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Health Risk Assessment on Pesticide Residues in Drinking Water in Shenzhen

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Abstract

Objectives: To conduct health risk assessment of drinking water pesticide residues and its annual trend analysis in Shenzhen City. **Methods:** The samples of product water, pipe water and secondary supply water from 2011 to 2013 were collected and analyzed. The evaluation models of health risk assessments for children and adults on the 12 non-carcinogenic materials (namely heptachlor, pentachlorophenol, hexachlorocyclohexane, hexachlorobenzene, DDT, malathion, glyphosate, dimethoate, bentazone, atrazine, chlorothalonil, furadan) were recommended by the U.S. Environmental Protection Agency (US EPA). **Results:** The results showed that the maximums of the measured indicators in the above were controlled in accordance with the National Health Standards (GB5749—2006) published by Ministry of Health in China. The adults and children's health indices (HIs) of the 12 non-carcinogenic materials were greater than 1 (2.323 - 6.312). Dimethoate residue in factory and peripheral water was the largest risks of harm among the non-carcinogenic pollutants measured. And its HI_i (Its HI_i) was also greater than 1 (1.995 - 5.094) and followed by hexachlorobenzene and heptachlor. Annual rising trend on health risk of the 12 pesticide residues indicated that their HI_T on adults was 2323.18×10^{-3} in 2011, 2340.18×10^{-3} in 2012 and 2431.97×10^{-3} in 2013, and HI_T on children was 2965.07×10^{-3} in 2011, 2986.77×10^{-3} in 2012 and 3103.93×10^{-3} in 2013, respectively. This study also suggested that the average risk of peripheral water samples (HI_T was equal to 2619.64×10^{-3}) was greater than the factory samples' (HI_T was the same as 2366.92×10^{-3}), and the children's health risk was greater than the adults'. **Conclusions:** Health risks of drinking water pesticide residues in Shenzhen have exceeded the threshold values. The dimethoate was the main hazard and had been rising annually, and the children's health risk was greater than the adults'.

Analysis

1. Introduction

The term "pesticide" is a general term that includes many compounds used for a wide variety of purposes to control a range of living organisms. It includes animal and bird repellents, food storage protectants, insecticides and acaricides, mould-killing substances, antifouling products, plant growth regulators, compounds used to prevent the growth of lichen and mosses etconbuildings, rodenticides, soil sterilants, weed killers and wood preservatives and so on [1] [2]. In the recent years, the pesticide industry in China has developed very rapidly, and the pesticide production has been the second largest producer in the world for many years, but overtook the United States and became the largest pesticide producer by breaking through the million tons in the world in 2005 [2] [3]. Some parts of the environmental pesticide residues couldn't be removed absolutely by the modern drinking water treatment technologies, such as precipitation, filtration and disinfection, which remained in the surface and groundwater after the atmosphere-water-soil-biosphere circulation [4] [5] [6] [7]. Organochlorine (OC) pesticides are the one reason of hepatocirrhosis swelling, which could accumulate in human body fat and induce the liver enzymes. Organophosphate (OP) pesticides are the neurotoxic substance, which could cause neurological dysfunction, tremor, delirium, speech disorders, etc. [5] [6]. Just as the following report, concentrations of the 12 kinds of heptachlor chemicals in drinking water have been measured and their health risk assessments have been analyzed from 2011 to 2013 in this study.

2. Materials and Methods

2.1. Experimental

Drinking water samples were collected in Shenzhen from January 2011 to December 2013, with a total of 84 factory samples, 11 peripheral samples and 1 secondary supplement samples to analyze their pesticide residues (see **Figure 1**). Detection indexes for these samples included heptachlor, pentachlorophenol, hexachlorocyclohexane, hexachlorobenzene, DDT, malathion, glyphosate, dimethoate, bentazone,Atrazine, chlorothalonil, carbofuran, etc. The Shenzhen Municipal Water Affairs Bureau is responsible for water quality detection of secondary supplement according to the duties of the functional departments of the Shenzhen Municipal Government. Collection, storage and testing of all water samples were in accordance with the Inspection methods of drinking water (GB 5750-2006) [8] for evaluation.

Heptachlor, hexachlorocyclohexane, hexachlorobenzene, DDT and chlorothalonil in drinking water were detected by GC2010 Gas Chromatograph which was made by Shimadzu Corporation in Japan. Pentachlorophenol by GC-17A
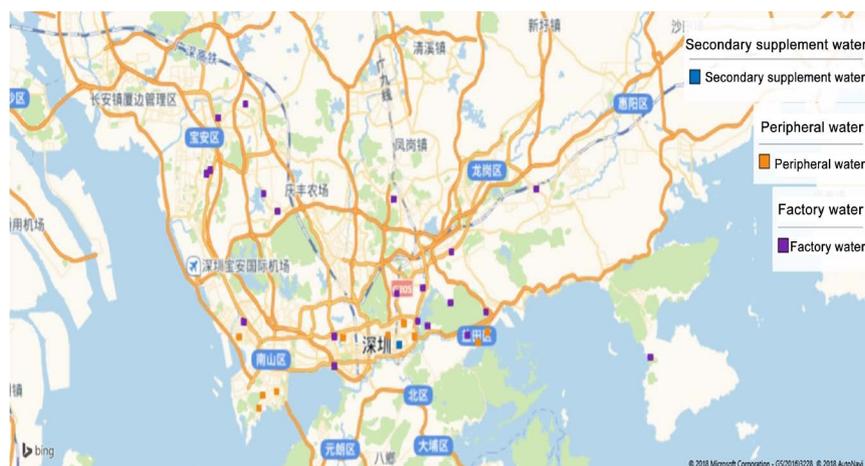
Figure 1. Sampling sites.

Gas Chromatograph of Shimadzu Corporation in Japan. Malathion and LeA by Agilent 7890 Gas Chromatograph of USA. Glyphosate, extinction Grass pine and atrazine by QTRAP 5500 liquid chromatography tandem mass spectrometry Instrument of American ABI company. Carbofuran by Agilent 1100 liquid color Spectrometer of USA. The test results were in accordance with the Standards for Drinking Water Hygiene (GB 5749-2006) for evaluation.

In view of data quality assurance, each sample was analyzed in triplicate and after every 10 samples two standards; one blank and another of 2.5 µg/L of respective metal were analyzed on atomic absorption. The reproducibility was found to be at 95 percent of confidence level, the relative standard deviations (RSD) were all less than 15 percent, conforming to the requirements of the US EPA which RSD is less than 30 percent [8]. Therefore, the average value of each water sample was used for further interpretation.

2.2. Pesticide Residues Health Risk Assessment

Pesticide residues health risk assessment was used to evaluate risks of heptachlor, pentachlorophenol, hexachlorocyclohexane, hexachlorobenzene, DDT, malathion, glyphosate, dimethoate, bentazone, Atrazine, chlorothalonil, carbofuran. Health risk index (HI) means average personal annual health risk values due to exposure to individual non-genetic toxicant. The calculation method is shown in formula (1) to (3) [4] [5] [6]:

$$HIT = \sum HIi \tag{1}$$

$$HIi = (CDIi/RfDi) \tag{2}$$

$$CDIi = (Ci \times IR \times EF \times ED)/(BW \times AT) \tag{3}$$

HIT—average personal health risk of total non-genetic toxicants by drinking, annual; HIi—average personal health risk of individual non-genetic toxicant by drinking, annual; CDIi—chronic daily exposure dose through drinking, mg/(kg·d); RfDi—reference dose of individual non-genetic toxicant by drinking, mg/(kg·d); Ci—concentration of individual measured substance in water, mg/L;

IR—intake rate of water, adult 2.2 L/d, children 1.0 L/d [9]; EF—exposure frequency (d/y); ED—exposure duration (y); BW—body weight (kg) [10]; AT—average exposure day (d).

There are no basic data on exposure parameters in China at present, so we generally introduced the RfDi, from USEPA to conduct risk assessment (see Table 1) [11].

2.3. Evaluation Standard

Testing pesticide residues of water samples were in accordance with the Inspection methods of drinking water (GB 5750-2006) [8] for evaluation. HIi > 1 indicated a potential of the pollutant for an adverse effect on human health or the necessity for further study. HIi < 1 indicated a relative safety for human health.

2.4. Data Analysis

Data entry for water quality monitoring database were performed by Excel 2003. The all data were analyzed descriptive statistically by software program (SPSS 13.0 for windows). After the normality test, the water quality monitoring data was non-normally distributed, so the range and median were used. There was statistical significance if P value was below to 0.05.

3. Results

1) Determination quality inspection indicators on pesticide residues in drinking water

Testing the pesticide residues of water samples, the minimum detection limit and the recovery rate were in accordance with the Inspection methods of drinking water (GB 5750-2006) [8] for evaluation (see Table 2).

Table 1. Noncarcinogenic reference dose on pesticides in drinking water in US.

Pesticide types	Pesticides	RfD [mg/(kg·d)]
Organochlorine (OC)	Heptachlor	0.0001
	Pentachlorophenol	0.0050
	Hexachlorocyclohexane	0.0030
	Hexachlorobenzene	0.0008
	DDT	0.0200
Organophosphate (OP)	Malathion	0.0200
	Glyphosate	0.1000
	Dimethoate	0.0002
Herbicide	Bentazone	0.0300
	Atrazine	0.0350
Fungicide	Chlorothalonil	0.0150
Carbamate	Carbofuran	0.0010

Table 2. Determination parameters on pesticide residues in drinking water.

Pesticide/(mg/L)	Minimum detection limit (mg/L)	Recovery rate/%
Heptachlor	0.0004	83.0 - 97.0
Pentachlorophenol	0.001	93.6 - 98.1
Hexachlorocyclohexane	0.005	85.8 - 108.0
Hexachlorobenzene	0.00001	81.1 - 91.0
DDT	0.001	91.3 - 102.0
Malathion	0.01	87.1 - 90.0
Glyphosate	0.00015	91.8 - 99.2
Dimethoate	0.01	85.2 - 92.0
Bentazone	0.0005	97.6 - 100.0
Atrazine	0.001	89.0 - 97.0
Chlorothalonil	0.001	83.0 - 112.0
Carbofuran	0.002	81.0 - 120.0

2) Concentrations of pesticide residues in product water, pipe water and secondary supply water in Shenzhen from 2011 to 2013

The samples with a total of 84 factory water, 11 peripheral water and 1 secondary supplement water from 2011 to 2013 have been tested, and the maximum value of the samples did not exceed the limit values according to the Standards for Drinking Water Hygiene (GB 5749-2006). The 6 samples of heptachlor (Upper limit is 0.4 μg/L), 10 samples of hexachlorocyclohexane (Upper limit is 5.0 μg/L), and 5 samples of DDT (Upper limit is 1.0 μg/L) approached to the critical values (see Table 3).

3) Health risk assessment and annual trend for children and adults on the 12 pesticides in drinking water from 2011 to 2013 in Shenzhen

The HIi of children and adults on the 12 pesticides in factory water from 2011 to 2013 in Shenzhen were more than 1 (2.323 - 6.312), the HIi of dimethoate were the most important and more than 1 (1.995 - 5.094).There was an upward trend year by year on HIi, and the HIi of adults in 2011 < HIi in 2012 < HIi in 2013, and the HIi of children in 2011 < HIi in 2012 < HIi in 2013. This study also showed that the health risks of children were higher than risks of adults from the pesticides in drinking water (see Table 4, Figure 2, Figure 3).

4) Non-carcinogenic risk assessment on the 12 pesticides in factory water, pipe water and secondary supply water in Shenzhen

We have detected the median and maximum values of concentrations on the 12 pesticides in 84 factory waters, 11 pipe waters and 1 secondary supply water from 2011 to 2013, which were employed to analyze the non-carcinogenic risk assessment from pesticides. No maximum values of concentrations on secondary supply water because of only one sample. The HIT resulted from the median values of concentrations of the 12 pesticides in the pipe water were higher than

Table 3. Detection on pesticide residue in drinking water from 2011 to 2013 in Shenzhen μg/L.

Pesticide	Factory water			Peripheral water			Secondary supplement water		Standard limit value
	n	mean with SD	Range	n	mean with SD	Range	n	Median	
Heptachlor	83	0.204 ± 0.186	0.200 - 0.400	11	0.216 ± 0.168	0.200 - 0.400	1	0.200	0.4
Pentachlorophenol	84	0.617 ± 0.406	0.500 - 5.000	11	0.952 ± 0.617	0.500 - 5.000	1	5.000	9.0
Hexachlorocyclohexane	84	2.122 ± 1.745	0.200 - 5.000	11	2.514 ± 1.921	0.200 - 5.000	1	2.500	5.0
Hexachlorobenzene	53	0.071 ± 0.053	0.002 - 0.500	11	0.095 ± 0.0643	0.005 - 0.850	1	0.005	1.0
DDT	84	0.453 ± 0.319	0.200 - 1.000	11	0.543 ± 0.368	0.500 - 1.000	1	0.500	1.0
Malathion	84	6.128 ± 4.227	5.000 - 50.000	11	5.451 ± 3.817	5.000 - 10.000	1	5.000	250.0
Glyphosate	84	0.154 ± 0.106	0.080 - 0.250	11	0.128 ± 0.104	0.100 - 0.200	1	0.080	700.0
Dimethoate	83	5.014 ± 4.581	0.500 - 10.000	11	5.452 ± 4.167	5.000 - 10.000	1	5.000	80.0
Bentazone	84	0.206 ± 0.164	0.150 - 0.500	11	0.452 ± 0.282	0.160 - 2.500	1	0.250	300.0
Atrazine	84	0.277 ± 0.201	0.025 - 1.000	11	0.413 ± 0.375	0.025 - 1.000	1	0.500	2.0
Chlorothalonil	84	0.503 ± 0.452	0.500 - 1.000	11	0.542 ± 0.382	0.500 - 1.000	1	0.500	10.0
Carbofuran	84	1.011 ± 0.926	1.000 - 2.000	11	1.089 ± 0.837	1.000 - 2.000	1	1.000	7.0

Table 4. Non-carcinogenic risk assessment for children and adults on the 12 pesticides in drinking water from 2011 to 2013 in Shenzhen ($\times 10^{-3}$).

(a)

Pesticides	The risk of adults					
	2011		2012		2013	
	Median	Maximum	Median	Maximum	Median	Maximum
Heptachlor	159.67	239.50	159.67	159.67	239.50	319.34
Pentachlorophenol	9.58	79.83	12.77	15.97	9.58	79.83
Hexachlorocyclohexane	42.58	133.06	66.53	66.53	66.53	133.06
Hexachlorobenzene	6.79	9.98	0.20	0.20	8.08	49.90
DDT	1.60	2.00	2.00	2.00	2.04	3.99
Malathion	23.95	199.59	19.96	19.96	24.35	199.59
Glyphosate	0.19	0.20	0.20	0.20	0.06	0.12
Dimethoate	1995.85	3991.71	1995.85	1995.85	1995.85	3991.71
Bentazone	0.43	0.45	0.45	0.45	0.69	1.33
Atrazine	0.05	0.06	0.06	0.06	1.14	2.28
Chlorothalonil	2.66	2.66	2.66	2.66	2.71	5.38
Carbofuran	79.83	79.83	79.83	79.83	81.43	159.67
Total	2323.18	4738.86	2340.18	2343.37	2431.97	4946.18

(b)

Pesticides	The risk of children					
	2011		2012		2013	
	Median	Maximum	Median	Maximum	Median	Maximum
Heptachlor	203.78	305.68	203.78	203.78	305.68	407.57
Pentachlorophenol	12.23	101.89	16.30	20.38	12.23	101.89

Continued

Hexachlorocyclohexane	54.34	169.82	84.91	84.91	84.91	169.82
Hexachlorobenzene	8.66	12.74	0.25	0.25	10.32	63.68
DDT	2.04	2.55	2.55	2.55	2.60	5.09
Malathion	30.57	254.73	25.47	25.47	31.08	254.73
Glyphosate	0.24	0.25	0.25	0.25	0.08	0.15
Dimethoate	2547.31	5094.61	2547.31	2547.31	2547.31	5094.61
Bentazone	0.54	0.58	0.58	0.58	0.88	1.70
Atrazine	0.07	0.07	0.07	0.07	1.46	2.91
Chlorothalonil	3.40	3.40	3.40	3.40	3.46	6.86
Carbofuran	101.89	101.89	101.89	101.89	103.93	203.78
Total	2968.07	6048.21	2986.77	2990.85	3103.93	6312.81
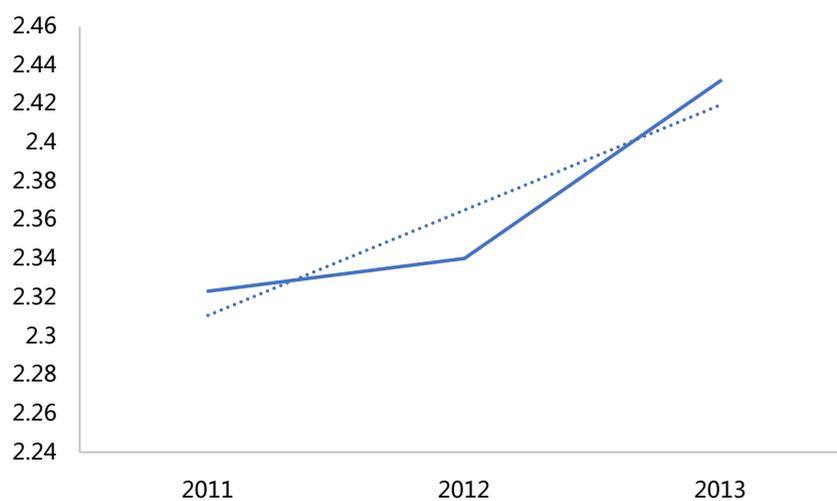
Figure 2. Annual rising trend on health risk of the 12 pesticide residues indicated that their HIT on adults.
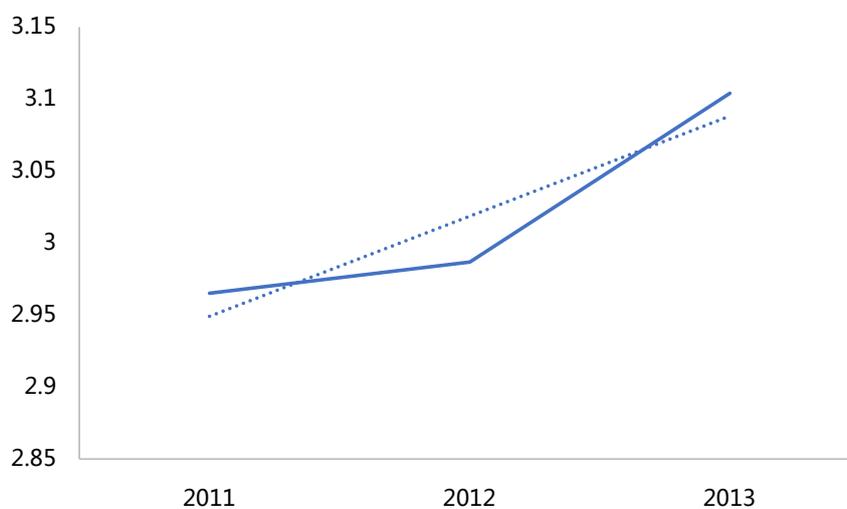
Figure 3. Annual rising trend on health risk of the 12 pesticide residues indicated that their HIT on children.

that in the factory water (2.619 vs 2.366), but lower HIT by the maximums values in pipe water comparing to that in factory water (5.240 vs 5.395). Dimethoate was the main resource of in factory water (HI = 2.003 - 3.991) and pipe water (HI = 2.177 - 3.991) on non-carcinogenic risk assessment. The total HIT from the median values of concentrations on the rest 11 pesticides except dimethoate was less than one (HI = 0.363 - 0.442), but the total HIT greater than one by analysing their maximum values of concentrations of the 11 pesticides either in factory water or in pipe water (HI = 1.249 - 1.403). Therefore, hexachlorobenzene and heptachlor ranked the second and third among the 12 pesticides on health risk (see Table 5).

4. Discussion

The study indicated that the maximum values of all the samples measured do not exceed the limit specified in Standards for Drinking Water Hygiene (GB 5749-2006), but the annual non-carcinogenic risks of adults and children on 12 pesticide residues such as heptachlor were greater than 1 (2. 323 - 6. 312) by the US EPA recommended risk assessment model. Meanwhile, the approximate 80% of the risks came from dimethoate, of which the annual personal non-carcinogenic risks were greater than 1 (1.995 - 5.094) with a slow upward trend year by year, and which risks on children were higher than those on adults. The reasons of this could be related to the increase in the production and use of pesticides in China, and the dimethoate was the main pesticide of fruit trees and vegetables

Table 5. Non-carcinogenic risk assessment on the 12 pesticides in factory water, pipe water and secondary supply water in Shenzhen ($\times 10^{-3}$).

Pesticides	The risk of factory water		The risk of pipe water		The risk of secondary supply water	
	Median	Maximum	Median	Maximum	Median	Maximum
Heptachlor	165.26	319.34	174.04	319.34	159.67	
Pentachlorophenol	9.88	79.83	15.25	79.83	79.83	
Hexachlorocyclohexane	56.50	133.06	67.01	133.06	66.53	
Hexachlorobenzene	21.16	498.96	69.66	498.96	0.50	
DDT	1.82	3.99	2.18	3.99	2.00	
Malathion	24.47	199.59	21.77	39.92	19.96	
Glyphosate	0.12	0.20	0.10	0.16	0.06	
Dimethoate	2003.04	3991.71	2177.48	3991.71	1995.85	
Bentazone	0.55	1.33	1.21	6.65	0.67	
Atrazine	0.64	2.28	0.95	2.28	1.14	
Chlorothalonil	2.69	5.32	2.90	5.32	2.66	
Carbofuran	80.79	159.67	87.10	159.67	79.83	
Total	2366.92	5395.27	2619.64	5240.89	2408.70	
Total except dimethoate	363.88	1403.56	442.16	1249.18	412.85	

in the south of China [11] [12] [13] [14]. The reason of mean risk values of pipe water being greater than that of factory water is related to the secondary chlorination in the water transportation and the pipe material and the long pipeline transportation [4] [10].

Crentsil *et al.* [3] reported the organochlorine pesticide residues and health risk assessment results of Accra Metropolis in Ghana, which indicated that residual levels of pesticides on self-produced papaya, tomatoes and imported apples were <0.01 to 0.11 μg/g, and the risk values of heptachlor, epoxy heptachlor, endrin aldehyde and endrin ketone were greater than 1, and the risk values of children were greater than those of adults.

Guohong Zhou *et al.* [15] reported that the non-carcinogenic health risk index of trihalomethane, a by-product of drinking water disinfection in Shenzhen, was less than 1, but its carcinogenic risk was close to the critical value, and the risk values of cancer in 3 factory waters exceeded the critical value. Our previous research on the health risk analysis of three kinds of toxic substances, such as arsenic and other toxic substances in drinking water in Shenzhen, indicated that the health risks of these 15 chemicals were still in control by the International Radiation Protection Committee, and the risk of carcinogens was greater than that of non-carcinogens, and the risk of children was higher than that of adults [16].

Of course, this study was referenced to the methods of pesticide residues in drinking water exposure to human health risk assessment in USEPA, and we only considered the exposure way of water intake average, and other exposure ways, such as food intake, swimming, bathing, and skin exposure pathway, were out of consideration. The reference dose selection, ethnicity, living habits, and the mechanism of various pesticides are different because of the lack of exposure parameter standards in China. Therefore, there may be some bias in the results of this evaluation [17] [18] [19]. The comprehensive study of environmental exposures to pesticides, including food and water intake, skin and respiratory exposures, is needed in the further.

Acknowledgements

Conflicts of Interest

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