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Atmospheric Deposition Impact of Polychlorinated Dibenzo-*p*-dioxin and Dibenzofuran on an Aquatic System in Central Taiwan

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ABSTRACT

The content and congener profiles of 17 PCDD/Fs were measured in an aquatic system in central Taiwan that included both aquatic animals and sediment. The total averaged mass content of PCDD/Fs was 0.432, 0.671, 0.244 and 0.727 pg g^{-1}, and the corresponding total PCDD/Fs-WHO_{2005}-TEQ were 0.070, 0.015, 0.024 and 0.018 pg WHO_{2005}-TEQ g^{-1} in *O. mossambica*, *M. meretrix*, *L. calcarifer* and *O. gigas*, respectively. The total PCDD/Fs in the sediment in the coastal area (72.63 ng kg^{-1}) was much higher than that in the fishponds (29.53 ng kg^{-1}), but the corresponding WHO_{2005}-TEQ values in the coastal area (0.545 ng WHO_{2005}-TEQ kg^{-1}) were lower than those in the fishponds (0.655 ng WHO_{2005}-TEQ kg^{-1}). The mass PCDD/F congener profiles of the aquatic animals and sediment samples in the fishponds and coastal area were dominated by more highly chlorinated PCDD/F congeners such as OCDD and OCDF, while the WHO_{2005}-TEQ PCDD/F congener profiles were dominated by the less-chlorinated PCDD/F congeners such as 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF. Over a time periods ranging from 30 and 60 years, the modeled PCDD/F contents in the sediment ranged between 9.27 and 18.53 ng WHO_{2005}-TEQ kg^{-1} in scenario A, and the PCDD/F contents ranged from 6.18–12.36 ng WHO_{2005}-TEQ kg^{-1} in scenario B. Compared with the observed average values of sediment in this study, the ratios of the modeled and observed values ranged from 9.4–34.0.

Keywords: PCDD/Fs; Aquatic animal; Sediment; POPs; Atmospheric deposition; Fishpond; Biota.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxin and dibenzofuran (PCDD/Fs) are persistent organic pollutants (POPs) that can remain for long periods and transport long distances in the environment due to their persistence and lipophilic properties (Welsch-Pausch *et al*., 1995). Generally, PCDD/Fs are the by-products of thermal industrial-chemical processes (Bumb *et al*., 1980), and can be produced by natural like forest fires (Prange *et al*., 2003). PCDD/Fs include 75 PCDD and 135 PCDF according to the different number and position of the chlorines. However, only the chlorine substitution in 2,3,7,8 has been shown to be much more harmful than the others. The health risk of PCDD/Fs include immunotoxicity, carcinogenicity, chloracne and endocrine disruptor (Bertazzi *et al*., 2001; Schecter *et al*., 2006; Yang *et al*., 2015). Among the 17 toxic congeners of PCDD/Fs, the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) is the most toxic (Mitrou *et al*., 2001), and it will take at least 7 years to remove the half-life of 2,3,7,8-TCDD in humans (Cole *et al*., 2003).

Ambient air is the most important pathway for the transfer of PCDD/Fs (Lohmann and Jones, 1998; Kao *et al*., 2006; Lee *et al*., 2009). Since they are released into the atmosphere, they can disperse long distances and eventually reach water systems, soil and plants via dry and wet deposition, and can eventually bioaccumulate in the food chain, particularly in the case of fat (Wild *et al*., 1994; Welsch-Pausch *et al*., 1995; McLachlan *et al*., 1996; Halsall *et al*., 1997; Lohmann and Jones, 1998; Ren *et al*., 2007). There are two types of atmospheric deposition, dry and wet, that serve as the major removal mechanism for PCDD/Fs (Koester and Hites, 1992). Gas-particle partitioning plays an important role on the deposition process of PCDD/Fs, while meteorological conditions such as the ambient temperature, wind direction and velocity, humidity and total

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suspended particle concentration can affect gas-particle partitioning (Pankow, 1994; Lohmann *et al*., 1999; Chang *et al*., 2004; Zhu *et al*., 2017a). Ingestion, inhalation and dermal contact are the PCDD/Fs exposure pathway, but previous studies have suggested that more than 90% of human exposure occurs through the food-chain (Charnley and Doull, 2005; Correa *et al*., 2006). Because of the high risk to human health, tolerable daily intake (TDI) values for PCDD/Fs were set by the World Health Organization (WHO) as 1–4 pg WHO_{2005}-TEQ kg^{-1} bw day^{-1} (WHO, 1998). Many studies have evaluated the potential health risks associated with daily intake of PCDD/Fs (Eduljee and Jackson, 1994; Liem *et al*., 2000; Fernandez *et al*., 2004; Kiviranta *et al*., 2004).

Since the daily intake of PCDD/Fs were studied for the first time in Taiwan (Hsu *et al*., 2002), the potential adverse effect on the ambient environment and human health due to the emission of PCDD/Fs has been a public concern in Taiwan. Taiwan's government chose incineration as the main waste disposal method due to the high population density. Generally, the PCDD/Fs are produced by the human activities, including industrial and heat-treatment processes (Addink and Altwicker, 2001; Lin *et al*., 2014; Cheruiyot *et al*., 2016). Previous studies have indicated that the highest PCDD/Fs TEQ levels occurred in fish and shellfish (Hsu *et al*., 2002; Sasamoto *et al*., 2006). In this context, it is important to measure the POPs in aquatic systems, and to evaluate the content of POPs accumulated in animals and sediments.

The objectives of this study were to investigate the potential effect of atmospheric dry and wet deposition of PCDD/Fs on the animals in Taiwan. Two biotas and four aquatic animals were chosen as the study objects: *Ostrea gigas* Thunberg on the coast, and *Meretrix meretrix* Linnaeus, *Oreochromis mossambica* and *Lates calcarifer* in fishponds. We analyzed 17 PCDD/Fs in the sediment and aquatic animal samples in the two biotas, and modeled the total deposition over a 30 to 60 year period to test the effect of PCDD/F deposition (dry and wet deposition) on the sediment and aquatic animals in the two biotas. The results of this study can provide further understanding of human exposure to PCDD/Fs.

MATERIAL AND METHOD

Biota and Sample Collection

Two biotas and four aquatic animal samples were used to evaluate PCDD/F contents in Taiwan: *O. gigas* on the coast, and *M. meretrix*, *O. mossambica* and *L. calcarifer* in fishponds, and the corresponding sediment samples in the two biotas were also analyzed. *O. gigas* is a large oyster native to Japan that was introduced along the Pacific coast of the United States. The marine clam *M. meretrix* is a species of mollusk that lives on the loose bottom of water bodies. *O. mossambica* is a kind of freshwater teleost and native to southern Africa. *L. calcarifer* is native to coastal areas in the Indian and Western Pacific Oceans. It is a species of freshwater fish that migrates to salt water to breed. These aquatic animals are a favorite food not only in Taiwan, but also in other countries. For example, 40000 lbs *L. calcarifer* were shipped per week in Australia in 2006 (Pierce, 2006). The *O. gigas* were collected from the shallow sea in Taisi, Sihhu and Kouhu from November 2013 to August 2014. The *M. meretrix, O. mossambica* and *L. calcarifer* were collected from fishponds in Mailiao, Taisi, Sihhu, Kouhu, Dongshih and Shueilin from November 2013 to May 2014. The detailed collection data is provided in Table 1.

Sample Pretreatment

Fresh tissues from the aquatic animal samples were cut into small pieces with stainless steel scissors after brushing any dirt and soil particles from their surfaces. All samples

Table 1. The sample locations in Taiwan.

Biota	Sample type	Longitude	Latitude	Location
Fishpond	sediment/animal	120.23	23.80	Mailiao
	sediment/animal	120.21	23.76	Mailiao
	sediment/animal	120.25	23.78	Mailiao
	sediment/animal	120.19	23.75	Taisi
	animal	120.19	23.73	Taisi
	sediment/animal	120.18	23.71	Taisi
	sediment/animal	120.21	23.73	Taisi
	sediment/animal	120.18	23.70	Sihhu
	sediment	120.15	23.62	Kouhu
	sediment/animal	120.15	23.60	Kouhu
	sediment	120.21	23.55	Shueilin
	sediment/animal	120.21	23.52	Shueilin
	sediment/animal	120.24	23.71	Dongshih
Sea	sediment/animal	120.17	23.72	Taisi
	sediment/animal	120.16	23.72	Taisi
	sediment/animal	120.15	23.66	Sihhu
	sediment/animal	120.15	23.66	Sihhu
	sediment/animal	120.14	23.62	Kouhu
	sediment/animal	120.14	23.56	Kouhu

were freeze-dried, ground in a mill and homogenized before solvent extraction. Each tissue sample (30–100 g) then was ground and homogenized in a food mill with anhydrous sodium sulfate and extracted for 10 h with dichloromethane, in a Soxhlet apparatus. After extraction, the samples were concentrated through rotary evaporation and the lipid content was determined gravimetrically. The extracted lipids re-dissolved in hexane, were treated with concentrated sulfuric acid, followed by a series of sample cleanup and fractionation procedures, including multilayered silica, alumina, and activated carbon columns. The soil samples were passed through a 2 mm (16 mesh) sieve to remove large rocks and other matters. Then the samples were freeze-dried, ground in a mill and homogenized. Finally, the samples were Soxhlet-extracted using toluene for 24 h. Subsequently, the sample solutions were treated with the above cleanup and fractionation procedures.

Analysis of PCDD/Fs

All chemical analyses were conducted in the Super Micro Mass Research and Technology Centre at Cheng Shiu Institute of Technology, which has passed the international intercalibration standards test on PCDD/Fs in fly ash, sediment, mother's milk, human blood, and cod liver. Each sample was cleaned up with the use of an activated carbon column, and elution with 25 mL of DCM: hexane (1:24, v/v); the activated carbon column was sequentially eluted with 5 mL of toluene: methanol: ethyl acetate: hexane (1:1:2:16, v/v/v/v), followed by 40 mL of toluene. Finally, the eluate was concentrated to approximately 1 mL and transferred to a vial and then further concentrated to near dryness using a stream of nitrogen. Immediately before injection, 10 mL of the standard solution for evaluating recovery was added to the sample extract to minimize the possibility of loss.

High-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for the PCDD/F analysis. The HRGC (Hewlett-Packard 6970 Series gas, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA) with splitless injection, and the HRMS (Micromass Autospec Ultima, Manchester, UK) had a positive electron impact (EI+) source. The selected ion monitoring (SIM) analyzer mode was used with the resolution power set at 10,000. The electron energy and source temperature were set at 35 eV and 250°C, respectively. Helium was used as the carrier gas. The protocol for quality analysis/quality control was strictly followed.

RESULTS AND DISCUSSION

PCDD/F Content in Aquatic Animals

Considering the difference in the fishpond and coastal areas, aquatic fauna in the two different habitants were chosen for analysis. For all samples in the fishponds, the PCDD/F content differed between sample sites and species. There were six sample sites for *M. meretrix*, four sample sites for *O. mossambica*, and only one sample site for *L. calcarifer.* The mass content of 17 PCDD/Fs in the animals in the fishponds are present in Table 2. In November 2013, the mass content of the PCDD/Fs in the three animals was in the following order: *M. meretrix* (0.687 pg g^{-1}), *O. mossambica* (0.321 pg g^{-1}) and *L. calcarifer* (0.168 pg g^{-1}); the total WHO_{2005}-TEQ PCDD/Fs was in the following order: *O. mossambica* (0.068 pg WHO_{2005}-TEQ g^{-1}), *L. calcarifer* (0.024 pg WHO_{2005}-TEQ g^{-1}) and *M. meretrix* (0.013 pg WHO_{2005}-TEQ g^{-1}). In May 2014, the total PCDD/Fs in the three animals was in the following order: *M. meretrix* (0.655 pg g^{-1}), *O. mossambica* (0.541 pg g^{-1}) and *L. calcarifer* (0.320 pg g^{-1}); the total WHO_{2005}-TEQ was in the following

Table 2. Observed PCDD/Fs content in aquatic animal from the fishpond.

	November 2013			May 2014		
	O. mossambica	*M. meretrix*	*L. calcarifer*	*O. mossambica*	*M. meretrix*	*L. calcarifer*
2,3,7,8-TeCDD	0.016	0.002	0.004	0.015	0.002	0.005
1,2,3,7,8-PeCDD	0.019	0.002	0.009	0.019	0.004	0.008
1,2,3,4,7,8-HxCDD	0.004	0.002	0.002	0.005	0.003	0.002
1,2,3,6,7,8-HxCDD	0.008	0.003	0.004	0.010	0.006	0.005
1,2,3,7,8,9-HxCDD	0.003	0.002	0.002	0.005	0.005	0.003
1,2,3,4,6,7,8-HpCDD	0.008	0.015	0.009	0.037	0.069	0.035
OCDD	0.015	0.095	0.048	0.069	0.162	0.076
2,3,7,8-TeCDF	0.126	0.009	0.027	0.119	0.009	0.024
1,2,3,7,8-PeCDF	0.033	0.008	0.011	0.039	0.008	0.010
2,3,4,7,8-PeCDF	0.055	0.009	0.019	0.062	0.009	0.018
1,2,3,4,7,8-HxCDF	0.004	0.010	0.004	0.007	0.009	0.004
1,2,3,6,7,8-HxCDF	0.007	0.009	0.005	0.013	0.009	0.006
2,3,4,6,7,8-HxCDF	0.005	0.007	0.003	0.012	0.016	0.007
1,2,3,7,8,9-HxCDF	0.001	0.001	0.002	0.004	0.002	0.002
1,2,3,4,6,7,8-HpCDF	0.009	0.058	0.010	0.045	0.104	0.036
1,2,3,4,7,8,9-HpCDF	0.002	0.009	0.002	0.012	0.020	0.006
OCDF	0.007	0.445	0.008	0.070	0.220	0.072
PCDD/Fs (pg g^{-1})	0.321	0.687	0.168	0.541	0.655	0.320
PCDD/Fs (pg WHO_{2005}-TEQ g^{-1})	0.068	0.013	0.024	0.071	0.017	0.024

order: *O. mossambica* (0.071 pg WHO_{2005}-TEQ g^{-1}), *L. calcarifer* (0.024 pg WHO_{2005}-TEQ g^{-1}) and *M. meretrix* (0.017 pg WHO_{2005}-TEQ g^{-1}). The averaged mass content of PCDD/Fs was 0.432, 0.671 and 0.244 pg g^{-1} in *O. mossambica*, *M. meretrix* and *L. calcarifer*, and the corresponding WHO_{2005}-TEQ total PCDD/Fs was 0.070, 0.015 and 0.024 pg WHO_{2005}-TEQ g^{-1} in *O. mossambica*, *M. meretrix* and *L. calcarifer*. On the whole, both the mass and WHO_{2005}-TEQ content of total PCDD/Fs are higher in the warm season (May 2014) than in the cold season (November 2013). Unlike the other two animals, the mass content of the PCDD/Fs in *M. meretrix* showed a minor decrease from November 2013 to May 2014, which may have been due to its low fat content. The WHO_{2005}-TEQ content showed a small increase.

The mass content of 17 PCDD/Fs in animals in the coastal areas are present in Table 3. The total mass and WHO_{2005}-TEQ content of PCDD/Fs fluctuated significantly over a year. The total mass content of PCDD/Fs are in the following order in February 2014 (1.162 pg g^{-1}), November 2013 (0.781 pg g^{-1}), August 2014 (0.633 pg g^{-1}) and May 2014 (0.331 pg g^{-1}). The results for the WHO_{2005}-TEQ content were slightly different from that for mass content, for which the values were in the following order: August 2014 (0.024 pg WHO_{2005}-TEQ g^{-1}), February 2014 (0.022 pg WHO_{2005}-TEQ g^{-1}), May 2014 (0.016 pg WHO_{2005}-TEQ g^{-1}) and November 2013 (0.008 pg WHO_{2005}-TEQ g^{-1}). Generally, concentrations in the ambient air fluctuate with environmental temperature. The results indicated that the PCDD/F concentrations were higher in the cold season than in the warm season (Chen *et al*., 2017; Zhu *et al*., 2017a, b). The averaged mass content of PCDD/Fs was 0.727 pg g^{-1}, and the corresponding WHO_{2005}-TEQ content was 0.018 pg WHO_{2005}-TEQ g^{-1}. However, the PCDD/F content in the aquatic animals in the fishponds and coastal areas did not exhibit the similar trends in ambient air, which may have been due to the fact that the PCDD/F content in animals is not only affected by the atmospheric deposition from air, but also by feed (Isosaari *et al*., 2002; Hoogenboom *et al*., 2010, 2015).

PCDD/Fs Congener Profile in Aquatic Animals

The PCDD/F congener profiles in the animals in the fishponds based on WHO_{2005}-TEQ fraction are shown in Fig. 1. The results show that the congener profiles in *O. mossambica* were dominated by the 1,2,3,7,8-PeCDD (27.7% and 26.2% in November 2013 and May 2014, respectively), 2,3,4,7,8-PeCDF (24.3% and 26.1% in November 2013 and May 2014, respectively), 2,3,7,8-TeCDD (23.3% and 20.4% in November 2013 and May 2014, respectively) and 2,3,7,8-TeCDF (18.6% and 16.7% in November 2013 and May 2014, respectively). The WHO_{2005}-TEQ PCDD/F congener profiles in *M. meretrix* were largely dominated by 2,3,4,7,8-PeCDF (21.7% and 26.1% in November 2013 and May 2014, respectively), 1,2,3,7,8-PeCDD (19.2% and 21.5% in November 2013 and May 2014, respectively), 2,3,7,8-TeCDD (15.2% and 13.1% in November 2013 and May 2014, respectively). The WHO_{2005}-TEQ PCDD/F congener profiles in *L. calcarifer* were largely dominated by 1,2,3,7,8-PeCDD (36.3% and 32.7% in November 2013 and May 2014, respectively), 2,3,4,7,8-PeCDF (23.8% and 22.2% in November 2013 and May 2014, respectively), 2,3,7,8-TeCDD (17.2% and 18.4% in November 2013 and May 2014, respectively). The PCDD/F congener profiles in the animals in the fishponds based on mass fraction are shown in Fig. 2. The mass PCDD/F congener profiles in *O. mossambica* were dominated by the 2,3,7,8-TeCDF (39.4% and 22.0% in November 2013 and May 2014, respectively), 2,3,4,7,8-PeCDF (17.2% and 11.5% in November 2013 and May 2014, respectively), 1,2,3,7,8-PeCDF (10.1% in November 2013), OCDF (13.0% in May 2014) and OCDD (12.8% in May 2014). The mass

Table 3. Observed PCDD/Fs content in *Ostrea gigas* in the coastal areas.

	Nov. 2013	Feb. 2014	May. 2014	Aug. 2014
2,3,7,8-TeCDD	0.001	0.003	0.003	0.004
1,2,3,7,8-PeCDD	0.002	0.004	0.005	0.005
1,2,3,4,7,8-HxCDD	0.001	0.002	0.003	0.003
1,2,3,6,7,8-HxCDD	0.002	0.005	0.003	0.007
1,2,3,7,8,9-HxCDD	0.002	0.004	0.003	0.004
1,2,3,4,6,7,8-HpCDD	0.023	0.042	0.019	0.032
OCDD	0.585	0.721	0.155	0.324
2,3,7,8-TeCDF	0.005	0.023	0.014	0.036
1,2,3,7,8-PeCDF	0.004	0.019	0.012	0.025
2,3,4,7,8-PeCDF	0.006	0.015	0.011	0.017
1,2,3,4,7,8-HxCDF	0.003	0.013	0.005	0.008
1,2,3,6,7,8-HxCDF	0.004	0.016	0.005	0.009
2,3,4,6,7,8-HxCDF	0.004	0.015	0.008	0.013
1,2,3,7,8,9-HxCDF	0.001	0.007	0.005	0.011
1,2,3,4,6,7,8-HpCDF	0.021	0.046	0.019	0.024
1,2,3,4,7,8,9-HpCDF	0.003	0.038	0.008	0.011
OCDF	0.114	0.189	0.055	0.101
PCDD/Fs (pg g^{-1})	0.781	1.162	0.331	0.633
PCDD/Fs (pg WHO_{2005}-TEQ g^{-1})	0.008	0.022	0.016	0.024

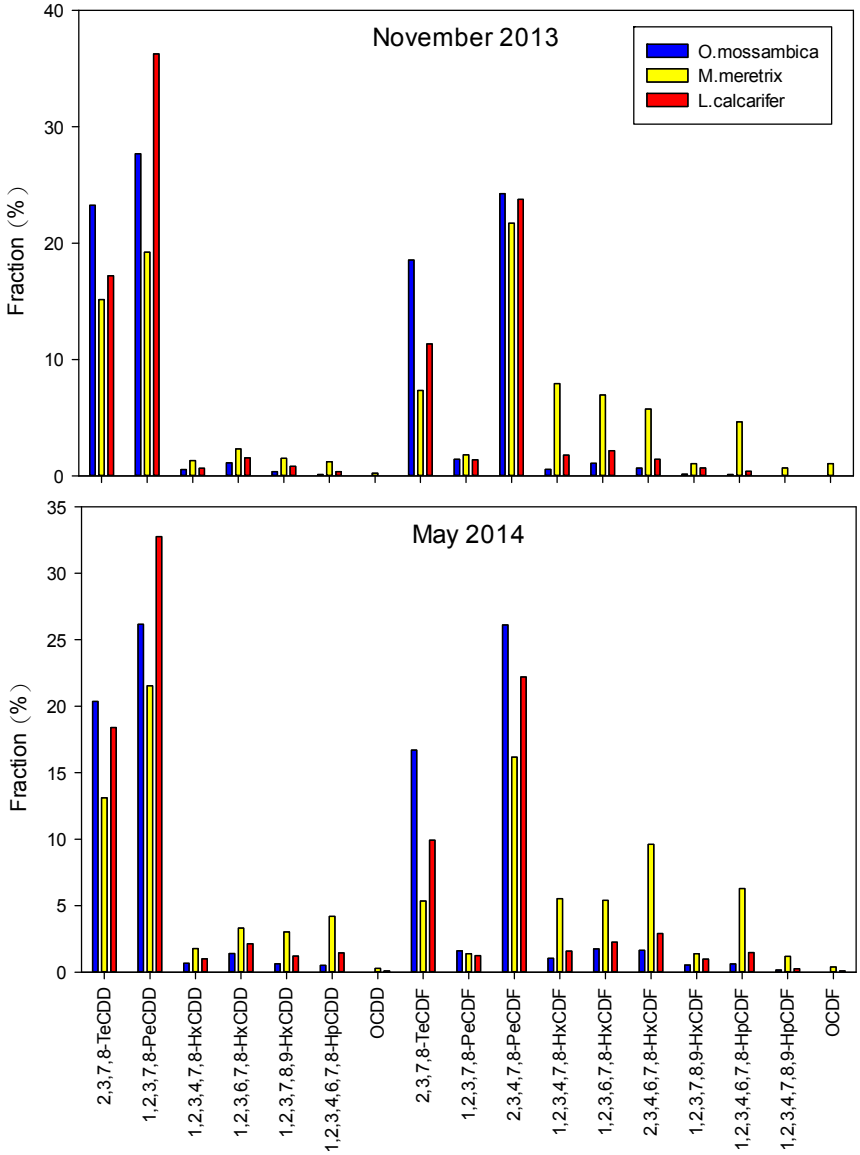

Fig. 1. The congener profile of WHO_{2005}-TEQ PCDD/Fs in aquatic animals from the fishpond.

PCDD/F congener profiles in *M. meretrix* were dominated by the higher chlorinated PCDD/F congeners like OCDD (64.9% and 33.6% in November 2013 and May 2014, respectively), OCDF (13.8% and 24.8% in November 2013 and May 2014, respectively) and 1,2,3,4,6,7,8-HpCDF (8.5% and 15.8% in November 2013 and May 2014, respectively). The mass PCDD/F congener profiles in *L. calcarifer* were dominated by the OCDD (28.6% and 23.8%), OCDF (22.6% in May 2014), 2,3,7,8-TeCDF (16.2% in November 2013), 2,3,4,7,8-PeCDF (11.3% in November 2013), 1,2,3,4,6,7,8-HpCDF (11.2% in May 2014).

The PCDD/F congener profiles in the animals in the coastal areas based on WHO_{2005}-TEQ fraction are shown in Fig. 3. The WHO_{2005}-TEQ PCDD/F congener profiles in *O. gigas* were dominated by the lower chlorinated PCDD/F congeners such as 1,2,3,7,8-PeCDD (24.5%, 17.6%, 28.1% and 19.2% in November 2013, February 2014, May 2014 and August 2014, respectively) and 2,3,4,7,8-PeCDF (20.9%, 20.2%, 21.4% and 21.7% in November 2013, February 2014, May 2014 and August 2014, respectively), 2,3,7,8-TeCDD (17.4%, 14.8%, 16.8% and 14.6% in November 2013, February 2014, May 2014 and August 2014, respectively) and 2,3,7,8-TeCDF (6.1%, 10.3%, 8.9% and 15.2% in November 2013, February 2014, May 2014 and August 2014, respectively). The PCDD/F congener profiles in the animals in the coastal area based on mass fraction are shown in Fig. 4. The mass PCDD/F congener profiles in *O. gigas* were dominated by the higher chlorinated PCDD/F congeners such as OCDD (75.0%, 62.2%, 46.8% and 51.1% in November 2013, February 2014, May 2014 and August 2014, respectively) and OCDF (14.6%, 16.3%, 16.6% and 16.0% in November 2013, February 2014, May 2014 and August 2014, respectively).

On the whole, the mass PCDD/F congener profiles of the animal samples in the fishponds and coastal area were dominated by more highly chlorinated PCDD/F congeners

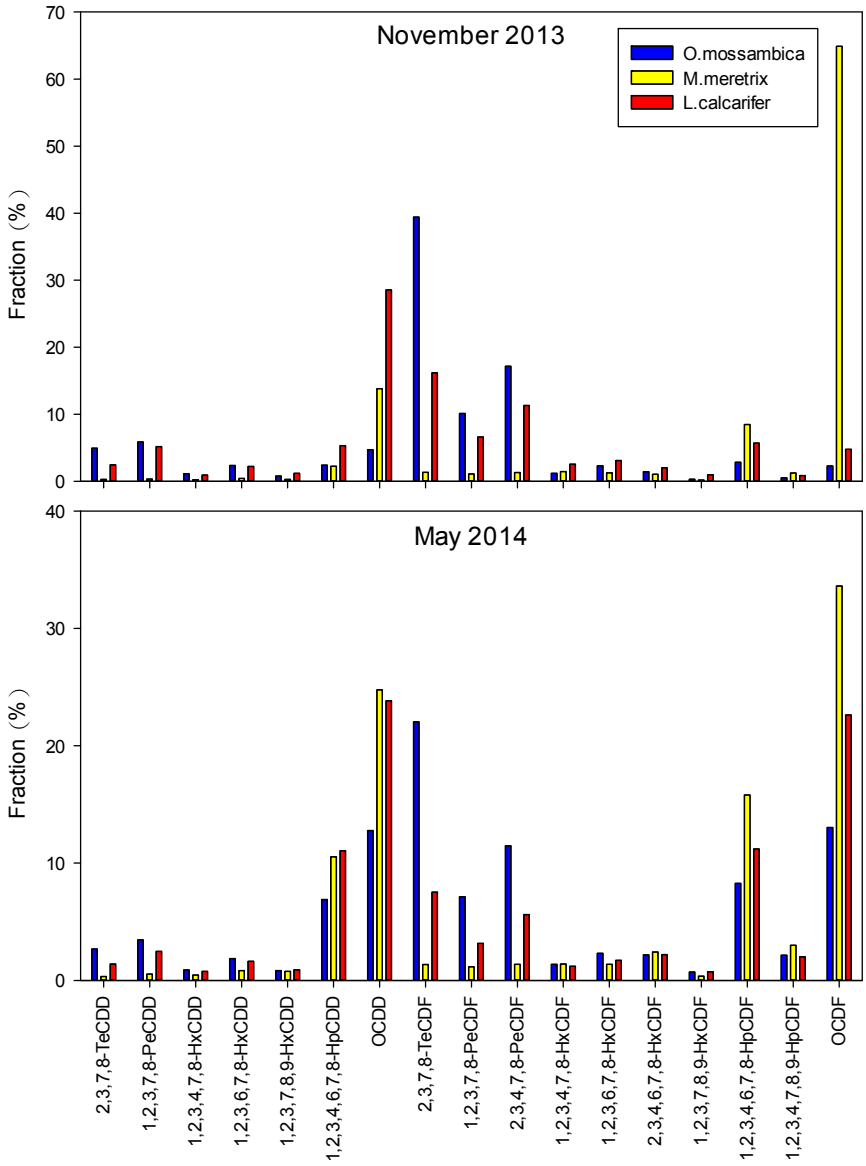

Fig. 2. The congener profile of mass PCDD/Fs in aquatic animals from the fishpond.

such as OCDD and OCDF, for which the similar results have been detected in krill and fish (Corsolini *et al*., 2002; Bengtson Nash *et al*., 2008), but for which different results have been obtained for penguin and eggs of the Adélie penguin (Kumar *et al*., 2002; Mwangi *et al*., 2016). The WHO_{2005}-TEQ PCDD/F congener profiles of the animal samples in the fishponds and coastal area were dominated by less chlorinated PCDD/F congeners such as 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF.

PCDD/F Content in Sediment

The mass content of 17 PCDD/Fs in the sediment in the fishponds are presented in Table 4. The mass content of total PCDD/Fs in the sediment was 56.09, 22.10, 17.99 and 21.94 ng kg^{-1} in November 2013, March 2014, May 2014 and July 2014, respectively, with an average of 29.53 ng kg^{-1}. The corresponding WHO_{2005}-TEQ content of PCDD/Fs was 1.309, 0.426, 0.426 and 0.457 ng WHO_{2005}-TEQ kg^{-1} in November 2013, March 2014, May 2014 and July 2014, respectively, with an average of 0.655 ng WHO_{2005}-TEQ kg^{-1}.

The content of 17 PCDD/Fs in the sediment in the coastal areas is presented in Table 5. The mass content of total PCDD/Fs in the sediment was 63.73, 74.10 and 80.05 ng kg^{-1} in November 2013, February 2014 and May 2014, respectively, with an average of 72.63 ng kg^{-1}. The corresponding WHO_{2005}-TEQ content of PCDD/Fs was 0.623, 0.446 and 0.565 ng WHO_{2005}-TEQ kg^{-1} in November 2013, February 2014 and May 2014, respectively, with an average of 0.545 ng WHO_{2005}-TEQ kg^{-1}. For most sites along the coast line, the mass concentrations of total PCDD/Fs increased with time, which may have been due to the accumulation of PCDD/F deposition.

The mass content of total PCDD/Fs in the sediment did not show seasonal variations in the fishponds, which may have been because the PCDD/F content in the fishponds in mainly influenced by anthropogenic factors. The total mass content of

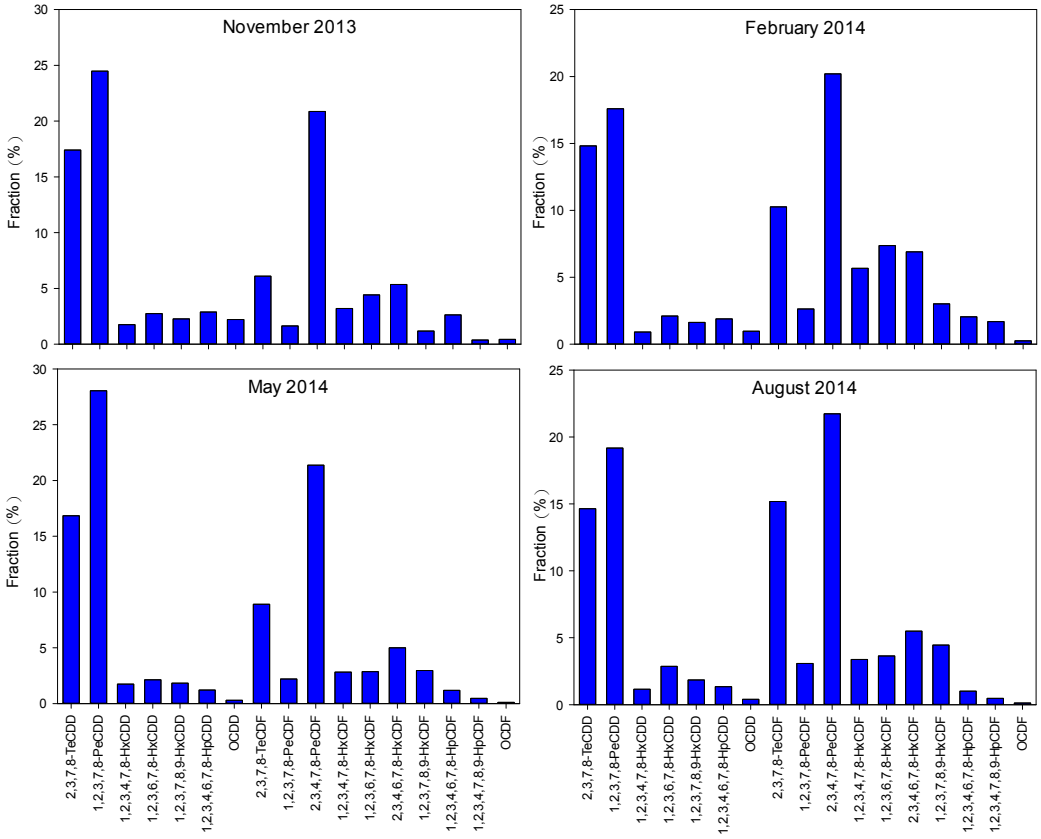

Fig. 3. The congener profile of WHO_{2005}-TEQ PCDD/Fs in aquatic animals from the coastal area.

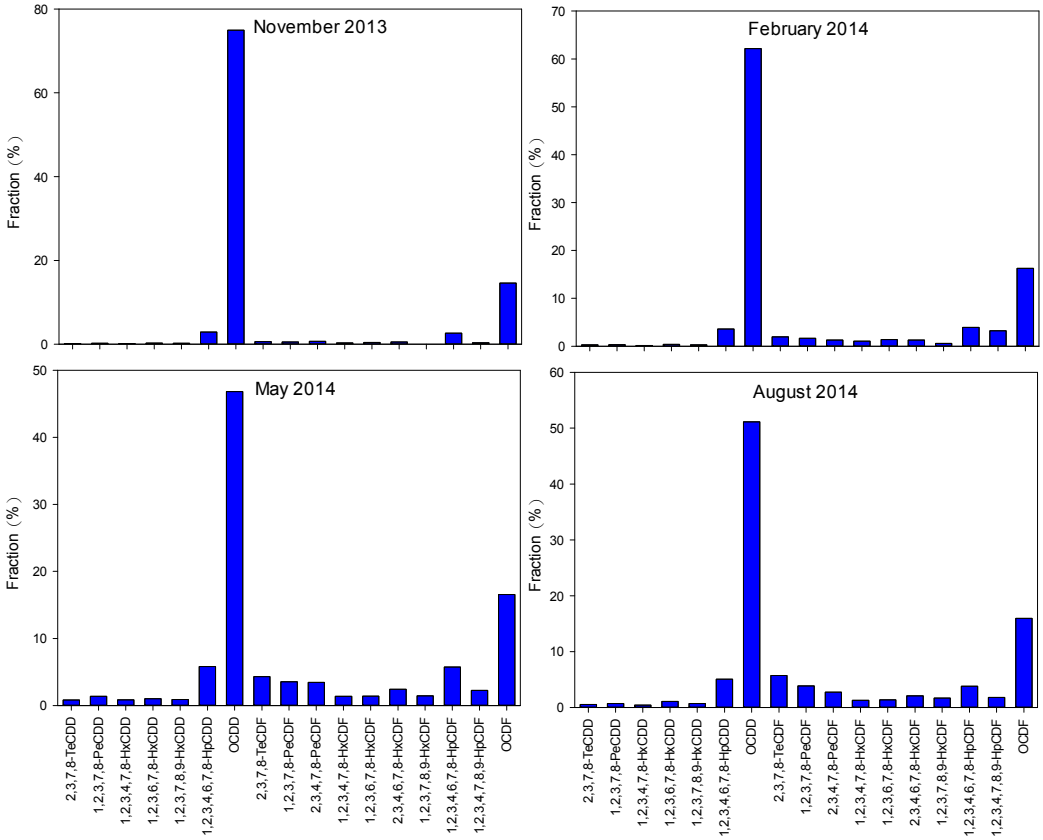

Fig. 4. The congener profile of mass PCDD/Fs in aquatic animals from the coastal area.

Table 4. Observed PCDD/Fs content in sediment from the fishpond.

	Nov. 2013	Mar. 2014	May. 2014	Jul. 2014
2,3,7,8-TeCDD	0.089	0.064	0.068	0.088
1,2,3,7,8-PeCDD	0.214	0.095	0.100	0.129
1,2,3,4,7,8-HxCDD	0.164	0.052	0.052	0.066
1,2,3,6,7,8-HxCDD	0.416	0.109	0.116	0.123
1,2,3,7,8,9-HxCDD	0.301	0.086	0.105	0.109
1,2,3,4,6,7,8-HpCDD	3.623	0.913	0.783	1.407
OCDD	15.909	7.558	5.943	7.862
2,3,7,8-TeCDF	0.384	0.174	0.246	0.193
1,2,3,7,8-PeCDF	0.762	0.259	0.254	0.226
2,3,4,7,8-PeCDF	0.788	0.195	0.229	0.188
1,2,3,4,7,8-HxCDF	1.758	0.463	0.369	0.364
1,2,3,6,7,8-HxCDF	1.381	0.360	0.299	0.269
2,3,4,6,7,8-HxCDF	1.477	0.296	0.258	0.246
1,2,3,7,8,9-HxCDF	0.141	0.067	0.052	0.038
1,2,3,4,6,7,8-HpCDF	8.555	2.224	1.665	1.446
1,2,3,4,7,8,9-HpCDF	1.161	0.407	0.295	0.245
OCDF	18.994	8.785	7.169	8.927
PCDD/Fs (ng kg^{-1})	56.09	22.10	17.99	21.94
PCDD/F (ng WHO_{2005}-TEQ kg^{-1})	1.309	0.426	0.426	0.457

Table 5. Observed PCDD/Fs content in sediment in the coastal areas.

	Nov. 2013	Feb. 2014	May. 2014
2,3,7,8-TeCDD	0.077	0.055	0.074
1,2,3,7,8-PeCDD	0.148	0.089	0.130
1,2,3,4,7,8-HxCDD	0.085	0.074	0.088
1,2,3,6,7,8-HxCDD	0.204	0.213	0.236
1,2,3,7,8,9-HxCDD	0.198	0.181	0.222
1,2,3,4,6,7,8-HpCDD	2.723	3.245	3.420
OCDD	46.030	53.400	60.500
2,3,7,8-TeCDF	0.264	0.180	0.243
1,2,3,7,8-PeCDF	0.412	0.201	0.234
2,3,4,7,8-PeCDF	0.325	0.201	0.294
1,2,3,4,7,8-HxCDF	0.514	0.292	0.366
1,2,3,6,7,8-HxCDF	0.485	0.264	0.296
2,3,4,6,7,8-HxCDF	0.379	0.317	0.329
1,2,3,7,8,9-HxCDF	0.061	0.042	0.074
1,2,3,4,6,7,8-HpCDF	2.248	2.356	2.207
1,2,3,4,7,8,9-HpCDF	0.304	0.257	0.278
OCDF	9.313	12.662	11.220
PCDD/Fs (ng kg^{-1})	63.73	74.10	80.05
PCDD/Fs (ng WHO_{2005}-TEQ kg^{-1})	0.623	0.446	0.565

PCDD/Fs in the sediment in the coastal areas (72.63 ng kg^{-1}) was much higher than that in the fishponds (29.53 ng kg^{-1}), but the corresponding WHO_{2005}-TEQ values in the coastal areas (0.545 ng WHO_{2005}-TEQ kg^{-1}) were lower than those in the fishponds (0.655 ng WHO_{2005}-TEQ kg^{-1}). The mass values were higher than those for the soil samples in the Antarctic coastal environment, for which the mass values were 18.5 ± 49.5 and 3.73 ± 3.51 pg g^{-1}, but they were similar to the soil samples in urban areas of Taiwan (52.4 ± 3.10 pg g^{-1}), while the WHO_{2005}-TEQ value were slightly higher than those found in the Antarctic coastal environment (0.424 ± 1.09 pg WHO_{2005}-TEQ g^{-1}) and the urban area in Taiwan (0.481 ± 2.05 pg WHO_{2005}-TEQ g^{-1}) (Kuo *et al*., 2015; Mwangi *et al*., 2016).

PCDD/F Congener Profiles in Sediment

The PCDD/F congener profiles in the sediment in the fishponds based on the WHO_{2005}-TEQ fraction are shown in Fig. 5. As the results show, the PCDD/F congener profiles of the sediment in the fishponds were dominated by 2,3,4,7,8-PeCDF (18.1%, 13.7%, 16.2% and 12.3% in November 2013, March 2014, May 2014 and July 2014, respectively), 1,2,3,7,8-PeCDD (16.4%, 22.3%, 23.6% and 28.3% in November 2013, March 2014, May 2014 and July 2014, respectively), 2,3,7,8-TeCDD (14.9%, 16.0% and 19.2% in March 2014, May 2014 and July 2014,

respectively), 1,2,3,4,7,8-HxCDF (13.4%, 10.9%, 8.7% and 8.0% in November 2013, March 2014, May 2014 and July 2014, respectively) and 2,3,4,6,7,8-HxCDD (11.3% in November 2013). The PCDD/F congener profiles in the sediment in the fishponds based on mass fraction are shown in Fig. 6. The mass PCDD/F congener profiles of the sediment in the fishponds were dominated by the higher chlorinated PCDD/F congeners such as OCDF (33.8%, 39.7%, 39.8% and 40.7% in November 2013, March 2014, May 2014 and July 2014, respectively), OCDD (28.3%, 34.2%, 33.0% and 35.9% in November 2013, March 2014, May 2014 and July 2014, respectively) and 1,2,3,4,6,7,8-HpCDF (15.2%, 10.1%,9.2% and 6.6% in November 2013, March 2014, May 2014 and July 2014, respectively).

The PCDD/F congener profiles in the sediment in the shallow sea based on the WHO_{2005}-TEQ fraction are show in Fig. 7. As the results show, the PCDD/F congener profiles of sediment in the shallow sea were dominated by the 1,2,3,7,8-PeCDD (23.8%, 20.0% and 23.0% in November 2013, February 2014 and May 2014, respectively), 2,3,4,7,8-PeCDF (15.7%, 13.6% and 15.6% in November 2013, February 2014 and May 2014, respectively) and 2,3,7,8-TeCDD (12.3%, 12.4% and 13.1% in November 2013, February 2014 and May 2014, respectively). The PCDD/F congener profiles in the sediment in the shallow sea based on mass fraction are shown in Fig. 8. Unlike the WHO_{2005}-TEQ PCDD/F congener profiles, the mass PCDD/F congener profiles in the sediment in the shallow sea are largely dominated by the more highly chlorinated PCDD/F congeners OCDD (72.2%, 72.1% and 75.4% in November 2013, February 2014 and May 2014, respectively), followed by the OCDF (14.6%, 17.1% and 14.0% in November 2013, February 2014 and May 2014, respectively).

On the whole, the mass PCDD/F congener profiles of the sediment samples in the fishponds and coastal areas were dominated by more highly chlorinated PCDD/F congeners such as OCDD and OCDF, and similar results were found in the soil samples in the vicinity of an industrial complex and the Antarctic coastal environment (Kuo *et al*., 2015; Mwangi *et al*., 2016). The WHO2005-TEQ PCDD/F congener profiles were dominated by the less chlorinated PCDD/F congeners such as 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF.

The Effects of Atmospheric Deposition on Sediment

As we know, the atmosphere is important for transferring PCDD/Fs via deposition from air to the soil and water system (Welsch-Pausch *et al.*, 1995). Previous studies have suggested the annual dry, wet and total deposition in Yunlin County to be 4955, 254 and 5209 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2014, 4524, 348 and 4872 and 4742 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2015, and 4224, 518 and 4742 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$ in 2016 (Chen *et al*., 2017). According to the results of Chen *et al*. (2017), we obtained the annual average total deposition which was 4941 pg WHO_{2005}-TEQ m^{-2} $year^{-1}$. We used the annual average deposition value to model the content of PCDD/Fs in a time range from 30 to 60 years. In a context with a sediment density of 1.6 g cm^{-3}, scenario A was constructed

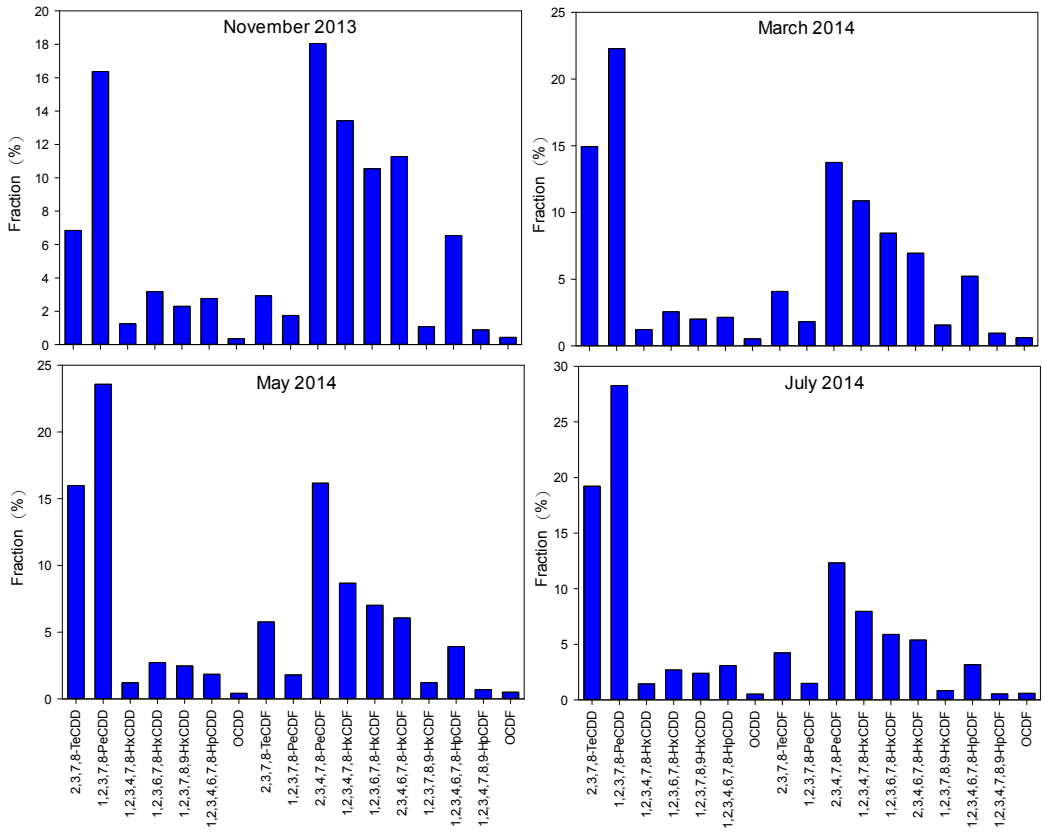

Fig. 5. The congener profile of WHO_{2005}-TEQ PCDD/Fs in the fishpond sediment.

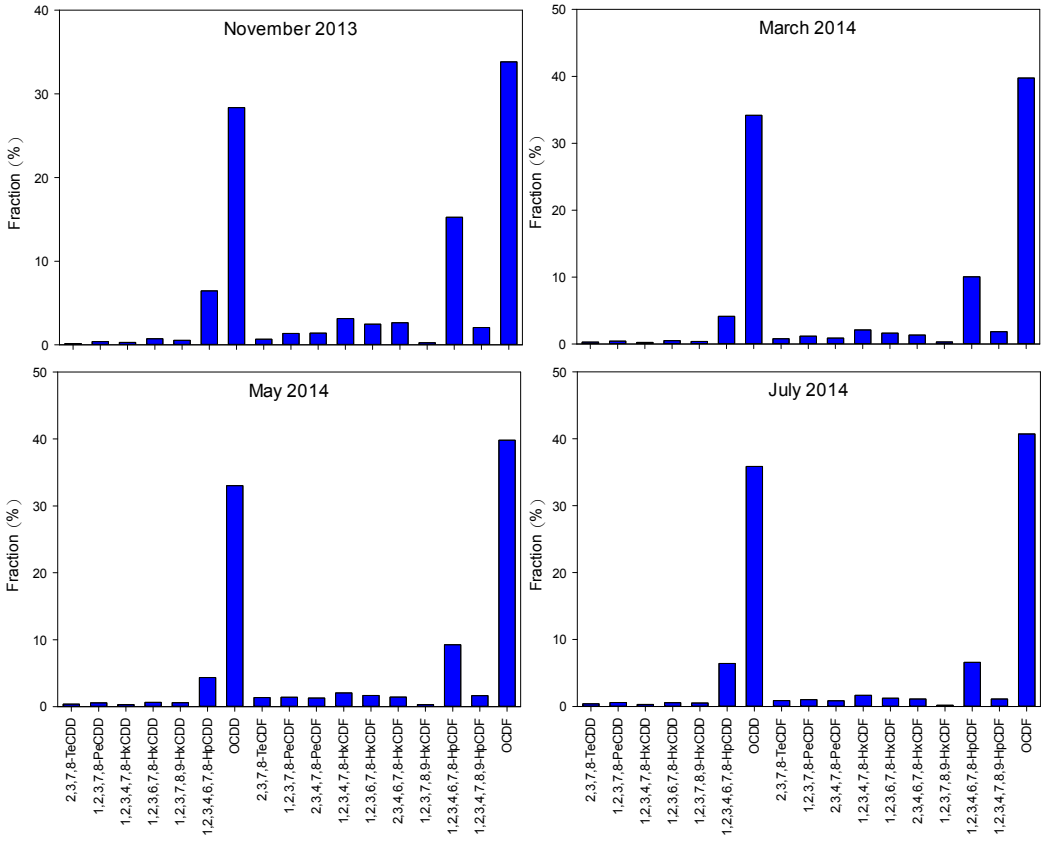

Fig. 6. The congener profile of mass PCDD/Fs in the fishpond sediment.

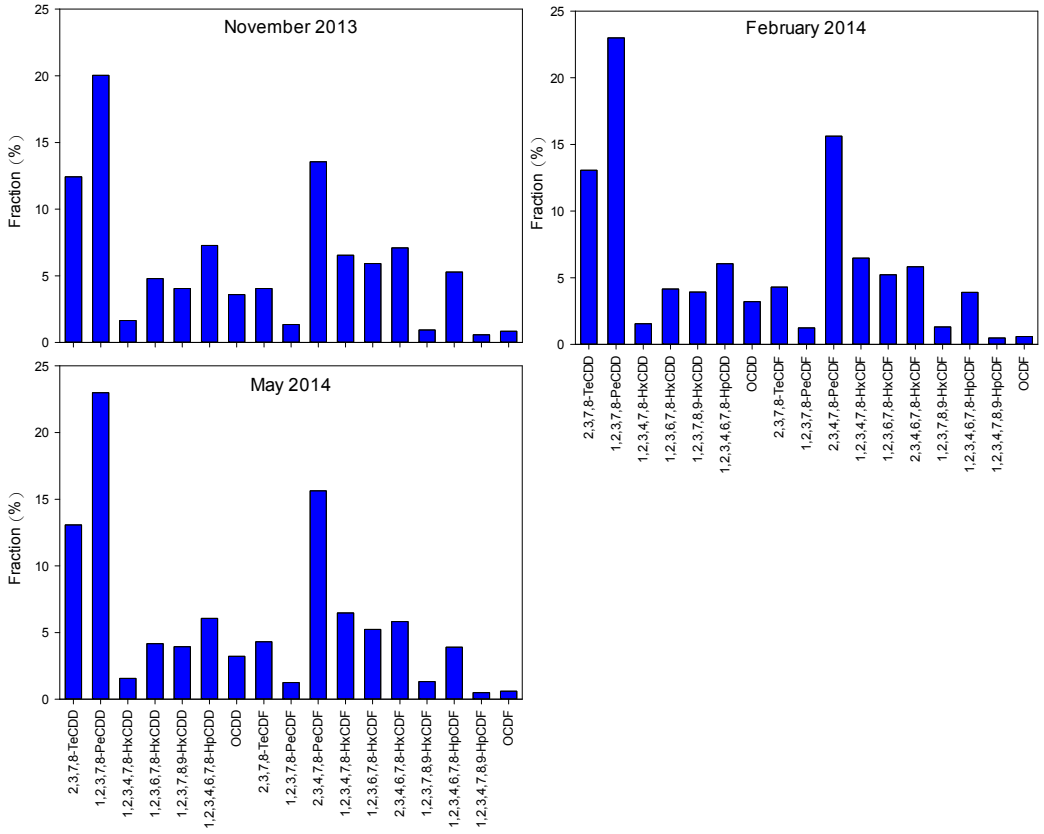

Fig. 7. The congener profile of WHO_{2005}-TEQ PCDD/Fs in the coastal area sediment.

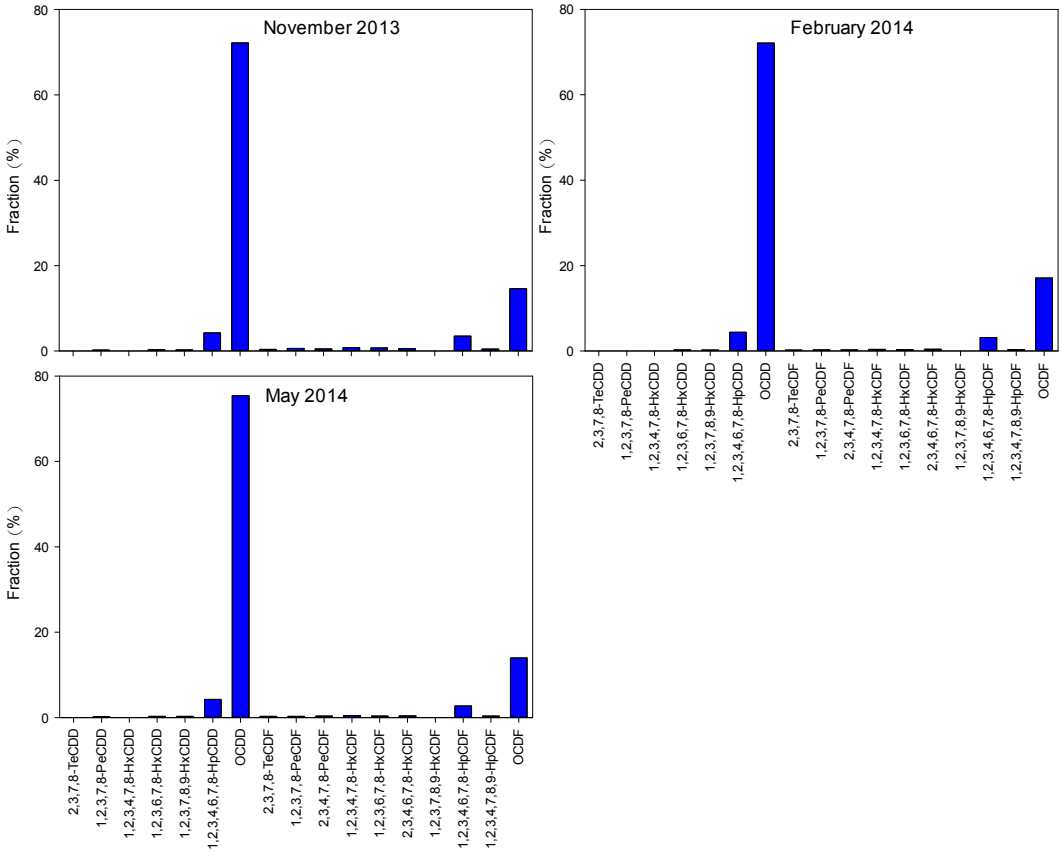

Fig. 8. The congener profile of mass PCDD/Fs in the coastal area sediment.

Table 6. The modeled total content in the sediment and the ratio of the modeled and observed values over a 30–60 year period.

Years	Total Deposition	Scenario A	Scenario B	Fishpond		Coastal area	
	(pg WHO_{2005}-TEQ m^{-2})	(ng WHO_{2005}-TEQ kg^{-1})		A	B	A	B
30	148260	9.27	6.18	14.2	9.4	17.0	11.3
40	197680	12.36	8.24	18.9	12.6	22.7	15.1
50	247100	15.44	10.3	23.6	15.7	28.3	18.9
60	296520	18.53	12.36	28.3	18.9	34.0	22.7

with a soil depth of 10 cm, and scenario B was constructed with a soil depth of 15 cm. The results of the modeled PCDD/F content are listed in Table 6. At a depth of 10 cm, the modeled PCDD/F content in the sediment was 9.27, 12.36, 15.44 and 18.53 ng WHO_{2005}-TEQ kg^{-1} for 30, 40, 50 and 60 years. When the depth was 15 cm, the modeled PCDD/F content in the sediment was 6.18, 8.24, 10.30 and 12.36 ng WHO_{2005}-TEQ kg^{-1} for 30, 40, 50 and 60 years. The PCDD/F content increased with the accumulation of PCDD/Fs through atmospheric deposition from the atmosphere. Compared with the observed average values of sediment in this study, the ratios of the modeled and observed values ranged from 9.4–34.0. For the sediment in the fishponds, the ratios of the modeled and observed values ranged between 14.2 and 28.3 for scenario A, and between 9.4 and 18.9 for scenario B. For the sediment in coastal area, the ratios of modeled and observed values ranged between 17.0 and 34.0 for scenario A, and between 11.3 and 22.7 for scenario B. With economic and industrial development, the POPs pollution will get worse. Therefore, the content of PCDD/Fs in soil and animals will increase and it will eventually accumulate in the human body and affect human health. The results of this study provide useful information for prevention and control strategies for POPs.

CONCLUSION

1. In the fishponds, the averaged mass content total PCDD/Fs was 0.432, 0.671 and 0.244 pg g^{-1}, and the corresponding WHO_{2005}-TEQ total PCDD/Fs content was 0.070, 0.015 and 0.024 pg WHO_{2005}-TEQ g^{-1} in *O. mossambica*, *M. meretrix* and *L. calcarifer*, respectively. For *O. gigas* in the coastal areas, the averaged mass content of PCDD/Fs was 0.727 pg g^{-1}, and the corresponding WHO_{2005}-TEQ content was 0.018 pg WHO_{2005}-TEQ g^{-1}.

2. The mass PCDD/F congener profiles of the animal samples in the fishponds and coastal areas were dominated by more highly chlorinated PCDD/F congeners, such as OCDD and OCDF, while the WHO_{2005}-TEQ PCDD/F congener profiles of the animal samples were dominated by less chlorinated PCDD/F congeners such as 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF.
3. The total mass content of PCDD/Fs in the sediment in the coastal area (72.63 ng kg^{-1}) was much higher than that in the fishponds (29.53 ng kg^{-1}), but the corresponding WHO_{2005}-TEQ values at coastal area (0.545 ng WHO_{2005}-TEQ kg^{-1}) were lower than that those in the fishponds (0.655 ng WHO_{2005}-TEQ kg^{-1}).
4. For sediment samples, the mass PCDD/F congener profiles at fishpond and coastal area are dominated by more highly chlorinated PCDD/F congeners, such as OCDD and OCDF, while the WHO_{2005}-TEQ PCDD/F congener profiles were dominated by less chlorinated PCDD/F congeners such as 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF.
5. The modeled PCDD/F content in the sediment ranged between 9.27 and 18.53 ng WHO_{2005}-TEQ kg^{-1} when the soil depth was 10 cm, and the PCDD/F content range from 6.18–12.36 ng WHO_{2005}-TEQ kg^{-1} over a 30 to 60 years period when the depth was 15 cm.
6. Compared with the observed average values of sediment in this study, the ratios of the modeled and observed values ranged between 9.4 and 34.0.

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