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Health Risk from Exposure of Organic Pollutants Through Drinking Water Consumption in Nanjing, China

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Abstract Human health risk analysis for 24 organic pollutants in drinking water of Nanjing was conducted. For non-carcinogenic risk, the 95th percentile hazard quotient (HQ) values of pollutants were all less than the unacceptable level of one. Considering the lifetime carcinogenic risk (LCR), however, the 95th percentile LCR values of 2,6-dinitrotoluene (1.30E-05), benzo(b)fluoranthene (3.10E-05), benzo(a)pyrene (3.37E-05) and dibenz(a,h)anthracene (2.09E-05) exceeded the unacceptable level of 1.00 E-05. These results suggest that organic pollutants in drinking water of Nanjing might pose potential lifetime carcinogenic risk for local consumers, and concerted efforts are required to ensure safety of consumers.

Keywords Organic pollutant · Non-carcinogenic risk · Carcinogenic risk · Drinking water

Drinking water pollution is one of the most important environmental problems in the world. Organic pollutants in drinking water, which can cause adverse effects on the human and wildlife, have received great attention. In China, during the past two decades, researchers focused their attention on quantitative investigation of organic pollutants in drinking water (Zhao et al. 2004; Shao et al. 2005). Nanjing, which has a population of 6.4 million inhabitants, is one of the most important industrial cities in the south-east China (31° and 32°N, 118° and 119°E). However, comprehensive determination of organic pollutants in drinking water of Nanjing has not been reported in previous literatures.

For effective evaluation of water quality, it is important to identify potential human health effects of organic pollutants in drinking water. Traditional method for evaluating health effects is to directly compare the measured values with permissible limits, but it is not sufficiently reliable to provide detailed hazard levels and identify contaminants of the most concern. Human health risk assessment is an effective approach to determine health risk levels posed by specific chemicals. This method has been applied to assess the potential adverse health effects exposing to contaminated water (Hartley et al. 1999; Sun et al. 2007; Kavcar et al. 2009).

This study was carried out as a survey and risk analysis for 24 organic pollutants in drinking water of Nanjing. The main objectives of this study were (1) to determine the levels of the organic pollutants in the drinking water of Nanjing, (2) to evaluate their potential health risk and quantify the uncertainty of risk assessment.

Materials and Methods

Drinking water sampling was performed in July, September, October, December 2007 and March, June, September, December 2008 from effluent water of the largest tap water plant (BHK plant) in Nanjing, which provides 50% tap water for Nanjing city. A total of 32 water samples were screened for 24 organic pollutants, including 12 polycyclic aromatic hydrocarbons (PAHs) (acenaphthylene, anthracene, benz(a) anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo (k)fluoranthene, chrysene, dibenz(a,h)anthracene, fluorene,

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phenanthrene, pyrene, indeno(1,2,3-cd)pyrene), 6 phthalates (PAEs) (bis(2-ethylhexyl)adipate, bis(2-ethylhexyl)phthalate, butyl benzyl phthalate, dibutyl phthalate, diethyl phthalate, dimethyl phthalate) and six other organic pollutants (isophorone, hexachlorocyclopentadiene, 2,4-dinitrotoluene, 2,6-dinitrotoluene, hexachlorobenzene, pentachlorophenol). The standard organic pollutants were obtained from Supelco (Bellefont, PA, USA). A liquid–liquid extraction method (EMC 2006) was used to extract the target contaminants in water samples. Quality assurances were conducted according to our previous reports (Wu et al. 2009a, b). Detection limits for all compounds ranged from 0.4 to 2 ng L^{-1} (a signal-to-noise ratio of 3).

Human health risk assessment is based on reliable exposure pathways of contaminants (USEPA 1989, 1997). The organic pollutants in water exposed to human body mainly through the following pathways: (1) direct ingestion of water consumption, (2) dermal absorption of contaminants in water adhered to exposed skin. So both pathways were studied in this study.

The expose doses through ingestion and dermal absorption pathway were calculated by use of Eq. 1 and 2 respectively, which were adapted from US Environmental Protection Agency (USEPA 1989):

$$D_i = \frac{C_w \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

$$D_d = \frac{C_w \times SA \times K_p \times ET \times EF \times ED \times CF_1 \times CF_2}{BW \times AT} \quad (2)$$

where D_i (μg kg^{-1} day^{-1}), exposure dose through ingestion of water; D_d (μg kg^{-1} day^{-1}), exposure dose through dermal absorption; C_w (μg L^{-1}), concentration of organic pollutant in drinking water; IR (L day^{-1}), ingestion rate; EF (day $year^{-1}$), exposure frequency; in this study, 365 day $year^{-1}$; ED (year), exposure duration, in this study, equal to the lifetime; BW (kg), average body weight; AT (day), averaging time, for non-carcinogens and carcinogens, ED × 365 days; SA (cm^2), exposed skin area; K_p (cm h^{-1}), dermal permeability coefficient; ET (min day^{-1}), exposure time during bathing and shower; CF_1, unit conversion factor, L 1,000 cm^{-3}; CF_2, unit conversion factor, h 60 min^{-1}.

The hazard quotient (HQ) was calculated by Eq. 3 to estimate non-carcinogenic risk (USEPA 1989):

$$HQ = D/RfD \quad (3)$$

where D (μg kg^{-1} day^{-1}): the exposure dose obtained from the Eq. 1 and 2.

RfD (μg kg^{-1} day^{-1}): the reference dose of the contaminant. The ingestion reference dose (RfD_i) values were obtained from the USEPA (2008). The RfD_i was multiplied by a gastrointestinal absorption factor (ABS_g) to yield the corresponding dermal absorption reference dose (RfD_d) (USEPA 2004).

To assess the overall non-carcinogenic risk posed by all chemicals, the HQ calculated for each chemical was summed and expressed as hazard index (HI) by the Eq. 4 (USEPA 1989):

$$HI = HQ_1 + HQ_2 + \cdots + HQ_n \quad (4)$$

Lifetime carcinogenic risk (LCR) associated with ingestion and dermal exposure was calculated using the following Eq. 5 (USEPA 1989):

$$LCR = D \times SF \quad (5)$$

where LCR, the probability of developing cancer over a lifetime as a result of exposure to a contaminant; D (μg kg^{-1} day^{-1}), the exposure dose obtained from the Eq. 1 and 2; SF (μg kg^{-1} $day^{-1})^{-1}$, the carcinogenic slope factor of the contaminant.

The ingestion slope factor (SF_i) values were obtained from the USEPA (2008). The SF_i was divided by a gastrointestinal absorption factor (ABS_g) to yield the corresponding dermal absorption slope factor (SF_d). Because the SF obtained from USEPA was derived assuming a body weight of 70 kg and 70 year lifetime, which was different from the actual conditions of the exposure population in this study. The SF values used in this study were adjusted according to the recommended method by USEPA (1997).

Risk index (RI) of all chemicals were calculated by summing the LCR of individual chemical using the Eq. 6 to assess the overall potential for carcinogenic risk effects (USEPA 1989):

$$RI = LCR_1 + LCR_2 + \cdots + LCR_n \quad (6)$$

Uncertainty existing in health risk assessment mainly arises from the estimation of exposure level and toxicity values of the analysis. Because of limited reference data about toxicity values, the recommended toxicity values (RfD, K_p and SF) for contaminants from USEPA (2008) were directly applied. The variables of C_w, IR, ED, SA, BW and ET were evaluated on the basis of the local statistical data and USEPA reference data. Lognormal distribution was considered as the probability distribution of above variables according to the reports of literature (Chen and Liao 2006). Monte Carlo simulation (n = 50,000) was employed to estimate the uncertainties of the risk assessment. The Monte Carlo simulation was implemented by R language (Version 2.7.0).

Results and Discussion

The summary statistics of analytical results including mean, standard deviation, median and rang values for organic pollutants are presented in Table 1. The concentrations of ∑PAHs ranged from 23.7 to 399.1 ng L^{-1} with

Table 1 Summary statistics of organic pollutants in drinking water (ng L^{-1})

	Min	Median	Mean	SD	Max
Isophorone	ND	ND	3.06	6.80	25.05
Hexachlorocyclopentadiene	ND	15.64	15.12	10.67	34.56
Dimethyl phthalate	ND	21.43	25.52	25.53	84.70
2,6-Dinitrotoluene	ND	252.01	300.29	191.00	718.34
Acenaphthylene	ND	ND	1.91	2.49	7.31
2,4-Dinitrotoluene	ND	ND	5.24	11.14	36.84
Diethyl phthalate	ND	23.90	28.76	31.18	133.91
Fluorene	ND	15.18	37.21	48.52	205.64
Hexachlorobenzene	ND	ND	4.06	10.39	38.73
Pentachlorophenol	ND	ND	8.98	20.06	54.89
Phenanthrene	5.57	27.61	36.52	25.02	90.77
Anthracene	ND	28.76	28.80	24.18	79.82
Dibutyl phthalate	10.17	1,734.29	2,363.55	1,902.09	5,513.44
Pyrene	ND	7.54	8.01	6.53	27.67
Butyl benzyl phthalate	ND	ND	3.18	5.89	19.48
Bis(2-ethylhexyl)adipate	ND	14.18	234.19	371.83	1,188.52
Chrysene	ND	1.80	10.69	16.06	64.39
Benzo(a)anthracene	ND	2.91	7.96	12.72	45.73
Bis(2-ethylhexyl)phthalate	488.75	2,014.18	1,914.43	904.29	3,672.72
Benzo(b)fluoranthene	ND	11.77	53.65	67.77	196.05
Benzo(k)fluoranthene	ND	2.32	22.91	47.10	171.94
Benzo(a)pyrene	ND	3.22	5.98	7.56	28.39
Indeno(1,2,3-cd)pyrene	ND	ND	7.42	14.49	46.07
Dibenz(a,h)anthracene	ND	ND	13.90	39.94	149.78

SD standard deviation, *ND* no detection

a mean value of 234.9 ng L^{-1}. Total PAEs levels in drinking water ranged from 655.5 to 6,080.6 ng L^{-1} with a mean value of 4,569.6 ng L^{-1}. The concentration ranges of $\sum$PAHs, $\sum$PAEs and other organic pollutants found in this study area were similar to the levels found in drinking water in other regions (Chen 2007; Shao et al. 2008). However, direct comparison of literature data obtained from other investigations was complicated by the disparity in sampling protocols and digestion procedures of the samples. So, this comparison needs to be treated with caution.

In the risk assessment process, ED and BW were calculated according to the data of local residents obtained from the Ministry of Health of the People's Republic of China (MHPRC 2007). Since the reference data of IR, SA and ET for local residents were limited, they directly derived from the reference values of USEPA (1997). The mean and standard deviation values of population parameters are shown in Table 2. Since the toxicity values for some pollutants are limited, the health risks of pollutants, which could obtain the efficient toxicity values, are analyzed.

The HQ of individual pollutant and HI obtained from Monte Carlo simulation are listed in Table 3. Because Monte Carlo simulation covers all possible scenarios which might not be encountered during sampling, some of highest model values may be overestimated. Hence, in this study, the 95th percentile values were used as high-end estimates instead of the maxima in the risk characterization (Masago et al. 2006; Kavcar et al. 2009). In general, if HQ or HI value is greater than one, it indicates potential adverse health effects and needs for further study.

For ingestion pathway, the 95th percentile HQ values of each pollutant were all less than one, and the 95th percentile HI value was 2.42E-02. Considering dermal adsorption pathway, the 95th percentile HQ and HI values were also below unity. These results indicate that measured pollutants in drinking water may pose little or no non-carcinogenic health risk for local consumers though ingestion and dermal adsorption exposure.

Lifetime carcinogenic risk (LCR) is defined as the incremental probability that an individual will develop cancer during ones lifetime due to chemical exposure under specific scenarios (Chen and Liao 2006). Under most regulatory programs, a LCR value over 1.00E-05 indicates potential carcinogenic risk (De Miguel et al. 2007).

LCRs of contaminants by ingestion and dermal exposure were calculated and showed in Table 4. For ingestion pathway, the 95th percentile LCR values of each pollutant were all lower the 1.00E-05 with the exception of 2,6-dinitrotoluene (1.30E-05). For dermal adsorption pathway, the LCR

Table 2 Population parameters as random variables

Definition	Units	Mean	SD
Ingestion rate (IR)[a]	L day^{-1}	1.43	0.64
Exposure duration (ED)[b]	year	74.06	2.84
Surface area (SA)[a]	cm^2	20,091	1,912
Body weight (BW)[b]	kg	57.3	5.59
Exposure time during bathing and shower (ET)[b]	min day^{-1}	17.21	9.51

[a] Adapted from USEPA (1997)

[b] Adapted from the MHPRC (2007)

Table 3 Non-carcinogenic risk of organic pollutants

	Ingestion				Dermal			
	Minimum	Mean	95%	Maximum	Minimum	Mean	95%	Maximum
Isophorone	8.44E-10	3.83E-07	1.45E-06	1.79E-05	–	–	–	–
Hexachlorocyclopentadiene	2.25E-06	6.37E-05	1.71E-04	6.60E-04	–	–	–	–
2,6-Dinitrotoluene	4.42E-04	7.52E-03	1.91E-02	7.31E-02	2.24E-06	7.56E-05	2.07E-04	7.47E-04
2,4-Dinitrotoluene	1.98E-07	6.66E-05	2.39E-04	5.67E-03	3.67E-09	9.90E-07	3.61E-06	1.31E-04
Diethyl phthalate	1.29E-08	8.87E-07	2.78E-06	1.67E-05	1.55E-10	1.57E-08	5.02E-08	4.92E-07
Fluorene	2.68E-07	2.32E-05	7.58E-05	6.63E-04	1.90E-07	3.28E-05	1.06E-04	1.06E-03
Hexachlorobenzene	2.11E-07	1.22E-04	5.02E-04	3.93E-03	2.88E-07	4.26E-04	1.69E-03	2.67E-02
Pentachlorophenol	4.33E-08	6.58E-05	2.47E-04	6.23E-03	1.51E-08	5.18E-05	2.06E-04	2.86E-03
Anthracene	5.61E-08	2.37E-06	6.66E-06	4.25E-05	4.94E-08	1.78E-06	5.15E-06	2.80E-05
Dibutyl phthalate	2.38E-05	5.98E-04	1.63E-03	1.93E-02	1.62E-06	7.32E-05	2.13E-04	1.20E-03
Pyrene	2.19E-07	6.81E-06	1.90E-05	7.25E-05	9.29E-07	2.90E-05	8.43E-05	6.35E-04
Bis(2-ethylhexyl)adipate	7.10E-08	9.49E-06	3.24E-05	2.36E-04	6.72E-10	1.39E-07	5.11E-07	6.71E-06
Bis(2-ethylhexyl)phthalate	1.42E-04	2.38E-03	5.55E-03	1.68E-02	9.50E-05	1.28E-03	3.24E-03	2.12E-02
HI	1.12E-03	1.09E-02	2.42E-02	8.35E-02	1.44E-04	1.98E-03	4.91E-03	2.96E-02

Table 4 Carcinogenic risk of organic pollutants

	Ingestion				Dermal			
	Minimum	Mean	95%	Maximum	Minimum	Mean	95%	Maximum
Iophorone	1.60E-13	7.27E-11	2.76E-10	3.41E-09	–	–	–	–
2,6-Dinitrotoluene	3.00E-07	5.11E-06	1.30E-05	4.97E-05	1.52E-09	5.14E-08	1.41E-07	5.08E-07
2,4-Dinitrotoluene	2.69E-10	9.06E-08	3.25E-07	7.71E-06	4.99E-12	1.35E-09	4.91E-09	1.78E-07
Hexachlorobenzene	2.70E-10	1.57E-07	6.42E-07	5.03E-06	3.69E-10	5.45E-07	2.17E-06	3.42E-05
Pentachlorophenol	1.56E-11	2.37E-08	8.88E-08	2.24E-06	5.44E-12	1.87E-08	7.42E-08	1.03E-06
Bis(2-ethylhexyl)adipate	5.11E-11	6.83E-09	2.33E-08	1.70E-07	4.84E-13	1.00E-10	3.68E-10	4.83E-09
Chrysene	1.05E-11	1.94E-09	6.70E-09	6.04E-08	2.48E-11	1.22E-08	4.23E-08	1.30E-06
Benz[a]anthracene	9.06E-10	1.38E-07	4.92E-07	3.51E-06	4.48E-09	8.61E-07	3.09E-06	2.23E-05
Bis(2-ethylhexyl)phthalate	3.98E-08	6.66E-07	1.55E-06	4.71E-06	2.66E-08	3.60E-07	9.08E-07	5.95E-06
Benzo[b]fluoranthene	1.34E-08	9.86E-07	3.24E-06	1.81E-05	9.49E-08	8.91E-06	3.10E-05	2.16E-04
Benzo[k]fluoranthene	1.55E-10	4.06E-08	1.53E-07	1.84E-06	2.03E-09	6.29E-07	2.51E-06	2.65E-05
Benzo[a]pyrene	8.36E-09	1.09E-06	3.46E-06	3.12E-05	1.19E-07	1.01E-05	3.37E-05	2.58E-04
Indeno[1,2,3-cd]pyrene	1.57E-10	1.38E-07	5.16E-07	5.56E-06	1.79E-09	1.74E-06	6.61E-06	5.09E-05
Dibenz[a,h]anthracene	1.99E-09	2.69E-06	9.96E-06	1.95E-04	3.13E-09	5.28E-06	2.09E-05	3.48E-04
RI	1.19E-06	1.11E-05	2.54E-05	2.08E-04	2.12E-07	2.84E-05	9.91E-05	9.31E-04

values of benzo[b]fluoranthene (3.10E-05), benzo[a]pyrene (3.37E-05) and dibenz[a,h]anthracene (2.09E-05) exceeded the acceptable level. Considering the integrated carcinogenic health risk induced by combinative effects of pollutants, the RI values through the both exposure pathways were all higher than the 1.00E-05 (Table 4). These results indicate that the organic pollutants in the drinking water might pose potential carcinogenic health risk on the local consumers.

Although the Monte Carlo simulation was used to evaluate the uncertainties of risk assessment, there are many areas in which further research could be conducted to reduce the uncertainties. Firstly, periodic determination of pollutant contents in the tap water plant is needed to reflect the pollutant levels and characterize the distribution types of concentrations more comprehensively. Secondly, the toxicity values, such as Kp and SF, might have high influence on risk estimates (Chen and Liao 2006). Therefore, there is a need to conduct a more extensive characterization of the distribution of toxicity values for given pollutants, which would require the collection of more detailed information on toxicity data of contaminants from references. Finally, in this study, the IR and ET values were directly obtained from the USEPA, which might not be specific to Chinese. It is assumed that persons in Nanjing have a bath everyday, which could overestimate the health risk values through dermal exposure. Questionnaires could be a good method to collect the detailed information on exposure population and reduce variability of the results.

In conclusion, this study provides a primary health risk of organic pollutants in drinking water of Nanjing by Monte Carlo simulation. The results show that organic pollutants in drinking water of Nanjing might exert potential carcinogenic risk for local residents under worst-base scenario. The uncertainties notwithstanding, on basis of the monitoring data, health risk assessment might be a very useful tool to reveal the true meaning and relevance of organic pollutants in drinking water.

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