Science of the Total Environment 488–489 (2014) 168–175

Contents lists available at ScienceDirect

Science of the Total Environment

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

Distribution of artificial sweeteners in dust and soil in China and their seasonal variations in the environment of Tianjin

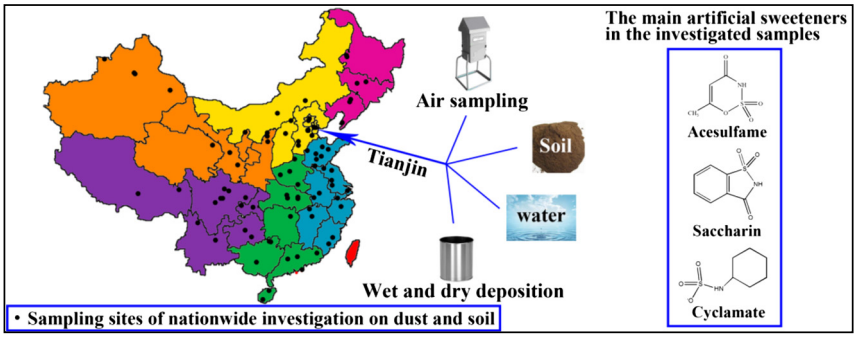
Zhiwei Gan, Hongwen Sun *, Yiming Yao, Yangyang Zhao, Yan Li, Yanwei Zhang, Hongwei Hu, Ruonan Wang

MOE Key Laboratory of Pollution Processes and Environmental Criteria, College of Environmental Science and Engineering, Nankai University, Tianjin 300071, China

HIGHLIGHTS

- Artificial sweeteners (ASs) were widely detected in Chinese outdoor dust and soil.
- Gas (particulate) phase levels of ASs were relatively higher in summer (winter).
- The AS factory and WWTP could/may act as atmospheric sources for ASs.
- Wet and dry deposition could act as sources for ASs in surface environment.
- Seasonal variations of AS levels in different waters were investigated in Tianjin.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 17 January 2014
Received in revised form 10 April 2014
Accepted 22 April 2014
Available online xxxx

Editor: Thomas Kevin V.

Keywords:
Artificial sweeteners
Atmosphere
Outdoor dust
Soil
Seasonal variations
Deposition fluxes

ABSTRACT

A nationwide investigation on the occurrence of artificial sweeteners (ASs) was conducted by collecting 98 paired outdoor dust and soil samples from mainland China. The ASs were widely detected in Chinese atmospheric dry deposition and soil samples, at concentrations up to 6450 and 1280 ng/g, respectively. To give a picture on AS distribution and source in the whole environment, the concentrations and seasonal variations of ASs in Tianjin were studied, including atmosphere, soil, and water samples. The AS levels were significantly higher in Haihe river at TJW (a sampling site in central city) in winter, while no obviously seasonal trends were obtained at BYL (close to a AS factory) and the site at a wastewater treatment plant. Saccharin, cyclamate, and acesulfame were the dominant ASs in both gas and particulate phase, with concentrations varying from 0.02 to 1940 pg/m^3. Generally, gas phase concentrations of the ASs were relatively higher in summer, while opposite results were acquired for particulate phase. Wet and dry deposition fluxes were calculated based on the measured AS levels. The results indicated that both wet and dry deposition could efficiently remove ASs in the atmosphere and act as important pollutant sources for the ASs in surface environment.

1. Introduction

Starting in 2007, great concern has been given to a new class of emerging pollutants, artificial sweeteners (ASs) (Brorström-Lundén et al., 2008; Kokotou et al., 2012; Lange et al., 2012). These sugar substitutes are used in food, beverages, sanitary products, pharmaceuticals and animal feeds over the world (Kokotou et al., 2012; Lange et al., 2012). Recent publications indicate that ASs occur widespread in surface waters, groundwater, drinking water, and wastewater treatment plants (WWTPs), at concentrations up to 120, 34, 7.2, and 200 μg/L, respectively (Kokotou et al., 2012; Lange et al., 2012; Gan et al., 2013). Among the ASs, saccharin (SAC), cyclamate (CYC), acesulfame (ACE), and sucralose (SUC) have raised more concerns due to their high

* Corresponding author at: College of Environmental Science and Engineering, Nankai University, 94 Weijin Street, Tianjin, 300071, China. Tel.: +86 22 23509241; fax: +86 22 23508807, +86 22 23509241.
E-mail address: sunhongwen@nankai.edu.cn (H. Sun).

http://dx.doi.org/10.1016/j.scitotenv.2014.04.084
0048-9697/

detection frequency or great concentrations in variety of environmental media (Kokotou et al., 2012; Lange et al., 2012).

As ASs are food additives, comprehensive toxicological tests have been conducted on human health, and they seem nontoxic to humans within acceptable daily intake values (Kroger et al., 2006). However, little is known on their potential impact on ecosystems, which merits further investigation (Lange et al., 2012; Stolte et al., 2013).

Majority of field investigations and laboratory studies on ASs have been concentrated on aquatic environment due to their high water solubility. The studies include environmental distribution, possible source, sorption, biodegradation, and photodegradation (Buerge et al., 2010; Soh et al., 2011; Kokotou et al., 2012; Lange et al., 2012; Sang et al., 2013). Owing to sewage irrigation and the use of pig manure or sludge as fertilizer that contains ASs, ASs may reach topsoil, and these also may be the potential sources of ASs in groundwater especially in agricultural areas (Buerge et al., 2010; Kokotou et al., 2012; Lange et al., 2012). Previous study implied that SAC, CYC, ACE, and SUC could be degraded in soil, with half-lives of 3–12, 0.4–6, 3–49, and 8–124 days, respectively (Buerge et al., 2010). However, to our knowledge, no information is available regarding on the environmental level of ASs in soil, which is important for understanding the fate and eco-risk of ASs.

Quite recently, we and another study reported that ASs even exist in rainwater, storm water runoff, and atmosphere samples at concentrations up to 1.3 μg/L, 1.1 μg/L, and 99 pg/m^3, respectively (Oppenheimer et al., 2012; Gan et al., 2013). It was very surprising that these polar pollutants with high water solubility could be found in rainfall and atmosphere samples. A report from the U.S. National Library of Medicine Toxnet Data Network (TOXNET, http://toxnet.nlm.nih.gov) indicated that SUC, neotame (NEO), and aspartame (ASP) are expected to exist solely in the atmospheric particulate phase, while ACE, SAC, and CYC are expected to exist in both gas phase and particulate phase in the ambient atmosphere based on their estimated vapor pressure. The potential source of the ASs in rainfall samples was grossly assessed in our previous study (Gan et al., 2013), and we suggested that ASs may reach the atmosphere by airborne dust, and more efforts are needed to understand the potential sources and temporal trends of the ASs in the atmosphere.

The objectives of this study were: 1) to confirm the extensive occurrence of ASs in soil and atmospheric samples by a nationwide investigation containing 98 paired outdoor dust samples and soil samples obtained across mainland China and Hong Kong; 2) to get a picture on AS distribution and seasonal variation in multimedia of environment by measuring the concentrations of the selected ASs in atmosphere samples (both gas and particulate phase), soil, and waters during two sampling campaign (winter and summer) in Tianjin, the third largest city in China; and 3) to grossly analyze the source and sink of ASs by estimating dry and wet deposition fluxes using ambient concentrations and assumed deposition velocities.

2. Materials and methods

2.1. Chemicals and reagents

Chemicals and materials used in this study are described in Section 1 in the Supporting Information (SI).

2.2. National survey

A total of 98 paired soil and outdoor dust samples were collected across the mainland China (one was from Hong Kong) during February to March in 2013. The detailed information of sampling campaign and sites was described in our previous study (Gan et al., 2014), and is also presented in Fig. 1, Fig. S1, Table S1, and Section 2 in the SI. The sampling sites covered most regions of China, except for Macau and Taiwan. Briefly, settled dust on exterior window sill or building surface above 1 m of the ground was sampled using a hand-held brush and a paper. Simultaneously, soil samples were collected using a polypropylene plastic (PP) tube near the dust sampling site, and 4 sub-samples of the topsoil (0–3 cm) were taken and then mixed thoroughly to obtain a bulk sample for each sampling site. They were then air dried, sieved, transferred to clean PP bottles, and stored at −20 °C until analysis.

2.3. Sampling sites

To grasp the distribution and seasonal variation of ASs in multiple media and their relationship of sink and source, ASs in different environmental media in Tianjin were investigated during two sampling campaigns. Tianjin is the third largest city in China and it currently has the most rapid economic development in China. The prevailing wind is southwest and northwest in summer and winter, respectively. Three sampling sites were chosen to evaluate the potential occurrence of the ASs based on our previous study (Fig. 1) (Gan et al., 2013). One site (BYL) is located on the southeast of one of the five national largest factories manufacturing SAC and CYC. Another site (WWTP) is situated in a WWTP alongside its aeration tank on the northeast (downwind direction in summer) of BYL. Previous investigation found high levels of ASs in rainfall samples from these two sites (Gan et al., 2013). An urban sampling site (TJW) is near the source of Haihe river in Tianjin and on the northwest of BYL, with lower AS levels in wet precipitation based on our previous study (Gan et al., 2013). This site played a role as the city background site due to that it had no known potential pollutant source.

2.4. Sample collection and preparation

Concurrent air, water, and soil samples were collected during two campaigns conducted on December 13–26, 2012 (winter) and

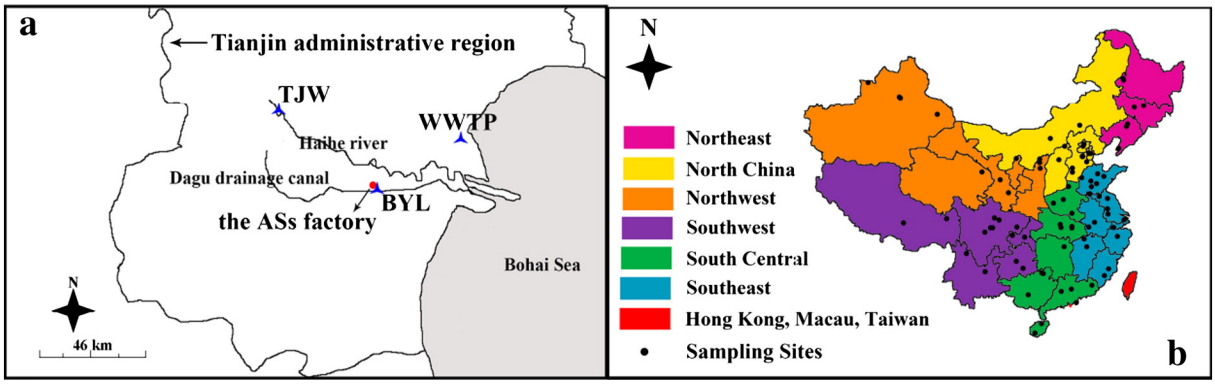


Fig. 1. Map of the sampling sites (a: sampling sites in Tianjin, b: paired dust and soil samples from mainland China). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)