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OCCURRENCE OF DIOXINS AND PCBS IN A TEMPORARY STORAGE SITE OF EARTHQUAKE AND TSUNAMI DISASTER WASTES

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Introduction

The Great East Japan Earthquake and its resultant tsunami occurred in March 2011 generated huge amounts of tsunami sediment and a multitude of wastes on the Pacific coast of the Tohoku district, Japan[1]. Disposal of non-recyclable materials and recovery of recyclable/reusable materials from the earthquake and tsunami disaster wastes were of considerable significance, both from an environmental and economic perspective. At that time, several temporary storage sites (TSSs) were provided for collections, storages, and separations of a huge amount of the disaster wastes in the Tohoku district. However, there is a concern about the potential risks for environmental and human health posed by persistent organic pollutants (POPs), which might be present in tsunami sediment, the rubble, and waste consumer products[2]. To reduce and control environmental contaminations and human exposure to POPs, particular attention should be paid to emissions and dissipation of POPs to the surrounding environments during the collection, storage, and separation of the disaster waste. However, so far there is no survey focused on levels of POPs in the environments inside and outside the TSSs.

This study investigated the levels and profiles of polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyls (PCBs) in tsunami sediment and surface soil samples collected from inside and outside of a TSS of the Great East Japan Earthquake in order to provide information on environmental emissions and dissipation of POPs during the waste treatment. Furthermore, this study attempted to estimate concentrations of dioxin TEQs in ambient air with application of pine needle biomonitoring[3], in order to provide retroactive information on emissions and dissipation of POPs from the TSS during the treatment periods.

Materials and methods

Sample collection and pretreatment.

In October 2013, two groups of tsunami sediments, eleven groups of surface soils, and six groups of pine needles were collected from inside and outside of one of the TSSs in the Tohoku district, Japan (Table 1). Each tsunami sediment and surface soil sample was composed of five subsamples, which were collected with a stainless steel shovel from a site of approximately 5 m^2 and were placed in re-sealable polyethylene zipper storage bags. The tsunami sediment and surface soil samples were air-dried and manually homogenized with a wooden hammer after removal of pebbles, weeds, and twigs. The tsunami sediment and surface soil samples were sieved to < 2.0 mm prior to long-term storage at -20°C and extraction. Each of 0.5-year-old and 1.5-year-old pine needle sample was collected from the branches from three sites around the TSS. The pine needle samples were washed with purified water to remove dusts on the surfaces, and then manually homogenized prior to extraction.

Chemical analysis.

PCDDs, PCDFs, and PCBs in tsunami sediment and surface soil samples were extracted with toluene by soxhlet extraction. PCDDs, PCDFs, and PCBs in pine needles were extracted with acetone and toluene by soxhlet extraction. A portion of each extract was cleaned up with concentrated sulfuric acid, and then it was passed through three glass columns, which were packed with multilayered silica gel, alumina, and carbon-impregnated silica gel, respectively. The cleaned-up extract was concentrated nearly to dryness and was redissolved in n-decane. PCDDs, PCDFs, and PCBs were measured using a gas chromatography system equipped with a high resolution mass spectrometer.

Estimation of TEQ concentration in ambient air around TSS.

Concentrations (pg-TEQ/m^3) of dioxin TEQs in ambient air were estimated using concentrations (pg/g) of dioxin congeners in pine needle samples and the following expression[3]:

The ages of pine needles opened in the spring of 2012 and in the spring of 2013 were 18 and 6 months, respectively. The average monthly vapor phase ratios of pine needles opened in the spring of 2012 and in the spring of 2013 were calculated using 13.87 and 18.52°C of average atmospheric temperatures[4], respectively. Detailed information on the method for estimation of dioxin TEQ concentration in ambient air is given elsewhere[3].

Results and discussion

Levels in tsunami sediments and surface soils.

Table 1 shows concentrations of dioxin TEQs, PCDDs, PCDFs, and PCBs in tsunami sediment, surface soil, and pine needle samples collected from inside and outside of a TSS in Tohoku district in this study. Concentrations of dioxin TEQs and PCBs in tsunami sediment and surface soil samples ranged 1.3–12 pg-TEQ/g and 0.035–0.29 mg/kg, respectively. In Japan, the regulation standard of dioxin TEQs includes the standard value of soil (1,000 pg-TEQ/g), the examination standard of soil (250 pg-TEQ/g to request necessary monitoring examination), and the environmental standard of sediment (150 pg-TEQ/g). For PCBs, there is a provisional removal standard (greater than 10 mg/kg: a standard whereby one is encouraged to remove polluted bottom sediment that would cause pollution of public water and fishery products). In addition, the treatment standard of PCBs for waste oil is set as 0.5 mg/kg, and waste with PCBs below this threshold is not considered as a specially controlled waste in Japan. Concentrations of dioxin TEQs and PCBs in tsunami sediment and surface soil samples in this study were lower than those of the regulation standards. According to the results of environmental investigations in Iwate, Miyagi, and Fukushima prefectures of the Tohoku district in 2006–2010[5], it was reported that concentration of dioxin TEQs and PCBs in sea sediments from the Pacific coast ranged 0.078–21 pg-TEQ/g and 0.0038–0.15 mg/kg, respectively. It was also reported that concentration of dioxin TEQs in soils ranged 0.000033–53 pg-TEQ/g. Concentrations of dioxin TEQs and PCBs in tsunami sediment and surface soil samples from inside and outside of the TSS in this study were comparable to those in sea sediments in the bottom of Pacific coast and soils from the Tohoku district in the previous reports, respectively.

Comparison of the profiles of PCDD/DF homologues indicate that tetra- and octa-CDD were the predominant homologues, accounting for 29 ± 8.9 and 40 ± 7.5% of the total PCDD/DFs in surface soil samples from inside of the TSS (Fig. 1). The extensive use of chloronitrofen (CNP) and pentachlorophenol (PCP) as herbicides for rice farming has resulted in contaminations by tetra- and octa-CDD as impurities of CNP and PCP technical formulations in sea sediments in Japan[6]. The profiles of PCDD/DF homologues in surface soil samples from inside of the TSS reflected those in sea sediments contaminated by tetra- and octa-CDD, suggesting that occurrence of dioxins and PCBs in surface soil samples from inside of the TSS might be closely associated with the sea sediments in the bottom of Pacific coast of the Tohoku district. On the other hand, the profiles of PCDD/DF homologues in the surface soil of outside the TSS reflected the atmospheric depositions[6], suggesting that dioxins and PCBs found in surface soil of outside the TSS were associated with ambient air and deposition around the Tohoku district.

Levels in ambient air.

Concentrations of dioxin TEQs in the 2012 and 2013 samples of pine needles ranged 0.39–0.53 and 0.15–0.21 pg-TEQ/g, respectively (Table 1). Concentrations of PCBs in the 2012 and 2013 samples of pine needles ranged 2,100–4,700 and 680–1,500 pg/g, respectively (Table 1). The differences of concentrations between the 2012 and 2013 samples reflected the differences in dioxin accumulations in pine needles during the time periods since the pine needles opened.

Using the concentrations of dioxins in pine needle samples, concentrations of dioxin TEQs in ambient air around the TSS during the spring of 2012 through October 2013 were estimated (Fig. 2). The mean concentrations of dioxin TEQs in ambient air nearby the TSS were 0.024 ± 0.002 pg-TEQ/m^3, whereas those at the sampling sites located 1 km north and south of the TSS were 0.014 ± 0.0015 and 0.0095 ± 0.015 pg-TEQ/m^3, respectively. According to the results of environmental surveys in Iwate and Miyagi prefectures of the Tohoku district in 2006–2013[5], it was reported that concentrations of dioxin TEQs in ambient air ranged 0.0045–0.076 pg-TEQ/m^3. Although the mean concentrations of dioxin TEQs in ambient air nearby the TSS were slightly higher than those at the sampling sites located 1 km north and south of the TSS, the mean concentrations of dioxin TEQs in ambient air around the TSS were comparable to those in Iwate, Miyagi, and Fukushima prefectures of the Tohoku district. The

results of this study suggest that the pine needle biomonitoring might be useful to understand retroactive information on emissions and dissipation of dioxin TEQs and PCBs from the TSS.

Conclusion

The results of this study provided information on occurrence of dioxins and PCBs inside and outside of the TSS during treatments of large amounts of the disaster wastes. Although the number of tsunami sediment, surface soil, and pine needle samples collected around the TSS was limited, no severe contamination by dioxins and PCBs was apparently found from this study. However, the differences of dioxin TEQ concentrations in ambient air at the sampling sites between nearby the TSS and 1 km far from the TSS were found. The differences of dioxin TEQ concentrations in ambient air suggest that the tsunami sediments and soils containing dioxins inside of the TSS might be dissipated by winds during the treatment periods. The emissions and dissipations of the sediments and soils containing POPs by winds during open storage of disaster wastes should be regarded as important contributors to occurrence of POPs in the surrounding environments of the TSS. Appropriate measures to prevent emissions and dissipations of the sediments and soils, specifically protections of winds by covering the sediments and wastes using sheets and roofs during open storage of disaster wastes, would be effective to reduce and control environmental contaminations by POPs. Furthermore, concentrations of dioxin TEQs and PCBs in the sediments after removal of wood wastes were lower than those of the regulation standards. Although other hazardous substances in the sediments, such as heavy metals and flame retardants, must be evaluated from the viewpoint of environmental and human health risks, we expected that tsunami sediments would be effectively utilized as recyclable/reusable materials, such as cement in building construction.

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Concentrations (pg-TEQ/m^3) of dioxin TEQs in ambient air were estimated using concentrations (pg/g) of dioxin congeners in pine needle samples and the following expression:

$$\text{TEQ concentration in ambient air (pg-TEQ/m}^3\text{)} = \frac{\text{concentration in pine needle (pg/g)} \times 0.1852 \times 100 \times 0.0107}{\text{needle age (month)} \times \text{average monthly vapor phase ratio in ambient air (\%)}}$$

Table 1. Concentrations of dioxin TEQs, PCDDs, PCDFs, and PCBs in tsunami sediment, surface soil, and pine needle samples collected from inside and outside of a TSS in the Tohoku district, Japan.

Sample	TEQ pg-TEQ/g	PCDDs pg/g	PCDFs pg/g	PCBs pg/g
tsunami sediments				
mix sediments with wood wastes (n = 3)	5.3	860	160	81000
sediments after removal of wood wastes (n = 1)	7.2	630	120	170000
surface soils				
mix sediments with wood waste site (n = 3)	7.9	1400	190	290000
tsunami sediment site (n = 2)	7.7	1900	310	41000
home electric appliance waste site (n = 2)	6.6	300	62	180000
fishnet waste site (n = 3)	6.7	1900	190	140000
metal waste site (n = 3)	12	1500	170	290000
polyvinyl chloride product waste site (n = 3)	2.2	470	77	45000
tire waste site (n = 2)	5.0	910	180	210000
wire and cable waste site (n = 2)	5.0	1000	130	74000
plastic waste site (n = 2)	7.5	1400	170	35000
nearby entrance (n = 1)	2.3	360	48	110000
outside of TSS (n = 1)	1.3	91	31	88000
pine needles				
2012 sample nearby TSS (n = 1)	0.53	17	26	4500
2012 sample from 1 km north of TSS (n = 1)	0.39	14	21	4700
2012 sample from1 km south of TSS (n = 1)	0.48	15	20	2100
2013 sample nearby TSS (n = 1)	0.21	6.0	9.5	1500
2013 sample from 1 km north of TSS (n = 1)	0.16	5.2	7.1	1200
2013 sample from1 km south of TSS (n = 1)	0.15	6.0	7.4	680

Fig. 1. Profiles of PCDD/DF homologues in surface soil samples collected from (a) inside (n = 23) and (b) outside (n = 1) of a TSS in the Tohoku district, Japan.

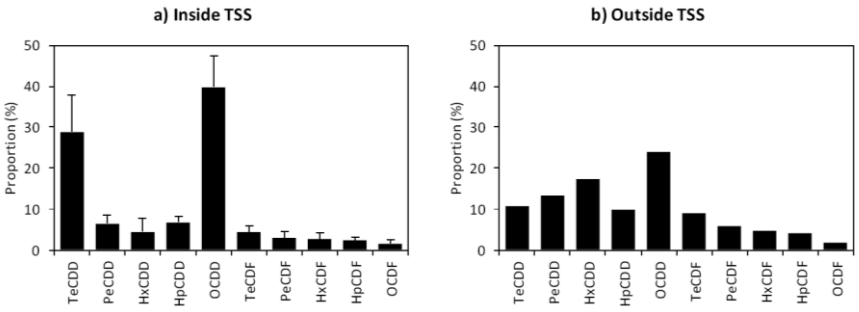

Fig. 2. Estimated concentrations of dioxin TEQs around a TSS in this study and mean concentrations of dioxin TEQs in the Tohoku district in 2006–2013.