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Exchange of organohalogen compounds between air and tree bark in the Yellow River region

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HIGHLIGHTS

- Air and tree bark OHCs concentrations in the Yellow River region were determined.
- Constant B was firstly and simultaneously determined for lots of individual OHCs.
- Reliability of tree bark to be used as air passive sampler was proved.
- Exchange model was verified and the optimum condition for compound was discussed.

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ABSTRACT

Organohalogen compound concentrations in paired air and bark samples from the Yellow River region were determined. Overall, the organohalogen compound concentrations were higher in the samples from the lower than from the upper Yellow River region. The polybrominated diphenyl ether, polychlorinated biphenyl, and organochlorine pesticide concentrations were 310–5200, 0.92–3.8, and 120–6700 pg m^{-3}, respectively, in the air samples and 29,000–190,0000, 220–1400, and 49,000–220,0000 pg g^{-1} lipid weight, respectively, in the bark samples. The concentrations in the air samples were significantly positively correlated with the concentrations in the bark samples. Constant B, related to the partitioning of a contaminant between the gas and particle phases in the air, was calculated for each compound. This was the first time constant B was simultaneously been determined for a range of different organohalogen compounds. An air-tree bark exchange model was calibrated and verified. The exchange coefficients (K_{BA}) that were determined were compared with the model results, and the optimum K_{OA} values for use in the model were found to be 10^9–10^{16}. The compound of interest needed to be detected in more than 50% of the samples for the model results to be valid.

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1. Introduction

Organohalogen compounds (OHCs) are widely used around the world, are persistent, can bioaccumulate, and can be transported over long distances in the environment (Covaci et al., 2011a; Yang et al., 2016). Organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), and brominated flame retardants, such as polybrominated diphenyl ethers (PBDEs), Dechlorane Plus (DP), and some novel brominated flame retardants (NBFRs), are all OHCs (Dirtu et al., 2013). PBDEs, which have been widely used as flame retardants (Covaci et al., 2011b), have been found in various environmental matrices and human tissues (Hoh et al., 2006; Qiu et al., 2010) and have been found to cause serious ecological and environmental problems (Van den Berg et al., 2006). The pentabromodiphenyl ether and octabromodiphenyl ether commercial products have been banned worldwide because their components are potentially toxic and have been found in the environment (Stapleton et al., 2009). The use of the decabromodiphenyl ether commercial product has been banned in the EU and in some states in the USA (de Wit et al., 2010). Some NBFRs have been considered for use as replacements for PBDEs, although it has been suggested that these NBFRs may also be toxic (Covaci et al., 2011b). PCBs, which were used widely as dielectric fluids, hydraulic fluids, and organic diluents in electrical transformers and capacitors (Jones and de Voogt, 1999), have been classed as persistent organic

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pollutants (POPs) by the Stockholm Convention (Harrad et al., 1994). A number of OCPs have also been classed as POPs because they are poorly biodegradable, highly toxic, can bioconcentrate, can undergo long-range atmospheric transport, and have been found in environmental matrices (Norstrom et al., 1988). OCPs have been widely used in China because they are cheap and very efficient pesticides with low acute toxicities. The use of OCPs has gradually been phased out and banned since 1983, but considerable amounts of OCP residues are still present in the environment because OCPs are persistent (Feng et al., 2011).

The concentrations of OHCs in the atmosphere are important air quality indicators. The concentrations of OHCs and other pollutants in air are usually measured in samples collected using active air sampling equipment. However, conventional high-volume air sampling equipment is expensive, and using it is relatively labor intensive. Passive air sampling is a much less expensive and labor intensive alternative way of estimating air concentrations of OHCs and other pollutants. Tree bark acts as a natural passive air sampler, continuously collecting pollutants from the surrounding air. Compounds with high octanol-air partition coefficients, in particular, tend to accumulate in tree bark because bark has a high lipid content and a large surface area (Salamova and Hites, 2010, 2013). OHC concentrations in tree bark therefore reflect the OHC concentrations in the atmosphere in the area around the tree the bark is on. The use of tree bark as a passive bio-sampler for determining atmospheric concentrations of PBDEs (Zhu and Hites, 2006), OCPs (Àngels Olivella et al., 2012), PCBs (Meredith and Hites, 1987; Hermanson and Hites, 1990), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (Di Lella et al., 2006), and DP (Qiu and Hites, 2008) has been considered.

The Yellow River is the second longest river in China. It originates in the Bayan Har Mountains and flows through nine provinces (Qinghai, Sichuan, Gansu, Ningxia, Inner Mongolia, Shaanxi, Shanxi, Henan, and Shandong) and empties into the Bohai Sea. Most recent studies of OHCs in the Yellow River region have focused on the lower reaches of the river (He et al., 2006; Ao et al., 2009), and little information is available on OHC concentrations in the upper reaches of the river. In this study, we analyzed air and tree bark samples collected across the whole Yellow River region to determine the OHC concentrations and identify potential local sources of OHCs throughout the region. We then used the results to determine the effectiveness of using tree bark as a passive air sampler for determining OHC concentrations in air.

2. Materials and methods

2.1. Sample information

Eight paired air and tree bark samples were collected from sites near the Yellow River in July and August 2012. More detailed information is given in SI. Particle and gas phase samples were collected using a high volume air sampler. Tree bark samples were collected from near the air sampling sites in the same period as the air samples were collected in.

2.2. Chemicals

The chemicals used included acetone, *n*-hexane, and dichloromethane (all of pesticide grade; J.T. Baker, Phillipsburg, NJ, USA), ultra-pure water (produced using a Milli-Q system; Millipore, Billerica, MA, USA), high purity nitrogen, helium, and methane (Chen Wei Xin, Beijing, China), anhydrous sodium sulfate (analytical grade, baked for 5 h at 450 °C; Sinopharm, Beijing, China), silica gel (100–200 mesh; Merck, Darmstadt, Germany), and neutral alumina (60 mesh; Alfa Aesar, Ward Hill, MA, USA). The silica gel and alumina were extracted with dichloromethane and then activated at 105 °C (silica gel) or 130 °C (alumina) for 12 h.

Unlabeled PBDE standards were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). Unlabeled PCB standards and a hexabromobenzene (HBB) standard were purchased from Dr. Ehrenstorfer (Augsburg, Germany). Unlabeled *anti*-DP, *syn*-DP, 1,2,3,4,5-penta-bromobenzene (PBBz), and 2,3,5,6-tetrabromo-*p*-xylene (pTBX) were purchased from Wellington Laboratories (Guelph, Canada). An unlabeled OCP standard mixture and unlabeled 2,3,5,5,6-pentabromoethylbenzene (PBEB) and pentabromotoluene (PBT) standards were purchased from AccuStandard (New Haven, CT, USA).

Standards of $^{13}C_{12}$-labeled PCBs, $^{13}C_{12}$-labeled BDE-139, $^{13}C_{12}$-labeled BDE-209, $^{13}C_{10}$-labeled *syn*-DP, and $^{13}C_{6}$-labeled HBB were purchased from Cambridge Isotope Laboratories. Standards of ^{13}C-labeled OCPs were purchased from Wellington Laboratories.

2.3. Sample preparation

A 10 g aliquot of each tree bark sample was cut into small pieces (each piece with an area of <1 cm^2), added to a Soxhlet extraction apparatus, and covered with 20.0 g anhydrous sodium sulfate. Each air sample was placed directly into a Soxhlet extraction apparatus. Each sample was spiked with internal standards and then Soxhlet extracted with 200 mL of a mixture of hexane and acetone (1:1 v/v) for 24 h. Each extract was then concentrated to about 2 mL by rotary evaporation, and then divided into two equal aliquots. One aliquot was cleaned using a multilayer silica column (packed, from bottom to top, with 1 g activated silica, 4 g silica containing 30% w/w 1 M $NaOH_{(aq)}$, 1 g activated silica, 8 g silica containing 44% w/w $H_2SO_{4(conc)}$, 2 g activated silica, and 4 g granular anhydrous sodium sulfate), which was eluted with 100 mL of a mixture of hexane and dichloromethane (97:3 v/v). The second aliquot was cleaned using a silica and alumina column (packed, from bottom to top, with 2 g anhydrous sodium sulfate, 8 g silica gel deactivated with 3% water, 8 g neutral alumina, and 2 g anhydrous sodium sulfate), which was eluted with 100 mL of a mixture of hexane and dichloromethane (7:3 v/v). The extracts were evaporated to 100 μL by rotary evaporation and then under a gentle stream of nitrogen. The first aliquot of each extract was analyzed for PBDEs, PCBs, and NBFRs, and the second aliquot of each extract was analyzed for the other analytes.

2.4. Analytical methods

The instrumental analytical methods that were used have been described previously (He et al., 2014a,b), and available in SI. Briefly, all of the analytes were determined using an Agilent 6890 gas chromatograph (Agilent Technologies, Santa Clara, CA, USA) equipped with an Agilent 5975 mass spectrometer.

2.5. Quality control and assurance

Method and field blank samples were analyzed. The analyte concentrations in the blank samples were satisfactory (<5% of the concentration found in a typical sample) for all of the analytes except for the HCB, so blank correction was only necessary for the HCB. The recoveries of the surrogate standards spiked into the samples were >70%, and the method limits of detection (MLDs) were 0.005–2.5 pg m^{-3} for the air samples and 20–10,000 pg g^{-1} lipid weight (l.w.) for the bark samples.

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