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Health risk assessment of reclaimed wastewater: A case study of a conventional water reclamation plant in Nanjing, China

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ABSTRACT

Contaminated reclaimed wastewater has the potential to induce adverse effects on the health of wastewater workers and residents. However, few studies have focused on these health risks. In this study, we assessed the health risk of samples collected from different treatment processing units used in a typical water reclamation plant in Nanjing, China. Chemical analysis revealed that 40 semi-volatile organic compounds (SVOCs) and 6 N-nitrosamines (NAs) persisted after wastewater treatment. A health risk assessment revealed that the SVOCs in effluents pose negligible non-carcinogenic risk to wastewater workers and local residents as both the hazard quotient (HQ) and hazard index (HI) were all below 1.00. However, polycyclic aromatic hydrocarbons (PAHs), phthalate esters (PAEs) and NAs may present a carcinogenic risk, since their risk index via dermal exposure exceeded the safety limit (1.00×10^{6}), indicating that conventional treatment processes cannot effectively reduce the health risk in reclaimed wastewater. These results strongly suggest the need for the introduction of advanced treatment technologies capable of effectively removing SVOCs and NAs in water reclamation plants.

1. Introduction

To relieve the pressure on traditional water sources, exploring alternative water supplies has become a worldwide necessity, especially in regions with water shortages (Fatta-Kassinos et al., 2016; Jiménez and Asano, 2008). China, as the largest developing country in the world, faces increasing demands on its water supply, which have led to water shortages. Great efforts have been made to find alternative sources of water, with the improved utilization rate of reclaimed wastewater to be one alternative that has been explored (Yi et al., 2011). Increasingly, reclaimed wastewater has been used for irrigation, landscaping, and recreational and environmental supplies as well as a source to replenish groundwater. This alternative source of water has become especially important in the northern cities of China, such as Beijing, Tianjin and Qingdao (Pinjing et al., 2001; Shi et al., 2010).

Although reclaimed wastewater is used as a non-potable water resource, there is a potential health risk from the accumulation and exposure to trace pollutants. It has been demonstrated that most trace pollutants, including semi-volatile organic compounds (SVOCs), cannot be completely removed by conventional wastewater treatment processes (Garcia-Segura et al., 2015; Grandclement et al., 2017). Additionally, disinfection, an essential process for water reclamation, inevitably produces unwanted by-products (DBPs). Many DBPs are carcinogens and teratogens (Liu and Zhong, 2017; Marti et al., 2015), especially N-nitrosamines (NAs), which are the most toxic kind of contaminants generated during the water reclamation process (Fujioka et al., 2016).

The wide application, easy accessibility and potential toxicity of reclaimed wastewater makes it a vital health issue. A human health risk assessment is a useful approach to assess the potential risk of environmental contaminants in source water, drinking water, and sediments (Sany et al., 2014; Wu et al., 2011; Zhao et al., 2015).

In this study, the effluents of different treatment units were sampled from a water reclamation plant in Nanjing, China, and concentrations of 64 SVOCs and 9 NAs in these effluents were detected. Based on the concentrations of these chemicals, a health risk assessment was conducted to evaluate whether SVOCs and NAs in reclaimed wastewater present a potential health risk to wastewater workers and residents. According to the assessment results, an analysis of different reclamation treatments that could reduce the public health risk associated with these pollutants was conducted.

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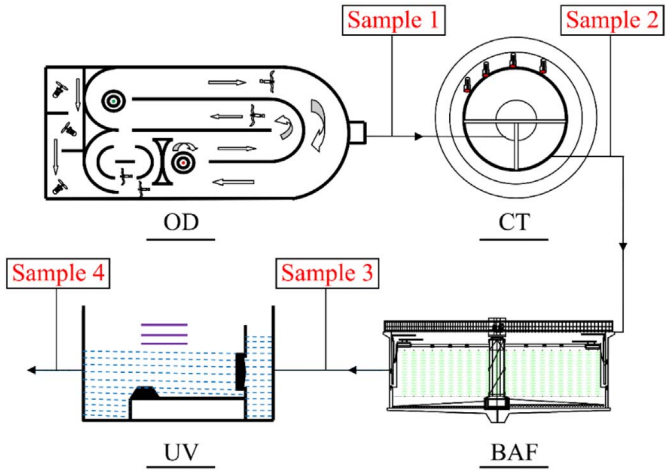

Fig. 1. Flow diagram of reclaimed wastewater treatment factory. OD, oxidation ditch; CT, coagulation tank; BAF, biological aerated filter; UV, ultraviolet disinfection pool.

2. Material and methods

2.1. Water sampling and analytical methods

2.1.1. Water sampling

The study was performed in a water reclamation plant in Nanjing, China. This plant receives secondary effluent from an oxidation ditch (OD) process and is located in domestic wastewater treatment facilities. The reclaimed wastewater is mostly used for landscaping. There are four treatment processes configured in a series in the plant. Reclaimed wastewater samples were taken from the effluent of each treatment process (Fig. 1). The water samples were collected in 5 L brown glass bottles once a month from October 2013 to September 2014 (12 sampling occasions and 48 samples in total). Before sample collection, all the bottles were initially immersed in chromosulfuric acid for 24 h, then washed with deionized water eight times and finally rinsed three times with raw water. One or two drops of concentrated hydrochloric acid and 1 g of ascorbic acid were added to each sample to reach a pH $<$ 2.0 and inhibit biodegradation. The samples were transferred to the laboratory within 2 h, filtered through 0.45-μm membrane filters and stored at 4 °C.

2.1.2. SVOC analysis

A total of 64 SVOCs were measured, including 18 polycyclic aromatic hydrocarbons (PAHs), 6 phthalate esters (PAEs), 19 organochlorine compounds (OCCs) and 21 other organic chemicals (Table S1). Standard SVOCs and a deuterated internal standard (IS) mixture (acenaphthene-d10, chrysene-d12 and phenanthrene-d10) were purchased from AccuStandard (USA) and Supeclo (USA), respectively.

The extraction of SVOCs was done using the liquid-solid extraction method US-EPA (1995) 525.2 with some modifications. Briefly, 5 mL of methanol was added to 1 L of water sample. Then, water samples were extracted through sample prep performance (SPE) cartridges (Supeclo, USA) filled with 2 g of coconut charcoal, at a flow rate of 10 mL min^{-1}. The cartridge was conditioned with 5 mL of ethyl acetate and 5 mL of methylene chloride. After extraction, the cartridges concentrated with organic compounds were connected to a drying tube, and the organic compounds were eluted with 5 mL of ethyl acetate, followed by 5 mL of methylene chloride and a 3 mL mixture of ethyl acetate and methylene chloride (1:1 of v/v). The combined extracts were concentrated to a volume of 1 mL using a nitrogen evaporation apparatus. SVOCs were measured on a GC/MS with electron ionization (EI) ion source (Agilent-7890, Germany). Quantification of individual compounds was done by comparing the peak areas with those of the standards.

2.1.3. NA analysis

The following nine NAs were analyzed: *n*-nitrosodi-n-butylamine (NDBA), *n*-nitrosodiethylamine (NDEA), *n*-nitrosodimethylamine (NDMA), *n*-nitrosodiphenylamine (NDPHA), *n*-nitrosodi-n-propylamine (NDPA), *n*-nitrosomethylethylamine (NMEA), *n*-nitrosomorpholine (NMOR), *n*-nitrosopiperidine (NPIP) and *n*-nitrosopyrrolidine (NPYR). Standard NAs and a deuterated IS mixture (NDMA-d_6 and NDPA-d_{14}) were purchased from AccuStandard (USA) and Cambridge Isotope Laboratories (USA), respectively. NAs were extracted from water samples (1 L each) by using an SPE cartridge filled with 2 g of coconut charcoal at a flow rate of l L h^{-1}. The cartridge was conditioned with methylene chloride, methanol and distilled water according to the US-EPA Method 521 (2005). The cartridges concentrated with NAs were connected to a drying tube, and NAs were eluted with 12 mL of methylene chloride. The extracts were concentrated using a nitrogen evaporation apparatus to a final volume of 1 mL before running samples on a GC/MS. Quantification of NAs was done by comparing the peak areas with those of the standards.

2.1.4. Quality assurance

All reagents used in this study were of chromatographic purity grade. Reagent blanks, standard reference materials and sample replicates were applied during the chemical analysis to assess contamination and precision. The mean recoveries for SVOCs and NAs were in the range of 80.9–96.3% and 90.5–102.5%, respectively. The detection limits were 0.4–1.0 ng L^{-1} and 0.2–5.0 ng L^{-1} for SVOCs and NAs, respectively. The chemical analysis was conducted in triplicate for each sample.

2.2. Statistical analysis

The results are expressed as the mean ± standard deviation (SD). The pollutant concentrations in different effluents were analyzed using one-way ANOVA followed by Tukey's HSD test. Statistical analysis was performed using SPSS 22 software (SPSS Inc., USA). Significance was set at $p < 0.05$.

2.3. Human exposure and health risk assessment

2.3.1. Problem formulation

In this study, we assumed that (i) the health risks (including non-carcinogenic risk and carcinogenic risk) of reclaimed wastewater were mainly caused by SVOCs and NAs, and (ii) the concentrations of these pollutants in each treatment unit were relatively stable.

2.3.2. Exposure assessment

Human exposure to wastewater contaminants mainly follow these pathways: inhalation of volatile pollutants, ingestion of food crops irrigated with wastewater or dermal contact of contaminated wastewater (Barker et al., 2013; Jan et al., 2010; Khan et al., 2008). Most water reclamation plants are open-air constructions. In China, reclaimed wastewater is mostly used as a non-potable source. Therefore, dermal contact is assumed to be the most likely pathway for human exposure to trace pollutants found in reclaimed wastewater. The two main populations that have the greatest risk of exposure to pollutants found in reclaimed wastewater are wastewater workers and residents (adults and children) that live near reclamation facilities. The different exposure parameters and probability distributions for these two populations are provided in Table S2. The exposure doses for pollutants were calculated using Eqs. (1) and (2), which were adapted from US-EPA (2004, 2011).

$$DAevent = 2FA \times Kp \times Cw\sqrt{\frac{6\tau event \times tevent}{\pi}} \quad (1)$$

where DA_{event} is dermal absorbed SVOCs or NAs per event (μg cm^{-1} $event^{-1}$), FA is the fraction of water absorbed, K_p is the dermal permeability coefficient of contaminant (cm h^{-1}), C_w is the