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Effect of household processing on reduction of pesticide residues in Tomato (*Lycopersicon esculentum* Mill)

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ABSTRACT

Tomatoes (Lycopersicon esculentum Mill.) contribute to a healthy, well-balanced diet. They are rich in minerals, vitamins, essential amino acids, sugars and dietary fibres. Tomato contains much vitamin B and C, iron and phosphorus. Tomato fruits are consumed fresh in salads or cooked in sauces, soup and meat or fish dishes. They can be processed into purées, juices and ketchup. Canned and dried tomatoes are economically important processed products. The major tomato producing states are Andhra Pradesh, Karnataka, West Bengal, Orissa, Maharashtra, and Gujarat. 978.44, 841.30 and 746.20 tones tomato produced by Gujarat state during 2010-11, 2009-10 and 2008-09 respectively (Source: National Horticulture Board (NHB)). Traditionally tomato preparation is eaten in the form of raw or cooked. Therefore, raw, washed and cooked form of tomato was selected for the study. The effects of household processing on pesticide residues were also studied. Analysis of tomato for pesticidal contamination was carried out on Gas Chromatograph-Electron Capture and TID Detector with capillary columns. Tomato was found contaminated with Phorate, malathion, parathion, quinalphos, profenophos, pendamethalin, aldrin, p,p' DDT, captafol, permethrin and , cypermethrin. The study revealed that tomato was found contaminated maximum with parathion and minimum with p, p' DDT in the range of 13.20-15.25 and 0.0065-0.0078 µgg^{-1} respectively. Findings show that washing and cooking process minimized the pesticide residues of eleven pesticides in the range of 1.74-64.78 and 38.40-90.15 percent respectively. The percentage reductions in the present study are supported by both early and most recent publications. These reductions are extremely important in evaluating the risk associated with ingestion of pesticide residues, especially in vegetables, which are eaten by almost all income groups' people. The present study showed that cooking was found more effective than washing and boiling.

Key words: Tomato, processing, pesticide residue, OP & OC.

INTRODUCTION

Vegetables are essential components of our diet due to their nutritional value. Fruits, nuts, and vegetables play a significant role in human nutrition, especially as sources of vitamins (C, A, B6, thiamine, niacin, E), minerals, and dietary fiber [1-3]. In near future, there is a need of around 5- 6 million tones of vegetables to feed over 1.3 billion Indian population expected by the year 2020 [4]. The total area under vegetables crops is 71, 31, 000 hectares with total annual production of 11, 01, 06000 tones [5, 6]. However, several factors limit their productivity, mainly insect pests and diseases, due to increased pest menace there is an average loss of 40% in different crops [7]. In order to combat the insect pest problem, lot of pesticides is used by the vegetable growers for better yield and quality. Insecticides are repeatedly applied during the entire period of growth and sometimes even at the fruiting stage. It accounts for 13-14% of total pesticide consumption as against 2.6% of cropped area [8]. Pesticide exposure has been

associated with human health risk of arthritis, skin disease, bone disorder, cancer and nerve disorder [9, 10]. Indiscriminate use of pesticides particularly at fruiting stage and non-adoption of safe waiting period leads to accumulation of pesticides residues in consumable vegetables. Contamination of vegetables with pesticide residues has been reported by many researchers [11-13]. Scientists and food processors have long been interested in the effect of processing on pesticide residues in food commodities. The extent to which pesticide residues are removed by processing depends on a variety of factors, such as chemical properties of the pesticides, the nature of food commodity, the processing step and the length of time the compound has been in contact with the food [14-16]. The presence of pesticide residues is a major bottleneck in the international trade of food commodities.

Tomato (*Lycopersicon esculentum* Mill.) is one of the most important vegetables worldwide. As it is a relatively short duration crop and gives a high yield, it is economically attractive and the area under cultivation is increasing daily. Tomato belongs to the *Solanaceae* family. This family also includes other well-known species, such as potato, tobacco, peppers and eggplant (aubergine). Tomato has its origin in the South American Andes. The cultivated tomato was brought to Europe by the Spanish conquistadors in the sixteenth century and later introduced from Europe to southern and eastern Asia, Africa and the Middle East. More recently, wild tomato has been distributed into other parts of South America and Mexico.

Tomatoes (*Lycopersicon esculentum* Mill.) contribute to a healthy, well-balanced diet. They are rich in minerals, vitamins, essential amino acids, sugars and dietary fibres. Tomato contains much vitamin B and C, iron and phosphorus. Tomato fruits are consumed fresh in salads or cooked in sauces, soup and meat or fish dishes. They can be processed into purées, juices and ketchup. Canned and dried tomatoes are economically important processed products. The major tomato producing states are Andhra Pradesh, Karnataka, West Bengal, Orissa, Maharashtra, and Gujarat. 978.44, 841.30 and 746.20 tones tomato produced by Gujarat state during 2010-11, 2009-10 and 2008-09 respectively (Source: National Horticulture Board (NHB)). Traditionally tomato preparation is eaten in the form of raw or cooked. Therefore, raw, washed and cooked form of tomato was selected for the study.

EXPERIMENTAL SECTION

Chemicals

a. Reagents: Standard pesticides which were >98% pure were procured from RFCL, Delhi, India. HPLC grade hexane, acetone and ethyl acetate, and AR grade anhydrous sodium sulphate, sodium chloride, Florisil, Activated charcoal, Silica gel for column chromatography were procured from RFCL, Delhi, India.

b. Standard materials: Standard pesticides which were >98% pure were procured from RFCL, Delhi, India. The standard stock solutions (100 ppm) were prepared in ethyl acetate and stored at -4^0C. Working standard mixtures of six pesticides in ethyl acetate, containing 10 µg/ml of each pesticide, were used for spiking the samples and preparing calibration standards.

Instruments

a. Blender-Boss Appliances, Daman, India
b. Centrifuge-Kumar Industries, Bombay, India
c. Mechanical shaker -Modern Industrial corporation, Bombay, India
d. Rotary evaporator -Jain Scientific, India
e. GC- Thermofisher 1000 GC equipped with capillary columns using ^{63}Ni electron capture detector (ECD) and TID.
f. Capillary column- 1. SPB-5 of 5% diphenyl/ 95% dimethyl fused silica capillary column (30 m×0.32 mm ID, 0.25 μm film thickness) 2. HP-1 of methyl silicone (10 m×0.53 mm ID, 2.65 μm film thickness).

Instrument conditions

For OC: Temperatures (^{0}C):150 (5 min) → 8 ^{o}c min^{-1} → 190 (2 min) → 15 ^{o}c min^{-1} 280°c (10 min); injection port: 280 ^{o}c; detector: 300 ^{o}c; carrier gas: (N_2), flow rate 60 ml min^{-1}, 2 ml through column and split ratio 1:10. Carrier gas, N_2, flow rate 60 ml min^{-1}, 2 ml through column.

For op: Temperatures(^{0}C): Oven: 100 (1 min) → 10 ^{o}c min^{-1} → 200 ^{o}c (0 min) → 20 ^{o}c min^{-1} → 260 ^{o}c (3 min); injector port, 250 ^{o}c , detector, 275 ^{o}c , carrier gas N_2 18 ml min^{-1}, H_2, 1.5 ml min^{-1} and zero air 130 ml min^{-1}.

Sampling

A total of 45 samples of vegetables were commercially purchased from the local market of Rajkot city, Gujarat, India, during October 2010 and February 2011 and served as the blank or spiked sample. All the samples were extracted fresh. . The unit was generally more than 250 g [17]. For the analysis, only the edible portions were included, whereas bruised or rotten parts were removed.

Processing vegetables

Samples of tomato, were washed, sliced into a suitable size and cooked. Vegetable samples (raw) were dry, cleaned to remove soil contamination with a disposable paper towel and blended to mace a homogeneous sample for pesticide analysis.

Washing

Vegetables were washed by placing in a plastic colander and rinsed under normal tap water (25-30^{0}c) for 30 second [18] with gentle rotation by hands and blotted dry with a paper towel. These samples were divided into two portions, of which one was analyzed as such after homogenizing in blender and other was further boiled and cooked.

Boling

Sliced vegetables were boiled by placing 75 ml of water in saucepan. Vegetable (50g) was added immediately to boil for 5-10 min / boiled still softness was subjected to pesticide analysis.

Cooking

Sliced vegetables were cooked (Kilgore et al., 1970) by placing 15 ml of water in saucepan. Vegetable (50g) was added immediately to cook for 10-12 min was subjected to pesticide analysis. Washed, boiled and cooked samples were processed in a similar manner as of unprocessed samples

Extraction

Commercially purchased tomato served as the blank or spiked sample. All the samples were extracted fresh. Each sample was chopped into small pieces and after quartering, a representative sample (50g) was macerated with 5-10g anhydrous sodium sulphate in Warring blender to make a fine paste. The macerated sample was extracted with 100ml acetone on mechanical shaker for 1 h by using the method of Kumari *et al.[12]*. Extract was filtered, concentrated up to 40ml and subjected to liquid-liquid partitioning with ethyl acetate (50, 30, 20 ml) after diluting 4-5 times with 10% aqueous NaCl solution. Concentrated the organic phase up to 10ml on rotary evaporator and divide it into two equal parts. One part was kept for OC and second for OP.

Clean-up

For OC, clean-up was carried out by using column chromatography. Column (60cm × 22mm) was packed with, Florisil and activated charcoal (5:1 w/w) in between the two layers of anhydrous sodium sulphate. Extract was eluted with 125ml mixture of ethyl acetate: hexane (3:7 v/v). Eluate was concentrated to 2ml for residue analysis.

Residues of OP were also cleaned by adopting column chromatographic technique. Column was packed with silica gel and activated charcoal (5:1 w/w) in between the layers of anhydrous sodium sulphate. Extract was eluted with 125ml mixture of acetone: hexane (3:7 v/v). After concentrating the eluate on rotary evaporator, final volume was made to 2ml for analysis by gas liquid chromatography (GC).

Quantization

An external method was employed in the determination of the quantities of residues in the sample extracts. A standard mixture of known concentration of pesticide was run and the response of the detector for each compound ascertained. The area of the corresponding peak in the sample was compared with that of the standard. All analyses were carried out in triplicates and the mean concentrations computed accordingly.

Recovery rate and limit of detection

Tomato samples were fortifies at 0.01, 0.02 and 0.1 mg/kg by adding 5.0 mL of a mixed standard solution. Recovery and precision (expressed as relative standard deviation) were calculated for three replicate samples. Percent recoveries in spiked samples ranged 87.3% -104.0 % [19]. Accordingly, the sample analysis data were corrected for these recoveries. Detection limit(s) of the method were also assessed based on the lowest concentrations of the residues in each of the matrices that could be reproducibly measured at the operating conditions of the GC; which were 0.001 mg/kg. Blank analyses were also carried in order to check any interfering species in the reagents.

Estimation

The cleaned extracts were analyzed on Thermofisher 1000 GC equipped with capillary columns using ^{63}Ni electron capture detector (ECD) and TID. Operating conditions were as per details: For OC: Detector : ECD (^{63}Ni), column: SPB-5 of 5% diphenyl/ 95% dimethyl fused silica capillary column (30 m×0.32 mm ID, 0.25 μm film thickness) with split system. Temperatures (^{0}C):150 (5 min) → 8 ^{o}c min^{-1} → 190 (2 min) → 15 ^{o}c min^{-1} 280°c (10 min); injection port: 280 ^{o}c; detector: 300 ^{o}c; carrier gas: (N_2), flow rate 60 ml min^{-1}, 2 ml through column and split ratio 1:10. Carrier gas, N_2, flow rate 60 ml min^{-1}, 2 ml through column.

RESULTS AND DISCUSSION

The average percent recoveries at the spiking levels of 1µg/ml of each pesticide were in the range of 80–110. The data collected during this study is presented in Tables 1. In the analyzed samples, the detected pesticides comprised of Phorate, malathion, parathion, quinalphos, profenophos, pendamethalin, aldrin, p,p' DDT, captafol, permethrin and , cypermethrin. The study revealed that tomato was found contaminated maximum with parathion and minimum with p, p' DDT in the range of 13.20-15.25 and 0.0065-0.0078 µgg^{-1} respectively. In India, DDT has been banned with effect from April 1993. Practically, DDT is not phased out completely because it is still used to control the mosquito in public health programmes from where it could enter the agricultural soils and water systems and possibly find its way into crops. Presence of endosulfan in the present study is due to use of endosulfan in almost every crop in Gujarat, India among the OC pesticides after banning of use of DDT and HCH in 1993.

The study revealed that tomato was found contaminated with all the pesticides. Residues of phorate (1.97-3.07 µgg^{-1}), malathion (5.94-7.07), quinalphos (0.54-0.60 µgg^{-1}), profenophos (8.20-8.72 µgg^{-1}), pendamethalin (0.48-0.59 µgg^{-1}), aldrin (0.012-0.018 µgg^{-1}), captafol (1.37-1.48 µgg^{-1}), permethrin (0.13-0.18 µgg^{-1}) and cypermethrin (0.38-0.44 µgg^{-1}) were detected in tomato. The results obtained from the present study are consistent with an earlier study that shows residues of these pesticides are present in different vegetables [11-13, 20-23].

Effects of household processing

Among household processes washing process reduced the pesticide residues by 1.74-83.87 percent. Maximum reduction of residue was observed in case of cypermethrin, captafol and parathion where the residues decreased to the extent of 83.87, 64.78 and 54.14 percent by washing process respectively. In the present study washing was found effective in the decontamination of pesticide residues as it depends on a number of factors like, location and age of residues, water solubility, temperature and type of washing solution. In earlier studies also, effect of these factors were observed in different vegetables by various researchers [11- 13, 20-23]. Washing found comparatively less effective in reducing the residue of phorate (9.91) and quinalphos (1.74).

Cooking was observed to be more effective in reducing the residues. By this process, reduction of residues of six pesticides was observed in the range of 38.40-90.15 percent. The great variation in reduction of residues by boiling/cooking was observed which may be attributed to the rates of degradation and volatilization of residues as the concentration of residues increases by heat involved in boiling/cooking. Maximum reduction was observed in the case of cypermethrin, p,p' DDT and phorate where the residues decreased to the extent of 90.15, 82.54 and 74.50 percent respectively. Holland *et al.*, [15, 21-23] reported appreciably reduction in pesticide residues in different commodities by using different processing methods. Hence, the present results are in consistent with the earlier results.

Table-1: Effect of processing on pesticide residues (µg g^{-1}) in tomato

Sr. no	Name of Pesticide	Raw (Mean)	Washing (Mean) [% Reduction]	Cooking (Mean) [% Reduction]
1	Phorate	1.97-3.07 (2.22)	1.78-2.84 (2.00) [9.91]	0.52-0.67 (0.566) [74.50]
2	Malathion	5.94-7.07 (6.212)	3.60-4.70 (3.88) [37.54]	3.40-4.65 (3.768) [39.34]
3	Parathion	13.20-15.25 (14.38)	6.27-6.81 (6.594) [54.14]	5.29-5.45 (5.372) [62.64]
4	Quinalphos	0.54-0.60 (0.576)	0.54-0.58 (0.566) [1.74]	0.29-0.33 (0.308) [46.53]
5	Profenophos	8.20-8.72 (8.53)	4.20-4.55 (4.404) [48.37]	2.44-2.85 (2.676) [68.63]
6	Pendamethalin	0.48-0.59 -0.526	0.39-0.47 (0.422) [19.77]	0.31-0.34 (0.324) [38.40]
7	Aldrin	0.012-0.018 (0.0144)	0.007-0.0084 (0.0077) [46.53]	0.0015-0.006 (0.0044) [69.44]
8	P,p' DDT	0.0065-0.0078 (0.0071)	0.004-0.0045 (0.004) [40.85]	0.0011-0.0014 (0.00124) [82.54]
9	Captafol	1.37-1.48 (1.414)	0.39-0.58 (0.498) [64.78]	0.35-0.44 (0.398) [71.85]
10	Permethrin	0.13-0.18 (0.152)	0.10-0.15 (0.124) [18.42]	0.012-0.09 (0.0624) [58.95]
11	Cypermethrin	0.38-0.44 (0.408)	0.029-0.09 (0.0658) [83.87]	0.021-0.055 (0.0402) [90.15]

CONCLUSION

It can be concluded that processing sustainably lowers the residues of pesticides in tomato. It was found that washing and cooking process minimized the pesticide residues of eleven pesticides in the range of 1.74-64.78 and 38.40-90.15 percent respectively. The percentage reductions in the present study are supported by both early and most recent publications. These reductions are extremely important in evaluating the risk associated with ingestion of pesticide residues, especially in vegetables, which are eaten by almost all income groups' people. The present study showed that cooking was found more effective than washing.

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