2004. Pasta. In: Wrigley, C, Corke, H and Walker, C. (Eds), Encyclopedia of Grain Science, Elsevier Australia, pp. 409-418.

Sissons, M., Abecassis, J., Marchylo, B., 2012. Methods used to assess and predict quality of durum wheat, semolina, and pasta. In: Sissons, M., Abecassis, J., Marchylo, B., Carcea, M.

(Eds), Durum wheat, chemistry and technology. American Association of Cereal Chemists, St Paul, MN, pp 219-234.

Snelson, 1987. Grain protectants. Australian Centre for International Agricultural Research. Monograph number 3. ACIAR, Canberra, 1-456.

Spectrum Commodities, http://www.spectrumcommodities.com/education/commodity/statistics/wheat.html. Accessed on 14.03.2015

Sriranjini, V., Rajendran, S., 2008. Sorption of sulfuryl fluoride by food commodities. Pest Management Science 64, 873-879.

Stewart, D., 1957. Sulfuryl fluoride – a new fumigant for the control of the drywood termite, *Kalotermes minor* Hagen. Journal of Economic Entomology 50, 183-195.

Strong, R.G., Lindgren, D.L., 1959. Effect of methyl bromide and hydrocyanic acid fumigation on the germination of rice. Journal of Economic Entomology. 52, 4, 706-710.

Su, N. Y., Scheffrahn, R. H., 1990. Efficacy of sulfuryl fluoride against four beetle pests of museums (Coleoptera: Dermestidae, Anobiidae). Journal of Economic Entomology 83, 879-882.

Su, N.Y., Osbrink, W. L.A., Scheffrahn, R.H., 1989. Concentration time relationship for fumigant efficacy of sulfuryl fluoride against the formosan subterranean termite (Isoptera: Rhinotermitidae). Journal of Economic Entomology 82, 156-158.

Sun, Y.P.,1946. An analysis of some important factors affecting the results of fumigation on insects. St. Paul, Minnesota Agricultural Experiment Station. Technical Bulletin No 177.

Taxay, E.P., 1966. Vikane inhalation. Journal of Occupational and Environmental Medicine 56, 425-426.

Thain, W., 1980. Monitoring toxic gases in the atmosphere for hygiene and pollution control. Oxford, Pergamum Press, 1-159.

Torkelson, T.R., 1959. Summary report of toxicological studies with Vikane (sulfuryl fluoride,40 SO_2F_2). Single exposures of rats to Vikane. The Dow Chemical Company. Midland, MI, 27-31.

USDA, 2014. Treatment manual, 1-920. https://www.aphis.usda.gov/import_export/plants/manuals/ports/downloads/treatment.pdf

USDA, 2012. Foreign agricultural service. Global agricultural information network. Iraq, grain and feed annual, pp 1-10.

http://gain.fas.usda.gov/Recent%20GAIN%20Publications/Grain%20and%20Feed%20Annual_Baghdad_Iraq_4-19-2012.pdf

Vardell, H.H., 1975. Methyl bromide. Effect of multiple fumigation on residues in flour. Journal of Economic. Entomology 68, 69-70.

Warrick, C., 2011. Fumigating with phosphine, other fumigants and controlled atmospheres, GRDC, Grain Research and Development Corporation, stored grain project, 1-16. http://www.grdc.com.au/uploads/documents/GRDC-Fumigating-with-Phosphine-other-fumigants-and-controlled-atmospheres.pdf?shortcut=1

Weber, W.J, McGinley P.M and Katz L.E.1991. Sorption phenomena in subsurface systems: Concepts, models and effects on contaminants fate and transport. Water Research 25, 499-528.

Weller G.L., 2003. Sorption of carbonyl sulphied by stored products. In: Credland, P.F., Armitage, D.M., Bell, C.H., Cogan, P.M., Highley, E. (Eds), Stored Products Protection. Proceedings of the 8th International Working Conference on Stored-Product Protection, 22-26 July 2002, York, UK. CAB International, Wallingford, United Kingdom, pp. 493-497.

Whitney, W.K., Jatz, K.O., Bulger, S.C., 1958. Effects of methyl bromide fumigation on the viability of barley, corn, grain sorghum, oats and wheat seeds. Journal of Economic Entomology 51, 847-861.

Wilbur, D.A., Halazon, G., 1965. Pests of farm stored wheat and their control. Kansas Agricultural Experiment Station Bulletin 481, 1-31.

Williams, L. H., Sprenkel, R. J.,1990. Ovicidal activity of sulfuryl fluoride to anobiid and lyctid beetle egg of various ages. Journal of Entomological Science 25, 366-375.

Wilkin, D.R., Mumford, J.D., Norton, G.A., 1990. The role of expert systems in current and future grain protection. In: Fleurat-Lessard, F.; Ducom, P. (Eds), Modelling in Grain Protection Management. Proceedings of the 5th International Working Conference on Stored Product Protection, 9-14 September 1990, Bordeaux, France. Imprimerie du Médoc, Bordeaux, France, pp. 2039-2046.

Winteringham, F.P.W., Harrison, A., Bridges, R.G., Bridges, P.M., 1955. The fate of labelled insecticide residues in food products. 2. The nature of methyl bromide residues in fumigated wheat. Journal of the Science of Food and Agriculture 6, 251-261.

Winteringham, F.P.W., Harrison, A., 1946. The sorption of methyl bromide by wheat. Journal of the Society of Chemical Industry 65, 140–149.

World Bank, 1986. Poverty and hunger: issues and options for food security in developing countries. A World Bank Policy study. Washington, DC.

Wrigley, C.W., 2009. Wheat: A unique grain for the world. In: Khan, K., Shewry, P.R, (Eds). Wheat chemistry and technology. American Association of Cereal Chemists International. St Paul, Minnesota, USA, pp. 1-17.

Zlatnova J.S., Ivanov, P.V., Stoilov, L.M., Chimshirova, K.V., Stanchev, B.S., 1987. DNA repair precedes replicative synthesis during early germination of maize. Plant Molecular Biology 10, 139-144.

APPENDIX A RAW DATA OF THE THESIS

Table.A.1. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 0 h.

Commodity	At time of 0 (initial concentration)						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	5.00665	6.80665	5.90665
durum	26	4.167x360	15	12	5.935654	4.935654	6.935654
semolina	27	4.167x360	15	12	4.866192	6.066192	5.466192
flour	28	4.167x360	15	12	5.497582	5.897582	5.097582
wheat	29	4.167x360	15	15	5.860323	5.560323	6.160323
durum	30	4.167x360	15	15	6.819351	4.819351	5.819351
semolina	31	4.167x360	15	15	5.033786	5.433786	5.833786
flour	32	4.167x360	15	15	5.416538	5.016538	5.216538
wheat	33	4.167x360	25	12	5.906473	4.906473	6.906473
durum	34	4.167x360	25	12	5.911337	5.911337	5.911337
semolina	35	4.167x360	25	12	5.06691	5.86691	5.46691
flour	36	4.167x360	25	12	5.890933	5.090933	5.490933
wheat	37	4.167x360	25	15	5.855317	5.855317	5.855317
durum	38	4.167x360	25	15	5.813518	4.813518	6.813518
semolina	39	4.167x360	25	15	5.829628	5.029628	5.429628
flour	40	4.167x360	25	15	4.214332	5.214332	6.214332
wheat	41	4.167x360	35	12	5.401295	6.401295	5.901295
durum	42	4.167x360	35	12	5.928083	5.928083	5.928083
semolina	43	4.167x360	35	12	5.166192	5.466192	5.766192
flour	44	4.167x360	35	12	5.447998	5.527998	5.487998
wheat	45	4.167x360	35	15	5.850342	5.050342	6.650342
durum	46	4.167x360	35	15	5.784022	5.384022	6.184022
semolina	47	4.167x360	35	15	5.125497	5.725497	5.425497
flour	48	4.167x360	35	15	5.410495	5.010495	5.210495

T.NO. treatment number in the experimental design.

Table.A.2. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 4 h.

Commodity	After 4 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	5.4164	5.8582	5.7601
durum	26	4.167x360	15	12	5.7185	5.8793	5.8744
semolina	27	4.167x360	15	12	5.475	5.4722	5.0562
flour	28	4.167x360	15	12	5.4046	5.3069	5.1071
wheat	29	4.167x360	15	15	5.4292	5.7443	5.5245
durum	30	4.167x360	15	15	5.9658	4.677	5.6917
semolina	31	4.167x360	15	15	5.3725	3.5609	5.22
flour	32	4.167x360	15	15	4.0014	4.8493	4.8094
wheat	33	4.167x360	25	12	5.5774	5.7861	5.7466
durum	34	4.167x360	25	12	5.7689	4.1145	5.7441
semolina	35	4.167x360	25	12	4.0372	3.613	5.057
flour	36	4.167x360	25	12	4.0072	2.8665	4.8579
wheat	37	4.167x360	25	15	5.2828	1.5909	5.4172
durum	38	4.167x360	25	15	5.3121	4.6496	5.4762
semolina	39	4.167x360	25	15	4.9423	4.4847	4.9043
flour	40	4.167x360	25	15	4.48	4.2856	4.5917
wheat	41	4.167x360	35	12	6.0954	5.8947	5.9821
durum	42	4.167x360	35	12	5.9724	5.9284	5.9079
semolina	43	4.167x360	35	12	5.1123	4.7116	5.0925
flour	44	4.167x360	35	12	4.4906	3.5213	4.3196
wheat	45	4.167x360	35	15	5.1086	4.6943	4.9319
durum	46	4.167x360	35	15	4.7394	4.8073	5.3002
semolina	47	4.167x360	35	15	4.0404	4.0551	4.4111
flour	48	4.167x360	35	15	4.2158	3.411	3.9672

Table.A.3. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 24 h.

Commodity	After 24 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	5.3668	4.4983	5.5275
durum	26	4.167x360	15	12	5.5842	5.45	5.5471
semolina	27	4.167x360	15	12	4.5156	4.3269	4.2627
flour	28	4.167x360	15	12	3.8792	3.4716	4.0332
wheat	29	4.167x360	15	15	5.0766	4.2814	4.4446
durum	30	4.167x360	15	15	4.8278	3.8415	4.7285
semolina	31	4.167x360	15	15	3.9507	2.2297	3.7419
flour	32	4.167x360	15	15	2.7174	2.6361	3.1023
wheat	33	4.167x360	25	12	5.161	5.2978	5.1461
durum	34	4.167x360	25	12	5.0456	3.727	5.2886
semolina	35	4.167x360	25	12	2.8739	2.1561	3.8151
flour	36	4.167x360	25	12	2.0226	1.2375	2.6906
wheat	37	4.167x360	25	15	3.8136	1.028	3.3836
durum	38	4.167x360	25	15	4.9487	3.3758	3.815
semolina	39	4.167x360	25	15	4.0776	2.0826	2.4573
flour	40	4.167x360	25	15	1.3534	1.1649	1.6257
wheat	41	4.167x360	35	12	5.171	4.8369	4.6226
durum	42	4.167x360	35	12	5.1589	4.7821	4.6261
semolina	43	4.167x360	35	12	4.5112	2.0781	2.4998
flour	44	4.167x360	35	12	0.7406	0.6337	1.0719
wheat	45	4.167x360	35	15	2.551	2.0261	2.1087
durum	46	4.167x360	35	15	2.3936	2.3025	2.8852
semolina	47	4.167x360	35	15	1.1659	1.1306	1.1926
flour	48	4.167x360	35	15	0.361	0.3178	0.5226

Table.A.4. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 48 h.

Commodity	After 48 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	5.3348	4.5719	5.4576
durum	26	4.167x360	15	12	5.4755	4.8492	5.3994
semolina	27	4.167x360	15	12	4.1358	3.4246	3.8255
flour	28	4.167x360	15	12	2.702	2.038	2.9809
wheat	29	4.167x360	15	15	3.8265	3.2187	3.6244
durum	30	4.167x360	15	15	4.2955	2.7782	4.1103
semolina	31	4.167x360	15	15	3.1412	1.1594	2.6362
flour	32	4.167x360	15	15	0.165	1.0491	1.8737
wheat	33	4.167x360	25	12	4.1424	4.6541	4.7969
durum	34	4.167x360	25	12	4.7276	3.0113	4.7065
semolina	35	4.167x360	25	12	2.0465	1.4015	2.4666
flour	36	4.167x360	25	12	0.7582	0.518	1.2918
wheat	37	4.167x360	25	15	2.1152	0.5684	2.1612
durum	38	4.167x360	25	15	3.2777	2.2405	2.5998
semolina	39	4.167x360	25	15	2.0263	0.7743	1.0119
flour	40	4.167x360	25	15	0.2652	0.3943	0.4869
wheat	41	4.167x360	35	12	1.1196	3.4804	3.4192
durum	42	4.167x360	35	12	4.1751	3.6136	3.5024
semolina	43	4.167x360	35	12	1.9934	0.7368	1.1017
flour	44	4.167x360	35	12	0.1761	0	0
wheat	45	4.167x360	35	15	1.0284	0.9311	0.7639
durum	46	4.167x360	35	15	1.1624	1.0775	1.3892
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.5. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 72 h.

Commodity	After 72 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	5.2929	4.947	5.3587
durum	26	4.167x360	15	12	5.1072	4.8684	5.2103
semolina	27	4.167x360	15	12	3.6082	2.9455	3.3087
flour	28	4.167x360	15	12	1.6422	1.3176	2.1419
wheat	29	4.167x360	15	15	3.3641	2.8478	3.0915
durum	30	4.167x360	15	15	3.5388	2.7326	3.4551
semolina	31	4.167x360	15	15	2.381	0.8016	2.05
flour	32	4.167x360	15	15	0.7273	0.5089	1.3729
wheat	33	4.167x360	25	12	4.0145	4.276	4.3922
durum	34	4.167x360	25	12	4.0793	3.0576	4.4065
semolina	35	4.167x360	25	12	1.467	0.9932	1.9845
flour	36	4.167x360	25	12	0.2978	0.2997	0.8622
wheat	37	4.167x360	25	15	1.6237	0.3873	1.5116
durum	38	4.167x360	25	15	2.1034	1.6556	2.0115
semolina	39	4.167x360	25	15	1.0017	0.4154	0.7131
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	2.5976	3.2281	2.6583
durum	42	4.167x360	35	12	3.0356	3.2233	2.8327
semolina	43	4.167x360	35	12	0.9806	0.404	0.7026
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0.3625	0	0
durum	46	4.167x360	35	15	0.613	0.4736	0.9205
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.6. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 96 h.

Commodity	After 96 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	4.7296	4.8133	5.0989
durum	26	4.167x360	15	12	5.0643	4.8456	5.1562
semolina	27	4.167x360	15	12	3.3828	2.3447	2.8833
flour	28	4.167x360	15	12	1.545	0.8526	1.94
wheat	29	4.167x360	15	15	2.2908	2.3823	2.5765
durum	30	4.167x360	15	15	3.2957	2.382	3.2516
semolina	31	4.167x360	15	15	2.0581	0.4922	1.5919
flour	32	4.167x360	15	15	0	0.1524	0.8999
wheat	33	4.167x360	25	12	3.4102	3.922	3.9267
durum	34	4.167x360	25	12	4.0105	2.8461	3.8691
semolina	35	4.167x360	25	12	1.2037	0.6026	1.9019
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0.6864	0.0467	1.0105
durum	38	4.167x360	25	15	1.5356	1.2252	1.4509
semolina	39	4.167x360	25	15	0.5211	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	1.8288	2.2932	2.1106
durum	42	4.167x360	35	12	2.5447	2.6244	2.2097
semolina	43	4.167x360	35	12	0.0904	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.7. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 168 h.

Commodity	After 168 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	4.2885	4.5444	4.5012
durum	26	4.167x360	15	12	4.7325	4.3414	4.7297
semolina	27	4.167x360	15	12	2.2182	1.5984	1.88
flour	28	4.167x360	15	12	1.1465	0.4563	0.9383
wheat	29	4.167x360	15	15	1.5874	1.6099	1.4353
durum	30	4.167x360	15	15	2.0681	1.6991	1.9938
semolina	31	4.167x360	15	15	0.8145	0	0.6996
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	2.9708	3.1177	2.7499
durum	34	4.167x360	25	12	2.8961	2.2247	2.7432
semolina	35	4.167x360	25	12	0	0.4332	0.5493
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0.3216	0	0
durum	38	4.167x360	25	15	0.1379	0.7031	0.5968
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	1.2693	1.5457	0.9833
durum	42	4.167x360	35	12	1.2086	1.6534	1.0115
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.8. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 192 h.

Commodity	After 192 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	4.2545	4.408	4.5603
durum	26	4.167x360	15	12	4.5559	4.3809	4.4084
semolina	27	4.167x360	15	12	1.8696	1.2251	1.6411
flour	28	4.167x360	15	12	0.3101	0.2246	0.7921
wheat	29	4.167x360	15	15	1.4158	1.2332	1.2387
durum	30	4.167x360	15	15	1.761	1.3257	1.7892
semolina	31	4.167x360	15	15	0.5963	0	0.5225
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	2.6848	2.7702	2.4114
durum	34	4.167x360	25	12	2.6804	1.944	2.4693
semolina	35	4.167x360	25	12	0	0	0
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0.2406	0	0
durum	38	4.167x360	25	15	0	0.4033	0
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	0.9847	1.0562	0.6984
durum	42	4.167x360	35	12	0.8863	1.1484	0.7816
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.9. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 216 h.

Commodity	After 216 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	4.0944	4.2817	4.2698
durum	26	4.167x360	15	12	4.5956	4.3532	4.5715
semolina	27	4.167x360	15	12	1.7271	1.1105	1.4576
flour	28	4.167x360	15	12	0.2165	0	0.6488
wheat	29	4.167x360	15	15	1.2373	1.0946	1.0608
durum	30	4.167x360	15	15	1.5382	1.2565	1.5691
semolina	31	4.167x360	15	15	0.5468	0	0
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	2.607	2.6945	2.2428
durum	34	4.167x360	25	12	2.3264	1.889	2.0583
semolina	35	4.167x360	25	12	0	0	0
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0	0	0
durum	38	4.167x360	25	15	0	0.1285	0
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	0.7734	0.9233	0.705
durum	42	4.167x360	35	12	0.7885	1.0667	0.5774
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.10. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 240 h.

Commodity	After 240 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	3.6856	3.992	4.2145
durum	26	4.167x360	15	12	4.4569	4.0413	4.4782
semolina	27	4.167x360	15	12	1.6391	0.804	1.217
flour	28	4.167x360	15	12	0.1504	0	0.5664
wheat	29	4.167x360	15	15	1.0733	0.8168	0.9204
durum	30	4.167x360	15	15	1.4947	0.9572	1.479
semolina	31	4.167x360	15	15	0	0	0
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	2.187	2.2359	2.1501
durum	34	4.167x360	25	12	2.2559	1.4746	2
semolina	35	4.167x360	25	12	0	0	0
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0	0	0
durum	38	4.167x360	25	15	0	0.5017	0
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	0.663	0.6408	0.5321
durum	42	4.167x360	35	12	0.7088	0.7467	0.5612
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.11. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 264 h.

Commodity	After 264 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	3.5966	4.0486	4.1725
durum	26	4.167x360	15	12	4.0812	4.0024	4.3716
semolina	27	4.167x360	15	12	1.4439	0.7505	1.2037
flour	28	4.167x360	15	12	0	0	0
wheat	29	4.167x360	15	15	0.8679	0.8036	0.8483
durum	30	4.167x360	15	15	1.2734	0.9074	1.4106
semolina	31	4.167x360	15	15	0	0	0
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	2.0217	2.1919	2.0348
durum	34	4.167x360	25	12	2.0313	1.4817	1.9469
semolina	35	4.167x360	25	12	0	0	0
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0	0	0
durum	38	4.167x360	25	15	0	0	0
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	0.5115	0.609	0.5183
durum	42	4.167x360	35	12	0	0.6977	0.5679
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.12. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 336 h.

Commodity	After 336 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	3.4628	3.651	3.6088
durum	26	4.167x360	15	12	4.0351	3.6953	4.0106
semolina	27	4.167x360	15	12	1.1489	0.4774	0.8604
flour	28	4.167x360	15	12	0	0	0
wheat	29	4.167x360	15	15	0.6119	0.5213	0.5979
durum	30	4.167x360	15	15	0.9841	0.6506	0.9414
semolina	31	4.167x360	15	15	0	0	0
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	1.643	1.657	1.5239
durum	34	4.167x360	25	12	1.6283	1.1288	1.4275
semolina	35	4.167x360	25	12	0	0	0
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0	0	0
durum	38	4.167x360	25	15	0	0	0
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	0.3125	0.3979	0
durum	42	4.167x360	35	12	0	0.4285	0
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.13. Raw data for the applied concentration 4.167 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 360 h.

Commodity	After 360 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	25	4.167x360	15	12	3.288	3.599	3.4297
durum	26	4.167x360	15	12	3.6254	3.6147	3.8048
semolina	27	4.167x360	15	12	0.9577	0.4552	0.7169
flour	28	4.167x360	15	12	0	0	0
wheat	29	4.167x360	15	15	0.5463	0.4534	0
durum	30	4.167x360	15	15	0.7768	0.601	0.8387
semolina	31	4.167x360	15	15	0	0	0
flour	32	4.167x360	15	15	0	0	0
wheat	33	4.167x360	25	12	1.4815	1.5146	1.321
durum	34	4.167x360	25	12	1.4736	1.0612	1.2703
semolina	35	4.167x360	25	12	0	0	0
flour	36	4.167x360	25	12	0	0	0
wheat	37	4.167x360	25	15	0	0	0
durum	38	4.167x360	25	15	0	0	0
semolina	39	4.167x360	25	15	0	0	0
flour	40	4.167x360	25	15	0	0	0
wheat	41	4.167x360	35	12	0.253	0.3252	0
durum	42	4.167x360	35	12	0	0.4131	0
semolina	43	4.167x360	35	12	0	0	0
flour	44	4.167x360	35	12	0	0	0
wheat	45	4.167x360	35	15	0	0	0
durum	46	4.167x360	35	15	0	0	0
semolina	47	4.167x360	35	15	0	0	0
flour	48	4.167x360	35	15	0	0	0

Table.A.14. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 0 h.

Commodity	Concentration time of 0 h (initial concentration)						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	10.6554	14.6554	12.6554
durum	50	8.928x168	15	12	12.71743	10.71743	14.71743
semolina	51	8.928x168	15	12	13.71158	14.71158	6.711583
flour	52	8.928x168	15	12	7.778837	11.77884	15.77884
wheat	53	8.928x168	15	15	14.55603	12.55603	10.55603
durum	54	8.928x168	15	15	10.46824	14.46824	12.46824
semolina	55	8.928x168	15	15	14.64215	8.64215	11.64215
flour	56	8.928x168	15	15	10.17669	12.17669	11.17669
wheat	57	8.928x168	25	12	14.6549	10.6549	12.6549
durum	58	8.928x168	25	12	9.665327	14.66533	12.66533
semolina	59	8.928x168	25	12	13.71312	9.71312	11.71312
flour	60	8.928x168	25	12	14.76459	11.76459	8.76459
wheat	61	8.928x168	25	15	13.5453	8.545302	15.5453
durum	62	8.928x168	25	15	12.45574	14.45574	10.45574
semolina	63	8.928x168	25	15	11.63324	9.633241	13.63324
flour	64	8.928x168	25	15	12.17196	13.17196	8.17196
wheat	65	8.928x168	35	12	13.64381	8.643812	15.64381
durum	66	8.928x168	35	12	12.70121	14.70121	10.70121
semolina	67	8.928x168	35	12	11.71158	9.711583	13.71158
flour	68	8.928x168	35	12	13.7583	14.7583	6.758303
wheat	69	8.928x168	35	15	12.53464	8.534642	16.53464
durum	70	8.928x168	35	15	14.39255	12.39255	10.39255
semolina	71	8.928x168	35	15	10.62439	11.62439	12.62439
flour	72	8.928x168	35	15	13.16374	11.16374	9.163738

Table.A.15. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 4 h.

Commodity	After 4 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	13.1523	12.1799	11.0987
durum	50	8.928x169	15	12	12.3022	12.072	12.2428
semolina	51	8.928x170	15	12	11.6193	10.9265	9.8924
flour	52	8.928x171	15	12	11.5152	10.8123	9.6289
wheat	53	8.928x172	15	15	12.7422	11.5858	10.6283
durum	54	8.928x173	15	15	12.1919	11.4313	10.2918
semolina	55	8.928x174	15	15	11.4099	10.3618	9.3528
flour	56	8.928x175	15	15	11.3559	10.2417	9.3691
wheat	57	8.928x176	25	12	13.0276	12.1451	11.1191
durum	58	8.928x177	25	12	12.8998	11.6625	10.7291
semolina	59	8.928x178	25	12	10.0469	10.9625	10.0937
flour	60	8.928x179	25	12	9.023	9.8355	9.0012
wheat	61	8.928x180	25	15	10.9074	11.4772	9.5297
durum	62	8.928x181	25	15	12.7984	11.8433	10.7928
semolina	63	8.928x182	25	15	10.6186	9.4694	8.5197
flour	64	8.928x183	25	15	8.5316	8.5527	8.5619
wheat	65	8.928x184	35	12	12.9058	11.7558	10.8291
durum	66	8.928x185	35	12	12.7005	11.5476	10.6197
semolina	67	8.928x186	35	12	9.7797	9.9289	9.8291
flour	68	8.928x187	35	12	7.1917	7.5245	7.3799
wheat	69	8.928x188	35	15	10.4598	9.6385	8.5146
durum	70	8.928x189	35	15	11.7361	10.4385	9.5976
semolina	71	8.928x190	35	15	6.7612	6.4385	6.6176
flour	72	8.928x191	35	15	4.7888	4.3095	4.5139

Table.A.16. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 24 h.

Commodity	After 24 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	12.5161	11.7428	10.6461
durum	50	8.928x169	15	12	11.5526	11.8401	11.7892
semolina	51	8.928x170	15	12	10.1091	9.633	8.4223
flour	52	8.928x171	15	12	10.2307	9.6705	8.4458
wheat	53	8.928x172	15	15	11.6052	10.1304	9.9254
durum	54	8.928x173	15	15	10.3073	10.511	10.4462
semolina	55	8.928x174	15	15	9.8849	9.4155	9.6874
flour	56	8.928x175	15	15	8.0674	8.1669	8.1224
wheat	57	8.928x176	25	12	11.9673	11.1237	11.9987
durum	58	8.928x177	25	12	11.3264	10.8458	9.5614
semolina	59	8.928x178	25	12	8.9837	8.2884	8.5891
flour	60	8.928x179	25	12	7.2633	7.6045	7.4625
wheat	61	8.928x180	25	15	8.8384	8.1976	8.7456
durum	62	8.928x181	25	15	9.2902	9.7503	9.5917
semolina	63	8.928x182	25	15	6.6265	6.2649	6.4917
flour	64	8.928x183	25	15	4.3691	4.5053	4.4619
wheat	65	8.928x184	35	12	10.7018	10.9508	10.8219
durum	66	8.928x185	35	12	11.0054	11.6006	11.3548
semolina	67	8.928x186	35	12	6.3095	6.4662	6.3598
flour	68	8.928x187	35	12	3.6078	4.0374	3.8497
wheat	69	8.928x188	35	15	6.1881	6.6948	6.4621
durum	70	8.928x189	35	15	7.1226	7.774	7.4123
semolina	71	8.928x190	35	15	3.1675	3.527	3.3871
flour	72	8.928x191	35	15	1.2115	1.2114	1.1922

Table.A.17. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 48 h.

Commodity	After 48 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	12.1679	11.1729	10.1987
durum	50	8.928x169	15	12	13.5425	11.3148	9.4562
semolina	51	8.928x170	15	12	8.9246	8.2749	8.0012
flour	52	8.928x171	15	12	8.1836	8.0048	8.0193
wheat	53	8.928x172	15	15	10.2729	9.411	8.3729
durum	54	8.928x173	15	15	9.6852	9.3737	9.5176
semolina	55	8.928x174	15	15	7.4782	7.2632	7.3548
flour	56	8.928x175	15	15	5.4308	5.8568	5.4519
wheat	57	8.928x176	25	12	10.8608	10.791	10.7294
durum	58	8.928x177	25	12	10.5703	9.4172	8.5548
semolina	59	8.928x178	25	12	6.2509	6.3944	6.3354
flour	60	8.928x179	25	12	4.0291	4.0952	4.1024
wheat	61	8.928x180	25	15	6.0387	6.3669	6.1528
durum	62	8.928x181	25	15	6.8605	6.9983	6.8921
semolina	63	8.928x182	25	15	3.2129	3.7611	3.4489
flour	64	8.928x183	25	15	1.3783	1.5134	1.4721
wheat	65	8.928x184	35	12	8.5706	8.3692	8.4216
durum	66	8.928x185	35	12	8.9697	8.2604	8.5219
semolina	67	8.928x186	35	12	2.8388	2.2604	2.6378
flour	68	8.928x187	35	12	0.9206	0.9667	0.8921
wheat	69	8.928x188	35	15	2.7701	3.3599	3.0245
durum	70	8.928x189	35	15	3.7403	3.793	3.6921
semolina	71	8.928x190	35	15	0.7722	0.7908	0.7865
flour	72	8.928x191	35	15	0	0	0

Table.A.18. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 72 h.

Commodity	After 72 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	11.0513	10.2994	9.1234
durum	50	8.928x169	15	12	10.6667	10.5308	10.6348
semolina	51	8.928x170	15	12	7.7508	7.9104	7.8921
flour	52	8.928x171	15	12	6.6547	6.9309	6.7256
wheat	53	8.928x172	15	15	8.7559	8.8483	8.7245
durum	54	8.928x173	15	15	8.9836	8.7021	8.8297
semolina	55	8.928x174	15	15	5.8524	5.7198	5.8193
flour	56	8.928x175	15	15	3.7199	4.0228	3.7912
wheat	57	8.928x176	25	12	9.4416	9.2412	9.3344
durum	58	8.928x177	25	12	9.143	8.5879	7.8745
semolina	59	8.928x178	25	12	4.548	4.4816	4.4691
flour	60	8.928x179	25	12	2.5207	2.2561	2.3617
wheat	61	8.928x180	25	15	4.1123	4.5792	4.321
durum	62	8.928x181	25	15	5.0353	5.072	5.1032
semolina	63	8.928x182	25	15	1.7558	1.9648	1.8273
flour	64	8.928x183	25	15	0.4883	0.4955	0.4999
wheat	65	8.928x184	35	12	6.4999	6.2219	6.3591
durum	66	8.928x185	35	12	6.7411	6.5028	6.6672
semolina	67	8.928x186	35	12	1.567	1.5338	1.5554
flour	68	8.928x187	35	12	0	0	0
wheat	69	8.928x188	35	15	1.5075	1.5387	1.4998
durum	70	8.928x189	35	15	2.0875	2.0184	2.0999
semolina	71	8.928x190	35	15	0	0	0
flour	72	8.928x191	35	15	0	0	0

Table.A.19. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 96 h.

Commodity	After 96 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	9.8439	9.4855	9.6154
durum	50	8.928x169	15	12	10.164	9.4473	8.3461
semolina	51	8.928x170	15	12	6.4055	6.9507	6.7321
flour	52	8.928x171	15	12	5.1107	5.519	5.4123
wheat	53	8.928x172	15	15	7.1519	7.2079	7.2654
durum	54	8.928x173	15	15	7.2531	7.4651	7.3615
semolina	55	8.928x174	15	15	4.2428	4.3351	4.3649
flour	56	8.928x175	15	15	2.294	2.3722	2.3124
wheat	57	8.928x176	25	12	8.7672	8.6969	8.6954
durum	58	8.928x177	25	12	7.9112	7.8155	7.9134
semolina	59	8.928x178	25	12	3.1521	3.0889	3.1024
flour	60	8.928x179	25	12	1.3472	1.1909	1.2345
wheat	61	8.928x180	25	15	2.8253	3.229	3.0925
durum	62	8.928x181	25	15	3.8279	3.6476	3.7701
semolina	63	8.928x182	25	15	0.9016	0.8572	0.8995
flour	64	8.928x183	25	15	0	0	0
wheat	65	8.928x184	35	12	4.7035	4.8205	4.7519
durum	66	8.928x185	35	12	4.7972	4.528	4.6691
semolina	67	8.928x186	35	12	0.76	0.7421	0.75564
flour	68	8.928x187	35	12	0	0	0
wheat	69	8.928x188	35	15	0.8012	0.7721	0.8897
durum	70	8.928x189	35	15	1.2497	1.1956	1.1721
semolina	71	8.928x190	35	15	0	0	0
flour	72	8.928x191	35	15	0	0	0

Table.A.20. Raw data for the applied concentration 8.928 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 168 h.

Commodity	After 168 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	49	8.928x168	15	12	9.2283	9.0964	8.5564
durum	50	8.928x169	15	12	10.2566	9.5599	9.3312
semolina	51	8.928x170	15	12	5.3998	5.6299	5.4912
flour	52	8.928x171	15	12	2.5962	3.0145	2.8124
wheat	53	8.928x172	15	15	4.9377	4.7914	4.8891
durum	54	8.928x173	15	15	5.3538	5.8055	5.5674
semolina	55	8.928x174	15	15	1.4733	1.8084	1.6741
flour	56	8.928x175	15	15	0	0	0
wheat	57	8.928x176	25	12	5.1667	5.3279	5.2314
durum	58	8.928x177	25	12	5.9856	6.122	5.7763
semolina	59	8.928x178	25	12	0.5337	0.4925	0.5441
flour	60	8.928x179	25	12	0	0	0
wheat	61	8.928x180	25	15	0.3142	0.4125	0.3998
durum	62	8.928x181	25	15	1.2523	1.3382	1.2879
semolina	63	8.928x182	25	15	0	0	0
flour	64	8.928x183	25	15	0	0	0
wheat	65	8.928x184	35	12	1.8405	1.3549	1.6245
durum	66	8.928x185	35	12	1.609	1.5833	1.5887
semolina	67	8.928x186	35	12	0	0	0
flour	68	8.928x187	35	12	0	0	0
wheat	69	8.928x188	35	15	0	0	0
durum	70	8.928x189	35	15	0	0	0
semolina	71	8.928x190	35	15	0	0	0
flour	72	8.928x191	35	15	0	0	0

Table.A.21. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 0 h.

Commodity	concentration at 0 time (initial concentration)						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	40.29633	48.29633	44.29633
durum	98	31.25x48	15	12	44.51385	40.51385	48.51385
semolina	99	31.25x48	15	12	42.99316	35.99316	43.99316
flour	100	31.25x48	15	12	36.22857	46.22857	41.22857
wheat	101	31.25x48	15	15	43.9489	40.9489	46.9489
durum	102	31.25x48	15	15	37.64164	49.64164	43.64164
semolina	103	31.25x48	15	15	40.75013	35.75013	45.75013
flour	104	31.25x48	15	15	35.1209	43.1209	39.1209
wheat	105	31.25x48	25	12	46.295	40.295	46.295
durum	106	31.25x48	25	12	49.33148	49.33148	34.33148
semolina	107	31.25x48	25	12	35.99854	42.99854	43.99854
flour	108	31.25x48	25	12	36.1787	41.1787	46.1787
wheat	109	31.25x48	25	15	44.91137	40.91137	45.91137
durum	110	31.25x48	25	15	37.59789	49.59789	43.59789
semolina	111	31.25x48	25	15	40.71895	45.71895	35.71895
flour	112	31.25x48	25	15	30.10436	43.10436	44.10436
wheat	113	31.25x48	35	12	40.25617	47.25617	45.25617
durum	114	31.25x48	35	12	48.45707	44.45707	40.45707
semolina	115	31.25x48	35	12	43.99316	35.99316	42.99316
flour	116	31.25x48	35	12	41.1567	40.1567	42.1567
wheat	117	31.25x48	35	15	45.87405	40.87405	44.87405
durum	118	31.25x48	35	15	43.37669	40.37669	46.37669
semolina	119	31.25x48	35	15	45.68797	40.68797	35.68797
flour	120	31.25x48	35	15	41.07559	34.07559	42.07559

Table.A.22. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 4 h.

Commodity	After 4 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	43.8251	43.9454	43.6796
durum	98	31.25x48	15	12	43.6258	48.5696	46.5458
semolina	99	31.25x48	15	12	40.1998	40.5564	40.3068
flour	100	31.25x48	15	12	42.4083	39.961	37.6364
wheat	101	31.25x48	15	15	43.8784	42.3302	43.8446
durum	102	31.25x48	15	15	43.1844	43.4278	43.9537
semolina	103	31.25x48	15	15	43.6932	40.9612	41.4952
flour	104	31.25x48	15	15	42.7314	38.8754	37.8642
wheat	105	31.25x48	25	12	43.996	43.1529	44.5031
durum	106	31.25x48	25	12	43.9006	43.1831	43.8758
semolina	107	31.25x48	25	12	42.9818	42.1586	39.4902
flour	108	31.25x48	25	12	38.0475	39.467	39.8066
wheat	109	31.25x48	25	15	43.6457	43.3461	44.2204
durum	110	31.25x48	25	15	42.6756	42.2354	42.0327
semolina	111	31.25x48	25	15	39.6027	37.2181	38.1722
flour	112	31.25x48	25	15	39.1322	36.4356	35.8017
wheat	113	31.25x48	35	12	43.1285	43.4663	43.8296
durum	114	31.25x48	35	12	43.4994	43.1571	42.9083
semolina	115	31.25x48	35	12	40.4636	39.2213	39.0962
flour	116	31.25x48	35	12	25.4078	33.1036	33.2418
wheat	117	31.25x48	35	15	41.697	45.3696	41.6669
durum	118	31.25x48	35	15	42.9685	40.4814	43.6133
semolina	119	31.25x48	35	15	35.3992	35.1338	32.7686
flour	120	31.25x48	35	15	31.1048	33.5069	30.6536

Table.A.23. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 8 h.

Commodity	After 8 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	43.9585	43.4717	43.6578
durum	98	31.25x48	15	12	42.223	43.4704	44.9671
semolina	99	31.25x48	15	12	39.8278	39.6695	39.792
flour	100	31.25x48	15	12	38.3991	37.0157	39.8098
wheat	101	31.25x48	15	15	45.4891	41.9515	41.4609
durum	102	31.25x48	15	15	43.2663	43.3043	42.8773
semolina	103	31.25x48	15	15	39.1425	37.0011	38.6214
flour	104	31.25x48	15	15	36.4315	35.6254	34.5037
wheat	105	31.25x48	25	12	42.153	42.4473	42.7157
durum	106	31.25x48	25	12	42.7809	42.612	42.5523
semolina	107	31.25x48	25	12	41.4642	37.9179	36.2609
flour	108	31.25x48	25	12	31.788	32.1797	33.1604
wheat	109	31.25x48	25	15	34.8041	35.2087	36.0794
durum	110	31.25x48	25	15	42.3083	40.6492	41.0648
semolina	111	31.25x48	25	15	32.6429	32.1637	32.8226
flour	112	31.25x48	25	15	29.3043	28.19	30.3572
wheat	113	31.25x48	35	12	42.4518	42.1603	42.3056
durum	114	31.25x48	35	12	42.8236	42.2549	42.7533
semolina	115	31.25x48	35	12	35.0729	30.3883	31.6222
flour	116	31.25x48	35	12	19.6936	22.381	23.672
wheat	117	31.25x48	35	15	36.1276	37.8779	35.5881
durum	118	31.25x48	35	15	39.8766	37.9797	38.2233
semolina	119	31.25x48	35	15	27.921	22.5602	25.8591
flour	120	31.25x48	35	15	23.5416	17.0177	20.6759

Table.A.24. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 12 h.

Commodity	After 12 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	43.3174	42.4717	44.6872
durum	98	31.25x48	15	12	42.7567	40.4704	44.7408
semolina	99	31.25x48	15	12	38.2173	38.6695	38.5788
flour	100	31.25x48	15	12	35.02	37.0157	36.5952
wheat	101	31.25x48	15	15	43.5325	42..9515	41.2298
durum	102	31.25x48	15	15	41.2662	43.3043	42.293
semolina	103	31.25x48	15	15	38.7051	37.0011	37.3119
flour	104	31.25x48	15	15	34.2361	34.6254	34.9962
wheat	105	31.25x48	25	12	41.8929	41.9473	41.8039
durum	106	31.25x48	25	12	41.0956	41.612	41.8706
semolina	107	31.25x48	25	12	36.2603	37.9179	35.6241
flour	108	31.25x48	25	12	29.6254	32.1797	31.9956
wheat	109	31.25x48	25	15	34.1599	33.2087	32.4374
durum	110	31.25x48	25	15	40.2781	40.6492	40.1315
semolina	111	31.25x48	25	15	31.6673	28.1637	31.4813
flour	112	31.25x48	25	15	26.8218	26.19	24.8018
wheat	113	31.25x48	35	12	41.068	41.1603	41.4177
durum	114	31.25x48	35	12	41.9694	41.2549	41.8843
semolina	115	31.25x48	35	12	30.6075	29.3883	28.2158
flour	116	31.25x48	35	12	19.2508	19.381	19.0865
wheat	117	31.25x48	35	15	33.4675	34.8779	35.786
durum	118	31.25x48	35	15	34.2577	35.9797	33.2031
semolina	119	31.25x48	35	15	21.2255	22.5602	20.5736
flour	120	31.25x48	35	15	16.231	14.0177	15.7037

Table.A.25. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 24 h.

commodity	After 24 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	43.0232	43.0345	43.3804
durum	98	31.25x48	15	12	40.8028	42.2939	41.3642
semolina	99	31.25x48	15	12	37.7567	38.4429	36.9128
flour	100	31.25x48	15	12	31.7835	32.7406	30.7206
wheat	101	31.25x48	15	15	38.7445	38.5519	38.4006
durum	102	31.25x48	15	15	40.4783	39.1023	38.4961
semolina	103	31.25x48	15	15	35.3663	34.8521	33.653
flour	104	31.25x48	15	15	25.3444	25.3569	25.0592
wheat	105	31.25x48	25	12	40.3564	40.7797	40.5263
durum	106	31.25x48	25	12	40.9051	40.4294	39.5034
semolina	107	31.25x48	25	12	30.1844	29.6594	28.6669
flour	108	31.25x48	25	12	19.9258	20.3582	21.5736
wheat	109	31.25x48	25	15	27.4761	26.4333	25.0245
durum	110	31.25x48	25	15	33.5019	33.8149	34.6092
semolina	111	31.25x48	25	15	19.7046	18.014	20.9776
flour	112	31.25x48	25	15	14.8945	14.0124	14.373
wheat	113	31.25x48	35	12	40.8633	38.1867	38.6149
durum	114	31.25x48	35	12	36.2903	38.0536	38.9683
semolina	115	31.25x48	35	12	19.3708	19.5428	18.6841
flour	116	31.25x48	35	12	5.3319	9.3337	8.8315
wheat	117	31.25x48	35	15	22.4213	26.8852	23.0469
durum	118	31.25x48	35	15	26.0951	27.7564	30.6958
semolina	119	31.25x48	35	15	7.8725	10.9191	9.6748
flour	120	31.25x48	35	15	3.0521	4.5237	3.9778

Table.A.26. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 28 h.

Commodity	After 28 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	43.6841	42.7992	41.9542
durum	98	31.25x48	15	12	40.6706	40.6947	40.5577
semolina	99	31.25x48	15	12	36.0605	36.8423	36.3512
flour	100	31.25x48	15	12	32.289	29.0057	29.7367
wheat	101	31.25x48	15	15	38.5851	37.7602	36.7411
durum	102	31.25x48	15	15	40.6583	38.6964	38.7732
semolina	103	31.25x48	15	15	33.175	33.407	33.0294
flour	104	31.25x48	15	15	26.1606	25.3916	24.7251
wheat	105	31.25x48	25	12	39.5146	39.2974	39.8654
durum	106	31.25x48	25	12	40.509	39.1816	38.2043
semolina	107	31.25x48	25	12	29.2813	28.4219	27.9824
flour	108	31.25x48	25	12	18.3352	19.994	20.0661
wheat	109	31.25x48	25	15	26.75	25.5461	24.1413
durum	110	31.25x48	25	15	33.8266	33.7953	33.8838
semolina	111	31.25x48	25	15	18.0956	17.7217	19.3404
flour	112	31.25x48	25	15	11.8724	13.5951	11.4701
wheat	113	31.25x48	35	12	40.7722	37.944	37.6506
durum	114	31.25x48	35	12	35.876	36.2167	37.846
semolina	115	31.25x48	35	12	16.9897	18.4457	16.4823
flour	116	31.25x48	35	12	3.0804	7.6997	6.8023
wheat	117	31.25x48	35	15	19.9201	25.3631	21.3944
durum	118	31.25x48	35	15	24.2057	25.4809	28.0702
semolina	119	31.25x48	35	15	5.7983	8.1484	7.4198
flour	120	31.25x48	35	15	1.1714	3.7742	2.5573

Table.A.27. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 32 h.

Commodity	After 32 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	42.9113	41.1912	42.009
durum	98	31.25x48	15	12	40.7322	40.4958	40.1614
semolina	99	31.25x48	15	12	34.2073	35.3297	36.3109
flour	100	31.25x48	15	12	30.1666	29.1441	28.0963
wheat	101	31.25x48	15	15	38.2534	37.1516	36.2842
durum	102	31.25x48	15	15	38.1884	36.0934	37.2615
semolina	103	31.25x48	15	15	29.3672	29.5435	29.1436
flour	104	31.25x48	15	15	25.4914	23.0043	22.1562
wheat	105	31.25x48	25	12	37.8885	38.0735	39.304
durum	106	31.25x48	25	12	39.486	38.3125	37.7715
semolina	107	31.25x48	25	12	28.7214	27.0439	26.3823
flour	108	31.25x48	25	12	16.9809	18.283	17.6932
wheat	109	31.25x48	25	15	25.8878	24.1863	23.5479
durum	110	31.25x48	25	15	31.8191	31.3209	31.9143
semolina	111	31.25x48	25	15	15.6661	15.7028	15.88
flour	112	31.25x48	25	15	9.8517	9.4063	9.0639
wheat	113	31.25x48	35	12	37.3821	36.6394	35.9893
durum	114	31.25x48	35	12	35.421	35.2582	35.8269
semolina	115	31.25x48	35	12	14.9912	14.3601	14.4819
flour	116	31.25x48	35	12	2.8165	3.4247	4.7289
wheat	117	31.25x48	35	15	19.8095	19.8455	19.7503
durum	118	31.25x48	35	15	23.9717	23.2555	23.7489
semolina	119	31.25x48	35	15	5.8349	5.7504	5.4829
flour	120	31.25x48	35	15	0	2.3144	1.7075

Table.A.28. Raw data for the applied concentration 31.25 mg/L at stated temperatures and moistures showing the calculated concentration of the fumigant at time of 36 h.

Commodity	After 36 hours						
	T. No	SF mg/L x time h	Temperature °C	Moisture %	Replicate1	Replicate2	Replicate3
wheat	97	31.25x48	15	12	43.9571	42.7038	41.152
durum	98	31.25x48	15	12	40.1822	40.8083	40.9929
semolina	99	31.25x48	15	12	35.7854	34.1739	33.0895
flour	100	31.25x48	15	12	29.9438	28.3044	27.2605
wheat	101	31.25x48	15	15	37.1539	35.5369	36.5626
durum	102	31.25x48	15	15	37.6805	36.3568	35.6132
semolina	103	31.25x48	15	15	28.3079	28.8221	28.4057
flour	104	31.25x48	15	15	23.806	22.5338	21.6433
wheat	105	31.25x48	25	12	38.8487	37.9189	36.7541
durum	106	31.25x48	25	12	38.975	37.9339	36.5444
semolina	107	31.25x48	25	12	27.5031	26.6134	25.7586
flour	108	31.25x48	25	12	16.7738	15.3491	14.937
wheat	109	31.25x48	25	15	24.9014	23.1973	22.84
durum	110	31.25x48	25	15	32.1118	31.7079	30.2242
semolina	111	31.25x48	25	15	14.6146	13.6417	12.6257
flour	112	31.25x48	25	15	8.88	8.4952	7.9067
wheat	113	31.25x48	35	12	37.1162	36.6438	35.4665
durum	114	31.25x48	35	12	34.7312	33.3323	32.9459
semolina	115	31.25x48	35	12	13.1525	12.0172	11.3308
flour	116	31.25x48	35	12	0.7632	3.9041	3.7729
wheat	117	31.25x48	35	15	16.6562	19.4789	17.4403
durum	118	31.25x48	35	15	24.6575	23.881	22.0691
semolina	119	31.25x48	35	15	2.2768	4.8503	4.5562
flour	120	31.25x48	35	15	0	1.5169	1.348

Table.A.29. Raw data of applied concentrations to fumigation system has adults and egg of *R. dominica* and wheat of 12% moisture at 25.

Replicates	Exposure time h	Applied SF		
		0.5 mg/L	1 mg/L	2 mg/L
1	0	1	1	1
	24	0.6514	0.5534	0.5771
	48	0.428	0.3563	0.4186
	72	0.3304	0.2538	0.2562
	96	0.2198	0.1961	0.1978
	120	0.1672	0.1458	0.138
	144	0.1206	0.1302	0.1153
	168	0.0806	0.1102	0.1503
2	0	1	1	1
	24	0.6284	0.5403	0.6215
	48	0.4122	0.3603	0.4622
	72	0.3302	0.2885	0.3014
	96	0.2144	0.2594	0.1932
	120	0.1592	0.1923	0.1534
	144	0.117	0.1455	0.1092
	168	0.077	0.1255	0.0942
3	0	1	1	1
	24	0.6414	0.5621	0.5822
	48	0.4242	0.3949	0.3746
	72	0.3288	0.271	0.2117
	96	0.2026	0.2234	0.2034
	120	0.1728	0.1654	0.1441
	144	0.1348	0.1371	0.1302
	168	0.1148	0.1271	0.1101

Table.A.30. Raw data of percentage reduction of *R. dominica* egg survivors at three concentrations of SF and percentage growth of unfumigated egg survivors (control).

Replicates	Exposure time h	Applied SF			Control
		0.5 mg/L	1 mg/L	2 mg/L	
1	0	100	100	100	0
	24	97.59423	95.07476	90.00473	28
	48	95.30874	90.41337	80.98485	45
	72	93.06335	85.97186	77.86932	53
	96	90.85806	81.75022	69.5303	80
	120	88.69286	77.70449	63.99621	85
	144	86.56776	73.91821	53.07765	90
	168	76.56776	63.91821	43.07765	100
2	0	100	100	100	0
	24	95.38746	94.88536	90.22674	20
	48	94.85531	89.99118	81.34152	40
	72	92.36334	85.36155	75.35853	55
	96	89.99196	80.95238	68.17855	75
	120	87.6246	76.80776	59.70713	80
	144	85.32958	72.83951	53.85451	90
	168	75.32958	62.83951	43.85451	100
3	0	100	100	100	0
	24	91.82435	95.34884	90.43355	25
	48	95.74468	90.87319	81.76249	47
	72	90.70501	86.61694	76.89255	50
	96	91.58535	82.5362	68.82375	80
	120	84.74564	78.67486	65.4147	85
	144	87.82595	74.98903	54.61828	90
	168	77.82595	64.98903	44.61828	100

Table.A.31. Raw data of percentage reduction of *R. dominica* adults at three concentrations of SF.

Replicates	Exposure time h	Applied SF		
		0.5 mg/L	1 mg/L	2 mg/L
1	0	100	100	100
	24	95	87	77
	48	89	76	59
	72	84	67	45
	96	80	58	34
	120	75	51	27
	144	72	45	20
	168	68	39	15
2	0	100	100	100
	24	94	85	72
	48	88	77	54
	72	82	65	43
	96	79	54	32
	120	74	50	29
	144	70	42	25
	168	67	35	16
3	0	100	100	100
	24	93	82	78
	48	87	72	62
	72	83	70	47
	96	81	60	36
	120	75	51	25
	144	74	47	21
	168	65	42	13

Table.A.32. Raw data of impacting physical sorption (*K*) on adult mortality constant (*R*) at three concentrations.

Replicates	Applied SF					
	0.5 mg/L		1 mg/L		2 mg/L	
	K	*R*	*K*	*R*	*K*	*R*
1	0.242184	-0.0023	0.105308	-0.0055	0.021648	-0.0109
	1.581619	-0.0552	0.604375	-0.132	0.374942	-0.2616
	3.362566	-0.1104	1.201743	-0.264	0.779178	-0.5232
	6.099315	-0.1656	1.916776	-0.396	1.241684	-0.7848
	10.12867	-0.2208	2.772687	-0.528	1.770893	-1.0464
	16.06215	-0.276	3.797098	-0.66	2.376437	-1.308
	24.79757	-0.3312	5.02332	-0.792	3.06923	-1.5696
	37.67333	-0.3864	6.491146	-0.924	3.861924	-1.8312
2	0.242184	-0.002	0.105308	-0.0059	0.021648	-0.009
	1.581619	-0.048	0.604375	-0.1416	0.374942	-0.216
	3.362566	-0.096	1.201743	-0.2832	0.779178	-0.432
	6.099315	-0.144	1.916776	-0.4248	1.241684	-0.648
	10.12867	-0.192	2.772687	-0.5664	1.770893	-0.864
	16.06215	-0.24	3.797098	-0.708	2.376437	-1.08
	24.79757	-0.288	5.02332	-0.8496	3.06923	-1.296
	37.67333	-0.336	6.491146	-0.9912	3.861924	-1.512
3	0.242184	-0.0025	0.105308	-0.0051	0.021648	-0.0121
	1.581619	-0.06	0.604375	-0.1224	0.374942	-0.2904
	3.362566	-0.12	1.201743	-0.2448	0.779178	-0.5808
	6.099315	-0.18	1.916776	-0.3672	1.241684	-0.8712
	10.12867	-0.24	2.772687	-0.4896	1.770893	-1.1616
	16.06215	-0.3	3.797098	-0.612	2.376437	-1.452
	24.79757	-0.36	5.02332	-0.7344	3.06923	-1.7424
	37.67333	-0.42	6.491146	-0.8568	3.861924	-2.0328

Table.A.33. Raw data of impacting physical sorption (*k*) on adult mortality constant (*R*) at three concentrations.

Replicates	Applied SF					
	0.5 mg/L		1 mg/L		2 mg/L	
	k	*R*	*k*	*R*	*k*	*R*
1	-0.0161	-0.0023	-0.00749	-0.0055	-0.00561	-0.0109
	-0.3864	-0.0552	-0.17976	-0.132	-0.13464	-0.2616
	-0.7728	-0.1104	-0.35952	-0.264	-0.26928	-0.5232
	-1.1592	-0.1656	-0.53928	-0.396	-0.40392	-0.7848
	-1.5456	-0.2208	-0.71904	-0.528	-0.53856	-1.0464
	-1.932	-0.276	-0.8988	-0.66	-0.6732	-1.308
	-2.3184	-0.3312	-1.07856	-0.792	-0.80784	-1.5696
	-2.7048	-0.3864	-1.25832	-0.924	-0.94248	-1.8312
2	-0.0149	-0.002	-0.007	-0.0059	-0.00591	-0.009
	-0.3576	-0.048	-0.168	-0.1416	-0.14184	-0.216
	-0.7152	-0.096	-0.336	-0.2832	-0.28368	-0.432
	-1.0728	-0.144	-0.504	-0.4248	-0.42552	-0.648
	-1.4304	-0.192	-0.672	-0.5664	-0.56736	-0.864
	-1.788	-0.24	-0.84	-0.708	-0.7092	-1.08
	-2.1456	-0.288	-1.008	-0.8496	-0.85104	-1.296
	-2.5032	-0.336	-1.176	-0.9912	-0.99288	-1.512
3	-0.0169	-0.0025	-0.00788	-0.0051	-0.00531	-0.0121
	-0.4056	-0.06	-0.18912	-0.1224	-0.12744	-0.2904
	-0.8112	-0.12	-0.37824	-0.2448	-0.25488	-0.5808
	-1.2168	-0.18	-0.56736	-0.3672	-0.38232	-0.8712
	-1.6224	-0.24	-0.75648	-0.4896	-0.50976	-1.1616
	-2.028	-0.3	-0.9456	-0.612	-0.6372	-1.452
	-2.4336	-0.36	-1.13472	-0.7344	-0.76464	-1.7424
	-2.8392	-0.42	-1.32384	-0.8568	-0.89208	-2.0328

Table.A.34. Raw data of impacting physical sorption (*K*) on egg mortality constant (*E*) at three concentrations.

Replicates	Applied SF					
	0.5 mg/L		1 mg/L		2 mg/L	
	K	*E*	*K*	*E*	*K*	*E*
1	0.242184	-0.001	0.105308	-0.0021	0.021648	-0.0044
	1.581619	-0.024	0.604375	-0.0504	0.374942	-0.1056
	3.362566	-0.048	1.201743	-0.1008	0.779178	-0.2112
	6.099315	-0.072	1.916776	-0.1512	1.241684	-0.3168
	10.12867	-0.096	2.772687	-0.2016	1.770893	-0.4224
	16.06215	-0.12	3.797098	-0.252	2.376437	-0.528
	24.79757	-0.144	5.02332	-0.3024	3.06923	-0.6336
	37.67333	-0.168	6.491146	-0.3528	3.861924	-0.7392
2	0.242184	-0.0011	0.105308	-0.0022	0.021648	-0.0043
	1.581619	-0.0264	0.604375	-0.0528	0.374942	-0.1032
	3.362566	-0.0528	1.201743	-0.1056	0.779178	-0.2064
	6.099315	-0.0792	1.916776	-0.1584	1.241684	-0.3096
	10.12867	-0.1056	2.772687	-0.2112	1.770893	-0.4128
	16.06215	-0.132	3.797098	-0.264	2.376437	-0.516
	24.79757	-0.1584	5.02332	-0.3168	3.06923	-0.6192
	37.67333	-0.1848	6.491146	-0.3696	3.861924	-0.7224
3	0.242184	-0.0009	0.105308	-0.002	0.021648	-0.0042
	1.581619	-0.0216	0.604375	-0.048	0.374942	-0.1008
	3.362566	-0.0432	1.201743	-0.096	0.779178	-0.2016
	6.099315	-0.0648	1.916776	-0.144	1.241684	-0.3024
	10.12867	-0.0864	2.772687	-0.192	1.770893	-0.4032
	16.06215	-0.108	3.797098	-0.24	2.376437	-0.504
	24.79757	-0.1296	5.02332	-0.288	3.06923	-0.6048
	37.67333	-0.1512	6.491146	-0.336	3.861924	-0.7056

Table.A.35. Raw data of impacting physical sorption (k) on egg mortality constant (E) at three concentrations.

Replicates	Applied SF					
	0.5 mg/L		1 mg/L		2 mg/L	
	k	*E*	*k*	*E*	*k*	*E*
1	-0.0161	-0.001	-0.00749	-0.0021	-0.00561	-0.0044
	-0.3864	-0.024	-0.17976	-0.0504	-0.13464	-0.1056
	-0.7728	-0.048	-0.35952	-0.1008	-0.26928	-0.2112
	-1.1592	-0.072	-0.53928	-0.1512	-0.40392	-0.3168
	-1.5456	-0.096	-0.71904	-0.2016	-0.53856	-0.4224
	-1.932	-0.12	-0.8988	-0.252	-0.6732	-0.528
	-2.3184	-0.144	-1.07856	-0.3024	-0.80784	-0.6336
	-2.7048	-0.168	-1.25832	-0.3528	-0.94248	-0.7392
2	-0.0149	-0.0011	-0.007	-0.0022	-0.00591	-0.0043
	-0.3576	-0.0264	-0.168	-0.0528	-0.14184	-0.1032
	-0.7152	-0.0528	-0.336	-0.1056	-0.28368	-0.2064
	-1.0728	-0.0792	-0.504	-0.1584	-0.42552	-0.3096
	-1.4304	-0.1056	-0.672	-0.2112	-0.56736	-0.4128
	-1.788	-0.132	-0.84	-0.264	-0.7092	-0.516
	-2.1456	-0.1584	-1.008	-0.3168	-0.85104	-0.6192
	-2.5032	-0.1848	-1.176	-0.3696	-0.99288	-0.7224
3	-0.0169	-0.0009	-0.00788	-0.002	-0.00531	-0.0042
	-0.4056	-0.0216	-0.18912	-0.048	-0.12744	-0.1008
	-0.8112	-0.0432	-0.37824	-0.096	-0.25488	-0.2016
	-1.2168	-0.0648	-0.56736	-0.144	-0.38232	-0.3024
	-1.6224	-0.0864	-0.75648	-0.192	-0.50976	-0.4032
	-2.028	-0.108	-0.9456	-0.24	-0.6372	-0.504
	-2.4336	-0.1296	-1.13472	-0.288	-0.76464	-0.6048
	-2.8392	-0.1512	-1.32384	-0.336	-0.89208	-0.7056

Table.A.36. Raw data of 1 mg/L applied SF to durum wheat at 0.50, 0.75 and 0.95 filling ratios.

Replicates	Exposure time h	Filling ratios		
		0.95	0.75	0.50
1	0	1	1	1
	24	0.31	0.41	0.54
	48	0.27	0.33	0.49
	72	0.21	0.28	0.43
	96	0.16	0.24	0.37
	120	0.12	0.2	0.34
	144	0.065	0.15	0.28
	168	0.061	0.14	0.23
2	0	1	1	1
	24	0.3	0.43	0.53
	48	0.25	0.32	0.49
	72	0.19	0.22	0.44
	96	0.11	0.23	0.4
	120	0.061	0.19	0.36
	144	0.041	0.15	0.29
	168	0.025	0.12	0.245
3	0	1	1	1
	24	0.315	0.41	0.55
	48	0.27	0.3	0.46
	72	0.21	0.25	0.41
	96	0.15	0.22	0.38
	120	0.09	0.18	0.35
	144	0.07	0.14	0.31
	168	0.05	0.11	0.25

Table.A.37. Raw data of the relationships between time and the chemical sorption constant (*k*) at 0.50, 0.75 and 0.95 filling ratios for bulk density and grain density.

Replicates	Exposure time h	Filling ratio					
		0.50		0.75		0.95	
		Bulk density	Grain density	Bulk density	Grain density	Bulk density	Grain density
1	0	-0.005	-0.00023	-0.009	-0.00027	-0.0144	-0.00031
	24	-0.129	-0.00561	-0.225	-0.00653	-0.3456	-0.00752
	48	-0.259	-0.01122	-0.45	-0.01306	-0.6912	-0.01503
	72	-0.388	-0.01683	-0.675	-0.01959	-1.0368	-0.02255
	96	-0.517	-0.02244	-0.9	-0.02611	-1.3824	-0.03007
	120	-0.647	-0.02805	-1.126	-0.03264	-1.728	-0.03758
	144	-0.776	-0.03366	-1.351	-0.03917	-2.0736	-0.0451
	168	-0.906	-0.03927	-1.576	-0.0457	-2.4192	-0.05262
2	0	-0.006	-0.00024	-0.011	-0.00031	-0.0153	-0.00033
	24	-0.132	-0.00574	-0.259	-0.00752	-0.3672	-0.00799
	48	-0.264	-0.01147	-0.518	-0.01503	-0.7344	-0.01597
	72	-0.397	-0.01721	-0.778	-0.02255	-1.1016	-0.02396
	96	-0.529	-0.02294	-1.037	-0.03007	-1.4688	-0.03195
	120	-0.661	-0.02868	-1.296	-0.03758	-1.836	-0.03993
	144	-0.793	-0.03441	-1.555	-0.0451	-2.2032	-0.04792
	168	-0.926	-0.04015	-1.814	-0.05262	-2.5704	-0.05591
3	0	-0.005	-0.00023	-0.009	-0.00025	-0.0131	-0.00028
	24	-0.126	-0.00544	-0.206	-0.00599	-0.3144	-0.00684
	48	-0.251	-0.01089	-0.413	-0.01197	-0.6288	-0.01368
	72	-0.377	-0.01633	-0.619	-0.01796	-0.9432	-0.02051
	96	-0.502	-0.02178	-0.826	-0.02394	-1.2576	-0.02735
	120	-0.628	-0.02722	-1.032	-0.02993	-1.572	-0.03419
	144	-0.753	-0.03266	-1.238	-0.03591	-1.8864	-0.04103
	168	-0.879	-0.03811	-1.445	-0.0419	-2.2008	-0.04787

Table.A.38. Raw data of the relationships between time and the physical sorption (*K*) at 0.50, 0.75 and 0.95 filling ratios for bulk density and grain density.

Replicates	Exposure time h	Filling ratio					
		0.50		0.75		0.95	
		Bulk density	Grain density	Bulk density	Grain density	Bulk density	Grain density
1	0	2.0452	0.088701	1.3369	0.038771	0.863609	0.018783
	24	2.7964	0.121277	1.9248	0.055821	1.535028	0.033387
	48	3.6515	0.158363	2.6436	0.076664	2.482643	0.053997
	72	4.6244	0.200558	3.5225	0.102152	3.820178	0.083089
	96	5.7317	0.248583	4.5976	0.133329	5.707411	0.124136
	120	6.9918	0.303235	5.9118	0.171442	8.371955	0.18209
	144	8.4261	0.365438	7.5195	0.218067	12.13083	0.263846
	168	10.058	0.436209	9.4848	0.275059	17.43741	0.379264
2	0	1.9342	0.083885	1.2341	0.035789	0.797112	0.017337
	24	2.6529	0.115058	1.7724	0.0514	1.404782	0.030554
	48	3.4663	0.150334	2.418	0.070122	2.229135	0.048484
	72	4.3855	0.190198	3.1894	0.092493	3.330928	0.072448
	96	5.4236	0.235221	4.107	0.119102	4.773976	0.103834
	120	6.5947	0.286012	5.1916	0.150555	6.616566	0.14391
	144	7.9145	0.34325	6.4658	0.187509	8.890832	0.193376
	168	9.3993	0.407646	7.95	0.230551	15.58789	0.292037
3	0	2.1519	0.093329	1.4484	0.042004	0.899021	0.019554
	24	2.9349	0.127286	2.0932	0.060703	1.606185	0.034935
	48	3.8314	0.166169	2.8983	0.08405	2.626182	0.057119
	72	4.8582	0.210701	3.909	0.11336	4.111365	0.089422
	96	6.0358	0.261772	5.1861	0.150396	6.302855	0.137087
	120	7.3875	0.320394	6.8118	0.197543	9.604751	0.208903
	144	8.9412	0.387779	8.9041	0.258218	14.73008	0.320379
	168	10.729	0.465315	11.63	0.337275	20.08289	0.402053

Table.A.39. Raw data of the effect of the physical sorption (*K*) on chemical sorption (*k*) at 0.50 filling ratio for bulk density and grain density.

Replicates	Bulk Density		Grain density	
	K	*k*	*K*	*k*
1	2.0452	-0.005	0.088701	-0.00023
	2.7964	-0.129	0.121277	-0.00561
	3.6515	-0.259	0.158363	-0.01122
	4.6244	-0.388	0.200558	-0.01683
	5.7317	-0.517	0.248583	-0.02244
	6.9918	-0.647	0.303235	-0.02805
	8.4261	-0.776	0.365438	-0.03366
	10.058	-0.906	0.436209	-0.03927
2	1.9342	-0.006	0.083885	-0.00024
	2.6529	-0.132	0.115058	-0.00574
	3.4663	-0.264	0.150334	-0.01147
	4.3855	-0.397	0.190198	-0.01721
	5.4236	-0.529	0.235221	-0.02294
	6.5947	-0.661	0.286012	-0.02868
	7.9145	-0.793	0.34325	-0.03441
	9.3993	-0.926	0.407646	-0.04015
3	2.1519	-0.005	0.093329	-0.00023
	2.9349	-0.126	0.127286	-0.00544
	3.8314	-0.251	0.166169	-0.01089
	4.8582	-0.377	0.210701	-0.01633
	6.0358	-0.502	0.261772	-0.02178
	7.3875	-0.628	0.320394	-0.02722
	8.9412	-0.753	0.387779	-0.03266
	10.729	-0.879	0.465315	-0.03811

Table.A.40. Raw data of the effect of the physical sorption (*K*) on chemical sorption (*k*) at 0.75 filling ratio for bulk density and grain density.

Replicates	Bulk Density		Grain density	
	K	*k*	*K*	*k*
1	1.3369	-0.0094	0.038771	-0.00027
	1.9248	-0.2251	0.055821	-0.00653
	2.6436	-0.4502	0.076664	-0.01306
	3.5225	-0.6754	0.102152	-0.01959
	4.5976	-0.9005	0.133329	-0.02611
	5.9118	-1.1256	0.171442	-0.03264
	7.5195	-1.3507	0.218067	-0.03917
	9.4848	-1.5758	0.275059	-0.0457
2	1.2341	-0.0108	0.035789	-0.00031
	1.7724	-0.2592	0.0514	-0.00752
	2.418	-0.5184	0.070122	-0.01503
	3.1894	-0.7776	0.092493	-0.02255
	4.107	-1.0368	0.119102	-0.03007
	5.1916	-1.296	0.150555	-0.03758
	6.4658	-1.5552	0.187509	-0.0451
	7.95	-1.8144	0.230551	-0.05262
3	1.4484	-0.0086	0.042004	-0.00025
	2.0932	-0.2064	0.060703	-0.00599
	2.8983	-0.4128	0.08405	-0.01197
	3.909	-0.6192	0.11336	-0.01796
	5.1861	-0.8256	0.150396	-0.02394
	6.8118	-1.032	0.197543	-0.02993
	8.9041	-1.2384	0.258218	-0.03591
	11.63	-1.4448	0.337275	-0.0419

Table.A.41. Raw data of the effect of the physical sorption (*K*) on chemical sorption (*k*) at 0.95 filling ratio for bulk density and grain density.

Replicates	Bulk Density		Grain density	
	K	*k*	*K*	*k*
1	0.863609	-0.0144	0.018783	-0.00031
	1.535028	-0.3456	0.033387	-0.00752
	2.482643	-0.6912	0.053997	-0.01503
	3.820178	-1.0368	0.083089	-0.02255
	5.707411	-1.3824	0.124136	-0.03007
	8.371955	-1.728	0.18209	-0.03758
	12.13083	-2.0736	0.263846	-0.0451
	17.43741	-2.4192	0.379264	-0.05262
2	0.797112	-0.0153	0.017337	-0.00033
	1.404782	-0.3672	0.030554	-0.00799
	2.229135	-0.7344	0.048484	-0.01597
	3.330928	-1.1016	0.072448	-0.02396
	4.773976	-1.4688	0.103834	-0.03195
	6.616566	-1.836	0.14391	-0.03993
	8.890832	-2.2032	0.193376	-0.04792
	15.58789	-2.5704	0.292037	-0.05591
3	0.899021	-0.0131	0.019554	-0.00028
	1.606185	-0.3144	0.034935	-0.00684
	2.626182	-0.6288	0.057119	-0.01368
	4.111365	-0.9432	0.089422	-0.02051
	6.302855	-1.2576	0.137087	-0.02735
	9.604751	-1.572	0.208903	-0.03419
	14.73008	-1.8864	0.320379	-0.04103
	20.08289	-2.2008	0.402053	-0.04787

Table.A.42. Raw data of repeated fumigation for five times with 8.928 mg/L on bread wheat 12.5% moisture content.

Replicates	Exposure time h	Number of fumigations				
		1	2	3	4	5
1	0	8.9166	8.7709	8.2242	8.4541	8.343
	24	4.431	4.631	4.712	4.922	4.922
	48	3.931	4.231	4.364	4.622	4.652
	72	3.431	3.831	4.016	4.322	4.382
	96	2.931	3.431	3.668	4.022	4.112
	120	2.431	3.031	3.32	3.722	3.842
	144	1.931	2.631	2.972	3.422	3.572
	168	1.431	2.231	2.624	3.122	3.302
2	0	8.5485	8.5174	8.6299	8.167	8.03
	24	4.5485	4.597	4.6299	4.787	4.977
	48	4.0485	4.197	4.2819	4.487	4.707
	72	3.5485	3.797	3.9339	4.187	4.437
	96	3.0485	3.397	3.5859	3.887	4.167
	120	2.5485	2.997	3.2379	3.587	3.897
	144	2.0485	2.597	2.8899	3.287	3.627
	168	1.5485	2.197	2.5419	2.987	3.357
3	0	8.5485	8.0174	8.629	8.167	8.12
	24	4.5485	4.6485	4.6299	4.899	4.989
	48	4.0485	4.2485	4.2819	4.599	4.719
	72	3.5485	3.8485	3.9339	4.299	4.449
	96	3.0485	3.4485	3.5859	3.999	4.179
	120	2.5485	3.0485	3.2379	3.699	3.909
	144	2.0485	2.6485	2.8899	3.399	3.639
	168	1.5485	2.2485	2.5419	3.099	3.369

Table.A.43. Raw data of repeated fumigation for five times with 8.928 mg/L on soft wheat 12.5% moisture content.

Replicates	Exposure time h	Number of fumigations				
		1	2	3	4	5
1	0	8.9631	8.8506	8.54	8.6053	8.87
	24	5.1631	5.3631	5.24	5.334	5.434
	48	4.8631	5.1231	5.031	5.154	5.264
	72	4.5631	4.8831	4.822	4.974	5.094
	96	4.2631	4.6431	4.613	4.794	4.924
	120	3.9631	4.4031	4.404	4.614	4.754
	144	3.6631	4.1631	4.195	4.434	4.584
	168	3.3631	3.9231	3.986	4.254	4.414
2	0	8.8487	8.5763	8.7107	8.8745	8.83
	24	5.313	5.413	5.02	5.12	5.22
	48	5.013	5.173	4.811	4.94	5.05
	72	4.713	4.933	4.602	4.76	4.88
	96	4.413	4.693	4.393	4.58	4.71
	120	4.113	4.453	4.184	4.4	4.54
	144	3.813	4.213	3.975	4.22	4.37
	168	3.513	3.973	3.766	4.04	4.2
3	0	8.8487	8.5763	8.71	8.8745	8.857
	24	5.213	5.513	5.11	5.213	5.213
	48	4.913	5.273	4.901	5.033	5.043
	72	4.613	5.033	4.692	4.853	4.873
	96	4.313	4.793	4.483	4.673	4.703
	120	4.013	4.553	4.274	4.493	4.533
	144	3.713	4.313	4.065	4.313	4.363
	168	3.413	4.073	3.856	4.133	4.193

Table.A.44. Raw data for the effect of SF concentrations, temperatures and moisture on durum germination percentage.

Commodity	SF mg/L	Temperature°C	Moisture%	Replicates			
				1	2	3	4
durum	0	15	12	90	91	90	89
durum	0	15	15	91	90	90	90
durum	0	25	12	89	89	91	91
durum	0	25	15	90	91	90	89
durum	0	35	12	91	91	90	89
durum	0	35	15	89	90	88	88
durum	4.167	15	12	86	87	85	86
durum	4.167	15	15	42	42	41	41
durum	4.167	25	12	18	20	19	18
durum	4.167	25	15	6	7	6	6
durum	4.167	35	12	24	25	24	25
durum	4.167	35	15	9	9	8	7
durum	8.928	15	12	78	78	79	78
durum	8.928	15	15	10	11	10	11
Durum	8.928	25	12	36	37	36	38
Durum	8.928	25	15	12	12	13	13
Durum	8.928	35	12	8	9	8	7
Durum	8.928	35	15	1	2	2	1
Durum	31.25	15	12	42	43	44	42
Durum	31.25	15	15	8	10	9	10
Durum	31.25	25	12	50	50	51	50
Durum	31.25	25	15	12	13	13	13
Durum	31.25	35	12	2	1	3	1
Durum	31.25	35	15	2	1	2	1

Table.A.45. Raw data for the effect of SF concentrations, temperatures and moisture on durum hectolitre weight kg/hL.

Commodity	SF mg/L	Temperature°C	Moisture%	Replicates		
				1	2	3
Durum	0	15	12	82.18	82.31	81.97
Durum	0	15	15	82.33	82.54	82.25
Durum	0	25	12	82.43	82.29	82.18
Durum	0	25	15	82.30	82.11	82.42
Durum	0	35	12	82.31	82.39	82.11
Durum	0	35	15	82.24	82.40	82.43
Durum	4.167	15	12	82.13	81.99	82.41
Durum	4.167	15	15	79.99	80.05	80.08
Durum	4.167	25	12	81.91	82.46	82.23
Durum	4.167	25	15	79.98	80.18	79.85
Durum	4.167	35	12	82.41	82.32	82.73
Durum	4.167	35	15	79.65	79.94	79.75
Durum	8.928	15	12	82.26	82.33	82.43
Durum	8.928	15	15	80.35	79.79	78.32
Durum	8.928	25	12	82.43	82.48	82.33
Durum	8.928	25	15	80.22	80.07	78.42
Durum	8.928	35	12	82.28	82.47	82.54
Durum	8.928	35	15	80.32	79.84	78.96
Durum	31.25	15	12	82.03	82.01	82.31
Durum	31.25	15	15	80.22	80.30	78.59
Durum	31.25	25	12	82.39	82.07	82.10
Durum	31.25	25	15	79.84	79.92	79.62
Durum	31.25	35	12	82.24	82.28	82.32
Durum	31.25	35	15	80.17	80.03	79.15

Table.A.46. Raw data for the effect of SF concentrations, temperatures and moisture on durum 1000 grain weight g.

Commodity	SF mg/L	Temperature°C	Moisture%	Replicates		
				1	2	3
Durum	0	15	12	42.4	41.24	43.6
Durum	0	15	15	45.68	41.28	44.24
Durum	0	25	12	42.64	42.64	41.56
Durum	0	25	15	43.04	42.72	39.76
Durum	0	35	12	41.4	44.04	43.44
Durum	0	35	15	43.4	42.6	43.28
Durum	4.167	15	12	39.76	41.4	43.44
Durum	4.167	15	15	43.04	41.56	38.44
Durum	4.167	25	12	39.04	39.92	42.72
Durum	4.167	25	15	41.8	42.64	41.32
Durum	4.167	35	12	40.36	44.04	40.12
Durum	4.167	35	15	42.72	44.24	42.64
Durum	8.928	15	12	40.84	40.8	42.6
Durum	8.928	15	15	43.2	42	42.4
Durum	8.928	25	12	40.48	40	42
Durum	8.928	25	15	42.6	43.28	42.84
Durum	8.928	35	12	39.4	40.96	42.6
Durum	8.928	35	15	43.4	43.6	41.28
Durum	31.25	15	12	44.64	39.8	42.08
Durum	31.25	15	15	41.24	45.04	43.92
Durum	31.25	25	12	38.8	39.84	40.8
Durum	31.25	25	15	45.68	43.2	39.88
Durum	31.25	35	12	39.68	41.72	38.4
Durum	31.25	35	15	42.92	41.44	40.72

Table.A.47. Raw data for the effect of SF concentrations, temperatures and moisture on durum protein content.

Commodity	SF mg/L	Temperature°C	Moisture%	Replicates		
				1	2	3
Durum	0	15	12	13.4	12.8	12.9
Durum	0	15	15	13	13.1	13.7
Durum	0	25	12	12.9	13.1	13.2
Durum	0	25	15	13.2	13.2	13.2
Durum	0	35	12	13.2	13.1	13.3
Durum	0	35	15	12.9	13	13
Durum	4.167	15	12	13.2	13.2	13.1
Durum	4.167	15	15	13	12.8	13.1
Durum	4.167	25	12	13	12.9	13.2
Durum	4.167	25	15	13	13.7	13.5
Durum	4.167	35	12	13.2	12.9	13.1
Durum	4.167	35	15	13.4	13.2	13.2
Durum	8.928	15	12	12.9	13.3	13.2
Durum	8.928	15	15	12.9	13.2	13.1
Durum	8.928	25	12	13.1	13.3	13.2
Durum	8.928	25	15	13.3	13.3	13
Durum	8.928	35	12	13.1	13.4	13.4
Durum	8.928	35	15	13.1	13.2	13.2
Durum	31.25	15	12	13.1	13.1	13.5
Durum	31.25	15	15	13.4	13.1	13.4
Durum	31.25	25	12	13.2	13.4	13.2
Durum	31.25	25	15	13.7	13.3	13.2
Durum	31.25	35	12	13.3	13.5	13.4
Durum	31.25	35	15	13.4	13.4	13.5

Table.A.48. Raw data for the effect of commodity, SF concentrations, temperatures and moisture on grain hardness.

Commodity	SF mg/L	Temperature°C	Moisture%	Replicates		
				1	2	3
Durum	0	15	12	95	95	94
Durum	0	15	15	96	94	95
Durum	0	25	12	95	96	96
Durum	0	25	15	94	95	93
Durum	0	35	12	95	94	95
Durum	0	35	15	96	95	96
Durum	4.167	15	12	94	95	94
Durum	4.167	15	15	95	95	94
Durum	4.167	25	12	95	94	94
Durum	4.167	25	15	96	97	96
Durum	4.167	35	12	105	103	104
Durum	4.167	35	15	96	98	98
Durum	8.928	15	12	104	104	103
Durum	8.928	15	15	96	95	97
Durum	8.928	25	12	102	105	105
Durum	8.928	25	15	96	96	96
Durum	8.928	35	12	105	104	103
Durum	8.928	35	15	95	95	97
Durum	31.25	15	12	103	103	101
Durum	31.25	15	15	96	97	97
Durum	31.25	25	12	104	104	100
Durum	31.25	25	15	97	96	98
Durum	31.25	35	12	103	103	103
Durum	31.25	35	15	96	97	96

Table.A.49. Raw data for the effect of, SF concentrations, temperatures and moisture on semolina production during milling from durum grain.

SF mg/L	Temperature°C	Moisture%	Replicates		
			1	2	3
0	15	12	107	105.8	105
0	15	15	104.5	106.5	105.2
0	25	12	103.7	110	108.9
0	25	15	107.2	106.3	106.7
0	35	12	104.7	102.3	110.3
0	35	15	109.1	108.9	106.3
4.167	15	12	108.08	110.56	105.71
4.167	15	15	103.15	103.31	100.55
4.167	25	12	117.52	115.06	108.94
4.167	25	15	100.92	100.38	102.27
4.167	35	12	113.25	112.37	104.13
4.167	35	15	115.06	111.75	109.56
8.928	15	12	106.52	113.77	112.36
8.928	15	15	106.27	108.85	106.03
8.928	25	12	119.49	103.79	115.92
8.928	25	15	106.85	103.42	106.86
8.928	35	12	114.61	105.07	106.23
8.928	35	15	103.59	102.03	102.33
31.25	15	12	109.96	107.17	105.3
31.25	15	15	100.24	99.59	99
31.25	25	12	109.02	116.94	103.31
31.25	25	15	98.33	98.56	110.94
31.25	35	12	120.11	109.81	110.9
31.25	35	15	110.29	105.39	106.29

Table.A.50. Raw data for the effect of, SF concentrations, temperatures and moisture on bran production during milling from durum grain.

SF mg/L	Temperature°C	Moisture%	Replicates		
			1	2	3
0	15	12	81.4	82.2	88.8
0	15	15	81.3	81.4	82
0	25	12	85.6	76.8	83.4
0	25	15	81.2	83.3	79.9
0	35	12	88.6	84.9	78.8
0	35	15	83.1	81.6	86.1
4.167	15	12	82.06	81.5	83
4.167	15	15	78.39	79.85	83.62
4.167	25	12	76.53	75.79	87.28
4.167	25	15	82.8	82.58	82.25
4.167	35	12	79.58	79.83	83.37
4.167	35	15	75.42	74.41	78.47
8.928	15	12	87.51	76.94	79.63
8.928	15	15	81.82	77.51	78.74
8.928	25	12	73.51	90.11	73.51
8.928	25	15	76.67	82.65	81.06
8.928	35	12	79.12	86.49	87.14
8.928	35	15	80.07	80.13	78.93
31.25	15	12	80.68	84.98	87.66
31.25	15	15	87.27	85.97	86.4
31.25	25	12	82.2	79.86	84.4
31.25	25	15	87.88	86.19	75.21
31.25	35	12	72.3	78.55	80.55
31.25	35	15	76.26	82.16	80.12

Table.A.51. Raw data for the effect of, SF concentrations, temperatures and moisture on reel semolina production during milling from durum grain.

SF mg/L	Temperature°C	Moisture%	Replicates		
			1	2	3
0	15	12	26.3	24.9	22.7
0	15	15	24.5	25.3	24.8
0	25	12	22.9	26.9	18.9
0	25	15	25.6	24.3	25.3
0	35	12	21	25.1	24.4
0	35	15	25.1	24.9	25
4.167	15	12	22.67	21.01	25.1
4.167	15	15	24.35	23.52	21.96
4.167	25	12	19	22.8	17.75
4.167	25	15	22.49	22.74	24.15
4.167	35	12	20.19	21.13	24.09
4.167	35	15	18.94	20.64	20.81
8.928	15	12	19.42	20.28	21.23
8.928	15	15	18.64	21.19	21.69
8.928	25	12	21.12	19.32	23.8
8.928	25	15	23.56	22.67	20.79
8.928	35	12	19.94	20.32	18.71
8.928	35	15	23.5	24.34	25.49
31.25	15	12	22.02	20.79	20.04
31.25	15	15	19.42	19.38	20.75
31.25	25	12	22.04	16.28	23.35
31.25	25	15	19.44	21.49	21.11
31.25	35	12	20.55	22.8	21.45
31.25	35	15	21.46	20.11	20.54

Table.A.52. Raw data of the effect of SF, temperature, moisture and commodity on brightness of semolina (L*).

SF mg/L	Moisture %	Temperature°C	Commodity	Replicates		
				1	2	3
0	12	15	milled*	87.33	86.88	86.8
0	12	15	milled	86.42	86.63	87.01
0	12	15	milled	86.35	86.43	86.8
0	12	15	commercial**	81.98	81.81	82.12
0	12	15	commercial	81.53	81.52	81.44
0	12	15	commercial	82	81.07	81.61
0	12	25	milled	87.81	87.13	87.07
0	12	25	milled	86.57	86.68	85.82
0	12	25	milled	87.02	86.85	86.57
0	12	25	commercial	82.37	81.9	82.09
0	12	25	commercial	81.3	81.89	82.01
0	12	25	commercial	81.89	81.95	81.88
0	12	35	milled	87.16	87.1	87.04
0	12	35	milled	86.59	86.49	86.93
0	12	35	milled	85.72	86.64	86.33
0	12	35	commercial	82.05	81.75	81.91
0	12	35	commercial	81.39	81.37	81.5
0	12	35	commercial	81.84	81.28	81.35
4.167	12	15	milled	86.16	86.54	86.68
4.167	12	15	milled	81.62	81.54	81.33
4.167	15	15	milled	86.88	86.44	86.82
4.167	15	15	commercial	81.12	80.84	81.28
4.167	12	25	commercial	86.53	86.62	87.06
4.167	12	25	commercial	81.62	81.73	81.5
4.167	15	25	milled	86.56	86.41	86.77
4.167	15	25	milled	81	81.44	81.29
4.167	12	35	milled	86.29	86.63	87.09
4.167	12	35	commercial	81.39	81.39	81.79
4.167	15	35	commercial	86.53	86.61	86.52
4.167	15	35	commercial	80.48	80.93	80.38
8.928	12	15	milled	86.5	86.61	86.74
8.928	12	15	milled	82.09	81.7	81.71
8.928	15	15	milled	86.11	86.55	85.96
8.928	15	15	commercial	81.3	81.37	81.03
8.928	12	25	commercial	86.41	86.66	86.51
8.928	12	25	commercial	81.64	82.18	81.79
8.928	15	25	milled	86.44	86.88	86.06
8.928	15	25	milled	81.08	81.44	81.14
8.928	12	35	milled	86.96	86.61	86.68
8.928	12	35	commercial	81.92	81.48	82.18

8.928	15	35	commercial	86.52	86.63	86.2
8.928	15	35	commercial	80.74	80.85	80.6
31.25	12	15	milled	86.77	86.46	86.55
31.25	12	15	milled	82.06	81.98	82.05
31.25	15	15	milled	86.72	86.67	86.54
31.25	15	15	commercial	81.77	81.64	81.32
31.25	12	25	commercial	78.58	86.34	86.47
31.25	12	25	commercial	82.2	81.43	81.79
31.25	15	25	milled	86.86	86.68	86.48
31.25	15	25	milled	81.5	81.2	81.1
31.25	12	35	milled	86.51	86.39	86.43
31.25	12	35	commercial	82.46	81.48	81.83
31.25	15	35	commercial	86.39	86.4	86.73
31.25	15	35	commercial	80.91	81.22	81.05

* Milled is semolina milled from fumigated grains.

** Commercial is semolina supplied commercially and fumigated.

Table.A.53. Raw data of the effect of SF, temperature, moisture and commodity on redness of semolina (a*).

SF mg/L	Moisture %	Temperature°C	Commodity	Replicates		
				1	2	3
0	12	15	milled*	-2.65	-2.63	-2.58
0	12	15	milled	-2.55	-2.79	-2.81
0	12	15	milled	-2.73	-2.56	-2.72
0	12	15	commercial**	-2.03	-2.05	-2.46
0	12	15	commercial	-2.21	-2.11	-2.1
0	12	15	commercial	-2.09	-2.17	-2.2
0	12	25	milled	-2.78	-2.69	-2.63
0	12	25	milled	-2.66	-2.68	-2.59
0	12	25	milled	-2.74	-2.56	-2.66
0	12	25	commercial	-2.32	-2.25	-2.29
0	12	25	commercial	-2.24	-2.34	-2.37
0	12	25	commercial	-2.45	-2.48	-2.53
0	12	35	milled	-2.67	-2.72	-2.6
0	12	35	milled	-2.63	-2.65	-2.67
0	12	35	milled	-2.39	-2.7	-2.71
0	12	35	commercial	-2.28	-2.29	-2.22
0	12	35	commercial	-1.99	-2.02	-2.1
0	12	35	commercial	-2.21	-2.23	-2.16
4.167	12	15	milled	-2.44	-2.56	-2.64
4.167	12	15	milled	-2.32	-2.36	-2.42
4.167	15	15	milled	-2.42	-2.56	-2.67
4.167	15	15	commercial	-2.23	-2.3	-2.22
4.167	12	25	commercial	-2.4	-2.56	-2.65
4.167	12	25	commercial	-2.31	-2.3	-2.28
4.167	15	25	milled	-2.61	-2.69	-2.62
4.167	15	25	milled	-2.16	-2.21	-2.21
4.167	12	35	milled	-2.46	-2.57	-2.75
4.167	12	35	commercial	-2.25	-2.2	-2.32
4.167	15	35	commercial	-2.41	-2.5	-2.43
4.167	15	35	commercial	-2.01	-2.07	-1.88
8.928	12	15	milled	-2.61	-2.55	-2.52
8.928	12	15	milled	-2.26	-2.26	-2.21
8.928	15	15	milled	-2.37	-2.52	-2.5
8.928	15	15	commercial	-2.09	-2.06	-2.11
8.928	12	25	commercial	-2.47	-2.59	-2.48
8.928	12	25	commercial	-2.22	-2.32	-2.24
8.928	15	25	milled	-2.45	-2.56	-2.53
8.928	15	25	milled	-2.06	-2.09	-2.14
8.928	12	35	milled	-2.67	-2.62	-2.68
8.928	12	35	commercial	-2.24	-2.16	-2.28

8.928	15	35	commercial	-2.49	-2.57	-2.51
8.928	15	35	commercial	-1.98	-1.98	-2.01
31.25	12	15	milled	-2.67	-2.58	-2.59
31.25	12	15	milled	-2.35	-2.35	-2.32
31.25	15	15	milled	-2.55	-2.47	-2.58
31.25	15	15	commercial	-2.3	-2.28	-2.18
31.25	12	25	commercial	-2.09	-2.47	-2.51
31.25	12	25	commercial	-2.36	-2.29	-2.29
31.25	15	25	milled	-2.63	-2.58	-2.38
31.25	15	25	milled	-2.18	-2.17	-2.19
31.25	12	35	milled	-2.57	-2.51	-2.49
31.25	12	35	commercial	-2.42	-2.2	-2.32
31.25	15	35	commercial	-2.5	-2.46	-2.47
31.25	15	35	commercial	-2.08	-1.96	-2.07

* Milled is semolina milled from fumigated grains.

** Commercial is semolina supplied commercially and fumigated.

Table.A.54. Raw data of the effect of SF, temperature, moisture and commodity on yellowness of semolina (b*).

SF mg/L	Moisture %	Temperature°C	Commodity	Replicates		
				1	2	3
0	12	15	milled*	20.81	21.85	21.68
0	12	15	milled	21.45	21.32	20.93
0	12	15	milled	20.77	21.23	20.84
0	12	15	commercial**	26.69	26.75	29.15
0	12	15	commercial	27.61	27.4	27.21
0	12	15	commercial	27.2	27.36	27.74
0	12	25	milled	20.97	21.64	21.09
0	12	25	milled	21.29	21.58	21.14
0	12	25	milled	21.2	21.17	21.7
0	12	25	commercial	27.89	27.72	27.72
0	12	25	commercial	27.89	28.23	28.48
0	12	25	commercial	29.22	29.45	29.51
0	12	35	milled	21.51	21.96	21.45
0	12	35	milled	21.13	21.33	21.03
0	12	35	milled	21.23	21.2	21.35
0	12	35	commercial	27.67	27.66	27.43
0	12	35	commercial	26.84	26.66	27.14
0	12	35	commercial	27.59	27.61	27.48
4.167	12	15	milled	21.79	21.49	21.95
4.167	12	15	milled	29.75	28.89	30.03
4.167	15	15	milled	21.15	21.71	21.86
4.167	15	15	commercial	30.03	28.92	29.79
4.167	12	25	commercial	20.98	21.45	21.53
4.167	12	25	commercial	29.6	28.64	29.66
4.167	15	25	milled	21.47	21.81	21.54
4.167	15	25	milled	29.72	28.53	29.6
4.167	12	35	milled	21.35	21.73	21.77
4.167	12	35	commercial	29.67	28.23	29.84
4.167	15	35	commercial	20.4	20.85	20.93
4.167	15	35	commercial	28.59	28.5	28.11
8.928	12	15	milled	21.71	21.34	21.37
8.928	12	15	milled	27.98	27.99	28.05
8.928	15	15	milled	21.23	20.94	21.11
8.928	15	15	commercial	28.18	27.87	27.59
8.928	12	25	commercial	21.57	21.62	21.46
8.928	12	25	commercial	28.12	28	27.96
8.928	15	25	milled	21.66	21.08	21.13
8.928	15	25	milled	28.12	27.62	27.08
8.928	12	35	milled	21.48	21.76	21.8
8.928	12	35	commercial	27.85	27.76	28.04

8.928	15	35	commercial	21.04	21.12	20.82
8.928	15	35	commercial	27.51	27.55	26.91
31.25	12	15	milled	21.69	22.01	21.54
31.25	12	15	milled	28.24	28.44	28.34
31.25	15	15	milled	21.21	20.77	20.94
31.25	15	15	commercial	28.24	28.64	27.79
31.25	12	25	commercial	19.97	21.32	21.49
31.25	12	25	commercial	28.5	28.18	28.43
31.25	15	25	milled	21.3	21.38	20.58
31.25	15	25	milled	28.47	27.98	28.04
31.25	12	35	milled	21.46	21.73	21.01
31.25	12	35	commercial	28.4	27.92	28.17
31.25	15	35	commercial	21.31	21.34	20.35
31.25	15	35	commercial	27.93	26.67	27.45

* Milled is semolina milled from fumigated grains.
** Commercial is semolina supplied commercially and fumigated.

Table.A.55. Raw data of the effect of SF, temperature, moisture and commodity on whiteness index of semolina (WI).

SF mg/L	Moisture %	Temperature°C	Commodity	Replicates		
				1	2	3
0	12	15	milled*	-17	-20.92	-20.31
0	12	15	milled	-19.54	-19.03	-17.51
0	12	15	milled	-17.1	-18.74	-17.23
0	12	15	commercial**	-36.7	-36.82	-44.11
0	12	15	commercial	-39.29	-38.68	-38.09
0	12	15	commercial	-38.24	-38.4	-39.71
0	12	25	milled	-17.48	-20.12	-20.01
0	12	25	milled	-18.93	-19.95	-18.55
0	12	25	milled	-18.51	-18.43	-20.42
0	12	25	commercial	-40.45	-39.76	-39.84
0	12	25	commercial	-40.03	-41.27	-42.06
0	12	25	commercial	-44.2	-44.89	-45.03
0	12	35	milled	-19.61	-21.29	-19.43
0	12	35	milled	-18.33	-19.09	-17.91
0	12	35	milled	-18.87	-18.57	-19.18
0	12	35	commercial	-39.66	-39.53	-38.89
0	12	35	commercial	-36.95	-36.42	-37.88
0	12	35	commercial	-39.35	-39.21	-38.86
4.167	12	15	milled	-20.79	-19.67	-20.3
4.167	12	15	milled	-45.59	-43.08	-46.24
4.167	15	15	milled	-18.36	-20.47	-20.94
4.167	15	15	commercial	-46.14	-42.84	-45.55
4.167	12	25	commercial	-17.81	-19.5	-19.71
4.167	12	25	commercial	-45.17	-42.41	-45.29
4.167	15	25	milled	-19.59	-20.81	-19.8
4.167	15	25	milled	-45.19	-41.98	-45.01
4.167	12	35	milled	-19.19	-20.53	-20.58
4.167	12	35	commercial	-45.24	-41.07	-45.94
4.167	15	35	commercial	-15.7	-17.33	-17.51
4.167	15	35	commercial	-41.71	-41.67	-40.31
8.928	12	15	milled	-20.46	-19.12	-19.2
8.928	12	15	milled	-40.63	-40.49	-40.67
8.928	15	15	milled	-18.81	-17.67	-18.42
8.928	15	15	commercial	-40.89	-40	-39.05
8.928	12	25	commercial	-19.97	-20.13	-19.58
8.928	12	25	commercial	-40.86	-40.69	-40.44
8.928	15	25	milled	-20.3	-18.09	-18.45
8.928	15	25	milled	-40.63	-39.3	-37.58
8.928	12	35	milled	-19.56	-20.64	-20.76
8.928	12	35	commercial	-40.16	-39.71	-40.83

8.928	15	35	commercial	-18.05	-18.31	-17.32
8.928	15	35	commercial	-38.71	-38.87	-39.91
31.25	12	15	milled	-20.33	-21.53	-19.84
31.25	12	15	milled	-41.37	-41.93	-41.68
31.25	15	15	milled	-18.61	-17.02	-17.65
31.25	15	15	commercial	-41.24	-42.38	-39.74
31.25	12	25	commercial	-15.87	-19.09	-19.69
31.25	12	25	commercial	-42.22	-40.93	-41.83
31.25	15	25	milled	-18.9	-19.22	-16.37
31.25	15	25	milled	-41.82	-40.27	-40.38
31.25	12	35	milled	-19.58	-20.56	-17.93
31.25	12	35	commercial	-42.02	-40.2	-41.07
31.25	15	35	commercial	-19.06	-19.13	-15.46
31.25	15	35	commercial	-39.99	-36.39	-38.66

* Milled is semolina milled from fumigated grains.

** Commercial is semolina supplied commercially and fumigated.

Table.A.56. Raw data of the effect of SF, temperature, moisture and commodity on yellowness index of semolina (YI).

SF mg/L	Moisture %	Temperature°C	Commodity	Replicates		
				1	2	3
0	12	15	milled*	31.02	32.5	32.3
0	12	15	milled	32.1	31.87	31.25
0	12	15	milled	31.22	31.8	31.19
0	12	15	commercial**	40.22	40.36	43.22
0	12	15	commercial	41.53	41.28	41.07
0	12	15	commercial	40.85	41.38	41.66
0	12	25	milled	31.1	32.16	32.45
0	12	25	milled	31.85	32.19	31.86
0	12	25	milled	31.61	31.61	32.39
0	12	25	commercial	41.58	41.53	41.47
0	12	25	commercial	41.96	42.17	42.43
0	12	25	commercial	43.39	43.64	43.74
0	12	35	milled	31.97	32.58	31.93
0	12	35	milled	31.62	31.92	31.41
0	12	35	milled	32	31.7	31.98
0	12	35	commercial	41.42	41.51	41.17
0	12	35	commercial	40.61	40.39	40.95
0	12	35	commercial	41.4	41.62	41.43
4.167	12	15	milled	32.61	32.12	32.68
4.167	12	15	milled	44.12	43.11	44.57
4.167	15	15	milled	31.58	32.43	32.52
4.167	15	15	commercial	44.66	43.4	44.31
4.167	12	25	commercial	31.45	32.04	32.03
4.167	12	25	commercial	43.95	42.73	44.07
4.167	15	25	milled	32.09	32.52	32.12
4.167	15	25	milled	44.32	42.71	44.07
4.167	12	35	milled	32	32.41	32.33
4.167	12	35	commercial	44.11	42.35	44.17
4.167	15	35	commercial	30.68	31.26	31.34
4.167	15	35	commercial	43.12	42.86	42.57
8.928	12	15	milled	32.41	31.9	31.91
8.928	12	15	milled	41.8	41.95	42.01
8.928	15	15	milled	31.89	31.39	31.78
8.928	15	15	commercial	42.32	41.91	41.68
8.928	12	25	commercial	32.26	32.26	32.09
8.928	12	25	commercial	42.13	41.78	41.88
8.928	15	25	milled	32.37	31.48	31.77
8.928	15	25	milled	42.33	41.57	41
8.928	12	35	milled	31.99	32.46	32.48
8.928	12	35	commercial	41.69	41.73	41.84

8.928	15	35	commercial	31.54	31.61	31.33
8.928	15	35	commercial	41.69	41.7	40.98
31.25	12	15	milled	32.31	32.81	32.18
31.25	12	15	milled	42.12	42.39	42.26
31.25	15	15	milled	31.7	31.14	31.39
31.25	15	15	commercial	42.22	42.76	41.83
31.25	12	25	commercial	32.32	31.95	32.14
31.25	12	25	commercial	42.39	42.27	42.46
31.25	15	25	milled	31.78	31.93	30.94
31.25	15	25	milled	42.61	42.11	42.21
31.25	12	35	milled	32.09	32.48	31.51
31.25	12	35	commercial	42.18	41.94	42.12
31.25	15	35	commercial	31.93	31.95	30.56
31.25	15	35	commercial	42.15	40.45	41.5

* Milled is semolina milled from fumigated grains.
** Commercial is semolina supplied commercially and fumigated.

Table.A.57. Raw data of pasta optimum cooking time.

sample	Optimum cooking time min:sec	Fumigation conditions
27	12	semolina 15°C 12% moist 4.167SFx360 mg/l/h
35	12	semolina 25°C 12% moist 4.167SFx360h mg/l/h
43	12	semolina 35°C 12% moist 4.167SFx360h mg/l/h
99	13	semolina 15°C 12% moist 31.25SFx48h mg/l/h
107	13	semolina 25°C 12% moist 31.25SFx48h mg/l/h
115	12	semolina 35°C 12% moist 31.25SFx48h mg/l/h
11: 15d	11.3	control///semolina 25°C 12% moist 0SFx360h mg/l/h
11:7d	11.3	control/// semolina 25°C 12% moist 0SFx 168h mg/l/h
19:15d	12	control///semolina 35°C 12% moist 0SFx360h mg/l/h
19:7d	12	control/// semolina 35°C 12% moist 0SFx168h mg/l/h
3:15d	11.3	control/// semolina 15°C 12% moist 0SFx360h mg/l/h
3:7d	12	control//semolina 15°C 12% moist 0SFx168h mg/l/h
manildra	12	manilldra 2013

Table.A.58. Raw data of pasta cooking loss and water absorption.

Weight of solids after cooking(g)	Cooking Loss%	Diff bwtn rep1 and rep2	Mean Cooking Loss	Weight of uncooked sample (g)	Weight of cooked sample (g)	H2O Abs%	Diff bwtn rep1 and rep2	Mean H2O Abs%
0.19	3.68	-0.16	3.76	5.17	13.77	166.34	1.66	165.51
0.20	3.84			5.21	13.79	164.68		
				initial w of pasta	final w of pasta			
0.28	5.59	0.22	5.48	5.01	12.62	151.90	2.39	150.70
0.27	5.37			5.03	12.55	149.50		
0.30	5.98	0.00	5.98	5.02	12.93	157.57	-1.00	158.07
0.30	5.98			5.02	12.98	158.57		
0.31	6.09	-0.23	6.21	5.09	13.09	157.17	-0.14	157.24
0.32	6.32			5.06	13.02	157.31		
0.30	5.89	0.03	5.88	5.09	13.17	158.74	-0.63	159.06
0.30	5.86			5.12	13.28	159.38		
0.27	5.39	-0.53	5.65	5.01	12.72	153.89	-3.50	155.64
0.30	5.92			5.07	13.05	157.40		
0.27	5.29	-0.01	5.30	5.1	13.01	155.10	-1.29	155.74
0.27	5.30			5.09	13.05	156.39		
0.30	5.98	-0.08	6.02	5.02	12.93	157.57	1.32	156.91
0.31	6.05			5.12	13.12	156.25		
0.35	6.93	0.18	6.84	5.05	12.88	155.05	-5.27	157.68
0.34	6.75			5.04	13.12	160.32		
0.38	7.57	0.23	7.46	5.02	13.44	167.73	9.79	162.83
0.37	7.34			5.04	13	157.94		
0.36	7.07	-1.87	8.01	5.09	13.11	157.56	8.66	153.24
0.45	8.95			5.03	12.52	148.91		
0.40	7.94	-0.09	7.98	5.04	13.15	160.91	3.97	158.93
0.41	8.02			5.11	13.13	156.95		
0.43	8.57	0.43	8.35	5.02	12.34	145.82	-8.15	149.89
0.41	8.13			5.04	12.8	153.97		
0.46	9.13	1.06	8.60	5.04	12.6	150.00	-1.38	150.69
0.41	8.07			5.08	12.77	151.38		

Table.A.59. Raw data of uncooked pasta colour.

Sample	Fumigation conditions	minolta#	UC-L*	UC-a*	UC-b*	UC-WI	UC-YI
99	semolina 15°C 12% moist 31.25SFx48h mg/l/h	1514	67.47	2.04	46.29	-65.18	68.74
27	semolina 15°C 12% moist 4.167SFx360 mg/l/h	1515	67.8	1.79	45.93	-65.16	68.22
11:7d	control/// semolina 25°C 12% moist 0SFx 168h mg/l/h	1516	66.69	2.42	45.18	-62.41	68.08
19:15d	control///semolina 35°C 12% moist 0SFx360h mg/l/h	1517	65.94	2.77	44.79	-60.77	68.1
107	semolina 25°C 12% moist 31.25SFx48h mg/l/h	1518	66.62	2.07	45.78	-63.16	68.71
3: 15d	control/// semolina 15°C 12% moist 0SFx360h mg/l/h	1519	66.71	2.38	45.53	-62.94	68.42
19:7d	control/// semolina 35°C 12% moist 0SFx168h mg/l/h	1520	66.1	2.77	45.02	-61.33	68.24
115	semolina 35°C 12% moist 31.25SFx48h mg/l/h	1521	67.72	1.84	44.99	-63.66	67.34
manildra	manilldra 2013	1522	67.4	2.24	50.36	-70.65	72.53
35	semolina 25°C 12% moist 4.167SFx360h mg/l/h	1523	67.18	2.03	48.1	-67.25	70.6
43	semolina 35°C 12% moist 4.167SFx360h mg/l/h	1524	66.38	1.12	46.81	-64.21	69.82
11: 15d	control///semolina 25°C 12% moist 0SFx360h mg/l/h	1525	66.45	2.28	46.15	-63.41	69.15
3:7d	control//semolina 15°C 12% moist 0SFx168h mg/l/h	1526	66.6	2.47	44.27	-60.97	67.22

Table.A.60. Raw data of cooked pasta colour.

Sample	Fumigation conditions	minolta	CP-L*	CP-a*	CP-b*	CP-WI	CP-YI
3:7d	control//semolina 15°C 12% moist 0SFx168h mg/l/h	1572	75.47	-2.12	30.46	-43.93	47.4
27	semolina 15°C 12% moist 4.167SFx360 mg/l/h	1573	75.89	-2.67	32.21	-48.44	49.36
11: 15d	control///semolina 25°C 12% moist 0SFx360h mg/l/h	1574	75.81	-2.28	32	-47.88	49.14
99	semolina 15°C 12% moist 31.25SFx48h mg/l/h	1575	75.84	-2.41	32.8	-49.81	50.09
manildra	manilldra 2013	1576	76.42	-2.69	34.07	-53.29	51.34
19:7d	control/// semolina 35°C 12% moist 0SFx168h mg/l/h	1577	75.45	-1.78	30.6	-44.24	47.58
115	semolina 35°C 12% moist 31.25SFx48h mg/l/h	1578	75.79	-2.61	32.71	-49.55	50
35	semolina 25°C 12% moist 4.167SFx360h mg/l/h	1579	76.39	-2.94	32.34	-49.11	49.3
43	semolina 35°C 12% moist 4.167SFx360h mg/l/h	1580	75.71	-2.75	32.04	-47.89	49.22
11:7d	control/// semolina 25°C 12% moist 0SFx 168h mg/l/h	1581	75.57	-2.11	30.96	-45.21	47.98
19:15d	control///semolina 35°C 12% moist 0SFx360h mg/l/h	1582	75.21	-1.87	31.09	-45.28	48.29
3: 15d	control/// semolina 15°C 12% moist 0SFx360h mg/l/h	1583	75.93	-2.05	31.33	-46.35	48.28
107	semolina 25°C 12% moist 31.25SFx48h mg/l/h	1584	75.72	-2.46	32.1	-48.05	49.3

Table.A.61.Raw data of pasta firmness.

Test ID	Sample	Fumigation conditions	Force 1 g	Area-FT 1:2 g.s
6051401	19:15D	control///semolina 35°C 12% moist 0SFx360h mg/l/h	573.4	186.15
6051402			540.82	170.63
6051403			561.7	182.59
6051404			569.05	186.08
6051405	11:15D	control///semolina 25°C 12% moist 0SFx360h mg/l/h	590.42	188.11
6051406			561.74	177.72
6051407			582.47	186.14
6051408			574.78	182.84
6051409	43	semolina 35°C 12% moist 4.167SFx360h mg/l/h	598.91	190.89
6051410			596.71	190.6
6051411			610.01	198.4
6051412			634.33	210.92
6051413	35	semolina 25°C 12% moist 4.167SFx360h mg/l/h	576.89	187.51
6051414			567.28	184.4
6051415			570.63	187.22
6051416			555.01	181.09
6051417	115	semolina 35°C 12% moist 31.25SFx48h mg/l/h	563.42	183.58
6051418			550.19	178.37
6051419			531.94	172.76
6051420			570.04	192.16
6051421	11:7D	control/// semolina 25°C 12% moist 0SFx 168h mg/l/h	580.14	192.74
6051422			570.6	193.16
6051423			578.01	194.98
6051424			548.23	187.04
6051425	3:15D	control/// semolina 15°C 12% moist 0SFx360h mg/l/h	588.03	194.52
6051426			556.82	184.46
6051427			564.56	188.56

6051428			581.93	197.86
6051429	107	semolina 25°C 12% moist 31.25SFx48h mg/l/h	581.92	187.15
6051430			596.11	192.5
6051431			582.73	191.15
6051432			600.14	198.71
6051433	27	semolina 15°C 12% moist 4.167SFx360 mg/l/h	539.1	170.42
6051434			533.47	169.94
6051435			576.25	192.22
6051436			544.41	178.87
6051437	manildra2013	manildra2013	598.66	195.51
6051438			603.1	197.63
6051439			606.45	199.57
6051440			634.52	217.63
6051441	99	semolina 15°C 12% moist 31.25SFx48h mg/l/h	557.29	183.43
6051442			541.76	180.35
6051443			540.31	179.53
6051444			499.89	164.16
6051445	19:7D	control;/// semolina 35°C 12% moist 0SFx168h mg/l/h	539.11	183.62
6051446			524.49	179.84
6051447			534.41	187.09
6051448			525.08	182.81
6051449	3:7D	control//semolina 15°C 12% moist 0SFx168h mg/l/h	510.3	171.17
6051450			530.75	183.67
6051451			543.52	187.49
6051452			515.68	181.04

Table.A.62.Raw data of pasta stickiness.

Sample	OCT	Force 1	area1:2	area3:4	Fumigation conditions
19:7D	12	19.3	12.53	5.912	control/// semolina 35°C 12% moist 0SFx168h mg/l/h
19:7D		23.4	12.98	6.061	
19:7D		20.1	9.417	5.332	
11:7D	11:30	23.8	10.44	5.591	control/// semolina 25°C 12% moist 0SFx 168h mg/l/h
11:7D		21.8	11.16	5.712	
11:7D		21.1	10.19	5.578	
3:15D	11:30	21.8	10.92	6.406	control/// semolina 15°C 12% moist 0SFx360h mg/l/h
3:15D		19.5	12.01	5.519	
3:15D		19.9	11.41	5.533	
107	11:30	21	11.79	6.251	semolina 25°C 12% moist 31.25SFx48h mg/l/h
107		22.2	9.69	6.097	
107		22.8	9.946	5.877	
115	12	24.7	10.59	6.234	semolina 35°C 12% moist 31.25SFx48h mg/l/h
115		21.9	10.79	5.88	
115		21.1	11.35	6.191	
19:15D	12	20.6	10.41	6.254	control///semolina 35°C 12% moist 0SFx360h mg/l/h
19:15D		21.4	10.23	5.79	
19:15D		21.3	9.878	5.622	
3:7D	12	20.1	11.89	5.631	control//semolina 15°C 12% moist 0SFx168h mg/l/h
3:7D		23.1	11.2	5.73	
3:7D		20.5	10.48	5.924	
35	12	22.7	12.41	7.144	semolina 25°C 12% moist 4.167SFx360h mg/l/h
35		25.8	14.17	7.568	
35		26	13.2	7.88	
11:15D	11:30	21.2	11.52	5.696	control///semolina 25°C 12% moist 0SFx360h mg/l/h
11:15D		25.8	10.25	5.823	
11:15D		22.2	11.91	6.082	
mani2013	12	21.3	10.99	6.117	manilldra 2013
mani2013		19.9	12.73	6.482	

mani2013		22.2	11.86	6.35	
99	12	25.2	11.62	6.295	semolina 15°C 12% moist 31.25SFx48h mg/l/h
99		24.6	13.83	6.289	
99		24.4	15.83	6.417	
27	12	21.9	12.81	6.003	semolina 15°C 12% moist 4.167SFx360 mg/l/h
27		21.3	13.53	6.392	
27		23.3	13.84	6.145	
43	12	22.4	12.98	6.37	semolina 35°C 12% moist 4.167SFx360h mg/l/h
43		24.3	12.98	6.582	
43		23.8	12.95	6.226	

Table.A.63.Raw data of mixograph results as maximum peak time (MPT), maximum peak hight (MPH), width of mixogram at peak time (WAP), width of mixogram 8 minutes past peak mixing time (WA8) and resistance breakdown (RBD) of milled from durum grain and commercially supplied semolina fumigated with SF 0, 4.167, 8.928 and 31.25 mg/L at 12 and 15% moisture content and 15, 25 and 35°C .

SF mg/L	Tmperature°C	Moisture%	Commodity	Sample	Replicates	Mixo rep	Day	MPT	MPH	WAP	WA8	RBD
0	15	12	milled	2	1	A	2	4.2	65.1	19.7	10.5	47.0
0	15	12	milled	2	1	B	2	4.5	64.7	13.8	10.3	25.0
0	15	12	milled	2	2	A	2	3.8	64.2	18.1	10.0	44.9
0	15	12	milled	2	2	B	2	3.9	65.5	16.3	10.5	35.1
0	15	12	milled	2	3	A	2	4.8	69.2	13.1	11.2	14.4
0	15	12	milled	2	3	B	2	3.7	67.8	16.3	10.9	33.1
0	25	12	milled	10	1	A	2	3.4	69.9	37.0	10.2	72.4
0	25	12	milled	10	1	B	2	3.2	67.0	21.5	9.9	54.1
0	25	12	milled	10	2	A	2	4.1	63.2	11.8	9.1	22.6
0	25	12	milled	10	2	B	2	3.7	63.6	13.8	9.8	29.4
0	25	12	milled	10	3	A	2	2.9	67.4	25.0	10.0	59.9
0	25	12	milled	10	3	B	2	4.2	64.3	13.3	9.0	31.9
0	35	12	milled	18	1	A	2	3.9	64.1	17.9	13.0	27.3
0	35	12	milled	18	1	B	2	4.0	62.3	15.6	8.7	43.9
0	35	12	milled	18	2	A	2	4.9	66.9	16.5	13.1	20.9
0	35	12	milled	18	2	B	2	4.1	66.5	14.0	10.2	26.9
0	35	12	milled	18	3	A	2	3.8	80.7	20.6	12.7	38.2
0	35	12	milled	18	3	B	2	3.5	66.0	14.4	8.5	41.2
0	15	12	milled	2	1	A	7	3.7	64.3	13.2	8.9	32.5
0	15	12	milled	2	1	B	7	3.6	63.2	16.6	9.3	44.0
0	15	12	milled	2	2	A	7	3.9	65.2	16.0	9.0	43.3
0	15	12	milled	2	2	B	7	3.9	64.4	16.9	10.5	37.6
0	15	12	milled	2	3	A	7	4.3	63.0	19.6	9.4	52.2
0	15	12	milled	2	3	B	7	3.0	65.5	25.0	9.8	61.0
0	25	12	milled	10	1	A	7	3.6	66.8	21.6	10.1	52.9

0	25	12	milled	10	1	B	7	3.5	67.4	23.4	13.1	44.1
0	25	12	milled	10	2	A	7	3.7	64.9	20.4	11.2	45.3
0	25	12	milled	10	2	B	7	3.2	65.4	20.7	10.0	51.8
0	25	12	milled	10	3	A	7	3.2	70.8	35.3	13.9	60.6
0	25	12	milled	10	3	B	7	3.5	67.8	20.0	10.8	46.2
0	35	12	milled	18	1	A	7	4.0	65.2	15.1	11.3	25.0
0	35	12	milled	18	1	B	7	2.7	65.8	22.4	9.2	59.0
0	35	12	milled	18	2	A	7	3.2	66.2	21.4	9.2	57.3
0	35	12	milled	18	2	B	7	3.2	65.2	23.7	9.1	61.8
0	35	12	milled	18	3	A	7	4.6	64.2	12.8	9.0	29.4
0	35	12	milled	18	3	B	7	3.8	64.9	14.2	10.7	25.0
0	15	12	milled	2	1	A	15	3.8	64.4	15.7	9.0	42.3
0	15	12	milled	2	1	B	15	3.8	64.9	17.2	9.7	43.7
0	15	12	milled	2	2	A	15	4.0	63.5	12.2	12.6	-2.8
0	15	12	milled	2	2	B	15	3.2	63.8	25.1	12.1	52.0
0	15	12	milled	2	3	A	15	3.8	63.5	15.2	10.3	31.8
0	15	12	milled	2	3	B	15	3.5	64.1	12.9	8.4	34.6
0	25	12	milled	10	1	A	15	3.8	63.6	12.0	8.0	33.6
0	25	12	milled	10	1	B	15	3.5	62.5	12.2	8.4	31.2
0	25	12	milled	10	2	A	15	3.7	66.7	15.2	8.7	43.2
0	25	12	milled	10	2	B	15	3.4	64.3	13.3	10.6	20.3
0	25	12	milled	10	3	A	15	3.6	61.8	16.0	7.6	52.2
0	25	12	milled	10	3	B	15	3.4	62.9	17.4	9.1	48.0
0	35	12	milled	18	1	A	15	3.6	65.2	18.7	10.0	46.4
0	35	12	milled	18	1	B	15	3.1	64.4	37.0	10.9	70.6
0	35	12	milled	18	2	A	15	3.9	61.7	17.5	9.6	45.2
0	35	12	milled	18	2	B	15	3.4	63.3	14.8	9.3	37.2
0	35	12	milled	18	3	A	15	3.1	65.4	33.9	10.0	70.5
0	35	12	milled	18	3	B	15	4.0	61.7	12.1	10.5	13.0
0	15	12	commercial	3	1	B	2	4.5	54.7	13.2	10.8	18.3
0	15	12	commercial	3	2	A	2	2.7	57.0	33.6	9.9	70.5
0	15	12	commercial	3	2	B	2	4.0	57.6	20.9	9.4	54.9

0	15	12	commercial	3	3	A	2	5.0	59.2	15.2	10.7	29.5
0	15	12	commercial	3	3	B	2	5.0	58.3	12.0	10.5	13.1
0	25	12	commercial	11	2	A	2	5.0	58.2	18.3	9.5	48.2
0	25	12	commercial	11	2	B	2	5.0	60.3	16.7	11.1	33.4
0	25	12	commercial	11	2	A	2	5.9	54.3	10.5	9.4	10.3
0	25	12	commercial	11	2	B	2	6.4	53.8	11.8	9.7	17.6
0	25	12	commercial	11	3	A	2	5.7	58.8	14.3	10.0	30.0
0	25	12	commercial	11	3	B	2	5.9	58.6	13.3	9.3	30.3
0	35	12	commercial	19	1	A	2	5.5	61.3	13.1	9.8	25.2
0	35	12	commercial	19	1	B	2	5.9	59.4	11.0	8.7	21.3
0	35	12	commercial	19	2	A	2	5.2	54.4	11.7	8.9	24.1
0	35	12	commercial	19	2	B	2	6.1	54.3	10.4	8.4	18.6
0	35	12	commercial	19	3	A	2	4.7	59.6	19.5	9.2	52.9
0	35	12	commercial	19	3	B	2	5.6	58.3	13.5	9.6	28.6
0	15	12	commercial	3	1	A	7	4.8	72.0	20.1	17.0	15.6
0	15	12	commercial	3	1	B	7	5.6	57.3	9.4	7.5	20.2
0	15	12	commercial	3	2	A	7	5.0	54.9	10.7	8.9	17.3
0	15	12	commercial	3	2	B	7	4.3	58.1	22.4	7.3	67.3
0	15	12	commercial	3	3	A	7	5.3	53.3	12.5	8.8	29.2
0	15	12	commercial	3	3	B	7	4.3	55.7	17.0	8.2	51.9
0	25	12	commercial	11	1	A	7	4.2	62.7	15.6	10.1	35.3
0	25	12	commercial	11	1	B	7	4.1	58.9	20.7	8.6	58.3
0	25	12	commercial	11	2	A	7	4.4	60.7	16.6	9.7	41.3
0	25	12	commercial	11	2	B	7	4.2	60.1	12.8	7.4	42.4
0	25	12	commercial	11	3	A	7	4.8	56.7	10.8	7.7	28.0
0	25	12	commercial	11	3	B	7	4.6	56.1	13.4	9.4	30.2
0	35	12	commercial	19	1	A	7	5.7	54.2	12.2	10.0	18.1
0	35	12	commercial	19	1	B	7	5.5	55.2	11.0	8.8	20.0
0	35	12	commercial	19	2	A	7	5.6	56.1	14.8	10.2	31.3
0	35	12	commercial	19	2	B	7	6.1	54.2	11.3	10.7	5.2
0	35	12	commercial	19	3	A	7	5.1	53.4	13.4	10.0	25.3
0	35	12	commercial	19	3	B	7	6.4	52.9	12.1	9.9	18.6

0	15	12	commercial	3	1	A	15	3.5	58.4	35.9	9.0	75.0
0	15	12	commercial	3	1	B	15	5.2	56.1	10.8	8.3	22.9
0	15	12	commercial	3	2	A	15	4.5	54.6	14.6	9.0	38.1
0	15	12	commercial	3	2	B	15	4.1	55.6	14.6	7.2	50.7
0	15	12	commercial	3	3	A	15	4.4	54.3	14.5	7.7	47.1
0	15	12	commercial	3	3	B	15	5.0	55.5	10.3	6.4	37.8
0	25	12	commercial	11	1	A	15	4.8	56.6	11.5	7.2	37.1
0	25	12	commercial	11	1	B	15	4.8	55.5	9.9	7.5	23.8
0	25	12	commercial	11	2	A	15	3.8	56.3	21.1	9.7	54.1
0	25	12	commercial	11	2	B	15	4.0	60.7	16.8	10.6	36.8
0	25	12	commercial	11	3	A	15	5.0	50.9	9.8	7.8	20.3
0	25	12	commercial	11	3	B	15	3.3	63.8	18.9	11.5	38.9
0	35	12	commercial	19	1	A	15	3.8	55.9	18.9	6.7	64.3
0	35	12	commercial	19	1	B	15	6.5	54.9	8.7	8.0	8.1
0	35	12	commercial	19	2	A	15	3.9	53.5	10.5	8.4	19.7
0	35	12	commercial	19	2	B	15	4.8	54.4	12.2	7.9	35.1
0	35	12	commercial	19	3	A	15	6.3	54.7	10.9	9.9	9.5
0	35	12	commercial	19	3	B	15	5.8	57.2	14.9	12.1	18.7
0	15	12	commercial	3	1	A	2	5.4	54.7	14.2	8.5	40.0
4.167	15	12	milled	26	1	A		5.0	63.0	16.6	11.6	29.7
4.167	15	12	milled	26	1	B		4.0	65.7	21.8	12.8	41.2
4.167	15	12	milled	26	2	A		4.3	63.3	12.3	8.9	27.2
4.167	15	12	milled	26	2	B		4.5	62.2	16.3	10.0	38.8
4.167	15	12	milled	26	3	A		4.6	66.8	17.1	9.8	42.4
4.167	15	12	milled	26	3	B		4.3	65.7	17.7	12.0	32.3
4.167	25	12	milled	34	1	A		4.5	62.2	16.6	11.2	32.5
4.167	25	12	milled	34	1	B		4.9	62.4	13.3	9.6	28.4
4.167	25	12	milled	34	2	A		3.4	65.4	17.2	9.8	43.2
4.167	25	12	milled	34	2	B		3.9	61.4	24.1	12.0	50.2
4.167	25	12	milled	34	3	A		4.6	70.0	15.3	11.5	24.6
4.167	25	12	milled	34	3	B		4.7	69.9	22.1	10.4	52.9
4.167	35	12	milled	42	1	A		4.6	61.6	13.1	10.2	22.4

4.167	35	12	milled	42	1	B		4.5	64.0	14.1	11.0	21.8
4.167	35	12	milled	42	2	A		4.9	62.5	15.6	10.2	34.1
4.167	35	12	milled	42	2	B		4.6	63.8	17.9	11.7	34.7
4.167	35	12	milled	42	3	A		3.2	60.0	20.2	7.1	64.6
4.167	35	12	milled	42	3	B		4.2	62.4	13.7	8.9	35.1
4.167	15	12	commercial	27	1	A		4.8	66.8	15.4	11.7	24.2
4.167	15	12	commercial	27	1	B		4.3	63.8	15.7	10.8	31.1
4.167	15	12	commercial	27	2	A		4.3	56.6	14.1	7.7	45.4
4.167	15	12	commercial	27	2	B		3.9	58.3	10.3	7.0	32.2
4.167	15	12	commercial	27	3	A		5.3	61.1	11.4	10.6	7.2
4.167	15	12	commercial	27	3	B		3.5	63.1	30.8	9.0	70.9
4.167	25	12	commercial	35	1	A		4.6	62.2	17.3	11.8	32.0
4.167	25	12	commercial	35	1	B		4.7	62.3	16.0	10.6	34.1
4.167	25	12	commercial	35	2	A		3.9	56.7	11.5	7.9	31.9
4.167	25	12	commercial	35	2	B		3.9	57.0	10.4	4.7	54.7
4.167	25	12	commercial	35	3	A		4.8	59.7	11.0	7.6	30.3
4.167	25	12	commercial	35	3	B		4.9	59.1	13.5	7.3	46.3
4.167	35	12	commercial	43	1	A		5.5	64.0	10.6	7.2	31.9
4.167	35	12	commercial	43	1	B		4.0	63.6	11.5	7.9	31.9
4.167	35	12	commercial	43	2	A		4.4	57.8	13.8	8.0	42.0
4.167	35	12	commercial	43	2	B		4.6	60.6	11.3	7.5	33.9
4.167	35	12	commercial	43	3	A		3.4	65.9	46.6	11.5	75.4
4.167	35	12	commercial	43	3	B		4.3	63.3	16.8	11.5	31.7
8.928	25	12	milled	58	1	A		3.4	59.9	14.5	9.6	33.6
8.928	25	12	milled	58	1	B		4.5	61.6	14.5	8.9	38.2
8.928	25	12	milled	58	2	A		4.5	66.3	15.1	10.1	33.2
8.928	25	12	milled	58	2	B		4.7	62.7	15.2	11.2	25.9
8.928	25	12	milled	58	3	A		4.0	62.4	13.0	8.7	33.0
8.928	25	12	milled	58	3	B		4.8	61.4	12.4	9.1	26.8
8.928	35	12	milled	66	1	A		5.0	60.3	13.2	10.7	19.2
8.928	35	12	milled	66	1	B		4.6	62.0	13.7	10.0	27.1
8.928	35	12	milled	66	2	A		4.3	63.4	16.5	10.6	35.7

8.928	35	12	milled	66	2	B		4.6	64.3	17.5	10.5	40.0
8.928	35	12	milled	66	3	A		4.5	66.0	17.5	12.0	31.3
8.928	35	12	milled	66	3	B		4.6	65.9	16.9	11.2	33.9
8.928	15	12	milled	50	1	A		3.5	61.9	15.8	12.7	19.6
8.928	15	12	milled	50	1	B		3.8	64.5	15.2	10.7	29.8
8.928	15	12	milled	50	2	A		3.6	65.1	31.8	13.0	59.1
8.928	15	12	milled	50	2	B		4.8	62.5	14.1	9.6	31.8
8.928	15	12	milled	50	3	A		4.4	64.4	16.4	11.4	30.4
8.928	15	12	milled	50	3	B		3.3	64.1	28.3	10.1	64.3
8.928	15	12	commercial	51	1	A		5.3	59.5	12.2	6.1	49.9
8.928	15	12	commercial	51	1	B		3.8	60.7	17.1	6.8	60.2
8.928	15	12	commercial	51	2	A		5.1	59.1	16.8	9.5	43.5
8.928	15	12	commercial	51	2	B		3.9	59.4	17.7	10.7	39.9
8.928	15	12	commercial	51	3	A		4.3	57.5	11.3	6.8	39.8
8.928	15	12	commercial	51	3	B		4.4	58.6	24.9	10.3	58.6
8.928	25	12	commercial	59	1	A		8.9	36.8	5.6	10.1	-81.4
8.928	25	12	commercial	59	1	B		4.8	63.3	18.6	9.1	50.8
8.928	25	12	commercial	59	2	A		4.6	65.4	33.9	11.6	65.6
8.928	25	12	commercial	59	2	B		5.1	60.4	16.1	9.9	38.4
8.928	25	12	commercial	59	3	A		3.7	65.0	18.5	9.0	51.3
8.928	25	12	commercial	59	3	B		5.1	59.4	15.5	9.9	36.1
8.928	35	12	commercial	67	1	A		4.7	62.3	19.5	11.3	42.1
8.928	35	12	commercial	67	1	B		5.6	59.9	13.0	9.6	26.3
8.928	35	12	commercial	67	2	A		4.6	58.7	13.9	8.2	41.2
8.928	35	12	commercial	67	2	B		5.1	59.1	11.3	8.3	26.4
8.928	35	12	commercial	67	3	A		4.6	58.9	15.1	11.5	23.5
8.928	35	12	commercial	67	3	B		5.3	60.3	20.7	8.8	57.6
31.25	15	12	milled	98	1	A		4.3	63.3	12.4	8.8	28.7
31.25	15	12	milled	98	1	B		4.1	62.4	11.3	9.3	17.0
31.25	15	12	milled	98	2	A		4.4	68.1	13.4	11.1	17.7
31.25	15	12	milled	98	2	B		4.5	65.5	15.3	12.5	18.0
31.25	15	12	milled	98	3	A		3.9	60.1	14.5	10.1	30.2

31.25	15	12	milled	98	3	B		5.0	60.0	12.8	9.7	24.1
31.25	25	12	milled	106	2	A		2.7	57.4	30.2	9.7	67.9
31.25	25	12	milled	106	2	B		4.6	62.3	13.6	10.0	26.8
31.25	25	12	milled	106	3	A		4.5	63.4	14.2	11.6	18.7
31.25	25	12	milled	106	3	B		3.0	62.0	35.9	10.6	70.5
31.25	25	12	milled	106	6	A		3.8	65.7	32.0	9.8	69.3
31.25	25	12	milled	106	6	B		3.7	62.8	20.9	11.6	44.5
31.25	35	12	milled	114	1	A		4.2	61.7	11.9	8.6	27.3
31.25	35	12	milled	114	1	B						
31.25	35	12	milled	114	2	A		3.2	62.4	28.6	8.1	71.5
31.25	35	12	milled	114	2	B		4.1	60.5	12.0	9.5	20.7
31.25	35	12	milled	114	3	A		3.9	75.0	13.6	11.7	13.9
31.25	35	12	milled	114	3	B		4.4	61.6	12.2	10.2	16.5
31.25	15	12	commercial	99	1	A		3.6	65.0	19.7	6.4	67.7
31.25	15	12	commercial	99	2	B		3.9	59.1	15.2	9.5	37.8
31.25	15	12	commercial	99	2	A		4.3	55.2	12.1	7.1	41.0
31.25	15	12	commercial	99	2	B		4.8	54.4	13.0	7.9	39.3
31.25	15	12	commercial	99	3	A		4.6	58.0	15.9	9.5	40.3
31.25	15	12	commercial	99	3	B		2.9	54.4	14.5	9.0	37.7
31.25	25	12	commercial	107	1	A		4.2	57.7	14.8	8.6	41.8
31.25	25	12	commercial	107	1	B		4.8	58.6	15.2	7.5	50.3
31.25	25	12	commercial	107	2	A		4.0	61.4	19.0	9.4	50.4
31.25	25	12	commercial	107	2	B		5.0	65.7	14.9	10.9	27.4
31.25	25	12	commercial	107	3	A		4.7	59.1	11.4	8.2	28.5
31.25	25	12	commercial	107	3	B		4.5	57.9	17.1	6.3	62.9
31.25	35	12	commercial	115	1	A		3.9	55.9	12.9	8.4	34.5
31.25	35	12	commercial	115	1	B		4.3	57.5	11.1	5.8	47.9
31.25	35	12	commercial	115	2	A		5.2	60.7	18.6	9.3	50.1
31.25	35	12	commercial	115	2	B		4.4	62.5	39.9	8.2	79.4
31.25	35	12	commercial	115	3	A		3.5	62.8	29.4	11.0	62.6
31.25	35	12	commercial	115	3	B		5.2	60.1	19.4	9.3	51.8
4.167	15	15	milled	30	1	A		4.8	63.4	19.6	11.1	43.7

4.167	15	15	milled	30	1	B		4.0	63.5	32.9	9.8	70.1
4.167	15	15	milled	30	2	A		4.6	67.8	16.9	13.1	22.5
4.167	15	15	milled	30	2	B		4.8	68.1	17.8	11.3	36.5
4.167	15	15	milled	30	3	A		5.7	66.1	17.3	12.8	26.2
4.167	15	15	milled	30	3	B		4.7	64.4	14.6	12.1	17.2
4.167	25	15	milled	38	1	A		5.3	59.4	14.9	11.3	24.1
4.167	25	15	milled	38	1	B		4.8	59.3	11.6	8.3	28.6
4.167	25	15	milled	38	2	A		4.7	60.9	16.5	8.4	49.4
4.167	25	15	milled	38	2	B		4.3	62.3	16.2	10.0	38.3
4.167	25	15	milled	38	3	A		4.7	67.2	13.5	16.7	-24.0
4.167	25	15	milled	38	3	B		5.1	65.0	14.6	11.9	18.3
4.167	35	15	milled	46	1	A		6.3	55.8	12.5	10.8	14.2
4.167	35	15	milled	46	1	B		5.8	60.2	14.7	10.2	30.8
4.167	35	15	milled	46	2	A		6.0	61.8	12.1	10.0	18.0
4.167	35	15	milled	46	2	B		7.1	61.0	12.3	11.1	9.4
4.167	35	15	milled	46	3	A		7.2	59.4	11.4	9.4	17.5
4.167	35	15	milled	46	3	B		7.7	58.8	12.3	10.1	17.9
4.167	15	15	commercial	31	1	A		5.4	57.6	13.3	9.1	31.1
4.167	15	15	commercial	31	1	B		6.0	57.5	13.0	10.0	22.8
4.167	15	15	commercial	31	1	A		4.2	58.6	13.9	10.0	28.2
4.167	15	15	commercial	31	1	B		4.9	60.9	13.6	10.7	21.4
4.167	15	15	commercial	31	2	A		5.3	56.8	14.6	11.5	21.4
4.167	15	15	commercial	31	2	B		4.5	72.2	34.3	12.0	65.2
4.167	25	15	commercial	39	1	A		5.1	54.9	10.4	6.2	40.7
4.167	25	15	commercial	39	1	B		5.5	55.1	10.5	7.0	33.5
4.167	25	15	commercial	39	2	A		6.2	54.7	11.9	7.9	33.6
4.167	25	15	commercial	39	2	B		6.0	54.2	11.3	10.3	9.0
4.167	25	15	commercial	39	3	A		5.7	58.1	15.0	8.1	46.1
4.167	25	15	commercial	39	3	B		6.4	57.9	16.5	9.8	40.4
4.167	35	15	commercial	47	1	A		7.5	59.7	13.9	13.9	0.0
4.167	35	15	commercial	47	1	B		7.6	59.2	17.5	12.4	29.4
4.167	35	15	commercial	47	2	A		7.0	52.8	13.4	11.7	12.5

4.167	35	15	commercial	47	2	B						
4.167	35	15	commercial	47	3	A		8.1	54.3	11.5	10.1	12.2
4.167	35	15	commercial	47	3	B		6.0	56.9	13.7	10.6	22.5
8.928	25	15	milled	62	1	A		4.8	62.9	15.7	9.7	38.3
8.928	25	15	milled	62	1	B		4.9	61.9	11.2	8.7	21.9
8.928	25	15	milled	62	2	A		4.8	59.6	10.2	8.2	20.0
8.928	25	15	milled	62	2	B		5.0	59.0	9.7	8.0	16.9
8.928	25	15	milled	62	3	A		5.9	65.9	17.2	12.3	28.5
8.928	25	15	milled	62	3	B		5.1	64.9	22.1	11.5	48.1
8.928	35	15	milled	70	1	A		6.1	66.3	18.2	12.2	33.3
8.928	35	15	milled	70	1	B		5.5	65.4	17.8	11.6	34.9
8.928	35	15	milled	70	2	A		6.5	61.0	13.5	13.0	3.5
8.928	35	15	milled	70	2	B		6.1	61.2	13.4	14.5	-7.9
8.928	35	15	milled	70	3	A		6.9	60.3	15.6	15.1	3.3
8.928	35	15	milled	70	3	B		6.0	62.5	22.0	12.4	43.9
8.928	15	15	milled	54	1	A		4.4	62.7	15.6	11.0	29.4
8.928	15	15	milled	54	1	B		4.7	61.9	17.5	13.6	22.3
8.928	15	15	milled	54	2	A		4.9	67.3	20.6	12.0	41.7
8.928	15	15	milled	54	2	B		5.7	65.4	22.4	12.6	43.6
8.928	15	15	milled	54	3	A		4.8	59.4	15.1	10.0	34.2
8.928	15	15	milled	54	3	B		5.6	59.7	16.9	9.1	46.4
8.928	15	15	commercial	55	1	A		5.5	61.7	15.5	10.9	29.9
8.928	15	15	commercial	55	1	B		4.9	57.8	18.6	11.3	39.2
8.928	15	15	commercial	55	2	A		6.6	55.0	9.4	8.5	10.2
8.928	15	15	commercial	55	2	B		6.5	56.2	10.7	7.6	28.7
8.928	15	15	commercial	55	3	A		7.7	55.9	10.7	9.5	10.6
8.928	15	15	commercial	55	3	B		5.3	55.1	8.7	8.4	3.3
8.928	25	15	commercial	63	1	A		6.7	59.7	17.8	12.2	31.9
8.928	25	15	commercial	63	1	B		6.8	60.7	13.3	10.3	22.5
8.928	25	15	commercial	63	2	A		7.1	55.6	12.6	11.9	5.9
8.928	25	15	commercial	63	2	B		5.2	54.4	14.2	8.4	40.5
8.928	25	15	commercial	63	3	A		4.4	52.8	16.3	7.6	53.6

8.928	25	15	commercial	63	3	B		6.2	51.8	9.8	8.7	10.8
8.928	35	15	commercial	71	1	A		7.5	58.2	11.6	10.2	12.2
8.928	35	15	commercial	71	1	B		7.6	58.4	14.4	12.9	10.7
8.928	35	15	commercial	71	2	A		7.4	56.5	13.5	11.6	14.5
8.928	35	15	commercial	71	2	B		7.8	54.7	11.2	10.5	5.8
8.928	35	15	commercial	71	3	A		8.6	54.3	11.8	9.2	21.5
8.928	35	15	commercial	71	3	B		5.4	56.3	14.1	11.7	17.0
31.25	15	15	milled	102	1	A		5.2	60.6	13.6	11.9	12.3
31.25	15	15	milled	102	1	B		4.6	62.6	17.5	10.5	40.1
31.25	15	15	milled	102	2	A		4.3	66.0	26.2	11.3	56.9
31.25	15	15	milled	102	2	B		4.3	66.4	26.6	12.2	54.1
31.25	15	15	milled	102	3	A		5.8	60.9	15.1	9.4	37.9
31.25	15	15	milled	102	3	B		4.8	61.4	13.1	8.1	38.1
31.25	25	15	milled	110	1	A		5.2	67.8	19.9	15.1	24.3
31.25	25	15	milled	110	1	B		4.6	67.4	18.8	12.4	33.7
31.25	25	15	Milled	110	2	A		5.5	63.9	15.5	11.2	27.6
31.25	25	15	milled	110	2	B		4.9	63.7	13.6	10.4	23.7
31.25	25	15	milled	110	3	A		5.0	60.5	15.3	8.7	42.7
31.25	25	15	milled	110	3	B		5.4	59.5	11.9	10.7	10.1
31.25	35	15	milled	118	1	A		4.9	62.5	17.5	11.4	34.7
31.25	35	15	Milled	118	1	B		5.1	63.2	21.8	9.8	54.9
31.25	35	15	Milled	118	2	A		6.7	57.1	11.6	9.3	19.4
31.25	35	15	Milled	118	2	B		6.6	57.7	12.8	11.2	12.5
31.25	35	15	Milled	118	3	A		6.1	62.4	18.5	11.5	37.8
31.25	35	15	Milled	118	3	B		5.3	61.9	17.7	10.4	41.1
31.25	15	15	milled	103	1	A		5.7	53.3	12.2	10.3	15.6
31.25	15	15	milled	103	1	B		6.3	54.7	13.3	8.3	37.1
31.25	15	15	milled	103	2	A		5.3	53.7	12.3	9.7	21.7
31.25	15	15	milled	103	2	B		6.2	56.6	12.8	8.6	33.0
31.25	15	15	milled	103	3	A		6.7	55.5	14.5	10.4	28.4
31.25	15	15	milled	103	3	B		6.5	54.0	10.4	11.7	-13.0
31.25	25	15	milled	111	1	A		5.4	46.8	12.2	9.5	22.1

31.25	25	15	milled	111	1	B		4.5	48.3	16.3	6.9	57.6
31.25	25	15	milled	111	2	A		5.9	54.3	10.5	9.4	10.3
31.25	25	15	milled	111	2	B		6.4	53.8	11.8	9.7	17.6
31.25	25	15	milled	111	3	A		5.4	51.4	16.9	13.4	20.7
31.25	25	15	milled	111	3	B		4.1	46.2	10.8	8.9	17.4
31.25	35	15	milled	119	1	A		4.9	58.8	14.6	8.5	41.9
31.25	35	15	milled	119	1	B		4.7	53.8	17.2	9.7	44.0
31.25	35	15	milled	119	2	A		8.7	51.0	10.7	9.3	12.9
31.25	35	15	milled	119	2	B		5.9	50.8	9.9	9.2	6.7
31.25	35	15	milled	119	3	A		5.6	53.1	9.2	9.7	-5.6
31.25	35	15	milled	119	3	B		6.8	51.8	12.3	8.4	32.2

* Milled is semolina milled from fumigated grains. ** Commercial is semolina supplied commercially and fumigated. Mixo rep is mixograph repeat. MPT is maximum peak time. MPH is maximum peak height. WAP is width of mixogram at peak time. WA8 is width of peak at 8 minutes. RBD is resistance breackdown.

Table.A.64.Raw data of ash content%.

Commodity	SF mg/L	Temperature°C	Moisture%	replicates		
				1	2	3
flour	4.167	15	12	1.80	1.76	1.64
flour	8.928	15	12	1.67	1.69	1.68
flour	31.25	15	12	1.70	1.70	1.61
flour	control	15	12	1.86	1.99	1.90
semolina	4.167	15	12	0.98	0.97	0.94
semolina	8.928	15	12	0.89	0.95	0.93
semolina	31.25	15	12	0.89	0.91	0.90
semolina	control	15	12	0.99	0.97	0.92
flour	4.167	15	15	1.72	1.76	1.69
flour	8.928	15	15	1.97	1.93	1.83
flour	31.25	15	15	1.67	1.69	1.75
flour	control	15	15	1.90	1.94	1.91
semolina	4.167	15	15	0.95	0.97	0.95
semolina	8.928	15	15	0.99	0.89	0.92
semolina	31.25	15	15	0.90	0.97	1.00
semolina	control	15	15	0.98	0.99	0.97
flour	4.167	25	12	1.71	1.68	1.70
flour	8.928	25	12	1.70	1.77	1.67
flour	31.25	25	12	1.70	1.71	1.62
flour	control	25	12	1.95	2.03	1.99
semolina	4.167	25	12	0.97	0.95	0.96
semolina	8.928	25	12	0.94	0.92	0.93
semolina	31.25	25	12	0.87	0.88	0.91
semolina	control	25	12	0.95	0.98	0.94
flour	4.167	25	15	1.70	1.71	1.68
flour	8.928	25	15	1.93	1.92	1.99
flour	31.25	25	15	1.72	1.71	1.78
flour	control	25	15	2.00	1.98	1.99
semolina	4.167	25	15	0.86	0.96	0.89
semolina	8.928	25	15	0.92	0.85	0.90
semolina	31.25	25	15	1.01	0.87	0.96
semolina	control	25	15	0.97	0.97	0.96
flour	4.167	35	12	1.67	1.62	1.70
flour	8.928	35	12	1.69	1.64	1.75
flour	31.25	35	12	1.76	1.66	1.88
flour	31.25	35	12	1.70	1.88	0.88
flour	control	35	12	1.89	0.93	0.95
semolina	4.167	35	12	0.96	0.95	0.94
semolina	8.928	35	12	0.99	0.94	0.92
semolina	31.25	35	12	0.99	1.00	1.73
semolina	control	35	12	1.00	1.73	1.92
flour	4.167	35	15	1.83	1.79	1.69
flour	8.928	35	15	1.91	1.85	1.70
flour	control	35	15	1.90	1.88	1.89

semolina	4.167	35	15	0.86	0.86	0.90
semolina	8.928	35	15	0.91	0.93	0.93
semolina	31.25	35	15	0.98	0.90	0.92
semolina	control	35	15	0.97	0.94	0.93

Table.A.65. Raw data of over cooking time firmness.

Sample ID	Fumigation conditions	Force 1	area1:2
43001	semolina 35°C 12% moist 4.167SFx360h mg/l/h	305.97	101.33
43002		280.04	90.73
43003		307.02	103.82
43004		292.22	93.12
99005	semolina 15°C 12% moist 31.25SFx48h mg/l/h	300.53	96.76
99006		299.38	97
99007		284.4	89.6
99008		298.74	98.29
27009	semolina 15°C 12% moist 4.167SFx360 mg/l/h	340.3	114.2
27010		348.9	120.1
27011		343.1	117.1
27012		346.4	118.1
MANI013	manildra2013	353.49	119.61
MANI014		370.37	126.03
MANI015		358.02	122.69
MANI016		350.66	117.72
1115D017	control///semolina 25°C 12% moist 0SFx360h mg/l/h	326.66	109.57
1115D018		320.77	105.53
1115D019		328.39	111.22
1115D020		342.08	117.03
35021	semolina 25°C 12% moist 4.167SFx360h mg/l/h	319.96	106.18
35022		314.56	105.78
35023		306.28	103.43
35024		312.14	102.5
3025	control//semolina 15°C 12% moist 0SFx168h mg/l/h	331.22	115.7
3026		342.96	120.9
3027		336.9	119.37
3028		336.69	118.35
1915D029	control///semolina 35°C 12% moist 0SFx360h mg/l/h	324.29	108.35
1915D030		341.92	116.98
1915D031		344.24	122.67
1915D032		324.62	108.44
115033	semolina 35°C 12% moist 31.25SFx48h mg/l/h	324.6	108.4
115034		329.6	108.4
115035		349.7	118.8
115036		329.9	111.4
107037	semolina 25°C 12% moist 31.25SFx48h mg/l/h	342.9	117.9
107038		354.3	117.9

107039		359.7	121.8
107040		347.1	117.2
315d041	control/// semolina 15°C 12% moist 0SFx360h mg/l/h	356.1	121.1
315d042		363	123.3
315d043		360.7	124.4
315d044		347.2	116.9
11046	control/// semolina 25°C 12% moist 0SFx 168h mg/l/h	353.77	122.72
11047		340.45	116.32
11048		341.81	116.7
11049		348.55	120.45
19050	contro;/// semolina 35°C 12% moist 0SFx168h mg/l/h	336.85	114.57
19051		345.36	120.59
19052		339.22	117.31
19053		356.35	124.58

Table.A.66. Raw data for baking study.

SF mg/L, 25°C and 12% Moisture	Sample,	loaf #	av loaf vol	app.	vol score	oven spring score	dist	Struc	Soft & Res	total score	total score	L*	a*	b*
0	sample 9	6	413	12	16.5	2.8	6	4	7	48.3	53.6	71.2	3.8	19.4
0	sample 9	10	408	12	16.3	4.9	6	5	7	51.2	56.9	72.1	3.5	19.0
0	sample 9	12	438	12	17.5	5.0	6	6	7	53.5	59.4	72.3	3.5	19.0
4.167	sample 33	4	425	12	17	4.2	6	6	7	52.2	58.0	72.0	3.5	19.1
4.167	sample 33	7	420	12	16.8	5.0	6	5	7	51.8	57.6	72.0	3.5	19.0
4.167	sample 33	15	413	12	16.5	2.8	6	4	7	48.3	53.6	71.9	3.5	18.8
8.928	sample 57	5	418	12	16.7	4.2	6	4	7	49.9	55.5	71.8	3.5	19.1
8.928	sample 57	8	420	12	16.8	4.2	6	6	7	52.0	57.8	71.9	3.5	19.0
8.928	sample 57	13	428	12	17.1	6.1	6	4	7	52.2	58.0	72.0	3.5	19.0
31.25	sample 105	3	438	12	17.5	2.8	6	5	7	50.3	55.9	72.4	3.4	19.1
31.25	sample 105	9	423	12	16.9	5.0	6	4	7	50.9	56.5	71.6	3.6	19.1
31.25	sample 105	14	433	12	17.3	4.7	6	6	7	53.0	58.8	72.1	3.4	18.9
	Base Flour	11	540	14	21.6	10.2	10	10	10	75.8	84.3	82.2	-0.1	14.9

Table.A.67. Raw data for fluoride residues study.

Sample	SF mg/L	Temperature°C	Moisture%	interpretation	Fumigation conditions	Residue mg/kg
50 percent filled				50% filling ratio	Durum grains fumigated with 1mg/l SF at 12% and 25°C for 168 hours	6.8
70 percent filled				75% filling ratio		7.5
95 percent filled				95% filling ratio		9.2
Mf cont				Multiple fumigation control	Wheat grains fumigated with 8.928 mg/l at 12% moisture and 25°C for 168 hors	<0.2
Mf1				Grains fumigated for one time		<0.2
Mf2				Grains fumigated for two times		3.3
Mf3				Grains fumigated for three times		5.1
Mf4				Grains fumigated for four times		8.5
Mf 5				Grains fumigated for five times		13
Ds 50	8.9	15	12	Durum sorption sample 50	8.928, 168 15c 12%	7.4
Ds 58	8.9	25	12	Durum sorption sample 58	8.928, 168 25c 12%	9.9
Ds 66	8.9	35	12	Durum sorption sample 66	8.928, 168 35c 12%	13
Grain-milling - semolina						
Ds 58	8.9	25	12	Durum sorption	8.928, 168 25c 12%	9.9

				sample 58		
Ms 58	8.9	25	12	Milled semolina sample 58	8.928, 168h 25 12% mc	3.3
Bran 58	8.9	25	12	Bran from milled sample 58	8.928, 168h 25 12% mc	10
Ms 34	4.1	25	12	Milled semolina sample 34	4.167, 360h, 25c 12% mc	2.2
Ms 34	4.1	25	12	Bran from milled sample 34	4.167, 360h, 25c 12% mc	9.4
Ms 34	31	35	12	Milled semolina sample 114	31.25, 48h 35c 12%mc	3.9
Bran 114	31	35	12	Bran from milled sample 114	31.25, 48h 35c 12%mc	33
Commercial semolina with and without fumigation						
Ss cnt 3	0	15	12	Semolina sorption control sample 3	168h, 15c 12%	1.1
Ss cnt 11	0	25	12	Semolina sorption control sample 11	168h, 25c 12%	0.2
Ss cont 19	0	35	12	Semolina sorption control sample 59	168, 35c 12 %	0.4
Ss 51	8.9	15	12	Semolina sorption sample 51	8.928, 168h, 15c 12%	4.4
Ss 59	8.9	25	12	Semolina sorption sample	8.928, 168, 25c 12 %	6.1

				59		
Ss 67	8.9	35	12	Semolina sorption sample 67	35c 12% mc	7.2
Fumigated uncooked pasta t59	8.9	25	12	Its uncooked psata sample no 59	8.928, 168h , 25c , 12%	2.3
cooking Water for fumigated sample 59 (fdried)	8.9	25	12		8.928, 168h , 25c , 12%	53
control uncooked pasta for (59) rod 1	0	25	12	Is control for T59	168h , 25c , 12%	0.4
Cooking Water for control sample 59 (fdried)	0	25	12		168h , 25c , 12%	5.3
Cooked pasta control 59	0	25	12	Not fumigated and cooked pasta	8.928, 168h , 25c , 12%	0.2
Fumigated cooked pasta 59	8.9	25	12		8.928, 168h , 25c , 12%	6.7