International Journal of Chemical Studies

P-ISSN: 2349–8528
E-ISSN: 2321–4902
IJCS 2017; 5(5): 1499-1504

Received: 03-07-2017
Accepted: 04-08-2017

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Fumigation of stored-products: Current needs and practices, and future outlook

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Abstract
Food grains if stored in proper conditions can be stored for several years, with very minimal detectable level of quality loss. But under improper storage conditions, the losses may be very severe. Fumigation is one of the most effective and quick insect control techniques which have become popular all over the world in the recent years. Fumigants are gaseous pesticides that are slightly heavier than air and have the ability to diffuse to all areas of the sealed container. Methyl bromide had been used as a quarantine chemical for the treatment of agricultural products and for the control of insects in buildings and commodities since 1930s. As methyl bromide is identified as an ozone depleting chemical, many other fumigants like chloropicrin, sulfuryl fluoride, phosphine and carbon dioxide are proven by various researchers as an effective replacement for the methyl bromide. Major insects all around the world developed various levels of resistance for chemicals due to the extensive use of these fumigants. Use of extreme temperatures by application of heat and cold to the stored product is also an effective technique for insect control to some extent. Modified atmospheric storage (MAS) which involves altering of the storage environment is by far the best method to control the insect infestation in stored product.

Keywords: Fumigation, Methyl Bromide, Chloropicrin, Sulfuryl Fluoride, Phosphine, Carbon dioxide, Modified Atmospheric Storage (MAS).

Introduction
Food grains if stored in proper conditions can be stored for several years, with very minimal detectable level of quality loss. But under improper storage conditions, the losses may be very severe. According to Food and Agricultural Organisation, the yearly global quantitative loss of the cereal grains and oil seeds is approximately 30% and 20% respectively (Anon., 2012) [3]. A major loss of food grains occurs during storage due to storage environment and spoilage organisms. Written records about the infestation of stored products had been reported in written records since 3000BC (Levinson and Levinson, 1994) [22]. High moisture content, high temperature along with excessive broken kernels and dockage provide a favourable condition for the infestation. Controlling of these external factors and the use of proper sanitary conditions before storage will prevent initial infestation in stored products. In the recent years, various prophylactic measures to prevent insects from attacking stored products like grains has been developed and successfully implemented. Attack of insects on stored grain can be prevented by doing proper harvest (avoid mechanical damage of the grains) and by maintaining the storage environment clean and dry before storage.

Insects once enter into the stored product; there are few practical techniques to get rid of these insects. Small quantities of products can be heated or frozen to kill all the insects. Grains are always handled in large quantities; control of infestation is more challenging. Ideal conditions for most of the stored product insects are 25-32 °C and 65-75% relative humidity (Gould and McGuire, 2000) [6], so the growth of insects in stored product depends on the habitat in which they live. The manipulation of the stored product habitat can slow down the increase in pest populations or can eliminate infestations. If anything above or below these ideal conditions, insect’s growth and fitness are reduced, and in more extreme conditions insects eventually die (Fields and White, 2002) [15]. The main advantage of this physical control approach is unlike chemical insecticides, no regulatory approach is needed. These techniques will not leave any residue on the product or the treated area and are having low risk to the workers than chemical methods. However, these techniques need hermetic sealing of grain storage bin in order to maintain the desirable temperature and relative humidity for the required time. During recent years, various studies are conducted to understand the behaviour of insects in response to the parasitoids on them. Over 50 parasitoids and predators attack over 75 stored-product insect pests (Fields and White, 2002) [15]. Use of predators will not leave any chemical residues, and registration is simpler than chemical insecticides. The main backdrop of this technique to control the infestation in stored product is that the predators or parasitoids will contaminate the grain bulk with the body parts and excreta. This disadvantage can be overcome by the use of contact insecticides that are less toxic to mammals. The grain protectants like organophosphorus compounds are applied to the grain before infestation. The use of contact chemicals such as malathion, pirimiphos-methyl, chlorpyrifos-methyl, fenitrothion and dichlorvos, has led to extensive insect resistance throughout the world (Subramanyam and Hagstrum, 1996) [34]. Contact insecticides degrade and produce the components that are effective against insects, this process is relatively slow and time consuming. Due to this problem with contact insecticides, there is an increase in popularity for the use of gaseous chemicals called fumigants. Fumigants are gaseous pesticides that are slightly heavier than air and have the ability to diffuse to all areas of the sealed container.

In the recent years, fumigants have proven to be a very effective and quick insect control technique. Therefore, the aim of the paper is to review the potential effect and use of various fumigants for controlling stored product infestation on the basis of published studies done so far.

Fumigant advantages

- highly toxic to all forms of life; hence kill all stages of the insect
- heavier than air, can seep into smallest of cracks and crevices
- produce lesser residues when compared with contact insecticides
- applied without distributing the commodity
- usually available and economic to use

Fumigants disadvantages

- highly toxic to humans and other mammals, hence needed to be applied with proper care
- proper handling of fumigant is needed, hence need trained applicators
- for higher efficiency, application area should be air tight
- when used in higher concentrations, can reduce seed germination rate and leave residues
- will not prevent re-infestation after fumigation

Fumigants for the control of stored product insects

Fumigants are the gaseous pesticides with high capability to seep through the smallest cracks and crevices. This seepage ability of the fumigant also poses three major problems for the applicators:

- the pest in the stored product will not be killed without the enough quantity of the fumigant inside the structure
- loss of fumigant to the atmosphere, is directly loss of economy
- this seepage ability of the fumigant also means that it can also escape easily through electrical conduits, pipes, augers, and other passageways into adjacent buildings and may cause adverse effects on the humans and animals

Various chemical fumigants that are used worldwide for insect control in stored products include:

Methyl bromide

Methyl bromide has been used as a quarantine chemical for the treatment of agricultural products and for the control of insects in buildings and commodities since 1930s (Fields and White, 2002) [15]. Since then this chemical is used as a soil fumigant; being effective against insects, nematodes, weeds and soil-borne pathogens, including fungi, viruses and bacteria. It is also used as a quarantine purposes for durable and perishable products (Tylor, 1994) [35]. Paster *et al*. (1979) [28] had studied the effect of concentration and time of exposure of methyl bromide on different fungal species. Fumigation of wheat grains with methyl bromide at concentration of 40 mg/l for 24h had caused 100% mortality of spores of Aspergillus and Penicillium species. Various reports published (Anonymous 1990a, 1990b) [1, 2] have drawn attention of the world towards ozone depletion property of methyl bromide. Consequently, in Copenhagen, 1992, the Fourth Meeting of the Parties to the Montreal Protocol on Substances that Deplete the Ozone Layer (an international agreement that controls the production and consumption of ozone-depleting substances) decided to place methyl bromide in ozone depleting substances and all developed countries are scheduled to eliminate the bulk of their consumption of methyl bromide by 2005 (Fields and White, 2002) [15]. Methyl bromide was scheduled for worldwide withdrawal from routine use as a fumigant in 2015, under the directive of the Montreal Protocol on Substances that Deplete the Ozone Layer (Schneider *et al*., 2003) [32]. The loss of such an important insecticide that served the world for about 60 years has forced managers in food industry, pest control operators, and entomologists to find alternative chemicals.

Chloropicrin

Chloropicrin is a colourless, non-flammable liquid fumigant that is toxic to fungi, insects, mites and rodents in the storage area. It can also control plant parasites, nematodes, and certain soil bacteria. The chemical name of this fumigant is Trichloronitromethane, and commonly called as tear gas as it causes irritation in the eyes when exposed. Chloropicrin was scientifically known hundreds of years ago, but it was commercially got popularity only during 1920s. For many years, chloropicrin was very popular in grain and milling industries because the residual left on the grain kernel continued to kill the insects and rodents. Johnson (1937) [20] conducted experiments on flour warehouse infected with insects of all stages and stated that the proper concentration of chloropicrin can kill all the life stages of the insects. Concentration of this fumigant effects the penetration of the chemical into the grain to kill the various life stages of weevils. He also identified that the ¼ pound of chloropicrin per 1000 cubic feet of warehouse can cause irritation and finally death due to putrefaction for rodents. Witherspoon, and Garber (1922) [38] identified that even a low concentration of 1 to 3ppm of chloropicrin can cause tears in the eyes of conscious person, as the concentration is increased irritation of the membranes in nose, throat, and bronchial tubes occur progressively. Experimental work indicates the limit of human voluntary toleration for a 2-minute period as about 22 ppm. Some times when higher concentration of this chemical are used for fumigation, the chemical residues continues through the processing chain and contaminates the final product.

Sulfuryl fluoride

Sulfuryl fluoride is a popular fumigant that is considered as a feasible alternative to the methyl bromide for the fumigation of storage structures like empty flourmills and harvested agricultural products (Drinkall *et al*., 1996) [13]. Sulfuryl fluoride is a biologically active inorganic chemical that is odourless, colourless, non-corrosive and non-flammable and essentially non-reactive at working airborne concentrations (Reichmuth *et al*., 1997) [29]. Sulfuryl fluoride is having the ability to penetrate into infested matrices like intact wood and other grain commodities. This fumigant controls the insect infestation by disturbing the glycolysis cycle leading to less metabolic energy production (Meikle *et al*., 1963) [23]. Bell *et al*., (1999) [8] studied the effect of sulfuryl fluoride concentration and temperature of exposure on different age groups of the eggs of Mediterranean flour moth (Ephestia kuehniella). They have identified that at all the concentrations and temperatures used in the experiment, eggs aged 1-2 days were more tolerant than other age groups, followed by 2-3 day-old, 0-1 day-old and 3-4 day-old eggs. At higher temperature of 25 °C, eggs of Mediterranean flour moth needs lesser concentration of sulfuryl fluoride (1000 mg.h/l to prevent hatch and 800 mg.h/l to prevent adult emergence) than at lower temperatures of 15°C (about 4000 mg.h/l to prevent hatch and 3000 mg.h/l to prevent emergence). Baltaci *et al*., (2009) [5] studied the lethal effects of sulfuryl fluoride on different life stages of the warehouse moth Ephestia elutella (Hübner) exposed to different concentrations of sulfuryl fluoride for varying exposure times. They have identified that 11.6g/m3 concentration of the fumigant, exposure for 18h killed all the larvae and pupae of warehouse moth. When compared to larvae and pupae, eggs require more concentration for more exposure time to completely kill them. Athanassiou *et al*., (2012) [4] has evaluated the effect of sulfuryl fluoride for controlling all life stages of psocids (Liposcelis sps.). A highest concentration of 31.25g/m3 is needed for 48h for obtaining complete mortality of all the life stages of the psocids. Calvert *et al*., (1998) [10] studied the about health of the structural fumigation workers associated with the use of sulfuryl fluoride as a fumigant. Although sulfuryl fluoride was an effective chemical for fumigation of storage structures, it had been understood that long term exposure of humans to sulfuryl fluoride may cause adverse effects on olfactory function and some cognitive functions.

Phosphine

Phosphine (chemical name: phosphane) is the most widely used fumigant that gained popularity after the worldwide withdrawal of methyl bromide in 1990s. Phosphine is sold in tablets or pellets form of aluminium phosphide or magnesium phosphide, which react with the water vapour in the air (when moisture content is more than 10% wet weight) to form phosphane gas and aluminium hydroxide or magnesium hydroxide (Fields and White, 2002) [15]. Commercial phosphine solid fumigants contain other minor ingredients like ammonium carbonate, ammonium bicarbonate, urea and paraffin to regulate the gas release and decrease flammability (Harein and Davis, 1992) [17]. Hole *et al*., (1976) [18] have studied the toxicity of phosphine fumigation on the mortality of all the life stages of Acanthoscelides obtectus, Caryedon serrutus, Cryptolestes ferrugineus, C. pusillus, Lasioderma serricorne, Oryzaephilus surinamensis. Ptinus tectus, Rhyzopertha dominica, Sitophilus granarius, S. oryzae, S. zeamais, Tribolium castaneum and Trogoderma granarium. Test was conducted with storage temperature from 10 to 30°C, relative humidity of 70%, with concentration ranging from 0.013 to 2.96 mg/l and exposure time from 1 to 16 days. They have identified that pupa and larvae were the most resistant life stages. Sitophilus spp. was the most tolerant among all the above species. Their study final reviled that phosphine was most effective at the higher temperatures, for long exposures at low concentrations were far more effective than short exposures at high concentrations. Ridley *et al*., 2011 [30], studied the effectiveness on fumigation of silo bag with phosphine gas. Silo bags containing wheat were artificially infested with lesser grain borer R. dominica (F.). Complete control was achieved by fumigating the sample in silo bag with phosphine gas at a rate of 1.5 g/m3 for a fumigation period of 17 days. One major back drop of using phosphine as a fumigant is its slow activity time. Instead of using phosphine as a solid pellets, various other combination of techniques are used in order to decrease the duration needed for treatment and to increase the insect mortality. In Australia for Siroflor, phosphine is used as a compressed gas in combination with 98% carbon dioxide was used to reduce combustion (Winks and Ryan, 1990) [37]. For the purpose space fumigation, phosphine was used along with heat and carbon dioxide (Mueller, 1995) [26]. Other main drawbacks of this fumigant are that it is highly inflammable at higher concentrations above 1.8% v/v, corrosive character on most of the metals like copper and silver. During recent years, due to the rapid use of phosphine, stored product pests developed resistance to this chemical worldwide (Mills, 2001) [24].

Carbon dioxide

Carbon dioxide (CO2) is a colourless, odourless and tasteless gas than is heavier than air. It is a non-combustible gas, so is generally used as a fire-extinguishing material. It is usually found in the air at a concentration of 0.03% by volume. However, at higher concentrations, it is poisonous. CO2 is used for the fumigation of stored product at higher concentrations of 60% by volume. Newton 1993 [27], studied the effect of CO2 on the stored product insects and pests at different exposure time. Mixed cultures of Sitophilus orzae (L.), T. castaneum (Herbst), Liposcelis bostrychophila Badonne, Tyrophagus putrescentiae (Schrank) and Acarus siro L. were exposed to 60% CO2 concentration at different exposure time of 4, 7 and 14 days. Complete control of T. castaneum required 7 days exposure; S. orzae, L. bostrychophila and A. siro required 14 days. T. putrescentiae was highly tolerant to CO2 fumigation and survives even after 14 days of exposure time. Wang *et al*., (2009) [36] studies the combined effect of carbon monoxide and carbon dioxide on the mortality of rusty grain beetle, C. ferrugineus (Stephens), red flour beetle, T. castaneum (Herbst), and granary weevil, S. granarius (L.) at different conctrations of CO and CO2. In their experiment, they have determined that carbon monoxide has no effect on the mortality of adults of all the three species. Mixture of CO and CO2 had shown the better morality in all the three species than CO2 alone at lower concentrations. The main advantage of using CO2 as a fumigant is that it will not leave any toxic residue or off-odours on the stored product. Also CO2 will not change the germination potential of the treated grains. However, fumigation of stored product using CO2 requires fairly longer exposure time and the effectiveness of the fumigant is greatly influenced by the storage temperature. At lower temperatures, effectiveness is greatly reduced. In environmental safety point of view CO2 is also seen as a greenhouse gas. Carbon dioxide is a by-product of existing industrial processes, such as the production of hydrogen gas from petroleum. Therefore, CO_2 fumigation does not itself add to the ‘‘greenhouse effect’’ (Newton 1993) [27].

Fumigation: Future outlook

For many years, chemical control measures have dominated the methods of pest control in grains, but recently more interest has been directed towards non-chemical methods (Mourier and Poulsen, 2000) [25]. Due to the extensive use of chemicals worldwide, all major species of insects have developed varying degrees of resistance to almost all the popular insecticides (Champ and Dyte, 1976) [11]. At the same time, new chemicals that are safe enough for use on foodstuffs are only slowly becoming available, because of the high cost of developing and registering them. Increasingly sophisticated markets are expecting that cereals and other foodstuffs is “residue free”, and in organic food production the use of insecticides is ruled out (Mourier and Poulsen, 2000) [25]. So here comes the need for development of alternative techniques for fumigation of grains. Unlike most other insect pests, stored-product pests live in an environment that is largely determined by humans. The manipulation of this stored product environment can slowdown the increase in pest population or even used to eliminate them (Fields and White, 2002) [15]. These techniques to vary the stored product environment can solve all the problems that are associated with chemical fumigants.

Heat

A technique that has been used successfully for many years to control the stored product pests is the use of elevated temperatures. The recorder use of elevated for stored product was heating of grains to 69 °C to control Sitotroga cerealella in France in 1762. The popularity of using heat to control the insects was reduced due to the increase in use of chemical fumigants in early 90s. Extensive research has been conducted to obtain the correct control temperature and exposure time for the control of each individual insect species. Generally stored products pests are controlled at 40 °C for 24h or 45 °C for 12h or 50 °C for 5 min or 55°C for 1 min or 60 °C for 30s (Fields, 1992). During the recent times there is increase in popularity for the use of high temperature short time technique for controlling stored grain pest. Mourier and Poulsen (2000) [25] have proven that this technique can control different stored product insects and mites. They had conducted experiments with grain mite (Acari) and S. granarius infested wheat sample and Prostephanus truncates infested maize sample. In this test they had observed the complete control of grain mites and adult of S. granarius was obtained at 300-350 °C for 6s. Complete control of P. truncatus in maize was obtained at 700 °C for 19s. Denlinger and Yocum (1998) [12] postulated that exposure of stored product pest to higher temperatures hampers the cell functioning in critical tissues, rather than general damage of all cells. Using of elevated temperature for the control of stored product sometimes bring out undesirable changes in the grain sample like reduction in moisture content and germination rate of the sample. So the use of correct temperature and correct exposure time are needed for obtaining the complete control infestation without harming the functional properties of the grain.

Cold

Reproduction of stored product insect is generally stopped below 18°C except S. granarius, which can reproduce at temperature down to 15 °C (Fields and White, 2002) [15]. Below 5 °C they show only restricted moment. Application of lower temperatures is also used in certain parts of the world for the control of the insects. In temperate countries, where temperature goes down to -20 °C, cold treatment for the storage structures is practiced by forcing the outer cold air into the structure using fans, but this method of control was gradually replaced by spot treatments with a contact insecticides and fumigants. In some parts of the world, alternative to the cooling of the storage structure, only the stored product might be disinfested using lower temperature. Infestation in dried fruits and nuts is controlled using cold treatment of the sample. Commercial grain chillers are used in countries like Australia, Europe and United states to cool the seeds, usually to 15 °C, to prevent the losses due to insect infestation and grain respiration (Burks *et al*., 2000) [9].

Modified Atmospheric Storage (MAS)

Modified atmospheric storage is the food preservation technique that preserves the food natural quality as well as extends the storage life. Modified atmosphere storage has an important role to be played in the integrated pest management (IPM) which ensures no pesticide residues (Banks *et al*., 1991) [7]. Since Montreal Protocol had phased-out methyl bromide in 2005, modified atmospheric storage is the most effective and environmentally friendly technique to preserve food grains and other perishables from insects and other pests. Disinfestation of stored grains using modified atmosphere (MA) is done by altering the storage atmosphere gases such as carbon dioxide (CO_2), oxygen (O_2) and nitrogen (N_2), to render the atmosphere in the stores lethal to pests (Jayas and Jeyamkondan, 2002) [19]. Modified atmospheres can be achieved by adding solid or gaseous CO_2 or by removing O_2 (replacing with CO_2 or N_2 gas) or by creating an airtight conditions after removing the O_2 from the atmosphere. These three conditions are referred to as high CO_2, low O_2 and hermetical storage respectively and collectively called as modified atmospheric storage (Banks & Fields, 1995) [6]. The main infectious organisms on grains are insects and moulds. Most of the insects and moulds cannot grow well at reduced oxygen concentration in the atmosphere. Modified atmospheric storage in grains is mainly concerned with reducing the oxygen concentration by either replacing it with CO_2 or N_2 gases. Stored product insects develop resistance to chemical fumigants as well as modified atmospheric storage. But the resistance developed is relatively low in case of modified atmosphere when compared to the chemical fumigants (Krishnamurthy *et al*., 1986) [21]. Jayas and Jeyamkondan (2002) [19] have reviewed the potentiality in the use of modified atmospheres for the storage of grains, meat, fruits and vegetables and opportunities for further research. Spratt (1979) [33] studied the effect of oxygen, carbon dioxide and nitrogen gases in a ratio of 1:1:8 on the mean longevity of the adults of maize weevil (Sitophilus zeamais). In this particular study he had observed that the longevity of adult males of maize weevil was reduced from 56 days in air to 49 days in gas mixture and of females from 75 to 63 days under same conditions. Riudavets *et al*., (2009) [31] had studied the effect of two modified atmospheres (50% and 90% CO_2) on the different life stages of ten stored product pests (L. serricorne, C. ferrugineus, O. surinamensis, Tribolium confusum, R. dominica, Sitophilus oryzae, E. kuehniella, Plodia interpunctella, L. bostrychophila and T. putrescentiae). All the ten pest species and their developmental stages have shown different sensitivity to the two modified atmospheres.

The beetles S. oryzae, R. dominica, C. ferrugineus and L. serricorne were the species that had shown highest tolerance and moths are the most easily killed species among the all. Many research studies have proven that modified atmospheric storage is a very effective technique for the control of pests and moulds in stored products.

Conclusion

Fumigation is the very effective and quick insect control technique. Due the extensive use of chemical fumigants worldwide, all major species of insects have developed varying degrees of resistance to almost all the popular insecticides. At the same time, new chemicals that are safe enough for use on foodstuffs are only slowly becoming available, because of the high cost of developing and registering them. Increasingly sophisticated markets are expecting that cereals and other foodstuffs is “residue free”, and in organic food production the use of insecticides is ruled out. Safer methods like the manipulation of this stored product environment can slowdown the increase in pest population or even used to eliminate them. Use of physical control methods like heat and cold are very useful in controlling the stored product pest. During recent years, modified atmospheric storage (MAS) which involves the variation of stored grain environment by altering the storage atmosphere gases such as carbon dioxide (CO2), oxygen (O2) and nitrogen (N2), to render the atmosphere in the stores lethal to pests has gained popularity to control the stored product pest.

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