Atmospheric levels and distribution of Dechlorane Plus in an E-waste dismantling region of East China

Huizhong Sun^{1,2}, Yingming Li¹, Pu Wang¹, Shucheng Zheng^{1,2}, Julius Matsiko^{1,2},
Dou Wang^{1,2}, Weiwei Zhang^{1,2}, Yanfen Hao^{1,2}, Qinghua Zhang^{1,2*} & Guibin Jiang^{1,2}

¹State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences,
Chinese Academy of Sciences, Beijing 100085, China

²University of Chinese Academy of Sciences, Beijing 100049, China

Received June 16, 2016; accepted August 3, 2016; published online October 27, 2016

Atmospheric concentrations of Dechlorane Plus (DP) were investigated in Taizhou, an electronic-waste (E-waste) dismantling region in East China. Passive air samplers with polyurethane foam (PUF) disks were deployed every three months during the sampling period of September 2009–August 2010. The total DP (*syn*- and *anti*-DP) concentrations in air ranged from not detected to 277 pg/m³, with a mean concentration of 53.9 pg/m³. A generally declining trend of DP levels was found from the E-waste dismantling region to the peripheral areas. The median values of total DP concentrations in autumn, winter, spring and summer were 52.2, 28.8, 39.7 and 30.1 pg/m³, respectively. The seasonal variations of DP concentrations were mainly associated with the intensity of E-waste dismantling activities and meteorological conditions. The mean value of *anti*-DP fractional abundance (f_{anti}) was 0.74±0.08, which was consistent with those in the commercial DP products. This study confirmed a significant emission source related to the distribution of atmospheric DP in the E-waste dismantling area and supplied information for the seasonal variation of DP in the atmosphere.

Dechlorane Plus, air, E-waste dismantling, seasonal variation

Citation: Sun H, Li Y, Wang P, Zheng S, Matsiko J, Wang D, Zhang W, Hao Y, Zhang Q, Jiang G. Atmospheric levels and distribution of Dechlorane Plus in an E-waste dismantling region of East China. *Sci China Chem*, 2017, 60: 305–310, doi: 10.1007/s11426-016-0261-6

1 Introduction

Dechlorane Plus (DP; molecular formula: C₁₈H₁₂Cl₁₂; two stereo isomers: *syn*-DP and *anti*-DP) is a chlorinated flame retardant that is added into polymer materials to enhance their flame retardant properties. Due to the advantages of less discoloration and non-blooming, DP has been widely applied to electronic equipment, textile industry and plastic decoration materials [1]. As a substitute product of Dechlorane (Mirex), DP was once produced by Hooker Chemical (now known as OxyChem) in the 1970s. Now two major

DP manufacturers are located in Niagara Falls, New York, USA [2] and in Huai'an, Jiangsu Province, China [3], respectively. Over the past decades, the volume of DP production and consumption has increased with years. The annual production of DP approximately reached 10 million pounds [4].

In 2006, Hoh *et al.* [2] first identified DP in ambient air, fish, and sediment samples from the Great Lakes region nearby a DP manufactory. Thereafter DP has been known as a ubiquitous contaminant and detected in various environmental and biological media, such as ambient air [4], water [5], sediment [1], indoor dust [6], plant [7,8], aquatic biota [9] and amphibian [10], terrestrial wildlife [11,12], even in human serum [13] and breast milk [14]. DP

*Corresponding author (email: qhzhang@rcees.ac.cn)

has attracted great interest due to its similar properties (e.g., environmental persistence, bioaccumulation [9,15,16], long-distance transportation [5]) to persistent organic pollutants (POPs), which may pose a potential risk to human health.

As one of long-distance transportation intermediaries, atmosphere prominently dominates in transporting DP from sources to remote areas. In recent years, the detections of DP in air samples have increased on a global scale. The DP atmospheric concentrations have been reported in the North American Great Lakes region [17,18], urban and rural sites in Asian countries including Japan [19], Korea [20], China [4] and Pakistan [21], the remote locations in the Arctic and sub-Arctic regions [22,23], as well as the marine atmosphere from Arctic to Antarctica [5,24]. During the past decade, air pollution has raised concerns in China [25,26]. The first study reported DP atmospheric concentrations in 97 urban and rural sites covering the most domestic provinces in 2008 [4]. Subsequently, some studies have investigated DP levels in the atmosphere of several large cities (Beijing [20], Shanghai [27], Harbin [28], Dalian [29], etc.), the areas surrounding DP manufacturing plants [3,30], electronic-waste (E-waste) dismantling site (Qingyuan) [31], and remote location in Tibetan Plateau [32].

E-waste dismantling region attracted great attention due to the serious environmental hazard related to E-waste dismantling activities. Many studies have observed high POPs concentrations in the environmental and biological samples from the E-waste recycling regions [33,34]. As an additive flame retardant, DP is easily released into environment during the production, use and disposal processes of DP-containing products. Moreover, the recycling process of E-waste may also lead to the release of DP into the surrounding environment, which was identified as a major emission source of DP [35]. Taizhou is one of the major E-waste dismantling regions in East China which started E-waste dismantling activities in the late 1970s. The E-waste dismantling industry employs around forty thousand people, and the annual dismantling capacity has exceeded 2.2 million metric tons [36]. In our previous study, high DP levels were measured in earthworm and agricultural soil samples collected from the E-waste recycling region in Taizhou and the concentrations in most soil samples were higher than those reported in urban areas of China [37]. However, the studies on the atmospheric DP levels in Taizhou and its spatial-temporal distributions were scarce.

In this study, we collected air samples at various locations of E-waste dismantling region in four seasons to investigate the seasonal variation of DP concentrations between the E-waste dismantling sites and peripheral sites. The potential influences on DP distribution were also discussed involving the weather and geographical conditions, as well as the E-waste dismantling activity.

2 Experimental

2.1 Materials

The reagents, dichloromethane (DCM), *n*-hexane, and acetone were pesticide grade and purchased from J. T. Baker (USA). Nonane was of chromatographic grade and purchased from Sigma-Aldrich (USA). Silica gel 60 (70–230 mesh) was supplied from Merck (Darmstadt, Germany) and baked at 550 °C for 12 h to activate it. Sulfuric acid and sodium hydroxide were analytically pure and obtained from Beijing Chemical Factory (China) to prepare acid silica gel (30%, w/w) and basic silica gel (1.2%, w/w) from activated silica gel. Anhydrous sodium sulfate was also obtained from Beijing Chemical Factory (China) and baked at 660 °C for 6 h before use. Carbon (Carbopak C) and Celite (545 coarse) were from Supelco (USA) and baked at 130 °C for 6 h after mixing (18%, w/w). The standard solutions of *syn*-DP, *anti*-DP, ¹³C₁₂-PCB-138 (injection standard) and ¹³C₁₂-PCB-209 (surrogate standard) were purchased from Wellington Laboratories (Canada).

2.2 Sample collection

Passive air samplers (PASs) with polyurethane foam (PUF) disks deployed in the present study have been described previously [38,39]. In brief, a PUF disk was housed inside a stainless steel chamber consisting of a pole and two domes with different external diameters. The PASs were placed under the eaves, roped to trees or poles (1.5 m above the ground). Air flowed into the chamber through the gap between two domes and several holes in the surface of the bottom dome. The contaminants and fine/ultrafine particles in the air were adsorbed on the surface of PUF disks. The effective concentrations of passive air samples were calculated by equivalent air volumes (effective sampling rate × sampling time) which were related to the properties of the analytes, sampling days and planar surface area of PUF disks. The PUF disks in this study are 14 cm in diameter and 1.35 cm thick, with the planar surface area of 365 cm². According to the previous studies [3], the effective sampling rate for DP was set as 3.5 m³ per day.

The PASs were deployed in seven sites to collect air samples every three months (approximately 90 d for collecting each air sample) between September 2009 and August 2010. The sampling period covered four distinct seasons: autumn (from September to November), winter (from December to February of the next year), spring (from March to May) and summer (from June to August). Seven sampling sites were clustered within a 15 km radius of the E-waste dismantling center, and situated in two regions: E-waste recycling region (S1–S4) and peripheral region (S5–S7) (Figure 1). Site 1 (S1), S2, S3 and S4 were located around the E-waste recycling plant; S5 was seated in the adjacent urban center; S6 and S7 were rural sites which

situated in the south and east direction of recycling center, respectively.

Before sampling, PUF disks were extracted twice by the accelerated solvent extraction device (Dionex ASE 300, USA) with acetone and DCM/*n*-hexane (1:1, *v/v*) as solvents at 100 °C, 1500 psi and two cycles to remove impurities. After sample collection, PUF disks were wrapped by aluminum foil and sealed in package, then transported to the laboratory and stored in refrigerator at −20 °C awaiting further analysis.

2.3 Extraction and purification

The extraction and purification procedures followed our previous studies [3,40] with a minor modification. Briefly, PUF samples were extracted using Dionex ASE 300 with DCM: *n*-hexane (1:1, *v/v*) at 100 °C, 1500 psi and two cycles. Prior to extraction, 10 μL $^{13}\text{C}_{12}$ -PCB-209 (100 pg/μL) was spiked as the internal standard. The extract was concentrated to 2 mL by vacuum rotary evaporation and purified by multilayer silica gel column (from bottom to top: 1 g activated silica gel, 4 g basic silica gel, 1 g activated silica gel, 8 g of acidic silica gel, 2 g activated silica gel and 4 g anhydrous sodium sulfate). The column was pre-rinsed with 80 mL *n*-hexane, and then rinsed with 100 mL *n*-hexane after sample loading. The eluent was concentrated to 2 mL and further purified by activated carbon column (1.5 g activated carbon mixture at the bottom and 4 g anhydrous sodium sulfate at the top). The column was pre-rinsed with 50 mL toluene and 20 mL *n*-hexane in turn, and finally eluted with 50 mL *n*-hexane. The final eluent was concentrated to 1 mL and transferred into a vial containing 20 μL nonane. The content in the vial was further evaporated to approximately 20 μL under a gentle stream of nitrogen and later

spiked with 10 μL $^{13}\text{C}_{12}$ -PCB-138 (100 pg/μL) as injection standard prior to instrumental analysis.

2.4 Instrumental analysis

The instrumental analysis of DP was performed with gas chromatography coupled with negative chemical ionization mass spectrometer (GC-NCI-MS, SHIMADZU 2010 Ultra, Japan). The GC system applied to a DB-5MS column (15 m×0.25 mm ID, film thickness of 0.1 μm, J & W Scientific, USA). Detailed information about analytical parameters was provided previously [3]. A splitless mode was selected for injection. The temperatures of injection port and the transfer line were held at 285 °C. The oven temperature was set at 100 °C initially for 4 min, then ramped to 150 °C at 8 °C/min and held for 4 min, subsequently ramped to 300 °C at 12 °C/min and held for 13 min. Helium was used as the carrier gas with the column flow rate of 1 mL/min. The MS source temperature was set to 200 °C. Methane was used as the moderating gas. The MS system was operated in selected ion monitoring (SIM) mode with the monitoring ions: *m/z* 653.8 and 651.8 (*syn*- and *anti*-DP), *m/z* 507.9 and 509.9 ($^{13}\text{C}_{12}$ -PCB-209), *m/z* 369.7 and 371.7 ($^{13}\text{C}_{12}$ -PCB-138).

2.5 Quality assurance and quality control (QA/QC)

The limits of detection (LOD) for sample analysis of *syn*- and *anti*-DP (mean±standard deviation) were 0.21±0.12 and 0.32±0.13 pg/m³, respectively. The recoveries of $^{13}\text{C}_{12}$ -PCB-209 were 96.4%±16.2% for all the samples. Four procedural blanks were paralleled along with every batch of the seven samples during experiment process for quality control. In the procedural blanks, the two isomers of DP were detected at <5% of the levels determined in all samples except for one sample (S7 in summer) in which the concentration was extremely low and the blanks reached up to around 20% of that. The value of S7 (summer) was therefore not presented here.

3 Results and discussion

3.1 Overall concentrations of DP

In the present study, the concentrations of ΣDP (sum of *syn*- and *anti*-DP) ranged from not detected to 277 pg/m³ with median and algebraic mean values of 39.6 and 53.9 pg/m³. The DP levels determined in Taizhou were obviously higher than the air samples collected by PASSs from urban (15.6±15.1 pg/m³) and rural (3.5±5.6 pg/m³) sites across China. In the past few years, high-volume air samplers (HVASs) were used to measure the DP atmospheric concentrations as well. The DP levels in air determined close to the Chinese DP manufacturing plant were 7737–26734 and

Figure 1 The sampling sites and seasonal distribution of atmospheric DP concentrations (color online).

3332 pg/m³ [3,30]. Meanwhile, the atmospheric concentrations of DP were reported in several Chinese large cities with dense population and developed industry, such as Beijing (1.98±3.90 pg/m³) [20], Shanghai (5.48±1.28 pg/m³) [27], Harbin (0.35±0.21 pg/m³) [28] and Dalian (3±6 pg/m³) [29]. Although different sampling techniques (passive and active air samplers) may lead to deviation in the measured concentrations, field-based calibration studies have showed the comparability of passive and active samplers for measuring particle-associated chemicals in air [41]. The DP levels determined in Taizhou were significantly lower than DP manufacturing plant surrounding areas but higher than urban average levels. The concentrations of total DP and the *anti*-isomer fractional abundance (f_{anti}) in the atmosphere from different areas are summarized in Table S1 (Supporting Information online). The atmospheric concentrations of DP in Taizhou were higher than most reported regions in Asia, Europe and North America. The relatively high atmospheric DP levels suggested that some DP emission sources occurred in the E-waste recycling area.

3.2 Spatial distribution

Among the seven sampling sites, the maximum annual Σ DP concentrations were measured in S1 adjacent to an E-waste recycling plant. The concentrations in E-waste region (S1–S4) ranged from 28.8–277 pg/m³ with median and algebraic mean values of 63.1 and 77.7 pg/m³. Meanwhile, the lower concentrations were measured in the urban (S5) and rural (S6, S7) sites (not detected–52.2 pg/m³ with the median and algebraic mean values of 16.3 and 22.2 pg/m³) which were located several miles away from the E-waste recycling center. The annual mean concentration of E-waste region was approximately 4 times greater than the peripheral regions (S5–S7). The DP concentrations showed an obviously declining trend with the increasing distance from the E-waste recycling plant. This result indicated that local E-waste recycling activity was probably the primary DP emission source in study area, which adhered to our previous summarization about DP emission sources [35]. The similar result was also found in the North American Great Lakes region where the higher atmospheric DP concentrations were observed in a rural site (Sturgeon Point, NY, USA) located near the DP manufacturing facility and the concentrations of doubling distance from DP plant decreased by about 30% [18].

The atmospheric concentrations in the E-waste dismantling sites (S1–S4) were obviously lower than those from Qingyuan (13.1–1794 pg/m³, an average of 363 pg/m³) [31], another E-waste site in South China. Moreover, the similar results were obtained for some other Halogenated flame retardants (HFRs). The levels of several HFRs including polybrominated diphenyl ethers (PBDEs), hexachlorobenzene (HBB) and pentabromoethylbenzene (PBEB) detected in Taizhou were much lower than those reported in Qing-

yuan [34]. Since the past decade, some measures have been taken by Chinese government in Taizhou to regulate informal E-waste recycling activities. The lower atmospheric levels of flame retardants in Taizhou may be caused by the increasingly stringent regulations on E-wastes as well as recycling methods.

3.3 Seasonal variation

The seasonal distribution of total DP during the sampling period (September 2009 to August 2010) is shown in Figure 1. The median values (range) of Σ DP concentrations in autumn, winter, spring and summer were 52.2 pg/m³ (16.1–277 pg/m³), 28.8 pg/m³ (9.3–149 pg/m³), 39.7 pg/m³ (16.5–69.3 pg/m³) and 30.1 pg/m³ (not detected–77.7 pg/m³), respectively. The maximum atmospheric DP level was observed in autumn, followed by spring and summer, whereas the DP overall concentration in winter was the minimum. Moreover, the highest concentrations in the E-waste recycling region (S1–S4) and a rural site (S6) were detected in autumn, while the highest level in the other peripheral sites (S5 and S7) was in spring.

The intensity of local dismantling activities and variable meteorological conditions were probably the main factors on the seasonal variation in Taizhou. According to previous study, the atmospheric DP concentrations at E-waste sites were highly dependent upon the strength of the emission source [31]. The historical data of the amount of dismantled E-waste in Taizhou showed that more E-wastes were recycled from April to June and October to December [42]. The higher DP concentrations in autumn and spring were consistent with the larger dismantled E-wastes in these seasons.

Due to the high octanol-air partition coefficient ($\log K_{\text{OA}}=12.26$ [43]), DP is a particle-associated chemical and mostly detected in particle phase [3]. However, the PASs used in this study could mainly collect gas phase samples with rarely fine/ultrafine particles absorbed by PUF disks [44]. As the particle number concentrations and size distributions in air are affected by the meteorological conditions, the different weather conditions (precipitation, particulate pollution and seasonal winds) influenced the atmospheric DP concentrations to some degree. Since Taizhou is located on the eastern coast of China, it has a subtropical monsoon climate with four distinctive seasons. Northerly winds prevail in winter and southerly winds reign in summer. The mean annual temperatures were between 17.2 to 18 °C with the highest temperature in July and the lowest in January. The annual rainfalls ranged from 1744 to 2483 mm and more precipitations were observed in June and July [45,46]. According to the monitoring data, the heavy particulate pollution generally occurred from April to June and October to December [47,48]. Less precipitation, heavy particulate pollution as well as frequent E-waste dismantling activities in autumn resulted in the highest DP concentration in that season. As the wind direction in Taizhou changes with the

seasons, the seasonal wind affected the DP levels of peripheral sites to varying degrees. S5 and S6 are situated in the downwind area at spring and autumn, where the highest concentrations were observed in spring and autumn, respectively. Since S7 was not present in the main wind direction area, the higher concentrations were measured in both spring and autumn which were mainly influenced by the frequent E-waste recycling activities in these seasons.

The seasonal variations of atmospheric DP concentrations in Taizhou were different from another E-waste recycling region. Chen *et al.* [31] investigated the monthly variabilities of DP in Qingyuan and detected the relatively high DP levels from January to April in the E-waste site and in February and March at the reference site. The different seasonal pattern of DP in the air between two E-waste recycling regions was probably caused by local dismantling activities. The higher DP concentrations in Qingyuan during January to April depended on the frequent dismantling activities in that period. Additionally, the different air-sampling methods and meteorological parameters were other factors influencing these levels.

Although the atmospheric DP concentrations in Taizhou were scarce, seasonal variations of some other HFRs were measured in previous studies. The total PBDEs levels in the atmosphere at E-waste recycling region were strikingly higher than those at the reference site and the winter levels of PBDEs was about 3 times of that in summer, which should be mainly due to the different meteorological conditions in two seasons [49]. Nevertheless, in a subsequent research, Zhang *et al.* [34] reported that the higher atmospheric concentrations of PBDEs, HBB and PBEB observed in summer and the various PBDEs congener profiles in summer and winter may reflect the different emission mechanisms in two seasons. Despite the different results, the influence of weather conditions and emission source were speculated as the main factors, which is in correspondence with our study. However, the relationships among DP concentrations, meteorological conditions (temperature, precipitation and wind speed) and intensity of dismantling activities require further investigation.

3.4 DP isomer ratios

The fractional abundance (f_{anti}) value is used to represent the composition of DP isomers, which is defined as follow [13]:

$$f_{\text{anti}} = [\text{anti-DP}] / ([\text{anti-DP}] + [\text{syn-DP}])$$

The f_{anti} values of technical DP products determined in previous studies were not constant. The values of the products from OxyChem were ranged from 0.64 to 0.80 [2,9,50], whereas the values from the Chinese DP manufacture plant were 0.70 ± 0.003 [15] and 0.68 ± 0.01 [3]. During the long-range atmospheric transport from the pollution source to the remote area, the sun-irradiation and biological effects will change the two stereo isomers proportions to some ex-

tent. The apparent decline of f_{anti} values (0.36 ± 0.12) [24] were observed in the marine atmosphere, which could reflect the influence of atmospheric transport on DP isomer composition. Therefore, the proportion of a DP isomer can reflect the presence of local sources.

In this study, the annual f_{anti} value (mean $\pm$ standard deviation) was 0.74 ± 0.08 (Figure 2). The f_{anti} values in E-waste recycling region (0.75 ± 0.06) were higher than the peripheral region (0.73 ± 0.10), but there was no significant difference observed in two regions (Mann-Whitney U test, $p = 0.33$). In our previous study, the f_{anti} values in soil samples collected from various farmland locations in Taizhou ranged from 0.57 to 0.80, with a mean value of 0.71 [37]. The f_{anti} values in the air were in agreement with the soil samples collected at the same period. The values of f_{anti} in Taizhou were comparable to those obtained in another E-waste site (0.726 ± 0.037) [31] and were closed to the commercial products. The values were higher than the mean value of many Chinese urban (0.67 ± 0.10) and rural (0.66 ± 0.11) areas [4]. The higher f_{anti} values in E-waste recycling region were coincident with the presence of local emission source. Furthermore, the f_{anti} values of autumn, winter, spring and summer were 0.78 ± 0.05 , 0.70 ± 0.07 , 0.71 ± 0.09 and 0.79 ± 0.05 , respectively. The value of f_{anti} in autumn and summer were higher than spring and winter, but the differences among four seasons were not significant (one-way ANOVA on ranks, $p = 0.055$). This result suggested that both DP isomers were stable in the atmosphere and basically invariable with weather conditions.

4 Conclusions

In this study, atmospheric DP concentrations were measured in an E-waste dismantling region in East China. Relatively high DP levels were found in this study which suggested that E-waste dismantling activity was the major DP emission source in the local area. DP concentrations in the air showed a decreasing trend with the increasing distance from the E-waste dismantling plant and the average DP

Figure 2 The f_{anti} value of air samples (color online).

concentrations near the E-waste site were several times higher than those in the peripheral regions. Moreover, the seasonal variation was also observed in the study areas. The higher DP levels were measured in autumn around E-waste sites while in spring and autumn at the peripheral regions. The meteorological conditions and the intensity of dismantling activities were probably the primary reasons for the seasonal variation of DP levels. The f_{anti} values were similar to DP commercial products, indicating there was no stereo selectivity of atmospheric DP occurred in this E-waste recycling area.

Acknowledgments This work was supported by the National Natural Science Foundation of China (21477156, 21277165), the National Basic Research Program of China (2015CB453101) and the Strategic Priority Research Program of the Chinese Academy of Sciences (XDB14010100).

Conflict of interest The authors declare that they have no conflict of interest.

Supporting information The supporting information is available online at chem.scichina.com and link.springer.com/journal/11426. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

- 1 Qiu XH, Marvin CH, Hites RA. *Environ Sci Technol*, 2007, 41: 6014–6019
- 2 Hoh E, Zhu LY, Hites RA. *Environ Sci Technol*, 2006, 40: 1184–1189
- 3 Zhang QH, Zhu CF, Zhang HD, Wang P, Li YM, Ren DW, Jiang GB. *Sci Bull*, 2015, 60: 792–797
- 4 Ren NQ, Sverko E, Li YF, Zhang Z, Harner T, Wang DG, Wan XN, McCarry BE. *Environ Sci Technol*, 2008, 42: 6476–6480
- 5 Moeller A, Xie ZY, Sturm R, Ebinghaus R. *Environ Sci Technol*, 2010, 44: 8977–8982
- 6 Zhu JP, Feng YL, Shoeib M. *Environ Sci Technol*, 2007, 41: 7694–7698
- 7 Qiu XH, Hites RA. *Environ Sci Technol*, 2008, 42: 31–36
- 8 Na GS, Wei W, Zhou SY, Gao H, Ma XD, Qiu LN, Ge LK, Bao CG, Yao ZW. *J Environ Sci*, 2015, 28: 8–13
- 9 Tomy GT, Pleskach K, Ismail N, Whittle DM, Helm PA, Sverko E, Zaruk D, Marvin CH. *Environ Sci Technol*, 2007, 41: 2249–2254
- 10 Li L, Wang WY, Lv QX, Ben YJ, Li XH. *J Environ Sci*, 2014, 26: 636–642
- 11 Gauthier LT, Hebert CE, Weseloh DVC, Letcher RJ. *Environ Sci Technol*, 2007, 41: 4561–4567
- 12 Guerra P, Fernie K, Jimenez B, Pacepavicius G, Shen L, Reiner E, Eljarrat E, Barcelo D, Alae M. *Environ Sci Technol*, 2011, 45: 1284–1290
- 13 Ren GF, Yu ZQ, Ma ST, Li HR, Peng P, Sheng GY, Fu JM. *Environ Sci Technol*, 2009, 43: 9453–9457
- 14 Ben YJ, Li XH, Yang YL, Li L, Di JP, Wang WY, Zhou RF, Xiao K, Zheng MY, Tian Y, Xu XB. *Environ Pollut*, 2013, 173: 176–181
- 15 Wu JP, Zhang Y, Luo XJ, Wang J, Chen SJ, Guan YT, Mai BX. *Environ Sci Technol*, 2010, 44: 606–611
- 16 Tomy GT, Thomas CR, Zidane TM, Murison KE, Pleskach K, Hare J, Arsenaault G, Marvin CH, Sverko E. *Environ Sci Technol*, 2008, 42: 5562–5567
- 17 Venier M, Hites RA. *Environ Sci Technol*, 2008, 42: 4745–4751
- 18 Salamova A, Hites RA. *Environ Sci Technol*, 2011, 45: 9924–9930
- 19 Kakimoto K, Nagayoshi H, Takagi S, Akutsu K, Konishi Y, Kajimura K, Hayakawa K, Toriba A. *Ecotox Environ Safe*, 2014, 99: 69–73
- 20 Kakimoto K, Nagayoshi H, Akutsu K, Konishi Y, Kajimura K, Hayakawa K, Toriba A. *Environ Sci Pollut Res*, 2015, 22: 14600–14605
- 21 Syed JH, Malik RN, Li J, Wang Y, Xu Y, Zhang G, Jones KC. *Chemosphere*, 2013, 93: 1646–1653
- 22 Xiao H, Shen L, Su YS, Barresi E, DeJong M, Hung HL, Lei YD, Wania F, Reiner EJ, Sverko E, Kang SC. *Environ Pollut*, 2012, 161: 154–161
- 23 Salamova A, Hermanson MH, Hites RA. *Environ Sci Technol*, 2014, 48: 6133–6140
- 24 Moeller A, Xie ZY, Cai MH, Zhong GC, Huang P, Cai MG, Sturm R, He JF, Ebinghaus R. *Environ Sci Technol*, 2011, 45: 6793–6799
- 25 Wang S, Li GG, Gong ZY, Du L, Zhou QT, Meng XY, Xie SY, Zhou L. *Sci China Chem*, 2015, 58: 1435–1443
- 26 Chen HS, Li J, Ge BZ, Yang WY, Wang ZF, Huang S, Wang YL, Yan PZ, Li JJ, Zhu LL. *Sci China Chem*, 2015, 58: 1403–1415
- 27 Yu ZQ, Liao RE, Li HR, Mo LG, Zeng XY, Sheng GY, Fu JM. *Environ Pollut*, 2011, 159: 2982–2988
- 28 Ma WL, Liu LY, Hong Q, Sun DZ, Shen JM, Wang DG, Li YF. *Environ Int*, 2011, 37: 66–70
- 29 Yang M, Jia HL, Ma WL, Qi H, Cui S, Li YF. *Atmos Environ*, 2012, 55: 73–79
- 30 Wang DG, Yang M, Qi H, Sverko E, Ma WL, Li YF, Alae M, Reiner EJ, Shen L. *Environ Sci Technol*, 2010, 44: 6608–6613
- 31 Chen SJ, Tian M, Wang J, Shi T, Luo Y, Luo XJ, Mai BX. *Environ Pollut*, 2011, 159: 1290–1296
- 32 Yu Y, Hung H, Alexandrou N, Roach P, Nordin K. *Environ Sci Technol*, 2015, 49: 8623–8630
- 33 Zhu CF, Wang P, Li YM, Chen ZJ, Li WJ, Ssebugere P, Zhang QH, Jiang GB. *Environ Sci Process Impacts*, 2015, 17: 693–699
- 34 Zhang T, Huang YR, Chen SJ, Liu AM, Xu PJ, Li N, Qi L, Ren Y, Zhou ZG, Mai BX. *J Environ Monit*, 2012, 14: 3156–3163
- 35 Wang P, Zhang QH, Zhang HD, Wang T, Sun HZ, Zheng SC, Li YM, Liang Y, Jiang GB. *Environ Int*, 2016, 88: 206–220
- 36 Tang W, Cheng JP, Zhao WC, Wang WH. *J Environ Sci*, 2015, 34: 107–132
- 37 Xiao K, Wang P, Zhang HD, Shang HT, Li YM, Li XH, Ren DW, Chen WH, Zhang QH. *Environ Geochem Health*, 2013, 35: 625–631
- 38 Pozo K, Harner T, Shoeib M, Urrutia R, Barra R, Parra O, Focardi S. *Environ Sci Technol*, 2004, 38: 6529–6537
- 39 Harner T, Shoeib M, Diamond M, Stern G, Rosenberg B. *Environ Sci Technol*, 2004, 38: 4474–4483
- 40 Zhang HD, Wang P, Li YM, Shang HT, Wang YW, Wang T, Zhang QH, Jiang GB. *Environ Sci Technol*, 2013, 47: 10567–10573
- 41 Markovic MZ, Prokop S, Staebler RM, Liggio J, Harner T. *Atmos Environ*, 2015, 112: 289–293
- 42 <http://lqhbluqiao.gov.cn/YGGC/ArticleList.aspx?CategoryID=2047&CurrentPageIndex=1>, accessed on 2016-07-29
- 43 Sverko E, Tomy GT, Reiner EJ, Li YF, McCarry BE, Arnot JA, Law RJ, Hites RA. *Environ Sci Technol*, 2011, 45: 5088–5098
- 44 Klanova J, Eupr P, Kohoutek J, Harner T. *Environ Sci Technol*, 2008, 42: 550–555
- 45 http://www.zjtz.gov.cn/art/2014/5/14/art_3327_90277.html, accessed on 2016-07-29
- 46 http://www.zjtz.gov.cn/art/2014/5/14/art_3353_90110.html, accessed on 2016-07-29
- 47 http://www.zjtz.gov.cn/art/2014/4/18/art_3374_90192.html, accessed on 2016-07-29
- 48 http://www.zjtz.gov.cn/art/2014/4/18/art_3343_90354.html, accessed on 2016-07-29
- 49 Han W, Feng J, Gu Z, Chen D, Wu M, Fu J. *Bull Environ Contam Toxicol*, 2009, 83: 783–788
- 50 Sverko E, Tomy GT, Marvin CH, Zaruk D, Reiner E, Helm PA, Hill B, McCarry BE. *Environ Sci Technol*, 2008, 42: 361–366