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*Supplement of*

# Spatial distribution of the persistent organic pollutants across the Tibetan Plateau and its linkage with the climate systems: a 5-year air monitoring study

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**Summary of the number of pages, figures, and tables**

## Text S1 Sampling program of the active air sampling

The sampling period was from November 2008 to September 2011, and the samples were collected every 2 weeks. Polyurethane foam (PUF, 7.5 cm × 6 cm diameter) was used to collect the gas-phase OCPs and PCBs. The particulates were collected on glass fiber filters. A pair of PUF plugs (7.5 cm × 3 cm) was installed in series inside the sampling cylinder to monitor potential breakthrough. Typically, 500-700 $m^3$ of air was collected over a two-week period. Before sampling, all of the PUF plugs were pre-cleaned by a Soxhlet extraction using dichloromethane (DCM) for 24 hours and the filters were baked at 450 °C for 5 hours. After sampling, the harvested PUF plugs and filters were stored in sealed, solvent-rinsed, glass jars and stored at -20 °C until extraction.

**Text S2 Chemical extraction, cleanup and details on the gas chromatographic temperature**

The retrieved XAD sample was transferred into the Soxhlet body and spiked with the recovery standards of 2,4,5,6-tetrachloro-m-xylene (TCmX), and decachlorobiphenyl (PCB-209). Samples were Soxhlet-extracted using DCM for 24 hours. The extracts were first concentrated by a rotary evaporator and solvent exchanged to hexane. Then, the samples were loaded on the top of a chromatography column (from the top to bottom: 1g of anhydrous sodium sulfate, 2 g of 3% deactivated alumina, and 3 g of 6% deactivated silica gel). The column was eluted with 30 mL of a mixture of DCM and hexane (1:1). The elute was further cleaned using gel-permeation chromatography (GPC, containing 6 g of Biobeads SX3), and the samples were finally solvent exchanged and concentrated in 20 μL of dodecane containing a known quantity of pentachloronitrobenzene (PCNB) and PCB 54 as the internal standards. Details on the gas chromatographic temperature are given in text S2 (Supporting Information).

The samples were analyzed on a Finnigan-TRACE GC-MS system with a CP-Sil 8CB capillary column (50 m, 0.25 mm, 0.25 μm), operating under single-ion monitoring (SIM) mode. Helium was used as the carrier gas at 1 ml $min^{-1}$ under constant-flow mode. The oven temperature began at 100 °C for 2 min and was increased at a rate of 20 °C $min^{-1}$ to 140 °C, then increased at a rate of 4 °C $min^{-1}$ to 200 °C, held for 10 min, and finally increased at a rate of 4 °C $min^{-1}$ to 300 °C and held for 17 min.

**Table S1. Data of field blank concentrations and method detection limits (MDL, ng/sample) for each sampling year**

| Sample name | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | ∑PCBs |
|---|---|---|---|---|---|---|---|---|
| 2007-08 blank[a] | 0.27 | 0.68 | ND | 0.44 | 0.34 | ND | ND | 0.19 |
| 2007-08 MDL[a] | 0.28 | 1.2 | 0.03 | 0.47 | 0.36 | 0.03 | 0.03 | 0.18 |
| 2008-09 blank1 | 0.01 | 0.01 | 0.78 | ND | 0.21 | ND | ND | 0.16 |
| 2008-09 blank2 | ND | 0.02 | 0.39 | 0.12 | 0.23 | ND | ND | 0.12 |
| 2008-09 blank3 | ND | ND | 0.16 | ND | 0.14 | 0.28 | ND | 0.08 |
| 2008-09 blank4 | ND | ND | 0.35 | 0.02 | 0.25 | ND | ND | 0.14 |
| 2008-09 blank5 | ND | 0.10 | 0.33 | 0.14 | ND | ND | ND | 0.13 |
| Blank average | 0.01 | 0.04 | 0.40 | 0.09 | 0.21 | 0.28 | ND | 0.13 |
| 2008-09 MDL | 0.20 | 0.19 | 1.09 | 0.29 | 0.35 | 0.20 | 0.20[b] | 0.21 |
| 1/2 MDL | 0.1 | 0.1 | 1.55 | 0.15 | 0.17 | 0.1 | 0.1 | 0.1 |
| 2009-10 blank 1 | ND | ND | 0.42 | ND | 0.40 | ND | 0.41 | 0.05 |
| 2009-10 blank 2 | ND | ND | 0.63 | ND | 0.26 | ND | ND | 0.11 |
| 2009-10 blank 3 | ND | ND | 0.55 | ND | 0.27 | ND | ND | 0.08 |
| 2009-10 blank 4 | 0.42 | 0.14 | 0.26 | 0.13 | 0.39 | 0.23 | 0.53 | 0.07 |
| 2009-10 blank 5 | 0.05 | 0.04 | 0.51 | ND | 0.29 | ND | 0.25 | 0.09 |
| Blank average | 0.24 | 0.09 | 0.47 | 0.13 | 0.32 | 0.23 | 0.40 | 0.08 |
| 2009-10 MDL | 1.02 | 0.30 | 0.90 | 0.20 | 0.52 | 0.20 | 0.82 | 0.15 |
| 1/2 MDL | 0.5 | 0.15 | 0.45 | 0.1 | 0.26 | 0.1 | 0.41 | 0.08 |
| 2010-11 blank 1 | 0.05 | 0.03 | 0.37 | ND | 0.22 | ND | 0.19 | 0.24 |
| 2010-11 blank 2 | 0.02 | 0.02 | 0.22 | 0.01 | 0.10 | 0.03 | 0.02 | 0.11 |
| 2010-11 blank 3 | 0.08 | ND | 0.47 | ND | 0.17 | 0.07 | 0.18 | 0.16 |
| 2010-11 blank 4 | 0.06 | 0.06 | 0.33 | ND | 0.26 | 0.19 | 0.15 | 0.17 |
| 2010-11 blank 5 | 0.05 | 0.07 | 0.59 | ND | 0.24 | ND | 0.18 | 0.15 |
| 2010-11 blank 6 | 0.03 | 0.05 | 0.40 | ND | 0.09 | 0.06 | 0.05 | 0.08 |
| Blank average | 0.05 | 0.05 | 0.40 | 0.01 | 0.18 | 0.09 | 0.13 | 0.15 |
| 2010-11 MDL | 0.12 | 0.10 | 0.77 | 0.20 | 0.40 | 0.30 | 0.35 | 0.32 |
| 1/2 MDL | 0.06 | 0.05 | 0.38 | 0.1 | 0.2 | 0.15 | 0.17 | 0.16 |
| 2011-12 blank 1 | 0.04 | ND | 0.29 | 0.01 | 0.07 | 0.03 | 0.04 | 0.04 |
| 2011-12 blank 2 | 0.04 | ND | 0.27 | ND | 0.08 | 0.02 | 0.04 | 0.06 |
| 2011-12 blank 3 | 0.03 | ND | 0.23 | 0.02 | 0.08 | 0.02 | 0.04 | 0.03 |
| 2011-12 blank 4 | 0.05 | 0.03 | 0.39 | 0.01 | 0.09 | 0.07 | 0.01 | 0.07 |
| 2011-12 blank 5 | 0.07 | 0.04 | 0.61 | 0.01 | 0.05 | 0.17 | 0.19 | 0.05 |
| 2011-12 blank 6 | 0.03 | 0.01 | 0.26 | 0.02 | 0.09 | 0.16 | 0.16 | 0.02 |
| Blank average | 0.04 | 0.03 | 0.34 | 0.01 | 0.08 | 0.08 | 0.08 | 0.05 |
| 2011-12 MDL | 0.09 | 0.07 | 0.76 | 0.04 | 0.12 | 0.29 | 0.30 | 0.10 |
| 1/2 MDL | 0.05 | 0.04 | 0.4 | 0.02 | 0.06 | 0.15 | 0.15 | 0.05 |

The methods detection limits (MDLs) were derived as the mean field blank concentration plus 3 times of its standard deviation.

a: The data have been reported in Wang et al. 2010.

**Table S2 Concentration presented as amounts of POPs sequestered over the sampling period (ng/sampler)**

| Sampling site | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | PCB-28 | PCB-52 | PCB-101 | PCB-153 | PCB-138 | PCB-180 | $\sum_6$ PCBs |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **2007-2008** | | | | | | | | | | | | | | |
| Xigaze | 3.1 | 1.2 | 24.7 | 3.1 | 4.9 | 26 | 10.7 | 0.51 | 0.48 | 0.34 | 0.42 | 1.04 | *ND* | 2.81 |
| Nam co | 0.1 | *0.6* | 2.5 | 2.3 | *0.18* | 2.9 | 0.5 | 0.1 | 0.06 | *0.02* | 0.07 | 0.18 | *ND* | 0.45 |
| Lhasa | 4.1 | 1.4 | 56.2 | 2.2 | 5.7 | 22.3 | 6.2 | 0.54 | 0.15 | *0.02* | 0.23 | 0.54 | *ND* | 1.5 |
| Lhaze | 1.6 | 0.8 | 22 | 1 | 1 | 9.4 | 2.5 | 0.4 | 0.14 | 0.11 | 0.08 | 0.2 | *ND* | 0.95 |
| Lulang | 1.6 | 0.5 | 16.9 | 2.9 | 3.9 | 28.5 | 6.5 | 0.53 | 0.44 | *0.02* | 0.12 | 0.6 | *ND* | 1.73 |
| Everest | 4.5 | 5.4 | 22 | 0.1 | 0.3 | 3.1 | 0.5 | 2.1 | 0.1 | *0.02* | *0.02* | *0.02* | *ND* | 2.28 |
| Naqu | 7.3 | 1.9 | 27.4 | 1.7 | 0.8 | 15.3 | 4.1 | 0.28 | 0.25 | 0.1 | 0.07 | 0.65 | *ND* | 1.37 |
| Qamdo | 1.2 | 1.1 | 16.1 | 6.7 | 11 | 34.4 | 9.4 | 1.06 | 0.22 | *0.02* | 0.21 | 1.04 | *ND* | 2.57 |
| Saga | 4.9 | 1.6 | 20.2 | 3.2 | 5.7 | 28.5 | 9 | 0.4 | 0.3 | 0.16 | 0.08 | 0.25 | *ND* | 1.21 |
| Rawu | 16.4 | 18.7 | 11.8 | 1.1 | 4.1 | 38.9 | 7.2 | 2.38 | 0.17 | *0.02* | *0.02* | *0.02* | *ND* | 2.63 |
| Bomi | 6.5 | 3.6 | 60.7 | 2 | 4.2 | 11.5 | 3 | 0.89 | 0.48 | *0.02* | *0.02* | 1.38 | *ND* | 2.81 |
| GBJD | 6.9 | 2.1 | 23.1 | 3.4 | 6.8 | 28.1 | 6.5 | 0.4 | 0.23 | *0.02* | 0.26 | 0.93 | *ND* | 1.86 |
| Golmud | 2.9 | 1 | 44.2 | 1.5 | 1.2 | 1.4 | 0.5 | 0.63 | 0.27 | *0.02* | *0.02* | *0.02* | *ND* | 0.98 |
| Gar | 3.7 | 13.0 | 8.4 | 0.9 | 1.9 | 9.3 | 2.1 | 1.25 | 0.11 | *0.02* | *0.02* | *0.02* | *ND* | 1.44 |
| Chayu | / | / | / | / | / | / | / | / | / | / | / | / | / | / |
| Muztagata | / | / | / | / | / | / | / | / | / | / | / | / | / | / |
| **2008-2009** | | | | | | | | | | | | | | |
| Xigaze | 6.63 | 2.59 | 12.06 | 0.97 | 1.33 | 2.55 | 3.28 | *0.06* | 0.41 | *0.12* | 0.03 | *0.05* | *ND* | 0.69 |
| Nam co | 3.61 | 1.32 | 26.45 | 0.31 | 0.36 | 1.33 | 0.99 | 0.22 | 0.62 | *0.12* | 0.07 | *0.05* | *ND* | 1.09 |
| Lhasa | 4.96 | 2.91 | 29.92 | *0.15* | 0.93 | 4.85 | 2.44 | 0.22 | 0.54 | *0.12* | *0.01* | *0.05* | *ND* | 0.95 |
| Lhaze | 4.35 | 1.07 | 18.66 | *0.15* | 0.51 | 2.26 | 2.50 | 0.22 | 0.70 | *0.12* | 0.04 | 0.11 | *ND* | 1.22 |
| Lulang | 2.89 | 0.88 | 8.49 | 0.84 | 0.80 | 4.66 | 5.61 | *0.06* | 0.37 | *0.12* | 0.02 | *0.05* | *ND* | 0.63 |

| | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Everest | 5.01 | 1.73 | 24.24 | *0.15* | 0.55 | 5.83 | 4.72 | 0.11 | 0.47 | *0.12* | *0.01* | *0.05* | *ND* | 0.79 |
| Naqu | 11.01 | 2.64 | 12.56 | *0.15* | *0.17* | 1.18 | 2.12 | *0.06* | 0.39 | *0.12* | 0.03 | *0.05* | *ND* | 0.67 |
| Qamdo | 2.23 | 0.89 | 11.08 | *0.15* | 1.26 | 0.40 | 2.15 | 0.21 | 0.56 | *0.12* | 0.06 | *0.05* | *ND* | 1.04 |
| Saga | 4.66 | 1.32 | 14.54 | *0.15* | 0.96 | 3.14 | 3.39 | 0.19 | 0.50 | *0.12* | 0.03 | *0.05* | *ND* | 0.91 |
| Rawu | 1.65 | 0.34 | 6.32 | *0.15* | *0.17* | 0.70 | 1.58 | *0.06* | 0.13 | *0.12* | *0.01* | *0.05* | *ND* | 0.38 |
| Bomi | 14.00 | 3.91 | 26.95 | 0.60 | 4.14 | 9.65 | 21.91 | 0.15 | 0.49 | *0.12* | 0.03 | *0.05* | *ND* | 0.87 |
| GBJD | / | / | / | / | / | / | / | / | / | */* | / | / | | / |
| Golmud | 4.84 | 2.30 | 33.85 | *0.15* | *0.17* | *0.10* | *0.10* | 0.25 | 0.70 | *0.12* | *0.01* | *0.05* | *ND* | 1.14 |
| Gar | 6.55 | 1.98 | 30.14 | 0.65 | 0.97 | *0.10* | 1.07 | 0.27 | 0.74 | *0.12* | 0.03 | *0.05* | *ND* | 1.22 |
| Chayu | 3.34 | 0.84 | 16.10 | *0.15* | 1.09 | 5.97 | 15.54 | *0.06* | 0.30 | *0.12* | *0.01* | *0.05* | *ND* | 0.55 |
| Muatagata | / | / | / | / | / | / | / | / | / | / | / | / | / | / |
| **2009 - 2010** | | | | | | | | | | | | | | |
| Xigaze | 2.03 | 0.63 | 9.83 | 3.16 | 4.70 | 8.97 | 3.85 | *0.07* | *0.02* | 0.14 | *0.02* | 0.05 | *ND* | 0.31 |
| Nam co | 3.14 | 0.84 | 15.98 | 0.66 | 1.46 | 11.46 | 6.37 | *0.07* | 0.12 | 0.70 | 0.16 | 0.22 | *ND* | 1.36 |
| Lhasa | 1.73 | 0.53 | 9.99 | 0.42 | 2.64 | 7.74 | 3.46 | *0.07* | 0.05 | 0.28 | *0.02* | 0.09 | *ND* | 0.52 |
| Lhaze | 3.88 | 1.33 | 11.52 | *0.10* | 1.56 | 4.53 | 3.20 | *0.07* | 0.05 | 0.20 | *0.02* | 0.05 | *ND* | 0.40 |
| Lulang | 3.10 | 0.75 | 8.50 | 0.35 | 2.07 | 11.73 | 5.81 | *0.07* | 0.06 | 0.17 | *0.02* | 0.07 | *ND* | 0.41 |
| Everest | 4.15 | 1.24 | 14.37 | 0.59 | 1.98 | 11.21 | 4.88 | *0.07* | 0.09 | 0.24 | *0.02* | 0.04 | *ND* | 0.47 |
| Naqu | 10.78 | 2.44 | 15.33 | *0.10* | 1.07 | 4.33 | 1.95 | *0.07* | 0.06 | 0.14 | *0.02* | 0.03 | *ND* | 0.33 |
| Qamdo | 2.99 | 1.18 | 11.58 | 0.23 | 1.86 | 3.24 | 2.67 | 0.16 | 0.16 | 0.21 | *0.02* | *0.02* | *ND* | 0.58 |
| Saga | 2.52 | 0.65 | 9.58 | *0.10* | 1.89 | 4.71 | 4.73 | *0.07* | 0.05 | 0.15 | *0.02* | *0.02* | *ND* | 0.32 |
| Rawu | 3.89 | 0.97 | 15.70 | 0.22 | 1.34 | 7.19 | 3.67 | *0.07* | 0.06 | 0.11 | *0.02* | *0.02* | *ND* | 0.29 |
| Bomi | 6.92 | 2.16 | 7.31 | 1.23 | 15.05 | 26.49 | 21.70 | 0.25 | *0.02* | 0.32 | *0.02* | 0.06 | *ND* | 0.68 |
| GBJD | / | / | / | / | / | / | / | / | / | / | / | / | / | / |
| Golmud | 6.03 | 1.59 | 33.31 | 0.20 | 1.22 | 5.15 | 1.74 | 0.24 | 0.28 | 0.32 | *0.02* | 0.08 | *ND* | 0.95 |
| Gar | 1.27 | 0.86 | 13.20 | *0.10* | 1.01 | 1.49 | 1.96 | *0.07* | 0.08 | 0.27 | *0.02* | 0.12 | *ND* | 0.57 |
| Chayu | 6.70 | 1.83 | 26.73 | 1.43 | 7.45 | 31.77 | 17.19 | 0.18 | 0.14 | 0.31 | *0.02* | 0.05 | *ND* | 0.71 |

| | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Muztaga | 1.83 | 0.37 | 16.28 | *0.10* | 1.00 | 1.01 | 1.30 | *0.07* | *0.02* | 0.17 | *0.02* | *0.02* | *ND* | 0.31 |
| **2010 - 2011** | | | | | | | | | | | | | | |
| Xigaze | 1.62 | 0.24 | 11.64 | 0.54 | 1.41 | 0.92 | *0.18* | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Nam co | 1.13 | 0.17 | 8.54 | *0.10* | *0.20* | 0.83 | 0.63 | 0.35 | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.54 |
| Lhasa | 0.73 | 0.11 | 5.27 | 0.24 | 0.99 | 1.16 | 0.59 | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Lhaze | 1.23 | 0.17 | 9.90 | *0.10* | *0.20* | 0.36 | *0.18* | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Lulang | 1.41 | 0.16 | 7.39 | 0.24 | 0.71 | 2.06 | 0.93 | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Everest | 2.30 | 0.55 | 22.23 | 0.28 | 0.73 | 2.39 | 0.87 | *0.12* | 0.08 | *0.07* | *0.02* | *0.05* | *ND* | 0.35 |
| Naqu | 6.04 | 1.07 | 12.83 | *0.10* | *0.20* | 0.71 | *0.18* | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Qamdo | 1.36 | 0.22 | 9.03 | *0.10* | 0.41 | 0.44 | *0.18* | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Saga | 3.42 | 0.54 | 16.28 | *0.10* | 0.67 | 0.95 | 1.06 | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Rawu | 1.53 | 0.33 | 7.11 | 0.21 | 0.55 | 1.26 | 0.41 | 0.38 | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.57 |
| Bomi | 3.83 | 0.58 | 5.05 | 0.26 | 4.13 | 1.96 | 1.12 | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| GBJD | 3.59 | 0.84 | 9.04 | 0.25 | 1.08 | 3.28 | 1.50 | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Golmud | 1.28 | 0.17 | 19.88 | *0.10* | *0.20* | 0.33 | *0.18* | *0.12* | 0.07 | *0.07* | *0.02* | *0.05* | *ND* | 0.34 |
| Gar | 0.67 | 0.11 | 8.97 | *0.10* | *0.20* | *0.15* | *0.18* | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Chayu | 1.31 | 0.16 | 9.02 | 0.66 | 4.94 | 8.06 | 5.36 | *0.12* | *0.04* | *0.07* | *0.02* | *0.05* | *ND* | 0.31 |
| Muztaga | 2.14 | 0.22 | 25.31 | *0.10* | 0.90 | *0.15* | *0.18* | *0.12* | 0.09 | *0.07* | *0.02* | *0.05* | *ND* | 0.36 |
| **2011 - 2012** | | | | | | | | | | | | | | |
| Xigaze | 1.10 | 0.27 | 6.71 | 0.31 | 0.78 | 1.56 | 0.77 | *0.03* | 0.03 | *0.05* | *0.01* | *0.01* | *ND* | 0.14 |
| Nam co | 1.31 | 0.34 | 10.07 | 0.09 | 0.22 | 1.98 | 0.75 | *0.03* | 0.03 | *0.05* | *0.01* | 0.02 | *ND* | 0.15 |
| Lhasa | 0.60 | 0.14 | 3.65 | 0.31 | 1.39 | 2.60 | 1.65 | *0.03* | 0.02 | 0.12 | 0.02 | 0.03 | *ND* | 0.23 |
| Lhaze | 1.15 | 0.21 | 5.77 | 0.10 | 0.37 | 1.51 | 1.03 | *0.03* | 0.02 | *0.05* | 0.02 | 0.02 | *ND* | 0.15 |
| Lulang | 1.75 | 0.42 | 5.22 | 0.29 | 0.89 | 5.33 | 3.15 | *0.03* | 0.04 | *0.05* | *0.01* | 0.02 | *ND* | 0.16 |
| Everest | 1.80 | 0.56 | 11.97 | 0.22 | 0.75 | 4.11 | 2.29 | *0.03* | 0.05 | 0.12 | 0.02 | 0.03 | *ND* | 0.26 |
| Naqu | 6.48 | 1.45 | 14.72 | 0.13 | 0.39 | 1.73 | 1.58 | 0.06 | 0.06 | *0.05* | 0.02 | 0.02 | *ND* | 0.22 |
| Qamdo | 0.83 | 0.25 | 4.08 | 0.12 | 0.64 | 0.96 | 0.79 | *0.03* | 0.03 | 0.11 | 0.02 | 0.03 | *ND* | 0.23 |

| | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Saga | 1.23 | 0.37 | 6.45 | 0.09 | 0.29 | 0.97 | 0.53 | *0.03* | 0.03 | *0.05* | *0.01* | *0.01* | *ND* | 0.14 |
| Rawu | 1.09 | 0.29 | 4.46 | 0.19 | 0.52 | 2.81 | 1.60 | *0.03* | 0.03 | *0.05* | *0.01* | 0.02 | *ND* | 0.15 |
| Bomi | 1.13 | 0.28 | 2.76 | 0.40 | 2.95 | 5.47 | 3.79 | *0.03* | *0.01* | 0.15 | 0.08 | 0.11 | *ND* | 0.39 |
| GBJD | 2.19 | 0.48 | 7.36 | 0.27 | 1.14 | 5.11 | 22.68 | 0.06 | 0.04 | 0.13 | *0.01* | 0.02 | *ND* | 0.27 |
| Golmud | 1.97 | 0.73 | 18.98 | 0.05 | 0.49 | 0.41 | 0.56 | 0.14 | 0.08 | 0.20 | 0.02 | 0.05 | *ND* | 0.50 |
| Gar | 0.82 | 0.21 | 6.82 | 0.04 | 0.19 | 0.44 | 0.35 | *0.03* | 0.03 | *0.05* | *0.01* | *0.01* | *ND* | 0.14 |
| Chayu | 1.56 | 0.31 | 6.12 | 0.41 | 2.34 | 7.82 | 6.37 | *0.03* | 0.03 | 0.11 | 0.02 | 0.02 | *ND* | 0.22 |
| Muztagata | 2.25 | 0.33 | 22.00 | *0.02* | 1.20 | 0.80 | 1.30 | *0.03* | 0.02 | 0.15 | 0.02 | *0.01* | *ND* | 0.24 |

**Entries in italics are substituted values at 1/2 MDL.**

If the concentration of a compound after blank correction was below the MDL, the concentration was substituted with 1/2 MDL.

PCB 180 was never detected in samples

**Table S3. The concentrations of duplicates (ng/sample) and average RSD for each compound**

| Sampling year | Sample name | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | ∑PCBs |
|---|---|---|---|---|---|---|---|---|---|
| 2007-2008 | Xigaze a | 3.7 | 1.3 | 22 | 2.7 | 4.4 | 29 | 12 | 6.4[a] |
| | Xigaze b | 2.5 | 1.1 | 27 | 3.5 | 5.6 | 23 | 9.4 | 5.6 |
| | Lhasa a | 3.7 | 1.2 | 57 | 1.9 | 5.2 | 25.9 | 6.6 | 3.6 |
| | Lhasa b | 4.5 | 1.6 | 61.4 | 2.5 | 6.2 | 18.7 | 5.7 | 4.2 |
| | Lhaze a | 1.4 | 0.7 | 21.7 | 0.9 | 1.2 | 10.5 | 2.6 | 2.6 |
| | Lhaze b | 1.8 | 0.9 | 23.2 | 1.1 | 0.8 | 8.2 | 2.4 | 3.2 |
| | RSD | 20% | 17% | 8% | 17% | 19% | 19% | 11% | 12% |
| 2008-2009 | Xigaze a | 6.55 | 2.14 | 10.04 | 0.95 | 1.40 | 2.19 | 3.35 | 0.67[b] |
| | Xigaze b | 6.72 | 3.03 | 14.07 | 1.00 | 1.27 | 2.90 | 3.22 | 0.59 |
| | Namco a | 4.03 | 1.29 | 26.50 | 0.31 | 0.36 | 0.55 | 1.12 | 1.56 |
| | Namco b | 3.19 | 1.35 | 26.40 | 0.15 | 0.17 | 2.10 | 0.87 | 0.35 |
| | Lhaze a | 3.22 | 0.82 | 13.36 | 0.15 | 0.52 | 2.29 | 3.77 | 1.08 |
| | Lhaze b | 5.48 | 1.32 | 23.97 | 0.52 | 0.51 | 2.23 | 1.22 | 1.09 |
| | Lulang a | 2.78 | 0.84 | 7.94 | 0.91 | 0.42 | 6.36 | 5.23 | 0.29 |
| | Lulang b | 3.01 | 0.91 | 9.04 | 0.77 | 1.19 | 2.96 | 5.99 | 0.90 |
| | Everest a | 5.70 | 1.88 | 23.51 | 0.10 | 0.85 | 4.94 | 6.10 | 0.98 |
| | Everest b | 4.31 | 1.59 | 24.98 | 0.15 | 0.25 | 6.72 | 3.35 | 0.33 |
| | RSD | 16% | 15% | 16% | 34% | 41% | 36% | 29% | 48% |
| 2009-2010 | Namco a | 2.49 | 0.77 | 11.40 | 0.44 | 1.35 | 5.05 | 3.05 | 1.11[b] |
| | Namco b | 3.78 | 0.92 | 20.57 | 0.88 | 1.57 | 17.86 | 9.69 | 1.59 |
| | Lhasa a | 1.75 | 0.60 | 11.43 | 0.42 | 2.57 | 8.12 | 3.71 | 0.54 |
| | Lhasa b | 1.72 | 0.46 | 8.54 | 0.41 | 2.72 | 7.37 | 3.21 | 0.37 |
| | Everest a | 3.55 | 1.40 | 9.80 | 0.64 | 1.91 | 12.45 | 5.26 | 0.50 |
| | Everest b | 4.75 | 1.09 | 18.95 | 0.54 | 2.06 | 9.96 | 4.50 | 0.47 |
| | RSD% | 17% | 16% | 35% | 20% | 7% | 34% | 32% | 19% |
| 2010-2011 | Xigaze a | 1.74 | 0.29 | 11.36 | 0.68 | 1.38 | 1.05 | 0.26 | 0.16[b] |
| | Xigaze b | 1.50 | 0.19 | 11.92 | 0.40 | 1.45 | 0.78 | 0.22 | 0.16 |
| | Namco a | 0.97 | 0.10 | 7.64 | 0.08 | 0.40 | 0.73 | 1.13 | 0.67 |
| | Namco b | 1.29 | 0.24 | 9.45 | 0.13 | 0.19 | 0.93 | 0.12 | 0.16 |
| | Lhasa a | 0.70 | 0.10 | 5.43 | 0.24 | 1.12 | 1.25 | 0.81 | 0.16 |
| | Lhasa b | 0.75 | 0.11 | 5.10 | 0.24 | 0.87 | 1.07 | 0.38 | 0.16 |
| | Lhaze a | 1.44 | 0.21 | 9.73 | 0.07 | 0.21 | 0.34 | 0.04 | 0.16 |
| | Lhaze b | 1.03 | 0.13 | 10.06 | 0.05 | 0.09 | 0.39 | 0.03 | 0.16 |
| | Lulang a | 1.83 | 0.21 | 6.71 | 0.26 | 0.59 | 2.19 | 0.92 | 0.16 |
| | Lulang b | 0.98 | 0.11 | 8.06 | 0.22 | 0.83 | 1.94 | 0.95 | 0.16 |
| | Everest a | 2.59 | 0.64 | 26.61 | 0.34 | 0.96 | 3.05 | 1.05 | 0.16 |
| | Everest b | 2.01 | 0.46 | 17.86 | 0.22 | 0.51 | 1.74 | 0.68 | 0.16 |
| | RSD | 20% | 32% | 11% | 23% | 33% | 18% | 38% | 12% |
| 2011-2012 | Xigaze a | 1.19 | 0.30 | 7.06 | 0.32 | 0.82 | 1.77 | 0.87 | 0.17[b] |
| | Xigaze b | 1.02 | 0.24 | 6.36 | 0.29 | 0.75 | 1.34 | 0.67 | 0.14 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Namco a | 1.40 | 0.35 | 11.22 | 0.09 | 0.25 | 2.09 | 0.81 | 0.18 |
| Namco b | 1.23 | 0.32 | 8.93 | 0.09 | 0.18 | 1.86 | 0.68 | 0.17 |
| Lhasa a | 0.62 | 0.15 | 3.57 | 0.25 | 1.27 | 2.20 | 1.32 | 0.19 |
| Lhasa b | 0.57 | 0.14 | 3.72 | 0.36 | 1.51 | 3.00 | 1.99 | 0.22 |
| Lhaze a | 1.08 | 0.22 | 5.47 | 0.10 | 0.41 | 1.21 | 1.08 | 0.21 |
| Lhaze b | 1.22 | 0.19 | 6.07 | 0.11 | 0.33 | 1.81 | 0.98 | 0.16 |
| Lulang a | 1.35 | 0.36 | 4.59 | 0.22 | 0.65 | 4.64 | 2.44 | 0.18 |
| Lulang b | 2.16 | 0.49 | 5.84 | 0.36 | 1.12 | 6.03 | 3.86 | 0.25 |
| Everest a | 1.81 | 0.49 | 11.17 | 0.17 | 0.53 | 3.94 | 1.68 | 0.22 |
| Everest b | 1.80 | 0.63 | 12.78 | 0.27 | 0.97 | 4.28 | 2.89 | 0.29 |
| RSD | 11% | 13% | 10% | 19% | 22% | 17% | 23% | 15% |
| Average RSD for all duplicates | 17% | 20% | 15% | 16% | 24% | 24% | 27% | 21% |

a: $\sum_{15}$PCBs (data were from Wang et al., 2010);

b: $\sum_{6}$PCBs.

Duplicate samples were deployed at 6 sites (Lhasa, Xigaze, Lhaze, Mt. Everest, Lulang and Namco), however some samples were destroyed during extraction and cleanup process. Thus data of 3 duplicates were reported for sampling periods from 2007 to 2008, and from 2009-2010; data of 5 duplicates were reported for sampling periods from 2008 to 2009.

**Table S4**. **Estimated Volumetric Air Concentrations (pg m-3) from the Tibetan Plateau Sites**

| Sampling site | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | PCB-28 | PCB-52 | $\sum_6$ PCBs |
|---|---|---|---|---|---|---|---|---|---|---|
| **2007-2008** | | | | | | | | | | |
| Xigaze | 3.89 | 1.50 | 30.96 | 3.89 | 6.14 | 32.59 | 13.41 | 0.64 | 0.60 | 3.52 |
| Nam co | 0.12 | 0.72 | 2.99 | 2.76 | 0.22 | 3.47 | 0.60 | 0.12 | 0.07 | 0.54 |
| Lhasa | 4.84 | 1.65 | 69.84 | 2.60 | 6.72 | 26.31 | 7.31 | 0.64 | 0.18 | 1.77 |
| Lhaze | 1.90 | 0.95 | 26.19 | 1.19 | 1.19 | 11.19 | 2.98 | 0.48 | 0.17 | 1.13 |
| Lulang | 2.14 | 0.67 | 22.61 | 3.88 | 5.22 | 38.12 | 8.70 | 0.71 | 0.59 | 2.31 |
| Everest | 4.70 | 5.64 | 22.99 | 0.10 | 0.31 | 3.24 | 0.52 | 2.19 | 0.10 | 2.38 |
| Naqu | 9.70 | 2.52 | 36.39 | 2.26 | 1.06 | 20.32 | 5.45 | 0.37 | 0.33 | 1.82 |
| Qamdo | 1.55 | 1.42 | 20.84 | 8.67 | 14.24 | 44.52 | 12.17 | 1.37 | 0.28 | 3.33 |
| Saga | 5.11 | 1.67 | 21.07 | 3.34 | 5.95 | 29.73 | 9.39 | 0.42 | 0.31 | 1.26 |
| Rawu | 17.65 | 20.13 | 12.70 | 1.18 | 4.41 | 41.87 | 7.75 | 2.56 | 0.18 | 2.83 |
| Bomi | 9.10 | 5.04 | 85.00 | 2.80 | 5.88 | 16.10 | 4.20 | 1.25 | 0.67 | 3.93 |
| GBJD | 8.27 | 2.52 | 27.70 | 4.08 | 8.15 | 33.69 | 7.79 | 0.48 | 0.28 | 2.23 |
| Golmud | 4.93 | 1.70 | 75.17 | 2.55 | 2.04 | 2.38 | 0.85 | 1.07 | 0.46 | 1.67 |
| Gar | 4.13 | 14.51 | 9.37 | 1.00 | 2.12 | 10.38 | 2.34 | 1.39 | 0.12 | 2.18 |
| Chayu | / | / | / | / | / | / | / | / | / | / |
| Muztagata | / | / | / | / | / | / | / | / | / | / |
| **2008-2009** | | | | | | | | | | |
| Xigaze | 6.17 | 2.41 | 11.23 | 0.90 | 1.24 | 2.37 | 3.05 | 0.06 | 0.38 | 0.64 |
| Nam co | 3.13 | 1.14 | 22.90 | 0.27 | 0.31 | 1.15 | 0.86 | 0.19 | 0.54 | 0.94 |
| Lhasa | 4.51 | 2.65 | 27.20 | 0.14 | 0.85 | 4.41 | 2.22 | 0.20 | 0.49 | 0.86 |
| Lhaze | 3.89 | 0.96 | 16.70 | 0.13 | 0.46 | 2.02 | 2.24 | 0.20 | 0.63 | 1.09 |
| Lulang | 3.05 | 0.93 | 8.96 | 0.89 | 0.84 | 4.92 | 5.92 | 0.06 | 0.39 | 0.66 |
| Everest | 4.75 | 1.64 | 22.99 | 0.14 | 0.52 | 5.53 | 4.48 | 0.10 | 0.45 | 0.75 |

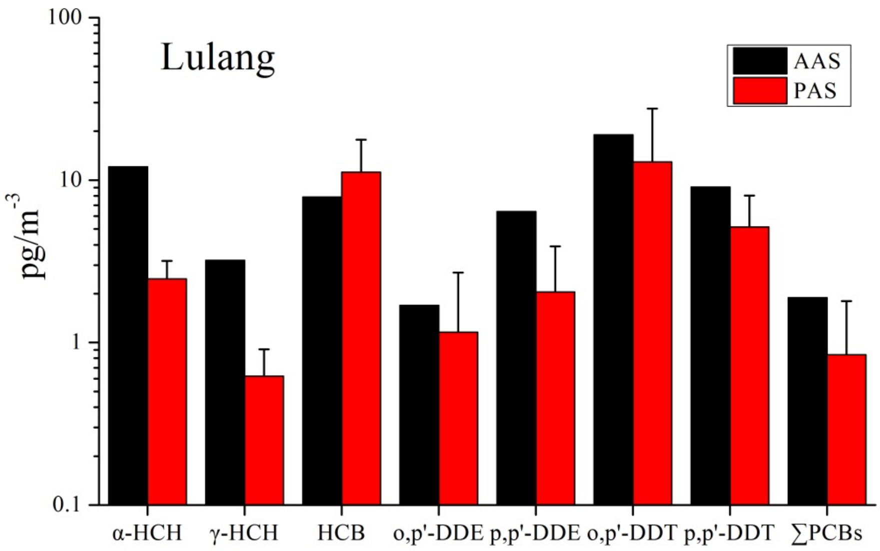
| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Naqu | 9.49 | 2.27 | 10.82 | 0.13 | 0.15 | 1.02 | 1.83 | 0.05 | 0.34 | 0.58 |
| Qamdo | 2.25 | 0.90 | 11.20 | 0.15 | 1.27 | 0.40 | 2.17 | 0.21 | 0.57 | 1.05 |
| Saga | 3.38 | 0.96 | 10.55 | 0.11 | 0.70 | 2.28 | 2.46 | 0.14 | 0.36 | 0.66 |
| Rawu | 1.81 | 0.37 | 6.93 | 0.16 | 0.19 | 0.77 | 1.73 | 0.07 | 0.14 | 0.42 |
| Bomi | 16.58 | 4.63 | 31.92 | 0.71 | 4.90 | 11.43 | 25.95 | 0.18 | 0.58 | 1.03 |
| GBJD | / | / | / | / | / | / | / | / | / | / |
| Golmud | 6.15 | 2.92 | 43.03 | 0.19 | 0.22 | 0.13 | 0.13 | 0.32 | 0.89 | 1.45 |
| Gar | 5.72 | 1.73 | 26.31 | 0.57 | 0.85 | 0.09 | 0.93 | 0.24 | 0.65 | 1.07 |
| Chayu | 5.35 | 1.35 | 25.81 | 0.24 | 1.75 | 9.57 | 24.91 | 0.10 | 0.48 | 0.88 |
| Muatagata | / | / | / | / | / | / | / | / | / | / |
| **2009 - 2010** | | | | | | | | | | |
| Xigaze | 1.95 | 0.61 | 9.46 | 3.04 | 4.52 | 8.63 | 3.70 | 0.07 | 0.02 | 0.30 |
| Nam co | 2.70 | 0.72 | 13.73 | 0.57 | 1.25 | 9.85 | 5.47 | 0.06 | 0.10 | 1.17 |
| Lhasa | 1.80 | 0.55 | 10.41 | 0.44 | 2.75 | 8.07 | 3.61 | 0.07 | 0.05 | 0.54 |
| Lhaze | 3.55 | 1.22 | 10.54 | 0.09 | 1.43 | 4.15 | 2.93 | 0.06 | 0.05 | 0.37 |
| Lulang | 3.40 | 0.82 | 9.33 | 0.38 | 2.27 | 12.88 | 6.38 | 0.08 | 0.07 | 0.45 |
| Everest | 3.70 | 1.10 | 12.80 | 0.53 | 1.76 | 9.98 | 4.35 | 0.06 | 0.08 | 0.42 |
| Naqu | 9.94 | 2.25 | 14.14 | 0.09 | 0.99 | 3.99 | 1.80 | 0.06 | 0.06 | 0.30 |
| Qamdo | 3.14 | 1.24 | 12.18 | 0.24 | 1.96 | 3.41 | 2.81 | 0.17 | 0.17 | 0.61 |
| Saga | 2.23 | 0.57 | 8.46 | 0.09 | 1.67 | 4.16 | 4.18 | 0.06 | 0.04 | 0.28 |
| Rawu | 3.16 | 0.79 | 12.75 | 0.18 | 1.09 | 5.84 | 2.98 | 0.06 | 0.05 | 0.24 |
| Bomi | 8.32 | 2.60 | 8.79 | 1.48 | 18.09 | 31.85 | 26.09 | 0.30 | 0.02 | 0.82 |
| GBJD | / | / | / | / | / | / | / | / | / | / |
| Golmud | 7.04 | 1.86 | 38.90 | 0.23 | 1.42 | 6.01 | 2.03 | 0.28 | 0.33 | 1.11 |
| Gar | 1.31 | 0.88 | 13.57 | 0.10 | 1.04 | 1.53 | 2.01 | 0.07 | 0.08 | 0.59 |
| Chayu | 7.87 | 2.15 | 31.41 | 1.68 | 8.75 | 37.33 | 20.20 | 0.21 | 0.16 | 0.83 |
| Muztaga | 2.33 | 0.47 | 20.72 | 0.13 | 1.27 | 1.29 | 1.65 | 0.09 | 0.03 | 0.39 |

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| **2010 - 2011** | | | | | | | | | | |
| Xigaze | 1.72 | 0.25 | 12.37 | 0.57 | 1.50 | 0.98 | 0.19 | 0.13 | 0.04 | 0.33 |
| Nam co | 1.10 | 0.17 | 8.30 | 0.10 | 0.19 | 0.81 | 0.61 | 0.34 | 0.04 | 0.52 |
| Lhasa | 0.80 | 0.12 | 5.79 | 0.26 | 1.09 | 1.27 | 0.65 | 0.13 | 0.04 | 0.34 |
| Lhaze | 1.27 | 0.18 | 10.23 | 0.10 | 0.21 | 0.37 | 0.19 | 0.12 | 0.04 | 0.32 |
| Lulang | 1.73 | 0.20 | 9.09 | 0.30 | 0.87 | 2.53 | 1.14 | 0.15 | 0.05 | 0.38 |
| Everest | 2.20 | 0.53 | 21.26 | 0.27 | 0.70 | 2.29 | 0.83 | 0.11 | 0.08 | 0.33 |
| Naqu | 6.08 | 1.08 | 12.91 | 0.10 | 0.20 | 0.71 | 0.18 | 0.12 | 0.04 | 0.31 |
| Qamdo | 1.63 | 0.26 | 10.85 | 0.12 | 0.49 | 0.53 | 0.22 | 0.14 | 0.05 | 0.37 |
| Saga | 3.10 | 0.49 | 14.75 | 0.09 | 0.61 | 0.86 | 0.96 | 0.11 | 0.04 | 0.28 |
| Rawu | 1.55 | 0.33 | 7.19 | 0.21 | 0.56 | 1.27 | 0.41 | 0.38 | 0.04 | 0.58 |
| Bomi | 5.21 | 0.79 | 6.87 | 0.35 | 5.62 | 2.67 | 1.52 | 0.16 | 0.05 | 0.42 |
| GBJD | 4.23 | 0.99 | 10.65 | 0.29 | 1.27 | 3.86 | 1.77 | 0.14 | 0.05 | 0.37 |
| Golmud | 1.97 | 0.26 | 30.61 | 0.15 | 0.31 | 0.51 | 0.28 | 0.18 | 0.11 | 0.52 |
| Gar | 0.70 | 0.11 | 9.33 | 0.10 | 0.21 | 0.16 | 0.19 | 0.12 | 0.04 | 0.32 |
| Chayu | 1.88 | 0.23 | 12.94 | 0.95 | 7.09 | 11.56 | 7.69 | 0.17 | 0.06 | 0.44 |
| Muztaga | 2.69 | 0.28 | 31.78 | 0.13 | 1.13 | 0.19 | 0.23 | 0.15 | 0.11 | 0.45 |
| **2011 - 2012** | | | | | | | | | | |
| Xigaze | 1.12 | 0.28 | 6.84 | 0.32 | 0.79 | 1.59 | 0.78 | 0.03 | 0.03 | 0.14 |
| Nam co | 1.21 | 0.31 | 9.31 | 0.08 | 0.20 | 1.83 | 0.69 | 0.03 | 0.03 | 0.14 |
| Lhasa | 0.62 | 0.14 | 3.74 | 0.32 | 1.43 | 2.67 | 1.69 | 0.03 | 0.02 | 0.24 |
| Lhaze | 1.10 | 0.20 | 5.52 | 0.10 | 0.35 | 1.44 | 0.99 | 0.03 | 0.02 | 0.14 |
| Lulang | 2.03 | 0.49 | 6.06 | 0.34 | 1.03 | 6.19 | 3.66 | 0.03 | 0.05 | 0.19 |
| Everest | 1.72 | 0.53 | 11.42 | 0.21 | 0.72 | 3.92 | 2.18 | 0.03 | 0.05 | 0.25 |
| Naqu | 6.15 | 1.38 | 13.98 | 0.12 | 0.37 | 1.64 | 1.50 | 0.06 | 0.06 | 0.21 |
| Qamdo | 0.96 | 0.29 | 4.74 | 0.14 | 0.74 | 1.12 | 0.92 | 0.03 | 0.03 | 0.27 |
| Saga | 1.05 | 0.32 | 5.50 | 0.08 | 0.25 | 0.83 | 0.45 | 0.03 | 0.03 | 0.12 |

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Rawu | 1.06 | 0.28 | 4.35 | 0.19 | 0.51 | 2.74 | 1.56 | 0.03 | 0.03 | 0.15 |
| Bomi | 1.46 | 0.36 | 3.57 | 0.52 | 3.82 | 7.08 | 4.90 | 0.04 | 0.01 | 0.50 |
| GBJD | 2.47 | 0.54 | 8.31 | 0.30 | 1.29 | 5.77 | 25.62 | 0.07 | 0.05 | 0.30 |
| Golmud | 2.58 | 0.96 | 24.90 | 0.07 | 0.64 | 0.54 | 0.73 | 0.18 | 0.10 | 0.66 |
| Gar | 0.79 | 0.20 | 6.58 | 0.04 | 0.18 | 0.42 | 0.34 | 0.03 | 0.03 | 0.14 |
| Chayu | 2.21 | 0.44 | 8.66 | 0.58 | 3.31 | 11.06 | 9.01 | 0.04 | 0.04 | 0.31 |
| Muztagata | 2.85 | 0.42 | 27.85 | 0.03 | 1.52 | 1.01 | 1.65 | 0.04 | 0.03 | 0.30 |

**Text S3 The comparison between AAS and PAS**

To test if the R used in this study is rational, a direct concentration comparison between PAS and AAS is imperative owing to the relatively accurate air volume of AAS. There are one site, Lulang, where two kinds of samplers were both deployed, thus provides the possibility for the comparison. Data of AAS were collected from the previous study (Sheng et al., 2013). The sampling period of AAS was from September 2008 to August 2011, which overlapped with PAS sampling. AAS data were then averaged and compared with data obtained from PAS. The results are presented below.




The DDTs derived from PAS had no much difference from the corresponding AAS concentrations; whereas the low molecular weight OCPs, such as α-HCH and γ-HCH, showed some discrepancy (Figure above). This may be caused by different air masses being sampled and different adsorption characteristics for two kinds of samplers. Under these restrictions, the concentration variability within a factor of 2 or 3 is deemed to be acceptable (Gouin et al., 2005). Therefore, the differences in the present PAS/AAS comparison (Figure above) were acceptable, which demonstrate that the R obtained in our study was reasonable.

**Table S5 Average air concentrations (ng/sample) for each sampling site during the 5 sampling years**

| Sampling site | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | PCB-28 | PCB-52 | ∑6 PCBs |
|---|---|---|---|---|---|---|---|---|---|---|
| **Xigaze** | 2.9 | 1.0 | 13.0 | 1.6 | 2.6 | 8.0 | 3.8 | 0.2 | 0.2 | 0.9 |
| **Nam co** | 1.9 | 0.7 | 12.7 | 0.7 | 0.5 | 3.7 | 1.8 | 0.2 | 0.2 | 0.7 |
| **Lhasa** | 2.4 | 1.0 | 21.0 | 0.7 | 2.3 | 7.7 | 2.9 | 0.2 | 0.2 | 0.7 |
| **Lhaze** | 2.4 | 0.7 | 13.6 | 0.3 | 0.7 | 3.6 | 1.9 | 0.2 | 0.2 | 0.6 |
| **Lulang** | 2.2 | 0.5 | 9.3 | 0.9 | 1.7 | 10.5 | 4.4 | 0.2 | 0.2 | 0.6 |
| **Everest** | 3.6 | 1.9 | 19.0 | 0.3 | 0.9 | 5.3 | 2.7 | 0.5 | 0.2 | 0.8 |
| **Naqu** | 8.3 | 1.9 | 16.6 | 0.4 | 0.5 | 4.7 | 2.0 | 0.1 | 0.2 | 0.6 |
| **Qamdo** | 1.7 | 0.7 | 10.4 | 1.5 | 3.0 | 7.9 | 3.0 | 0.3 | 0.2 | 0.9 |
| **Saga** | 3.3 | 0.9 | 13.4 | 0.7 | 1.9 | 7.7 | 3.7 | 0.2 | 0.2 | 0.6 |
| **Rawu** | 4.9 | 4.1 | 9.1 | 0.4 | 1.3 | 10.2 | 2.9 | 0.6 | 0.1 | 0.8 |
| **Bomi** | 6.5 | 2.1 | 20.6 | 0.9 | 6.1 | 11.0 | 10.3 | 0.3 | 0.2 | 1.0 |
| **GBJD** | 4.2 | 1.1 | 13.2 | 1.3 | 3.0 | 12.2 | 10.2 | 0.2 | 0.1 | 0.8 |
| **Golmud** | 3.4 | 1.2 | 30.0 | 0.4 | 0.7 | 1.5 | 0.6 | 0.3 | 0.3 | 0.8 |
| **Gar** | 2.6 | 3.2 | 13.5 | 0.4 | 0.9 | 2.3 | 1.1 | 0.3 | 0.2 | 0.7 |
| **Chayu** | 3.2 | 0.8 | 14.5 | 0.7 | 4.0 | 13.4 | 11.1 | 0.1 | 0.1 | 0.4 |
| **Muztagata** | 2.1 | 0.3 | 21.2 | 0.1 | 1.0 | 0.7 | 0.9 | 0.1 | 0.0 | 0.3 |

**Table S6 Average volumetric concentrations (pg/m$^3$) for each sampling site during the 5 sampling years**

| sampling site | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | PCB-28 | PCB-52 | ∑6 PCBs |
|---|---|---|---|---|---|---|---|---|---|---|
| Xigaze | 3.0 | 1.0 | 14.2 | 1.7 | 2.8 | 9.2 | 4.2 | 0.2 | 0.2 | 1.0 |
| Nam co | 1.7 | 0.6 | 11.4 | 0.8 | 0.4 | 3.4 | 1.6 | 0.1 | 0.2 | 0.7 |
| Lhasa | 2.5 | 1.0 | 23.4 | 0.8 | 2.6 | 8.5 | 3.1 | 0.2 | 0.2 | 0.8 |
| Lhaze | 2.3 | 0.7 | 13.8 | 0.3 | 0.7 | 3.8 | 1.9 | 0.2 | 0.2 | 0.6 |
| Lulang | 2.5 | 0.6 | 11.2 | 1.2 | 2.0 | 12.9 | 5.2 | 0.2 | 0.2 | 0.8 |
| Everest | 3.4 | 1.9 | 18.3 | 0.2 | 0.8 | 5.0 | 2.5 | 0.5 | 0.2 | 0.8 |
| Naqu | 8.3 | 1.9 | 17.7 | 0.5 | 0.6 | 5.5 | 2.2 | 0.1 | 0.2 | 0.6 |
| Qamdo | 1.9 | 0.8 | 12.0 | 1.9 | 3.7 | 10.0 | 3.7 | 0.4 | 0.2 | 1.1 |
| Saga | 3.0 | 0.8 | 12.1 | 0.7 | 1.8 | 7.6 | 3.5 | 0.2 | 0.2 | 0.5 |
| Rawu | 5.0 | 4.4 | 8.8 | 0.4 | 1.4 | 10.5 | 2.9 | 0.6 | 0.1 | 0.8 |
| Bomi | 8.1 | 2.7 | 27.2 | 1.2 | 7.7 | 13.8 | 12.5 | 0.4 | 0.3 | 1.3 |
| GBJD | 5.0 | 1.3 | 15.6 | 1.6 | 3.6 | 14.4 | 11.7 | 0.2 | 0.1 | 1.0 |
| Golmud | 4.5 | 1.5 | 42.5 | 0.6 | 0.9 | 1.9 | 0.8 | 0.4 | 0.4 | 1.1 |
| Gar | 2.5 | 3.5 | 13.0 | 0.4 | 0.9 | 2.5 | 1.2 | 0.4 | 0.2 | 0.9 |
| Chayu | 4.3 | 1.0 | 19.7 | 0.9 | 5.2 | 17.4 | 15.5 | 0.1 | 0.2 | 0.6 |
| Muztagata | 2.6 | 0.4 | 26.8 | 0.1 | 1.3 | 0.8 | 1.2 | 0.1 | 0.1 | 0.4 |

**Text S4 A comparison between concentrations of the current study and the previous Global Atmospheric Passive Sampling (GAPS) study**

To further identify the POPs contamination level on the global scale, a comparison was performed between the current study and the previous Global Atmospheric Passive Sampling (GAPS) study which also relied on the XAD-PAS (Shunthirasingham et al., 2010). The levels of HCB in the Tibetan atmosphere ranged from 2.5 to 60.7 ng/sample (Table S2), which is higher than the background areas of Asia (4.6-27.4 ng/sample) and South America (8.9-17.8 ng/sample), but similar to those in Europe (12.6-78.1 ng/sample) (Shunthirasingham et al., 2010). The level of α-HCH (0.1-16.4 ng/sample) and γ-HCH (0.1-18.7 ng/sample) is comparable to the remote stations in North America (Below detection limit-15.3 ng/sample) and Africa (1.4-18.7 ng/sample), respectively (Shunthirasingham et al., 2010). Regarding the PCBs, its value ranged from 0.1 to 2.8 ng/sample in this study, which is much lower than those of Arctic Region (1.7-14 ng/sample) (Shunthirasingham et al., 2010). These comparisons indicate that POPs concentrations in the atmosphere of TP are akin to those observed in the background sites of the world, indicating the remoteness of TP. However, it is worth noting that the DDTs levels of TP (0.1-38.9 ng/sample for o.p’-DDT and 0.9-22.7 ng/sample for p-p’-DDT, Table S2) were considerably higher than other remote regions (mostly not detected) (Shunthirasingham et al., 2010), which may suggest closer proximity of the TP to the DDT source regions.

**Table S7. *P* values of randomized block ANOVA test for the concentrations of DDTs, HCHs, HCB and PCBs in the different sampling sites. ($p < 0.05$ indicates that the concentrations significantly differ between two sites.)**

| **Sampling site** | | **DDTs** | **HCHs** | **HCB** | **PCBs** |
|---|---|---|---|---|---|
| Xigaze | Nam co | 0.306 | 0.621 | 0.764 | 0.560 |
| | Lhasa | 0.788 | 0.898 | 0.312 | 0.671 |
| | Lhaze | 0.326 | 0.787 | 0.970 | 0.499 |
| | Lulang | 0.777 | 0.798 | 0.744 | 0.735 |
| | Everest | 0.407 | 0.703 | 0.650 | 0.773 |
| | Naqu | 0.421 | 0.078 | 0.702 | 0.539 |
| | Qamdo | 0.916 | 0.719 | 0.808 | 0.801 |
| | Saga | 0.700 | 0.952 | 0.817 | 0.403 |
| | Rawu | 0.799 | 0.120 | 0.553 | 0.798 |
| | Bomi | 0.139 | 0.053 | 0.154 | 0.525 |
| | GBJD | 0.319 | 0.556 | 0.895 | 0.976 |
| | Golmud | 0.233 | 0.547 | ***0.003*** | 0.863 |
| | Gar | 0.255 | 0.558 | 0.900 | 0.666 |
| | Chayu | 0.090 | 0.706 | 0.566 | 0.530 |
| | Muztagata | 0.271 | 0.808 | 0.232 | 0.347 |
| Nam co | Lhasa | 0.449 | 0.714 | 0.191 | 0.874 |
| | Lhaze | 0.966 | 0.822 | 0.792 | 0.925 |
| | Lulang | 0.193 | 0.811 | 0.979 | 0.807 |
| | Everest | 0.844 | 0.383 | 0.452 | 0.768 |
| | Naqu | 0.826 | ***0.026*** | 0.495 | 0.974 |
| | Qamdo | 0.260 | 0.893 | 0.955 | 0.405 |
| | Saga | 0.521 | 0.664 | 0.946 | 0.798 |
| | Rawu | 0.441 | ***0.043*** | 0.769 | 0.743 |
| | Bomi | ***0.014*** | ***0.016*** | 0.086 | 0.225 |
| | GBJD | 0.062 | 0.311 | 0.695 | 0.635 |
| | Golmud | 0.863 | 0.275 | ***0.001*** | 0.451 |
| | Gar | 0.907 | 0.282 | 0.861 | 0.880 |
| | Chayu | ***0.009*** | 0.400 | 0.392 | 0.936 |
| | Muztagata | 0.829 | 0.852 | 0.147 | 0.660 |
| Lhasa | Lhaze | 0.475 | 0.887 | 0.294 | 0.801 |
| | Lulang | 0.581 | 0.898 | 0.183 | 0.931 |
| | Everest | 0.575 | 0.611 | 0.574 | 0.891 |
| | Naqu | 0.591 | 0.060 | 0.527 | 0.849 |
| | Qamdo | 0.708 | 0.816 | 0.211 | 0.499 |
| | Saga | 0.908 | 0.946 | 0.215 | 0.679 |
| | Rawu | 0.988 | 0.094 | 0.111 | 0.866 |
| | Bomi | 0.082 | ***0.039*** | 0.673 | 0.290 |
| | GBJD | 0.220 | 0.485 | 0.455 | 0.736 |
| | Golmud | 0.354 | 0.466 | ***0.039*** | 0.551 |
| | Gar | 0.383 | 0.476 | 0.256 | 0.994 |

| | | | | | |
|---|---|---|---|---|---|
| | Chayu | 0.052 | 0.619 | 0.702 | 0.819 |
| | Muztagata | 0.384 | 0.895 | 0.747 | 0.565 |
| Lhaze | Lulang | 0.208 | 0.989 | 0.772 | 0.735 |
| | Everest | 0.877 | 0.516 | 0.624 | 0.698 |
| | Naqu | 0.859 | ***0.044*** | 0.675 | 0.951 |
| | Qamdo | 0.278 | 0.928 | 0.836 | 0.355 |
| | Saga | 0.549 | 0.834 | 0.845 | 0.871 |
| | Rawu | 0.466 | 0.070 | 0.578 | 0.674 |
| | Bomi | ***0.016*** | ***0.028*** | 0.144 | 0.192 |
| | GBJD | 0.068 | 0.412 | 0.087 | 0.579 |
| | Golmud | 0.830 | 0.384 | ***0.002*** | 0.397 |
| | Gar | 0.873 | 0.394 | 0.929 | 0.807 |
| | Chayu | ***0.010*** | 0.529 | 0.543 | 0.993 |
| | Muztagata | 0.801 | 0.993 | 0.220 | 0.720 |
| Lulang | Everest | 0.268 | 0.525 | 0.436 | 0.960 |
| | Naqu | 0.278 | ***0.045*** | 0.479 | 0.782 |
| | Qamdo | 0.859 | 0.917 | 0.934 | 0.555 |
| | Saga | 0.505 | 0.845 | 0.925 | 0.617 |
| | Rawu | 0.591 | 0.072 | 0.789 | 0.934 |
| | Bomi | 0.228 | ***0.029*** | 0.081 | 0.331 |
| | GBJD | 0.451 | 0.419 | 0.679 | 0.793 |
| | Golmud | 0.142 | 0.392 | ***0.001*** | 0.610 |
| | Gar | 0.157 | 0.401 | 0.841 | 0.925 |
| | Chayu | 0.151 | 0.537 | 0.379 | 0.756 |
| | Muztagata | 0.180 | 0.983 | 0.141 | 0.515 |
| Everest | Naqu | 0.981 | 0.164 | 0.943 | 0.743 |
| | Qamdo | 0.351 | 0.460 | 0.486 | 0.590 |
| | Saga | 0.656 | 0.660 | 0.494 | 0.582 |
| | Rawu | 0.565 | 0.238 | 0.297 | 0.974 |
| | Bomi | ***0.023*** | 0.116 | 0.327 | 0.356 |
| | GBJD | 0.089 | 0.795 | 0.794 | 0.826 |
| | Golmud | 0.712 | 0.824 | ***0.009*** | 0.645 |
| | Gar | 0.754 | 0.838 | 0.563 | 0.885 |
| | Chayu | ***0.015*** | 0.986 | 0.883 | 0.720 |
| | Muztagata | 0.700 | 0.568 | 0.419 | 0.488 |
| Naqu | Qamdo | 0.363 | ***0.035*** | 0.532 | 0.387 |
| | Saga | 0.673 | 0.069 | 0.539 | 0.823 |
| | Rawu | 0.581 | 0.830 | 0.331 | 0.719 |
| | Bomi | ***0.024*** | 0.852 | 0.293 | 0.213 |
| | GBJD | 0.093 | 0.341 | 0.842 | 0.615 |
| | Golmud | 0.695 | 0.241 | ***0.008*** | 0.432 |
| | Gar | 0.736 | 0.234 | 0.611 | 0.854 |
| | Chayu | ***0.016*** | 0.195 | 0.831 | 0.961 |
| | Muztagata | 0.685 | 0.078 | 0.385 | 0.681 |
| Qamdo | Saga | 0.624 | 0.764 | 0.991 | 0.278 |
| | Rawu | 0.719 | 0.058 | 0.726 | 0.612 |

| | | | | | |
|---|---|---|---|---|---|
| | Bomi | 0.168 | ***0.023*** | 0.096 | 0.700 |
| | GBJD | 0.365 | 0.369 | 0.732 | 0.804 |
| | Golmud | 0.195 | 0.337 | ***0.001*** | 0.937 |
| | Gar | 0.214 | 0.346 | 0.906 | 0.495 |
| | Chayu | 0.110 | 0.475 | 0.422 | 0.387 |
| | Muztagata | 0.234 | 0.945 | 0.161 | 0.247 |
| Saga | Rawu | 0.897 | 0.107 | 0.718 | 0.560 |
| | Bomi | 0.064 | ***0.046*** | 0.099 | 0.143 |
| | GBJD | 0.185 | 0.522 | 0.739 | 0.487 |
| | Golmud | 0.416 | 0.508 | ***0.001*** | 0.314 |
| | Gar | 0.449 | 0.519 | 0.915 | 0.684 |
| | Chayu | ***0.041*** | 0.665 | 0.429 | 0.872 |
| | Muztagata | 0.441 | 0.849 | 0.164 | 0.827 |
| Rawu | Bomi | 0.084 | 0.689 | ***0.046*** | 0.373 |
| | GBJD | 0.225 | 0.443 | 0.519 | 0.848 |
| | Golmud | 0.346 | 0.336 | ***0.000*** | 0.669 |
| | Gar | 0.375 | 0.328 | 0.640 | 0.860 |
| | Chayu | 0.054 | 0.273 | 0.259 | 0.698 |
| | Muztagata | 0.378 | 0.113 | 0.090 | 0.470 |
| Bomi | GBJD | 0.769 | 0.267 | 0.268 | 0.561 |
| | Golmud | ***0.009*** | 0.175 | 0.096 | 0.643 |
| | Gar | ***0.010*** | 0.170 | 0.121 | 0.287 |
| | Chayu | 0.759 | 0.143 | 0.436 | 0.221 |
| | Muztagata | ***0.019*** | 0.055 | 0.966 | 0.138 |
| GBJD | Golmud | ***0.045*** | 0.947 | ***0.012*** | 0.857 |
| | Gar | ***0.050*** | 0.935 | 0.810 | 0.731 |
| | Chayu | 0.582 | 0.816 | 0.705 | 0.600 |
| | Muztagata | 0.063 | 0.458 | 0.340 | 0.415 |
| Golmud | Gar | 0.955 | 0.986 | ***0.002*** | 0.546 |
| | Chayu | ***0.006*** | 0.848 | ***0.021*** | 0.429 |
| | Muztagata | 0.947 | 0.445 | 0.137 | 0.276 |
| Gar | Chayu | ***0.007*** | 0.861 | 0.489 | 0.824 |
| | Muztagata | 0.909 | 0.454 | 0.193 | 0.569 |
| Chayu | Muztagata | ***0.013*** | 0.574 | 0.519 | 0.726 |

## Text S5 Calculation equations for fugacity, fugacity ratio and fugacity fluxes

Air-soil exchange directions and fluxes are very important for understanding the global cycling of POPs, which will help to understand whether the chemical can be evaporated from soil and how much the exchange fluxes will be.

Air fugacity of the chemicals can be represented as:

$f_a = C_a RT$ (1).

Soil fugacity of the chemicals can be presented as:

$f_s = C_s \rho_s RT/0.41\varphi_{om}K_{oa}$ (2)

where $C_s$ is the soil concentration (ng/g), $\rho_s$ is the density of soil solids (g/m3), $\varphi_{om}$ is the fraction of organic matter in the soil on a dry soil basis, $K_{oa}$ is the octanol-air partition coefficient of the compound, R is the gas constant (8.31 pa $m^3$/mol/K) and T is the average air temperature (K).

Fugacity fraction(*ff*) could be calculated by $ff = f_s/(f_s + f_a)$ (3)

Ideally, the *ff* = 0.5 indicates that soil fugacity and air fugacity is same and the exchange reach equilibrium. However, assuming an error of approximately 20% in $K_{OA}$ and 35% in analysis results in a propagated error of approximately 40% in the *ff*, a fugacity fraction in the range 0.5± 0.2 may not represent a significant departure from equilibrium. Therefore, the criterion range representing the equilibrium can vary from 0.3 to 0.7 (Harner et al., 2001 and Li et al., 2009).

Flux quantification (*F*) can provide detailed data on how much POP has been volatilizing from air or depositing into soil.

$F = (f_a - f_s)/(RT/K_{13} + L_3 * (RT/B_1 + H/B_2))$, （4）

where $K_{13}$, $L_3$, $B_1$ and $B_2$ are air-soil mass transfer coefficient (m/h), diffusion path length in soil (m), molecular diffusivities in air and water ($m^2$/h), respectively ($K_{13}$= 3.75m/h, $L_3$= 0.1m, $B_1$=0.0179 $m^2$/h, $B_2$=1.79×$10^{-6}$ $m^2$/h, these values were cited from McLachlan et al.( 2002). H is Henry constant (Pa m3/mol).

**Table S8 The soil-air equilibrium status (*ff*) of Tibetan Plateau sites.**

| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **2007-2008** | | | | | | | | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Namco** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | 0.66 | 0.58 | 0.59 | 0.70 | 0.86 | 0.74 | 0.77 | 0.91 | 0.60 | 0.81 | 0.79 | 0.65 | 0.94 |
| **PCB-28** | 0.58 | 0.52 | 0.68 | 0.06 | 0.82 | 0.99 | 0.90 | 1.00 | 0.97 | 0.85 | 0.98 | 0.96 | 0.97 |
| **PCB-52** | 0.94 | 0.75 | 0.95 | 0.18 | 0.89 | 0.99 | 0.44 | 0.93 | - | - | 0.59 | 0.38 | - |
| **α-HCH** | 0.29 | 0.87 | 0.81 | 0.31 | 0.97 | 0.88 | 0.98 | 0.99 | 0.64 | 0.86 | 0.59 | 0.58 | 0.90 |
| **γ-HCH** | 0.94 | 1.00 | 0.96 | 0.58 | 0.95 | 1.00 | 0.97 | 1.00 | 0.99 | 0.98 | 0.98 | 0.97 | 0.95 |
| **p,p'-DDE** | 0.003 | 0.05 | 0.07 | 0.003 | 0.01 | 0.14 | 0.11 | 0.17 | 0.01 | 0.21 | 0.07 | 0.16 | 0.30 |
| **o,p'-DDT** | 0.004 | 0.001 | 0.004 | 0.000 | 0.08 | 0.000 | - | 0.002 | - | - | - | - | - |
| **p,p'-DDT** | 0.15 | 0.07 | 0.31 | 0.14 | 0.55 | 0.41 | 0.17 | 0.49 | - | - | 0.04 | 0.05 | - |
| **2008-2009** | | | | | | | | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Namco** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | - | 0.78 | 0.79 | 0.81 | 0.92 | 0.88 | 0.30 | 0.94 | 0.80 | 0.81 | 0.88 | 0.86 | 0.85 |
| **PCB-28** | - | 0.92 | 0.94 | 0.69 | 0.97 | 1.00 | 0.86 | 1.00 | 1.00 | 0.99 | 0.99 | 0.99 | 1.00 |
| **PCB-52** | - | 0.82 | 0.95 | 0.22 | 0.80 | 0.99 | 0.09 | 0.79 | - | - | 0.55 | 0.38 | - |
| **α-HCH** | - | 0.82 | 0.70 | 0.82 | 0.95 | 0.88 | 0.70 | 0.98 | 0.53 | 0.86 | 0.69 | 0.59 | 0.86 |
| **γ-HCH** | - | 0.99 | 0.96 | 0.99 | 0.97 | 0.99 | 0.96 | 1.00 | 0.98 | 0.99 | 0.99 | 0.97 | 0.99 |
| **p,p'-DDE** | - | 0.26 | 0.08 | 0.06 | 0.14 | 0.56 | 0.08 | 0.35 | 0.05 | 0.14 | 0.39 | 0.58 | 0.51 |
| **o,p'-DDT** | - | 0.01 | 0.01 | 0.01 | 0.91 | 0.003 | - | 0.01 | - | - | - | - | - |
| **p,p'-DDT** | - | 0.11 | 0.07 | 0.43 | 0.87 | 0.70 | 0.12 | 0.56 | - | - | 0.14 | 0.15 | - |
| **2009-2010** | | | | | | | | | | | | | |
| **HCB** | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Namco** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **PCB-28** | - | 0.77 | 0.93 | 0.70 | 0.92 | 0.95 | 0.42 | 0.96 | 0.83 | 0.88 | 0.90 | 0.83 | 0.92 |
| **PCB-52** | - | 0.91 | 0.90 | 0.72 | 0.97 | 1.00 | 0.95 | 1.00 | 1.00 | 1.00 | 1.00 | 0.99 | 1.00 |
| **α-HCH** | - | 0.96 | 1.00 | 0.45 | 0.93 | 1.00 | 0.35 | 0.98 | - | - | 0.91 | 0.79 | - |

| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **γ-HCH** | - | 0.80 | 0.82 | 0.72 | 0.93 | 0.95 | 0.73 | 0.98 | 0.78 | 0.88 | 0.77 | 0.58 | 0.97 |
| **p,p'-DDE** | - | 0.99 | 0.98 | 0.97 | 0.95 | 1.00 | 0.97 | 1.00 | 1.00 | 1.00 | 0.99 | 0.97 | 1.00 |
| **o,p'-DDT** | - | 0.11 | 0.02 | 0.01 | 0.09 | 0.28 | 0.02 | 0.15 | 0.02 | 0.04 | 0.21 | 0.17 | 0.46 |
| **p,p'-DDT** | - | 0.002 | 0.002 | 0.001 | 0.54 | 0.001 | - | 0.01 | - | - | - | - | - |
| | - | 0.10 | 0.07 | 0.30 | 0.84 | 0.58 | 0.02 | 0.49 | - | - | 0.09 | 0.15 | - |
| **2010-2011** | | | | | | | | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Namco** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | 0.83 | 0.78 | 0.95 | 0.80 | 0.92 | 0.97 | 0.54 | 0.96 | 0.79 | 0.82 | 0.84 | 0.84 | 0.94 |
| **PCB-28** | 0.82 | 0.84 | 0.94 | 0.28 | 0.98 | 1.00 | 0.77 | 1.00 | 0.99 | 0.99 | 0.99 | 0.99 | 1.00 |
| **PCB-52** | 0.99 | 0.97 | 1.00 | 0.49 | 0.98 | 1.00 | 0.59 | 0.98 | - | - | 0.93 | 0.84 | - |
| **α-HCH** | 0.44 | 0.89 | 0.88 | 0.84 | 0.96 | 0.98 | 0.87 | 0.99 | 0.80 | 0.93 | 0.71 | 0.69 | 0.98 |
| **γ-HCH** | 0.98 | 1.00 | 0.99 | 0.99 | 0.99 | 1.00 | 0.99 | 1.00 | 1.00 | 1.00 | 0.99 | 0.99 | 1.00 |
| **p,p'-DDE** | 0.02 | 0.25 | 0.07 | 0.02 | 0.29 | 0.50 | 0.12 | 0.54 | 0.04 | 0.11 | 0.43 | 0.50 | 0.81 |
| **o,p'-DDT** | 0.04 | 0.01 | 0.02 | 0.01 | 0.88 | 0.01 | - | 0.07 | - | - | - | - | - |
| **p,p'-DDT** | 0.44 | 0.38 | 0.55 | 0.76 | 0.99 | 0.89 | 0.17 | 0.94 | - | - | 0.30 | 0.63 | - |
| **2011-2012** | | | | | | | | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Namco** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | 0.87 | 0.84 | 0.97 | 0.87 | 0.97 | 0.98 | 0.51 | 0.98 | 0.87 | 0.89 | 0.93 | 0.83 | 0.96 |
| **PCB-28** | 0.91 | 0.96 | 0.99 | 0.84 | 0.99 | 1.00 | 0.98 | 1.00 | 1.00 | 1.00 | 1.00 | 0.99 | 1.00 |
| **PCB-52** | 0.99 | 0.97 | 1.00 | 0.57 | 0.99 | 1.00 | 0.67 | 0.99 | - | - | 0.95 | 0.78 | - |
| **α-HCH** | 0.57 | 0.87 | 0.96 | 0.88 | 0.98 | 0.98 | 0.86 | 0.99 | 0.86 | 0.94 | 0.88 | 0.69 | 0.98 |
| **γ-HCH** | 0.99 | 1.00 | 1.00 | 0.99 | 0.99 | 1.00 | 0.99 | 1.00 | 1.00 | 1.00 | 1.00 | 0.98 | 1.00 |
| **p,p'-DDE** | 0.02 | 0.22 | 0.10 | 0.02 | 0.21 | 0.43 | 0.11 | 0.41 | 0.08 | 0.10 | 0.64 | 0.35 | 0.83 |
| **o,p'-DDT** | 0.02 | 0.004 | 0.01 | 0.002 | 0.78 | 0.004 | - | 0.02 | - | - | - | - | - |
| **p,p'-DDT** | 0.05 | 0.16 | 0.28 | 0.45 | 0.94 | 0.75 | 0.15 | 0.74 | - | - | 0.48 | 0.17 | - |

**Table S9. The soil-air net volatilization fluxes (ng/m$^2$/h) of PCB-28, PCB-52, HCB, α-HCH and γ-HCH at the Tibetan Plateau sites. (air data from 2007-2008 PAS)**

| 2007-2008 | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Nam co** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | -0.098 | -0.035 | -0.142 | -0.063 | -0.419 | -0.495 | -0.026 | -0.840 | -0.055 | -0.271 | -0.216 | -0.118 | -0.538 |
| **PCB-28** | -0.001 | 0.000 | -0.005 | 0.009 | -0.018 | -0.352 | -0.004 | -0.390 | -0.089 | -0.039 | -0.077 | -0.032 | -0.176 |
| **PCB-52** | -0.015 | -0.004 | -0.042 | 0.001 | -0.008 | -0.123 | 0.000 | -0.008 | - | - | -0.001 | 0.000 | - |
| **α-HCH** | 0.018 | -0.044 | -0.110 | 0.036 | -0.163 | -0.111 | -0.027 | -0.612 | -0.012 | -0.089 | -0.009 | -0.014 | -0.120 |
| **γ-HCH** | -0.147 | -0.501 | -0.400 | -0.031 | -0.092 | -1.557 | -0.097 | -2.799 | -0.457 | -1.103 | -0.330 | -0.263 | -1.042 |
| **2008-2009** | | | | | | | | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Nam co** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | - | -0.086 | -0.342 | -0.085 | -0.455 | -0.655 | 0.049 | -0.875 | -0.129 | -0.271 | -0.255 | -0.214 | -0.474 |
| **PCB-28** | - | -0.003 | -0.009 | 0.000 | -0.022 | -0.353 | -0.003 | -0.392 | -0.091 | -0.047 | -0.078 | -0.033 | -0.180 |
| **PCB-52** | - | -0.005 | -0.042 | 0.000 | -0.007 | -0.122 | 0.002 | -0.006 | - | - | 0.000 | 0.000 | - |
| **α-HCH** | - | -0.041 | -0.081 | -0.023 | -0.160 | -0.112 | -0.016 | -0.604 | -0.003 | -0.088 | -0.015 | -0.015 | -0.114 |
| **γ-HCH** | - | -0.500 | -0.401 | -0.105 | -0.094 | -1.553 | -0.096 | -2.799 | -0.453 | -1.118 | -0.333 | -0.264 | -1.090 |
| **2009-2010** | | | | | | | | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Nam co** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | - | -0.084 | -0.428 | -0.063 | -0.452 | -0.718 | 0.014 | -0.898 | -0.135 | -0.309 | -0.263 | -0.202 | -0.522 |
| **PCB-28** | - | -0.003 | -0.009 | 0.000 | -0.023 | -0.354 | -0.004 | -0.392 | -0.091 | -0.047 | -0.078 | -0.033 | -0.181 |
| **PCB-52** | - | -0.006 | -0.044 | 0.000 | -0.008 | -0.123 | 0.000 | -0.009 | - | - | -0.002 | -0.001 | - |
| **α-HCH** | - | -0.039 | -0.112 | -0.018 | -0.157 | -0.122 | -0.017 | -0.605 | -0.019 | -0.092 | -0.020 | -0.013 | -0.131 |
| **γ-HCH** | - | -0.500 | -0.409 | -0.103 | -0.093 | -1.561 | -0.097 | -2.798 | -0.460 | -1.120 | -0.334 | -0.264 | -1.093 |
| **2010-2011** | | | | | | | | | | | | | |

| | GBJD | Lulang | Bomi | Rawu | Qamdo | Lhasa | Nam co | Lhaze | Xigaze | Everest | Saga | Naqu | Gar |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **HCB** | -0.162 | -0.085 | -0.435 | -0.084 | -0.457 | -0.735 | -0.006 | -0.900 | -0.124 | -0.277 | -0.239 | -0.206 | -0.538 |
| **PCB-28** | -0.002 | -0.002 | -0.009 | 0.001 | -0.023 | -0.354 | -0.003 | -0.392 | -0.091 | -0.047 | -0.078 | -0.033 | -0.180 |
| **PCB-52** | -0.015 | -0.006 | -0.044 | 0.000 | -0.008 | -0.123 | 0.000 | -0.009 | - | - | -0.002 | -0.001 | - |
| **α-HCH** | 0.003 | -0.046 | -0.124 | -0.024 | -0.162 | -0.126 | -0.023 | -0.614 | -0.020 | -0.098 | -0.016 | -0.028 | -0.133 |
| **γ-HCH** | -0.153 | -0.503 | -0.416 | -0.105 | -0.097 | -1.562 | -0.099 | -2.802 | -0.461 | -1.122 | -0.334 | -0.269 | -1.096 |
| | | | | | | | **2011-2012** | | | | | | |
| | **GBJD** | **Lulang** | **Bomi** | **Rawu** | **Qamdo** | **Lhasa** | **Nam co** | **Lhaze** | **Xigaze** | **Everest** | **Saga** | **Naqu** | **Gar** |
| **HCB** | -0.171 | -0.097 | -0.448 | -0.094 | -0.479 | -0.743 | -0.002 | -0.917 | -0.145 | -0.314 | -0.274 | -0.202 | -0.548 |
| **PCB-28** | -0.002 | -0.003 | -0.010 | 0.000 | -0.023 | -0.354 | -0.004 | -0.392 | -0.092 | -0.048 | -0.078 | -0.033 | -0.181 |
| **PCB-52** | -0.015 | -0.006 | -0.044 | 0.000 | -0.009 | -0.123 | 0.000 | -0.009 | - | - | -0.002 | -0.001 | - |
| **α-HCH** | -0.003 | -0.045 | -0.138 | -0.026 | -0.165 | -0.127 | -0.023 | -0.615 | -0.022 | -0.100 | -0.024 | -0.028 | -0.133 |
| **γ-HCH** | -0.155 | -0.502 | -0.417 | -0.105 | -0.097 | -1.562 | -0.099 | -2.802 | -0.461 | -1.122 | -0.335 | -0.267 | -1.095 |

**Table SI-10. Relative composition data (%) used for cluster analysis**

| Samples | α-HCH | γ-HCH | HCB | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT | $\Sigma_6$PCBs |
|---|---|---|---|---|---|---|---|---|
| Xigaze 1 | 4.1 | 1.6 | 32.3 | 4.1 | 6.4 | 34.0 | 14.0 | 3.7 |
| Namco 1 | 1.0 | 6.3 | 26.2 | 24.1 | 1.9 | 30.4 | 5.2 | 4.7 |
| Lhasa 1 | 4.1 | 1.4 | 56.4 | 2.2 | 5.7 | 22.4 | 6.2 | 1.5 |
| Lhaze 1 | 4.1 | 2.0 | 56.1 | 2.5 | 2.5 | 23.9 | 6.4 | 2.4 |
| Lulang 1 | 2.6 | 0.8 | 27.0 | 4.6 | 6.2 | 45.6 | 10.4 | 2.8 |
| Everest 1 | 11.8 | 14.1 | 57.6 | 0.3 | 0.8 | 8.1 | 1.3 | 6.0 |
| Naqu 1 | 12.2 | 3.2 | 45.8 | 2.8 | 1.3 | 25.6 | 6.8 | 2.3 |
| Qamdo 1 | 1.5 | 1.3 | 19.5 | 8.1 | 13.3 | 41.7 | 11.4 | 3.1 |
| Saga 1 | 6.6 | 2.2 | 27.2 | 4.3 | 7.7 | 38.4 | 12.1 | 1.6 |
| Rawu 1 | 16.3 | 18.5 | 11.7 | 1.1 | 4.1 | 38.6 | 7.1 | 2.6 |
| Bomi 1 | 6.9 | 3.8 | 64.4 | 2.1 | 4.5 | 12.2 | 3.2 | 3.0 |
| GBJD 1 | 8.8 | 2.7 | 29.3 | 4.3 | 8.6 | 35.7 | 8.3 | 2.4 |
| Golmud 1 | 5.4 | 1.9 | 82.3 | 2.8 | 2.2 | 2.6 | 0.9 | 1.8 |
| Gar 1 | 9.1 | 31.9 | 20.6 | 2.2 | 4.7 | 22.8 | 5.2 | 3.5 |
| Chayu 2 | 7.7 | 1.9 | 36.9 | 0.3 | 2.5 | 13.7 | 35.7 | 1.3 |
| Bomi 2 | 17.1 | 4.8 | 32.9 | 0.7 | 5.0 | 11.8 | 26.7 | 1.1 |
| Xigaze 2 | 22.0 | 8.6 | 40.1 | 3.2 | 4.4 | 8.5 | 10.9 | 2.3 |
| Namco 2 | 10.2 | 3.7 | 74.6 | 0.9 | 1.0 | 3.8 | 2.8 | 3.1 |
| Lhasa 2 | 10.5 | 6.2 | 63.5 | 0.3 | 2.0 | 10.3 | 5.2 | 2.0 |
| Lhaze 2 | 14.2 | 3.5 | 60.7 | 0.5 | 1.7 | 7.4 | 8.1 | 4.0 |
| Lulang 2 | 11.7 | 3.5 | 34.2 | 3.4 | 3.2 | 18.8 | 22.6 | 2.5 |
| Everest 2 | 11.6 | 4.0 | 56.3 | 0.3 | 1.3 | 13.6 | 11.0 | 1.8 |
| Naqu 2 | 36.1 | 8.7 | 41.2 | 0.5 | 0.6 | 3.9 | 7.0 | 2.2 |
| Qamdo 2 | 11.6 | 4.6 | 57.7 | 0.8 | 6.6 | 2.1 | 11.2 | 5.4 |
| Saga 2 | 16.0 | 4.5 | 50.0 | 0.5 | 3.3 | 10.8 | 11.7 | 3.1 |
| Rawu 2 | 14.6 | 3.0 | 56.0 | 1.3 | 1.5 | 6.2 | 14.0 | 3.4 |
| Golmud 2 | 11.3 | 5.4 | 79.4 | 0.4 | 0.4 | 0.2 | 0.2 | 2.7 |
| Gar 2 | 15.3 | 4.6 | 70.6 | 1.5 | 2.3 | 0.2 | 2.5 | 2.9 |
| Xigaze 3 | 6.1 | 1.9 | 29.4 | 9.4 | 14.0 | 26.8 | 11.5 | 0.9 |
| Namco 3 | 7.6 | 2.0 | 38.7 | 1.6 | 3.5 | 27.8 | 15.4 | 3.3 |
| Lhasa 3 | 6.4 | 2.0 | 37.0 | 1.6 | 9.8 | 28.6 | 12.8 | 1.9 |
| Lhaze 3 | 14.6 | 5.0 | 43.4 | 0.4 | 5.9 | 17.1 | 12.1 | 1.5 |
| Lulang 3 | 9.5 | 2.3 | 26.0 | 1.1 | 6.3 | 35.8 | 17.8 | 1.3 |
| Everest 3 | 10.7 | 3.2 | 37.0 | 1.5 | 5.1 | 28.8 | 12.5 | 1.2 |
| Naqu 3 | 29.7 | 6.7 | 42.2 | 0.3 | 2.9 | 11.9 | 5.4 | 0.9 |
| Qamdo 3 | 12.3 | 4.8 | 47.6 | 0.9 | 7.6 | 13.3 | 11.0 | 2.4 |
| Saga 3 | 10.3 | 2.7 | 39.1 | 0.4 | 7.7 | 19.2 | 19.3 | 1.3 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Rawu 3 | 11.7 | 2.9 | 47.2 | 0.7 | 4.0 | 21.6 | 11.0 | 0.9 |
| Bomi 3 | 8.5 | 2.6 | 9.0 | 1.5 | 18.5 | 32.5 | 26.6 | 0.8 |
| Golmud 3 | 12.0 | 3.2 | 66.4 | 0.4 | 2.4 | 10.3 | 3.5 | 1.9 |
| Gar 3 | 6.2 | 4.2 | 64.5 | 0.5 | 4.9 | 7.3 | 9.6 | 2.8 |
| Chayu 3 | 7.1 | 2.0 | 28.5 | 1.5 | 7.9 | 33.9 | 18.3 | 0.8 |
| Muztaga 3 | 8.2 | 1.7 | 73.3 | 0.5 | 4.5 | 4.5 | 5.9 | 1.4 |
| | | | | | | | | |
| Xigaze 4 | 9.6 | 1.4 | 69.0 | 3.2 | 8.4 | 5.5 | 1.1 | 1.8 |
| Namco 4 | 9.3 | 1.4 | 70.3 | 0.8 | 1.6 | 6.8 | 5.2 | 4.4 |
| Lhasa 4 | 7.8 | 1.2 | 56.1 | 2.6 | 10.5 | 12.3 | 6.3 | 3.3 |
| Lhaze 4 | 9.9 | 1.4 | 79.5 | 0.8 | 1.6 | 2.9 | 1.4 | 2.5 |
| Lulang 4 | 10.7 | 1.2 | 55.9 | 1.8 | 5.4 | 15.6 | 7.0 | 2.3 |
| Everest 4 | 7.7 | 1.9 | 74.8 | 0.9 | 2.5 | 8.0 | 2.9 | 1.2 |
| Naqu 4 | 28.2 | 5.0 | 59.8 | 0.5 | 0.9 | 3.3 | 0.8 | 1.4 |
| Qamdo 4 | 11.3 | 1.8 | 74.9 | 0.8 | 3.4 | 3.7 | 1.5 | 2.6 |
| Saga 4 | 14.7 | 2.3 | 69.8 | 0.4 | 2.9 | 4.1 | 4.5 | 1.3 |
| Rawu 4 | 12.8 | 2.8 | 59.4 | 1.8 | 4.6 | 10.5 | 3.4 | 4.8 |
| Bomi 4 | 22.2 | 3.4 | 29.3 | 1.5 | 24.0 | 11.4 | 6.5 | 1.8 |
| GBJD 4 | 18.0 | 4.2 | 45.4 | 1.3 | 5.4 | 16.5 | 7.5 | 1.6 |
| Golmud 4 | 5.7 | 0.8 | 88.4 | 0.4 | 0.9 | 1.5 | 0.8 | 1.5 |
| Gar 4 | 6.3 | 1.0 | 83.9 | 0.9 | 1.9 | 1.4 | 1.7 | 2.9 |
| Chayu 4 | 4.4 | 0.5 | 30.2 | 2.2 | 16.6 | 27.0 | 18.0 | 1.0 |
| Muztaga 4 | 7.3 | 0.7 | 86.2 | 0.3 | 3.1 | 0.5 | 0.6 | 1.2 |
| | | | | | | | | |
| Xigaze 5 | 9.5 | 2.3 | 57.6 | 2.7 | 6.7 | 13.4 | 6.6 | 1.2 |
| Namco 5 | 8.8 | 2.3 | 67.5 | 0.6 | 1.5 | 13.3 | 5.0 | 1.0 |
| Lhasa 5 | 5.7 | 1.3 | 34.5 | 2.9 | 13.2 | 24.6 | 15.6 | 2.2 |
| Lhaze 5 | 11.2 | 2.0 | 56.1 | 1.0 | 3.6 | 14.7 | 10.0 | 1.5 |
| Lulang 5 | 10.2 | 2.4 | 30.3 | 1.7 | 5.2 | 31.0 | 18.3 | 0.9 |
| Everest 5 | 8.2 | 2.6 | 54.5 | 1.0 | 3.4 | 18.7 | 10.4 | 1.2 |
| Naqu 5 | 24.3 | 5.4 | 55.1 | 0.5 | 1.5 | 6.5 | 5.9 | 0.8 |
| Qamdo 5 | 10.5 | 3.2 | 51.6 | 1.5 | 8.1 | 12.2 | 10.0 | 2.9 |
| Saga 5 | 12.2 | 3.7 | 64.1 | 0.9 | 2.9 | 9.6 | 5.3 | 1.4 |
| Rawu 5 | 9.8 | 2.6 | 40.1 | 1.7 | 4.7 | 25.3 | 14.4 | 1.4 |
| Bomi 5 | 6.6 | 1.6 | 16.1 | 2.3 | 17.2 | 31.9 | 22.1 | 2.3 |
| GBJD 5 | 5.5 | 1.2 | 18.6 | 0.7 | 2.9 | 12.9 | 57.4 | 0.7 |
| Golmud 5 | 8.3 | 3.1 | 80.1 | 0.2 | 2.1 | 1.7 | 2.4 | 2.1 |
| Gar 5 | 9.1 | 2.3 | 75.7 | 0.4 | 2.1 | 4.9 | 3.9 | 1.6 |
| Chayu 5 | 6.2 | 1.2 | 24.3 | 1.6 | 9.3 | 31.1 | 25.3 | 0.9 |
| Muztaga 5 | 8.0 | 1.2 | 78.2 | 0.1 | 4.3 | 2.8 | 4.6 | 0.9 |

The naming convention of the samples is the site name plus the sampling year. For example, “Xigaze 1” is the sample for site of Xigaze in the first sampling year of 2007 to 2008.

**Table S11. Results of the grouping using cluster analysis**

| 3 groups | Group 1 | Group 2 | Group 3 |
|---|---|---|---|
| Number of sites | 28 | 17 | 30 |
| Sample name | Xigaze 1 | Golmud 1 | Lhasa 1 |
| | Namco 1 | Namco 2 | Lhaze 1 |
| | Lulang 1 | Golmud 2 | Everest 1 |
| | Naqu 1 | Gar 2 | Bomi 1 |
| | Qamdo 1 | Muztaga 3 | Xigaze 2 |
| | Saga 1 | Xigaze 4 | Lhasa 2 |
| | Rawu 1 | Namco 4 | Lhaze 2 |
| | GBJD 1 | Lhaze 4 | Everest 2 |
| | Gar 1 | Everest 4 | Naqu 2 |
| | Lulang 2 | Qamdo 4 | Qamdo 2 |
| | Chayu 2 | Saga 4 | Saga 2 |
| | Bomi 2 | Muztaga 4 | Rawu 2 |
| | Xigaze 3 | Golmud 4 | Lhaze 3 |
| | Namco 3 | Gar 4 | Naqu 3 |
| | Lhasa 3 | Golmud 5 | Qamdo 3 |
| | Lulang 3 | Gar 5 | Golmud 3 |
| | Everest 3 | Muztaga 5 | Gar 3 |
| | Saga 3 | | Lhasa 4 |
| | Chayu 3 | | Lulang 4 |
| | Rawu 3 | | Naqu 4 |
| | Bomi 3 | | Rawu 4 |
| | Chayu 4 | | Bomi 4 |
| | Lhasa 5 | | GBJD 4 |
| | Lulang 5 | | Xigaze 5 |
| | Chayu 5 | | Namco 5 |
| | Rawu 5 | | Lhaze 5 |
| | Bomi 5 | | Everest 5 |
| | GBJD 5 | | Naqu 5 |
| | | | Qamdo 5 |
| | | | Saga 5 |

| **composition (%)** | **Group 1** | **Group 2** | **Group 3** |
|---|---|---|---|
| **α-HCH** | 8.0 (1.0-17.1) | 9.3 (5.4-15.3) | 14.1 (4.1-36.1) |
| **γ-HCH** | 4.0 (0.5-31.9) | 2.2 (0.7-5.4) | 4.3 (1.2-14.1) |
| **HCB** | 29.6 (9.0-47.2) | 77.1 (69.0-88.4) | 54.6 (29.3-67.5) |
| **DDTs** | 56.5 (34.9-79.1) | 9.3 (1.2-18.1) | 24.7 (5.6-43.3) |
| **$\Sigma_6$ PCBs** | 1.9 (0.7-4.7) | 2.1 (0.9-4.4) | 2.4 (0.8-6.0) |

The average and range (in brackets) of composition in each group were listed.

**Table S12. The representative sites in each group**

| Group 1 | Group 2 | Group 3 |
|---|---|---|
| Chayu (100%)[a] | Muztaga (100%) [b] | Lhaze (80%) |
| Lulang (80%) | Golmud (80%) | *Naqu (80%)*[c] |
| GBJD (67%) | Gar (60%) | Everest (60%) |
| Rawu (60%) | | Qamdo (60%) |
| Bomi (60%) | | Lhasa (60%) |

a. The percentages in the brackets after the site name represent the frequency of the sampling site appears in this group, which is derived through dividing the times of the sampling site appears in this group to the total sampling times in this site.

b. Most sites have 5 samples of the consecutive 5 years, except that Chayu sampled for 4 years; GBJD sampled for 3 years and Muztaga for 3 years. (see Table S1)

c. Due to the possible local contamination of Naqu, Naqu was not included as a representative site of this group.

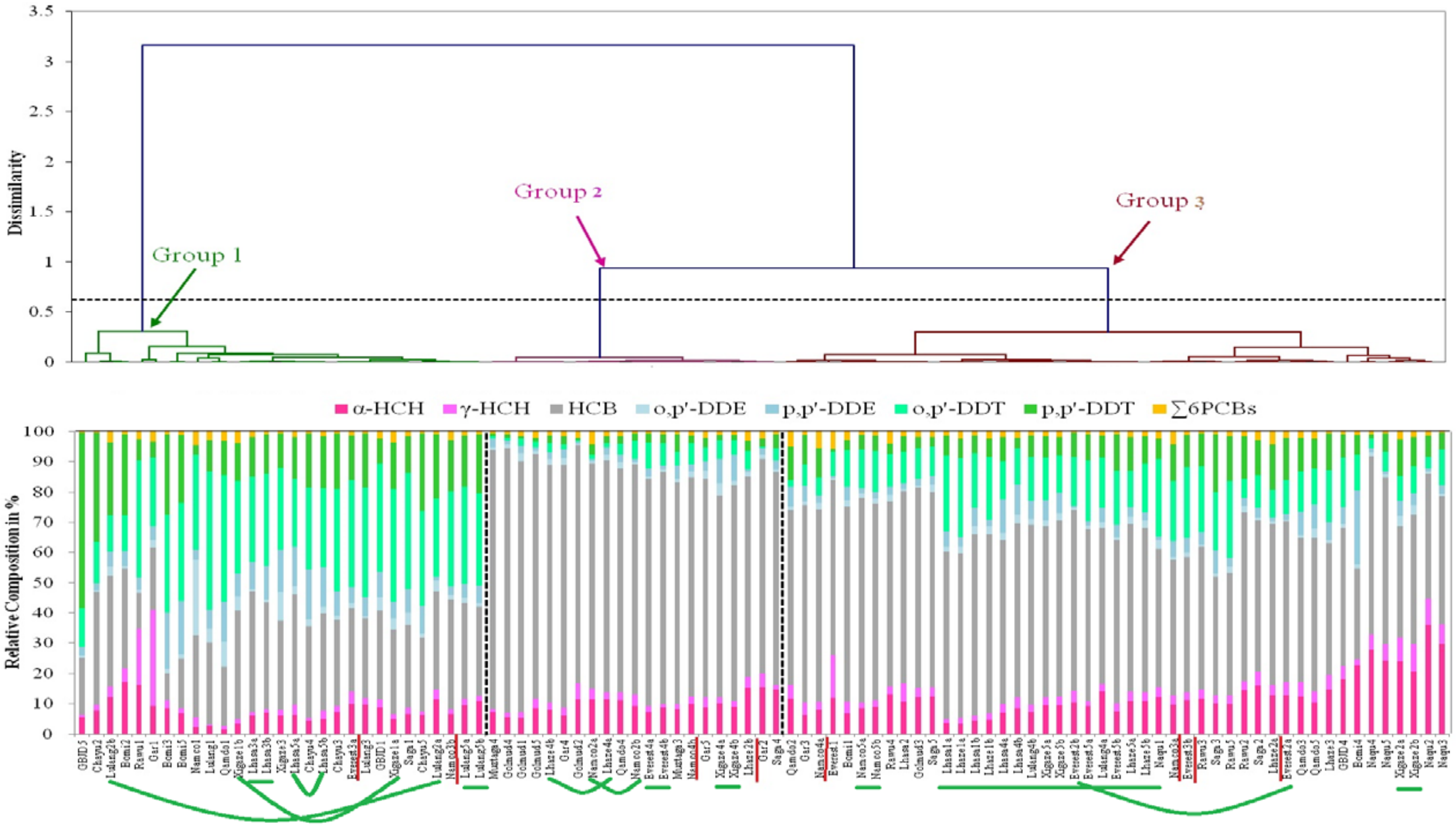



**Figure S1 Result of cluster analysis after including the duplicate samples**

In this study, 23 pairs of duplicates were obtained (Table S4). Duplicates were named by the name of sampling site followed by sampling year and alphabet a or b. For example, Lhasa 1a and Lhasa 1b means the Lhasa duplicates employed for the first sampling year (2007-2008) and Lhasa 2a and 2b means the Lhasa duplicates employed for the second sampling year (2008-2009) and so on. From the figure above, we found that most of samples appeared just shoulder by shoulder, such as the duplicates labeled by green horizontal lines, and the duplicates grouped in the same group were connected by green curves. The samples labeled with red vertical lines were duplicate samples (4 pairs) dispersed in different groups. From Liu and Wania (2014), they suggested only when less than 20% of the duplicates in a study fail to be in the same group, may the analytical uncertainties be acceptable for clustering results. In our study, only 4 of 23 pairs of duplicate samples were dispersed in different groups, which is less than the threshold of 20%. This suggested that the results of cluster analysis were from the real difference/similarity between sampling sites but not the random error of the analytical procedure.

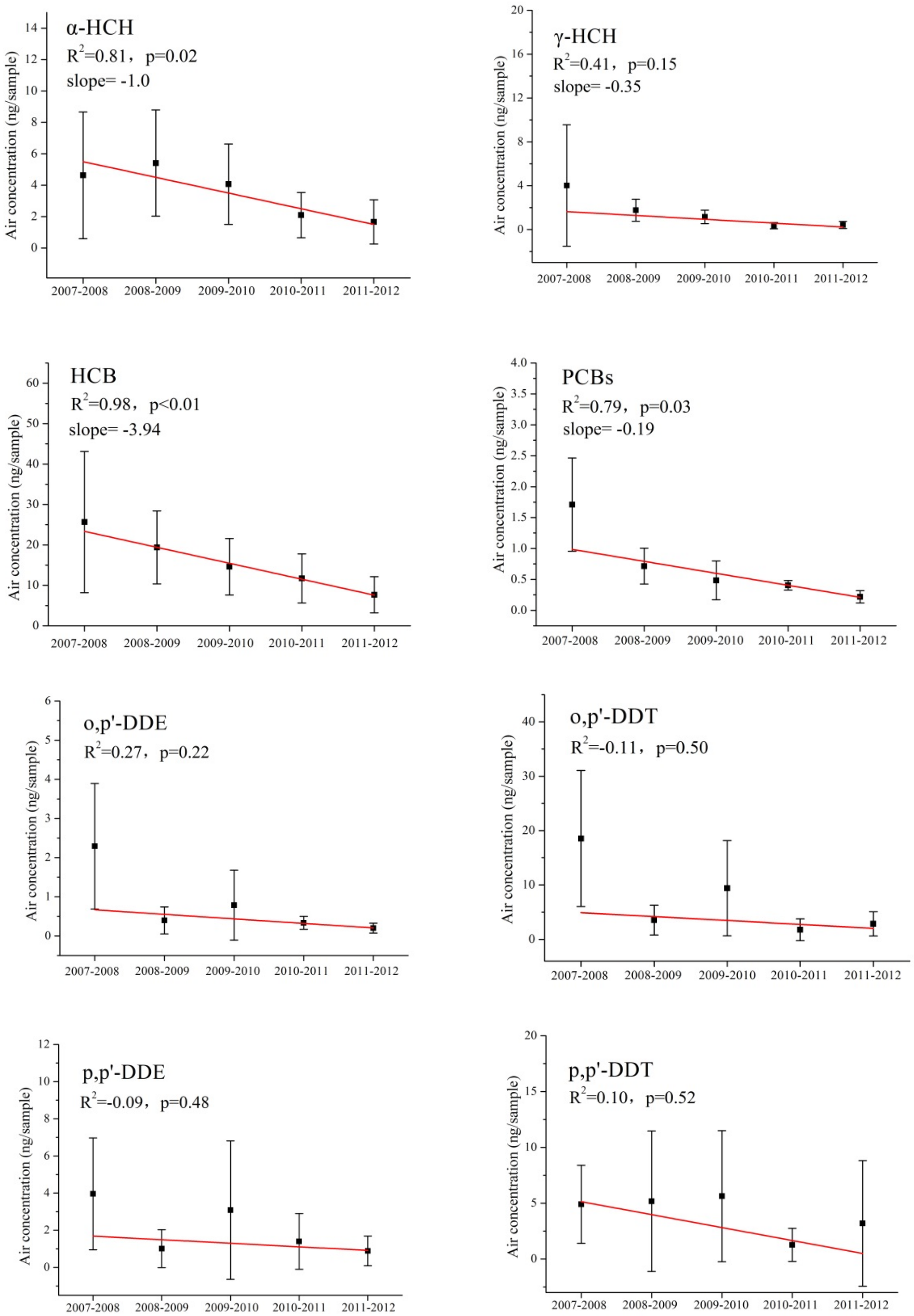



**Figure S2 Inter-annual concentration variation from 2007 to 2012**

**Table S13 . Results of randomized block ANOVA test for the concentrations of DDTs in the monsoon region, westerly region and transition region from 2007 to 2012. ($p < 0.05$ indicates that the levels of DDTs significantly differ between two years.)**

| | | p value | | | |
|---|---|---|---|---|---|
| Year | | o,p'-DDE | p,p'-DDE | o,p'-DDT | p,p'-DDT |
| **Monsoon region** | | | | | |
| 2007~2008 | 2008~2009 | 0.005 | 0.45 | 0.008 | 0.46 |
| | 2009~2010 | 0.006 | 0.51 | 0.12 | 0.39 |
| | 2010~2011 | 0.001 | 0.48 | 0.004 | 0.40 |
| | 2011~2012 | 0.002 | 0.41 | 0.007 | 0.63 |
| 2008~2009 | 2009~2010 | 0.93 | 0.17 | 0.15 | 0.90 |
| | 2010~2011 | 0.44 | 0.96 | 0.63 | 0.13 |
| | 2011~2012 | 0.52 | 0.95 | 0.94 | 0.23 |
| 2009~2010 | 2010~2011 | 0.39 | 0.19 | 0.07 | 0.11 |
| | 2011~2012 | 0.46 | 0.16 | 0.13 | 0.19 |
| 2010~2011 | 2011~2012 | 0.89 | 0.91 | 0.68 | 0.72 |
| **Westerly region** | | | | | |
| 2007~2008 | 2008~2009 | 0.02 | 0.04 | 0.12 | 0.33 |
| | 2009~2010 | 0.008 | 0.24 | 0.49 | 0.38 |
| | 2010~2011 | 0.009 | 0.01 | 0.12 | 0.14 |
| | 2011~2012 | 0.004 | 0.01 | 0.13 | 0.20 |
| 2008~2009 | 2009~2010 | 0.36 | 0.20 | 0.31 | 0.10 |
| | 2010~2011 | 0.42 | 0.35 | 0.98 | 0.53 |
| | 2011~2012 | 0.13 | 0.41 | 0.94 | 0.70 |
| 2009~2010 | 2010~2011 | 0.90 | 0.06 | 0.32 | 0.04 |
| | 2011~2012 | 0.47 | 0.06 | 0.34 | 0.06 |
| 2010~2011 | 2011~2012 | 0.40 | 0.91 | 0.96 | 0.80 |
| **Transition region** | | | | | |
| 2007~2008 | 2008~2009 | 0.007 | 0.03 | 0.001 | 0.12 |
| | 2009~2010 | 0.01 | 0.16 | 0.008 | 0.24 |
| | 2010~2011 | 0.009 | 0.03 | 0.000 | 0.001 |
| | 2011~2012 | 0.007 | 0.03 | 0.001 | 0.01 |
| 2008~2009 | 2009~2010 | 0.79 | 0.41 | 0.37 | 0.68 |
| | 2010~2011 | 0.89 | 0.91 | 0.61 | 0.04 |
| | 2011~2012 | 0.98 | 0.99 | 0.84 | 0.24 |
| 2009~2010 | 2010~2011 | 0.90 | 0.35 | 0.17 | 0.02 |
| | 2011~2012 | 0.82 | 0.41 | 0.28 | 0.12 |
| 2010~2011 | 2011~2012 | 0.92 | 0.90 | 0.75 | 0.37 |

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