Supporting information

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Analytical Methods of PBDEs

Sample Pretreatment. The air-dried soil samples were homogenized and sieved through an 80-mesh (i.d. 0.18 mm) mesh. About 15 g soil of each sample was Soxhlet-extracted with a mixture of acetone and hexane (1:1 in volume) for 72h after spiked with two surrogate standards (^{13}C-PCB141 and PCB209), and copper granules were added for desulfuring. The extract was concentrated, and solvent-exchanged to hexane to approximately 1mL, and then loaded on a 1 cm i.d. multi-layer silica/alumina glass column, packed with, from bottom to top, neutral alumina (6 cm, 3% deactivated), neutral silica gel (2 cm, 3% deactivated), sodium hydroxide silica (5 cm, 25%), neutral silica gel (2 cm, 3% deactivated), sulfuric acid silica (10 cm, 44%), and anhydrous sodium sulfate (1 cm). The PBDEs fractions were eluted with 30 mL hexane, and followed by 60 mL mixture solvent of dichloromethane and hexane (1:1 in volume). The elution was concentrated to 0.2 mL under a gentle nitrogen stream. Internal standard (^{13}C-PCB208) was added into vials before instrumental analysis.

Instrumental Analysis. BDE85 was not quantified because of the chromatographic interferences in some of the samples. The pretreated samples were analyzed by a Shimadzu Model 2010 gas chromatograph coupled with a QP2010 mass spectrometer (Shimadzu, Japan). A DB-XLB capillary column (30 m × 0.25 mm i.d. and 0.25 μm film thickness) was used for determination of ten PBDE congeners. BDE209 was analyzed separately on a much shorter CP-Sil 13CB (12.5 m × 0.25 mm i.d. and 0.2 μm film thickness) capillary column as it was prone to degradation at high column temperature and long-term analysis during the GC elution (*1*). For $\sum_{10}$PBDEs (include BDE85), the GC system was operated in splitless injection mode, and the split mode was turned on

after 1min, helium was used as the carrier gas, with a column flow of 1.0 mL/min .The oven and injection temperature were110 ^{o}C and 290 ^{o}C, respectively, column temperature was initiated at 110 ^{o}C (held for 1 min), then increased to 180 ^{o}C at 8 ^{o}C /min (held for 1min), 240 ^{o}C at 2 ^{o}C /min (held for 5 min), 280 ^{o}C at 2 ^{o}C /min (held for 15 min), and finally reached to 300 ^{o}C at 10 ^{o}C /min (held for 20 min). For BDE209, only column temperature program was changed, with an initial temperature of 110 ^{o}C (held for 1 min) to 300 ^{o}C at 8 ^{o}C /min (held for 17 min). The injective volume was 1μL, manual injected.

For MS system, negative chemical ionization (NCI) in the selected ion monitoring (SIM) mode was processed. Methane was used as reagent gas at an ion source pressure of 2.4 MPa. The temperature of Ion source and Interface were 250 ^{o}C and 285 ^{o}C, respectively. The m/z 79 and 81($[Br]^{-}$) were monitored for tri- to hepta- BDEs, and m/z 486.7, 488.7 for BDE209. For surrogate standards, m/z 372, 374 and 376 were monitored for ^{13}C-PCB141 and m/z 496, 498, and 500 for PCB209, moreover, m/z 474, 476, and 478 were monitored for ^{13}C-13PCB208.

Literature cited:

(1) Covaci, A.; Voorspoels, S.; de Boer, J. Determination of brominated flame retardants, with emphasis on polybrominated diphenyl ethers (PBDEs) in environmental and human samples–a review. *Environ. Int.* **2003**, *29*, 735–756.

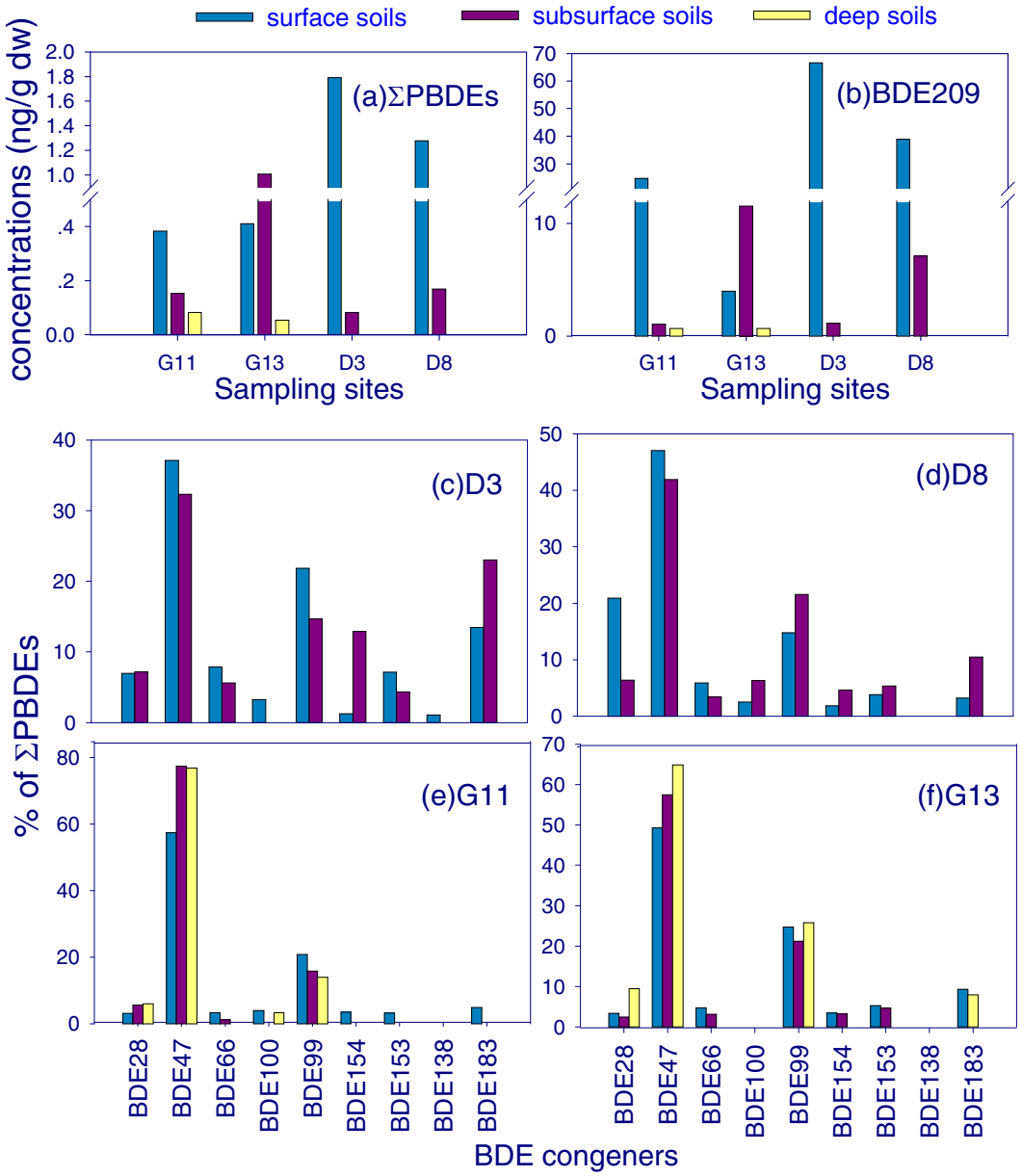

FIGURE S1. The vertical distribution of concentrations and compositional profiles for the surface, subsurface, and deep soils (G11, G13, D3, and D8) (a) The concentrations of PBDEs, (b) The concentrations of BDE209; (c)-(f) are the compositional profiles of $\sum_9$PBDEs for the surface, subsurface, and deep soils at C11 (c), C13 (d), D3 (e), and D8 (f) sampling sites, respectively. Only subsurface soils were collected at D3 and D8.

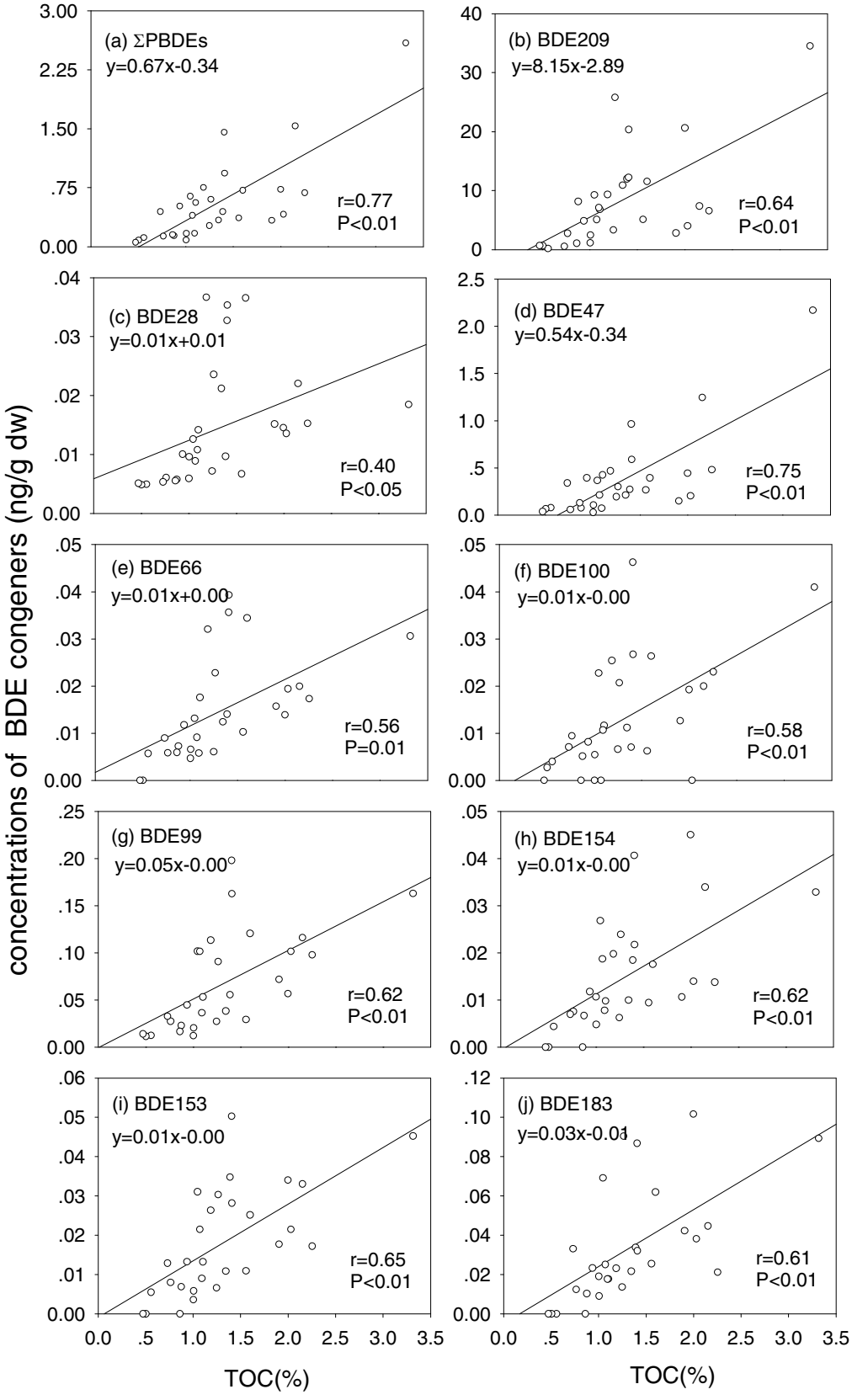

FIGURE S2. Correlation between BDE congeners and TOC. (a) $\sum_9$PBDEs, (b) BDE209, (c) BDE28, (d) BDE47, (e) BDE66, (f) BDE100, (g) BDE99, (h) BDE154, (i) BDE153, and (j) BDE183.