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Effects of Different Household Treatment Methods on Minimizing Pesticide Residue Levels in Apple and Strawberry Fruits

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Abstract

Consumption of fruits with detectable pesticides residues is rising various human health concerns including endocrine disruption, cancer and Parkinson's disease. The purpose of this study was to assess the effectiveness of different household processes in reducing pesticide residues from two different types of fruits: apple and strawberry. Samples were spiked with known concentrations of commonly used eight fungicides and insecticides including vinclozoline, metalaxyl, methiocarb, cyprodinil, thiabendazole, myclobutanil, bifenthrin, and fenpropathrin. In apple fruit samples, the effectiveness of soaking in tap water, and soaking in Zamzam water (alkaline water) were tested in reducing eight pesticides. For the strawberry samples, three household processes including soaking in tap water, tap water + baking soda, and tap water + lemon juice were tested in reducing bifenthrin insecticide. Samples were blended and extracted using QuEChER. The extracted and concentrated samples were finally analyzed using Gas Chromatography Mass Spectrophotometer (GC-MS). Different household processes were compared based on the processing factor. The results revealed that soaking in Zamzam water reduced more pesticide residues compared with tap water. In case of washing strawberry fruits, tap water were most effective in reducing bifenthrin by more than 50% as compared to tap water with baking soda or tap water with lemon juice. This findings confirm that washing of fruits can remove pesticide residues, but the effectiveness depends on the physicochemical properties of a pesticide compound and type of fruits.

Keywords: Pesticide residue, household treatment methods, percentage recover, apple, strawberry

INTRODUCTION

Use of synthetic pesticides has well recognize benefit in terms of improving productivity, but excessive use of pesticides on agricultural fields have serious environmental and human health issues. Frequent consumption of fruits and vegetables is recommended to improve public health due to their known sources of antioxidants compounds such as phenolics and vitamin C (CDC, 2013; Agarwal *et al*., 2015), however these essential health benefits can be offset by the presence of significant levels of synthetic pesticide residues (Park, 2018; Wołejko *et al*., 2014). Contamination of fruits and vegetables with pesticides is often linked to intensive agriculture or inorganic farming, which uses mineral fertilizer and synthetic pesticides to increase crop yields (Oerke, 2005). Excess use of pesticides (insecticide or herbicide) in an intensive agriculture has adverse effects on the environment, and ultimately on human health. Various human health concerns are linked to pesticide uses including endocrine disruption, cancer, and Parkinson's disease (Cecchi *et al*. 2012; Gerage *et al*., 2017). Specially, people with weak immune system including children can be at risk due to chronic exposure to dietary sources of pesticides.

According to the Environmental Working Group (EWG), 70% of all fruits and vegetables contain up to 230 different pesticides that may affect human health and the environment (Park, 2018). This implies that chronic exposure of human to

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cocktails of pesticide compounds is possible at any age. Most importantly, the effect of cocktail of pesticides on human health is not known. Chronic exposure to pesticides may show various symptoms including neurotoxicity, chromosome changes, dermatitis, kidney damage, allergies, peripheral neuropathy, liver damage, heart rhythms, asthma, Parkinson's disease, teratogenic effects, tumors and hearing loss. (Pesticide National Network UK, 2017). On the other hands, acute pesticide poisoning may cause symptoms such as stomach cramps, fatigue, seizures, loss of appetite, vomiting, muscle spasms, conjunctive discomfort, headache, difficult respiration and nosebleed, each substance can have distinct symptoms or effects (Kumari, 2008; Jang *et al*., 2010). Dietary exposure to pesticides mainly linked to symptoms of chronic effects such as respiratory illnesses.

As it is difficult for the society to avoid agricultural pesticide uses, countries around the world have established maximum residue limits (MRL) for pesticides in food commodities to ensure food safety for consumers and protect human health (Solomon *et al*., 2002; Hazrat and Ezzat, 2019; USDA, 2020). It is important to explore methods that can reduce the pesticide level to below the MRL.

In case of fruits and vegetables, limited studies have assessed the effectiveness of common household processing such as washing, peeling, refrigeration, ozone treatment, cooking and ultrasonic waves in reducing some pesticides in fruits and vegetables (Kentish and Feng, 2014; Shabeer *et al*., 2015). According to George (2017), washing apples, tomatoes, lettuce, cucumber, and apricots with tap water could significantly reduce some compounds of pesticides residuals. Even if washing fresh produces with tap water is a common practice, it is not clear how this practice applies to most types of fruits and pesticide compounds.

The purpose of this study is to assess the effectiveness of different household processing in reducing pesticides from apple and strawberry fruits. Selected pesticides for this study include vinclozoline, metalaxyl, *methiocarb*, cyprodinil, thiabendazole, myclobutanil, bifenthrin and fenpropathrin. These compounds are fungicides except methiocarb, which is carbamate pesticide, and fenpropathrin and bifenthrin, which are pyrethroid insecticides. Even if it is classified by U.S. EPA as a possible human carcinogen, bifenthrin has been used on various agricultural crops and in homes since 1985. The study is particularly important as it aims to contribute to the knowledge gap on the potential effect of household food processing in reducing dietary exposure of human to synthetic pesticide residues.

MATERIALS AND METHODS

Pesticide Solutions and calibration curves

Multi-residue working solutions containing pesticides compounds were purchased from Restek® (Figure 1 and Table 1). Stock solution of each pesticide was prepared at 1000 µg/ml in ethyl acetate. Working standard mixtures of 300 µg/ml in ethyl acetate was used for spiking the samples and preparing calibration standards. Two different initial calibration standards were applied for different type of fruits. To analyze apple fruits, five-point calibration standards of 9 compounds applied. The concentrations include 1.5, 6, 12, 25 and 36 µg/ml. In Case of strawberry fruits, six-point calibration standards of bifenthrin was applied: 0.5, 1, 2, 5.0, 7.5, and 10 µg/ml. The internal standard concentration was 10 µg/ml. The linear regression correlation coefficients were applied to evaluate the calibration curves. For continuous calibration verification (CCV), 10 ug/mL was applied.

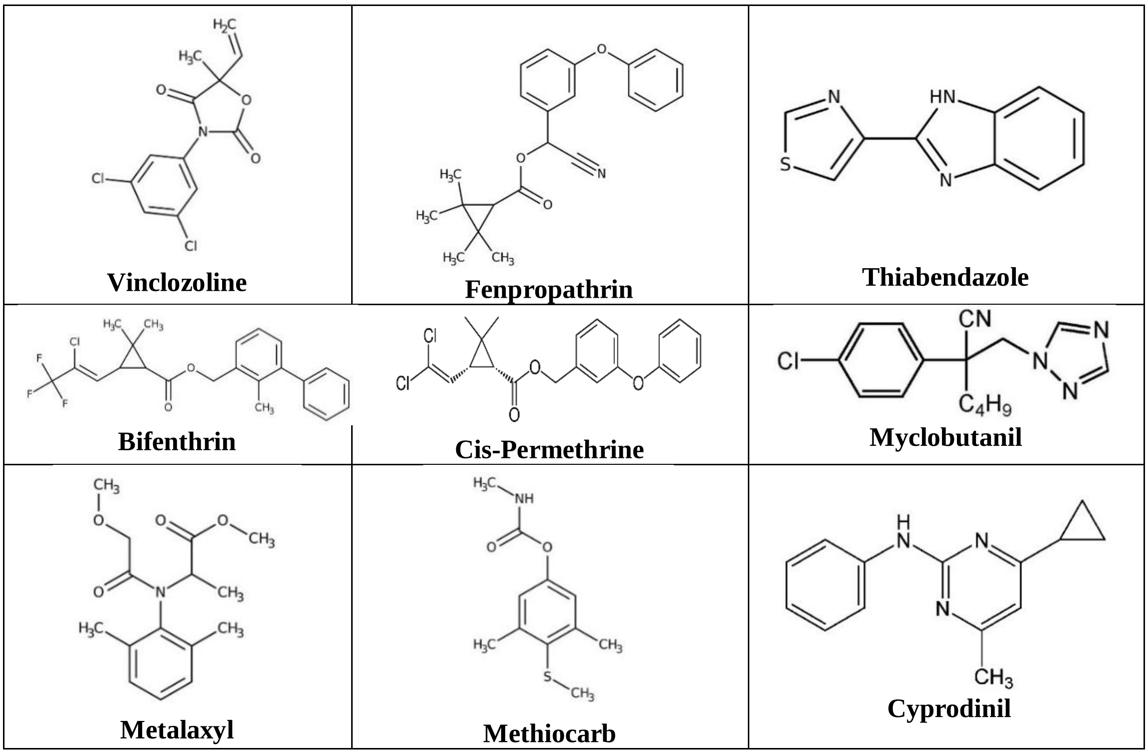

Figure 1. Chemical structures of target-pesticide compounds

Fruit sample spiking and extraction

Apple and strawberry fruit samples were purchased from the main supermarkets in Washington, DC and transported in a temperature-controlled container to be analyzed at the Environmental Quality Testing Laboratory (EQTL) of the University of the District of Columbia (UDC). Three replicates of fruit samples were spiked with 1mg/ml of stock solution. The samples were spiked with 100 ml of 300 ug/ml working standard solution and stored in a dark at room temperature for 24 hours before household processing or extraction. The samples were then treated with different house hold methods and then finally extracted and cleaned up using QuEChERS method (Lehotay *et al*., 2010). Prior to extraction, samples were blended and homogenized. Subsequently, a homogenized samples of 15 g was weighed into a 50 mL centrifuge tube. Then 15 mL acetonitrile was added. The tubes were capped and vigorously hand-shaken for 30 seconds. Salts (6.0 g Anhydrous $MgSO_4$ and 1.5 g NaCl) were added to each of the tubes, which were capped again and shaken for about one minute before centrifugation at 3000 rpm for 5 min. For the cleanup, 8 mL of the supernatant of sample extract was transferred to dispersive-SPE tubes containing 1200 mg $MgSO_4$, 400 mg PSA, and 400 mg C18. The tubes were shaken and centrifuged for 5 min. Then 4 mL of the supernatant of the extract was concentrated to 1 mL using evaporator. Then 0.5 mL of the cleaned-up and concentrated sample was transferred into a GC-MS vial for samples analysis.

Household processes

In this study, the effectiveness of two sets of household processing were tested for reducing pesticide residues from apple and strawberries fruits as follows:

I. Apple fruit samples were soaked in 2 L of tap water and Zamzam water for 15 minutes at pH 7.53 and 8.9, respectively.
II. Strawberry fruit samples were soaked in 2 L of tap water at pH of 7.3, and in tap water with lemon juice at pH 2.7, and tap water with baking soda pH of 8.2.

GC-MS analysis and calibration curves

Samples were analyzed using a PerkinElmer Clarus 600 GC-MS in full scan and selective ion mode. Turbo Mass software was applied for both ion identification and quantification. The identified ions were confirmed based on the built-in National Institute of Standards and Technology (NIST) library search. Each pesticide compound was quantified based on the peak height and area of internal standards (Tables 1 and 2, and Figures 2 and 3).

Table 1. Compounds identified and quantified in apple fruit analyses using full scan

Peak No.	Retention Time (min)	Compound Name	Quantifying Ion
1	17.03	2,4,4'-Trichlorobiphenyle (Internal Standard)	256
2	17.14	Vinclozoline	54
3	17.47	Metalaxyl	206
4	18	Methiocarb	168
5	18.13	2,2 ,5,5 Tetrachlor (Internal Standard)	220
6	19.68	Cyprodinil	224
7	20.17	Thiabendazole	174
8	22.08	Myclobutanil	179
9	26.34	Bifenthrin	181
10	26.61	Fenpropathrin	97

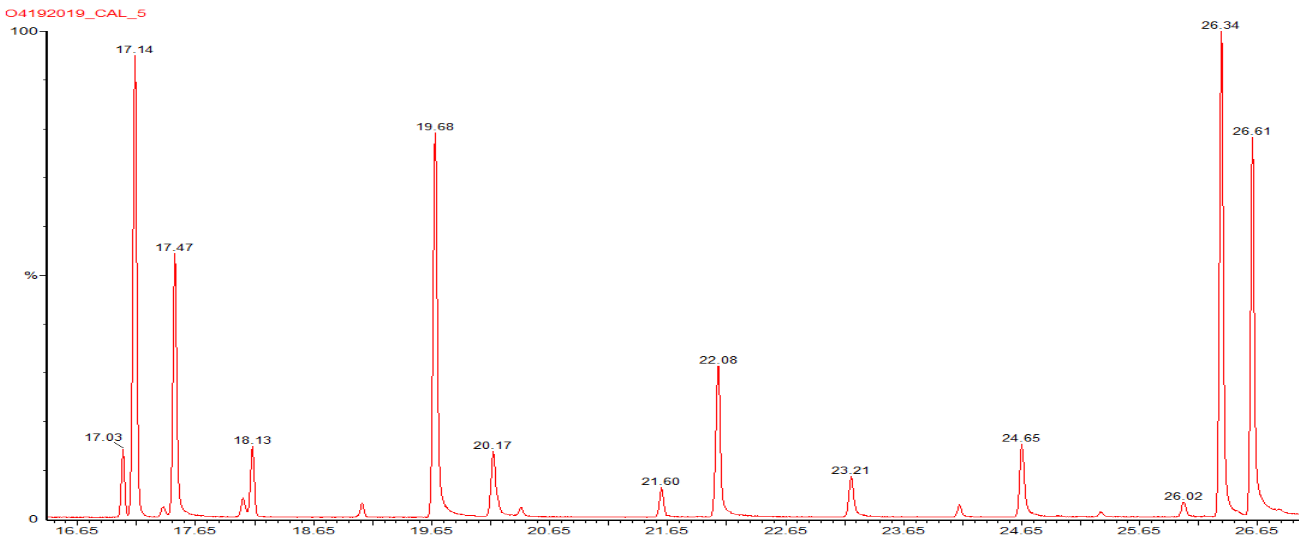

Figure 2. Identification for apple analysis by Gas Chromatography-Mass Spectrometry (GC-MS)

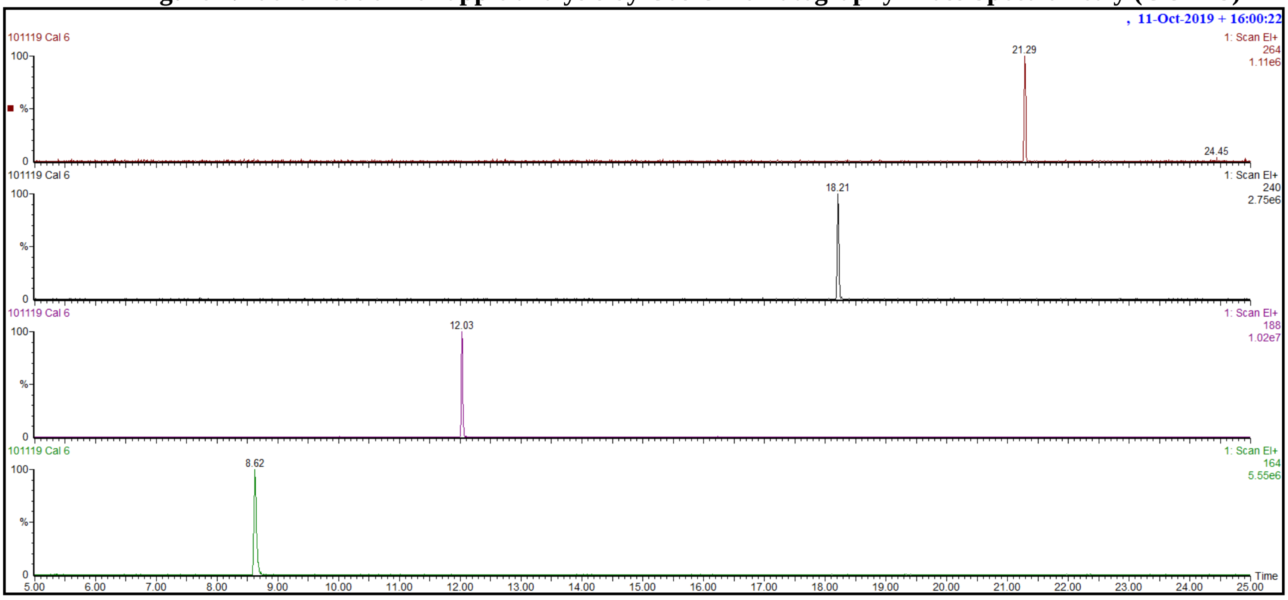

Figure 3. Identification for strawberry analysis by Gas Chromatography-Mass Spectrometry (GC-MS).

Table 2. Results of compounds identified and quantified in strawberry fruit analyses using selective ion mode

Peak No.	Retention Time (min)	Compound Name	Quantifying Ion
1	8.62	Acenaphthene-D10	164
2	12.03	Phenantheren_D6	188
3	18.21	Bifenthrin	181
4	21.29	Pyrene-D3	264

Figures 4 and 5 show the calibration curves for apple fruit analysis and strawberry fruit analysis, respectively. The linear regression coefficient (r^2) ranges from 0.84 to 0.99.

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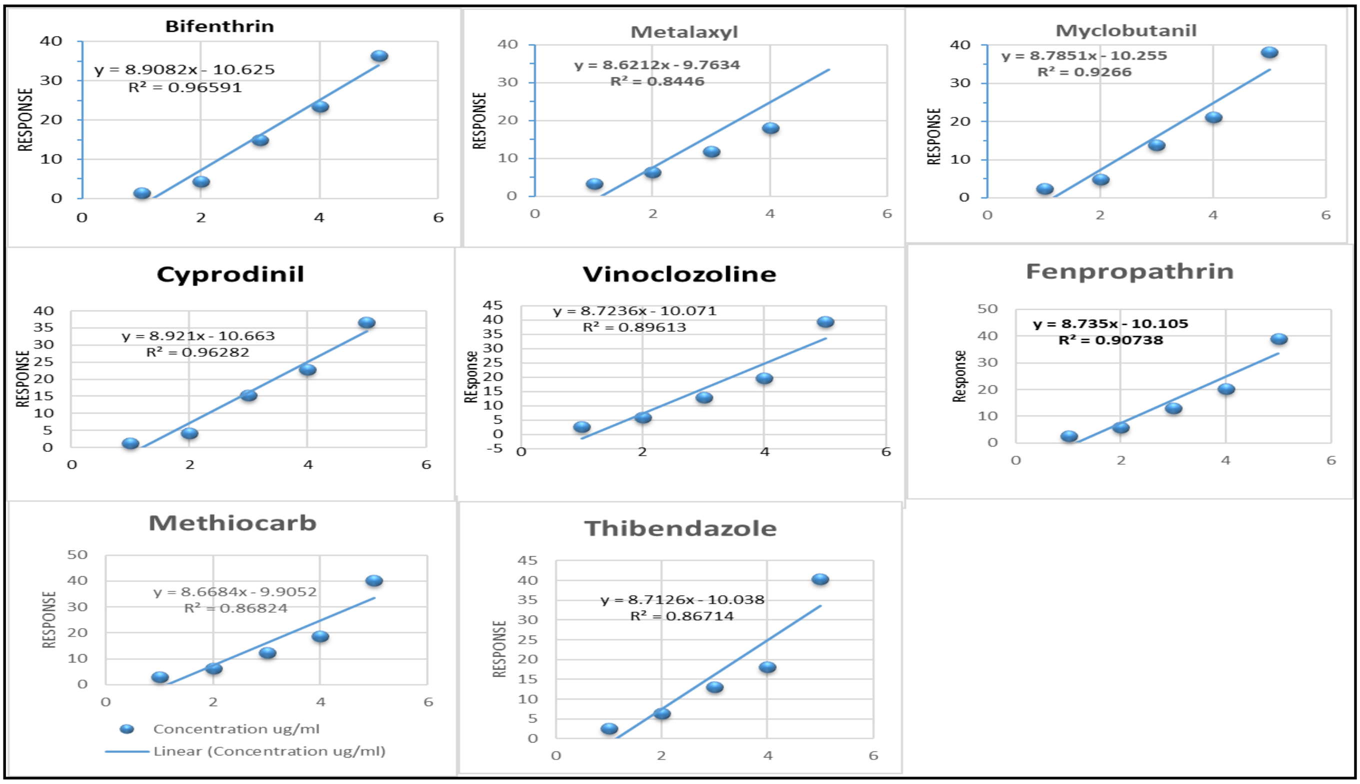

Figure 4. Calibration curve for apple analysis.

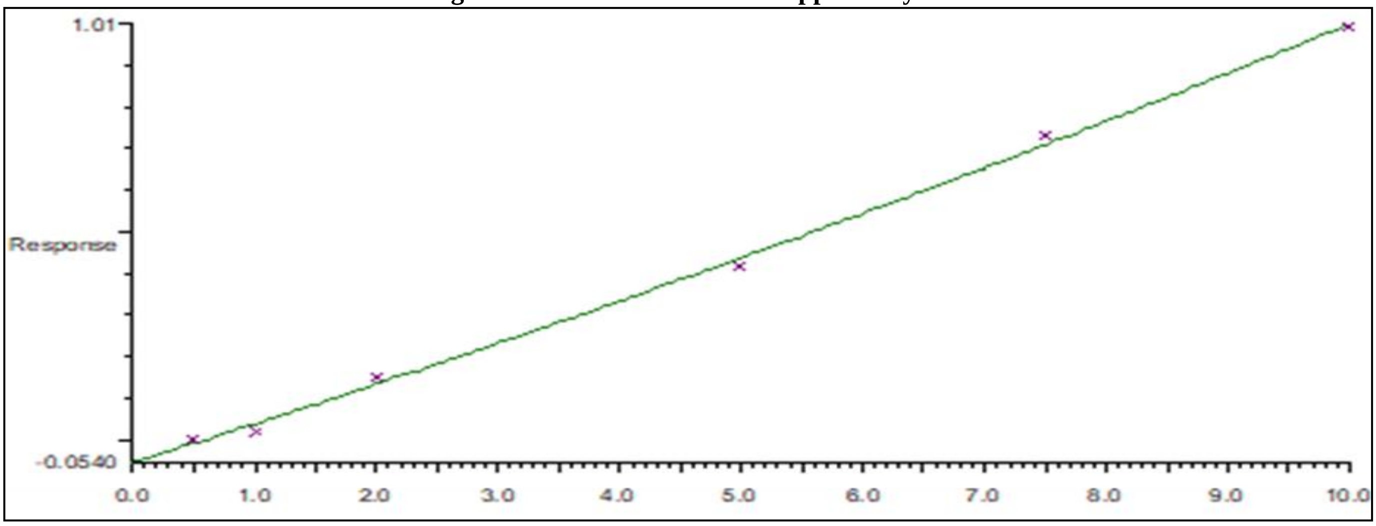

Figure 5. Bifenthrin calibration curve for strawberry fruit analysis (r^2 = 0.99)

Data analysis and calculation of processing factor

Priory to statistical analysis, the data sets were tested for normality distribution using histogram. The result shows that the data sets were not normally distributed, and thus a non-parametric statistical method was applied. Subsequently, depending on the number of samples analyzed, the Kruskal-Walis Method was selected for this study. The effectiveness of each method, also known as processing or reduction factor was calculated as follows (Bonnechere *et al*., 2012):

$$PF = \frac{A}{B} \times 100\%$$

Where PF is process factor[%]; A is the concentration of pesticide residue detected in a spiked and processed sample [ug/kg]; B is the concentration of pesticide residue in a spiked and unprocessed sample [ug/Kg].

The processing factor is expected to be less than 100% or the household process either does not work or results in concentration of the pesticide residues.

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RESULTS AND DISCUSSION

The process factor of soaking apple fruits in Zamzam water and tap water was assessed (Figure 6). The results showed that soaking in the Zamzam water resulted in more processing factor of bifenthrin, imazalil thiabendazole, myclobutanil, cyprodinil, trans permethrine and cispermethrine in washing apple fruits. In other words, tap water relatively shows less effective than the Zamzam water which is alkaline. This is consistent with the previous study that alkaline water is more effective in removing non-systemic pesticides from apple fruits surface than acidic or neutral water (Yang *et al.*, 2017). Systemic pesticides penetrate the surface of apple and become difficult to be washed off. It is also noted that most of pesticides are less stable in alkaline water than acidic water. This is due to alkaline hydrolysis that occurs when pesticides come in contact with alkaline water. Water with higher pH (> 8) can result in the breakdown of some pesticides through alkaline hydrolysis.

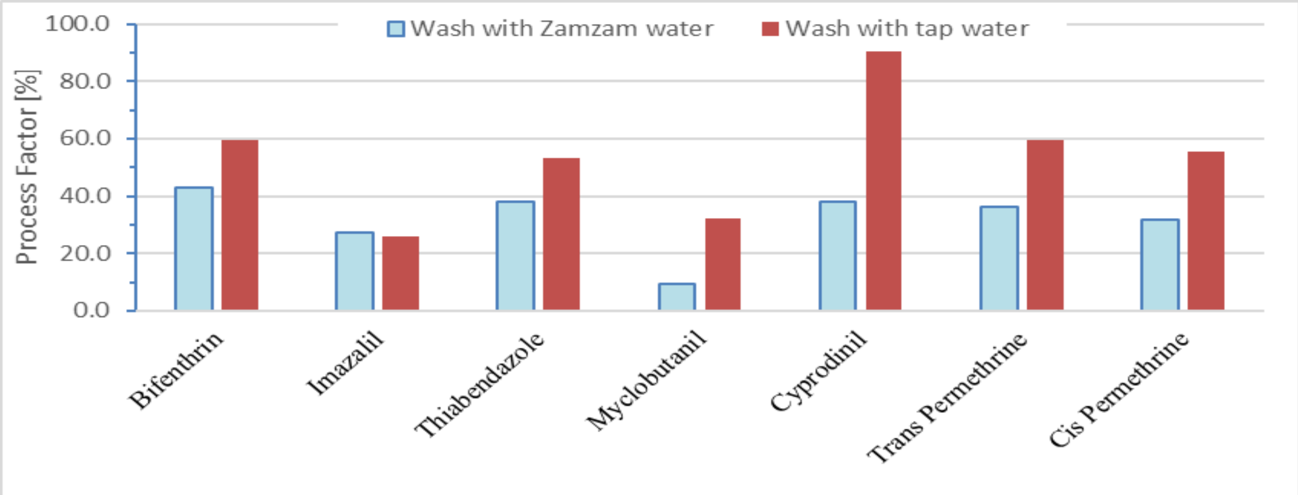

Figure 6. Comparison of average processing factor: Soaking in Zamzam water vs tap water

The effectiveness of three household processing methods in removing bifenthrin from strawberry fruits was also evaluated and the results are indicated in Table 3. The results showed that there is a significant difference among the three methods ($p < 0.05$): tap water, tap water with baking soda, and acidified tap water with lemon juice were evaluated. Tap water with near-neutral pH 7.3 resulted in more reduction in pesticide residues as compared to alkaline (tap water with baking soda with pH 8.3) or acidic tap water (tap water with lemon juice, pH 4). This indicates that lemon juice or baking soda is less effective in removing bifenthrin than tap water. The results showed that washing strawberries can significantly remove 50% of the pesticide residue, while washing with tap water + baking soda and tap water + lemon juice removed about 40% and 30% of the bifenthrin residue, respectively. In contrary, the acidic solution is effective in extracting organochlorin pesticides than other solutions (Bajwa and Sandhu, 2014).

Table 3 Pesticide residue levels in strawberry samples treated with different household methods

Household treatments methods	Spiked (μg/kg)	Sample Weight (g)	Measured (μg/Kg)	Processing Factor (% *PF)*
No treatment	156.50 ± 38.28	25.93 ±3.22	211.88±14.89	100.33± 28.31
Tap water	293.67± 33.93 [a]	20.07±1.55 [a]	215.37± 45.54	51.79 ± 10.19 [a]
Tap water + baking soda	293.03±91.66 [a]	19.80 ±2.87 [a]	242.36 ± 99.62	59.53±20.88 [a]
Tap water + lemon juice	359.53± 42.60 [a]	17.43±0.75 [a]	343.66 ± 10.93 [a, b]	79.17 ± 7.52

Data presented as mean± SD of three replicates. [a] significant difference compared with no treatment. [b] significant difference compared with tap water ($p < 0.05$).

This research finding is consistent with previous studies that washing fruits and vegetables before consumption reduces the level of pesticide residue. This study finding is consistent with the previous research findings. According to Tomer *et al.* (2014), washing with alkaline water can remove up to 89% of cypermethrin residues from okra. Moreover, soaking Chinese kale in 5 L of water containing half a teaspoon of vinegar for 10 min removed around 50% and 30% of cypermethrin and profenofos residues, respectively (Wanwimolruk *et al*., 2015). This study confirms that acidic solution is less effective, reducing some pesticide compounds as compared to alkaline solutions. It must also be noted that the effectiveness of the washing process depends on the type fruits and physicochemical properties of a pesticide compound. Furthermore, the stability of different pesticide depends on the pH and temperature conditions, the pesticide sprayed time, the pesticide solubility, nature of the fruit or vegetable, and treatment duration (Guardia-Rubio *et al*., 2007; Lopez-Fern´andez *et al*., 2013).

CONCLUSION

Dietary exposure to pesticides can be reduced by applying appropriate household processes that include washing. In this study, different household-processing methods were tested for their effectiveness in removing pesticide residue in fruits. These research findings confirmed that washing of fruits could remove pesticide residues, but the effectiveness of this household process depends on the physicochemical properties of a pesticide compound and type of fruits. Soaking fruits in alkaline water reduce more pesticides residues in some pesticide compounds, whereas acidified water such as adding lemon juice to tap water can reduce pesticides. Further research is required to study more household process as well as more fruits and vegetables.

List of abbreviations

QuEChERS: Quick, Easy, Cheap, Effective, Rugged, and Safe; GC-MS: Gas Chromatography-Mass Spectrometry; QC/QA: Quality control and Quality assurance; CCV: Continuous Calibration Verification; SPE: Solid Phase Extraction; CCB: Continuous verification of Calibration Blank.

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