Environmental Pollution xxx (2017) 1–9

Contents lists available at ScienceDirect

Environmental Pollution

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

Exposure assessment of PCDD/Fs for the population living in the vicinity of municipal waste incinerator: Additional exposure *via* local vegetable consumption☆

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ARTICLE INFO

Article history:
Received 24 November 2016
Received in revised form
15 February 2017
Accepted 16 February 2017
Available online xxx

Keywords:
Dioxin
Emission
Air
Vegetation
Local foods
Human exposure

ABSTRACT

While the exposure assessment of polychlorinated dibenzo-p-dioxin and polychlorinated dibenzofuran (PCDD/Fs) for people living in the vicinity of municipal solid waste incinerators (MSWI) has been investigated, indirect exposure to MSWI-emitted PCDD/Fs *via* consumption of local foods has not been well assessed. In this study, the PCDD/F concentration in the local vegetables grown near a MSWI located in Shenzhen, South China, was determined to be 0.92 ± 0.59 pg/g wet weight (ww), significantly higher than that (0.25 ± 0.35 pg/g ww) in commercial vegetables ($p < 0.05$). The PCDD/F concentrations in Banyan leaf (*Ficus microcarpa*) samples collected from 5 sampling sites at 1 km intervals from the MSWI were found to be significantly decreased with increasing distance, suggesting that the local plants would be impacted by emissions from the MSWI. The exposure assessment of PCDD/Fs for the population living in the vicinity of MSWI was carried out by simultaneously analyzing PCDD/Fs in other food groups that were commonly consumed by the residents. If only the local vegetables were consumed and other foods were acquired commercially, the total dietary intake for a general adult was 0.94 ± 0.41 pg TEQ/kg bw/day, of which consumption of local vegetables accounted for 52.3%. If all foods consumed including vegetables were from a commercial source, the total dietary intake was 0.56 ± 0.30 pg TEQ/kg bw/day, of which consumption of commercial vegetables accounted for 20.1%. The present study for the first time reported the additional human exposure to PCDD/Fs *via* consumption of local vegetables impacted by emissions from MSWI.

1. Introduction

Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs), commonly known as dioxins, are byproducts generally derived from industrial manufacture and incomplete combustion processes, and are especially of concern due to their immunotoxicity, reproductive and developmental toxicity, and endocrine disruption, as well as carcinogenicity toward humans (USEPA, 2004; WHO, 1998). Municipal solid waste incineration (MSWI) has been applied widely as a main municipal solid waste disposal treatment all over the world (Demirarslan et al., 2016). Even though advanced technology has been widely applied and has greatly reduced PCDD/F emissions from MSWI, MSWI is still one of the important PCDD/F emission sources due to the rapid growth of solid waste production (Dopico and Gomez, 2015; Wang et al., 2016).

Exposure assessments of PCDD/Fs for people living in the vicinity of MSWIs have been carried out, and it was concluded that MSWI-emitted PCDD/Fs should not lead to additional exposure after adequate controls are applied for PCDD/F emissions (Domingo et al., 2012; Mari et al., 2007; Nadal et al., 2004; Vilavert et al., 2010, 2012). In these exposure assessments, the direct exposure to MSWI-emitted PCDD/Fs generally included three pathways (ingestion, dermal contact with affected soil, and inhalation of affected air); however, various groups of foodstuffs collected from the market were used for the dietary exposure assessment since neither crops nor livestock in the neighborhood were available (Domingo et al., 2012; Mari et al., 2007; Nadal et al., 2004; Vilavert et al., 2010, 2012). Thus, human exposure to PCDD/Fs would be underestimated considering that MSWI-emitted PCDD/Fs can transfer to local plant-origin food through atmospheric gas-phase

☆ This paper has been recommended for acceptance by Dr. Chen Da.

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http://dx.doi.org/10.1016/j.envpol.2017.02.036
0269-7491/

partitioning and particle-phase deposition (Domingo et al., 2001; Schuhmacher et al., 2006; Welschpausch et al., 1995), and to local animal-origin food through direct exposure to MSWI-emitted PCDD/Fs and ingestion of the surrounding vegetation (Cangialosi et al., 2008; Karademir, 2004). On the other hand, some studies have made attempts to assess human exposure to MSWI-emitted PCDD/Fs *via* consumption of local foods using food-chain modeling (Fries and Paustenbach, 1990; Glorennec et al., 2005; Karademir, 2004; Nouwen et al., 2001). While such a modeling approach provides important information for understanding the exposure risk of MSWI-emitted PCDD/Fs, validation with measurements of on-site food samples has been lacking.

In this study, we determined the PCDD/F concentrations in 14 species of local vegetables collected from a garden near a MSWI, and 21 species of commercial vegetables from a market in order to assess the human exposure to PCDD/Fs for the population living in the vicinity of MSWIs. To understand whether vegetables grown near the MSWI were influenced by the emissions from the MSWI, the spatial associations of PCDD/Fs in atmospheric samples and leaves of Banyan (*Ficus microcarpa*) with distance from the MSWI were also analyzed. PCDD/Fs in other commercial food groups (meat, fish and shrimp, cereals, eggs, fruits, and milk) that were commonly consumed by the residents living around the MSWI were simultaneously analyzed, and the probable dietary exposure to PCDD/Fs for the population living in the vicinity of MSWI was evaluated. The present study can strengthen the scientific basis for understanding the health risk of human exposure to MSWI-emitted PCDD/Fs.

2. Materials and methods

2.1. Field study

The monitoring program was set up around a MSWI located in Shenzhen City, Guangdong Province, China, which was officially put into service in December 2005. The processing capacity is 1200 tons of waste per day, with annually generated power of 1.44 billion kilowatt hour. The combustion temperature of the burning treatment lines is higher than 850 °C, even as high as 1100 °C. This could guarantee the decomposition of dioxin in the incineration process. In addition, activated carbon is used in the flue gas purification system to absorb small amounts of dioxin generated in the flue gas cooling process, which would be further removed by a bag-house filter. The height of the stack is 80 m. The emission levels measured by the professional test authority are below 0.1 ng TEQ/m^3.

The study area is subject to the influence of subtropical oceanic monsoons and the four seasons are not distinct, but there is a clear dry period (from September to February) with an average precipitation of 67 mm and a wet period (from March to August) with an average precipitation of 239 mm (Meteorological Bureau of Shenzhen Municipality, 2013-2014). The mean annual temperature is 22.3 °C, and the mean annual wind speed is 2.1–3.0 m/s. North and northeast winds prevail in September to December, northeast and southeast winds prevail in January to May, as calmer winds, and southwest and northeast winds in June to August.

A vegetable garden is located about 1 km away from the MSWI (Fig. 1), and no other potential dioxin pollution sources nearby could be found based on the dioxin pollution sources listed in *Standardized Toolkit for Identification and Quantification of Dioxin and Furan Release* (United Nations Environment Programme, 2005). According to the environmental impact assessment (EIA) report (Fig. S1), the garden was close to the sensitive sites (residential area), where significantly high PCDD/F concentrations were predicted, especially in weather conditions unfavorable to air diffusion (Environmental Assessment Center of Peking University, 2003). The surface area of the vegetable garden is about 25000 m^2, and the species of commonly grown vegetables include cabbage, spinach, flowering cabbage, lettuce, sweet potato leaves, water spinach, garlic sprout, leaves of asparagus lettuce, green onion, leaf lettuce, pakchoi, cucumber, green bean and green pepper.

Based on investigation of the research areas, these local vegetables were consumed in significant amounts by the people who owned the vegetable garden. There were about 40 families living around the vegetable garden, a rough estimate of 100 people. They rarely bought any vegetables, and were generally self-sufficient. However, the rest of the population living in the area mainly acquired vegetables and other foods from a market located about 3 km from the MSWI. Almost all the commercial foodstuffs in the market are not local-grown food, and the commercial vegetables generally come from the commodity vegetable bases in Shandong and He'nan Province.

2.2. Sample collection

2.2.1. Vegetables and other food samples

In order to assess the PCDD/F dietary intake accurately, we first established a full understanding of the dietary habits of the local population through literature reading and field visits. Sample collection was then conducted from August, 2013 to August, 2014. The local vegetables planted in the garden near the MSWI were collected. Fourteen species of local vegetables were collected at two different times and the concentration of each species was expressed as the average of the two samples. Six soil samples were also collected from the garden (the top 5 cm after removing the surface litter layer, each approximately 250–400 g) to better understand the potential sources of PCDD/Fs in vegetables planted in the garden. Twenty-one species of commercial vegetables were randomly purchased from the market at two different times and the concentration of each species was expressed as the average of the two samples. The complete details of vegetables analyzed in the present study are summarized in Table 1. Other food samples that the local population usually consumed were all obtained in the market, including eight kinds of fruits which were collected at three different times, three kinds of eggs which were collected at four different times, three kinds of cereals (including rice collected at six different times, rice flour and wheat flour collected at two different times), three kinds of meat and one kind of milk which were collected at eight different times, and ten kinds of fish and shrimp, most of which were collected at three different times. All the concentrations of these samples were also expressed as the average concentrations.

2.2.2. Air samples

Air samples were collected from 5 sites monthly throughout a year (September, 2013 to August, 2014). The 5 sampling sites (A_1-A_5) were set at intervals of 1 km along the regional prevailing northeast wind direction considering the requirements of continuous electricity and permission from the residents, and A_1 was about 1 km away from the MSWI (Fig. 1). The EIA report supported that sites A_1 and A_2 were both within the areas impacted by the incinerator, and site A_2 was within the region with the maximum ground level concentration (Environmental Assessment Center of Peking University, 2003). There were some factories near these sampling sites, but according to *Standardized Toolkit for Identification and Quantification of Dioxin and Furan Release* (United Nations Environment Programme, 2005), none were potential dioxin sources. At each sampling site, a total of 1200–1500 m^3 of air was sampled using a HV-1000 sampler purchased from SIBATA Scientific Technologies, Ltd. (Saitama, Japan), drawing 500 L/min of air

Please cite this article in press as: Ben, Y., et al., Exposure assessment of PCDD/Fs for the population living in the vicinity of municipal waste incinerator: Additional exposure *via* local vegetable consumption, Environmental Pollution (2017), http://dx.doi.org/10.1016/j.envpol.2017.02.036