Chemosphere 205 (2018) 643–648

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Tracing the sources of iodine species in a non-saline wastewater

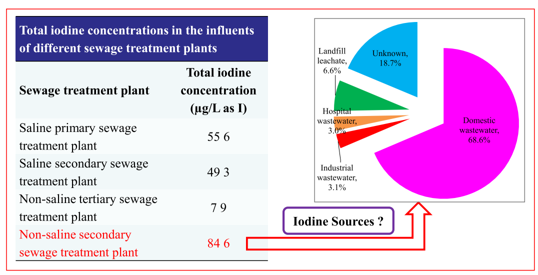
Tingting Gong [a, b, **], Xiangru Zhang [a, *], Wenqing Liu [a], Yun Lv [a], Jiarui Han [a], Ka Chun Choi [a], Wanxin Li [a], Qiming Xian [b]

[a] Department of Civil and Environmental Engineering, The Hong Kong University of Science and Technology, Hong Kong, China
[b] State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, Nanjing, China

HIGHLIGHTS

- A non-saline wastewater in Hong Kong contained an unexpected high level of iodine.
- The concentrations of iodine species in various types of wastewater were measured.
- A specific domestic wastewater was the major iodine source.
- The extensive use of salt resulted in high levels of iodine in domestic wastewater.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 January 2018
Received in revised form
20 April 2018
Accepted 22 April 2018
Available online 24 April 2018

Handling Editor: Shane Snyder

Keywords:
DBPs
Iodinated DBPs
Iodine
Iodide
Wastewater
Salt

ABSTRACT

There are two types of wastewater in Hong Kong, non-saline and saline wastewaters. When it comes to disinfection, iodide is an important inorganic ion in concern because it may involve in the formation of iodinated disinfection byproducts, which show significantly higher toxicity than their brominated and chlorinated analogues. In this study, it was found that a non-saline wastewater in Hong Kong contained an unexpected high level of iodine. To trace the iodine sources of this non-saline wastewater, the information of the corresponding area was collected to find the possible iodine sources; then, the water samples from the possible iodine sources were collected; the concentrations of iodine species (iodide, iodate and organic iodine) in these collected water samples were determined; finally, the contribution percentages of iodine species from different sources were calculated. The results revealed that a specific domestic wastewater was the major iodine source, contributing to 68.6% of total iodine, 66.3% of iodide, 57.0% of iodate, and 112% of organic iodine in the non-saline wastewater, while landfill leachate, industrial and hospital wastewaters were the minor iodine sources, contributing to 6.6%, 3.1%, and 3.0% of total iodine in the non-saline wastewater, respectively. Furthermore, it was found that the extensive use of salt might result in high levels of iodine in the domestic wastewater and thus lead to the high level of iodine in the non-saline wastewater.

1. Introduction

Iodine is universally present in natural waters such as seawater, freshwater and rain, and it has been detected in tap water (Blount et al., 2010; Gilfedder et al., 2008, 2009; Gong and Zhang, 2013; Pan and Zhang, 2013; Qin et al., 2014; Schwehr and Santschi, 2003).

* Corresponding author.
** Corresponding author.
E-mail addresses: ttgong@nju.edu.cn (T. Gong), xiangru@ust.hk (X. Zhang).

https://doi.org/10.1016/j.chemosphere.2018.04.147
0045-6535/

Iodine is an essential nutrient for mammals including humans, thus various physiological fluids, such as milk, serum, and urine, contain iodine (Hou et al., 2000; Verma et al., 1992). To ensure the proper daily iodine intake for humans, iodine has been added to commercial salt (Dasgupta et al., 2008; GB5461–2000; Pan et al., 2016). Besides, a large number of iodine-containing X-ray contrast media are used for medical imaging, introducing iodine to hospital wastewater (Duirk et al., 2011; Wendel et al., 2016; Xu et al., 2017). Iodine is also used in the manufacture of chemicals and pharmaceuticals (Piñeiro et al., 2011), ending up in industrial wastewater. Moreover, iodine is commonly used in the sanitation process in dairies (Hladik et al., 2016). The typical iodine concentrations in seawater, freshwater, rain, urine and tap water are 45–90, 0.5–20, 0.5–5.0, 0–300 and 6–13 μg/L, respectively (Gong and Zhang, 2013; Hou et al., 2000; Schwehr and Santschi, 2003; Ünak et al., 1999; Whitehead, 1984). With all these natural and artificial iodine sources, wastewater tends to contain iodine. The three dominant iodine species in waters are iodide, iodate and organic iodine (Gilfedder et al., 2008).

Commonly, municipal wastewater is non-saline wastewater with low levels of inorganic ions (Gong et al., 2016). When seawater is used for toilet flushing, high levels of inorganic ions (including iodide and iodate ions) are introduced into municipal wastewater, resulting in saline wastewater (Gong and Zhang, 2015; Gong et al., 2016). To reduce the local freshwater demand, Hong Kong has implemented the use of seawater for toilet flushing on an extensive scale since the 1950s (Tang et al., 2007). However, in some rural areas of Hong Kong, tap water is still used for toilet flushing due to the difficulties in seawater supply. Therefore, both non-saline and saline wastewaters are present in Hong Kong. Normally, the iodine concentrations in saline wastewater should be higher than those in non-saline wastewater due to the contribution of seawater (Gong and Zhang, 2013). In this study, the concentrations of iodine species in wastewater influents from different sewage treatment plants in Hong Kong were determined, including a saline primary sewage treatment plant, a saline secondary sewage treatment plant, a non-saline tertiary sewage treatment plant, and a non-saline secondary sewage treatment plant. The results are listed in Table 1. Notably, the wastewater influent of the non-saline secondary sewage treatment plant contained a high level of total iodine (84.6 μg/L), which was even higher than those of the saline sewage treatment plants (55.6 and 49.3 μg/L), and the dominant iodine species in this non-saline wastewater was iodide (70.3 μg/L). It was out of expectation that a non-saline wastewater contained such a high level of iodine. Since non-saline wastewater is widely present all over the world, the presence of high levels of iodine in non-saline wastewater may be a critical issue which should be concerned.

Wastewater effluents of sewage treatment plants are generally disinfected to inactivate pathogens before discharge. Chlorination may be the most cost-effective method for wastewater disinfection. During chlorination of wastewater effluents, iodinated disinfection byproducts (DBPs) may be generated in the presence of iodide. Besides, iodine-containing X-ray contrast media are also precursors of iodinated DBPs during water chlorination (Matsushita et al., 2015, 2016; Tian et al., 2017; Xu et al., 2017). Previous studies have reported the formation of iodinated DBPs in chlorinated saline wastewater effluents (Gong and Zhang, 2015; Yang and Zhang, 2013). When iodinated DBPs along with wastewater effluents are discharged into the receiving water body, they may pose adverse effects to the ecosystem (Liu and Zhang, 2014; Yang and Zhang, 2013). It has been demonstrated that iodinated DBPs are much more toxic than their brominated and chlorinated analogues (Cemeli et al., 2006; Li et al., 2016; Liu and Zhang, 2014; Richardson et al., 2007, 2018; Yan et al., 2016a; Yang and Zhang, 2013). To control the formation of iodinated DBPs, the level of iodide in wastewater needs to be controlled. Since source control is an effective strategy to control the iodide level in wastewater, there is a critical need to trace the iodine sources of wastewater. Therefore, the purpose of this study was to trace the iodine sources of the non-saline wastewater containing an unexpected high level of iodine.

2. Materials and methods

2.1. Chemicals and reagents

Ascorbic acid (≥99.7%), ammonium chloride (≥99.5%) and potassium nitrate (≥99.0%) were purchased from Riedel-deHaën. A sodium hypochlorite stock solution from Sigma Aldrich was diluted to around 2000 mg/L as Cl_2 and periodically standardized by the N,N-diethyl-p-phenylene diamine (DPD) ferrous titrimetric method (APHA et al., 1995). All other chemicals used in this study were purchased at the highest purities available from Sigma Aldrich. Ultrapure water (18.2 MΩ cm) was supplied by a NANOpure Diamond purifier system (Barnstead).

2.2. Preparation of solutions

An ammonium chloride solution (1.0 g/L as N), a phenol solution (10.0 g/L as C), a sodium arsenite solution (13.0 g/L) and an ascorbic acid solution (1.0 g/L) were prepared and stored in amber glass bottles at 4 °C. The phenol, ascorbic acid and sodium arsenite solutions were newly prepared every week. A monochloramine solution was freshly prepared prior to use by gradually adding the sodium hypochlorite solution to the ammonium chloride solution at a Cl_2/N molar ratio of 0.8. Iodoacetic acid was dissolved in ultrapure water to prepare standard solutions with total organic iodine (TOI) concentrations of 5, 10, 20, 50 and 100 μg/L as I. A potassium nitrate solution (5000 mg/L as NO_3^-) was prepared as the rinsing solution for removing inorganic halides from activated carbon.

2.3. Collection of water samples

The average influent flow rate of the non-saline secondary sewage treatment plant was 81,000 m^3 per day (Hong Kong Drainage Services Department, 2009), which was mainly from Districts A and B of this area (Fig. S1 in the Supporting Information (SI)). Three wastewater influent samples were collected from the non-saline secondary sewage treatment plant in March, July and

Table 1
Concentrations of iodine species in the wastewater influents of different sewage treatment plants.

Sewage treatment plant	Total iodine (μg/L as I)	Iodide (μg/L as I)	Iodate (μg/L as I)	Organic iodine (μg/L as I)
Saline primary sewage treatment plant	55.6	43.0 (±1.5)	6.9 (±0.3)	5.7 (±0.2)
Saline secondary sewage treatment plant	49.3	44.3 (±1.2)	1.2 (±0.1)	3.8 (±0.1)
Non-saline tertiary sewage treatment plant	7.9	6.3 (±0.3)	1.6 (±0.1)	<2.5[a]
Non-saline secondary sewage treatment plant	84.6	70.3 (±2.4)	3.1 (±0.2)	11.2 (±0.2)

[a] Below the quantitation limit.