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Comparison of Removal Efficiencies of Different Household Cleaning Methods in Reducing Imidacloprid and Triabendazole Residues on Cherry Tomatoes

Zhi-Yuan Meng[1,2], Yue-Yi Song[1,2], Xiao-Jun Chen[1,2], Ya-Jun Ren[1,2], Chun-Liang Lu[2], Li Ren[1,2], Hua-Chen Gen[1,2], Jia-Xin Zhu[1,2], Quan Yuan[1,2], Teng-Fei Li [1,2] & Zhi-Ying Xu[3]

[1] School of Horticulture and Plant Protection, Yangzhou University, Yangzhou, Jiangsu, P.R. China

[2] Joint International Research Laboratory of Agriculture & Agri-Product Safety, Yangzhou University, Yangzhou, Jiangsu, P.R. China

[3] Yangzhou Polytechnic College, Yangzhou, Jiangsu, P.R. China

Correspondence: Xiao-Jun Chen, School of Horticulture and Plant Protection, Yangzhou University, Yangzhou, Jiangsu, P.R. China. E-mail: cxj@yzu.edu.cn

Zhi-Ying Xu, Yangzhou Polytechnic College, Yangzhou, Jiangsu 225000, P.R. China. E-mail: yzxuzhiying@163.com

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Abstract

People have paid much attention on the pesticide residues in agricultural products at present. However there are less concern on processed agricultural products or on final consumption of processed foods, even though most processed foods are being finally consumed. In this paper, pesticide residues such as imidacloprid and triabendazole on the cherry tomatoes were cleaned by conducting different household cleaning methods in the way of normal vegetable cleaning. Results showed that it can effectively remove the residual imidacloprid on cherry tomatoes by soaking first them in water and then rinsing them with running tap water, wherein the removal rates were 30.59%-65.24% and processing factor were 0.3476-0.6941. While to remove the residual triabendazole on cherry tomatoes, we first soaked them in 0.1% edible vinegar solution and then rinsed them with running tap water, which can also effectively remove the triabendazole residues on the cherry tomatoes, with removal rates reaching 29.31%-74.01% and processing factor reaching 0.2599-0.7069. Our research provides an inherent relationship between pesticide residues and cleaning approaches as well as important theoretical basis for risk assessments of agricultural food.

Keywords: cherry tomatoes, imidacloprid, triabendazole, removal efficiency

1. Introduction

Compared with ordinary tomato or vegetable, cherry tomato is rich in nutrients such as organic acids, sugar, vitamins and minerals vitamin C (Li et al., 2012). Cherry tomato is a type of leguminous vegetable, which is being daily consumed in China. Cherry tomato is the host of thrips, pod borer, cutworm, aphids, leaf miner and other major pests, therefore chemical pesticides will be inevitably applied during its growth process. As a kind of neonicotinoid insecticide, imidacloprid enjoys advantages including high efficiency, broad spectrum against insects such as *Thysanoptera Homoptera, Coleoptera, Diptera* and *Lepidoptera* and other insects, and good uptake characteristics in protecting plants. Moreover, imidacloprid is of excellent effect in controlling sucking pests (Zhang et al., 2009; Liu, 2012). Belonging to benzimidazole fungicides thiabendazole has high efficiency, low toxicity, broad-spectrum and good uptake characteristics in protecting plants and plays as fresh-keeping sterilizing agent in the process of fruit storage (Zhao et al., 2009; Li et al., 2011). The application of imidacloprid and thiabendazole in cherry tomatoes production will inevitably cause pesticide residues, which may threat consumers' health.

Currently, a strict regulation has been implemented regarding pesticide residues on agricultural products such as fruits, crops and vegetable. Although some pesticide residues still remain at harvest, the residues tend to decline with the decomposition of pesticide. Great attention has been paid on the monitoring of pesticide residues, however insufficient emphasis has been placed on its cleaning process or on the processed agricultural products today. Most agricultural products are eaten after being cleaned. Therefore, it is very necessary to monitor these novel pesticides (imidacloprid and triabendazole) before and after cleaning. The two pesticides may degrade during food processing or storage processes, and some degradation products may be even more toxic than their parent compounds (St-Amand et al., 2004; Luís et al., 2005; Muhammad et al., 2006; Chai et al., 2009). Therefore, it is in great need to seek an appropriate cleaning method to remove the residual pesticides on agricultural products for both household and factory processing, so as to reduce the potential threat to human health. A systematic study of removal efficiencies of different cleaning methods is helpful for establishing guidelines for food cleaning and handling, with the purpose of minimizing human exposure to pesticide residues on food. Furthermore, it is a critical step for people to understand the removal efficiencies of different household cleaning methods in reducing imidacloprid and triabendazole residues on cherry tomatoes, because this is the basis for characterizing degradation products in the cherry tomatoes during processing, and for revealing the inherent relationship between pesticide residues and processing approaches, so that the theoretical basis for risk assessments of food can be reached.

2. Materials and Methods

2.1 Chemicals and Materials

Dr. Ehrenstorfer GmbH provided the technical grade analytical standard of imidacloprid (purity 98.0%) and triabendazole (purity 98.5%). 70% imidacloprid water dispersible granule (70% imidacloprid WDG) was obtained from Shanghai Dupont Agricultural Chemicals Co., Ltd, and 15% triabendazole suspension concentrate (15% triabendazole SC) was obtained from Jiangsu Bailing Agrochemical Co., Ltd. N-propyl ethylene diamine (PSA) and C18 were obtained from DIKMA Technology Co., Ltd. (USA). Edible vinegar, edible iodine salt, edible sodium bicarbonate, fruits and vegetables cleaning agent were obtained from Zhenjiang Hengguan Industry Co., Ltd., Jiangsu Guhuai Salt Co., Ltd. and Golden Bridge Salt Group, Nanjing Honeydew Park Sugar Co., Ltd., and Xi'an Kaimi Corporation, respectively.

2.2 Preparation of Imidacloprid and Triabendazole Residues on Cherry Tomatoes

The crisp, full pod, non-damage and non-mildewed cherry tomatoes, which were grown in Yangzhou University Experimental Farms, were selected as experimental materials. The upper and lower ends of the cherry tomatoes were removed. A 100 mg kg^{-1} soaking solutions of imidacloprid or triabendazole (70% imidacloprid WDG or 15% triabendazole SC) were prepared in deionized water, respectively. Totally 10 kg of cherry tomatoes were fully immersed in 60 L of the two soaking solutions respectively for 30 min at room temperature, and then dried in open air for 1 h.

2.3 Cleaning Methods

In treatment one (T_1), all cherry tomatoes were cleaned using a sieve with running tap water for 2 min, so that it can guarantee the cherry tomatoes were uniformly cleaned. After that the cherry tomatoes samples were air-dried in a fume hood at room temperature. In the second treatment (T_2), all cherry tomatoes remained uncleaned. Three replicates were applied for each treatment.

We investigated the soaking effects when using different solutions such water, 0.1% edible vinegar, 0.1% edible salt solution, 0.1% edible sodium bicarbonate solution and 0.1% fruit and vegetable cleaning solution. By soaking cherry tomatoes (1 kg) in 4 L of water, 0.1% water, 0.1% edible vinegar solution, 0.1% edible salt solution, 0.1% edible sodium bicarbonate solution, and 0.1% fruit and vegetable cleaning solution, for 5, 10, 15, 30, 45 and 60 min, respectively. Cherry tomatoes in control group remained uncleaned. After that the cherry tomatoes were rinsed with running tap water for 2 min for three times, and then air-dried in a fume hood. Imidacloprid or triabendazole residues on cherry tomatoes were extracted, cleaned-up and analyzed using HPLC. Removal efficiency and processing factor of imidacloprid and triabendazole were calculated respectively according to the contents of their residues on cherry tomatoes.

2.4 Extraction of Imidacloprid and Triabendazole Residues from Cherry Tomatoes

Cherry tomatoes (5 g) were homogenized in 30 mL of acetonitrile for 3 min using high-speed homogenizer. For recovery experiment, imidacloprid and triabendazole were added to the cherry tomatoes samples, respectively, and the final concentration of imidacloprid was 5.00, 1.00, and 0.20 μg g^{-1}, so was that of triabendazole. In addition, a blank control group was set up and all experiments were repeated three times.

Imidacloprid and triabendazole were extracted from cherry tomatoes using QuEChERS method after applied with different cleaning treatments. Sodium chloride (1.5 g) and anhydrous magnesium sulfate (6 g) were added to the samples before conducting centrifuging treatment at 4000 rpm for 5 min. Afther that, it can collect about 2 mL of supernatant, which was then added into the centrifuge tubes containing $MgSO_4$ (150 mg), PSA (25 mg), and C_{18} (25 mg) for 5 min of centrifuging. Finally, the sample solution was filtered using a 0.22 μm membrane and analyzed by HPLC.

2.5 Instrumental Analysis

Treatments of separation and detection were carried out using L-2000 Series HPLC (Hitachi Co., Japan) which was equipped with UV detector. The imidacloprid residues were quantitatively determinded by HPLC as conducted by Lu and Wang (Wang et al., 2012; Lu et al., 2013). It is worth noting that the L-2000 HPLC system used in the experiment was equipped with a binary pump, auto plate-sampler, column oven, and UV detector. The treatment of separation was performed using ODS chromatographic columns C_{18} (250 mm× 4.6 mm (i.d.), 5 μm) under the conditions of 25 °C, wavelength 270 nm, mobile solvents CH_3CN:H_2O = 30:70 (V:V) and isocratic at 1.0 mL min^{-1}. 10 µL of aliquot was injected directly into the HPLC system to test imidacloprid and quantified with external standard peak area. The triabendazole residues were quantitatively determined using HPLC by Wang and Zhao (Wang et al., 2008; Zhao et al., 2009). The treatment of separation was conducted using ODS chromatographic columns C_{18} (250 mm× 4.6 mm (i.d.), 5 µm) under the conditions of 25 °C, wavelength 298 nm, mobile solvents CH_3OH:H_2O = 60:40 (V:V) and isocratic at 1.0 mL min^{-1}.

2.6 Calculation Methods for Removal Rate and Processing Factor

Removal rate and processing factor of each treatment were calculated. The removal rate and processing factor can be calculated by formulas as below, respectively.

$$\text{Removal rate}(\%) = \frac{\text{Initial concentration - Residues concentration}}{\text{Initial concentration}} \times 100 \qquad (1)$$

$$\text{Processing factor} = \frac{\text{Residues concentration}}{\text{Initial concentration}} \qquad (2)$$

All data were analyzed through analysis of Duncan multiple comparison. All the experiments were repeated three as means±standard error of mean. Different lowercase letters after the number was of 5% significant difference, while different uppercase letters after the number was of 1% significant difference.

3. Results

3.1 Fortified Recoveries of Imidacloprid and Triabendazole from the Cherry Tomatoes

To figure out the extrication efficiency and clean-up procedures, recovery experiments were conducted at different fortification levels to verify the reliability and validity of analytical method. The control samples of cherry tomatoes were spiked at 5, 1.0, and 0.2 mg kg^{-1} respectively followed by methodology as described above. According to Table 1, it can be seen that the average recovery rate was over 80%. In addition, the coefficients of variation were changed from 1.89% to 4.08% and changed from 1.89% to 4.35%, respectively.

Table 1. The recoveries of imidacloprid and triabendazole from the cherry tomatoes

Pesticides	Fortification levels (mg kg^{-1})	Average recoveries (%)	Standard deviation	Coefficient of variance (%)
	5.0	92.72	0.0878	1.89
imidacloprid	1.0	89.52	0.0358	4.00
	0.2	82.03	0.0067	4.08
	5.0	93.71	0.0884	1.89
triabendazole	1.0	88.67	0.0226	2.55
	0.2	84.20	0.0073	4.35

Note. The remaining amount was the average of three replicates.

3.2 Removal Efficiencies of Different Cleaning Methods in Removing the Residual Pesticide on Cherry Tomatoes

3.2.1 Removal Efficiency of the Cleaning Method Using Running Tap Water

According to Table 2 that the removal rate and the processing factor were 49.37% and 0.5043, respectively, after the cherry tomatoes with imidacloprid residue were cleaned with running tap water for 5 min. In constrat, the removal rate and the processing factor for cherry tomatoes with triabendazole residues were 33.93% and 0.6607, respectively after cleaning with simply running tap water for 5 min.

Table 2. Removal efficiencies of residual imidacloprid and triabendazole on cherry tomatoes by cleaning with running tap water

Samples	Treatment method	Initial concentration (mg kg^{-1})	Residues concentration (mg/kg)	Removal rate (%)	Processing factor
Imidacloprid	Tap water 5 min	4.473±0.4435	2.2648±0.0892	49.37±1.9953	0.5063±0.0199
Triabendazole	Tap water 5 min	4.3640±0.2564	2.8833±0.4289	33.93±9.8271	0.6607±0.0983

Note. The experiments were set up in a completely randomized design. All the experiments were repeated thrice as means ± standard error of mean.

3.2.2 Removal Effects of Residual Imidacloprid and Triabendazole on Cherry Tomatoes by First Soaking in Tap Water and Then Cleaned with Running Tap Water

According to Figure 1, it can be seen that the removal rates were 30.59%-65.24% and the processing factors were 0.3476-0.6941 after the cherry tomatoes with imidacloprid residues were first soaked in the water for several times and then cleaned with running tap water for 2 min. In contrast, the removal rates reach 19.13%-60.73% and the processing factors reach 0.3927-0.8087 after the cherry tomatoes with triabendazole residue were first soaked in the water for several times and then cleaned with running tap water for 2 min.

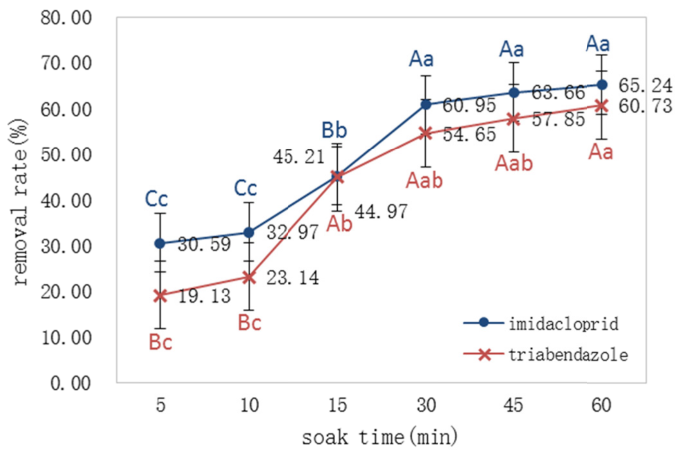

Figure 1. Removal efficiencies of residual imidacloprid and triabendazole on the cherry tomatoes by first soaking in tap water and then cleaned with running tap water

3.2.3 Removal Efficiency of Residual Imidacloprid and Triabendazole on Cherry Tomatoes by First Soaking in Edible Salt Solution and Then Cleaned with Running Tap Water

According to Figure 2, it can be seen that the removal rate were 23.61%-50.76% and the processing factors were 0.4924-0.7639 after the cherry tomatoes with imidacloprid residues were first soaked in the 0.1% edible salt solution for different times and then cleaned with running tap water for 2 min. In constrat the removal rates were 22.62%-63.95% and the processing factors were 0.3605-0.7738 after cherry tomatoes with triabendazole residues were first soaked in the 0.1% edible salt solution for different times and then cleaned with running tap water for 2 min.

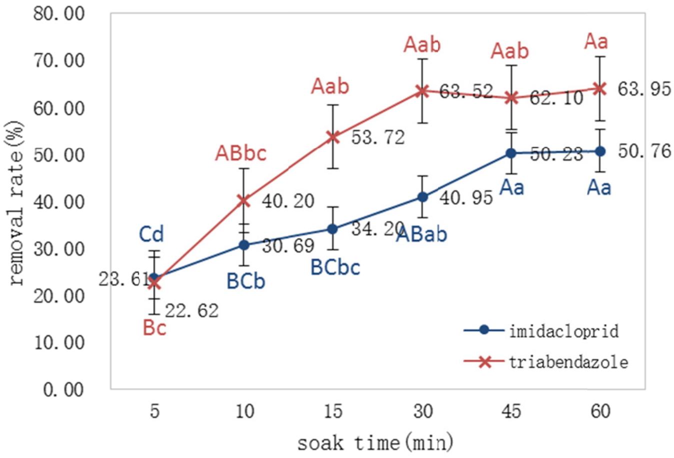

Figure 2. Removal efficiencies of residual imidacloprid or triabendazole on cherry tomatoes by first soaking in the salt solution for different times and then cleaned with running tap water

3.2.4 Removal Efficiency of Residual Imidacloprid or Triabendazole on the Cherry Tomatoes by First Soaking in the Edible Vinegar Solution and Then Cleaning with Running Tap Water

According to Figure 3, it can be seen that the removal rate were 19.97%-64.79% and the processing factors were 0.3521-0.8003 after the cherry tomatoes with imidacloprid residues were first soaked in the 0.1% edible vinegar solution for different times and then cleaned with running tap water for 2 min. In contrast, the removal rates were 29.21%-74.01% and the processing factors were 0.2599-0.7069 after the cherry tomatoes with triabendazole residues were first soaked in the 0.1% edible vinegar solution for different times and then cleaned with running tap water for 2 min.

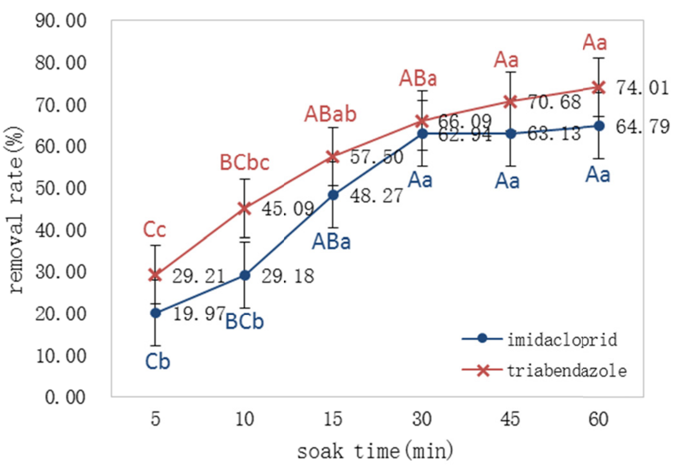

Figure 3. Removal efficiencies of residual imidacloprid and triabendazole on cherry tomatoes by first soaking in the 0.1% edible vinegar solutions for different times and then cleaned with running tap water

3.2.5 Removal Efficiency of Residual Imidacloprid and Triabendazole on the Cherry Tomatoes by First Soaking in the Edible Sodium Bicarbonate Solution and Then Cleaned with Running Tap Water

As shown in Figure 4, the removal rate were 15.42%-61.04% and the processing factors were 0.3896-0.8458 after the cherry tomatoes with imidacloprid residues were first soaked in the 0.1% edible sodium bicarbonate solution for different times and then cleaned with running tap water for 2 min. In contrast, the removal rates were 21.81%-59.55% and the processing factors were 0.4045-0.7819 after the cherry tomatoes with triabendazole residue were first soaked in the 0.1% edible sodium bicarbonate solution for different times and then cleaned with running tap water for 2 min.

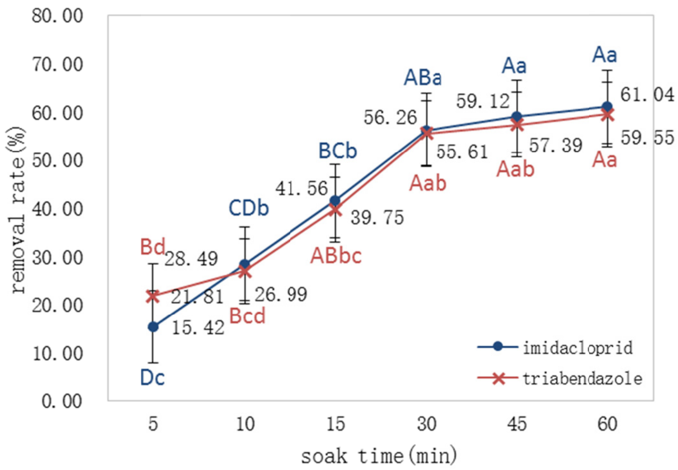

Figure 4. Removal efficiencies of residual imidacloprid or triabendazole on cherry tomatoes by first soaking in the 0.1% sodium bicarbonate solution for different times and then cleaned with running tap water

3.2.6 Removal Efficiencies of Residual Imidacloprid or Triabendazole on Cherry Tomatoes by First Soaking in the Fruit and Vegetable Cleaning Solution and Then Cleaned with Running Tap Water

As shown in Figure 5, the removal rate were 17.67%-56.26% and the processing factors were 0.4376-0.8232 after the cherry tomatoes with imidacloprid residues were first soaked in the 0.1% fruit and vegetable cleaning solution for different times and then cleaned with running tap water for 2 min. In contrast, the removal rates were 22.83%-57.81% and the processing factors were 0.4219-0.7717 after the cherry tomatoes with triabendazole residues were first soaked in the 0.1% fruit and vegetable cleaning solution for different times and then cleaned with running tap water for 2 min.

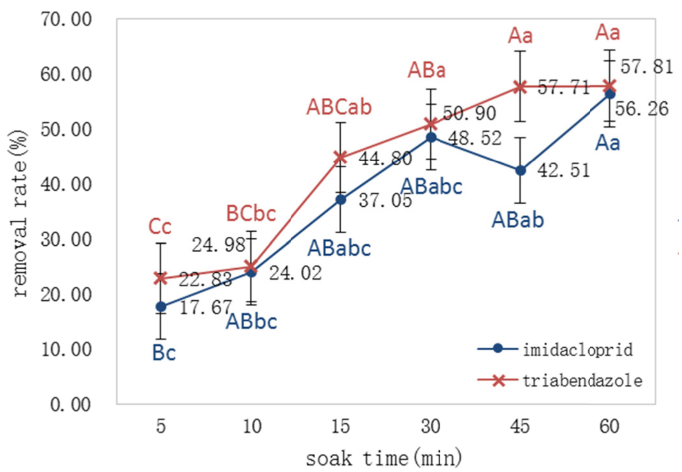

Figure 5. Removal efficiencies of residual imidacloprid or triabendazole on cherry tomatoes by first soaking in the 0.1% fruit and vegetable cleaning solution for different times and then cleaned with running tap water

As shown in Figure 6, it can conclude that cleaning method of first soaking in the water and cleaning with running tap water is the preferred method. After being soaked in water for 30 min, the repeated times for soaking will not affect the removal efficiency of residual imidacloprid. Imidacloprid enjoys good solubility in water which is 0.61 g/L and KowlogP = 0.57 with low lipid water distribution coefficient (Jeschke et al., 2011; Liu, 2012). The residual imidacloprid may easily enter into the water when the cherry tomatoes are soaked in the water.

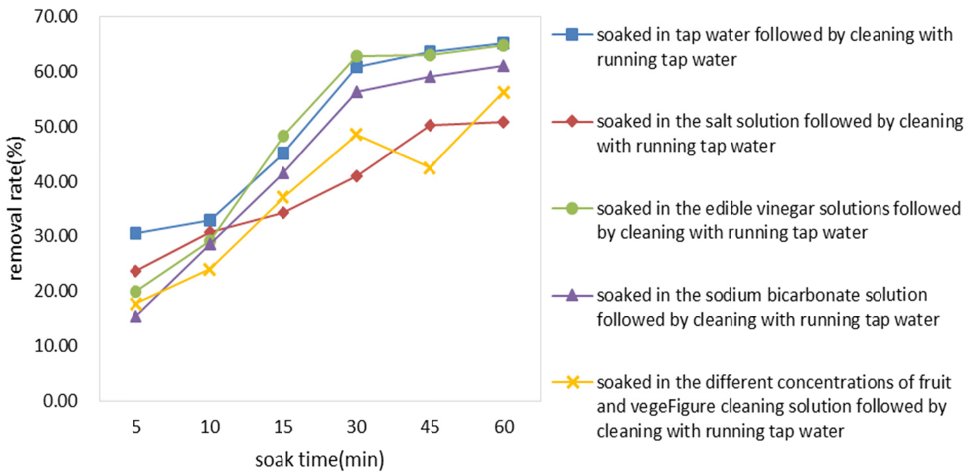

Figure 6. Removal efficiency of residual imidacloprid on cherry tomatoes by soaking in different solutions for different times

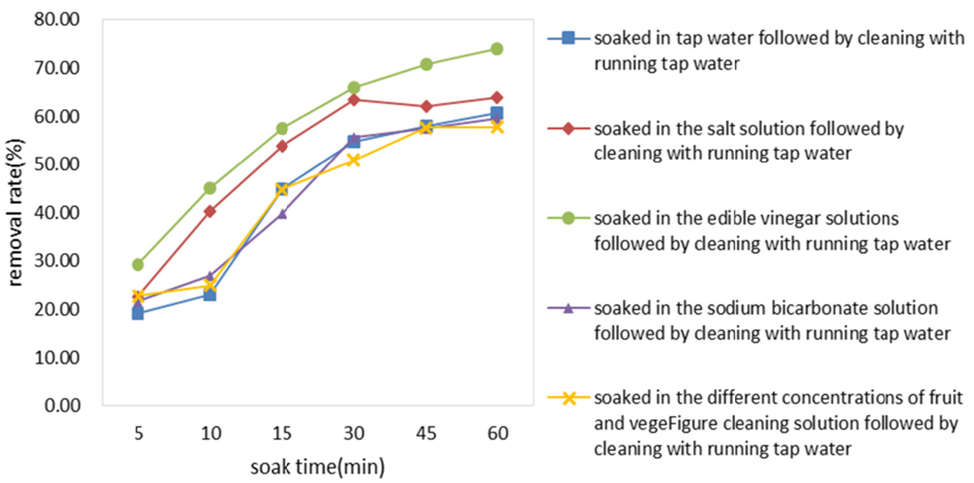

Figure 7. Removal efficiency of residual triabendazole on cherry tomatoes by soaking in different solutions for different times

As shown in Figure 7, it can conclude that the cleaning method of first soaking in the 0.1% edible vinegar and then cleaned with running tap water was the preferred method. After the residue triabendazole cherry tomatoes being soaked in the 0.1% edible vinegar for 30 min, the repeated times for soaking will not affect the removal efficiency of residue triabendazole on cherry tomatoes. Triabendazole enjoys good solubility in the water which is 0.05 g/L and KowlogP = 2.39 (pH = 7) with high lipid water distribution coefficient (Liu, 2012). The residual triabendazole on cherry tomatoes may easily enter into 0.1% edible vinegar when being soaked in 0.1% edible vinegar.

4. Conclusions

In this study, pesticide residues such as imidacloprid and triabendazole on the cherry tomatoes were cleaned using different cleaning techniques in the way of vegetable cleaning habits. The cleaning methods included rinsing with running water, cleaning after soaking in water, edible vinegar, edible salt, sodium bicarbonate solution or fruit and vegetable cleaning solution, respectively. It concluded that using the method of first soaking in water and then rinsing with running tap water, it could remove the imidacloprid residues on the cherry tomatoes most effectively, with removal rates reaching 30.59%-65.41% and processing factor reaching 0.3476-0.6941. In contrast, based on the method of first soaking in 0.1% edible vinegar solution and then rinsing with running tap water, it can remove the triabendazole residues on the cherry tomatoes most effectively, with removal rates reaching 29.31%-74.01% and processing factor reaching 0.3521-0.7069. Our research provides the inherent relationship between pesticide residues and cleaning approaches, as well as the important theoretical basis for risk assessments of agricultural food.

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