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Processing of Raw Agricultural Produce and Its Effect on Pesticide Residues: A Review

S.R. Padaliya, K.D. Parmar[1], Suchi Chawla[1]

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ABSTRACT

Due to ever increasing population and limited natural resources use of pesticides as a major input to increase agricultural productivity is indispensible today. However, their inadequate application may produce large quantities of residues in the environment leading to environmental contamination. These residues may leach in the soil, water and even remain in the crop residues and food products. From there they enter in human food chain posing serious health hazards. Various food processing techniques, at industrial and/or domestic level, have been found to reduce the contents of pesticide residues in most food materials. These techniques alone or in combination reduce the levels of pesticides significantly in the food items. In the present review effect of processes like washing, peeling, cooking, drying, milling and industrial techniques like, baking, dairy products, fermentation, malting etc are reviewed. The review of literature suggests there is a need to systematize and optimize these techniques so that these can be used to provide a safer food culture for humans.

Key words: Food processing, Pesticides, Residues, Raw agricultural produce.

With the increase in consumer awareness and food safety regulations the concerns about various residue contaminations in food and water are highlighted. To feed an ever increasing population it is essential that crop production shall increase. It has been suggested by Nishant and Upadhyay (2016) that application of insecticides was the major protective measure against the attacks by devastating pests. There are numerous insect pests which feed on crops leading to huge crop losses ranging from 10-30% in the developed and 40-75% in developing countries (Roy, 2002). Storage of crops after harvest may cause even greater losses, particularly in the tropics (FAO, 1985). To prevent these crop losses and to increase crop production, the use of pesticides has increased worldwide. Although the use of pesticides are beneficial for the protection of food, fiber, human health and comfort yet their excessive use/ misuse, their volatile nature, long-distance transports eventually results in serious health hazards for humans and severely impact local and global environment (Ecobichon, 2001). Only a small fraction of applied pesticides are directly involved in the pesticidal action. Most of it find their way as 'residue' in the environment into the terrestrial and aquatic food chains where they undergo concentration and exert potential, long term, adverse health effects on animals and humans (Winteringham, 1971). However the localization of the pesticide in the food crops depends on the type of pesticide, crop and/or interaction of both (Bajwa and Sandhu, 2014). Pesticide residues in fruits and vegetables are a major concern to consumers due to the health hazards imposed by them. Residues of pesticides have been found in both raw/fresh produce and processed food. However, food processing techniques have been found to significantly reduce the pesticide residues in fruits and vegetables in several studies (Chavarri *et al.*, 2005). The food processing techniques generally used in different studies focused on commercial as well as home processing including washing, blanching, peeling, pureeing, cooking, roasting, frying and boiling (Ayam *et al*, 2017; Bajwa and Sandhu, 2014). The effects of food processing on pesticide residues have been extensively reviewed by researchers such as Holland *et al.,* 1994; Kaushik *et al.* 2009; Bajwa and Sandhu, 2014. These authors established that processing of food lead to reduction in the levels of residues of pesticides in most of the food materials except fruit juices and vegetable oils where the food got concentrated.

Department of Entomology, B. A. College of Agriculture, Anand Agricultural University, Anand-388 110, Gujarat, India.
[1]AINP on Pesticide Residues, ICAR Unit-9, Anand Agricultural University, Anand-388 110, Gujarat, India.

Corresponding Author: Suchi Chawla, AINP on Pesticide Residues, ICAR Unit-9, Anand Agricultural University, Anand-388 110, Gujarat, India. Email: suchisood10@yahoo.co.uk

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Food processing techniques

Food processing is the transformation of agricultural products into food, or of one form of food into other forms. Food processing includes many forms of processing foods, from grinding grain to make raw flour to home cooking to complex industrial methods used to make convenience foods such as bread, pastas etc. Primary food processing turns agricultural products, such as raw wheat kernels or

livestock, into something that can eventually be eaten. Food processing dates back to the prehistoric ages when crude processing incorporated fermenting, sun drying, preserving with salt and various types of cooking (such as roasting, smoking, steaming and oven baking). However, food processing techniques have been revolutionized depending upon the needs of the modern working conditions leading to advent of ready to eat foods, frozen foods etc.

Common processing of vegetables and fruits

Vegetables are generally consumed after cooking. General procedure followed for before cooking involves washing, peeling, blanching and then cooking. Some vegetables are consumed as salad without cooking. Many studies have been done in the past to assess effect of these pre-cooking steps on residues of pesticides. Many researchers have also reviewed these effects. Farrow *et al*, 1969 were of the view that washing removes loose surface residues and major portions of polar compounds such as carbaryl whereas hot water blanching increases pesticide removal and may hydrolyze substantial fractions of non-persistent compounds. Peeling and juicing operations usually result in almost complete removal of chlorinated hydrocarbons. The processing of food commodities lead to reduction or removal of residues of insecticides and other pesticides those are present in them. These operations such as washing, peeling, blanching and cooking play a role in the reduction of residues (Elkins, 1989).

Andrade *et al.* (2015) in a study on tomato samples have shown that Diflubenzuron (Dimax 480 SC), azoxystrobin (Amistar 50 SP) and procymidone (Sumilex 500 WP), were removed by peeling. They have observed that washing with 10% of sodium bicarbonate solution is efficient for reducing dimethoate residues (treatment with formulation Perfekthion 400 SP), 10% vinegar solution for acetamirpid (Convence 20 SP) and procymidone (Sumilex 500 WP), bicarbonate and 10% vinegar solution for imidacloprid (Confidor 700 WG) and thiamethoxam (Actara 750 SP). Washing with water showed the lowest residue concentration for diflubenzuron (Dimax 480 SC) and washing with water and vinegar, did not differ significantly for azoxystrobin (Amistar 50 SP). However, fipronil (Regent 800 WG) residues were not removed from the tomato samples in their study. Chauhan *et al*, 2014 have reported that peeling vegetables or fruit reduced pyrethroid residues by 60-100%. Washing raw agricultural commodities with saline water and detergent was more effective (34-60%) in reducing residues than was simple washing under tap water (10-70%). Freezing is also effective in reducing residue levels and achieved reductions between 24% and 94%. Cooking of food products eliminated 75-98% of the pesticide residues present. When foods were cooked in oils, however, reductions in pesticide residues were less (45%). Soliman *et al*, 2001, have reported reduction of residues of organochlorines and organophosphates by 22.9-47.3% from potatoes. In contrast to Chauhan *et al*, 2014 who reported low reduction of residues on cooking in oil, Soliman *et al.*, 2001 have shown 30-35% reduction in the levels of organochlorines and 49-53% in organophosphates. Heshmati and Nazemi, 2018 have shown that in tomato samples levels of DDVP reduced by 30.7, 91.9, 70.7 and 88.9%, respectively after 15 min of washing with tap and ozonated water, a detergent solution and ultrasonic cleaner. In a study on tomato Kwon *et al.*, 2015 have proposed that non-systemic pesticide residues on tomato surfaces could be largely removed through washing and peeling. They have observed that chlorothalonil (Thalonil 75 WP), oxadixyl (oxadixylcopper 8 WP) and thiophanate-methyl (Thiophan 70 WP) levels reduced by 92, 52 and 84%, respectively after washing of tomato. Washing brinjal samples (treated with the dimethoate98.5% purity, methylparathion-98.5% purity, quinalphos-99% purity, endosulfan-97% purity and profenofos-92% purity by preparing 0.2% solution by diluting 2 ml of formulations in 1 liter of water) in 2% salt solution reduced residues of endosulfan, dimethoate, methyl parathion, quinalphos and profenophos by 78-91%. This reduction was almost 100% when this was followed by cooking (Vemuri *et al.*, 2015). Shakoori *et al* 2018 studied the effect of washing and washing followed by cooking on residues of 41 different pesticides in rice samples. They found that washing process removed the residues of 34 pesticides in the range between 12.0-88%, whereas azinphos-ethyl (organophosphate), carbofuran (carbamate), cinosulfuron (sulfonylurea), coumaphos (organothiophosphate), etrimfos (organophosphate), fluometuron (phenylurea) and primiphos-methyl (phosphorothioate) were not significantly removed by washing. The most reduction occurred in oxadiazon (88.1%) and propiconazole (87.1%). However, the least removal occurred in the levels of malathion (12%), carbaryl (24.1%) and phosphamidon (25.7%). Washing followed by cooking (Iranian cooking method Kateh which involved boiling and steaming) lead to 20.7-100% decrease in residues of pesticides. Oxydemeton-methyl residues were completely removed. Another study from Iran has shown 78%, 55% and 35% reduction in rice samples for carbaryl, propoxur and pirimicarb respectively on cooking (Shoeibi *et al.*, 2011). Krol *et al.*, 2000 have shown that out of twelve pesticides captan, chlorothalonil, iprodione, vinclozolin, endosulfan, permethrin, methoxychlor, malathion, diazinon, chlorpyrifos, bifenthrin and DDE levels of nine pesticides showed significant decrease on rinsing. Vinclozolin, bifenthrin and chlorpyrifos did not get reduced. They have also established that the rinsability of a pesticide is not correlated with its water solubility. On washing of apples along with continuous hand rubbing for 10-15 seconds, captan residues were found to 50% lower as compared to those samples where post-harvest washing has not taken place. The same study has also shown 98% reduction in captan residues when apple samples were washed and peeled (Rawn *et al.*, 2008). Initial levels of 0.51 ppm of deltamethrin were removed completely on washing and steaming of chickpea grains (Lal and Dikshit, 2001). The initial residues of 0.68 ppm of iprodione in prune were reduced to 0.30 ppm after washing with water for 5 min

followed by oven drying and rehydration (Cabras *et al.,* 1998a).

The juices and preparation of purees of vegetables and fruits have also been reported to reduce pesticide levels significantly. Chlorthalonil (Thalonil 75 WP) remained to 3.66%, 0.32% and no detection levels in peeled tomatoes, tomato juice and puree respectively (Kwon *et al.*, 2015). In the same study the levels of oxadixyl (oxadixylcopper 8 WP) and thiophanate methyl (Thiophan 70 WP) remained to concentration of 40% and 6.2% in peeled tomatoes, 54% and 8.7% in juice and 77% and 16.2% in puree respectively. Chauhan *et al.*, 2014 have reported that juicing of vegetables and fruits was nearly as effective in reducing pyrethroid residues by 70-100%.

Baking is the technique of prolonged cooking of food by dry heat normally in an oven. It is primarily used for the preparation of bread, cakes, pastries and pies, tarts and quiches. It is also used for the preparation of baked potatoes, baked apples, baked beans. In a study from Egypt to investigate the effect of baking on pesticide residues, Habiba *et al.* (1992) have shown that the oven baking lead to decrease in residues of profenofos (Selecron 72% EC) from 11.48 ppm in fresh potatoes to 0.22 and 0.19 ppm in microwave-baked and oven-baked potatoes, respectively. Sharma *et al.*, 2005 prepared the bread from wheat flour by spiking different concentrations (1, 2, 3 and 4 mg/kg) of six pesticides (endosulfan, hexaconazole, propiaconazole, malathion, chlorpyriphos and deltamethrin) belonging to different chemical families and have shown that both yeast mediated fermentation and high temperature baking of wheat flour for bread preparation has lead to reduction in residues of hexaconazole, chlorpyrifos, propiconazole, malathion, deltamethrin, endosulfan by 46%, 51%, 60%, 63% and 70% respectively.

Drying is the oldest method of preserving food. Drying leads to decrease in the moisture content and may lead to concentrated residues. However, drying has been reported to decrease in residue levels. Athanasopoulos *et al*, 2005 have reported losses of 64.2-71.9% of methamidofos (Tamaron 60 SL) on drying of grapes. Another study on grapes after treatment with pesticides [chlorpyrifos (98.5%) 45µL/kg, diazinon (99.0%) 50µL/kg, dimethoate (98.5%) 50µL/kg, methidathion (98.5%) 45µL/ kg standarts] through spraying onto surface of grapes has reported 73, 92, 82 and 39% decrease in residues of chlorpyrifos, diazinon, methidathion and dimethoate, respectively on sun drying. When these grapes samples were dried in oven >90% reduction was observed in the residues of these pesticides in grapes (Ozbey *et al.*, 2017). Effect of sun drying followed by grinding on the residues of pesticides in chilli showed that the residues of fipronil 5 SC (Sonic Flo), λ- cyhalothrin 5 EC (Reeva-5), ethion 40 EC + cypermethrin 5 EC (Nagata), fenpropathrin 30 EC (Meothrin), fenazaquin 10 EC (Magister) concentrated by a factor of 5.02 to 8.57 (Patil *et al.*, 2018). Freezing of food is a common method of food preservation which slows both food decay and most chemical reactions. Freezing of tomatoes containing 1 ppm of residues of hexachlorobenzene, lindane, p, p'-DDT, dimethoate, profenofos and pirimiphos-methyl showed reduction of residues by 5.28%, 7.02%, 5.74%, 28.5%, 26.6% and 26.2% after six days and 10.6%, 16.3%, 13.0%, 32.6%, 28.2% and 31.4% after 12 days of freezing respectively (Abou-Arab, 1999a).

Processing of cereals

Pesticide-free wheat was placed in a small-scale model of a commercial storage vessel was treated with malathion and milled (Uygun *et al.*, 2005). Milling was found to reduce the malathion residues in wheat by 95%. When this was followed by baking further 82% reduction was found in residues of malathion. A study from Ethiopia where 5 g teff flour for each household processing method (doughing and backing) was spiked with eight pesticides including metabolites (DDT and its metabolites, cypermethrin, deltamethrin, permethrin and chlorpyrifos ethyl) at the rate of with 40 µl of 100 mg/L of each pesticide, showed that doughing decreased the residues of permethrin, cypermethrin, deltamethrin, chlorpyrifos ethyl, p,p'-DDE, p,p'-DDD, o,p'-DDT and p,p'-DDT by 59.9-86.4% and baking by 63.2–90.2% of the initial levels. The study also showed that the reduction of pesticide residues by baking is significantly different from doughing (p-value < 0.0001). There is also a significant difference between non-fermented and fermented dough (p-value = 0.012) (Mekonen *et al.*, 2019).

Malolactic fermentation is a process in winemaking in which tart-tasting malic acid, naturally present in grape must, is converted to softer-tasting lactic acid. The effect of red wine malolactic fermentation on the fate of seven fungicides (carbendazim, chlorothalonil, fenarimol, metalaxyl, oxadixyl, procymidone and tridimenol) and three insecticides (carbaryl, chlorpyrifos, dicofol) showed that the concentrations of active compound of chlorpyrifos and dicofol were reduced significantly wehereas the concentrations of chlorothalonil and procymidone diminished only slightly (Ruediger *et al.*, 2005). Cabras *et al.* (1998b) studied the effect of wine making on various pesticides by spiking different concentrations of pesticides in grapes. The fate of five fungicide residues from vine to wine was studied. After wine making (15 days) fluazinam (Ohayo 39.5 %p/p), mepanipyrim (Kif 3535 50 WP) and tetraconazole (Concorde 4 EC) had negligible residues in all samples. This was due to fermentation in case of fluazinam and mepanipyrim and to removal during the formation of must in the case of tetraconazole. In a study on effect of processing on pesticide residues in raisin, Cabras *et al.*, 1998c have shown that different drying processes affect pesticide residues level differently in raisin however the drying process caused a fruit concentration factor of 4. While sunlight-drying was more effective for phosalone (Zolone 33.6 EC) and vinclozolin (Ronilan 50 WP), oven-drying was more effective for iprodione (Rovral 50 SP) and procymidone (Sumisclex 50 WP).

Malting of cereals involves three steps, namely, steeping, germination and kiln drying. In a study on malting of barley, where barley grains (100 g, n=3), once spiked with the pesticides (0.2-1 mg/kg) were subjected to the process according to the common malting operations (steeping; germination; and kilning) after evaporation of the spiking solvent (3 h), Navarro *et al* (2007) have shown that pesticides i.e., fenitrothion and nuarimol (>98% purity standards), decline in the process was differential at different stages. Steeping was the most important stage in the removal of pesticide residues (52%) followed by germination (25%) and kilning (drying and curing, 23%). There was no significant decrease in the pesticide content during 3 months of malt storage. Another study on barley after application of mepronil (Basitac WP), propiconazole (Tilt EC), triadimefon (Bayleton WP), triflumizole (Trifmine WP), ethiofencarb (Arylmate EC), phenthoate (Elsane EC) and fenitrothion (Sumithion EC) followed by malting showed that carryover of residues after steeping was 3–50% and the losses of hydrophilic pesticides were significant. Very small loss was observed after germination and kilning. More than 80% of residues of phenthoate and fenitrothion (organophosphorous insecticides) remained after two months of storage (Miyake *et al.*, 2002).

Processing of milk

Liska (1968) have shown that Spray drying of milk destroyed in excess of 80% of the lindane residues in raw milk while sterilization caused essentially no change in lindane residues. Heptachlor was destroyed more easily than heptachlor epoxide. Of the insecticides studied, rnethoxychlor was the most stable to heat treatments used in milk processing. Abou-Arab (1999b), have shown that heat treatment of milk such as pasteurization, boiling and sterilization has decreased the residues of Lindane and its metabolites by 65-85%. He also showed that the reduction in residues of lindane was more in Domiati cheese made by acid-enzyme coagulation rather than when made by enzyme coagulation alone. The storage of yogurt and cheese also showed reduction in residue content. Using natural and spiked milk samples (α, β, γ and δ HCH at 1 parts per million [ppm]) were processed Singh and Nelapati, 2017 have shown that the total HCH (hexachlorohexane) content of 0.5984 ppm in natural milk, reduced to 0.5374 ppm in pasteurized milk, 0.3737 ppm in boiled, 0.307 ppm in sterilized milk accounting 10.19%, 37.55% and 48.69%, respectively. The initial total HCH level of 3.505 ppm (spiked milk, spiked at 1 ppm) degraded to 2.878 ppm in pasteurized milk, 1.618 ppm in boiled milk and 1.221 ppm in sterilized milk accounting 17.88%, 66.85% and 65.16% reductions, respectively. In natural and spiked milk samples, the residue levels differed significantly ($p<0.01$) within treatments, whereas in spiked milk boiling and sterilization treatments did not differ significantly. Processing of milk by different methods such as sterilization, fermentation, pasteurization etc lead to significant decrease in the levels of dimethoate and malathion. Pasteurization leads to complete degradation of malathion. However, dimethoate decreased by 73.42% on pasteurization. A degradation of 75.72% and 95.99% was observed for dimethoate and malathion respectively on sterilization of milk. On fermentation dimethoate and malathion degraded by 86.5% and 97.17% respectively (Abd-Rabo *et al*, 2016). Pasteurization of milk lead to loss of DDT and its metabolites by 15.6 to 58.5 % (Jordral *et al.*, 1995). Two different studies on effect of pasteurization on tomato showed nearly total degradation of dimethoate and maneb (Severini *et al*, 2003; Kontou *et al.*, 2004). However, Marudov *et al.* (1999) observed very little effect of pasteurization on peach puree on chlorophyll degradation. Sterilization lead to 37.4-49.6% and 78.8-79.4% decrease in primiphos-methyl and chlorpyrifos-methyl levels in wheat (Dordevic and Durovic-Pejcev, 2015; Dordevic and Durovic-Pejcev, 2016).

Processing of herbs and tea

Decoction of different herbs and varieties of tea are consumed in different parts of the world. These decoctions have medicinal values also. Different infusion processes or brewing are followed to prepare these. Water is boiled at different temperatures. Different studies have reported affect of infusion on the levels of pesticide residues. Xiao *et al*, 2017 investigated affect of infusion on transfer of pyrethroids [fenpropathrin (99.2%), beta-cypermethrin (99.2%), lambda-cyhalothrin (99.2%) and fenvalerate (99.0%) standards] in honeysuckle, chrysanthemum, wolfberry and licorice at different water temperatures, tea/water ratios and infusion intervals/times. They sprayed the sample crops with pesticides and no formulations were used. They have reported that only 0-6.70% of pyrethroids were transferred under the different tea brewing conditions leading to over 90% reduction in pyrethroid content in infusion. They have also proposed that the pesticides with high water solubility and low octanol–water partition coefficients (log K_{ow}) exhibited high transfer rates. A study on levels of pesticide, after soil application of recommended (3.2 and 2.1, 4.3 and 4.3 kg had1 for metalaxyl-M, fludioxonil, cyantraniliprole and thiamethoxam, respectively) and double the recommended dose of mixed pesticide solutions twice with an interval of 7 days at the squaring stage of chrysanthemum, showed that 59.9%, 9.8%, 29.4%, 88.2% and 68.4% of metalaxyl-M (99.5% purity), fludioxonil (99.0% purity), cyantraniliprole (99.2% purity), thiamethoxam (98.2% purity) and clothianidin (99.0% purity), respectively got transferred to tea decoction on infusion (Jiang *et al.*, 2020). Kumar *et al.*, 2005 have reported transfer of 23.6 to 40% levels of propargite on tea infusion. The remaining 35.7-53.2% remained in tea leaves. These studies show that the residues of pesticide significantly leach into decoctions of herbs and tea on infusion.

CONCLUSION

Pesticide residues are influenced by post-harvest processing. These processing involves both home

processing techniques as well as industrial processing. Both these ways were found to equally helpful in decreasing pesticide residue contents in food commodities. Different studies have shown that the processing significantly reduces the pesticide levels in different commodities. Cooking alone can reduce the residues to non-detectable levels. With an increased awareness in the field of food safety, it can be proposed that using combination of different food processing techniques specifically cooking and high temperature processing like baking etc may offer a safer food consumption culture, decreasing the health hazards posed due to use of pesticide residues on different food. Although different studies have been conducted to evaluate the effect of processing on pesticide residues levels in food, there is a need to develop a systematic approach to understand the effect of combination of different processing techniques on levels of pesticide residues including their metabolites and to optimize these techniques so as to provide a safer food for human beings.

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