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SHORT RESEARCH AND DISCUSSION ARTICLE

Different senescent HDPE pipe-risk: brief field investigation from source water to tap water in China (Changsha City)

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Abstract Semi-volatile organic compounds (SVOCs) derived from plastic pipes widely used in water distribution definitely influence our daily drinking water quality. There are still few scientific or integrated studies on the release and degradation of the migrating chemicals in pipelines. This investigation was carried out at field sites along a pipeline in Changsha, China. Two chemicals, 2, 4-tert-buthylphenol and 1, 3-diphenylguanidine, were found to be migrating from high density polyethylene (HDPE) pipe material. New pipes released more of these two compounds than older pipes, and microorganisms living in older pipes tended to degrade them faster, indicating that the aged pipes were safer for water transmission. Microorganism degradation in water plays a dominant role in the control of these substances. To minimize the potential harm to human, a more detailed study incorporating assessment of their risk should be carried out, along with seeking safer drinking pipes.

Keywords Plastic pipes · SVOCs · Drinking water · Microorganism degradation · 2, 4-Tert-buthylphenol · Water distribution

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Introduction

With the progress of urbanization in developing countries, plastic pipes made of high-molecular weight polymers are more and more widely used for drinking water distribution (Stern and Lagos 2008). To improve drinking water quality, most efforts have been focusing on the treatment process in water supply plant (Tang et al. 2013), while there are few systematic studies on the impact of water transmission conduit components on water quality. Some chemicals in plastic materials, including additives as well as their degradation products, can migrate into drinking water, with plasticizers counting as one of the most controversial additives (Wei et al. 2015). Anticorrosive or antioxidant coatings on pipe made of polyethylene and epoxy resins may also leach into the aqueous phase.

Chemicals migrating from pipe components frequently occur in drinking water and obviously deteriorate water quality (Stern and Lagos 2008). For example, Skjevrak et al. (2003) identified a range of esters, aldehydes, ketones, aromatic hydrocarbons, and terpenoids in Norwegian drinking water as having migrated from high-density polyethylene (HDPE) pipes. Moreover, urban residents are increasingly complaining about the significant taste and odour of drinking water, e.g. in the USA (Dietrich and Burlingame 2015) and China (Zhang et al. 2013). Organic compounds migrating from polymeric pipes into drinking water may be one of the causes of the poor taste of drinking water, which in turn is often accompanied by increase of total organic carbon (TOC) (Skjevrak et al. 2003). Some volatile organic compounds (VOCs) produced by fresh water algae (Stern and Lagos 2008) and bacteria (Skjevrak et al. 2003) are also found widely in distributed drinking water (Dietrich and Burlingame 2015). However, no practical investigation of VOCs or semi-VOCs (SVOCs) in drinking water along pipelines with sections of different age (different senescent pipelines) has been undertaken. Thus, the aim of this project was to investigate the presence and distribution of selected SVOCs from source water to tap water in Changsha, China. The potential migration of specific compounds from pipe material into water at different periods of pipeline life was specifically studied.

Methods and analyses

The pipeline investigated is located in Changsha City, Hunan Province, China. Water extracted from the Laodao River flows through a series of buildings in the Xingsha Water Supply Plant. The HDPE pipes in these buildings are of different age. Samples were collected from sites along the pipeline in the direction of flowing water (Fig. 1). In the water supply plant, water from Laodao River is chlorinated, and then mainly flows sequentially through an air flotation tank, a sedimentation tank, and different kinds of filter tanks in three parallel procedures (phase I, phase II and phase III), where phase I, phase II and phase III are fitted with siphon, inverse and backwash filters, respectively. Phase II was not in service at the time of this study, so there was no sampling of this process stream. Subsequently, the treated water from phase I and phase III was mixed together as product water. The ages of the pipes in the three phases and downstream built environments differ (Fig. 1): Phase I pipes in the water supply plant and the commercial building have been in service for about 20 years; Phase III pipes in the water supply plant and the residential district came into service approximately 5 years ago. The office building has been used for 8 years. The pipelines at the end of the water supply plant (denoted with purple in Fig. 1) were installed 3 years ago.

A gas chromatography-mass spectrometry (GC/MS) analytical method (Tang et al. 2015; Lai et al. 2012) was used to quantitatively determine SVOCs concentrations in the sampled water. Of the SVOCs screened, 2,4-tert-buthylpheno (2,4-DTBP) has an alkyl phenolic odour and can cause symptoms varying from headache, dizziness, eye damage to severe destruction of tissue by exposure (Skjevrak et al. 2003, 2005), and its LD50 (Rat) by oral is 2000 mg/kg (Sigma-Aldrich MSDS-137731 2015). As for 1,3-diphenylguanidine (1,3-DPG), it irritates the skin, and its oral LD50 (Rat-male) is 111 mg/kg (Sigma-Aldrich MSDS-D207756 2015). Though 2,4-DTBP and 1,3-DPG have relatively low toxicity, their potential or cumulative harm to humans may be larger when people are frequently exposed via drinking water.

In order to investigate the relationship between microorganisms and these two SVOCs, a small-scale simulation experiment was carried out. *Escherichia coli* (*E. coli*), widespread in natural water, was used as the model organism. All batch experiments were performed in glass conical flasks undergoing shaking at 37 °C (Sondi and Salopek-Sondi 2004; Dasari and Hwang 2010). Aliquots of bacterial suspension (approximately 40 CFU/mL) were added to the sterile water or Luria-Bertani liquid medium containing 2,4-DTBP or 1,3-DPG. The initial concentration of *E. coli* was chosen according to environmental quality standards for surface water (GB 3838-2002) in China. Then, aliquots of the solutions were withdrawn by a syringe at regular time intervals to test *E. coli* and SVOC concentrations. The concentration of *E. coli* was determined by dilution plate count method (Mortimer et al. 2000). Briefly, aliquots of 100 μL of all diluted samples were spread on the eosin-methylene blue agar plates, and the number of *E. coli* colonies was counted after 24 h of incubation at 37 °C. SVOC-free bacterial suspensions cultured under the

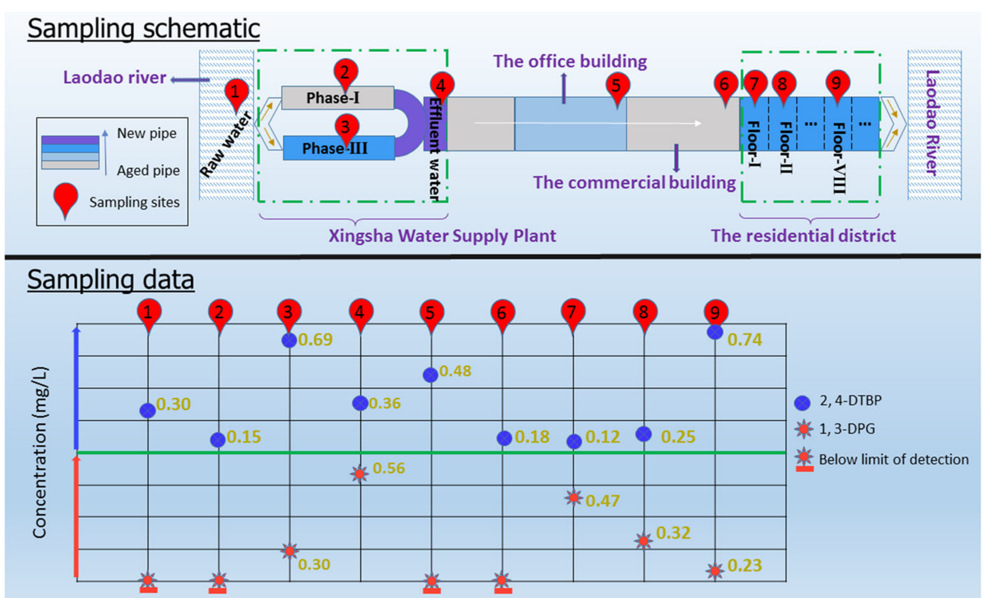

Fig. 1 The sampling schematic and the investigated sampling data in drinking water pipeline

Table 1 The concentrations of bacterial suspension exposed to different amounts of additional SVOCs for 2.5 h (CFU/mL)

Cultivation matrix	Blank	1 mg/L 2,4-DTBP	0.5 mg/L 2,4-DTBP	1 mg/L 1,3-DPG	0.5 mg/L 2,4-DTBP and 1 mg/L 1,3-DPG
Sterile water	570	150	1310	710	620
Luria-Bertani liquid medium	10,400	700	26,300	21,000	2000

same conditions were used as a control; the aforementioned GC/MS analytical method was used to quantitatively determine SVOCs concentrations.

Results and discussion

2,4-DTBP was observed in the raw water (0.30 mg/L), which may be due to the generation of 2,4-DTBP by fresh water algae (Jüttner 1995) or sewage discharge into the river upstream of the offtake. The concentrations of 2,4-tert-buthylphenol (2,4-DTBP) and 1,3- diphenylguanidine (1,3-DPG) in drinking water at the different sampling sites were in the range from 0.12 to 0.74 mg/L, respectively (Fig. 1). The level of 2,4-DTBP in the product water (0.36 mg/L) was higher than that in water supplied by the Second Water Supply Plant of Changsha (0.22 mg/L). That water supply plant has added activated carbon filter and ozone disinfection procedures, and their source water is from Xiangjiang River, suggesting that water source and treatment process might make a difference to the amount of 2,4-DTBP in product water. 2,4-DTBP may be a degradation product of an antioxidant, mainly produced from new HDPE pipes (Skjevrak et al. 2005). In the residential district, all the pipes on all the floors are of the same age, and the drinking water flows from the lower floor to the higher floor. It is reasonable that the concentration of this phenolic substance is higher in the pipeline at higher floors due to the accumulation of sustained release from the new pipes. Anaerobic bacteria and fungi with oxidizing ability possess greater potential to degrade phenolic compounds. The amount of 2, 4-DTBP in drinking water in the commercial building is reduced, possibly as a result of the microorganism in aged pipes metabolizing 2,4-DTBP.

The chemical 1,3-DPG was first detected in phase III, with concentrations highest in the product water, an observation which again may be caused by the new pipes in that part of the plant. 1,3-DPG is probably released by the breakdown of some additives in the new HDPE pipes. In the new pipes, the 1,3-DPG is released into drinking water more quickly than 2, 4-DTBP, probably because it is about ten times more soluble than 2,4-DTBP (Sigma-Aldrich MSDS-137731 2015; Sigma-Aldrich MSDS-D207756 2015). That 1,3-DPG is just present in the water supply plant and the residential district and there is a decreasing trend of the 1, 3-DPG concentrations in the pipes at higher floors in the residential district may be due to microorganisms residing in pipes preferentially metabolizing 1,3-DPG rather than 2,4-DTBP.

The concentration of bacterial suspension exposed to additional 2,4-DTBP or 1,3-DPG for 2.5 h is shown in Table 1. Obviously, the population of *E. coli* in the Luria-Bertani liquid medium was much higher than the population in the sterile water because of the presence of nutrients in the Luria-Bertani medium. At higher concentration, 2,4-DTBP was observed to inhibit the growth of *E. coli*, even though lower concentrations promoted reproduction, no matter whether the *E. coli* were in sterile water of Luria-Bertani medium. This observation can be explained by SVOCs at the higher concentrations being harmful to bacteria like *E. coli*, but being utilized as a food source at lower concentrations. At 0.5 mg/L, both 2,4-DTBP and 1,3-DPG favour *E. coli* growth (Fig. 2). The number of *E. coli* decreases in the first hour of exposure as the bacteria adapt to the SVOCs, then increases until the culture reaches a reproduction peak after 2.5 h, before falling to around 200 CFU/mL. The concentrations of both 2,4-DTBP and 1, 3-DPG gradually reduced in the presence of *E. coli*, suggesting that common bacteria can degrade both these SVOCs. Furthermore, the degradation of 1,3-DPG is preferential and almost complete in 6 h, whereas degradation of 2,4-DTBP is slower, albeit the microorganisms also keep the concentration of this chemical at low levels.

In summary, new pipes release 2,4-DTBP and 1,3-DPG and microorganisms tend to degrade them in the aged pipes. In brand new pipe, there are few microorganisms on the pipe

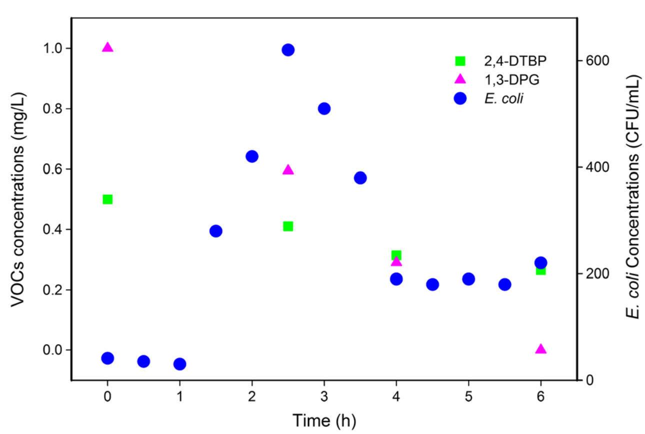

Fig. 2 The plots of concentration changes of 2,4-DTBP, 1,3-DPG and *E. coli* in sterile water versus time. Conditions: *E. coli* suspension (initial concentration of 40 CFU/mL) exposed in 0.5 mg/L 2,4-DTBP and 1 mg/L 1,3-DPG sterile water with shaking at 37 °C

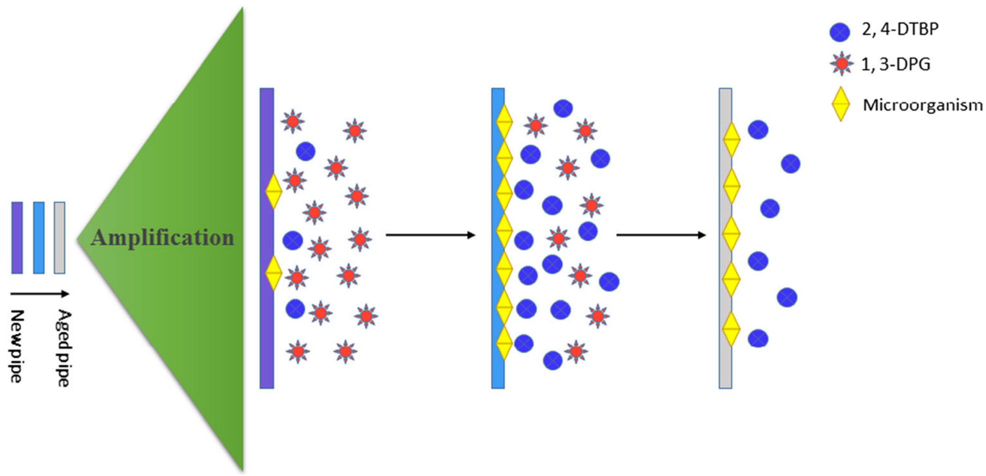

Fig. 3 The brief schematic diagram about variation of two migrating compounds and microorganisms in different senescent pipelines

inner wall (Fig. 3), and 1,3-DPG migrates faster than 2,4-DTBP into the water. When the pipe becomes senescent, 1,3-DPG is preferentially metabolized by the microorganisms and the amount of 2,4-DTBP in water increases. In more aged pipe, the degradation of 1,3-DPG is almost complete, and the microorganisms keep the concentration of 2,4-DTBP at low levels. Thus, it can be seen that the perception that new pipes are safer than aged pipes may not be a good one. Even so, the reason why some aged pipes are better than new pipes remains unclear. It is possible that migrating organic compounds may serve as nutrients for bacteria, and thus promote microbial growth in aged pipelines, which leads to the decrease of organic compounds and then the decrease of microorganisms to a low level. More detailed investigations on plastic pipes at different ages need to be proceeded as well to seek the low-risk period of all kinds of pipes.

Conclusion

An investigation on the distribution of SVOCs from source water to tap water in different senescent HDPE pipes was carried out in Changsha, China. 2,4-DTBP and 1,3-DPG migrated from HDPE pipe material into the water. The results possibly indicate that there is a close relationship between this phenomenon and the deterioration of HDPE pipes, and the different substances' degradation rates by microorganisms in water play a dominant role in the amount of these substances in the aqueous phase. Microorganisms could be used at water outlets to remove toxicities derived from new pipes. However, more detailed investigations on the specific toxicity of these unnoticed organic chemicals (2,4-DTBP and 1,3-DPG) in drinking water should be undertaken. The risk assessment of plastic pipes cannot be ignored, which can promote the progress about governmental management of plastic pipe materials for drinking water distribution. Moreover, more environmentally friendly pipes would be also a better choice.

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References

Dasari TP, Hwang H-M (2010) The effect of humic acids on the cytotoxicity of silver nanoparticles to a natural aquatic bacterial assemblage. Sci Total Environ 408:5817–5823

Dietrich AM, Burlingame GA (2015) Critical review and rethinking of USEPA secondary standards for maintaining organoleptic quality of drinking water. Environ Sci Technol 49(2):708–720

Jüttner F (1995) Physiology and biochemistry of odorous compounds from freshwater cyanobacteria and algae. Water Sci Technol 31(11):69–78

Lai B, Zhou Y-X, Yang P, Wang K (2012) Comprehensive analysis of the toxic and refractory pollutants in acrylonitrile–butadiene–styrene resin manufacturing wastewater by gas chromatography spectrometry with a mass or flame ionization detector. J Chromatogr A 1244:161–167

Mortimer FC, Mason DJ, Gant VA (2000) Flow cytometric monitoring of antibiotic-induced injury in escherichia coli using cell-impermeant fluorescent probes. Antimicrob Agents Chemother 44:676–681

Sigma-Aldrich MSDS-137731 (2015) Material Safety Data Sheet. http://www.sigmaaldrich.com/catalog/product/aldrich/137731?lang=en&region=US. Accessed 30 June

Sigma-Aldrich MSDS-D207756 (2015) Material Safety Data Sheet. http://www.sigmaaldrich.com/catalog/product/aldrich/d207756?lang=en&region=US. Accessed 30 June

Skjevrak I, Due A, Gjerstad KO, Herikstad H (2003) Volatile organic components migrating from plastic pipes (HDPE, PEX and PVC) into drinking water. Water Res 37(8):1912–1920

Skjevrak I, Lund V, Ormerod K, Herikstad H (2005) Volatile organic compounds in natural biofilm in polyethylene pipes supplied with lake water and treated water from the distribution network. Water Res 39(17):4133–4141

Sondi I, Salopek-Sondi B (2004) Silver nanoparticles as antimicrobial agent: a case study on E. coli as a model for Gram-negative bacteria. J Colloid Interface Sci 275:177–182

Stern BR, Lagos G (2008) Are there health risks from the migration of chemical substances from plastic pipes into drinking water? Hum Ecol Risk Assess 14(4):753–779

Tang R, Clark JM, Bond T, Graham N, Hughes D, Freeman C (2013) Assessment of potential climate change impacts on peatland dissolved organic carbon release and drinking water treatment from laboratory experiments. Environ Pollut 173:270–277

Tang L, Tang J, Zeng G-M, Yang G-D, Xie X, Zhou Y-Y, Pang Y, Fang Y, Wang J-J, Xiong W-P (2015) Rapid reductive degradation of aqueous p-nitrophenol usingnanoscale zero-valent iron particles immobilized on mesoporoussilica with enhanced antioxidation effect. Appl. Surf Sci 33:220–228

Wei G-L, Li D-Q, Zhuo M-N, Liao Y-S, Xie Z-Y, Guo T-L, Li J-J, Zhang S-Y, Liang Z-Q (2015) Organophosphorus flame retardants and plasticizers: sources, occurrence, toxicity and human exposure. Environ Pollut 196:29–46

Zhang L, Liu S, Liu W (2013) Investigation of organic matter migrating from polymeric pipes into drinking water under different flow manners. Environ Sci Proc Imp 15(2):280–290