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Health risk assessment of haloacetonitriles in drinking water based on internal dose☆

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ABSTRACT

To estimate the health risk of haloacetonitriles in different kinds of drinking water, the concentrations of haloacetonitriles in tap water, boiled water and direct drinking water were detected. The physiologically based pharmacokinetic (PBPK) model was used to calculate internal dose in the human body for haloacetonitriles through ingestion, and the probability distributions of the non-carcinogenic risk of haloacetonitriles for human via drinking water were assessed. This study found that the mean concentrations of dichloroacetonitrile (DCAN) in tap water, boiled water and direct drinking water were 0.955 μg/L, 0.207 μg/L and 0.127 μg/L, and those of dibromoacetonitrile (DBAN) were 0.221 μg/L, 0.104 μg/L, 0.089 μg/L, respectively. In China, direct drinking water is used most frequently, so the concentrations of haloacetonitriles in direct drinking water were used to obtain data on the internal dose of haloacetonitriles. In addition, the simulation results for the PBPK model showed that the highest and lowest concentrations of DCAN occurred in the liver and venous blood, respectively. The peak concentrations of DBAN in each tissue were in the decreasing order liver > rapidly perfused tissue > kidney > slowly perfused tissues > fat > arterial blood (venous blood). In addition, the highest 95th percentile hazard quotients (*HQ*) value of haloacetonitriles via drinking water for humans was 8.89×10^{-3}, much lower than 1. The 95th percentile hazard index (*HI*) was 0.046, which was also lower than 1, suggesting that there was no obvious non-carcinogenic risk.

1. Introduction

In recent years, disinfection by-products in drinking water have become a concern around the world (Monarca et al., 2004). To control disinfection by-products in drinking water, a variety of disinfection processes have been researched. However, some toxic disinfection by-products are produced in the disinfection process, and haloacetonitriles (HANs) are one of the most representative disinfection by-products (Chu et al., 2011; Plewa et al., 2008). Due to stronger cytotoxicity, genetic toxicity and the teratogenic abilities of HANs (Ahmed et al., 2000; Huang et al., 2007; Smith et al., 1989), the HANs in drinking water are a threat to human health (Nouraldeen and A, 1996). Thus, although the concentrations of HANs in drinking water were quite low, the toxicity of HANs was very high (Muellner et al., 2007). Thus, the risk for humans via long-term water drinking cannot be ignored. Therefore, it is very important to estimate the health effect of HANs in drinking water.

Other research has focused on analysis methods, production, distributions and levels of HANs in water. For example, Kristiana et al. (2012) determined the concentration of HANs in drinking water using head-space solid-phase micro-extraction combined with gas chromatography and mass spectrometry; Ma et al. (2014) studied the method of salt-assisted dispersive liquid-liquid micro-extraction combined with programmed temperature vaporization by gas chromatography mass spectrometry for the determination of HANs in drinking water; Zhang et al. (2013) researched the influence of temperature on the production of HANs and found that the yield of HANs decreased with increased temperature, and the concentration of HANs decreased with increased storage time. Many studies have shown that with decreased pH, HANs production increased during disinfection after adding chloramine and chlorine (Xu et al., 2012).

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There is increasing focus on HANs in water, but few studies have paid attention to the toxic effects of HANs to human health. Ahmed et al. (2005) found that pregnant mice exposed to DCAN can lead to foetal brain effects, such as oxidative stress, apoptosis and neurological adverse effects. Ileka-Priouzeau et al. (2015) evaluated the influence of multiple routes of exposure to HANs on small-for-gestational-age neonates in Quebec City, Canada. Most studies focus on the establishment of a detection method of HANs in environmental media, the influence factors of the production of HANs in disinfection and the toxicology of HANs. Although there are many studies on the toxicity of HANs, the metabolic mechanism and direct influence of HANs to human health remain unclear.

In view of the potential health hazards of HANs, more attention should be paid to their health risk assessment. Increasingly, scholars use internal dose to describe the human health risk of disinfection by-products, due to its accuracy in human health assessment (Ciffroy et al., 2016; Dewoskin, 2007; McLanahan et al., 2014). Internal dose refers to doses of chemical substances after absorption into the human body, which has become a core concept in toxicology and risk assessment in recent years. The Physiologically Based Pharmacokinetic model (PBPK model) can accurately simulate the absorption, degradation and concentration of chemicals in different tissues in various situations (Schuhmacher et al., 2014). The PBPK model can be used characterize the human health risk of pollutants more accurately, and the results may be more realistic than a conventional risk assessment method (Kenyon et al., 2016; Knutsen et al., 2013).

The PBPK model has been used to simulate the internal dose and calculate health risk in recent years (Krewski and Andersen, 1994). For example, Cox and Ricci (1992) used a PBPK model to evaluate the risk of the metabolite of benzene; Niu et al. (2015) calculated the cancer risk of trihalomethanes in reclaimed water based on PBPK model. As noted earlier, scholars applied the PBPK model of disinfection by-products to the human health risk assessment of hexachlorobutadiene and noted that the study is also applicable to other chemicals (Trevor et al., 2003). Therefore, this method of human health risk assessment is also suitable for the pollutants in the study. Although many scholars use a PBPK model to calculate the health risk of DBPs, most research mainly focused on common components or traditional disinfection by-products in drinking water; little studies has concentrated on the human health risk assessment of HANs based on PBPK-Monte Carlo.

Above all, HANs have potential health risks to humans, but there are few studies of these risks. Most scholars only used concentrations of pollutants in water to calculate human health risk; few studies have focused on using concentrations of pollutants in human tissues to conduct human risk assessment.

The purpose of this study is to analyse the concentration distributions of HANs in tap water, direct drinking water and boiled water and to estimate the non-carcinogenic risk of DCAN and DBAN in drinking water based on internal dose. This work can provide a scientific and theoretical basis for the effective control of disinfection by-products in drinking water, and may be helpful for controlling the risk to the drinking water provision.

2. Materials and methods

2.1. Sample collection and analysis

According to drinking habits in China, tap water is usually not suitable for drinking directly. People often boil tap water for tea or coffee, and the direct drinking water is used more frequently. Therefore, three kinds of water are sampled in this study: tap water, direct drinking water (treated tap water; the treatment process is illustrated in Fig. 1), and boiled water (tap water after boiling).

DBAN and DCAN are chosen to represent HANs in this study, considering their high toxicity and high detected concentrations in drinking water.

All samples were collected at three sample points in Tianjin (a city in North China) from April to July in 2016 (16 times per point). Tap water and direct drinking water were sampled directly from each tap. Boiled water was obtained after the boiled tap water cooling naturally. Before sampling, the bottles were soaked in concentrated nitric acid for 8 h and rinsed with sterile water several times to neutral pH. All bottles were dried in the vacuum box. The brown bottles (1 L) were filled until overflowing and tightly sealed with Parafilm M to quantify HANs in the water samples. The Parafilm M coating is inert, non-contaminating, and flexible (Tsubouchi et al., 1985). The samples were preserved at 4 °C and analysed within 24 h.

The samples were pre-treated through liquid-liquid extraction and analysed with a gas chromatography with an electron capture detector (Agilent 6890N system; HP-5 capillary column: 30.0 m × 320 μm × 0.25 μm; Agilent 7683B automatic sampler). The specific analysis process complied with the USEPA standard method 551.1 (USEPA, 1995).

All samples and blanks were analysed in duplicate for quality assurance control of laboratory analysis in this study. Only relative standard deviation (RSD) values below 10% were accepted, while samples outside this range were reanalysed. A standard reference material, (SRM) GSB 07-1982-2005, from the Institution for Environmental Reference Material Ministry of Environmental Protection was applied for calibration and analytical control (Niu et al., 2015). HANs were identified relative to external standards. The recoveries of SRM and external standards were 93.5% for DBAN and 95.2% for DCAN. In addition, SRM was continuously diluted and analysed until the peak had a signal-to-noise ratio (S/N) of 3. The corresponding concentrations were regarded as the detection limit, which was 0.01 μg/L for DBAN and 0.005 μg/L for DCAN.

2.2. PBPK model of HANs

The internal dose of HANs (such as in blood or other tissues) is difficult to measure. The measurement methods are not easily satisfied (Batterman et al., 2016), and biological samples are hard to obtain. Thus, we use the PBPK model to simulate the internal dose of HANs.

The PBPK model can be used to simulate the uptake, distribution, and metabolism of the pollutants, and can also be used to simulate the concentration change properties using time of exposure to contaminants in organs or target tissues. Moreover, the dose simulated by the PBPK model is known as the internal dose, which is a promising tool applied to risk assessment studies for different contaminants.

The physiological parameters used in the PBPK model were listed in Table S1. The distribution coefficients and metabolic parameters were taken from the relevant literature and listed in Table 1. In this study, we only introduce several typical equations in the details.

In this model, all HANs can be metabolized in the liver and kidney. The metabolism was described in equation (1).

$$RAM = \frac{V_{max} C_{vl}}{K_m + C_{vl}} \quad (1)$$

where RAM is metabolism conversion of pollutants in liver, μg/min; V_{max} is maximum metabolic rate constant of pollutants, μg/min; C_{vl} is concentration of pollutants in venous blood leaving the liver, μg/L; and K_m is the Michaelis constant, μg/L. In this system, the expression of the metabolic conversion of pollutants in the kidney

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