Evaluation of the distribution and transport of polychlorinated biphenyls and polybrominated diphenyl ethers in an e-waste dismantling region by using Apple snail as bioindicator

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Introduction

Contaminations of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) brought by primitive recycling of electrical and electronic waste (E-waste) have become a global environmental problem and is a hotspot in environmental research in recent years (Ni and Zeng, 2009, Ogunseitan, et al, 2009, Wong, et al, 2007). Large amounts of E-waste have been imported to developing countries such as China and India via unregulated and illegal trade (UNEP, 2005). The recycling techniques in these countries are usually very primitive, which include burning, melting, and acid chemical bath, etc. Therefore, typical persistent organic pollutants (POPs) such as PCBs, PBDEs, dioxins, and brominated dioxins have been found to be released to the ambient aquatic and terrestrial environment, and to the atmosphere.

In the present work, we collected apple snail (*Ampullariidae*) and soil samples in a radius of 70 kilometers around an E-waste recycling site, which is located in the southeast China, to investigate the spatial distribution, congener profiles, and assess the environmental risk of PCBs and PBDEs in this region to further illustrate the possibilities of using apple snail as a suitable bioindicator for these two groups of POPs in this region.

Materials and Methods

Sampling 40 composite apple snail (*Ampullariidae*) samples and 30 soils samples were collected in June, 2008 from 25 towns around an E-waste recycling site, which is located at a small town in Zhejiang province, Southern China. Apple snails were taken from rice paddies, ditches and ponds. Each sample included at least 30 apple snails, and soil samples were collected from the upper 0-10 cm surface soil layer at the same sampling sites. According to the distance from the E-waste recycling site, both the biota and soil samples were separated into 5 groups (Group 1-5 referred to 0-5, 5-10, 10-20, 20-30 and above 30 km, respectively.). Samples in group 1 (0-5 km) were all collected in the E-waste site.

Sample Analysis All samples were freeze-dried, grinded to fine powder and sealed in polyethylene bottles at 4 ^{o}C until analysis. Sample pretreatment, and instrumental analysis followed our previously established methods (Liu, et al, 2006 a, b). 14 PBDE congeners (BDEs 17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, and 209) and 25 PCB congeners, including 12 coplanar congeners (CBs 77,

81, 105, 114, 118, 123, 126, 156, 157, 167, and 169), six indicator congeners (CBs 28, 52, 101, 138, 153, and 180), and other congeners (CBs 3, 15, 19, 202, 205, 208, and 209) were analyzed.

Result and Discussion

All the results in this study were expressed on a dry weight basis. PCBs and PBDEs were found in all apple snail samples.$\sum_{25}$PCBs concentrations in apple snails ranged from 3.78 to 1812 ng/g with the geometric mean (GM) of 24.6 ng/g. The highest concentration of $\sum_{25}$PCBs (1812 ng/g) was found in Group 1. Ten apple snail samples and all four samples from Group 1 exceeded the USEPA screening value of 20 ng/g (wet weight)(USEPA, 2000).

Contaminant levels of spatial distributions varied depending on the sampling location. $\sum_{25}$PCBs concentrations in apple snails decreased dramatically with increasing distance from the E-waste recycling site, which indicated that this site is the primary source of PCBs in this region. $\sum_{13}$PBDEs (excluding BDE-209) concentrations were in the range of 0.09 and 27.66 ng/g with the GM of 0.72 ng/g in apple snails, which were also negatively related to the distance from the E-waste recycling site generally. BDE-209 could be detected in 67.5% of the snail samples. The highest concentration of BDE-209 (45040 ng/g dw) in snail samples was found in Group 1. On the whole, $\sum_{25}$PCB and $\sum_{13}$PBDE concentrations (GM) in Group 1 were about 100 times higher than that in Group 5. Compared to previous study in edible sea food (Domingo and Bocio, 2007; Jaikanlaya, et al, 2009, Wang et al, 2008; Meng, et al, 2007), PCBs and PBDEs in apple snails from Group 1 were at the high end of the global scale. This result also showed that PCB pollutions were more serious than PBDEs since obsolete transformers and electromotors have consistently been recycled in this E-waste recycling area in the past 30 years.

CB-28 was the most abundant congener, which accounted for 41.3% of $\sum_{25}$PCBs in apple snails. Six indicator PCBs (CB- 28, 52, 101, 138, 153, and 180) represented 79.5% of $\sum_{25}$PCBs. For PBDEs, BDE-209 was the most predominant congener, which accounted for 83.8% of the 14 PBDE congeners in apple snail samples. BDE-47, 71, 99, 153, and 183 were found prevalent in apple snail samples. Deca-BDE technical mixture was considered to be the major potential source of PBDEs in this region.

Generally, spatial distributions of PCBs and PBDEs in soil samples were consistent with that of apple snail samples. $\sum_{25}$PCBs and $\sum_{13}$PBDE in soils ranged from 0.48 to 90.1 and 0.06 to 31.2 ng/g, respectively. Higher concentrations of $\sum_{25}$PCBs and $\sum_{13}$PBDEs were present in soil samples in Group 1 and Group 2, and then gradually decreased from Group 3 to 5. It is therefore suspected that relative high concentrations of PCBs and PBDEs have diffused to surrounding regions from the E-waste recycling site, and it also proved that the direct influencing area of PCB and PBDE pollutions from the E-waste dismantling activities is over a radius of 30 km.

Significant linear relationship were found between PCB concentrations and PBDE concentrations in soil and biota samples (R is up to 0.99 and 0.96, respectively, $p<0.05$). The result implied that congener profiles of PCBs and PBDEs in snails could reflect the levels and characteristics of these two groups of contaminants in the ambient environment. Apple snails are easy to catch due to their wide

distribution, abundance, and their range of movement during their lifespan was comparative narrow. All these attributes makes Apple snails, as according to the criterion by Tanabe and Subramanian (Tanabe and Subramanian, 2006), ideal bioindicators. The results in this paper implied that apple snail should perhaps be considered as an appropriate bioindicator to indicate contamination profiles of these two groups of chemicals in this region.

Reference

Domingo, J. L., Bocio, A., 2007. Environ. Int. 33: 397-405.

Liu, H. X., Zhang, Q. H., Cai, Z. W., Li, A., Wang, Y. W., Jiang, G. 2006, Anal. Chim. Acta 557 : 314-320.

Liu, H. X., Zhang, Q. H., Song, M. Y., Jiang, G. B., Cai, Z. W. 2006, Talanta 70: 20-25.

Meng, X. Z., Zeng, E. Y., Yu, L. P., Mai, B. X., Luo, X. J., Ran, Y. 2007. Environ. Sci. Technol. 41: 1821-1827.

Ni, H. G., Zeng, E. Y., 2009. Environ. Sci. Technol. 43 : 3991-3994.

Ogunseitan, O. A., Schoenung, J. M., Saphores, J. D. M., Shapiro, A. A.2009, Science 326: 670-671.

Tanabe, S., Subramanian, A. , Bioindicators of POPs: Monitoring in developing countries. Kyoto University Press: Kyoto **2006**.

UNEP DEWA/GRID-Europe, E-waste, the hidden side of IT equipment's manufacturing and use. Chapter 5: Early Warning on Emerging Environmental Threats. **2005**

USEPA., Guidance for assessing chemical contaminant data for use in fish advisories. USEPA,. **2000**.

Wang, Y. W., Wang, T. H., Li, A., Fu, J. J., Wang, P., Zhang, Q. H., Jiang, G. B. 2008. Environ. Sci. Technol. 42 : 7159-7165.

Wong, M. H., Wu, S. C., Deng, W. J., Yu, X. Z., Luo, Q., Leung, A. O. W., Wong, C. S. C., Luksemburg, W. J., Wong, A. S.2007, Environ. Pollut. 149: 131-140.

Jaikanlaya, C., Settachan, D., Denison, M. S., Ruchirawat, M., van den Berg, M. 2009. Chemosphere 76: 239-249.