

1    *Supplement of*

2    **Atmospheric processes of persistent organic pollutants over**

3    **a remote lake of the central Tibetan Plateau: Implications**

4    **for regional cycling**

5

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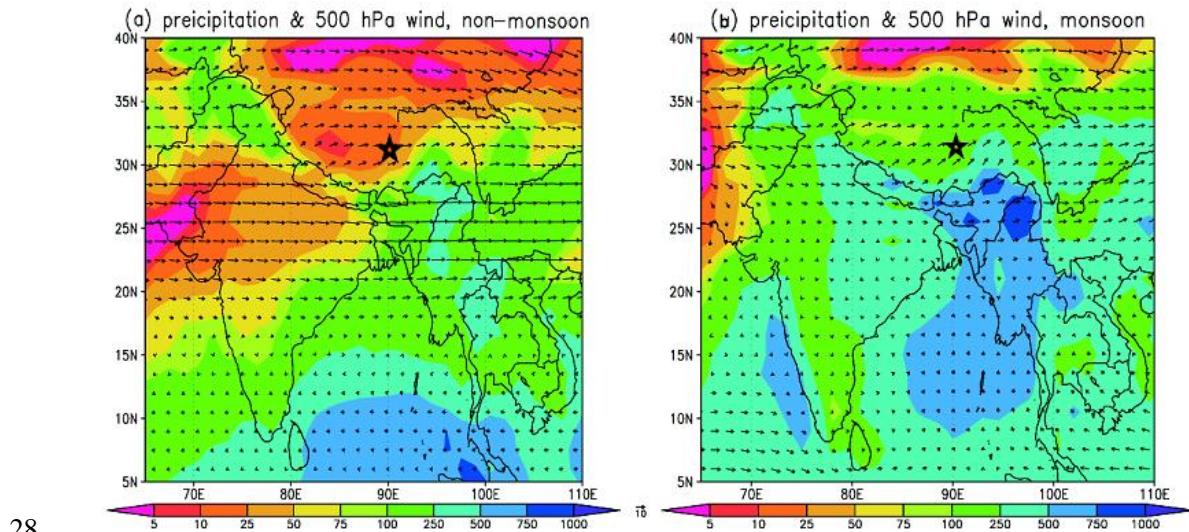
17    **Introduction**

18    [This supplement file includes the details about climate of sampling site, methods on  
19    the extraction and analysis of samples, detection limits and QA/QC results. Dataset of  
20    concentrations for each target compounds in this study and comparisons with  
21    literature values from other remote regions are also presented. Furthermore, method  
22    on air-water exchange calculation, localized physical-chemical properties of POPs and  
23    derived degradation fluxes are also detailed here. In total, this supplement includes six  
24    texts (Text S1 to S6), five figures (Figure S1 to S5) and seventeen tables (Table S1 to  
25    S17). They are listed in the order of appearance in the manuscript; please see the  
26    summary table below.]

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

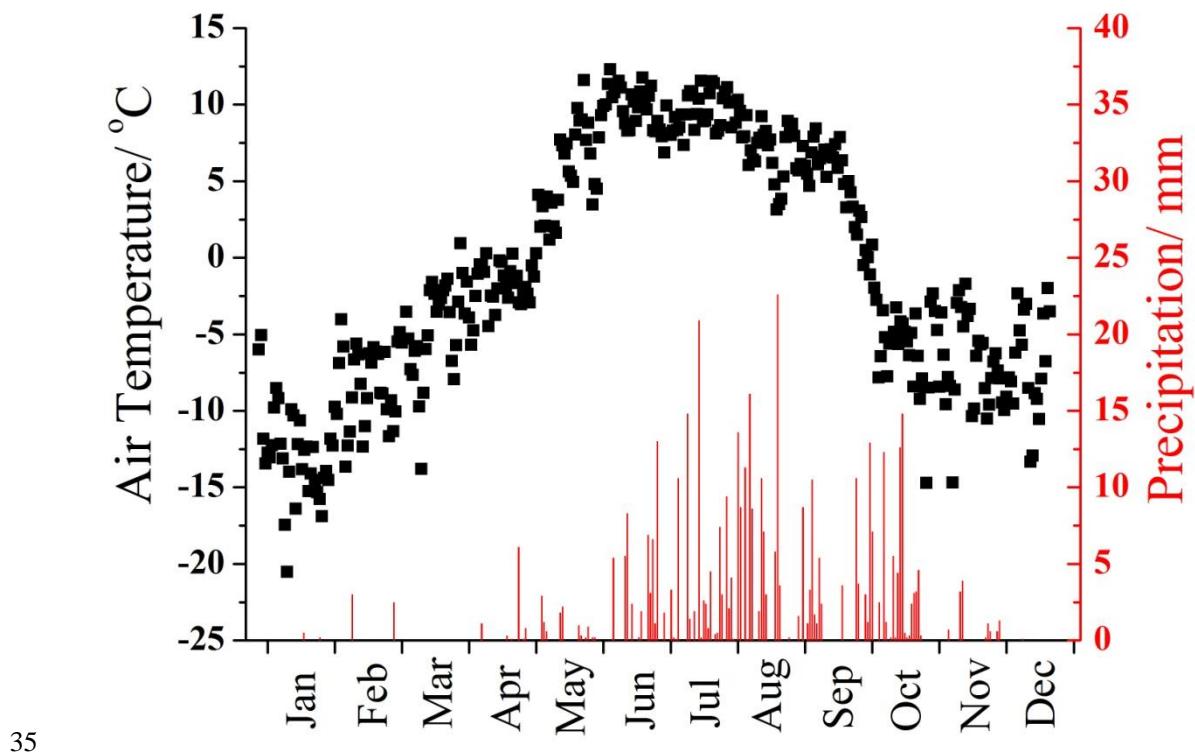
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**Figure S1. The dominated air circulation in Nam Co region for monsoon and non-monsoon season (average from 1980-2010). The star is the location of Nam Co Monitoring and Research Station.**

(Cited from Zhang et al., 2013: A study of Zhadang glacier energy and mass balance and its hydrological process, in NamCo basin, central Tibetan Plateau, Doctoral Dissertation)



35  
36 **Figure S2. Temporal variations of air temperature and precipitation in Nam Co**  
37 **station in 2012.**

38 The meteorological parameters were measured by an automatic weather station in the  
39 Nam Co station, and the data were downloaded in the website of High-cold Region  
40 Observation and Research Network for Land Surface Processes & Environment of  
41 China (HORN, <http://www.horn.ac.cn/>)

**Table S1. Air temperature and wind speed during the sampling period**

Start Date Y/M/D	End Date Y/M/D	Temperature (°C)	Wind speed (m/s)
2012.09.03	2012.09.15	6.7	3.0
2012.09.16	2012.09.20	6.4	3.1
2012.10.01	2012.10.15	-0.7	3.5
2012.10.24	2012.10.31	-5.5	3.0
2012.11.01	2012.11.15	-7.2	4.3
2012.11.16	2012.11.30	-5.9	NA
2012.12.01	2012.12.15	-8.3	NA
2012.12.16	2012.12.31	-6.6	NA
2013.01.03	2013.01.15	-12.6	3.3
2013.01.16	2013.01.31	-13.0	2.9
2013.02.06	2013.02.16	-8.9	3.6
2013.02.21	2013.03.01	-8.7	3.0
2013.03.02	2013.03.15	-6.1	2.2
2013.03.15	2013.03.31	-5.4	3.2
2013.04.01	2013.04.15	-3.2	3.5
2013.04.15	2013.04.30	-1.1	3.2
2013.05.01	2013.05.15	-0.2	3.1
2013.05.15	2013.06.04	5.2	3.3
2013.06.06	2013.06.15	8.0	2.8
2013.06.15	2013.06.30	9.7	3.2
2013.07.01	2013.07.19	10.0	2.8
2013.07.19	2013.08.03	8.5	3.0
2013.08.04	2013.08.15	8.6	2.4
2013.08.15	2013.08.31	8.9	2.9
2013.09.01	2013.09.15	5.2	2.7
2013.09.15	2013.09.30	4.9	2.9
2013.10.01	2013.10.15	-0.7	3.5
2013.10.15	2013.10.30	-5.5	3.0
2013.11.01	2013.11.15	-8.3	3.7
2013.11.15	2013.11.30	-9.7	3.0
2013.12.01	2013.12.15	-12.6	3.1
2013.12.16	2013.12.30	-9.1	4.1
2014.01.05	2014.01.31	-12.6	3.3
2014.02.01	2014.02.15	-8.7	3.7
2014.02.15	2014.02.28	-9.1	3.8
2014.03.01	2014.03.15	-6.1	2.2
2014.03.15	2014.03.31	-5.4	3.2
2014.04.01	2014.04.15	-2.4	3.2
2014.04.15	2014.04.30	-1.0	3.6
2014.05.01	2014.05.15	2.3	3.4
2014.05.15	2014.05.28	5.2	3.2

2014.06.02	2014.06.15	8.0	2.8
2014.06.22	2014.06.30	9.7	3.2
2014.07.01	2014.07.15	10.0	2.8
2014.07.15	2014.07.31	9.2	2.7
2014.08.01	2014.08.15	9.3	2.5
2014.08.15	2014.08.31	6.8	2.5
2014.09.02	2014.09.15	7.9	2.8
2014.09.15	2014.09.30	4.5	3.1

43

NA: not available

44 **Table S2. Information about the water samples.**

45 The information include water temperature, pH, electric conductivity, salinity,  
 46 dissolved oxygen and contents of suspended particulate matter (SPM).

<b>Spatial sample</b>	<b>T (°C)</b>	<b>pH</b>	<b>Electric conductivity (ms cm<sup>-1</sup>)</b>	<b>Salinity (ppt)</b>	<b>Dissolved Oxygen (mg L<sup>-1</sup>)</b>	<b>SPM (mg L<sup>-1</sup>)</b>
S1	14.5	9.48	1.96	1.1	9.2	11.0
S2	NA	9.51	1.92	1.0	9.0	3.2
S3	15.6	9.39	1.85	1.1	8.6	5.5
S4	NA	NA	NA	NA	NA	5.7
S5	14.4	9.48	1.76	1.0	9.3	7.0
S6	12.7	9.56	1.92	1.1	10.2	1.7
S7	11.7	9.44	1.54	0.8	9.4	5.3
S8	12.4	9.56	1.86	1.0	9.9	7.2
S9	12.4	9.56	1.93	1.1	9.6	6.7
S10	8.5	9.63	1.99	1.1	9.7	2.1
S11	8.1	9.49	2.02	1.1	8.9	6.1
S12	14.8	9.50	1.86	1.1	10.1	3.2
S13	14.6	9.40	1.83	1.1	10.2	5.2
S14	14.9	9.34	1.93	1.0	9.5	11.7
S15	14.8	9.38	0.74	1.0	NA	8.2

<b>Seasonal sample</b>	<b>T (°C)</b>	<b>PH</b>	<b>Electric conductivity (ms cm<sup>-1</sup>)</b>	<b>Salinity (ppt)</b>	<b>Dissolved Oxygen (mg L<sup>-1</sup>)</b>	<b>SPM (mg L<sup>-1</sup>)</b>
May-1	8.2	8.98	1.92	1.0	9.8	2.6
May-2	8.5	9.01	1.91	1.1	11.2	2.2
May-3	8.9	9.03	1.89	1.2	12.6	3.0
June-1	10.1	9.13	1.9	1.2	11.7	0.9
June-2	10.3	9.14	1.88	1.1	10.3	2.2
June-3	10.9	9.14	1.86	1.0	8.9	0.8
July-1	16.7	9.16	1.83	0.8	17.3	2.5
July-2	15.9	9.10	1.82	0.9	17.8	4.0
July-3	12.8	9.16	1.83	0.9	12.6	1.9
Aug-1	15.6	9.25	1.75	0.8	20.0	2.4
Aug-2	14.0	9.27	1.80	1.0	20.0	0.4
Aug-3	13.2	9.28	1.85	1.1	NA	0.5
Sep-1	12.4	9.16	1.82	0.8	13.4	1.0
Sep-2	12.2	9.18	1.82	1.0	11.9	2.0
Sep-3	12.3	9.17	1.82	0.9	12.7	0.6

47 NA: not available

48    **Text S1. Preparation, extraction and cleanup method for different samples**

49    **Preparation.** Polyurethane foam (PUF) and glass fiber filter (GFF 0.7 µm, Whatman)  
50    were used to absorb the gas and particulate phase POPs in air, respectively; while  
51    GFFs and XAD-2 resin were used to accumulate the POPs in suspended particulate  
52    matter (SPM) and dissolved phases in water, respectively. Prior to field sampling,  
53    PUF and XAD resin was pre-cleaned by Soxhlet extraction using dichloromethane  
54    (DCM) for 16 h; GFF were pre-combusted at 450 °C for 4 h. Glass adsorption column  
55    (internal diameter of 2.5 cm and length of 30 cm) was prepared and each one was  
56    filled with about 200 cm<sup>3</sup> of XAD resin.

57    **Extraction.** Each collected sample was spiked with a mixture of 2, 4,  
58    6-trichlorobiphenyl (PCB 30), Mirex and perylene-D12 as recovery surrogates before  
59    extraction. Different extraction methods were used for XAD-resin and other samples,  
60    but the procedure for cleanup is the same. The XAD column after passing through the  
61    water samples was first eluted with 100 mL of DCM. Then XAD resin was transferred  
62    to conical flask and mixed with enough anhydrous sodium sulfate for dehydration.  
63    Ultrasonically extraction was repeated for 3 times (20 min with 50 mL DCM for each  
64    time) to extract remaining chemicals. For the PUF and GFF filters, Soxhlet extraction  
65    was conducted using DCM at 48 °C for 16 h.

66    **Cleanup.** Extracts of the above samples were all concentrated to 1 mL by a rotary  
67    evaporator, solvent exchanged to hexane and then cleanup. The samples were loaded  
68    on a chromatography column (from the bottom to top: 3 g of 6% deactivated silica gel,  
69    2 g of 3% deactivated alumina and 1 g of anhydrous sodium sulfate) and eluted with  
70    30 mL of a mixture of hexane/DCM (1:1 v/v). The elute was further cleaned on  
71    gel-permeation chromatography (GPC, containing 6 g of Biobeads SX3) with  
72    hexane/DCM (1:1). The first 16 mL was discarded and the subsequent 30 mL fraction  
73    was collected. After the cleanup, all samples were finally concentrated to 100 µL  
74    containing a known quantity of pentachloronitrobenzene (PCNB), decachlorobiphenyl  
75    (PCB 209), prylene-D10 and benzo[g,h,i]perylene-D12 as internal standards.

76 **Text S2. Details about the chromatographic conditions**

77 The POPs concentrations in all samples were analyzed on the gas  
78 chromatograph-mass spectrometer (GC-MS, Finnigan Trace GC/PolarisQ); however  
79 different chromatographic column and temperature program were used for OCPs,  
80 PCBs and PAHs. Additionally, the enantiomers of  $\alpha$ -HCH were analyzed separately.

81 **OCPs and PCBs.** The GC-MS was operated with a CP-Sil 8CB capillary column (50  
82 m  $\times$  0.25 mm, film thickness 0.25  $\mu\text{m}$ ) and under single-ion monitoring (SIM) mode.  
83 Helium was used as the carrier gas at 1  $\text{mL min}^{-1}$  under constant-flow mode. The oven  
84 temperature began at 100  $^{\circ}\text{C}$  for 2 min and was increased at a rate of 20  $^{\circ}\text{C min}^{-1}$  to  
85 140  $^{\circ}\text{C}$ , then increased at a rate of 4  $^{\circ}\text{C min}^{-1}$  to 200  $^{\circ}\text{C}$ , held for 10 min, and finally  
86 increased at a rate of 4  $^{\circ}\text{C min}^{-1}$  to 300  $^{\circ}\text{C}$  and held for 17 min.

87 **PAHs.** PAHs were analyzed on GC-MS system with a DB-5MS column (60 m  $\times$  0.25  
88 mm, film thickness 0.25  $\mu\text{m}$ ). The carrier gas and SIM mode are the same as OCPs  
89 and PCBs. The oven temperature began at 50  $^{\circ}\text{C}$  for 2 min and was increased at a rate  
90 of 20  $^{\circ}\text{C min}^{-1}$  to 180  $^{\circ}\text{C}$ , then increased at a rate of 3  $^{\circ}\text{C min}^{-1}$  to 250  $^{\circ}\text{C}$ , then  
91 increased at a rate of 2  $^{\circ}\text{C min}^{-1}$  to 300  $^{\circ}\text{C}$  and held for 10 min, and finally increased  
92 at a rate of 10  $^{\circ}\text{C min}^{-1}$  to 310  $^{\circ}\text{C}$  and held for 5 min.

93 **Chiral analysis for  $\alpha$ -HCH.** Determination of enantiomeric compositions of  $\alpha$ -HCH  
94 was performed on an Agilent 6890 gas chromatography equipped with a micro-cell  
95  $^{63}\text{Ni}$  electron capture detector ( $\mu$ -ECD) at State Key Laboratory of Environmental  
96 Chemistry and Ecotoxicology, Research Center of Eco-Environment Sciences,  
97 Chinese Academy of Sciences. Separation for enantiomers of  $\alpha$ -HCH was carried out  
98 using a BGB-172 chiral column (30 m  $\times$  0.25 mm i.d., 0.25  $\mu\text{m}$  film thickness, BGB  
99 Analytik AG, Switzerland). The carrier gas was nitrogen with a flow of 0.7 mL/min.  
100 The injector and detector temperatures were 250  $^{\circ}\text{C}$  and 300  $^{\circ}\text{C}$ , respectively. The  
101 temperature program was: 90  $^{\circ}\text{C}$  held for 1 min, increased to 140  $^{\circ}\text{C}$  at a rate of  
102 15  $^{\circ}\text{C}/\text{min}$ , followed by 2  $^{\circ}\text{C}/\text{min}$  to 210  $^{\circ}\text{C}$ , held for 5 min and then 20  $^{\circ}\text{C}/\text{min}$  to  
103 240  $^{\circ}\text{C}$ , held for 10 min. According to the previous study (Ding et al., 2007a), the  
104 elution order of  $\alpha$ -HCH on BGB column was (-)- $\alpha$ -HCH and (+)- $\alpha$ -HCH.

105

**Table S3.** Concentrations of POPs in the field blanks and procedural blanks.

106

**Part 1. OCPs and PCBs in the field blanks:**

<b>Field blank Air (pg m<sup>-3</sup>)</b>	<b><math>\alpha</math>-HCH</b>	<b><math>\beta</math>-HCH</b>	<b><math>\gamma</math>-HCH</b>	<b>HCB</b>	<b><i>o,p'</i>- DDE</b>	<b><i>p,p'</i>- DDE</b>	<b><i>o,p'</i>- DDT</b>	<b><i>p,p'</i>- DDT</b>	<b>PCB- 28</b>	<b>PCB- 52</b>	<b>PCB- 101</b>	<b>PCB- 153</b>	<b>PCB- 138</b>	<b>PCB- 180</b>
PUF field blank 1	0.10	ND <sup>a</sup>	0.06	1.97	ND	0.44	ND	ND	ND	0.02	ND	ND	ND	ND
PUF field blank 2	0.10	ND	0.03	1.84	ND	0.15	ND	ND	ND	0.04	ND	ND	ND	ND
PUF field blank 3	0.10	ND	0.03	1.50	ND	0.28	ND	ND	ND	0.03	ND	ND	ND	ND
PUF field blank 4	0.13	ND	0.04	0.9	ND	0.38	ND	ND	ND	0.05	ND	ND	ND	ND
PUF field blank 5	0.25	ND	0.03	1.14	ND	0.47	ND	ND	ND	0.07	ND	ND	ND	ND
PUF field blank 6	0.15	ND	0.09	1.43	ND	0.36	ND	ND	ND	0.05	ND	ND	ND	ND
Blank average	0.14	0	0.05	1.46	0	0.35	0	0	0	0.04	0	0	0	0
<b>Field blank Water (pg L<sup>-1</sup>)</b>	<b><math>\alpha</math>-HCH</b>	<b><math>\beta</math>-HCH</b>	<b><math>\gamma</math>-HCH</b>	<b>HCB</b>	<b><i>o,p'</i>- DDE</b>	<b><i>p,p'</i>- DDE</b>	<b><i>o,p'</i>- DDT</b>	<b><i>p,p'</i>- DDT</b>	<b>PCB- 28</b>	<b>PCB- -52</b>	<b>PCB- 101</b>	<b>PCB- 153</b>	<b>PCB- 138</b>	<b>PCB- 180</b>
XAD field blank 1	0.79	1.31	1.08	1.05	0.57	0.27	0.19	0.34	0.14	0.18	0.15	0.05	0.01	0.09
XAD field blank 2	0.44	1.20	0.93	1.03	0.58	0.44	0.22	0.30	0.08	0.16	0.27	0.15	0.07	0.03
XAD field blank 3	BDL	0.53	0.05	0.85	0.30	0.34	0.14	0.17	BDL	0.05	0.18	0.04	0.02	BDL
Blank average	0.61	1.01	0.69	0.98	0.48	0.35	0.18	0.27	0.11	0.13	0.20	0.08	0.03	0.06

107 <sup>a</sup>ND=chemical was not detected in blank.

108

109

**Part 2. PAHs in the field blanks:**

Field blank Air (pg m <sup>-3</sup> )	Acel	Ace	Phe	Flu	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	BaP	IcdP	DahA	BghiP
PUF field blank 1	0.95	0.21	0.76	ND	0.24	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PUF field blank 2	0.87	0.19	0.34	ND	0.15	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PUF field blank 3	0.73	0.45	0.12	ND	0.46	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PUF field blank 4	0.61	0.54	0.56	ND	0.31	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PUF field blank 5	0.92	0.23	0.92	ND	0.36	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
PUF field blank 6	0.59	0.52	0.43	ND	0.27	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Blank average	0.78	0.36	0.52	0	0.30	0	0	0	0	0	0	0	0	0	0
Field blank Water (pg L <sup>-1</sup> )	Acel	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	Bap	IcdP	DahA	BghiP
XAD field blank 1	8.7	12.2	87.0	348.0	350.6	111.5	80.3	7.3	15.6	ND	ND	ND	ND	ND	ND
XAD field blank 2	2.8	3.4	52.9	244.4	41.1	105.8	59.5	4.1	9.1	ND	ND	ND	ND	ND	ND
XAD field blank 3	4.1	6.2	24.1	73.2	218.9	4.0	14.8	2.3	4.6	ND	ND	ND	ND	ND	ND
Blank average	5.2	7.3	54.7	221.9	203.5	73.8	51.5	4.6	9.7	ND	ND	ND	ND	ND	ND

110

**Part 3. OCPs, PCBs and PAHs in the procedural blanks (ng sample<sup>-1</sup>):**

(ng sample <sup>-1</sup> )	$\alpha$ -HCH	$\gamma$ -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
procedural blank 1	0.07	0.02	0.78	ND	0.16	ND	ND	ND	0.03	ND	ND	ND	ND
procedural blank 2	0.10	0.02	0.91	ND	0.23	ND	ND	ND	0.02	ND	ND	ND	ND
procedural blank 3	0.08	0.04	0.64	ND	0.20	ND	ND	ND	0.02	ND	ND	ND	ND
procedural blank 4	0.08	0.04	1.04	ND	0.16	ND	ND	ND	0.02	ND	ND	ND	ND
procedural blank 5	0.07	0.04	0.86	ND	0.14	ND	ND	ND	0.02	ND	ND	ND	ND
procedural blank 6	0.11	0.03	0.89	ND	0.28	ND	ND	ND	0.04	ND	ND	ND	ND
average	0.08	0.03	0.85	0	0.20	0	0	0	0.02	0	0	0	0
(ng sample <sup>-1</sup> )	Acel	Ace	Phe	Flu	Ant	BaA	Chr	Bbf	Bkf	BaP	IcdP	DahA	BghiP
procedural blank 1	0.24	0.13	0.29	ND	0.14	ND	ND	ND	ND	ND	ND	ND	ND
procedural blank 2	0.28	0.38	0.38	ND	0.27	ND	ND	ND	ND	ND	ND	ND	ND
procedural blank 3	0.13	0.38	0.43	ND	0.08	ND	ND	ND	ND	ND	ND	ND	ND
procedural blank 4	0.19	0.26	0.40	ND	0.17	ND	ND	ND	ND	ND	ND	ND	ND
procedural blank 5	0.05	0.07	0.14	ND	0.07	ND	ND	ND	ND	ND	ND	ND	ND
procedural blank 6	0.25	0.37	0.41	ND	0.31	ND	ND	ND	ND	ND	ND	ND	ND
average	0.19	0.27	0.34	0	0.17	0	0	0	0	0	0	0	0

112 **Text S3. The ways to define method detection limits (MDLs) and treat with the**  
113 **concentrations below MDLs**

114 The MDLs were derived as the mean blank concentration plus 3 times its standard  
115 deviation; when a target compound was not detected in the blanks, the concentration  
116 of the lowest calibration standard was substituted for the MDL. Based on 600 m<sup>3</sup> and  
117 200 L, the corresponding MDLs for air and water samples were calculated  
118 respectively and presented in Table S4.

119 If the concentration of a compound after blank correction was below the MDL, the  
120 concentration was substituted with 1/2 MDL in cases where greater than 70% of data  
121 were greater than the MDL (Antweiler and Taylor, 2008); otherwise it will be marked  
122 with “BDL”.

123

**Table S4. Detection frequency and MDLs for individual compound in each kind of samples**

124

	Air-gas phase			Air-particulate phase			Water-dissolved phase			Water-particulate phase		
	detection frequency (%)	MDL (pg m <sup>-3</sup> )	1/2 MDL (pg m <sup>-3</sup> )	detection frequency (%)	MDL (pg m <sup>-3</sup> )	1/2 MDL (pg m <sup>-3</sup> )	detection frequency (%)	MDL (pg L <sup>-1</sup> )	1/2 MDL (pg L <sup>-1</sup> )	detection frequency (%)	MDL (pg L <sup>-1</sup> )	1/2 MDL (pg L <sup>-1</sup> )
<b><math>\alpha</math>-HCH</b>	100	0.31	0.16	2	0.23	0.12	100	1.4	0.7	10	0.70	0.35
<b><math>\beta</math>-HCH</b>	43	0.33	0.17	2	0.33	0.17	100	2.3	1.1	17	1	0.5
<b><math>\gamma</math>-HCH</b>	96	0.12	0.06	19	0.09	0.04	100	2.4	1.2	53	0.26	0.13
<b>HCB</b>	100	2.68	1.34	5	2.11	1.05	79	1.3	0.7	37	6.32	3.16
<b><i>o,p'</i>-DDE</b>	68	0.33	0.17	0	0.33	0.17	0	1.0	0.5	3	1	0.5
<b><i>p,p'</i>-DDE</b>	66	0.70	0.35	7	0.59	0.30	10	0.6	0.3	13	1.78	0.89
<b><i>o,p'</i>-DDT</b>	91	0.33	0.17	23	0.33	0.17	3	0.3	0.2	10	1	0.5
<b><i>p,p'</i>-DDT</b>	72	0.33	0.17	21	0.33	0.17	3	0.5	0.3	33	1	0.5
<b>PCB-28</b>	100	0.03	0.02	23	0.03	0.02	72	0.2	0.1	50	0.1	0.05
<b>PCB-52</b>	83	0.10	0.05	0	0.08	0.04	17	0.3	0.2	27	0.23	0.12
<b>PCB-101</b>	98	0.03	0.02	74	0.03	0.02	3	0.4	0.2	93	0.1	0.05
<b>PCB-153</b>	81	0.03	0.02	28	0.03	0.02	0	0.3	0.1	47	0.1	0.05
<b>PCB-138</b>	79	0.03	0.02	21	0.03	0.02	7	0.1	0.06	53	0.1	0.05
<b>PCB-180</b>	19	0.03	0.02	5	0.03	0.02	0	0.2	0.09	10	0.1	0.05
<b>Acel</b>	100	1.25	0.63	23	0.75	0.37	100	14	7	100	2.25	1.12
<b>Ace</b>	100	0.85	0.42	72	1.13	0.57	100	21	10	100	3.40	1.70
<b>Flu</b>	100	0.33	0.17	100	0.33	0.17	100	149	75	100	1	0.5
<b>Phe</b>	100	1.39	0.70	100	1.12	0.56	100	638	319	100	3.37	1.69
<b>Ant</b>	100	0.62	0.31	100	0.77	0.39	48	670	335	97	2.31	1.16
<b>Fla</b>	100	0.33	0.17	100	0.33	0.17	56	255	128	100	1	0.5

<b>Pyr</b>	100	0.17	0.08	100	0.17	0.08	93	152	76	100	0.5	0.25
<b>BaA</b>	100	0.17	0.08	100	0.17	0.08	48	12	6	97	0.5	0.25
<b>Chr</b>	100	0.17	0.08	100	0.17	0.08	56	26	13	97	0.5	0.25
<b>Bbf</b>	100	0.33	0.17	86	0.33	0.17	52	1	0.5	55	1	0.5
<b>Bkf</b>	98	0.17	0.08	100	0.17	0.08	100	0.5	0.3	97	0.5	0.25
<b>BaP</b>	98	0.17	0.08	98	0.17	0.08	100	0.5	0.3	97	0.5	0.25
<b>IcdP</b>	49	0.17	0.08	56	0.17	0.08	0	0.5	0.3	55	0.5	0.25
<b>DahA</b>	31	0.33	0.17	33	0.33	0.17	0	1	0.5	48	1	0.5
<b>BghiP</b>	96	0.33	0.17	100	0.33	0.17	100	1	0.5	97	1	0.5

125

**Table S5. Breakthrough estimation: the percentage of OCPs, PCBs and PAHs in the second PUF to the total Plugs (PUF<sub>2</sub>/(PUF<sub>1</sub>+PUF<sub>2</sub>))**

	<i>α</i> -HCH	<i>β</i> -HCH	<i>γ</i> -HCH	HCB	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	PCB-28	PCB-52	PCB-101	PCB-153	PCB-138	PCB-180	
	Acel	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	Bap	IcdP	DahA	BghiP
breakthrough 1	15%	0%	11%	24%	5%	13%	5%	6%	11%	10%	13%	12%	8%	0%	
breakthrough 2	20%	4%	14%	18%	0%	2%	1%	1%	14%	7%	7%	0%	3%	0%	
breakthrough 3	25%	8%	17%	25%	0%	12%	8%	6%	14%	11%	13%	0%	16%	11%	
breakthrough 4	26%	7%	22%	30%	4%	5%	3%	2%	19%	7%	5%	3%	5%	0%	
breakthrough 5	28%	0%	12%	26%	3%	0%	1%	1%	10%	5%	6%	3%	1%	0%	
breakthrough 6	16%	25%	31%	20%	26%	25%	10%	19%	24%	0%	13%	10%	9%	0%	
breakthrough 7	17%	15%	25%	25%	14%	16%	14%	16%	18%	17%	15%	18%	15%	26%	
breakthrough 8	13%	18%	22%	16%	22%	24%	14%	11%	17%	14%	15%	15%	15%	9%	
breakthrough 9	13%	16%	14%	20%	14%	17%	15%	16%	13%	16%	18%	18%	19%	19%	
breakthrough 10	22%	20%	19%	32%	27%	22%	20%	23%	23%	17%	20%	21%	45%	11%	
breakthrough 11	15%	20%	10%	20%	4%	5%	3%	13%	10%	7%	8%	16%	7%	9%	
<b>average</b>	<b>19%</b>	<b>12%</b>	<b>18%</b>	<b>23%</b>	<b>11%</b>	<b>13%</b>	<b>9%</b>	<b>10%</b>	<b>16%</b>	<b>10%</b>	<b>12%</b>	<b>11%</b>	<b>13%</b>	<b>8%</b>	
breakthrough 1	1%	1%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
breakthrough 2	33%	34%	32%	31%	26%	20%	19%	31%	25%	39%	33%	38%	0%	0%	37%
breakthrough 3	16%	17%	15%	19%	16%	23%	23%	12%	26%	6%	17%	16%	0%	0%	21%
breakthrough 4	15%	10%	22%	23%	23%	20%	18%	19%	14%	12%	8%	11%	0%	0%	22%
breakthrough 5	24%	23%	28%	34%	31%	28%	24%	21%	19%	21%	24%	32%	26%	19%	24%
breakthrough 6	20%	21%	20%	19%	23%	14%	22%	22%	22%	23%	19%	28%	21%	18%	20%
breakthrough 7	23%	21%	17%	13%	14%	13%	14%	14%	14%	12%	12%	10%	15%	14%	15%
breakthrough 8	22%	23%	24%	19%	13%	20%	23%	33%	30%	33%	35%	34%	35%	27%	32%
breakthrough 9	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
breakthrough 10	22%	22%	23%	32%	30%	30%	31%	17%	24%	26%	21%	31%	50%	0%	23%
breakthrough 11	6%	7%	10%	13%	12%	11%	8%	26%	25%	17%	18%	28%	19%	32%	13%
<b>average</b>	<b>17%</b>	<b>16%</b>	<b>17%</b>	<b>18%</b>	<b>17%</b>	<b>16%</b>	<b>17%</b>	<b>18%</b>	<b>18%</b>	<b>17%</b>	<b>17%</b>	<b>21%</b>	<b>15%</b>	<b>10%</b>	<b>19%</b>

127 **Text S4. Correction of the dissolved POPs concentrations by dissolved organic  
128 carbon (DOC)**

In order to obtain the concentrations of truly freely dissolved POPs ( $C_w$ ), results of dissolved concentrations retained by the XAD should be corrected by the estimated influence of sorption by DOC in water as follows (Ruge et al., 2015):

$$C_w = C_{XAD} / [1 + K_{DOC} * C_{DOC}]$$

where  $C_{XAD}$  ( $\text{pg L}^{-1}$ ) is the POPs concentration derived from XAD samples,  $K_{\text{DOC}}$  is the DOC-water equilibrium partitioning coefficient ( $\text{m}^3 \text{ kg}^{-1}$ ), and  $C_{\text{DOC}}$  is the concentration of DOC in water ( $\text{kg m}^{-3}$ ).  $K_{\text{DOC}}$  can be estimated from octanol-water partition coefficient ( $K_{\text{ow}}$ ) by using correlation of  $\log K_{\text{DOC}}$  versus  $\log K_{\text{ow}}$  derived by Burkhard et al. (2000):

$$\log K_{\text{DOC}} = 0.85 * \log K_{\text{OW}} - 0.25$$

139 For the Nam Co Lake, a  $C_{DOC}$  of  $4.23 \text{ mg L}^{-1}$  was adopted from previous literature  
140 (Liu et al., 2010).

141    **Text S5. Temperature and salinity-corrected Henry's law constant ( $H$ )**

142    (1) By using the equations below,  $H$  values (unit: Pa m<sup>3</sup> mol<sup>-1</sup>) were corrected for the  
 143    measured water temperature in this study.

Compound	Regression equations	References
$\alpha$ -HCH	$\log H = 10.13 - 3098/T(K)$	(Sahsuvar et al., 2003)
$\beta$ -HCH	$\log H = 9.96 - 3400/T(K)$	(Sahsuvar et al., 2003)
$\gamma$ -HCH	$\log H = 10.14 - 3208/T(K)$	(Sahsuvar et al., 2003)
HCB	$\log H = 11.6 - 3013/T(K)$	(Jantunen et al., 2006)
PCB-28	$\log H = 10.11 - 2547.1/T(K)$	(Paasivirta et al., 2009)
AceI	$\log H = 10.47 - 2798/T(K)$	(Ma et al., 2010)
Ace	$\log H = 9.37 - 2443.3/T(K)$	(Ma et al., 2010)
Flu	$\log H = 10.66 - 2887.8/T(K)$	(Ma et al., 2010)
Phe	$\log H = 8.13 - 2230.3/T(K)$	(Ma et al., 2010)
Pyr	$\log H = 8.61 - 2528.7/T(K)$	(Ma et al., 2010)
BaA	$\log H = 8.46 - 2568.7/T(K)$	(Ma et al., 2010)
Chr	$\log H = 17.71 - 5384.2/T(K)$	(Ma et al., 2010)
Bbf	$\log H = 6.76 - 2366.8/T(K)$	(Ma et al., 2010)
Bkf	$\log H = 7.3 - 2541.4/T(K)$	(Ma et al., 2010)
Bap	$\log H = 5.54 - 2051/T(K)$	(Ma et al., 2010)
IcdP	$\log H = 3.85 - 1580.5/T(K)$	(Ma et al., 2010)
DahA	$\log H = 11.23 - 3371/T(K)$	(Ma et al., 2010)
BghiP	$\log H = 3.167 - 1384/T(K)$	(Ma et al., 2010)

144    (2) The aqueous solubility of a nonelectrolyte has been found generally to be  
 145    dependent on the concentration and type of salt present in solution. The salt effect on  
 146     $H$  is frequently described by the Setschenow equation (Cetin et al., 2006):

147                          
$$\log(H^*/H) = K_S C_S$$

148    where  $H^*$  and  $H$  are the Henry's law constant in saline water and deionized water,  
 149    respectively;  $K_S$  is the Setschenow or the salting-out constant (L mol<sup>-1</sup>);  $C_S$  is the  
 150    molar concentration of salt in lake water (mol L<sup>-1</sup>).  $K_S$  was estimated from  $K_{OW}$  using  
 151    a linear relationship derived by Ni et al. (2003):

152                          
$$K_S = 0.040 * \log K_{OW} + 0.114$$

153    For Nam Co Lake, the measured salinity ranged between 0.8‰ and 1.2‰, thus a  
 154    corresponding  $C_S$  of 0.02 mol L<sup>-1</sup> was used. Finally, the corrected  $H^*/H = 1.01$  or 1.02.

155 **Text S6. Flux calculation of air-water gas exchange**

156 The fluxes of air-water gas exchange ( $F_{AW}$ , ng m<sup>-2</sup> day<sup>-1</sup>) were calculated using the  
157 two-film model which has been used in many previous studies (Qiu et al., 2008; Xie  
158 et al., 2011):

159 
$$F_{AW} = K_{ol} (C_w - C_a R T_a / H)$$

160 where  $C_w$  and  $C_a$  are the dissolved and gaseous concentrations of the target  
161 compounds, which are considered to be the measured air and DOC-corrected water  
162 concentrations;  $R$  is the gas constant (8.314 Pa m<sup>3</sup> mol<sup>-1</sup> K<sup>-1</sup>),  $T_a$  (K) is the absolute  
163 temperature, and  $H$  (Pa m<sup>3</sup> mol<sup>-1</sup>) is the Henry's law constant.  $K_{ol}$  (m s<sup>-1</sup>) is the overall  
164 mass transfer coefficient, which contains contributions from the mass transfer  
165 coefficients of the water layer and the air layer, namely  $K_w$  and  $K_a$ . The value of  $K_{ol}$  is  
166 given by (Khairy et al., 2014):

167 
$$\frac{1}{K_{ol}} = \frac{RT}{H} \times \frac{1}{K_a} + \frac{1}{K_w}$$

168 where  $K_w$  and  $K_a$  are the water-side and air-side mass transfer coefficients,  
169 respectively. They are related to the wind speed and compound-specific molecular  
170 diffusivity. In the present study,  $K_w$  and  $K_a$  of target compounds were estimated from  
171 those of reference substances as follows.

172  $K_a$  (cm s<sup>-1</sup>) for water vapour and  $K_w$  (cm h<sup>-1</sup>) for CO<sub>2</sub> can be calculated according to  
173 the following relations:

174 
$$K_a(\text{H}_2\text{O}) = 0.2 \times U_{10} + 0.3$$

175 
$$K_w(\text{CO}_2) = 0.45 \times U_{10}^{1.64}$$

176 where  $U_{10}$  (m s<sup>-1</sup>) is the wind speed above the water surface at 10 m. It was calculated  
177 by using the wind speed ( $U_Z$ , m s<sup>-1</sup>) at any given height ( $Z$ , m) by following equation  
178 (Schwarzenbach et al., 2003):

179 
$$U_{10} = U_Z \left( \frac{10.4}{\ln(Z) + 8.1} \right)$$

180 Because rates of transfer is related to the molecular diffusivity, these estimates for  $K_a$   
181 ( $\text{H}_2\text{O}$ ) and  $K_w$  (CO<sub>2</sub>) allow prediction of  $K_a$  and  $K_w$  for other compounds of interest  
182 with the following relationships (Schwarzenbach et al., 2003):

183 
$$K_a(\text{analyte}) = K_a(\text{H}_2\text{O}) \times \left\{ \frac{D_a(\text{analyte})}{D_a(\text{H}_2\text{O})} \right\}^{0.67}$$

184                   
$$K_w(\text{analyte}) = K_w(\text{CO}_2) \times \left\{ \frac{S_c(\text{analyte})}{S_c(\text{CO}_2)} \right\}^{-0.5}$$

185       where  $D_a$  is the molecular diffusivity in air and  $Sc$  is the Schmidt number, calculated  
186       by dividing the kinematic viscosity of water ( $V_w$ ,  $\text{cm}^2 \text{ s}^{-1}$ ) at a given temperature by  
187       the molecular diffusivity in water ( $D_w$ ):

$$S_c(\text{analyte}) = \frac{V_w}{D_w(\text{analyte})}$$

188        $D_a$  and  $D_w$  values were estimated using the compound molecular mass to simplify the  
189       relationship (Schwarzenbach et al., 2003). We can derive the unknown diffusivity of  
190       target compounds in the present study by using the diffusivity of a reference substance,  
191       such as  $D_a$  ( $\text{H}_2\text{O}$ ) and  $D_w$  ( $\text{CO}_2$ ) according to the following equation (Schwarzenbach  
192       et al., 2003):

193                   
$$\frac{D_a(\text{analyte})}{D_a(\text{H}_2\text{O})} \approx \left[ \frac{M(\text{analyte})}{M(\text{H}_2\text{O})} \right]^{-0.5}$$

194                   
$$\frac{D_w(\text{analyte})}{D_w(\text{CO}_2)} \approx \left[ \frac{M(\text{analyte})}{M(\text{CO}_2)} \right]^{-0.5}$$

195

**Table S6. Equations used for temperature-corrected  $P_L$  for each compound**

<b>Compound</b>	<b>Regression equations( log<math>P_L</math>)</b>	<b>References</b>
Acel	9.93-2855/T	(Odabasi et al., 2006)
Ace	10.17-2979/T	(Odabasi et al., 2006)
Flu	10.61-3233/T	(Odabasi et al., 2006)
Phe	11.43-3726/T	(Odabasi et al., 2006)
Ant	11.54-3780/T	(Lei et al., 2002)
Fla	12.47-4382/T	(Odabasi et al., 2006)
Pyr	11.7-4164/T	(Lei et al., 2002)
BaA	10.87-4269/T	(Lei et al., 2002)
Chr	13.87-5294/T	(Odabasi et al., 2006)
Bbf	12.48-5148/T	(Odabasi et al., 2006)
Bkf	12.5-5165/T	(Odabasi et al., 2006)
Bap	12.59-5252/T	(Odabasi et al., 2006)
IcdP	13.13-5691/T	(Odabasi et al., 2006)
DahA	13.31-5794/T	(Odabasi et al., 2006)
BghiP	13.15-5737/T	(Odabasi et al., 2006)

197

**Table S7. Concentrations of gas phase POPs (pg m<sup>-3</sup>) in the atmosphere of Nam Co**

198

**Part 1. Concentrations of gas phase OCPs and PCBs (pg m<sup>-3</sup>) in air:**

Start Date	End Date	$\alpha$ -HCH	$\beta$ -HCH	$\gamma$ -HCH	HCB	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	PCB-28	PCB-52	PCB-101	PCB-153	PCB-138	PCB-180
2012.09.03	2012.09.15	4.8	BDL	2.5	12.2	0.6	2.1	0.7	<b>0.2</b>	0.4	0.25	0.44	0.17	0.35	0.03
2012.09.16	2012.09.20	3.2	BDL	1.3	15.5	0.5	1.3	2.8	0.8	0.8	0.22	0.37	0.06	0.12	BDL
2012.10.24	2012.10.31	3.4	BDL	0.7	12.2	<b>0.17</b>	<b>0.35</b>	3.4	1.3	0.3	<b>0.05</b>	0.05	<b>0.02</b>	<b>0.02</b>	BDL
2012.11.01	2012.11.15	4.6	BDL	0.6	38.4	<b>0.17</b>	<b>0.35</b>	0.8	0.5	0.3	0.14	0.08	0.03	0.03	BDL
2012.11.16	2012.11.30	2.5	BDL	0.4	36.7	<b>0.17</b>	<b>0.35</b>	0.4	<b>0.17</b>	0.4	0.19	0.08	0.03	<b>0.02</b>	BDL
2012.12.01	2012.12.15	1.4	BDL	0.2	26.7	<b>0.17</b>	<b>0.35</b>	<b>0.17</b>	<b>0.17</b>	0.1	<b>0.05</b>	0.04	<b>0.02</b>	<b>0.02</b>	BDL
2012.12.16	2012.12.31	0.7	BDL	0.3	20.8	<b>0.17</b>	<b>0.35</b>	<b>0.17</b>	<b>0.17</b>	0.4	0.14	0.07	0.03	<b>0.02</b>	BDL
2013.01.03	2013.01.15	1.5	BDL	0.3	37.3	<b>0.17</b>	<b>0.35</b>	0.5	0.4	0.5	0.19	0.11	<b>0.02</b>	<b>0.02</b>	BDL
2013.01.16	2013.01.31	1.3	BDL	0.6	29.8	<b>0.17</b>	<b>0.35</b>	0.5	<b>0.17</b>	0.7	0.21	0.08	<b>0.02</b>	<b>0.02</b>	BDL
2013.02.06	2013.02.16	0.8	BDL	0.2	14.1	<b>0.17</b>	<b>0.35</b>	0.6	<b>0.17</b>	0.1	<b>0.05</b>	<b>0.02</b>	<b>0.02</b>	<b>0.02</b>	BDL
2013.02.21	2013.03.01	3.9	BDL	1.2	25.6	0.4	0.8	2.9	2.4	0.4	0.17	0.08	0.03	<b>0.02</b>	BDL
2013.03.02	2013.03.15	1.2	0.3	0.4	18.1	<b>0.17</b>	<b>0.35</b>	0.4	<b>0.17</b>	0.5	0.16	0.09	<b>0.02</b>	<b>0.02</b>	BDL
2013.03.15	2013.03.31	3.2	0.5	1.0	24.3	<b>0.17</b>	<b>0.35</b>	0.7	0.4	0.5	0.15	0.08	<b>0.02</b>	<b>0.02</b>	BDL
2013.04.01	2013.04.15	3.6	0.5	1.0	17.0	<b>0.17</b>	<b>0.35</b>	1.0	0.6	0.6	0.17	0.08	<b>0.02</b>	0.03	BDL
2013.04.15	2013.04.30	6.5	0.9	2.2	19.4	0.5	<b>0.35</b>	6.6	2.1	0.9	0.33	0.19	0.06	0.05	BDL
2013.05.01	2013.05.15	9.0	0.6	1.9	25.7	0.3	<b>0.35</b>	4.8	2.4	0.9	0.29	0.15	0.04	0.05	BDL
2013.05.15	2013.06.04	7.7	1.4	4.2	15.5	0.4	0.8	6.1	2.2	1.4	0.56	0.23	0.05	0.05	BDL
2013.06.06	2013.06.15	3.8	0.8	1.6	12.1	<b>0.17</b>	<b>0.35</b>	2.2	0.7	0.7	0.29	0.16	<b>0.02</b>	0.04	BDL
2013.06.15	2013.06.30	8.1	1.3	4.8	11.9	0.7	1.2	12.0	3.9	1.5	0.56	0.29	0.08	0.09	BDL
2013.07.01	2013.07.19	6.9	0.6	2.3	12.8	0.4	0.6	7.9	1.9	0.8	0.30	0.19	0.08	0.07	BDL
2013.07.19	2013.08.03	6.7	0.9	1.7	15.1	1.4	1.4	6.0	2.0	1.4	0.44	0.39	0.06	0.09	0.03
2013.08.04	2013.08.15	3.7	0.7	0.9	15.9	<b>0.17</b>	<b>0.35</b>	1.0	0.6	1.4	0.44	0.39	0.04	0.05	0.13
2013.08.15	2013.08.31	5.3	0.4	1.2	14.6	0.5	0.9	14.1	4.9	0.6	0.23	0.28	0.08	0.06	BDL

Start Date	End Date	$\alpha$ -HCH	$\beta$ -HCH	$\gamma$ -HCH	HCB	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	PCB-28	PCB-52	PCB-101	PCB-153	PCB-138	PCB-180
2013.09.01	2013.09.15	5.4	BDL	<b>0.06</b>	11.7	3.1	3.4	1.6	<b>0.17</b>	2.7	<b>0.05</b>	0.13	0.07	0.09	BDL
2013.10.01	2013.10.15	4.6	BDL	2.8	11.4	2.2	3.8	4.4	1.1	0.8	0.44	0.28	0.54	0.14	BDL
2013.10.15	2013.10.30	3.5	BDL	5.4	17.5	0.6	1.0	3.2	0.9	0.3	0.38	0.24	0.33	0.11	BDL
2013.11.01	2013.11.15	2.2	BDL	1.0	40.5	0.5	1.3	1.0	1.0	0.7	0.96	0.31	0.24	0.12	BDL
2013.11.15	2013.11.30	1.9	BDL	0.4	24.5	<b>0.17</b>	0.9	<b>0.17</b>	0.2	0.6	0.66	0.30	0.14	0.07	BDL
2013.12.01	2013.12.15	1.2	BDL	3.3	23.4	4.1	2.7	1.5	<b>0.17</b>	1.0	<b>0.05</b>	0.28	0.08	0.07	BDL
2013.12.16	2013.12.30	2.6	BDL	7.2	27.6	1.9	5.3	1.5	0.9	1.8	1.08	1.06	0.33	0.18	BDL
2014.01.05	2014.01.31	1.3	BDL	2.7	16.4	2.3	1.9	1.2	0.2	0.9	<b>0.05</b>	0.32	0.11	0.06	BDL
2014.02.01	2014.02.15	0.7	BDL	0.7	12.4	<b>0.17</b>	<b>0.35</b>	<b>0.17</b>	<b>0.17</b>	0.5	0.62	0.40	0.15	0.07	BDL
2014.02.15	2014.02.28	2.7	0.5	4.9	33.0	0.6	1.4	0.6	<b>0.17</b>	1.9	0.59	0.71	0.18	0.08	BDL
2014.03.01	2014.03.15	1.7	BDL	1.4	20.4	0.4	1.0	1.6	0.4	0.5	0.81	0.51	0.18	0.07	BDL
2014.03.15	2014.03.31	2.3	BDL	3.7	18.3	0.5	0.9	0.7	<b>0.17</b>	0.8	<b>0.05</b>	0.45	0.14	0.07	BDL
2014.04.01	2014.04.15	4.5	BDL	<b>0.06</b>	16.1	1.4	3.1	3.2	0.4	1.9	<b>0.05</b>	0.48	0.05	0.09	BDL
2014.04.15	2014.04.30	2.9	BDL	1.6	18.6	0.6	0.8	1.6	1.0	0.7	1.02	0.57	0.36	0.15	BDL
2014.05.01	2014.05.15	4.2	0.4	2.2	15.8	0.6	1.3	1.8	1.1	0.7	0.96	0.54	0.26	0.12	BDL
2014.05.15	2014.05.28	2.3	0.5	4.3	14.8	1.5	2.9	4.4	1.7	0.7	0.93	0.75	0.49	0.22	0.05
2014.06.02	2014.06.15	7.1	1.4	3.1	26.8	3.6	6.9	6.3	1.8	2.0	1.30	0.97	0.86	0.39	0.08
2014.06.22	2014.06.30	4.7	0.5	2.1	15.1	0.7	2.1	8.0	2.4	0.8	0.68	0.43	0.26	0.10	BDL
2014.07.01	2014.07.15	6.5	BDL	5.3	12.6	2.4	4.9	4.5	<b>0.17</b>	3.7	5.47	0.74	0.09	0.03	0.05
2014.07.15	2014.07.31	7.4	1.0	3.0	24.3	1.3	2.2	6.6	1.7	1.9	1.40	0.80	0.38	0.97	BDL
2014.08.01	2014.08.15	8.4	1.5	6.7	21.7	13.9	12.8	17.9	1.8	6.9	4.15	1.75	0.41	0.45	0.05
2014.08.15	2014.08.31	7.5	1.4	1.7	25.6	1.7	4.8	6.4	2.8	2.9	1.59	1.22	0.29	0.46	0.06
2014.09.02	2014.09.15	4.0	0.6	1.8	12.0	2.3	6.1	9.8	3.7	3.1	0.71	0.84	0.33	0.27	0.03
2014.09.15	2014.09.30	4.1	BDL	2.3	17.3	1.5	4.1	6.0	3.5	3.1	0.47	0.48	0.15	0.16	BDL

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

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## Part 2. Concentrations of gas phase PAHs (pg m<sup>-3</sup>) in air:

Start Date	End Date	Acel	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	Bap	IcdP	DahA	BghiP
2012.09.16	2012.09.20	52	123	607	1170	121	256	204	13	68	4	9	6	BDL	BDL	7
2012.10.01	2012.10.15	19	31	540	2037	347	383	223	3	18	1	1	1	BDL	BDL	1
2012.10.24	2012.10.31	8	21	115	264	24	51	30	2	6	1	1	1	BDL	BDL	1
2012.11.01	2012.11.15	4	6	85	267	20	65	40	3	8	1	3	2	BDL	BDL	2
2012.11.16	2012.11.30	5	7	80	291	21	52	31	2	5	0.3	1	1	BDL	BDL	1
2012.12.01	2012.12.15	4	6	85	252	19	53	33	3	7	0.3	1	1	BDL	BDL	1
2012.12.16	2012.12.31	14	55	273	701	118	274	167	9	12	2	2	1	BDL	BDL	2
2013.01.03	2013.01.15	11	37	189	328	32	59	38	4	10	2	4	2	BDL	BDL	4
2013.01.16	2013.01.31	27	62	544	1120	85	147	87	3	10	0.5	2	1	BDL	BDL	2
2013.02.06	2013.02.16	5	9	109	320	33	94	54	2	6	0.2	0.3	0.3	BDL	BDL	<b>0.17</b>
2013.02.21	2013.03.01	8	13	171	684	138	314	192	9	13	0.3	1	1	BDL	BDL	1
2013.03.02	2013.03.15	11	31	199	636	66	194	126	2	8	0.1	0.3	0.5	BDL	BDL	1
2013.03.15	2013.03.31	7	11	161	726	124	308	192	7	12	1	1	1	BDL	BDL	1
2013.04.01	2013.04.15	5	7	102	470	34	107	54	2	8	1	2	1	BDL	BDL	2
2013.04.15	2013.04.30	4	4	137	1048	195	453	274	10	16	0.4	1	1	BDL	BDL	1
2013.05.01	2013.05.15	4	5	102	540	69	148	120	32	53	9	22	31	1	1	28
2013.05.15	2013.06.04	4	5	98	817	97	269	157	8	17	1	3	3	BDL	BDL	3
2013.06.06	2013.06.15	3	4	96	694	181	415	262	14	18	1	1	1	BDL	BDL	1
2013.06.15	2013.06.30	4	5	147	1316	301	577	356	13	23	0.4	1	1	BDL	BDL	1
2013.07.01	2013.07.19	5	8	145	1016	225	530	350	14	24	2	1	1	BDL	BDL	2
2013.07.19	2013.08.03	19	44	286	817	100	178	150	13	27	2	6	7	BDL	BDL	5
2013.08.04	2013.08.15	44	87	675	1124	106	205	206	11	33	4	4	5	BDL	BDL	5
2013.08.15	2013.08.31	8	14	162	565	90	124	80	2	11	0.3	1	0.3	BDL	BDL	1
2013.09.01	2013.09.15	170	226	2068	3844	382	176	813	6	34	2	5	4	3	BDL	7
2013.10.01	2013.10.15	10	20	206	829	90	162	145	2	11	2	1	1	0.4	1	

<b>Start Date</b>	<b>End Date</b>	<b>Acel</b>	<b>Ace</b>	<b>Flu</b>	<b>Phe</b>	<b>Ant</b>	<b>Fla</b>	<b>Pyr</b>	<b>BaA</b>	<b>Chr</b>	<b>Bbf</b>	<b>Bkf</b>	<b>Bap</b>	<b>IcdP</b>	<b>DahA</b>	<b>BghiP</b>
2013.10.15	2013.10.30	3	6	142	453	33	106	60	7	14	9	7	5	4	1	3
2013.11.01	2013.11.15	14	22	265	550	31	75	40	2	8	2	1	0.4	1	BDL	1
2013.11.15	2013.11.30	15	25	236	613	39	115	63	3	8	2	2	0.4	1	BDL	1
2013.12.01	2013.12.15	37	71	491	659	67	58	80	4	8	3	2	1	2	0.4	2
2013.12.16	2013.12.30	53	92	709	1429	112	176	132	10	21	8	6	5	6	1	6
2014.01.05	2014.01.31	34	58	422	758	79	70	78	3	8	2	1	2	1	BDL	2
2014.02.01	2014.02.15	4	6	137	518	24	84	42	2	8	2	1	1	1	BDL	1
2014.02.15	2014.02.28	43	67	547	969	55	70	72	3	9	3	2	1	1	0.3	2
2014.03.01	2014.03.15	4	9	153	590	44	173	86	4	11	3	3	1	1	0.3	2
2014.03.15	2014.03.31	8	18	146	389	23	72	54	1	4	1	1	0.4	0.5	BDL	1
2014.04.01	2014.04.15	56	80	750	1454	119	148	182	9	20	5	5	5	6	1	7
2014.04.15	2014.04.30	10	18	165	436	93	112	68	4	9	3	2	1	1	0.3	2
2014.05.01	2014.05.15	8	16	124	449	40	111	65	3	10	3	3	1	1	0.4	2
2014.05.15	2014.05.28	5	8	100	540	62	178	111	13	28	18	13	9	7	1	7
2014.06.02	2014.06.15	11	15	236	989	107	154	119	19	32	27	13	19	9	2	9
2014.06.22	2014.06.30	15	19	586	1370	173	190	110	3	11	0.4	2	1	BDL	BDL	1
2014.07.01	2014.07.15	118	159	1632	4270	381	104	338	4	68	0.5	2	2	0.5	BDL	3
2014.07.15	2014.07.31	36	45	716	1687	199	176	111	2	11	0.3	1	1	0.3	BDL	2
2014.08.01	2014.08.15	198	311	2580	7057	1310	760	946	22	79	14	8	13	7	2	11
2014.08.15	2014.08.31	16	28	743	4716	1137	1621	1242	269	698	29	<b>0.08</b>	<b>0.08</b>	8	9	<b>0.17</b>

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

203 **Table S8. Concentrations of POPs (pg m<sup>-3</sup>) in the total suspended particulates (TSP) of Nam Co**204 **Part 1. Concentrations of particulate phase OCPs and PCBs (pg m<sup>-3</sup>) in air:**

Start Date	End Date	$\alpha$ -HCH	$\beta$ -HCH	$\gamma$ -HCH	HCB	$\text{o,p}'$ -DDE	$\text{p,p}'$ -DDE	$\text{o,p}'$ -DDT	$\text{p,p}'$ -DDT	PCB-28	PCB-52	PCB-101	PCB-153	PCB-138	PCB-180
2012.09.03	2012.09.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2012.09.16	2012.09.20	BDL	BDL	0.20	3.14	BDL	BDL	BDL	BDL	0.06	BDL	0.13	BDL	BDL	BDL
2012.10.01	2012.10.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2012.10.24	2012.10.31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2012.11.01	2012.11.15	BDL	BDL	0.10	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2012.11.16	2012.11.30	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2012.12.16	2012.12.31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.05	BDL	0.05	BDL	BDL	BDL
2013.01.03	2013.01.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.06	BDL	0.06	BDL	BDL	BDL
2013.01.16	2013.01.31	BDL	BDL	BDL	BDL	BDL	BDL	0.38	0.33	0.09	BDL	0.04	BDL	BDL	BDL
2013.02.06	2013.02.16	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2013.02.21	2013.03.01	BDL	BDL	0.16	BDL	BDL	BDL	BDL	0.55	BDL	BDL	0.04	BDL	BDL	BDL
2013.03.02	2013.03.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	0.04	BDL	BDL	BDL
2013.03.15	2013.03.31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2013.04.01	2013.04.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2013.05.01	2013.05.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2013.05.15	2013.06.04	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2013.06.06	2013.06.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2013.06.15	2013.06.30	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.04	BDL	<b>0.02</b>	BDL	BDL	BDL
2013.07.01	2013.07.19	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.04	BDL	BDL	BDL
2013.07.19	2013.08.03	0.40	BDL	0.14	3.71	BDL	BDL	0.43	BDL	BDL	BDL	0.04	BDL	0.08	0.03
2013.09.01	2013.09.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.60	BDL	BDL	0.04	0.05	BDL	BDL
2013.10.01	2013.10.15	BDL	BDL	BDL	BDL	BDL	BDL	1.01	1.58	2.38	BDL	BDL	0.04	0.04	BDL
2013.10.15	2013.10.30	BDL	BDL	0.34	BDL	BDL	BDL	0.47	BDL	BDL	BDL	0.03	BDL	BDL	BDL

<b>Start Date</b>	<b>End Date</b>	<b><math>\alpha</math>-HCH</b>	<b><math>\beta</math>-HCH</b>	<b><math>\gamma</math>-HCH</b>	<b>HCB</b>	<b><math>o,p'</math>-DDE</b>	<b>p,p'-DDE</b>	<b><math>o,p'</math>-DDT</b>	<b>p,p'-DDT</b>	<b>PCB-28</b>	<b>PCB-52</b>	<b><i>PCB-101</i></b>	<b>PCB-153</b>	<b>PCB-138</b>	<b>PCB-180</b>
2013.11.01	2013.11.15	BDL	BDL	BDL	BDL	BDL	BDL	0.39	0.43	BDL	BDL	0.05	0.03	0.03	BDL
2013.11.15	2013.11.30	BDL	BDL	BDL	BDL	BDL	BDL	0.43	BDL	0.03	BDL	0.08	0.09	0.04	BDL
2013.12.01	2013.12.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.09	0.11	0.04	BDL
2013.12.16	2013.12.30	BDL	BDL	BDL	BDL	BDL	0.70	BDL	BDL	BDL	BDL	0.14	0.16	0.08	0.04
2014.01.05	2014.01.31	BDL	BDL	BDL	BDL	BDL	BDL	0.36	0.58	BDL	BDL	0.04	BDL	BDL	BDL
2014.02.01	2014.02.15	BDL	BDL	0.16	BDL	BDL	1.31	0.93	0.51	0.08	BDL	0.12	0.07	0.05	BDL
2014.02.15	2014.02.28	BDL	BDL	BDL	BDL	BDL	BDL	0.85	0.66	0.03	BDL	0.07	0.09	0.04	BDL
2014.03.01	2014.03.15	BDL	BDL	BDL	BDL	BDL	BDL	0.91	1.01	0.03	BDL	0.07	0.17	0.06	BDL
2014.03.15	2014.03.31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.04	0.05	BDL	BDL
2014.04.01	2014.04.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.04	0.04	BDL	BDL
2014.04.15	2014.04.30	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2014.05.01	2014.05.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2014.05.15	2014.05.28	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2014.06.02	2014.06.15	BDL	0.47	0.42	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.05	0.06	BDL	BDL
2014.06.22	2014.06.30	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2014.07.01	2014.07.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2014.07.15	2014.07.31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2014.08.01	2014.08.15	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL
2014.08.15	2014.08.31	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.03	BDL	BDL	BDL
2014.09.02	2014.09.15	BDL	BDL	0.10	BDL	BDL	BDL	BDL	BDL	BDL	BDL	<b>0.02</b>	BDL	BDL	BDL

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

## Part 2. Concentrations of particulate phase PAHs (pg m<sup>-3</sup>) in air:

Start Date	End Date	Acel	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	Bap	IcdP	DahA	BghiP
2012.09.03	2012.09.15	1	1	51	519	33	360	239	11	19	2	4	3	BDL	BDL	8
2012.09.16	2012.09.20	4	6	148	1392	216	966	606	34	45	3	5	3	BDL	BDL	7
2012.10.01	2012.10.15	2	2	58	525	75	428	287	20	38	4	16	17	BDL	BDL	29
2012.10.24	2012.10.31	2	2	58	571	70	463	307	23	49	5	19	20	BDL	BDL	30
2012.11.01	2012.11.15	2	3	58	552	76	411	285	21	44	1	21	21	BDL	BDL	28
2012.11.16	2012.11.30	BDL	1	16	74	4	56	43	8	20	<b>0.17</b>	9	10	BDL	BDL	13
2012.12.16	2012.12.31	2	2	26	123	8	82	61	11	25	2	11	12	BDL	BDL	15
2013.01.03	2013.01.15	4	6	74	306	15	198	159	27	58	3	30	27	1	BDL	39
2013.01.16	2013.01.31	2	3	33	156	7	95	69	11	25	1	11	12	BDL	BDL	16
2013.02.06	2013.02.16	BDL	2	18	95	7	110	101	28	61	11	34	36	3	1	37
2013.02.21	2013.03.01	BDL	1	17	105	7	86	75	18	40	3	19	22	BDL	BDL	30
2013.03.02	2013.03.15	BDL	2	60	676	90	618	464	70	146	6	59	74	BDL	1	87
2013.03.15	2013.03.31	BDL	2	31	173	12	124	106	23	59	3	28	33	BDL	BDL	45
2013.04.01	2013.04.15	BDL	1	19	106	11	71	58	13	28	21	21	18	18	2	24
2013.05.01	2013.05.15	BDL	1	21	117	10	61	49	10	25	21	16	17	17	1	27
2013.05.15	2013.06.04	BDL	1	21	116	8	67	52	10	25	19	17	12	16	2	22
2013.06.06	2013.06.15	BDL	1	18	103	10	57	46	9	22	17	15	10	12	1	18
2013.06.15	2013.06.30	BDL	2	27	148	10	69	50	8	17	2	8	10	BDL	BDL	15
2013.07.01	2013.07.19	BDL	1	24	125	6	48	33	4	11	2	4	3	0.5	BDL	6
2013.07.19	2013.08.03	BDL	1	39	379	61	179	125	11	46	2	8	8	BDL	BDL	14
2013.09.01	2013.09.15	BDL	2	25	116	7	74	51	9	25	23	10	8	12	2	15
2013.10.01	2013.10.15	BDL	<b>0.57</b>	19	106	5	105	82	16	62	58	26	23	34	4	38
2013.10.15	2013.10.30	BDL	2	26	123	5	74	54	8	20	17	17	14	16	1	17
2013.11.01	2013.11.15	BDL	2	20	104	5	87	64	12	28	8	14	16	1	1	20
2013.11.15	2013.11.30	BDL	1	20	139	4	197	116	15	41	<b>0.17</b>	17	20	BDL	BDL	23

<b>Start Date</b>	<b>End Date</b>	<b>Acel</b>	<b>Ace</b>	<b>Flu</b>	<b>Phe</b>	<b>Ant</b>	<b>Fla</b>	<b>Pyr</b>	<b>BaA</b>	<b>Chr</b>	<b>Bbf</b>	<b>Bkf</b>	<b>Bap</b>	<b>IcdP</b>	<b>DahA</b>	<b>BghiP</b>
2013.12.01	2013.12.15	BDL	1	18	115	5	104	68	9	22	2	9	11	BDL	BDL	11
2013.12.16	2013.12.30	2	2	32	117	6	80	54	8	25	<b>0</b>	8	10	BDL	BDL	10
2014.01.05	2014.01.31	BDL	<b>0.57</b>	15	96	5	100	75	13	31	<b>0</b>	15	18	BDL	BDL	20
2014.02.01	2014.02.15	BDL	1	24	180	9	186	93	12	27	<b>0.17</b>	11	13	BDL	BDL	12
2014.02.15	2014.02.28	3	3	49	184	7	151	105	21	48	<b>0.17</b>	24	26	BDL	BDL	30
2014.03.01	2014.03.15	BDL	1	20	95	4	94	76	18	45	8	22	29	1	1	30
2014.03.15	2014.03.31	BDL	1	28	125	6	95	77	16	41	29	21	24	24	2	28
2014.04.01	2014.04.15	BDL	<b>0.57</b>	9	43	2	20	14	2	5	3	2	2	2	BDL	3
2014.04.15	2014.04.30	BDL	<b>0.57</b>	9	46	5	37	32	8	20	16	12	11	12	1	12
2014.05.01	2014.05.15	BDL	<b>0.57</b>	5	28	2	21	17	4	12	11	8	8	7	0.4	9
2014.05.15	2014.05.28	BDL	<b>0.57</b>	9	55	2	26	18	2	8	5	3	3	2	BDL	3
2014.06.02	2014.06.15	BDL	2	13	54	2	24	17	2	7	4	2	2	1	BDL	3
2014.06.22	2014.06.30	BDL	<b>0.57</b>	10	53	2	22	15	2	5	3	2	2	1	BDL	2
2014.07.01	2014.07.15	BDL	<b>0.57</b>	13	65	8	33	22	3	10	4	3	2	1	BDL	3
2014.07.15	2014.07.31	BDL	<b>0.57</b>	15	75	7	34	23	4	15	8	6	2	3	BDL	5
2014.08.01	2014.08.15	BDL	<b>0.57</b>	24	110	7	48	29	3	7	4	2	0.4	1	BDL	2
2014.08.15	2014.08.31	BDL	<b>0.57</b>	9	49	2	20	12	1	3	1	1	<b>0.08</b>	0.2	BDL	1
2014.09.02	2014.09.15	BDL	<b>0.57</b>	13	60	9	23	18	2	5	3	3	2	0.3	BDL	3

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

208 **Table S9. Concentrations of dissolved phase POPs (pg L<sup>-1</sup>) in the lake water of Nam Co (after corrected by DOC)**209 **Part 1. Concentrations of dissolved phase OCPs and PCBs (pg L<sup>-1</sup>) in the water from different sites of the lake (S1-S15):**

	<b>S1</b>	<b>S2</b>	<b>S3</b>	<b>S4</b>	<b>S5</b>	<b>S6</b>	<b>S7</b>	<b>S8</b>	<b>S9</b>	<b>S10</b>	<b>S11</b>	<b>S12</b>	<b>S13</b>	<b>S14</b>	<b>S15</b>
<b><math>\alpha</math>-HCH</b>	5.0	11.8		8.0	9.5	7.5	8.7	9.3	9.3	9.8	6.3	14.7	11.5	8.6	18.4
<b><math>\beta</math>-HCH</b>	79.9	104.2		100.3	105.5	116.9	77.9	76.2	70.3	71.7	44.0	100.2	82.7	85.2	77.2
<b><math>\gamma</math>-HCH</b>	5.4	7.5		5.8	10.2	8.2	6.7	7.3	3.7	4.4	2.7	9.0	7.5	8.0	11.6
<b>HCB</b>	2.9	4.3		4.7	3.7	5.1	5.3	15.1	6.6	3.3	4.3	5.8	18.0	7.2	19.6
<b><i>o,p'</i>-DDE</b>	BDL	BDL		BDL	BDL	BDL	BDL	BDL	BDL						
<b><i>p,p'</i>-DDE</b>	BDL	BDL		BDL	BDL	0.9	BDL	BDL	1.2						
<b><i>o,p'</i>-DDT</b>	BDL	BDL		BDL	BDL	BDL	BDL	BDL	BDL						
<b><i>p,p'</i>-DDT</b>	BDL	BDL		BDL	BDL	BDL	BDL	BDL	BDL						
<b>PCB-28</b>	2.1	1.8		1.6	1.6	1.6	0.9	1.2	1.8	0.9	1.0	1.8	1.0	2.9	4.3
<b>PCB-52</b>	0.3	0.2		BDL	BDL	0.2	BDL	1.6	1.0						
<b>PCB-101</b>	BDL	BDL		BDL	BDL	BDL	BDL	BDL	0.3						
<b>PCB-153</b>	BDL	BDL		BDL	BDL	BDL	BDL	BDL	BDL						
<b>PCB-138</b>	BDL	BDL		BDL	BDL	0.03	BDL	BDL	0.2						
<b>PCB-180</b>	BDL	BDL		BDL	BDL	BDL	BDL	BDL	BDL						

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**Part 2. Concentrations of dissolved phase PAHs (pg L<sup>-1</sup>) in the water from different sites of the lake (S1-S15):**

	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12	S13	S14	S15
<b>Acel</b>	338	685		685	291	543	571	309	1326	411	283		539	1076	990
<b>Ace</b>	274	244		282	167	183	150	102	236	155	103		195	468	815
<b>Flu</b>	1549	1833		1726	1742	1638	1376	2148	2801	1646	1619		3275	4219	4118
<b>Phe</b>	2515	4311		5065	5497	7423	5732	13822	73236	5791	7499		15907	9883	17748
<b>Ant</b>	BDL	1130		1178	1301	1500	1176	5159	1829	1045	1978		6781	3035	7778
<b>Fla</b>	934	1865		2118	1984	3314	2883	10946	2179	3085	4397		12413	5525	14099
<b>Pyr</b>	751	1266		1390	1311	1553	1293	7633	1589	2445	3585		8871	3738	10669
<b>BaA</b>	48	55		105	87	94	70	380	98	199	302		337	134	401
<b>Chr</b>	484	265		358	291	521	446	514	255	518	595		463	469	2078
<b>Bbf</b>	BDL	BDL		BDL	6	5	BDL	17	BDL	BDL	16		3	2	BDL
<b>Bkf</b>	15	14		52	32	42	36	89	41	88	97		18	15	62
<b>Bap</b>	3	7		27	19	17	9	84	34	22	78		11	8	38
<b>IcdP</b>	BDL	BDL		BDL		BDL	BDL	BDL							
<b>DahA</b>	BDL	BDL		BDL		BDL	BDL	BDL							
<b>BghiP</b>	7	7		19	12	15	7	51	16	24	40		11	4	19
<b>Σ<sub>15</sub>PAHs (pg L<sup>-1</sup>)</b>	6918	11680		13005	12739	16848	13747	41255	83641	15429	20592		48825	28575	58817
<b>Σ<sub>15</sub>PAHs (ng L<sup>-1</sup>)</b>	7	12		13	13	17	14	41	84	15	21		49	29	59

211 Part 3. Concentrations of dissolved phase OCPs and PCBs (pg L<sup>-1</sup>) in different months from May to September:

	<i><b>α-HCH</b></i>	<i><b>β-HCH</b></i>	<i><b>γ-HCH</b></i>	<b>HCB</b>	<i><b>o,p'-DDE</b></i>	<i><b>p,p'-DDE</b></i>	<i><b>o,p'-DDT</b></i>	<i><b>p,p'-DDT</b></i>	<b>PCB-28</b>	<b>PCB-52</b>	<b>PCB-101</b>	<b>PCB-153</b>	<b>PCB-138</b>	<b>PCB-180</b>
<b>May-1</b>	6.0	44.7	4.1	5.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>May-2</b>	4.4	35.2	2.6	<b>0.7</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>May-3</b>	4.0	32.3	2.4	1.5	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>June-1</b>	12.5	83.0	8.5	4.4	BDL	BDL	BDL	BDL	0.2	BDL	BDL	BDL	BDL	BDL
<b>June-2</b>	5.8	62.0	4.1	<b>0.7</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>June-3</b>	7.0	58.0	3.8	2.8	BDL	BDL	0.3	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>July-1</b>	0.7	60.0	4.6	1.8	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>July-2</b>	11.3	66.9	5.5	1.9	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>July-3</b>	9.7	66.0	4.8	2.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
<b>Aug-1</b>	7.2	45.4	4.0	1.7	BDL	BDL	BDL	BDL	0.3	BDL	BDL	BDL	BDL	BDL
<b>Aug-2</b>	7.2	66.8	4.6	<b>0.7</b>	BDL	BDL	BDL	BDL	0.2	BDL	BDL	BDL	BDL	BDL
<b>Aug-3</b>	9.8	83.2	4.3	1.8	BDL	BDL	BDL	BDL	0.4	BDL	BDL	BDL	BDL	BDL
<b>Sep-1</b>	10.6	108.9	7.2	<b>0.7</b>	BDL	0.6	BDL	0.4	1.7	BDL	BDL	BDL	BDL	BDL
<b>Sep-2</b>	9.2	93.0	5.1	<b>0.7</b>	BDL	BDL	BDL	BDL	0.9	BDL	BDL	BDL	BDL	BDL
<b>Sep-3</b>	3.5	32.3	1.8	<b>0.7</b>	BDL	BDL	BDL	BDL	0.2	BDL	BDL	BDL	BDL	BDL

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

213 Part 4. Concentrations of dissolved phase PAHs ( $\text{pg L}^{-1}$ ) in different months from May to September:

	Acel	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	Bap	IcdP	DahA	BghiP	$\sum_{15}\text{PAHs}$ ( $\text{pg L}^{-1}$ )	$\sum_{15}\text{PAHs}$ ( $\text{ng L}^{-1}$ )	
<b>May-1</b>	64	116	1510	2755	BDL	BDL	264	BDL	BDL	BDL	3	1	BDL	BDL	1	4715	4.7	
<b>May-2</b>	35	74	771	1677	BDL	BDL	226	BDL	BDL	BDL	4	3	1	BDL	BDL	1	2793	2.8
<b>May-3</b>	87	105	697	1608	BDL	BDL	172	BDL	BDL	BDL	2	2	1	BDL	BDL	1	2674	2.7
<b>June-1</b>	309	220	1913	3358	BDL	BDL	352	BDL	26	BDL	4	2	BDL	BDL	2	6187	6.2	
<b>June-2</b>	36	76	560	1153	BDL	BDL	<b>74</b>	BDL	BDL	BDL	1	1	BDL	BDL	1	1901	1.9	
<b>June-3</b>	186	131	885	1501	BDL	BDL	<b>74</b>	BDL	BDL	BDL	1	1	BDL	BDL	1	2779	2.8	
<b>July-1</b>	138	106	844	1034	BDL	BDL	<b>74</b>	BDL	BDL	BDL	1	0	1	BDL	BDL	1	2198	2.2
<b>July-2</b>	314	196	1744	2211	BDL	BDL	150	BDL	BDL	BDL	1	1	BDL	BDL	1	4617	4.6	
<b>July-3</b>	214	120	1562	2142	BDL	BDL	167	BDL	BDL	BDL	2	1	1	BDL	BDL	1	4211	4.2
<b>Aug-1</b>	281	131	900	1281	BDL	BDL	<b>74</b>	BDL	BDL	BDL	2	1	1	BDL	BDL	1	2673	2.7
<b>Aug-2</b>	316	140	1233	1578	BDL	BDL	<b>74</b>	BDL	BDL	BDL	3	3	1	BDL	BDL	1	3349	3.3
<b>Aug-3</b>	463	193	1226	1645	BDL	BDL	<b>74</b>	BDL	BDL	BDL	3	2	1	BDL	BDL	1	3608	3.6
<b>Sep-1</b>																		
<b>Sep-2</b>	448	188	1529	2120	1430	BDL	226	BDL	26	BDL	0	2	BDL	BDL	2	5972	6.0	
<b>Sep-3</b>	315	166	1220	1779	BDL	BDL	<b>74</b>	BDL	BDL	BDL	2	1	2	BDL	BDL	0	3560	3.6

214 Entries in bold and italics are substituted values at 1/2 MDL.

215 **Table S10. Concentrations of particulate phase POPs (pg L<sup>-1</sup>) in the SPM of the water samples from Nam Co Lake**216 **Part 1. Concentrations of particulate phase OCPs and PCBs (pg L<sup>-1</sup>) in the lake water:**

	$\alpha$ -HCH	$\beta$ -HCH	$\gamma$ -HCH	HCB	$\text{o,p}'$ -DDE	$\text{p,p}'$ -DDE	$\text{o,p}'$ -DDT	$\text{p,p}'$ -DDT	PCB-28	PCB-52	PCB-101	PCB-153	PCB-138	PCB-180	PCBs
<b>S1</b>	BDL	BDL	0.5	BDL	BDL	2.3	BDL	BDL	1.3	0.7	0.8	0.3	0.7	0.1	3.9
<b>S2</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.3	BDL	0.4	BDL	0.5	BDL	1.3
<b>S3</b>	BDL	1.1	BDL	BDL	BDL	BDL	BDL	BDL	0.5	BDL	0.2	BDL	BDL	BDL	0.7
<b>S4</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.2	BDL	0.3	BDL	0.1	BDL	0.6
<b>S5</b>	BDL	BDL	0.5	BDL	BDL	BDL	BDL	BDL	0.8	0.3	0.2	BDL	BDL	BDL	1.3
<b>S6</b>	BDL	BDL	0.7	BDL	BDL	BDL	BDL	BDL	0.6	BDL	0.3	BDL	0.2	BDL	1.1
<b>S7</b>	BDL	BDL	0.5	BDL	BDL	BDL	BDL	BDL	0.5	BDL	0.3	BDL	0.1	BDL	0.9
<b>S8</b>	BDL	BDL	0.4	BDL	BDL	BDL	BDL	BDL	0.5	0.2	0.3	BDL	0.2	BDL	1.3
<b>S9</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.4	BDL	0.2	BDL	BDL	BDL	0.6
<b>S10</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.2	0.3	0.4	BDL	0.2	BDL	1.1
<b>S11</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	1.8	0.2	BDL	0.2	BDL	BDL	0.4
<b>S12</b>	BDL	BDL	0.6	BDL	BDL	BDL	BDL	BDL	0.4	0.3	0.5	0.1	0.4	BDL	1.7
<b>S13</b>	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.3	BDL	0.3	BDL	BDL	BDL	0.5
<b>S14</b>	BDL	2.2	3.3	BDL	BDL	BDL	BDL	BDL	7.3	BDL	1.6	BDL	0.8	BDL	9.8
<b>S15</b>	BDL	BDL	1.0	BDL	BDL	BDL	BDL	BDL	0.7	0.5	0.7	0.2	0.7	0.1	3.0

**Part 2. Concentrations of particulate phase PAHs ( $\text{pg L}^{-1}$ ) in the lake water:**

	Acel	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	Bbf	Bkf	Bap	IcdP	DahA	BghiP	$\sum_{15}\text{PAHs}$ ( $\text{pg L}^{-1}$ )	$\sum_{15}\text{PAHs}$ ( $\text{ng L}^{-1}$ )
<b>S1</b>	12	29	145	641	136	927	777	240	980	114	185	232	8	20	297	4742.7	4.7
<b>S2</b>	11	27	161	727	107	549	442	107	248	BDL	102	81	BDL	BDL	140	2700.2	2.7
<b>S3</b>	9	19	128	639	107	338	229	38	141	BDL	31	25	BDL	BDL	28	1732.6	1.7
<b>S4</b>	13	37	169	870	170	1240	1052	406	768	BDL	277	380	6	BDL	336	5723.1	5.7
<b>S5</b>	46	135	387	9800	151	613	488	64	162	38	40	57	BDL	BDL	50	12030.6	12.0
<b>S6</b>	16	42	352	1607	329	920	717	187	521	32	165	184	BDL	BDL	241	5312.4	5.3
<b>S7</b>																	
<b>S8</b>	9	11	135	787	232	2185	2005	830	1380	BDL	605	827	19	BDL	910	9935.9	9.9
<b>S9</b>	11	17	111	597	121	796	665	281	561	BDL	266	363	8	BDL	396	4192.7	4.2
<b>S10</b>	9	15	252	1696	699	5404	4308	2835	4159	BDL	2050	3242	31	47	3437	28184.3	28.2
<b>S11</b>	6	11	188	1155	301	2163	1818	1025	1652	BDL	620	1052	BDL	9	1105	11104.4	11.1
<b>S12</b>	5	8	139	972	148	563	441	54	269	BDL	58	65	BDL	BDL	74	2794.8	2.8
<b>S13</b>	6	13	122	668	121	379	237	43	103	5	25	35	2	BDL	29	1788.2	1.8
<b>S14</b>	28	56	266	1517	316	1296	1299	149	1432	BDL	217	331	BDL	BDL	328	7235.7	7.2
<b>S15</b>	9	23	216	1313	204	753	628	49	599	88	23	92	BDL	BDL	120	4117.8	4.1

218

**Table S11. Comparisons of OCPs and PCBs in the air of this study and data reported for other remote areas ( $\text{pg m}^{-3}$ )**

Location	Nam Co Lake, TP	Nam Co, TP	Lhasa, TP	Lulang, southeast TP	Mt. Everest	Rocky Mountains	Central Pyrenees	Antarctic	Alert, Arctic
<b>Sampling Year</b>	2012-2014	2006-2008	2006-2007	2008-2011	2002	2000	2001-2003	2007-2010	2000-2003
<b>Sampler</b>	AAS	FTS	AAS	AAS	AAS	AAS	AAS	AAS	AAS
<b><math>\alpha</math>-HCH</b>	4 (0.7-9)	48.7 (4.0-144)	2.3 (BDL-9.2)	12.1 (0.61-51)	19.2	21	8	0.2	22 (1.4-66)
<b><math>\beta</math>-HCH</b>	0.8 (BDL-1.5)				7.7				0.1 (0.018-1.0)
<b><math>\gamma</math>-HCH</b>	2.1 (0.1-7.2)	7.9 (0.9-19.9)	10.3 (3.7-20.9)	(BDL-7.07)		2.3	22		5.6 (0.43-19)
<b>HCB</b>	20 (11.4-40.5)	17.1 (7.3-31.7)		7.87 (0.05-27.1)	8.9	42	49	22.9	64 (20-130) 0.089
<b><i>o,p'</i>-DDE</b>	1.2 (0.2-13.9)	0.6 (BDL-0.99)		(BDL-7.3)				0.03 (0.013-0.36)	
<b><i>p,p'</i>-DDE</b>	1.9 (0.3-12.8)	0.6 (BDL-1.0)	2.9 (BDL-12.1)	6.4 (0.08-23.8)	5.1		33		0.41 (0.051-3.6)
<b><i>o,p'</i>-DDT</b>	3.6 (0.2-17.9)	8.2 (0.3-19.7)	5.8 (BDL-34.3)	(BDL-60.5)	5.1				0.14 (0.016-0.56)
<b><i>p,p'</i>-DDT</b>	1.2 (0.2-4.9)	3.9 (0.3-12.67)	3.2 (BDL-9.7)	(BDL-33.6)	3.7		BDL		0.09 (0.015-1.1)
<b><math>\Sigma</math>PCBs</b>	2.5 (0.2-13.7)	1.4 (0.1-2.6)	10.6 (3.4-45.4)	(BDL-16.7)			34	0.5	
<b>Reference</b>	This study	(Xiao et al., 2010)	(Gong et al., 2010)	(Sheng et al., 2013)	(Li et al., 2006)	(Wilkinson et al., 2005)	(Van Drooge et al., 2004)	(Kallenborn et al., 2013)	(Su et al., 2006 and 2008)

219

AAS: active air sampler; FTS: flow-through sampler.

220  
221**Table S12. Comparisons of PAHs concentrations from this study and data reported for other remote areas (ng m<sup>-3</sup>)**

Location	Sampling Year	Sampler	Gaseous (G)/ Particulate (P)	$\Sigma$ PAHs (ng/m <sup>3</sup> )	Reference
Nam Co, central TP	2012-2014	AAS	G	2.2 (0.5-13)	This study
			P	0.6 (0.1-3.4)	
Nam Co, central TP	2006-2008	FTS	G+P	0.7 (0.08-2.2)	(Xiao et al., 2010)
Lulang, southeast TP	2008-2011	AAS	P	2.0 (0.2-5)	(Wang et al., 2015)
Lahsa, TP	2006-2007	AAS	G+P	35.7 (11.4-72.5)	(Gong et al., 2011)
Pyrenees, Europe	1996-1997	AAS	G	(1.3-2.6)	(Fernandez et al., 2002)
Alps, Europe	1996-1997	AAS	G	(2.7-3.7)	
Open water of Lake Superior	2011	PE-PAS	G	(BDL-1.9)	(Ruge et al., 2015)
Mediterranean Sea	2006-2007	AAS	G	38.38 (25.84-86.47)	(Castro et al., 2012)
			P	1.4 (0.9-1.8)	
Pacific and Arctic Ocean	2010	AAS	G	(0.91-7.4)	(Ma et al., 2013)
			P	(0.0002-0.36)	
Atlantic Ocean	2005	AAS	G+P	0.6 (0.02-2.8)	(Nizzetto et al., 2008)
Terra Nova Bay, Antarctica	2009-2010	AAS	G	0.004 (0.0004-0.01)	(Piazza et al., 2013)
			P	0.13 (0.02-0.3)	
Arctic	2003	AAS	G	3.3 (0.9-6.3)	(Ding et al., 2007b)
			P	0.5 (0.02-2.6)	

222

AAS: active air sampler; PE-PAS: polyethylene passive air sampler; FTS: flow-through sampler.

**Table S13. Dissolved concentrations of OCPs and PCBs in lake water of Nam Co Lake and other remote sites (pg L<sup>-1</sup>).**

<b>Location</b>	<b>Year</b>	<b><math>\alpha</math>-HCH</b>	<b><math>\beta</math>-HCH</b>	<b><math>\gamma</math>-HCH</b>	<b>HCB</b>	<b><i>o,p'</i>-DDE</b>	<b><i>p,p'</i>-DDE</b>	<b><i>o,p'</i>-DDT</b>	<b><i>p,p'</i>-DDT</b>	<b><math>\sum_6</math>PCBs</b>	<b>Reference</b>
Nam Co Lake	2013	9.9±3.4	85.2±18.8	7±2.5	7.6±5.6	BDL	BDL-1.2	BDL	BDL	2.0±1.4	This study
Yamdro, TP	2002	850	2400				260				(Zhang et al., 2003)
Co Ngoin, TP	2002	550	810						190		(Zhang et al., 2003)
Taihu Lake, China	2004-2005	1887±1372	1773±944	484±373		53±113	77±91	135±287	12±4		(Qiu et al., 2008)
Chaochu Lake, China	2010-2011	460±460	900±480	280±310	170±70	1640±1820	20±40	140±390	220±220		(Ouyang et al., 2013)
European mountain lake	2000	68±35		139±89	8.5±3.2			12±3			(Fernandez et al., 2005)
North Pacific	2010	101.6	27.7	17.9	2.8						(Cai et al., 2012)
Indian Ocean	2011	3.2±0.8	5.4±2.2	2.2±1.3		48±120	37±85	330±840	1600±4000		(Huang et al., 2013)
Ross Sea, Antarctic	2003-2004	1.4±0.9		2.9±2.2							(Cincinelli et al., 2009)
North Atlantic and Arctic	2004	13.0±16.3		4.7±5.6	0.9-9.6	BDL	0.15±0.08	BDL	BDL-0.2		(Lohmann et al., 2009)
										<1	(Gioia et al., 2008)

225

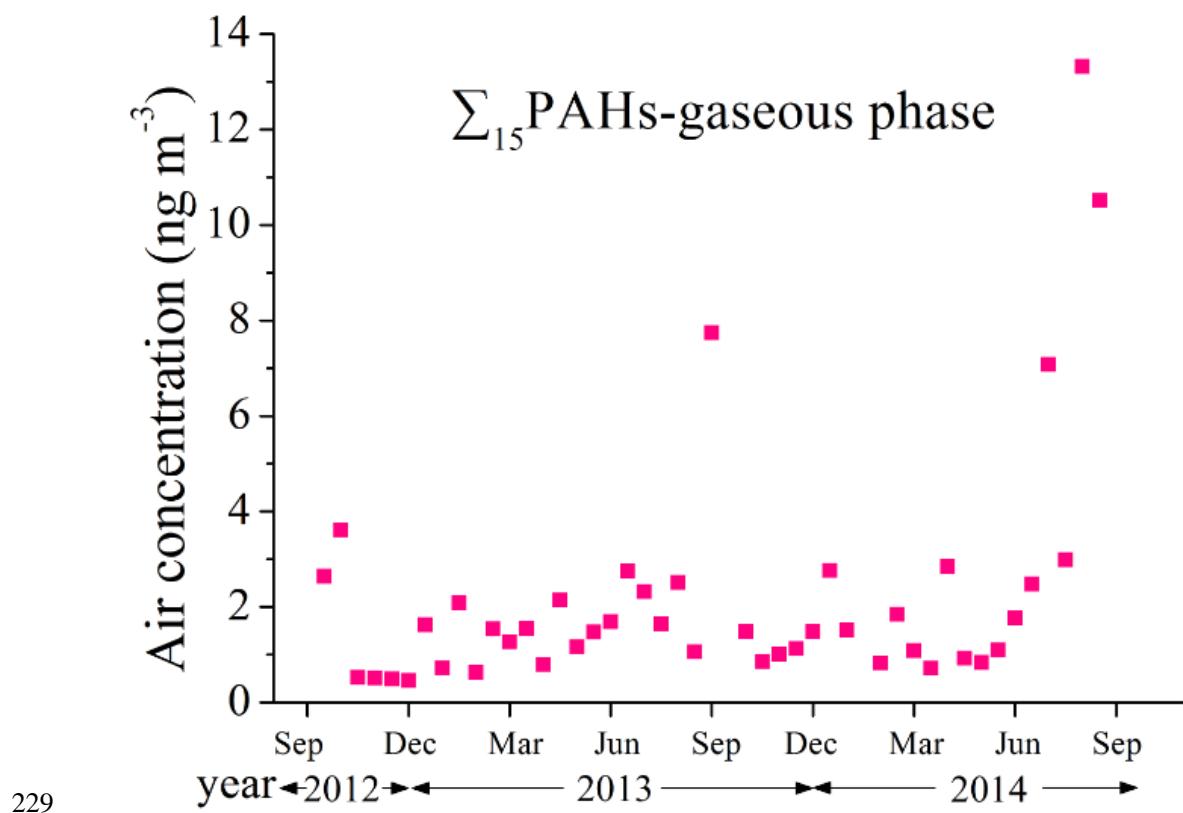
**Table S14. Concentrations of PAHs in lake water of Nam Co Lake and other remote sites (ng L<sup>-1</sup>).**

226

Location	Year	$\Sigma$ PAHs dissolved phase	$\Sigma$ PAHs particulate phase	Reference
Nam Co Lake	2013	28.6 (6.9-83.6)	7.3 (1.7-28)	This study
Lakes in Himalayan region, Nepal	2007	1.96 ± 1.86		(Guzzella et al., 2011)
Lake Redo, Pyrenees	2000	0.27 ± 0.19	0.41 ± 0.13	(Vilanova et al., 2001)
Lake Gossenkölle, Alps	2000	0.35 ± 0.19	0.57 ± 0.34	(Vilanova et al., 2001)
Lake Ladove, High Tatras	2000	3.4 ± 0.4	8.5 ± 0.7	(Fernandez et al., 2005)
Lake Ontario	2011-2012	5.5 ± 3.2		(Venier et al., 2014)
Lake Erie	2011-2012	4.8 ± 1.4		(Venier et al., 2014)
Lake Superior	2011-2012	1.1 ± 0.5		(Venier et al., 2014)
Atlantic Ocean	2005	(0.058-1.07)		(Nizzetto et al., 2008)
North Pacific and Arctic	2010		(0.014-0.76)	(Ma et al., 2013)
Gerlache Inlet Sea, Antarctica	2001	2.4 (2.1-2.9)	3.6 (2.8-4.7)	(Stortini et al., 2009)

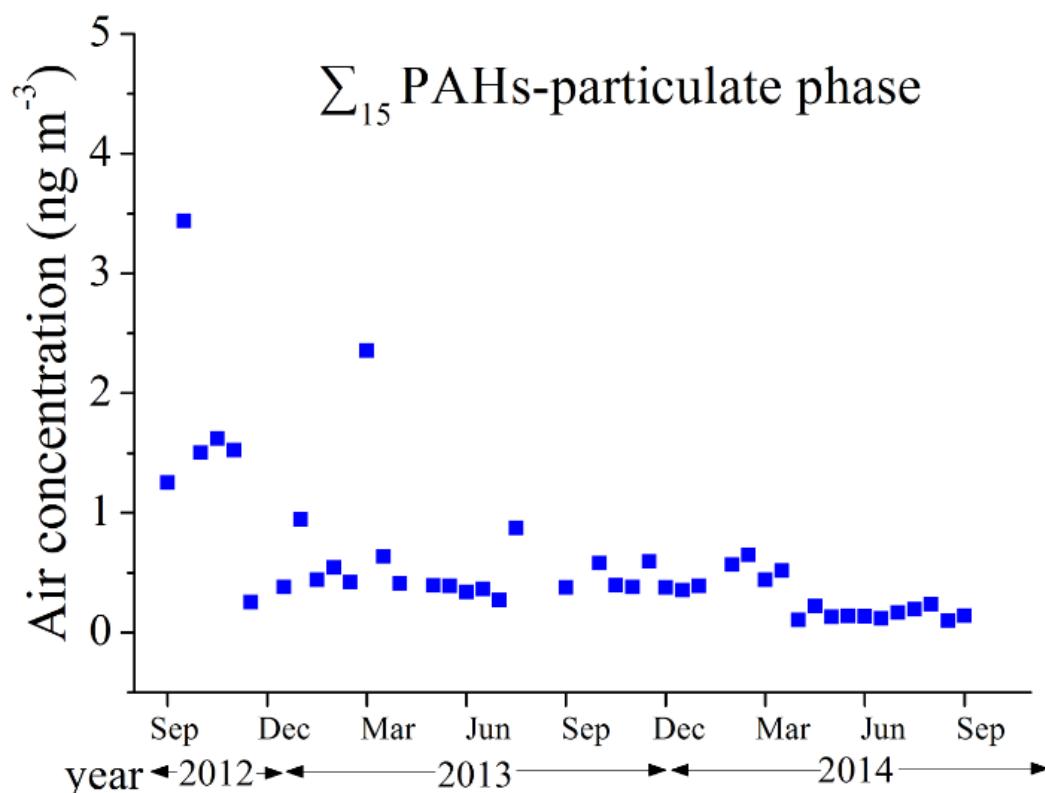
227 **Table S15. Comparison of isomer ratios of OCPs in this study and the source**  
 228 **region**

Sampling site	<i>a/r</i> -HCH	<i>p,p'</i> -DDT/ <i>p,p'</i> -DDE	Reference
Nam Co, TP	2.7±1.7	1.3±1.6	This study
Lulang, TP	3.7±1.99	>1	(Sheng et al., 2013)
Indian major cities	0.67±0.5	0.9±0.5	(Chakraborty et al., 2010)
Indian coastal region	0.11-4	2	(Zhang et al., 2008)
Bay of Bengal		>1	(Gioia et al., 2012)



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Figure S3. Seasonal patterns of the gaseous and particulate phase of  $\sum_{15}\text{PAHs}$  in the atmosphere of Nam Co.

233 **Table S16. Results of randomized block ANOVA to test the concentration**  
 234 **difference of POPs in the lake water from different regions of the Nam Co Lake.**

235 (a) Before the ANOVA analysis, the sampling sites of surface water in the present  
 236 study were classified as four regions according to the different locations:

<b>Site</b>	<b>Region</b>
S1	south
S2	south
S3	south
S4	south
S5	south
S6	south
S7	south
S8	northwest
S9	northwest
S10	north
S11	north
S12	north
S13	east
S14	east
S15	east

237 (2) *p* values derived from the ANOVA test:

		<b><i>p</i> values</b>		
<b>Location</b>		<b>HCHs</b>	<b>HCB</b>	<b>PAHs</b>
south	northwest	0.161	<b><i>0.05</i></b>	<b><i>0.001</i></b>
	north	0.106	0.96	0.303
	east	0.518	<b><i>0.002</i></b>	<b><i>0.005</i></b>
northwest	north	0.983	0.086	<b><i>0.007</i></b>
	east	0.424	0.25	0.16
north	east	0.361	<b><i>0.006</i></b>	<b><i>0.068</i></b>

238 (The values of *p* < 0.05 were highlighted in bold and italics, which indicates that the  
 239 levels of pollutants significantly differ between two regions)

240 **Table S17. Results of Clausius–Clapeyron regression**

241 Clausius-Clapeyron (C.C.) equation is often used for assessing the relative  
 242 contribution of re-volatilization from local surface versus long range atmospheric  
 243 transport:

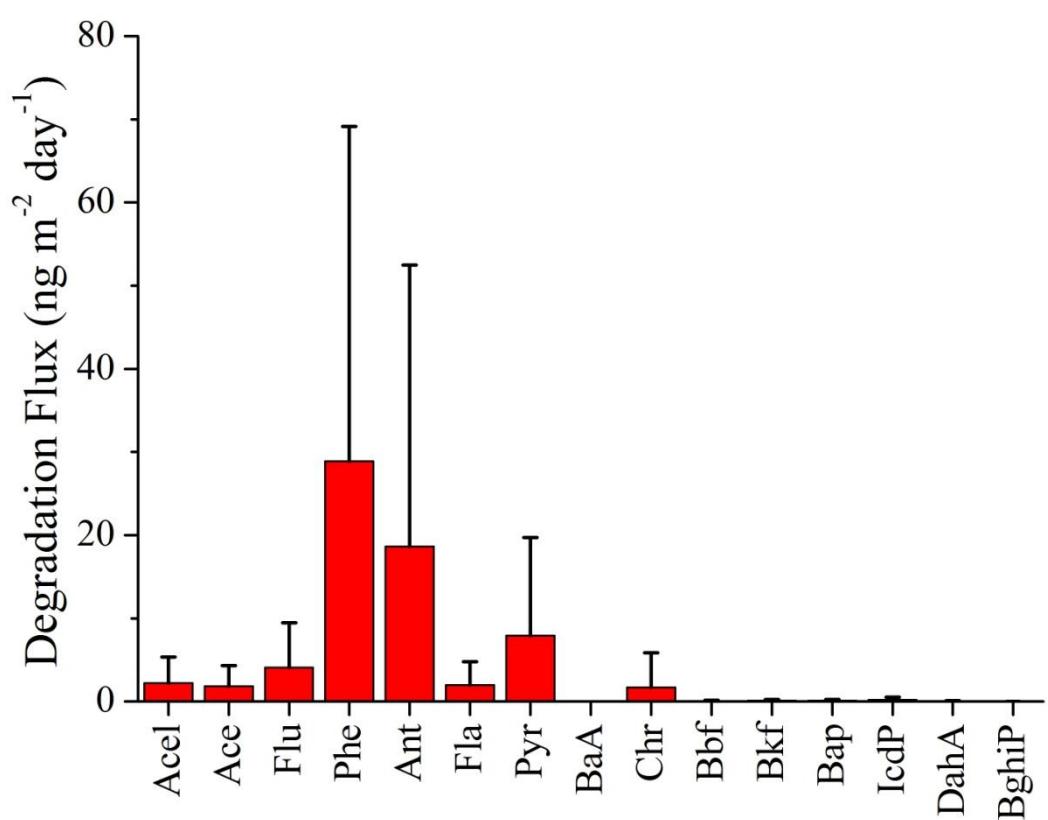
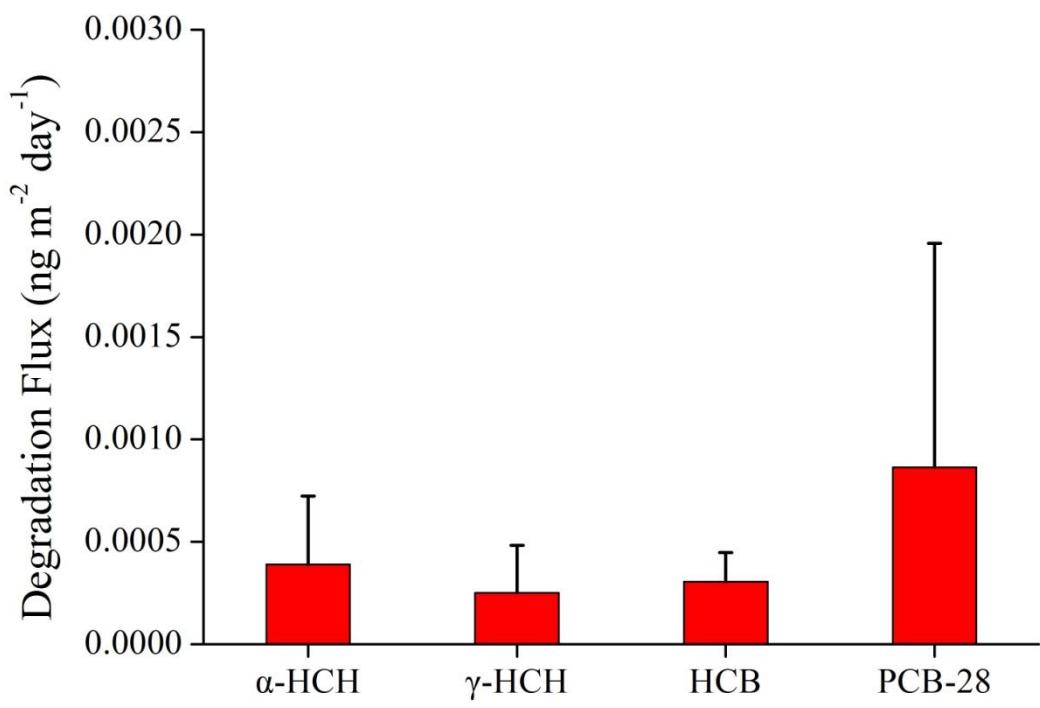
244  $\ln P = \frac{m}{T} + b$

245 where  $P$  is the partial pressure of pollutants in air (Pa),  $T$  is the air temperature (K), m  
 246 and b are slopes and intercepts derived from C.C. regression for each compound.  
 247 Carlson and Hites (2005) found that the temperature dependence will reduce or  
 248 disappear when  $T$  is below the freezing point. Thus, the sampling periods with  $T < 0$  °C  
 249 were excluded in the present study. Results included slope  $m$ ,  $R^2$ , p-value, and number  
 250 of data points,  $n$ , as shown in the table below.

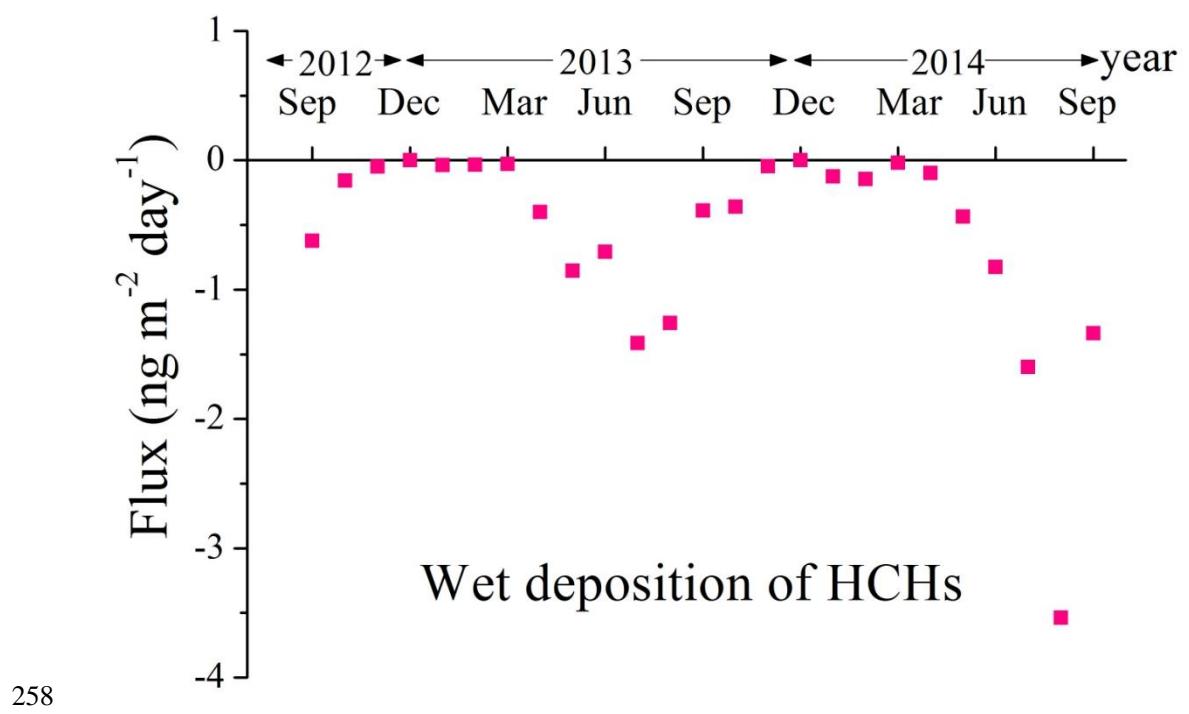
Compound	slope ( $m$ )	$R^2$	p-value	$n$	Compound	$m$	$R^2$	p-value	$n$
α-HCH	-7893	0.29	<b>0.01<sup>a</sup></b>	18	Acel	\	\	0.83	14
β-HCH	\ <sup>b</sup>	\	0.27	14	Ace	\	\	0.93	14
γ-HCH	\	\	0.50	17	Flu	\	\	0.56	14
HCB	\	\	0.55	18	Phe	\	\	0.37	14
o,p'-DDE	\	\	0.90	18	Ant	\	\	0.13	14
p,p'-DDE	\	\	0.58	18	Fla	\	\	0.17	14
o,p'-DDT	\	\	0.06	18	Pyr	\	\	0.37	14
p,p'-DDT	\	\	0.33	18	BaA	\	\	0.71	14
PCB-28	\	\	0.78	18	Chr	\	\	0.87	14
PCB-52	\	\	0.61	18	Bbf	\	\	0.28	14
PCB-101	\	\	0.96	18	Bkf	\	\	0.15	13
PCB-153	\	\	0.61	18	Bap	\	\	0.61	13
PCB-138	\	\	0.89	18	IcdP	\	\	0.91	6
PCB-180	\	\	0.60	18	DahA	\	\	0.28	4
					BghiP	\	\	0.60	13

251 a: Numbers in bold font stand for  $p < 0.05$ , indicating that the relationship is  
 252 significant.

253 b: “\” stands for the unavailable slope and  $R^2$  for some compounds which showed  
 254 insignificant correlation ( $p > 0.05$ ).



257 **Figure S4.** Average atmospheric degradation fluxes of individual POPs.



260 **References**

- 261 Antweiler, R. C., and Taylor, H. E.: Evaluation of statistical treatments of left-censored  
262 environmental data using coincident uncensored data sets: I. Summary statistics, *Environ. Sci.*  
263 *Technol.*, 42, 3732-3738, doi:10.1021/es071301c, 2008.
- 264 Burkhard, L. P.: Estimating dissolved organic carbon partition coefficients for nonionic organic  
265 chemicals, *Environ. Sci. Technol.*, 34, 4663-4668, doi:10.1021/es001269l, 2000.
- 266 Cai, M. H., Ma, Y. X., Xie, Z. Y., Zhong, G. C., Moller, A., Yang, H. Z., Sturm, R., He, J. F.,  
267 Ebinghaus, R., and Meng, X. Z.: Distribution and air-sea exchange of organochlorine  
268 pesticides in the North Pacific and the Arctic, *J. Geophys. Res.*, 117,  
269 doi:10.1029/2011jd016910, 2012.
- 270 Carlson, D. L., and Hites, R. A.: Temperature dependence of atmospheric PCB concentrations,  
271 *Environ. Sci. Technol.*, 39, 740-747, doi:10.1021/es049081f, 2005.
- 272 Castro-Jimenez, J., Berrojalbiz, N., Wollgast, J., and Dachs, J.: Polycyclic aromatic hydrocarbons  
273 (PAHs) in the Mediterranean Sea: Atmospheric occurrence, deposition and decoupling with  
274 settling fluxes in the water column, *Environ. Pollut.*, 166, 40-47,  
275 doi:10.1016/j.envpol.2012.03.003, 2012.
- 276 Cetin, B., Ozer, S., Sofuo glu, A., and Odabasi, M.: Determination of Henry's law constants of  
277 organochlorine pesticides in deionized and saline water as a function of temperature, *Atmos.*  
278 *Environ.*, 40, 4538-4546, doi: 10.1016/j.atmosenv.2006.04.009, 2006.
- 279 Chakraborty, P., Zhang, G., Li, J., Xu, Y., Liu, X., Tanabe, S., and Jones, K. C.: Selected  
280 Organochlorine Pesticides in the Atmosphere of Major Indian Cities: Levels, Regional versus  
281 Local Variations, and Sources, *Environ. Sci. Technol.*, 44, 8038-8043, doi:  
282 10.1021/Es102029t, 2010.
- 283 Cincinelli, A., Martellini, T., Del Bubba, M., Lepri, L., Corsolini, S., Borghesi, N., King, M. D.,  
284 and Dickhut, R. M.: Organochlorine pesticide air-water exchange and bioconcentration in  
285 krill in the Ross Sea, *Environ. Pollut.*, 157, 2153-2158, doi: 10.1016/j.envpol.2009.02.010,  
286 2009.
- 287 Ding, X., Wang, X. M., Xie, Z. Q., Xiang, C. H., Mai, B. X., Sun, L. G., Zheng, M., Sheng, G. Y.,  
288 and Fu, J. M.: Atmospheric hexachlorocyclohexanes in the North Pacific Ocean and the  
289 adjacent Arctic region: Spatial patterns, chiral signatures, and sea-air exchanges, *Environ. Sci.*  
290 *Technol.*, 41, 5204-5209, doi:10.1021/es070237w, 2007a.
- 291 Ding, X., Wang, X. M., Xie, Z. Q., Xiang, C. H., Mai, B. X., Sun, L. G., Zheng, M., Sheng, G. Y.,  
292 Fu, J. M., and Poschl, U.: Atmospheric polycyclic aromatic hydrocarbons observed over the  
293 North Pacific Ocean and the Arctic area: Spatial distribution and source identification, *Atmos.*  
294 *Environ.*, 41, 2061-2072, doi:10.1016/j.atmosenv.2006.11.002, 2007b.
- 295 Fernandez, P., Grimalt, J. O., and Vilanova, R. M.: Atmospheric gas-particle partitioning of  
296 polycyclic aromatic hydrocarbons in high mountain regions of Europe, *Environ. Sci. Technol.*,  
297 36, 1162-1168, doi: 10.1021/Es010190t, 2002.
- 298 Fernandez, P., Carrera, G., and Grimalt, J. O.: Persistent organic pollutants in remote freshwater  
299 ecosystems, *Aquat. Sci.*, 67, 263-273, doi:10.1007/s00027-005-0747-8, 2005.
- 300 Gioia, R., Lohmann, R., Dachs, J., Temme, C., Lakaschus, S., Schulz-Bull, D., Hand, I., and Jones,  
301 K. C.: Polychlorinated biphenyls in air and water of the North Atlantic and Arctic Ocean, *J.*  
302 *Geophys. Res.*, 113, doi: 10.1029/2007jd009750, 2008.
- 303 Gioia, R., Li, J., Schuster, J., Zhang, Y. L., Zhang, G., Li, X. D., Spiro, B., Bhatia, R. S., Dachs, J.,

- 304 and Jones, K. C.: Factors Affecting the Occurrence and Transport of Atmospheric  
305 Organochlorines in the China Sea and the Northern Indian and South East Atlantic Oceans,  
306 Environ. Sci. Technol., 46, 10012-10021, doi: 10.1021/Es302037t, 2012.
- 307 Gong, P., Wang, X. P., Sheng, J. J., and Yao, T. D.: Variations of organochlorine pesticides and  
308 polychlorinated biphenyls in atmosphere of the Tibetan Plateau: Role of the monsoon system,  
309 Atmos. Environ., 44, 2518-2523, doi: 10.1016/j.atmosenv.2010.04.025, 2010.
- 310 Gong, P., Wang, X. P., and Yao, T. D.: Ambient distribution of particulate- and gas-phase n-alkanes  
311 and polycyclic aromatic hydrocarbons in the Tibetan Plateau, Environmental Earth Sciences,  
312 64, 1703-1711, doi:10.1007/s12665-011-0974-3, 2011.
- 313 Gong, P., Wang, X. P., Xue, Y. G., Sheng, J. J., Gao, S. P., Tian, L. D., and Yao, T. D.: Influence of  
314 atmospheric circulation on the long-range transport of organochlorine pesticides to the  
315 western Tibetan Plateau, Atmospheric Research, 166, 157-164,  
316 doi:10.1016/j.atmosres.2015.07.006, 2015.
- 317 Gonzalez-Gaya, B., Zuniga-Rival, J., Ojeda, M. J., Jimenez, B., and Dachs, J.: Field  
318 Measurements of the Atmospheric Dry Deposition Fluxes and Velocities of Polycyclic  
319 Aromatic Hydrocarbons to the Global Oceans, Environ. Sci. Technol., 48, 5583-5592,  
320 doi:10.1021/es500846p, 2014.
- 321 Guzzella, L., Poma, G., De Paolis, A., Roscioli, C., and Viviano, G.: Organic persistent toxic  
322 substances in soils, waters and sediments along an altitudinal gradient at Mt. Sagarmatha,  
323 Himalayas, Nepal, Environ. Pollut., 159, 2552-2564, doi:10.1016/j.envpol.2011.06.015,  
324 2011.
- 325 Huang, Y. M., Xu, Y., Li, J., Xu, W. H., Zhang, G., Cheng, Z. N., Liu, J. W., Wang, Y., and Tian, C.  
326 G.: Organochlorine Pesticides in the Atmosphere and Surface Water from the Equatorial  
327 Indian Ocean: Enantiomeric Signatures, Sources, and Fate, Environ. Sci. Technol., 47,  
328 13395-13403, doi: 10.1021/Es403138p, 2013.
- 329 Jantunen, L. M., and Bidleman, T. F.: Henry's law constants for hexachlorobenzene, p,p'-DDE and  
330 components of technical chlordane and estimates of gas exchange for Lake Ontario,  
331 Chemosphere, 62, 1689-1696, doi: 10.1016/j.chemosphere.2005.06.035, 2006.
- 332 Kallenborn, R., Breivik, K., Eckhardt, S., Lunder, C. R., Mano, S., Schlabach, M., and Stohl, A.:  
333 Long-term monitoring of persistent organic pollutants (POPs) at the Norwegian Troll station  
334 in Dronning Maud Land, Antarctica, Atmospheric Chemistry and Physics, 13, 6983-6992, doi:  
335 10.5194/acp-13-6983-2013, 2013.
- 336 Khairy, M., Muir, D., Teixeira, C., and Lohmann, R.: Spatial Trends, Sources, and Air-Water  
337 Exchange of Organochlorine Pesticides in the Great Lakes Basin Using Low Density  
338 Polyethylene Passive Samplers, Environ. Sci. Technol., 48, 9315-9324,  
339 doi:10.1021/es501686a, 2014.
- 340 Lei, Y. D., Chankalal, R., Chan, A., and Wania, F.: Supercooled liquid vapor pressures of the  
341 polycyclic aromatic hydrocarbons, J. Chem. Eng. Data., 47, 801-806, doi:10.1021/je0155148,  
342 2002.
- 343 Li, C. L., Kang, S. C., Chen, P. F., Zhang, Q. G., and Fang, G. C.: Characterizations of  
344 particle-bound trace metals and polycyclic aromatic hydrocarbons (PAHs) within Tibetan  
345 tents of south Tibetan Plateau, China, Environmental Science and Pollution Research, 19,  
346 1620-1628, doi:10.1007/s11356-011-0678-y, 2012.
- 347 Li, J., Zhu, T., Wang, F., Qiu, X. H., and Lin, W. L.: Observation of organochlorine pesticides in

- 348           the air of the Mt. Everest region, *Ecotoxicol. Environ. Saf.*, 63, 33-41, doi:  
349           10.1016/j.ecoenv.2005.04.001, 2006.
- 350           Liu, X. B., Yao, T. D., Kang, S. C., Jiao, N. A. Z., Zeng, Y. H., and Liu, Y. Q.: Bacterial  
351           Community of the Largest Oligosaline Lake, Namco on the Tibetan Plateau, *Geomicrobiol. J.*,  
352           27, 669-682, doi:10.1080/01490450903528000, 2010.
- 353           Lohmann, R., Gioia, R., Jones, K. C., Nizzetto, L., Temme, C., Xie, Z., Schulz-Bull, D., Hand, I.,  
354           Morgan, E., and Jantunen, L.: Organochlorine Pesticides and PAHs in the Surface Water and  
355           Atmosphere of the North Atlantic and Arctic Ocean, *Environ. Sci. Technol.*, 43, 5633-5639,  
356           doi: 10.1021/Es901229k, 2009.
- 357           Ma, Y. G., Lei, Y. D., Xiao, H., Wania, F., and Wang, W. H.: Critical Review and Recommended  
358           Values for the Physical-Chemical Property Data of 15 Polycyclic Aromatic Hydrocarbons at  
359           25 degrees C, *J. Chem. Eng. Data.*, 55, 819-825, doi: 10.1021/Je900477x, 2010.
- 360           Ma, Y. X., Xie, Z. Y., Yang, H. Z., Moller, A., Halsall, C., Cai, M. H., Sturm, R., and Ebinghaus,  
361           R.: Deposition of polycyclic aromatic hydrocarbons in the North Pacific and the Arctic, *J.  
362           Geophys. Res.*, 118, 5822-5829, doi:10.1002/jgrd.50473, 2013.
- 363           Ni, N., and Yalkowsky, S. H.: Prediction of Setschenow constants, *Int. J. Pharm.*, 254, 167-172,  
364           doi:10.1016/S0378-5173(03)00008-5, 2003.
- 365           Nizzetto, L., Lohmann, R., Gioia, R., Jahnke, A., Temme, C., Dachs, J., Herckes, P., Di Guardo, A.,  
366           and Jones, K. C.: PAHs in air and seawater along a North-South Atlantic transect: Trends,  
367           processes and possible sources, *Environ. Sci. Technol.*, 42, 1580-1585,  
368           doi:10.1021/es0717414, 2008.
- 369           Odabasi, M., Cetin, E., and Sofuo glu, A.: Determination of octanol-air partition coefficients and  
370           supercooled liquid vapor pressures of PAHs as a function of temperature: Application to  
371           gas-particle partitioning in an urban atmosphere, *Atmos. Environ.*, 40, 6615-6625, doi:  
372           10.1016/j.atmosenv.2006.05.051, 2006.
- 373           Ouyang, H. L., He, W., Qin, N., Kong, X. Z., Liu, W. X., He, Q. S., Yang, C., Jiang, Y. J., Wang, Q.  
374           M., Yang, B., and Xu, F. L.: Water-gas exchange of organochlorine pesticides at Lake Chaohu,  
375           a large Chinese lake, *Environmental Science and Pollution Research*, 20, 2020-2032, doi:  
376           10.1007/s11356-012-1374-2, 2013.
- 377           Paasivirta, J., and Sinkkonen, S. I.: Environmentally Relevant Properties of All 209  
378           Polychlorinated Biphenyl Congeners for Modeling Their Fate in Different Natural and  
379           Climatic Conditions, *J. Chem. Eng. Data.*, 54, 1189-1213, doi: 10.1021/Je800501h, 2009.
- 380           Piazza, R., Gambaro, A., Argiriadis, E., Vecchiato, M., Zambon, S., Cescon, P., and Barbante, C.:  
381           Development of a method for simultaneous analysis of PCDDs, PCDFs, PCBs, PBDEs,  
382           PCNs and PAHs in Antarctic air, *Anal. Bioanal. Chem.*, 405, 917-932,  
383           doi:10.1007/s00216-012-6464-y, 2013.
- 384           Qiu, X. H., Zhu, T., Wang, F., and Hu, J. X.: Air-water gas exchange of organochlorine pesticides  
385           in Taihu Lake, China, *Environ. Sci. Technol.*, 42, 1928-1932, doi: 10.1021/Es071825c, 2008.
- 386           Ruge, Z., Muir, D., Helm, P., and Lohmann, R.: Concentrations, Trends, and Air-Water Exchange  
387           of PAHs and PBDEs Derived from Passive Samplers in Lake Superior in 2011, *Environ. Sci.  
388           Technol.*, 49, 13777-13786, doi:10.1021/acs.est.5b02611, 2015.
- 389           Sahsuvar, L., Helm, P. A., Jantunen, L. M., and Bidleman, T. F.: Henry's law constants for alpha-,  
390           beta-, and gamma-hexachlorocyclohexanes (HCHs) as a function of temperature and revised  
391           estimates of gas exchange in Arctic regions, *Atmos. Environ.*, 37, 983-992,

- 392 doi:10.1016/S1352-2310(02)00936-6, 2003.
- 393 Schwarzenbach, R. P., Gschwend, P. M., and Imboden, D. M.: Environmental Organic Chemistry,  
394 John Wiley and Sons, New Jersey, 2003.
- 395 Sheng, J., Wang, X., Gong, P., Joswiak, D. R., Tian, L., Yao, T., and Jones, K. C.: Monsoon-driven  
396 transport of organochlorine pesticides and polychlorinated biphenyls to the tibetan plateau:  
397 three year atmospheric monitoring study, Environ. Sci. Technol., 47, 3199-3208,  
398 doi:10.1021/es305201s, 2013.
- 399 Stortini, A. M., Martellini, T., Del Bubba, M., Lepri, L., Capodaglio, G., and Cincinelli, A.:  
400 n-Alkanes, PAHs and surfactants in the sea surface microlayer and sea water samples of the  
401 Gerlache Inlet sea (Antarctica), Microchemical Journal, 92, 37-43,  
402 doi:10.1016/j.microc.2008.11.005, 2009.
- 403 Su, Y., Hung, H., Blanchard, P., Patton, G. W., Kallenborn, R., Konoplev, A., Fellin, P., Li, H.,  
404 Geen, C., Stern, G., Rosenberg, B., and Barrie, L. A.: A circumpolar perspective of  
405 atmospheric organochlorine pesticides (OCPs): Results from six Arctic monitoring stations in  
406 2000-2003, Atmos. Environ., 42, 4682-4698, doi:10.1016/j.atmosenv.2008.01.054, 2008.
- 407 Su, Y. S., Hung, H., Blanchard, P., Patton, G. W., Kallenborn, R., Konoplev, A., Fellin, P., Li, H.,  
408 Geen, C., Stern, G., Rosenberg, B., and Barrie, L. A.: Spatial and seasonal variations of  
409 hexachlorocyclohexanes (HCHs) and hexachlorobenzene (HCB) in the Arctic atmosphere,  
410 Environ. Sci. Technol., 40, 6601-6607, doi: 10.1021/Es061065q, 2006.
- 411 Van Drooge, B. L., Grimalt, J. O., Camarero, L., Catalan, J., Stuchlik, E., and Garcia, C. J. T.:  
412 Atmospheric semivolatile organochlorine compounds in European high-mountain areas  
413 (Central Pyrenees and High Tatras), Environ. Sci. Technol., 38, 3525-3532, doi:  
414 10.1021/Es030108p, 2004.
- 415 Venier, M., Dove, A., Romanak, K., Backus, S., and Hites, R.: Flame Retardants and Legacy  
416 Chemicals in Great Lakes' Water, Environ. Sci. Technol., 48, 9563-9572,  
417 doi:10.1021/es501509r, 2014.
- 418 Vilanova, R. M., Fernandez, P., Martinez, C., and Grimalt, J. O.: Polycyclic aromatic  
419 hydrocarbons in remote mountain lake waters, Water Res., 35, 3916-3926, doi:  
420 10.1016/S0043-1354(01)00113-0, 2001.
- 421 Wang, X. P., Gong, P., Sheng, J. J., Joswiak, D. R., and Yao, T. D.: Long-range atmospheric  
422 transport of particulate Polycyclic Aromatic Hydrocarbons and the incursion of aerosols to  
423 the southeast Tibetan Plateau, Atmos. Environ., 115, 124-131,  
424 doi:10.1016/j.atmosenv.2015.04.050, 2015.
- 425 Wilkinson, A. C., Kimpe, L. E., and Blais, J. M.: Air-water gas exchange of chlorinated pesticides  
426 in four lakes spanning a 1,205 meter elevation range in the Canadian Rocky Mountains,  
427 Environ. Toxicol. Chem., 24, 61-69, doi: 10.1897/04-071r.1, 2005.
- 428 Xiao, H., Kang, S. C., Zhang, Q. G., Han, W. W., Loewen, M., Wong, F., Hung, H., Lei, Y. D., and  
429 Wania, F.: Transport of semivolatile organic compounds to the Tibetan Plateau: Monthly  
430 resolved air concentrations at Nam Co, J. Geophys. Res., 115, doi: 10.1029/2010jd013972,  
431 2010.
- 432 Xie, Z., Koch, B. P., Moller, A., Sturm, R., and Ebinghaus, R.: Transport and fate of  
433 hexachlorocyclohexanes in the oceanic air and surface seawater, Biogeosciences, 8,  
434 2621-2633, doi:10.5194/bg-8-2621-2011, 2011.
- 435 Zhang, G. S: A study of Zhadang glacier energy and mass balance and its hydrological process, in

- 436 NamCo basin, central Tibetan Plateau, Doctoral dissertation, 2013.
- 437 Zhang, W. L., Zhang, G., Qi, S. H. and Peng, P. A.: A preliminary study of  
438 organochlorinepesticides in water and sediments from two Tibetan Lakes. Geochimica. 32 (4),  
439 363-367 (in Chinese with English abstract), 2003.
- 440 Zhang, G., Chakraborty, P., Li, J., Sampathkumar, P., Balasubramanian, T., Kathiresan, K.,  
441 Takahashi, S., Subramanian, A., Tanabe, S., and Jones, K. C.: Passive Atmospheric Sampling  
442 of Organochlorine Pesticides, Polychlorinated Biphenyls, and Polybrominated Diphenyl  
443 Ethers in Urban, Rural, and Wetland Sites along the Coastal Length of India, Environ. Sci.  
444 Technol., 42, 8218-8223, doi: 10.1021/Es8016667, 2008.