OCCURRENCE OF PCDD/FS IN ENVIRONMENTAL MEDIA IN THE VICINITY OF A MUNICIPAL SOLID WASTE INCINERATOR IN EASTERN CHINA

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Abstract

The occurrence of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in environmental media in the vicinity of a municipal solid waste incinerator (MSWI) in Eastern China was discussed based on previous research work. A total of 33 agricultural soil sampling sites and 6 ambient air sampling sites located within 7 km of the MSWI were annually and seasonally monitored during 2006-2007 and 2007-2008, respectively. The concentrations and profiles of PCDD/F emission sources around the MSWI including open burning of wastes (OB), traffic and hot water boilers (HWBs) were also investigated to get a better understanding of the origins of the dioxins in the study area. The results revealed relative large temporal variations of PCDD/Fs in soils (up to 1.41 ng I-TEQ kg^{-1}) and high atmospheric PCDD/F pollution (0.495 pg I-TEQ m^{-3}, mean value) in the local area. Diffuse emission sources, i.e., OB, traffic and HWBs are identified as the major sources that accounts for the occurrence of PCDD/Fs in soil and air. By contrast, the impact of MSWI on surrounding environment seems to be limited.

Introduction

Since the first detection of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the flue gas of municipal solid waste incinerator (MSWI), the emission of these compounds has stirred public concerns worldwide due to their environmental toxics and associated adverse health effects[1]. In addition to MSWIs, PCDD/Fs can also be released unintentionally through various incineration, combustion, industrial and reservoir sources[2]. Identifying the major sources of PCDD/Fs in the environment is considered the preliminary step toward efficiently controlling and reducing PCDD/F pollution[3]. With respect to mainland China, the construction of MSWIs has only been booming since 2000 due to the lack of landfill sites and unsuccessful management of composting. As a result, 67 MSWIs were running by 2005, with a total daily treatment capacity of 33 000 t. It is expected that in 2015, 200 MSWI facilities with a total daily treatment capacity of 100,000 t will be operating in the country, and annually almost 200 g I-TEQ of PCDD/Fs will be released[4], assuming average emissions at the current national legal limit of 1.0 ng I-TEQ m^{-3}. However, to date, only a few domestic studies have examined the occurrence of PCDD/Fs in the environmental sinks (e.g. soil and sediment), and are mainly focused on the schistosomiasis and E-waste recycling areas[5]. Therefore, it is urgent to fill a large data gap in order to better understand the environmental impact of the MSWI. During 2006-2008, our research group initiated modeling and monitoring of PCDD/F concentrations in different environmental media (soil, air and emission sources) in the vicinity of an MSWI in Eastern China[6-10]. The mixed urban-agricultural setting around the MSWI is a typical example of an area undergoing rapid urbanization. As a result, the local environment has been deteriorating by intense anthropogenic activities including open burning of wastes (OB), traffic and operation of the hot water boilers (HWBs). Consequently, the main objective of this study is to interpret the occurrence of PCDD/Fs in the local area based on our previous research work.

Materials and Methods

Detailed descriptions of the study area were previously reported[9]. Briefly, it is a satellite town in Eastern China, with an area of 30 km^2 and a population of 41600. The MSWI is situated just in the center of the town, adjacent to two motorways with heavy traffic and has a daily treatment capacity of 800 t. Moreover, wood/cloth

residue-fueled HWBs are prevalent in town as an important hot water supplier for local habitants. Furthermore, a large amount of household garbage and cloth residue is discarded everyday and some is directly burnt in the open dumping sites. In order to better understand the impact of the MSWI and the occurrence of other dioxin emission sources in the local environment, 33 soil sampling sites and 6 ambient air sampling sites (A1-A6) in the vicinity of the MSWI were annually and seasonally monitored during 2006-2007 and 2007-2008, respectively. The concentrations and profiles of PCDD/Fs in potential sources, i.e., MSWI, Traff (traffic), OB-H/-C (open burning of household garbage/cloth residue) and HWB-W/-C (wood/cloth residue-fueled HWB) were also investigated in the local area (Fig. 1). Detailed information about the sampling sites, techniques and analysis of PCDD/Fs for soil, air and emission sources are referred to Yan et al. (2008)[6], Xu et al. (2009)[10] and Xu et al. (2009)[9], respectively. Statistical analysis was performed by SPSS 13.0 software package, yet only for 2,3,7,8-substituted isomers (i.e., congeners) with concentrations above the limit of detection (LOD).

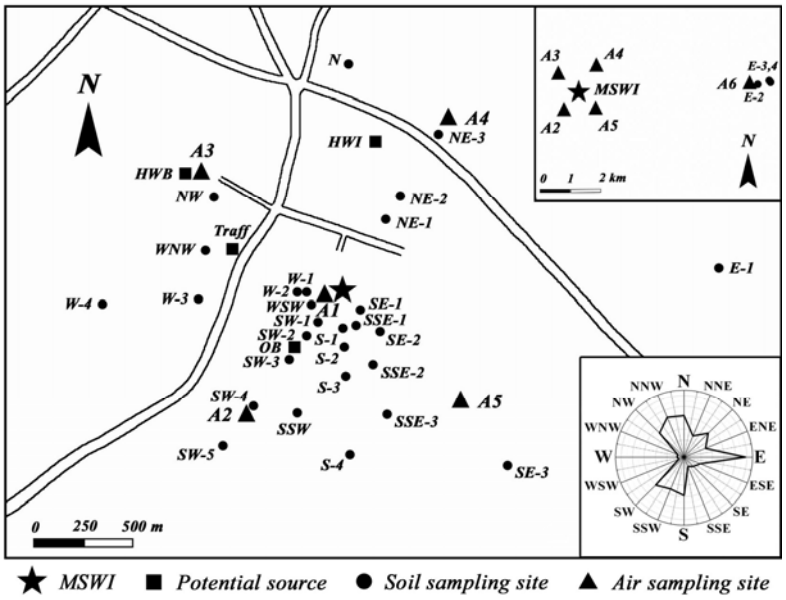

Figure 1: Distribution of soil, air and emission source samples in the vicinity of the MSWI

Results and Discussion

Concentrations of PCDD/Fs in different environmental media

The PCDD/F concentrations of soil in 2006 and 2007 ranged from 0.39 to 5.04 ng I-TEQ $kg^{-1}$and from 73.6 to 0.60 to 6.38 ng I-TEQ kg^{-1}, respectively[6,9]. According to the international guidelines and regulations for agricultural soil summarized by Leung et al. (2007)[11], PCDD/F contaminations in this study are comparatively light, with only two soil samples (i.e., WSW and NW) exceeding the stringent Canadian standard (4 ng I-TEQ kg^{-1}). However, in comparison to the initial investigation in 2006, the soil PCDD/F concentrations in 2007 as a whole increased significantly ($p < 0.01$), i.e., 33% and 39% for total concentration and I-TEQ (median value), respectively. With regard to ambient air, the concentrations varied over a factor of 51.4 from 0.059 to 3.03 pg I-TEQ m^{-3}, with an average value of 0.495 pg I-TEQ m^{-3}, and fell at the higher end of the range in the recorded peer investigations around the world[10]. Among the six monitoring sites, A2 and A3 showed comparatively high averaged PCDD/F levels, exceeding the Japanese ambient air quality standard (JAQS) of 0.6 pg I-TEQ m^{-3} for dioxins[12]. As for MSWI, the PCDD/F emission levels measured in 2007 are generally low (0.083-0.795 ng I-TEQ Nm^{-3}), compared with the national legal limit. By contrast, the diffuse emission sources show high dioxin levels[9]. The concentration of Traff was 0.439 pg I-TEQ m^{-3}, approx. 7-8 times greater than that in tunnel air in Taiwan[13]. The emission level of HWB-W was 1.95 ng I-TEQ Nm^{-3}, generally 2 orders of magnitude higher than various types of wood combustions[14], while the concentrations of OB-H (2.71 pg I-TEQ m^{-3}) and OB-C (6.14 pg I-TEQ m^{-3}) were approx. 2-5 times the backyard barrel burning of household garbage in Belgium[15] and in the

same degree of magnitude with the air around an E-waste dismantling area in China[16] and a landfill fire in Croatia[17], respectively. Fig. 2 shows the soil PCDD/F increases and atmospheric PCDD/F concentrations around the MSWI. It should be noted that only soil samples with temporal variations greater than 0.50 ng I-TEQ kg^{-1} were shown in Figure 2.

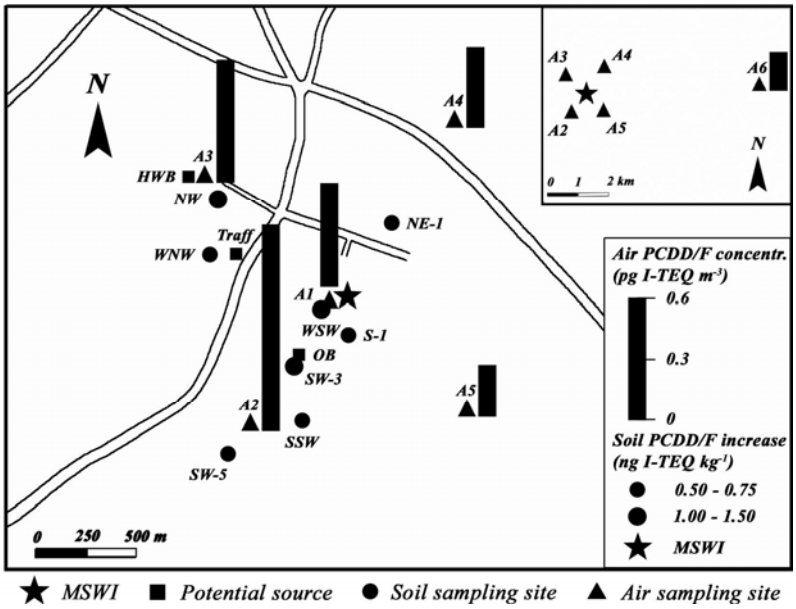

Figure 2: Soil PCDD/F increases and atmospheric PCDD/F concentrations around the MSWI

Source identification of PCDD/Fs in agricultural soils

The agricultural soils in the study area were previously proved to be almost free from two agrichemicals that have impurities of PCDD/Fs, i.e., pentachlorophenol/sodium pentachlorophenate (PCP/PCP-Na) and 1,3,5-trichloro-2-(4-nitrophenoxy) benzene (CNP)[9]. Consequently, these two agrochemicals were excluded from source identification of PCDD/Fs in soils. From a scientific point of view, literature data of unleaded gas-fueled vehicles (UGFV) from U.S. EPA[18] and city-driving diesel-fueled vehicles (DFV) from Ryan and Gullett[19] instead of sample Traff were used in statistical analysis. Actually, the congener profile of Traff was similar to that of UGFV and DFV[10]. Congener-specific factor analysis was applied to gain insight into the similarities and differences in congener profiles between the agricultural soils and reported emission sources. Since a large number of data that lower than the LOD (defined as censored values) in the variables may produce a large bias in the results, variables with more than 20% of their observations censored were excluded from the analysis[20]. Consequently, 15 congeners (except for 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD) and 62 soil samples collected in two investigations, i.e., 60 near-site ones and 2 averaged background controls and 7 emission sources, i.e., MSWI, HWB-W/-C, OB-H/-C, UGFV and DFV were regarded as variables and cases, respectively, and factor analysis was performed. Four major factors were extracted after varimax rotation with an eigenvalues greater than 1, together accounting for 84.5% of the total variance (Fig. 3). As indicated in Fig.3, soil samples as a whole has a closer relationship with OB-H/-C, DFV and UGFV than MSWI, indicating that agricultural soils within the study area were primarily influenced by diffuse emission sources including open burning of wastes and traffic. A perfect example is for sampling site SW-3. Since soil samples in the same direction from the stack, i.e., SW-1, SW-2 and SW-4, experienced even reductions of I-TEQ during 2006-2007, the highest I-TEQ increment observed in SW-3 (1.41 ng I-TEQ kg^{-1}) could not be primarily attributed to the depositions of PCDD/Fs from the MSWI. In fact, an open dumping site was discovered nearby emitting thick smoke plumes during the second soil investigation in 2007 (Fig. 1). Moreover, the SW-3 tends to be closer to the OB-H/-C in 2007 than that in 2006 (Fig. 3). It is, therefore, strong indications that soil sample SW-3 was significantly influenced by the OB. Furthermore, the road near site SW-3 was broadened several months after the initial investigation in 2006. Therefore, during 2006-2007, more impact from the emissions of traffic was expected for this site than ever

before. With respect to HWBs, soil samples seem to be more affected by wood-fueled HWBs, this is reasonable since most of the HWBs in the study area are wood-fueled.

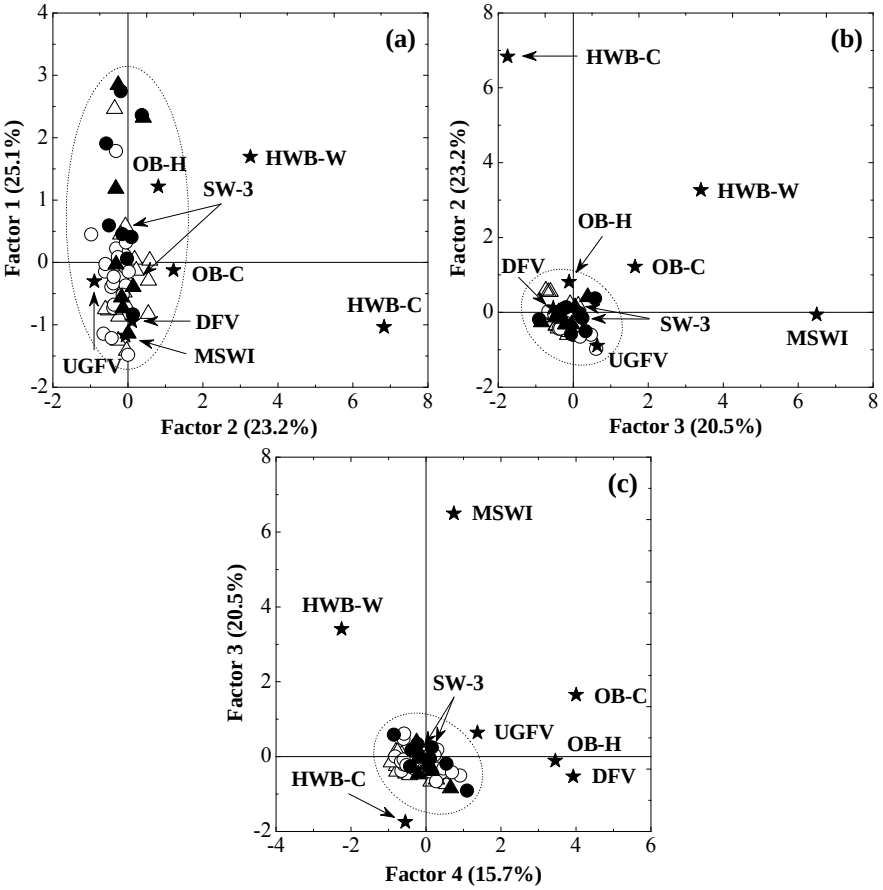

Figure 3: Congener-specific factor analysis of soils and emission sources: (a) vector 1 vs vector 2; (b) vector 2 vs vector 3; (c) vector 3 vs vector 4. The soil samples were marked with triangles (2006) and circles (2007), and those with variations lower/greater than 0.5 ng I-TEQ kg^{-1} were shown in white and dark colors, respectively; emission sources were marked with dark asterisks.

Source identification of PCDD/Fs in ambient air

Congener-specific factor analysis was also applied to gain insight into the similarities and differences in congener profiles between the ambient air and reported emission sources following the same data screening procedures as soils. Consequently, 16 congeners (except for 2,3,7,8-TCDD) and 24 ambient air samples and 7 emission sources mentioned above were regarded as variables and cases, respectively. Five major factors with eigenvalues greater than 1 were extracted after varimax rotation, accounting for 86.4% of the total variance (Fig. 4)[10]. Generally, all air samples are clustered into one large circle, with five exceeded the JAQS, i.e., A2-4, A3-4, A4-3, A1-3 and A1-4, further clustered into smaller circles with OB-C/-H. This indicates that the high atmospheric PCDD/F levels observed in this study are primarily attributed to OB. A case in study is for A2-4, which shows the highest dioxin (3.03 pg I-TEQ m^{-3}) and TSP (472 μg m^{-3}) concentration. Site A2 is situated adjacent to and downwind an open dumping site during the sampling period (Fig. 2). In fact, A2-4 shows high resemblance of congener concentration profiles to that of OB-C (also in its particulate phase)[10]. It was characterized by comparatively low abundance of OCDD and high proportions of 1,2,3,4,6,7,8-HpCDF and OCDF, being consistent with the congener pattern observed in the garden waste fire in Croatia[17]. Therefore, the high PCDD/F and TSP concentrations observed in A2-4 may mainly attribute to dispersion of particles from OB-C. The congener concentration profiles of the remaining four ambient air samples that exceeding the JAQS

were quite similar, with roughly equal amount of OCDD, 1,2,3,4,6,7,8-HpCDF and OCDF. This congener pattern was intermediate between those of OB-H/-C, indicating they might be affected by open burning of both household garbage and cloth residue. Moreover, UGFV, DFV and HWB-W/-C also exhibit good relationships with air samples, especially with lightly contaminated ones. This implies that traffic and HWBs may be significant PCDD/F contributors in ambient air. By contrast, the influence of the MSWI on the ambient air seems to remain limited. Therefore, we may conclude that the relatively large temporal variations of PCDD/Fs in agricultural soils and high atmospheric PCDD/F pollution observed in the study area were mainly attributed to OB, traffic and HWBs rather than MSWI. Consequently, effective management policies, especially for diffuse emission sources should be urgently undertaken to prevent the further deterioration of air quality in the local area.

Acknowledgements

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