REVIEW

Effect of handling and processing on pesticide residues in food- a review

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Abstract Pesticides are one of the major inputs used for increasing agricultural productivity of crops. The pesticide residues, left to variable extent in the food materials after harvesting, are beyond the control of consumer and have deleterious effect on human health. The presence of pesticide residues is a major bottleneck in the international trade of food commodities. The localization of pesticides in foods varies with the nature of pesticide molecule, type and portion of food material and environmental factors. The food crops treated with pesticides invariably contain unpredictable amount of these chemicals, therefore, it becomes imperative to find out some alternatives for decontamination of foods. The washing with water or soaking in solutions of salt and some chemicals e.g. chlorine, chlorine dioxide, hydrogen peroxide, ozone, acetic acid, hydroxy peracetic acid, iprodione and detergents are reported to be highly effective in reducing the level of pesticides. Preparatory steps like peeling, trimming etc. remove the residues from outer portions. Various thermal processing treatments like pasteurization, blanching, boiling, cooking, steaming, canning, scrambling etc. have been found valuable in degradation of various pesticides depending upon the type of pesticide and length of treatment. Preservation techniques like drying or dehydration and concentration increase the pesticide content many folds due to concentration effect. Many other techniques like refining, fermentation and curing have been reported to affect the pesticide level in foods to varied extent. Milling, baking, wine making, malting and brewing resulted in lowering of pesticide residue level in the end products. Post harvest treatments and cold storage have also been found effective. Many of the decontamination techniques bring down the concentration of pesticides below MRL. However, the diminution effect depends upon the initial concentration at the time of harvest, substrate/food and type of pesticide. There is diversified information available in literature on the effect of preparation, processing and subsequent handling and storage of foods on pesticide residues which has been compiled in this article.

Keywords Pesticides · Food · Localization · Handling · Processing · Product manufacture · Storage

Introduction

Food is a more basic need of man than shelter and clothing. It provides adequate nutrients for the body's growth, maintenance, repair and reproduction. India is the second most populous country in the world and its economic scenario is dominated primarily by agriculture sector. Nearly 64% of its population is dependent on agriculture. Food security is one of the major concerns for India, where the diverse agro climatic conditions necessitate different approaches for cultivation and crop protection. With the limitation of expansion of land area under cultivation, most of the gains in agricultural production have come from increased productivity through two major inputs i.e. fertilizers and pesticides.

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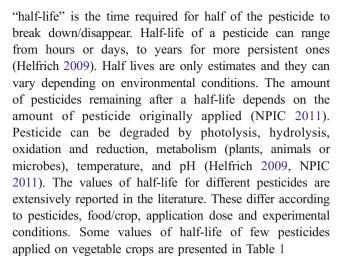Ludhiana 141 004, Punjab, India

The losses of crops caused by pests and plant diseases are quite high both in developed and developing countries. These are reported to be in the range of 10–30% in the former and 40–75% in latter case (Roy 2002). Even greater losses occur after the crop is harvested which are caused by the pests that attack the stored products, particularly in the tropics (FAO 1985). During the last 3–4 decades, chemical control of pests aimed at minimizing these losses has been introduced throughout the world. Approximately 70% of the pesticides used in the world are applied in developed countries and 30% in the developing countries (Pimentel 1987). India stands at 7th position in use of pesticides on crops (Singh 2002). Pesticides slowly start dissipating after these are sprayed. Every pesticide used on crops needs some waiting period before harvesting that differs from pesticide to pesticide and also from one crop to another. Food products become safe for consumption only after waiting period has lapsed. If fruits and vegetables are harvested before completion of the waiting period, they are likely to have higher level of residues, which are hazardous to health. Despite the restrictions and regulations on pesticide use, India accounts for one-third of pesticide poisoning cases in the world. The incidence of excessive pesticide residues may cause blindness, cancer, diseases of liver and nervous system etc. The long term effects could result in reduction of live sperm and fertility, increase in cholesterol levels, high infant mortality rates and several metabolic and genetic disorders (Gupta 2006).

Dissipation vis-à-vis- localization of pesticide residues in food commodities

Composition and properties of various food commodities vary according to their nature and group they belong. The same is true for pesticides. There are numerous factors that affect the extent of pesticide absorbance, penetration and degradation and differ from one category of food to another. The rate at which pesticides are moved and dissipated is closely related to the physico-chemical parameters of pesticide itself and surrounding environmental conditions. The ability to resist degradation (persistence) under various conditions is measured as half-life of the pesticide. The "half-life" is the time required for half of the pesticide to break down/disappear. Half-life of a pesticide can range from hours or days, to years for more persistent ones (Helfrich 2009). Half lives are only estimates and they can vary depending on environmental conditions. The amount of pesticides remaining after a half-life depends on the amount of pesticide originally applied (NPIC 2011). Pesticide can be degraded by photolysis, hydrolysis, oxidation and reduction, metabolism (plants, animals or microbes), temperature, and pH (Helfrich 2009, NPIC 2011). The values of half-life for different pesticides are extensively reported in the literature. These differ according to pesticides, food/crop, application dose and experimental conditions. Some values of half-life of few pesticides applied on vegetable crops are presented in Table 1

The retention of pesticides depends on the physiochemical properties of the pesticide molecules as well as food. In fruits and vegetables, most of the pesticide residues are retained on peel surface (Awasthi 1993). This is the reason that majority of the residues are removed by washing, peeling or treatments with chemical solutions like vinegar, turmeric, sodium bicarbonate, common salt or alcohol (Gupta 2006). Pesticides may be introduced to fruits and vegetables during different phases of production. Some pesticides are used before blooming, some while fruits are growing and others after harvesting. Therefore the location of pesticides in the same fruit may be different (Trewavas and Stewart 2003). Very little quantity of systemic pesticides may be absorbed into flesh (Lewis et al. 1998). The residue level in juices from fruit or must from grapes depended on the partitioning properties of the pesticide between the fruit skins and the juice. Therefore, lipophilic residues were poorly transferred to juice and a substantial amount was retained in the by-products, which frequently included the skin (Halland et al. 1994). In fruits and fruit-type vegetables, the concentration of pesticide residue was higher in the fruit stalk and near the epidermis (exocarp and fruit receptacle) than in the sarcocarp or pericarp. In leaf vegetables, concentration of the pesticide residue was higher in outer leaves than in inner ones (Yoshida et al. 1992). The leaching of pesticides from the surface of fruits and leafy commodities is due to their solubility in water (Jaggi et al. 2000). The effectiveness of washing is also

Table 1 Half-life values of few pesticides on vegetable crops

Crop	Name of pesticide	Application dose, g a.i/ha	$t_{1/2}$, days	Reference
Tomato	Flubendiamide	48	0.33	Kooner et al. (2010)
	Thiacloprid	48	1.18	Kooner et al. (2010)
Brinjal	β-cyfluthrin	18	1.74	Mandal et al. (2010)
	Imidacloprid	42	2.31	Mandal et al. (2010)
	Propargite	570	3.07	Kang et al. (2009)
Chilli	Flubendiamide	60	0.96	Sahoo 2009

dependent and may be reduced for insecticides, specifically on synthetic pyrethroids, due to strong bonding between the insecticide molecules and waxy layer of fruit skin and also their non-systemic and non-translaminar movement characteristics (Elliot 1980, Briggs 1985).The transfer of pesticide residue from dried (made) tea to tea infusion/brew depended on water solubility, partition coefficient and vapor pressure (Kumar et al. 2005).

In cereal grains, most of the pesticides are contained in outer layer of grain i.e. bran and therefore, milling and similar processing techniques remove the residues to varied extent (Udeaan and Bindra 1973). Also, cereal grains are invariably sprayed with insecticides for storage in bulk silos after harvest to reduce losses during storage for 1 year or more at ambient temperature. The residues of more lipophilic pesticides tend to remain on seed coat although a fraction can move to parts containing high levels of triglycerides ie bran and germ. Therefore, residue levels in bran were higher than in wheat by a factor of 2–6. For the pesticides which could enter the grain by translocation, residues were higher in the bran than in the flour (Halland et al. 1994). In other crops like pulses, residues of cypermethrin and deltamethrin could not be removed by washing and cooking in stored grains suggesting that residues penetrated the interior of grains (Hazarika and Dikshit 1992, Lal and Dikshit 2000). In pulses, maximum concentration of cypermethrin was found on seed coats (Dikshit 2001).

Milk and milk products sometimes are found contaminated with insecticides from feed and fodder which remain associated with its fat portion. Therefore, residues may be found in greater concentration (on a fat basis) in milk products (butter, cheese, *ghee, malai*) than milk from which these were manufactured (Li et al. 1970) Also, in meat, pesticides are mainly concentrated in fat component. Those pesticides which tend to be lipid soluble, are found in much higher concentration in egg yolk than in albumin. In contrast, those water soluble/polar, are found in higher concentrations in albumin (Krieger 2001). The most organic pesticides have high affinity for oils which makes their removal extremely difficult from different oils and fats.

The fat soluble insecticides, like organochlorines, after reaching a body of water tend to adhere to suspended organics which are consumed by small invertebrates that dwell upon them. Fish feeding upon small aquatic invertebrates accumulate the pesticides in their tissues, besides directly absorbing them from polluted water around. The organochlorine insecticides with their high lipid solubility easily accumulate in fat tissues of fish. This accumulation of pesticides in fish may be 10 to 10,000 times greater than their original concentrations in water (Kannan et al. 1997).

Effect of handling and processing

Foods after harvest/slaughter are subjected to various handling and processing operations both at home or industry level, involving a simple washing to more multistep and complex processing aimed to extend shelf-life, add variety, increase palatability and nutrient availability and to generate income. The various techniques and methods applied usually reduce residue levels because of washing or cleaning, peeling, blanching, juicing, cooking, milling baking, pasteurization, canning etc. However, some processes may lead to increase in the residue level due to concentration effect and/or affinity for lipid moiety.

Thus there is an increasing need for information about the effects of various processes on the fate of pesticide residues in foods both from a regulatory and public concern perspective. The various aspects covering different foods, preparatory steps, processing into different products and post harvest handling and storage for a large number of pesticides have been elaborated under the following heads:

The available information has been elaborated under the following heads:

I. Preparatory steps	II. Thermal treatment
III. Products manufacture	IV. Post harvest handling

I. Preparatory steps

The extent of pesticide reduction depends upon the washing operations, nature of pesticide molecule and other preparatory steps used. Loosely held residues of several pesticides are removed with reasonable efficiency by varied type of washing processes (Street, 1969). Moreover, majority of pesticides applied to crops are confined to the outer surfaces and undergo limited movement or penetration of the cuticle. Therefore, they are amenable to removal by washing, peeling and trimming operations (Toker and Bayindirli 2003). The effect of different preparatory steps on pesticide residues in food is being described under various subheads:

a) Washing with water: Fruits and vegetables are invariably washed before consumption. Vegetables are often peeled off and cooked prior to eating. Wallis et al. (1957) reported that only one minute washing of okra having an initial deposit of 15.20 ppm of malathion removed the residue to the extent that only traces were detected on washed okra. Dewan et al. (1967) reported that 11.83 cm rainfall washed the initial deposit of carbaryl in okra to nil thereby recording a 100% reduction. Nath et al. (1975) found that 30 s washing of

treated okra with tap water resulted in considerable removal of malathion deposits; recording a reduction of 89.15% and 79.48% respectively. In case of carbaryl the initial deposit as a result of two application dosages reduced to 5.94 and 12.24 ppm respectively by tap water washing of okra for 30 s. Thus, washing decreased the carbaryl deposit by 66.12 and 69.55% for lower and higher application dosage, respectively. Various pesticide decontamination processes like washing the fruits with water were reported to dislodge the residues to varying degrees depending on constitution of the fruit, chemical nature of the pesticide and environmental conditions. Washing was the most effective means of removing pesticide residues and minimizing dietary intakes from cabbage (Yuan et al. 2009). Elkins et al. (1968) also reported that cold water washing removed 96% malathion residue from beans.

Deshmukh and Lal (1969) reported that tap water washing of brinjals treated with carbaryl removed residue to a great extent. Bindra (1973) reported 80–83% reduction of carbaryl by washing of tomato. However, only 18–55% endosulfan was reduced by washing. The removal rates of pesticide residues of dieldrin and heptachlor epoxide in pumpkins and cucumbers, by washing with water or 0.1% liquid detergent were 8–52% and 19–67%, respectively (Yoshida et al. 1992). Removal of the fruit stalk, exocarp and tissue around stalk cavity of fruits and fruit-type vegetables and washing of leaves with water or dilute detergent solution were necessary to decrease the intake of pesticide residues from vegetables and fruits. Removal of methamidophos and carbofuran residue in broccoli during freezing processing was studied by Tsai et al. (1997). Washing for 3 min removed 44.1 and 32.8% of high and low doses of sprayed carbofuran, respectively. Residues in the juice prepared from washed commodities ranged from not detected to 0.83 mμg/g. In case of tomato, Singh and Lal (1966) reported a reduction of 86.20% in malathion deposit by one minute washing only. It was found that washing of tomato fruits in a stream of water for 1–3 h reduced pesticide residues in tomato products; tomato seeds showed higher levels of residues as they were not subjected to processing. (Ramadan et al. 1992).Washing removed more residues from carrots than from tomatoes (Burchat et al. 1998). Persistence of malathion in bell peppers was studied in field experiments (Bhagirathi et al. 2001). Washing of treated fruits with running tap water removed 67–

78% of malathion residues from samples. The level of chlorpyriphos and fanitrothion in artificially contaminated red pepper fruits after harvest were approximately 30–40% after shaking or sonicating the peppers for 5 min in water (Lee 2001).

The washing of the treated mango fruits has been reported to reduce surface residues to the extent of 66–68% for dimethoate and fenthion as compared to 21–27% for pyrethroids at the initial stages of spray treatment. The effectiveness of washing was, however, reduced at later stages of all the insecticides and more specifically on synthetic pyrethroids due to strong bonding between the insecticide molecules and waxy layer of fruit skin and also their non-systemic and non-translaminar movement characteristics (Elliot 1980, Briggs 1985). Approximately 50% of fenitrothion residue was removed on washing of apple fruit during production of concentrated juice from apples treated with 0.15% solution of fenitrothion (Lipowska et al. 1998). Cabras et al. (1998) observed a significant decrease in pesticide residue during the washing treatment of prunes. In case of iprodione, the residue at harvest time was 0.68 ppm and washing for 5 min resulted in 6 fold decline. The prolonged washing of the fruit did not affect the residue level. This was explained by considering that the pesticide penetrated the epicular layer and the cuticola after the treatment (Riederer and Schreiber 1995), thus avoiding direct contact with water and consequently solubilization. The decrease in residue after first washing was not attributable to a solubilization process due to pesticide adsorption by dust on the fruit during treatment. Washing removed both the dust and the adsorbed residue. The residue at harvest time was 0.21 ppm in case of phosalone which decreased to 0.07 ppm by washing. Prolonged washing did not cause any change in the residue level. Furthermore, washing caused an important decrease in iprodione and phosalone, while it did not affect the level of bitertanol and procymidone.

Tejada et al. (1990) reported that usual practice of washing rice and maize before cooking reduced chlorpyriphos residues by 59–100% coming from sprays on jute sacks. Miyahara and Saito (1994) reported that during Tofu production, the pesticides levels were reduced to about 10 and 20% of the initial levels by washing of soyabeans with water twice. Moreover, sprayed pesticides which remained as microparticles on the surface of the soyabeans, were easily removed by mechanical

stirring in water. Thus, 80% of pesticides on the surface were washed away. Decontamination processes such as washing and steaming dislodged the cypermethrin residues in pulses by 37–49% and 63–74%, respectively (Dikshit 2001).

b) *Washing with salt solution*: Washing with dilute salt (sodium chloride) solution is a convenient method to lower the load of contaminants from food surfaces particularly fruits and vegetables. This method could be equally effective for reducing the pesticide residue from other commodities too. This procedure is recommended as being practical for household use. Chlorothalonil was best removed from Chinese cabbage by 1% saline exposure for 10 min (Lee and Chou 1995). Decontamination of cabbage through different processes showed that residues were reduced to some extent by washing and cooking (Nagesh and Verma 1997). Washing the samples with salt water did not differ significantly with ordinary washing with tap water. However, dipping of green chillies in 2% salt solution for 10 min followed by washing in water removed 90.56 and 66.93% of residues from chillies at 0 and 5 days after final spraying, respectively (Phani-Kumar et al. 2000a,b).

c) *Washing with chemical solutions*: Chlorine water and dilute solutions of other chemicals are commonly used for disinfection of fruits and vegetables. These chemicals play an effective role in removing the pesticide residues. Efficacy of chlorinated and ozonated water dips in dissipation of azinphos-methyl, captan and formetanate-HCl was studied in a model system on apples and apple sauce (Ong et al. 1995). Pesticide residues on apples and in apple sauce were reduced by both washes, 500 ppm chlorine (calcium hypochlorite) being most effective. In 1996, Ong et al. (1996) observed that all 3 pesticides in model systems solution decreased by 50–100% with chlorine and ozone treatments. Captan and formetanate-HCl were rapidly degraded in 50 and 500 ppm chlorine solutions at pH 7 and 10.7. Rate of degradation of the pesticides increased at higher pH and temperature. Pesticide residues on fresh apples and in processed apple products were also reduced by chlorine and ozone washes; chlorine (500 ppm) being the most effective wash treatment. Use of ozone wash at 0.25 ppm was not as effective as chlorine wash owing to its low concentration, its instability in water and the high organic content of the wash water.

Fresh apples were treated with 2 different levels of mancozeb (1 and 10 mμg/mL,). Mancozeb residues decreased 56–99% with chlorine and 36–87% with chlorine dioxide treatments. Mancozeb residues decreased 56–97% with ozone treatment. Peroxyacetic acid (HPA) was also effective in degrading the mancozeb residues, with 44–99% reduction depending on treatment time and HPA concentration (Hwang et al. 2001).

Potatoes were washed for 10 min in tap water, an acid solution (5 or 10% solutions of an aqueous extract of radish leaves, acetic acid, citric acid, ascorbic acid or H_2O_2), a neutral solution (5 or 10% NaCl) or an alkaline solution (5 or 10% NaHCO$_3$) (Zohair 2001). Acidic solutions were found to be more effective for extraction of organochlorine pesticides from potatoes than the other washing solutions used. Soliman (2001) reported that washing potatoes with tap water or aqueous solutions of acetic acid and/or NaCl and blanching or frying of potatoes removed most of organochlorine and organophosphorous residues. No profenofos residues were detected in aubergines after washing with soap and acetic acid solutions, and a high percentage of profenofos residues were removed from peppers by acetic acid solution, potassium permanganate and tap water (Radwan et al. (2005).

The results of Wu et al. (2007) on the degradation of four pesticides by low concentration of dissolved ozone indicated that initial concentration of 1.4 ppm was effective to oxidize 60–99% of 0.1 ppm aqueous diazinon, parathion, methyl-parathion and cypermethrin shortly within 30 min. Ozonated water was mostly effective in cypermethrin removal (>60%). The efficacy highly depended on the dissolved ozone levels. Higher temperature enhanced the efficacy in pesticide removal with maximal efficacy for diazinon removal detected at 15–20 °C.

Washings from nectarines using aqueous solutions containing large amounts of hypochlorite, hydrogen peroxide and potassium permanganate contained large amounts of toxic by-products (oxons). Solutions containing sodium lauryl sulfate, glycerol and ethanol removed 50% of pesticide residues (Pugliese et al. 2004). Hadjikinova et al. (2006) showed that washing, particularly with sodium base (1%) or sodium hydrogen carbonate solutions (1.5%) markedly reduced concentration of chlorpyrifos-methyl insecticide and fenarimol) fungicide in cherries.

Kim et al. (2000) treated soybeans with 0.3 ppm ozone water for 30 min during the soaking period. Residues of carbendazim, captan, diazinon, fenthim,

dichlorvos and chlorpyriphos as affected by various soaking/ozonation treatments were examined. Ozone treatments destroyed more pesticide than soaking in pure water, captan being the most susceptible and chlorpyriphos least. Zhanggui et al. (2003) observed that pesticide residue levels were reduced in wheat and corn by ozone treatment at 15–20 ppm, with a maximum degradation rate of 12.3% per day.

d) Kitchen type and combination processing (Trimming, washing, peeling, cooking etc.): Kitchen-type processing techniques could remove 40–77% of diazinon residues and 37–82% of dimethoate residue in green beans and cauliflower (Bognar 1977). It has been reported that washing alone reduced the levels of dimethoate residues by approximately 25–80% whereas washing and cooking of cauliflower curds reduced the level of residues by 52–91% (Khaire and Deethe 1983). Sugibayashi et al. (1996) compared the effects of washing, peeling and cooking on the residue levels in white potatoes and carrots. Chlorfenvinphos-E, S-benzyl diisopropyl phosphorothiolate and monocrotophos-E were efficiently removed by washing alone. However, peeling was found to be the most effective way to remove the pesticide from the vegetables followed by frying. Boiling was effective in reducing the level of water-soluble pesticides. The residues of dimethoate were reduced by approximately 50% by boiling in water for 10 min in vegetables of the *Brassica rapa* type (Watanabe et al. 1988). Sharma et al. (1994) found that plain washing dislodged 20–52% mancozeb residues while washing coupled with cooking led to 53–79% decontamination on cabbage, knoll-khol, tomato, okra and brinjal.

According to Nagesh and Verma (1997) decontamination through different processes showed that the residues in cabbage were reduced to some extent by various home processing methods like washing and cooking. Cooking did not help much in reducing the residue below the MRLs of 0.25 and 0.05 ppm for quinalphos and chlorpyriphos respectively. In the study of dissipation of quinalphos and the effect of processing in cauliflower Lalitha et al. (1998) concluded that residues were considerably reduced from curds roasted in oil after boiling (36.3–68.6%) followed by simple boiling (10.1–60.0%) compared to the curds soaked in 2% salt water (14.7–43.2%) and soaked in tap water (13.0–40.5%).

The study conducted by Schattenberg et al. (1996) indicated that residue levels in most commodities were substantially reduced after typical household preparation. Washing and cooking treatments resulted in considerable reduction of lindane residues in brinjal and okra (Patel et al. 2001). Kadian et al. (2001) reported that cypermethrin residues declined in tomato, okra, bottle gourd and ridge gourd after all processing steps i.e. about 5–14% by washing, 6–26% by blanching, 6–19% by washing in brine solution and 15–33% by cooking. Kang and Lee (2005) brined Chinese cabbage samples in 8% brine for 4 h and boiled in water for 20 min, whereas spinach samples were blanched by dipping in water for 2 min and cooked in the same way as cabbage. Brining had little impact on pesticide levels in cabbage apart from those of diazinon and dichlorvos, decreased by about 20%. Residues undergoing greatest concentration decreased during cooking were diazinon and dichlorvos (80–90%), whereas levels of cypermethrin and deltamethrin and fenvalerate slightly increased. During blanching of spinach 72% decrease in dichlorvos concentration took place whereas decreases in levels of other pesticides ranged from 0–17%. During spinach cooking, decreases in pesticide concentration ranged from 0% for fenvalerate to 81% for dichlorvos. Lee and Jung (2009) observed that removal rates of the tested pesticides in hot pepper leaves by washing ranged from 27 to 90%. The blanching step increased their removal rates by 10–25%. Maximum reduction of fenazaquin residues in okra fruits (60–61%) was observed by washing+ boiling followed by boiling/cooking (38–40%) and then by washing (31–32%) (Duhan et al. 2010)

Peeling-off the fruit skin was reported to dislodge the residues to varying degree depending on constitution of the fruit, chemical nature of the pesticide and environmental conditions (Nath et al. 1975; Awasthi 1986). Awasthi (1993) found that washing reduced levels of dimethoate and fenthion on mangoes to 66–68% and levels of fenvalerate and cypermethrin to 21–27%. Peeling removed 100% of the residues in all cases.

Lee and Lee (1997) revealed that 45% of organophosphorous pesticide (OP) residues were eliminated when foods were washed in water, 56% with detergent washing, 91% with peeling, 51% with blanching-boiling and 90% in milling and processing. Peeling of potatoes caused a substantial decrease in chlorpropham residues (Singh et al. 2000). Soliman (2001) found that

malathion, lindane, HCB and p,p-DDD were present in highest levels in raw potatoes while chips had the lowest levels. Potato skins contained higher amounts of pesticides residues than potato pulp and peeling markedly decreased pesticide content of potatoes. Washing with tap water or aqueous solutions of acetic acid and/or NaCl, and blanching or frying of potatoes also removed most residues.

Washing and cooking were not found to be very effective in lowering residues in brinjals. However, reduction of residues was more due to cooking than simple washing (25–33%). In tomato both the processes reduced the residues almost to the same extent of 11–30% (Gill et al. (2001). The washing processing factor were 0.9± 0.3 for pyridaben, 1.1±0.3 for pyrifenox and 1.2±0.5 for tralomethrin, whereas the peeling processing factors were 0.3±0.2 for pyridaben and 0.0±0.0 for both pyrifenox and tralomethrin. (Boulaid et al. (2005).

Steeping in water followed by dehulling reduced residues of deltamethrin in chick peas (*Cicer arietinum L.*) by around 70–73%; however, this method was not sufficient to reduce the concentration to the prescribed maximum residue level of 0.1 ppm. On soaking, the transfer ratios (%, total pesticide residue amount in product/that in soybean) of soaked soybean were >60% for most of the pesticides investigated. The transfer ratio of soymilk ranged from 37 to 92%, and that of tofu ranged from 7 to 63% (Saka et al. 2008b).Washing and steaming removed 40 to 60% residues remaining on chick peas after storage (Lal and Dikshit 2000). Reddy et al. (2001) observed that washing with water followed by steam cooking removed triazophos to an extent of 64–88% in brinjals. Lindane residues were reduced to an extent of 42–56%. Radwan et al. (2005) reported that approximately 100% of profenofos residues were removed from aubergines and peppers by blanching and frying.

II. Thermal Treatment

Foods are invariably subjected to heat treatment during preparation and preservation. The heat treatment is given in many ways including pasteurization, boiling, cooking etc. depending upon the nature of food and aim of processing. The loss of pesticide residue during heat processing may be due to evaporation, co-distillation., thermal degradation which vary with the chemical nature of the individual pesticide (Sharma et al. 2005). A number of reports are available in literature on the effect of thermal treatment on

pesticides in foods are being summarized below under different sub-heads.

a) Pasteurization: Changes in the amount of pesticide residues in buffalo's milk on heat treatments were investigated by Abd-Rabo et al. (1989). Fresh buffalo milk was spiked with 40 ppm each of p'p'-DDT, p,p'-DDE or p,p'-DDD, 50 ppm carbaryl or 50 ppm fenvalerate pyrethroids. The increase in DDD after pasteurization was observed which was attributed to decomposition of DDT and DDE to DDD during heat processing. Pietrino (1991) reported that pesticide residues remained unaffected by pasteurization, sterilization and by storage under refrigerated conditions or at ambient temperature. Losses of p,p'-DDE, o,p'-DDD (TDE), p'p'-DDD and p'p'-DDT, respectively, were 15.59, 58.80, 23.17 and 24.28% in milk pasteurized at 65 °C for 30 min (Jordral et al. 1995). Residues of p,p'-DDT; p,p' DDE, diazinon, malathion and chlorpyriphos, respectively were reduced by 29.79, 21.11, 70.54, 51.94 and 44.68% by pasteurization (62.8 °C for 0.5 h). However, concentrations of p,p'-DDD increased and those of lindane and dieldrin were unaltered by the heat treatment used (El-Hoshy 1997).

Abou-Arab (1999) recorded that pasteurization reduced HCH content by 65–73%. They suggested that consumption of heat-treated milk and dairy products may be safer than consumption of raw milk in terms of HCH residue intake. Pasteurization had little effect on reduction (8–10%) of residues of Ronilan (vinclocolin) and Dursban 4E (chlorpyriphos) in peach puree and nectar (Marudov et al. 1999).

b) Cooking/boiling/Steaming: Nath et al. (1975) indicated that after 10 min open and steam cooking of okra, 86.82 and 75.97% malathion was decreased. In Chinese cabbage, boiled for 30 min, decomposition of diazenon, dieldrin, dimethoate, fenitrothion and chlorothalonil ranged from 72 to 99% (Wen et al. 1985). According to Nagesh and Verma (1997) cooking of cabbage did not reduce the residues below the MRLs of 0.25 and 0.05 ppm for quinolphos and chlorpyriphos respectively from a level of 500 ppm of each.

Nagayama (1996) observed that residual organophosphorous pesticides in green tea leaves and in crops of spinach, strawberries, oranges and grapefruit were decreased on leaching or cooking, respectively. Some residual pesticides passed into cooking water from the plant materials according to their water solubility. Incidence and stability of pesticide residues in some vegetable and fruits as affected by food

processing was studied by El-Nabarawy et al. (2002). Processing such as cooking caused marked reduction in insecticides, resulting in complete removal of insecticide residues in some cases.

The residue of organophosphorous pesticides in strawberries after cooking decreased with preparation process (Nagayama 1996). Same results were observed during marmalade preparation from oranges. However, marmalade prepared from grapefruit showed that the residual level in the grapefruit marmalade was 42%, being slightly different from that in orange marmalade. In case of candied peel made from orange zest after cooking, to be used as confectionary material, the pesticide residue decreased with cooking process. Pectin is easily transuded from foodstuffs by cooking and appeared to play a role in reducing the amount of pesticide extracted by organic solvents with heating (Nagayama 1997).

Clower et al. 1985 indicated that the level of EDB residue in medium or long grain polished rice decreased dramatically when cooked. Raw rice containing from about 100 to 1600 ppb EDB contained from about 5 to 50 ppb EDB after cooking. The level of EDB in cooked rice appeared to be related to subtle differences in cooking techniques. Lee et al. (1991) reported that washing of rice grains spiked with chlorpyriphos (500 ppb) removed approximately 60% of residues. However, it was found to be 30% of original quantity in cooked rice. Nakamura et al. (1993) reported that organophosphorous pesticides could be mostly removed by washing with water followed by steaming. The residues of applied dichlorvos, chlorpyriphos methyl, malathion and fenitrothion were 0 to 5.6% in boiled rice and 0% in rice noodles.

Residues of organophosphorous pesticides in wheat and their levels during cooking were investigated by Hori et al. (1992). Bread, spaghetti, Chinese noodles, Japanese noodles, sponge cake, cookies and coating were prepared using wheat flour. Chlorpyriphos methyl and malathion residues decreased after processing, but were still detectable in foods. Arita (1994) found that with increase of boiling time from 0 to 15 min, pesticide residues in noodles gradually decreased, 40–70% for Udon, 70–80% for buckwheat noodles and 4–5% for Chinese noodles, respectively, of the original level in raw noodles. The total pesticide content in water+noodles was considerably lower after boiling compared with that in the original raw noodles.

Stability of deltamethrin on pulses during processing was investigated by Dikshit (2002). Imme-diately after treatment of pulses with deltamethrin, washing and steaming were done. Steaming removed 34–48% residues. Maximum concentration of the insecticide was found on seed coat (84–90%). Dichlorvos was easily removed in every stage of Tofu production (Miyahara and Saito 1994). Since dichlorvos is an unstable pesticide and very volatile, it may have been vaporized by heat in the cooking stages.

Effects of hard boiling and scrambling on p,p'-DDT, heptachlor and chlorpyriphos residues in whole eggs and egg yolks as studied by Hsu et al. (1995) showed that cooking by scrambling did not affect DDT and heptachlor concentration in eggs, but decreased chlorpyriphos content in egg yolks by 38%. Hard boiling shell eggs degraded all the chlorpyriphos and an average of 16% of DDT and its metabolites. Heptachlor and heptachlor epoxide were not significantly affected by hard boiling.

Among the 303 samples of spices and medicinal plants, Abou-arab and Abou-Donia (2001) showed that on immersion of the medicinal plants in hot water, transfer of the pesticide residues to the watery extract depended upon the type of plant, the nature of the pesticide and the applied treatment.

c) Frying: Lewis et al. (1996) found that residues of all thiabendazole, tecnazene and chlorpropham pesticides were significantly reduced to <2 and <10% of the maximum theoretical residue carry-through level for potato crisps and jacket potato crisps, respectively. Lewis et al. (1998) further demonstrated that approximately 56% of the maleic hydrazide residue in a potato could be carried through into the potato crisps, irrespective of which type of crisp was being manufactured. They also showed that, due to the concentration effect caused by the loss of moisture during crisp manufacture, levels of maleic hydrazide residues in crisps were approximately 2 times those measured in the original potatoes.

III. Products manufacture

A set of processing techniques are used to convert raw materials into a variety of products for consumption. The amount of residue in the final product may be reduced or enhanced depending upon a set of parameters employed and length of processing. In addition, micro-organisms/fermentation, if employed, also contribute to residual degradation of pesticides (Sharma et al. 2005)

a) Drying and dehydration: The drying process could cause an appreciable decline in pesticide

residues mainly due to evaporation, degradation and codistillation. Nath et al. (1975) observed that dehydration of treated okra resulted in the removal of malathion deposit by 91.86 and 91.79% and a residue of 0.21 and 0.38 ppm could be detected on dehydrated product for lower and higher application dosages, respectively. In case of endosulfan, the dehydration resulted in 57.59 and 57.35% removal of the initial deposit as the amount detected was 2.96 and 6.44 ppm for lower and higher treatment dosages respectively. It was concluded that dehydration brought down the residue below prescribed tolerance limit in case of malathion, while the endosulfan residue after dehydration was more than the tolerance of 2 ppm. Lee (2001) artificially contaminated Red pepper fruits with chlorpyriphos and fenitrothion after harvest, at levels of 4 and 10 ppm, respectively. Sun or hot air-drying eliminated a 20–30% of residues. Industrial dehydration reduced phosalone levels in the apples by greater than 80% regardless of initial dose of (Mergnat et al. 1996).

The different drying methods have different effect on different pesticides In the production of raisin, the sun drying process caused a 4 times concentration of residue level while oven drying which was preceded by washing lead to decrease in iprodione and procymidone (Cabras et al. 1998a). Cabras et al. (1998b) noticed during prune processing that iprodione residue at harvest time was 0.68 ppm and became approximately half after the drying process, while phosalone residue level was 3 times higher after drying. This could be attributed to the concentration factor of the fruit. In the drying process, the residue decrease due to washing was compensated by the residue increase due to drying; therefore, the residue level did not change. They further investigated that some of the residues were not reduced during the fruit washing stage, but the drying stage led to complete elimination of remaining residues. Drying process caused a decrease in iprodione and bitertanol, while it did not affect phosalone. The sunlight- and oven-drying processes caused the fruit to concentrate by a factor of approximately 6 times. Nevertheless, the pesticide residues present in the dried fruit were lower than in the fresh fruit. The residue decreases were higher in the sunlight process than in the oven process. In the former, on average, the residues on the dried fruits were about half those on the fresh fruits, whereas in the latter they were about equal (Cabras et al. 1998c, d).

Cabras and Angioni (2000) investigated the pesticide residues in grapes and their processing products. The residue levels of benalaxyl, phosalone, metalaxyl, and procymidone on sun-dried grapes equaled those on the fresh grapes whereas they were 1.6 times higher for iprodione and one-third and one-fifth lower for vinclozolin and dimethoate, respectively. In the oven-drying process, benalaxyl, metalaxyl and vinclozolin showed the same residue value in the fresh and dried fruit, whereas iprodione and procymidone residues were lower in raisins than in the fresh fruit.

b) *Nectar/puree:* Apples treated with tetrachlorvinphos, chlorpyriphos, cyhalothrin or hexythiazox were processed into puree by washing, boiling, pressing, heating to 85–90 °C and sterilization (Neicheva et al. 1993). Pesticide residue levels decreased during processing by approximately 43–91%, depending on pesticide. Marudov et al. (1996) reported that processing of cherry into puree reduced Ronilan and Dursban concentration to 16.6–30.7% and 22.8–42.4%, respectively of the initial concentration depending upon washing treatment used (water/detergent). Processing into juice resulted in concentration of <1.0%. They further (1999) reported that processing resulted in a decrease in Ronilan residues of 80.9 and 87.1% and in Dursban residues of 74.5 and 81.6%, for puree and nectar, respectively.

c) *Grain milling:* Benoualid and Gueriviere (1972) studied the pesticide residues in wheat and its milling products. Small–scale milling tests showed that as long as the treatment with pesticides was not excessive and the grain was properly cleaned at the mills, the lindane and malathion contents of wheat milling fractions were low. Abu–Elamayem et al. (1979) added leptophos added at a concentration of 100 ppm to wheat grains before processing and found that only trace amounts (0.99 ppm) was found in the flour and a level of 11.95 ppm was found in the bran.

Alnaji and Kadoum (1979) observed higher residues of methyl phoxim were found in the bran and shorts and very small amounts were in the flour. The losses during milling were 8–10%. Bengston et al. (1983) reported that residues of deltamethrin, fenvalerate, permethrin and phenothrin were highly persistent on stored wheat. During milling, residues accumulated in the bran fractions and were reduced in white flour. Joia et al. (1985) observed that highest amounts of cypermethrin and fenvalerate were present in bran and least in endosperm. Both the insecticides

degraded in treated wheat at slow rates. Reduction of residues in flour was low and 79–84% of cypermethrin and 87–88% of fenvalerate were present in flour. Alnaji (1987) observed highest residue concentration in seed coat (bran and shorts) with small amount in flour. The losses in residues during milling were 86.15%.

The scouring of wheat samples treated with pirimiphos-methyl removed 17–28% more pesticide residues than did conventional cleaning (Brown et al. 1991). Puccetti et al. (1993) observed that the half life of the residues in whole wheat was approximately 6 months. High quantities of pirimiphos-methyl were found in flour (up to 550 ppb) and bread (up to 138 ppb).

Cogburn et al. (1990) reported that parboiling reduced malathion and chlorpyriphos methyl residues on rough rice and hulls but tended to increase in the other fractions. Small amounts survived all the processing including cooking. Rice bran contained higher amount of residues followed by straw, husk and grains. Boiling rice grains did not result in the loss of any HCH residues (Battu et al. 1989). Saka et al. (2008a) observed that in the polishing process, the transfer ratio (%, total pesticide residue amount in product/that in brown rice) of rice bran ranged from 40 to 106%, and that of polished rice ranged from 9 to 65% in pre-harvest samples.

d) Baking: Losses of methylphoxim residues during chlorine bleaching at 2 oz/100 lb of flour were 41.2–55.5% compared with 32.3–45.6% of malathion residues (Alnaji et al. 1979). Bengston et al. (1980) reported that during processing of wheat to white bread, residues were reduced by 98% for carbaryl, >44% for (1R)-phenothrin, 98% for fenitrothion and 85% for pirimiphos-methyl. During baking, methylphoxim and malathion residues were found to be degraded from 79.1–100% and from 80.1–100%, respectively (Alnaji and Kadoum 1981). Bolletti et al. (1996) observed that residue concentrations of chlorpyriphos methyl, pirimifos and malathion were considerably lower in the breads than in the flours. The cookie processing also significantly reduced the concentrations of malathion and chlorpyrifos-methyl (Uygun et al. 2009).

e) Malting and brewing: Miyake et al. 2002 observed that phenthoate and fenitrothion were persistent during storage of barley. A concentration greater than or equal to 80% of the initial remained after 2 months of storage. Losses of other pesticides during storage were 25–85%. Concen-

tration of metabolites of triademefon and triflumizole increased slightly during storage. Carryover during steeping ranged from 3 to 50% that was related to the log P (partition coefficient between n-octanol and water) value of pesticides; residues of pesticides with log P value<2 would probably be eliminated during steeping. Concentration of residues of fenitrothion, phenthoate and triflumizole and its metabolites were considerably reduced by kilning; residues of the other pesticides were little affected by germination or kilning. It was concluded that steeping was the main factor reducing pesticide residue concentration on malt; however, residues of pesticides with log P>2 may persist in malt.

The fate of herbicides, insecticides and fungicides during processing of barley into malt as studied by Navarro et al. (2007) showed that pesticides concentration declined along the process, although in different proportions. The carryover of residues after steeping was 45–85%. A good correlation (r>0.92) was observed between percentages removed after steeping and the POW values of pesticides. The amount remaining after malting ranged from 13 to 51% for fenitrothion and nuarimol, respectively. Steeping was found to be the most important stage in the removal of pesticide residues (52%) followed by germination (25%) and kilning (drying and curing, 23%). During malt storage (3 months) the fall in pesticide residues was not significant. Applying the standard 1st-order kinetics equation (r>0.95), the half-lives obtained for pesticides during malt storage varied from 244 to 1,533 days for myclobutanil and nuarimol, respectively. In beer, the production process lead to a reduction of residues present in grain and hops used in brewing. The clarifying substances did not show any efficacy in pesticide residue reduction (Farris et al. 1992).

f) Canning of fruits and vegetables: Hasegawa et al. (1991) observed that 0% captan, 39.5% of iprodione and 51.11–150.2% of pyrethrin components were present in cherries after canning with syrup. Lentza-Rizos (1995) reported that mean concentration iprodione in field-treated peach fruits was 1.23 ppm, while in washed fruits for canning it fell to 0.61 ppm. Cold storage for up to 20 days did not affect residues. Chemical peeling removed 82.5–95% of the residues. In canned peaches, stored for 8 months, the concentration of iprodione was low of 0.01–0.10 ppm.

In most cases, operations leading to canning resulted in a gradual decrease in residue levels in

the finished products; the washing, blanching, peeling and cooking stages were particularly effective. A combination of washing and blanching led to >50% reduction in pesticide residue levels in all samples except peaches. Total amounts of pesticides removed by all of the combined canning operations ranged from 90 to 100% in most products. Pepper retained 61% of chlorpyriphos, but these residues disappeared during storage of cans for 3 months. Acephate showed a surprising tenacity in peaches, as 11% of the original residues were still present in cans stored for 2 years (Chavarri et al. 2005). There was significant difference ($P=0.05$) in residue concentrations of the parent compounds between the whole peach, the half peach with peel, and the half peach without the peel, during commercial canning process. However, there were no significant difference in residue concentrations found between the half peach without the peel and the canned peach samples. The dissipation rate of metabolite residues varied from compound to compound (Lennon et al. 2006). Ethylenebisdithiocarbamates were completely removed from tomatoes and spinach by washing followed by hot water blanching. Samples analyzed at each stage of industrial processing of tomato showed a progressive reduction in the contents of the 3 insecticides, only insignificant quantities of dimethoate remaining after the pasteurization stage (Severini et al. 2003). *f) Milk products:* Pietrino (1991) reported that spray and freeze drying reduce the concentration of pesticides to some degree. Low levels of pesticide residues had no significant effect on the processing of milk into cultured milk or cheese. This was related to the fact that the microflora used in cheese making may degrade pesticide residues during cheese ripening. The transfer factor of pesticides from milk during processing of milk products was found to be 50% lower in ewe and goat milk than in cow milk. Processing of milk into yoghurt reduced its natural pesticide residue content (Ali et al. 1993). When raw buffalo milk was spiked with DDT+DDD+DDE or carbamates (carbaryl) or fenvelerate and used to make butter, cheese and cream, more DDE than DDT or DDD was retained in the products. Retention of carbaryl was in the order cheese > cream > butter and was greater than that of fenvalerate, which showed the opposite trend. Zidan et al. (1994) monitored contaminants during the manufacture of some conventional dairy products. Initial levels of insecticides, 3 ppm, HCH, 1.0 ppm lindane, and

2.0 ppm p,p'-DDT respectively in milk were reduced by 0, 5.3 and 0.1% in cream; 20.5, 23.3 and 24.6% in butter; 80.4, 92.7 and 77.4% in s*amna*; 3.5, 3.8 and 2.8% in cheese and 77.3, 2.0 and 9.0% in yogurt. Misra et al. (1996) studied the degradation of fenvalerate (pyrethroid) in milk by lactic acid bacteria. Sterile whole milk samples containing 1 ppm of fenvalerate were inoculated with strains of *Lactococcus lactis* subsp. *Lactis, Lactococcus lactis* subsp. *Diacetylactis-DRC1, Steptococcus salivarius, S. thermophilus–H* and *Lactobacillus delbrueckii* subsp. *Bulgaricus-W* and incubated at their optimum temperature of growth for 120 h. A gradual decrease in the level of fenvalerate was observed with the increase in incubation period. The relative reduction after 24 h incubation was 24% which increased to more than 80% after 120 h. *L. lactis* subsp. *Lactis* degraded 98.6% of fenvalerate in whole milk at 32°C after 120 h.

Buffaloes' and cows' milks were used with and without addition of 1.75 ppm malathion to make *Domiati* cheese, ghee and butter. Approximately 92% of malathion added to cows' and buffaloes' milk was retained in butter, but retention in ghee was only 12.5 and 13.9%, respectively. Fresh cheese retained 80.0 and 72.2% of malathion added to buffaloes' and cows' milk, respectively, but these values decreased to 31.5 and 22.3%, respectively, after 90 days of pickling (Dabiza et al. 1999).

Madan and Kathpal (2001) studied the decontamination of pesticide residues in milk by different kitchen processes viz, boiling, *malai* removal, curd, *lassi,* butter and *ghee* formation and showed that residues of HCH isomers decreased by 11.54–26.78% due to boiling alone and 35.86–50.88% due to boiling followed by *malai* removal. Reduction in DDT residues was 15.58–35.09% due to boiling and 25.32–62.04% due to boiling followed by *malai* removal. Curd and *lassi* formation did not show further decrease in the levels of residues. The amounts of these insecticides decreased due to boiling as well as *malai* removal, at the same time, their concentration increased to several fold in *malai,* butter and ghee due to fat soluble nature of these insecticides. It was observed that *malai* contained about 3–5 times and butter and ghee contained about 11–13 times higher residues of HCH and DDT than unboiled milk. Butter obtained from the top of *lassi* also contained about 2–3 times higher residues than the unboiled milk.

Manufacture of yoghurt from spiked milk and refrigerated storage for 3 days caused a gradual reduction of HCH levels by 1.4–8.9% (Abou-Arab 1999). Manufacture of Ras Cheese from spiked milk removed 36.7% of HCH and its metabolites after a 6-month ripening period. Reduction of HCH was higher in Domiati Cheese when an acid-enzyme coagulation step was included than when an enzyme coagulation step was used (3.6 and 1.4%, respectively). Rajashekar et al. (2007) reported highest levels of pesticide residues in butter and lowest levels in *khoa*. Sterilized milk contained lower pesticide levels than pasteurized milk. The intensity of heat treatment influenced degradation of pesticides in milk products, which was highest for dicofol, followed by β-endosulfan and α-endosulfan.

g) Oil extraction and processing: The residues in olive oil showed great variability on processing (Farris et al. 1992). The most water soluble pesticides had lower level of residues (1/3 or 1/4) than those present in drupes, while other pesticides had analogous residues in olives and oil or higher residues in oil. Diazinon and methidathion were about 5 times higher in the oil than in the drupes, indicating that these residues were completely transferred from fruit into oil. Miyahara and Saito (1993) studied the pesticide removal efficiencies of soybean oil refining processes. Dichlorvos concentration in dry soybeans and the oils was decreased markedly at each stage of processing and malathion concentration was reduced at the alkali-refining stage. The 4 pesticides namely dichlorvos, malathion, chlorpyriphos and captan were removed or eliminated completely by deodorizing. Fukazawa et al. (1999) added five organophosphorous pesticides (dichlorvos, parathion-methyl, malathion, chlorpyriphos and chlorfenvinphos) to crude soybean oil and the oil was refined by degumming, alkali-refining, bleaching and deodorization. Levels of pesticides in crude oil decreased slightly following degumming. After alkali treatment, the dichlorvos level in degummed oil decreased significantly, whereas levels of the other pesticides remained at greater than or equal to 80% of their original values. Bleaching reduced dichlorvos, chlorfenvinphos, malathion and chlorpyriphos levels by approximately 70, 60, 30 and 5%, respectively; parathion-methyl levels were reduced significantly using an adsorbent containing activated carbon. All pesticides were removed completely by deodorization at 260 °C. It was concluded that these refining procedures were

useful for removal of pesticide residues from crude soybean oil.

Ruiz-Mendez et al. (2005) investigated the effects of various processing stages of olive oil refining on pesticide levels. Oil samples were spiked with endosulfan (α-endosulfan, β-endosulfan and endosulfan sulfate), simazine, oxifluorfen and diflufenican and subjected to physical refining (bleaching and deodorization). Bleaching using earths was effective only for the elimination of simazine. For removal of the other pesticides tested, a physical refining treatment was required at 240 °C in the deodorizing stage for a period of 1 to 3 h. Fukazawa et al. (2007) studied the behavior of N-methylcarbamate pesticides during refinement processing of spiked (5 ppm) soybean oil. By degumming, aldicarb, aldicarb sulfoxide, aldicarb sulfone, oxamyl, thiodicarb, carbosulfan and benfuracarb contents decreased by up to 70% with H_3PO_4 treatment and a decreases of <26% were noted for the other pesticides. With hot water treatment, the decrease in the content of any of the pesticides was <52%. With alkali refining, decrease varied with the pesticide in the range 8–100%. NaOH treatment was found effective in removing pesticides. With bleaching, aldicarb, aldicarb sulfoxide, aldicarb sulfone, oxamyl, methomyl, thiodicarb, carbosulfan, benfuracarb, bendiocarb and furathiocarb contents were decreased by >80%, with activated clay containing activated charcoal. Carbaryl content was decreased markedly using this clay. With deodorization, 40% furathiocarb, 14% carbosulfan, 11% benfuracarb and 3% carbofuran could still be detected following deodorization at 260 °C. Degumming with H_3PO_4 and bleaching with activated clay caused conversion of carbosulfan and benfuracarb to carbofuran.

Trials conducted on samples of lemon, orange and green mandarin essential oils by by Tateo and Crivelli (1994) showed that cold deterpenation considerably reduced concentration of organophosphorous pesticide residues, commonly by a factor of 10–20.

j) Juice/concentrate preparation: Apple juice concentration (7 times) did not increase residues of sulphur, fenvalerate and chlorpyriphos (El-Hadidi et al. 1996). Azinphos-methyl, formetanate-HCl and myclobutanil showed higher residue levels in the juice concentrate although they were still below or close to their residue levels on or in whole fresh fruit. Influence of technological process on fenitrothion residues during production of concentrated

apple juice was reported by Lipowska et al. (1998). Approximately 50% of residue was removed on washing of fruit and >90% during pressing and filtration (even in non-washed apples). Fenitrothion concentration averaged 0.97 ppm in chopped fresh apples, 0.03 ppm in pressed juice and below detectable levels in filtered juice; pomace contained up to 3.08 ppm. Zabik et al. (2000) concluded that processing of apples was effective in reducing the levels of pesticides. Apples were processed into frozen apple slices, apple sauce, single-strength juice and juice concentrate. Producing single-strength apple juice reduced azinphos-methyl, chlorpyriphos, fenvalerate and methomyl residues by 97.6, 100, 97.8 and 78.1%, respectively. Production of apple sauce reduced all 4 compounds by greater than or equal to 95%. Processing of contaminated cherries for juices resulted in decrease in initial pesticide concentration of approx. 90 and 70% for Reldan and Rubigan, respectively (Hadjikinova et al. 2006).

The distribution of 9 pesticides between the juice and pulp of carrots and tomatoes during home culinary practices was investigated by Burchat et al. (1998). Tomato and carrot pulp contained a higher percentage of all pesticide residues, except for mancozeb in tomato juice. Although there was a difference in the relative distribution of the pesticides between the commodities with greater amounts present in the pulp of tomatoes, the pesticides followed a similar trend in both. Pesticides with the highest water solubility were present to a greater extent in the juice. An exception was noted in the case of diazinon and parathion, which were present in higher amounts in the pulp than their water solubility suggested. The residue in the pulp ranged from 56.4 to 75.2% for carrots and 49.7 to 95.4% for tomatoes. Washing of the produce removed more residues from carrots than from tomatoes, but it did not affect the relative distribution of the residues. The behavior and fate of the chemical varied with the pesticide as well as the crop. Pesticide residues were greatly decreased in tomato juice under cold or hot break. A sharp decline in profenofos level was noted after treatment by pectinex ultra SP-L and benzyme M during tomato crushing (Romeh et al. 2009).

Lee and Chun (2003) investigated leaching ratio of pesticides during the juicing of artificially or naturally contaminated kale. Samples of washed kale and kale juice were analysed for chlorpyriphos, diazinon, dichlorvos, dimethoate, EPN, profenofos, cypermethrin, deltamethrin, endosulfan and fenval-

erate; leaching ratios of these pesticides into kale juice were 45.1, 41.1, 4.4, 25.1, 58.3, 51.5, 68.9, 59.9, 35.8 and 53.4%, respectively.

k) *Wine making*: Frank et al. (1990) observed a significant reduction in pesticide residues on fermentation of grapes into wine. During processing of grapes into wines, vinification with skins lead to a greater pesticide (86%) reduction than vinification without skins (50%) (Cabras et al. 1991). The reduction in pesticide concentration was more evident with red wine verses the white wine vinification process. Parathion level decreased quite rapidly, while metalaxl was the most persistent (Stozhoko et al. 2007).

Will et al. (1999) showed that must clarification, fermentation and racking were found to be important steps in reducing pesticide residues. Must clarification reduced the residues 40–50%. During winemaking a reduction of residues present in grapes occurred due to hydrolysis and absorption on suspended matter. Rate of reduction was significant in the must, especially when the suspended matter was separated from it. Use of clarifying substances, both in must and wine, did not lead to greater residue reduction than with natural settling of suspended matter, with the exception of BMC, which was almost completely eliminated by bentonite. Winemaking with maceration usually lead to greater pesticide reduction than winemaking without maceration (Farris et al. 1992).

Soleas and Goldberg (2000) conducted fermentation trials to assess the role of clarifying agents (bentonite or kieselsol, added at 0.25 or 0.5 g/L to the must or finished wine) on reduction of pesticide concentration in wines. The musts were spiked with 15 pesticides. When no clarifying agent was added, residues of 11 of the pesticides were present in finished wines at >40% of the initial concentration. Amount of the initial pesticide load removed by the clarifying agents varied between pesticides and treatments. Reductions of >90% were achieved in some cases. In general, kieselsol reduced pesticide concentration to a greater extent than bentonite and post-fermentation treatment was more effective than pre-fermentation treatment. The effect of red wine malolactic fermentation on the fate of seven fungicides and three insecticides was investigated (Ruediger et al. 2005). After malolactic fermentation using *Oenococcus oeni,* the concentrations of the active compounds chlorpyriphos and dicofol were the most significantly reduced, whereas the concentrations of chlorothalonil and procymidone diminished slightly only. Flori et al.

(2001) reported that pesticide decay constants (Kwm) during winemaking were in the range 0.195–1.887 and were much lower during storage indicating that decreases in residue concentration during storage were negligible.

m) *Sausage curing*: Bayarri et al. (1998) observed that curing did not exert any significant effect on hexachlorocyclohexane residue levels. However, *Micrococcus varians* was able to degrade and caused significant reductions ($P<0.05$ and $P<0.01$) of hexachlorobenzene (12.7%) and p, p'-DDE (17.7%).

IV. Postharvest handling

Pack-house operations are very important for preserving and improving the quality of food. These differ according to the type of food handled.

a) *Packing, cooling and brushing*: The packing house process reduced pesticide residue levels on fresh peaches to levels that were generally below detection limits (Taylor and Bush 2002). Carbaryl and captan residues from field packed fruits were 32.2 and 21.9 times, respectively, greater than levels present in peel of fruits processed in packing house. Carbaryl levels were not reduced by hydro cooling but post-harvest brushing reduced pesticide residues by up to 94% in peel. Across processing operations, hydrocooling, hydrocooling+brushing, and brushing alone removed 37, 62 and 53%, respectively, of encapsulated methyl parathion (parathion-methyl) residues from peel. Hydrocooling had the greatest impact (72.5% reduction) on phosmet removal from peel. After hydrocooling, levels of phosmet were 5.7 times greater following brushing in 50% of subsequent samples. It was suggested that periodic cleaning of brushes may be necessary to prevent later contamination of peel with pesticides. Propiconazole residue was removed most effectively (69%) by the brushing operation. Nearly 31% of propiconazole was removed in the hydro-cooler. The packing process before shipment to retail outlets was generally effective in removing pesticides that may be present on peel at the time of harvest. Tests on waxed and unwaxed apples contaminated with a cocktail of pesticides showed that treatments including wiping with paper towels were more effective than other procedures for pesticide removal (Michaels et al. 2003).

b) *Storage*: Lemons, oranges and grapefruits were dipped in a solution of diphenyl, o-phenyl phenol, 2,4-dichlorophenoxyacetic acid (2,4-D), imazalil, thiabendazole, benomyl and sec-butylamine or fumigated with methyl bromide and stored at 14 °C. After 91 days, half lives of pesticides ranged from 36 days to >6 months. Diphenyl and imazalil were extremely stable, while bromide gradually increased in citrus fruits. After 8 week storage, marmalade from such lemons contained 95, 41, 42 and 22% of diphenyl, o-phenyl phenol, imazalil and bromide, respectively, while no 2,4-D, thiabendazole, benomyl or sec-butylamine was detected (Tsumura et al. 1992).

Following 12 months of storage at 20 ± 5 or 4 ± 1 °C, pesticide residues in apple puree decreased by 36–85% and 20–70%, respectively (Neicheva et al. 1993). Captan residues present in strawberries stored at 5 °C for 3 and 7 days were significantly reduced to values of 27–99% lower than those observed prior to storage. No reduction in levels of chlorothalonil residues was observed. The effects of storage at 5 °C on pesticide residue reduction depend on the chemical structure of the contaminating pesticide (Oliveira and Toledo 1996).

Singh et al. 2000 treated potatoes with a 29% (1:6) mixture of propham and chlorpropham at a concentration of 60 ml/t prior to storage at 12 °C. At 2 days after treatment, propham was not detected in samples, while chlorpropham was detected in peel, whole tubers and pulp at concentration of 4.69, 0.5 and 0.085 mμg/g, respectively; these concentrations fell to 2.61, 0.15 and 0.032 mμg/g at 21 days after treatment. Sakaliene et al. (2009) observed that chlorpropham concentratiom on washed and unwashed tubers decreased from approx. 15 ppm after storage for 28 days to approx. 9 ppm after 85 days. Peel and cooking water concentration also decreased similarly.

Gill et al. (2001) sprayed brinjals and tomatoes with alphamethrin and stored at ambient (40 °C) and refrigerated conditions (5 °C). Dissipation of alpha-methrin was observed faster at room temperature as compared to cold conditions in both the vegetables. Patel et al. (2001) studied the dissipation of lindane (HCH) in brinjals and okra fruits. Residues persisted for up to 20 and 5 days in brinjal and okra fruits, respectively. Relatively higher initial deposits of lindane (1.64 and 5.87 mμg/g for EC and powder formulations, respectively) were recorded on okra, with half-life values ranging from 0.9 to 1.0 days. In brinjal, lower initial lindane residues were recorded, which dissipated at a slower rate than in okra; half-life values ranged from 2.5 to 4.6 days. Mahajan et al. (2008) reported that CIPC (Isopropyl-N (3-chlorophenyl) carbamates) residue concentration

decreased drastically during storage of potato at 10±1 °C and residue level fell below MRL level (30 ppm) from initial concentration of above 50 ppm after 1 month of storage.

There were relatively high residue levels in bran and middlings after storage of spring wheat for 6 months and those of iodofenphops in flour were lower than the other compounds. Pirimiphos methyl degraded at a relatively lower rate than other compounds. The levels of insecticide residues recovered from fractions of grain with 16% moisture content were generally lower than those from fractions of grain with 12% moisture content (Mensah et al. 1979). Alnaji and Kadoum (1979) treated soft winter wheat with methyl phoxim and stored for periods up to 365 days. The residues degraded rapidly during the first month thereafter, it degraded gradually. The residues in pirimiphos-methyl treated wheat also followed the same pattern of degradation (Alnaji 1987).

In buckwheat sprayed with a solution of DDVP (dichlorvos), chlorpyriphos-methyl, malathion or fenitrothion, or fumigated with methyl bromide, and stored at 15 °C (Tsumura et al. 1994), it was found that half-lives of the pesticides ranged from 13 days (DDVP) to 124 days (fenitrothion). DDVP gave a biphasic semilogarithmic dissipation curve, in which the residue level decreased rapidly in the 1st phase, and then more slowly in the second phase. In the noodles prepared from stored wheat, 61% of the initial chlorpyriphos-methyl, 40% of malathion, 42% of fenitrothion and 21% of methyl bromide remained, while no DDVP was detected. Papadopoulou and Tomazou (1991) treated wheat seeds with permethrin and stored for 20 months under ambient conditions (17–32°C and 40–60% RH). The residues in whole ground grains ranged from 1.378±0.190 (day 1) to 0.247±0.026 ppm (day 427) in the wheat treated at 2 ppm a.i. permethrin and from 7.400±0.234 (days 1) to 1.294±0.017 ppm (day 427) in the wheat treated at 8 ppm a.i. permethrin. After 35 days of storage 75–80% permethrin residues were found in the bran portions of seeds subjected to each of the three treatments, while after 427 days of wheat storage, no detectable levels of permethrin residues were found in flour from treated wheat at the rate of 2 ppm a.i.

Effects of storage interval and milling procedures on dissipation of deltamethrin residues in post-harvest treated wheat were studied (Balinova et al. 2007). After 180 days at an application rate of 0.5 ppm, residue concentrations were between 0.03 and 0.2 ppm in the 3 flours. After 270 days

treatment at a rate of 4 ppm, the residues in wheat flour were in the range 0.4–1.5 ppm. The storage period of 8 months was generally not effective enough to reduce the residues of malathion, fenitrothion, chlorpyrifos methyl, and pirimiphos methyl in durum wheat (Uygun et al. 2009).

No significant loss of EDB was observed in portions of cooked rice which had been stored frozen for about 2 weeks (Clower et al. 1985). Following deltamethrin application at concentration of 2 and 3 ppm, chick peas (Cicer arietinum L.) were stored in jute bags under ambient conditions (temp. 4.5–37.5 °C, RH 40–63%) for 6 months. Residues recovered from samples at the end of storage were 0.43 and 0.63 ppm at 2 and 3 ppm initial treatment dose, respectively; this concentration corresponded to cumulative reductions of 77.37 and 78.42%. Washing and steaming removed between 40 and 60% of residues remaining on chick peas after storage (Lal and Dikshit 2000). The persistence of cypermethrin on black gram (Phaseolus mungo), chick peas (Cicer arietinum), cowpeas (Vigna synensis) and lentils (Lens culinaris) was studied under laboratory conditions (Dikshit 2001). Samples were treated with cypermethrin at 3 and 5 ppm and stored for 6 months. The high recovery of extractable residues of cypermethrin in all studied pulses revealed its high persistence (62–71% of the applied insecticide) after storage. Dissipation of cypermethrin was biphasic, with an initial rapid decline up to 3 months followed by a phase of slow and steady dissipation.

Conclusion

It is clear from the preceding review that pesticide residues remain in almost all the food commodities, as a result of preharvest or postharvest application. The location of pesticides in different parts of food varies with the nature of molecule and type of food commodity and environmental conditions. Pesticide can be degraded by photolysis, hydrolysis, oxidation and reduction, metabolism, temperature, and pH. The level of pesticide residues is affected by washing, preparatory steps, heating or cooking, processing during product manufacturing and post harvest handling and storage. The extent of reduction varies with nature of pesticide molecule, point of location, type of commodity, processing steps and product prepared. The washing of raw materials is the simplest way to reduce the pesticide residue in the final product. The more effective and convenient alternative could be washing with chlorine water or with dilute solutions of other chemicals depending upon food

commodity. Special precautions should be taken to dislodge the residues from raw materials to be used for preparation of concentrated and dehydrated products. Judicious and systematic approach be followed to adopt preharvest practices and postharvest treatments to minimize the residue levels in finished products. There is urgent need to monitor the pesticide residues to standardize the application doses. Equally important is to develop or find new pesticide molecules with high effectiveness and fast degrading capabilities. Pest management is one of the major inputs in agricultural production; therefore, this area needs great attention to economize the production, to provide safe foods and to lower the medical expenses for treatment of resulting ailments. Moreover, the alternative means for pest management should be explored. In the present scenario of globally competitive trade, all concerted efforts should be made to ensure food safety as it has a direct bearing on human health and for boosting the export of food commodities.

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