We are grateful to the reviewers' thoughtful and constructive comments, because that will surely improve the quality of our manuscript. According to the reviewers' suggestions, we have now amended the manuscript carefully. All revisions are highlighted in the revised manuscript. Our point-to-point response to the reviewers' comments is provided below.

Reviewer #1:

1. 52-54. Please consider your statement: "Moreover, melted sea glaciers have released large quantities of POPs back into the atmosphere in the polar regions, which has led to increased POP levels in air and water (Jantunen et al., 2008; Wong et al., 2011)." It is true that melted glaciers have released POPs, but I don't think this has led to increased levels in arctic air and seawater. What is the evidence for this? Levels in arctic air are actually going down, although the decline may be slowed by release from "secondary sources", which could include glaciers. There is nothing in these two references to suggest that melting glaciers are a major source of POPs to the Arctic, and so the authors should revise this statement, the supporting references, or both.

Response: We agree with the reviewer's concern. In the revision, we rephrased the statement: "Moreover, melting ice and glaciers also have released OPs back into the atmosphere, which buffers the decline of OP levels in the polar regions (Ma et al., 2011; Geisz et al., 2008)", please see Page 3, Line 61-63.

References:

Ma, J. M., Hung, H. L., Tian, C., and Kallenborn, R.: Revolatilization of persistent organic pollutants in the Arctic induced by climate change, Nature Climate Change, 1, 255-260, 2011.

Geisz, H. N., Dickhut, R. M., Cochran, M. A., Fraser, W. R., and Ducklow, H. W.: Melting glaciers: A probable source of DDT to the Antarctic marine ecosystem, Environ. Sci. Technol., 42, 3958-3962, 2008.

2. 85, Site description. Nam Co is an interesting lake and more background information could be presented. For example, there is riverine inflow into the lake, but no outflow, which has implications for both the water and POPs budgets. Can a summary of annual riverine delivery of water be given (m^3 y^{-1})? Sources of POPs are probably direct atmospheric deposition and also delivery of snowmelt. The lake has a high pH >9 and this may have an effect on the fate of some POPs in the warmer season, particularly alpha- and gamma-HCHs which are subject to hydrolysis.

Response: We added some background information of Nam Co in section 2.1, please see Page 5, Line 104-107. Regarding the water budget, the Nam Co Lake is mainly supplied by precipitation and glacier meltwater. According to Wu et al. (2014), the annual riverine delivery of water to the lake is approximately 1.3×10^9 m^3 $year^{-1}$.

The hydrolysis rate constants of HCHs not only depend on pH, but also temperature (Ngabe et al., 1993). The higher pH in lake water of Nam Co may favor the hydrolysis of HCHs, whereas the low water temperature (about 10 ℃ even in the summer) will decrease the hydrolysis rate sharply. Under this combined effect, we

think the hydrolysis of HCHs does exist, but is not significant.

References:

Wu, Y. H., Zheng, H. X., Zhang, B., Chen, D. M., and Lei, L. P.: Long-Term Changes of Lake Level and Water Budget in the Nam Co Lake Basin, Central Tibetan Plateau, Journal of Hydrometeorology, 15, 1312-1322, 2014.

Ngabe, B., Bidleman, T. F., and Falconer, R. L.: Base Hydrolysis of Alpha-Hexachlorocyclohexanes and Gamma-Hexachlorocyclohexanes, Environ. Sci. Technol., 27, 1930-1933, 1993.

3. 106. Concerning 0.7 μm GFFs. This particle size collection capability refers to water, not air. The Whatman glass fiber filters collected smaller particles from air than water, due to electrostatic attraction of particles to the filter. The accepted collection efficiency of Whatman EPM 2000 filters is >99% for particles >0.3 μm.

Response: The pore size of GFFs used for air and water sampling is 0.45 μm and 0.7 μm, respectively. The previous statement got wrong and we feel very sorry for this. It was corrected in the revision, please see Page 5, Line 129.

4. 107. "suspended particulates". Please use "suspended particles", or else "suspended particulate matter", as you do in line 121.

Response: We corrected it, please see Page 5, Line 130.

5. 146-147. Splitting the PUFs in half to investigate breakthrough is a good step, but the results are strange. I would expect the more volatile PAHs like Ace, Acel and Fle to show more breakthrough that the less volatile 3-5 ring compounds, but there is really not much difference. Please refer to Table S5 in Supporting Information, which shows the same average breakthrough for all PAHs. Can the authors provide an explanation?

Response: We checked the original data and we found there is no significant gradient for the breakthrough of more volatile PAHs (Ace, Acel and Flu) and other less volatile PAHs. This may be caused by the low temperature in Nam Co (annual average temperature of ~0 ℃). Given that supercooled liquid vapor pressure (P_L) is good representative of volatility, P_L of 15 PAHs was calculated (Lei et al., 2002; Odabasi et al., 2006). As shown in the figure below, obvious decrease of P_L for more volatile PAHs (Ace, Acel and Flu) was observed when the temperature decreased. Therefore, the volatility gradient between different PAH congeners is reduced due to the cold temperature. That may be the reason why the similar breakthrough level of PAH congeners was observed in our study.
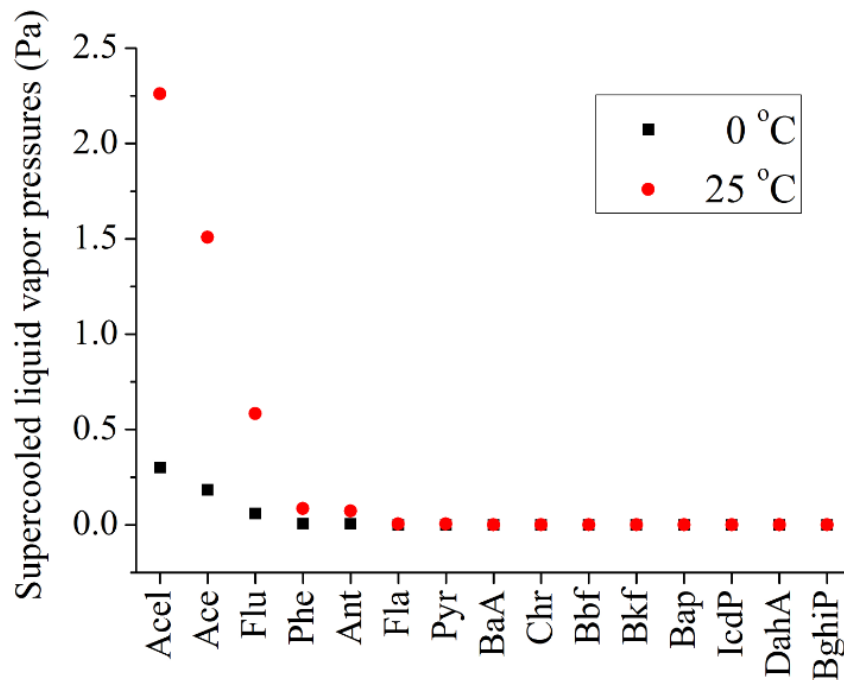
Figure. Supercooled liquid vapor pressures (P_L) of PAHs under different temperatures

References:

Lei, Y. D., Chankalal, R., Chan, A., and Wania, F.: Supercooled liquid vapor pressures of the polycyclic aromatic hydrocarbons, J. Chem. Eng. Data., 47, 801-806, 2002.

Odabasi, M., Cetin, E., and Sofuoglu, A.: Determination of octanol-air partition coefficients and supercooled liquid vapor pressures of PAHs as a function of temperature: Application to gas-particle partitioning in an urban atmosphere, Atmos. Environ., 40, 6615-6625, 2006.

6. Although much effort was given to evaluating collection efficiencies for atmospheric samples, I can see no similar effort to evaluate the XAD-2 collection efficiency. The authors pumped 200 L of water through their columns, which is a lot! Can they provide assurance that the relatively soluble HCH isomers were effectively retained under these conditions?

Response: Theoretically, 2 XAD columns deployed in series are used to evaluate the collection efficiency for water sample. Lohmann et al. (2009) did this by filtering around 1000 L of water and the result showed that: 1) the breakthrough of HCHs is limited (10-17%) although a large volume of water is pumped; 2) XAD has a great adsorption capacity, which can reach up to 20 ng for HCH [estimated by multiplying the volume of water sample (1000 L) by the concentration of HCH in water (1-20 pg L^{-1})]. Considering these existed results, the assessment of collection efficiency is usually omitted in the field sampling. Actually, in our study, we pumped 200 L of lake water with the concentrations of HCH around 10 pg L^{-1}, thus only about 2 ng of HCHs were accumulated which were not beyond the adsorption capacity of XAD. So we think HCHs are effectively retained.

Reference:

Lohmann, R., Gioia, R., Jones, K. C., Nizzetto, L., Temme, C., Xie, Z., Schulz-Bull, D., Hand, I., Morgan, E., and Jantunen, L.: Organochlorine Pesticides and PAHs in the Surface Water and Atmosphere of the North Atlantic and Arctic Ocean, Environ. Sci. Technol., 43, 5633-5639, 2009.

7. 147-150. "Certified surrogate standards…analysed alongside each sample". This would be more clear if stated: "Certified surrogate standards were added to each sample before extraction and analysis" (refer to Supporting Information, Text S1).
Response: We revised this sentence, please see Page 7, Line 177-179.

8. 159. Please give units of CG and CW. They should be mol m^{-3} as used in Eq. 1 and 2.
Response: We added the units of C_G and C_W, please see Page 8, Line 191-192.

9. 161-162. Please consider this statement: "According to Ruge et al. (2015) the POP concentrations retained by XAD were calibrated to derive the true freely dissolved POP concentrations in water (CW)". Ruge et al. collected PAHs and PBDEs from water using passive samplers with polyethylene strips, not XAD-2. The reference to XAD-2 for water sampling is Venier et al. (2014), in the authors' list. The Venier paper says: "The filters were not analyzed; therefore, results reported here represent the dissolved phase only. This operational definition of the dissolved phase has been adopted by regulatory agencies including the EPA, the USGS, and EC." However, there is a difference between "dissolved" (including sorption to dissolved organic carbon, DOC) and "truly dissolved" (not associated with DOC). The Venier statement only indicates that the filter-XAD-2 method discriminates between particulate POPs (found on the filter) and "dissolved" POPs on the XAD-2, which may (or may not!) include POPs associated with DOC. Can the authors provide other evidence that XAD-2 collects only "truly dissolved" POPs? This is an important point, because later in the paper (Table S4, Supporting Information) the authors use this mathematical correction for sorption of POPs to DOC. CW = CXAD/(1 + KDOC*CDOC). Doing this correction implies that CXAD includes BOTH the free POPs and POPs associated with DOC, in contradiction to the earlier statement (see above, "According to Ruge…").
Response: We aim to investigate the air-water gas exchange of POPs, so the concentrations of "truly freely dissolved POPs" are needed. However, XAD-2 resin collects both freely dissolved POPs and POPs associated with dissolved organic carbon (DOC) in water. As described in Gonzalez-Gaya et al. (2016), DOC-correction can remove the influence of possible sorption by DOC and derive the "truly freely dissolved POPs", thus we adopted this method. To make it clear, we changed the reference of Ruge et al. (2015) to Gonzalez-Gaya et al. (2016) in the revision, please see Page 8, Line 193-196.

Reference:

Gonzalez-Gaya, B., Fernandez-Pinos, M. C., Morales, L., Mejanelle, L., Abad, E., Pina, B., Duarte, C. M., Jimenez, B., and Dachs, J.: High atmosphere-ocean exchange of semivolatile aromatic hydrocarbons, Nature Geoscience, 9, 438-442, 2016.

10. 245. Possible sources. The discussion here suggests that LRAT is the main source to Nam Co and is supported by the uniform distribution of HCHs in lake water. Surprising, I thought snowmelt delivery would lead to a spring pulse in lake water, especially for HCHs near river discharge points. But no. So the authors' suggestion of the Indian Monsoon delivery is the best explanation, shown well in Figure 3. Some air samples contain high levels of o,p'-DDT and o,p'-DDE relative to the p,p'-isomers. Does this suggest dicofol contribution?

Response: Thanks for the reviewer's positive comment. We do think that the Indian Monsoon delivery is an important source of OCPs in Nam Co.

The higher levels of o,p'-DDT relative to p,p'-DDT can be attributed to two reasons. One is the fresh input of o,p'-DDT, i.e., dicofol contribution. This can be attributed to the long-range transport from South Asia, where has extremely higher o,p'-DDT/p,p'-DDT ratios (maximum 15, Zhang et al., 2008). The second reason is the higher volatility of o,p'-DDT than p,p' isomer (Spencer and Cliath, 1972), resulting in greater potential for long-range transport of o,p'-DDT and thus its higher level in receptor regions like the Tibetan Plateau.

References:

Zhang, G., Chakraborty, P., Li, J., Sampathkumar, P., Balasubramanian, T., Kathiresan, K., Takahashi, S., Subramanian, A., Tanabe, S., and Jones, K. C.: Passive Atmospheric Sampling of Organochlorine Pesticides, Polychlorinated Biphenyls, and Polybrominated Diphenyl Ethers in Urban, Rural, and Wetland Sites along the Coastal Length of India, Environ. Sci. Technol., 42, 8218-8223, 2008.

Spencer, W. F., and Cliath, M. M.: Volatility of DDT and Related Compounds, J. Agric. Food Chem., 20, 645-649, 1972.

11. 296. The alpha-HCH enantiomer section is well done and an excellent contribution to the study. The strongly nonracemic EFs in water contrast sharply with racemic signatures in air, suggesting that there is little volatilization, as the authors point out. Here the authors might alert the reader to the fugacity calculations (line 327) which also show net deposition of alpha-HCH. Also very interesting is the negative correlation between EF and bacterial abundance (Figure 5). Can this be quantified (r^2, p-value)?

Response: The negative correlation between EF in lake water and bacterial abundance can be quantified by R^2 and p-value (R^2=0.68, p-value<0.05), please see the figure below. In the revised manuscript, we added this figure as Figure S4 in supporting information.
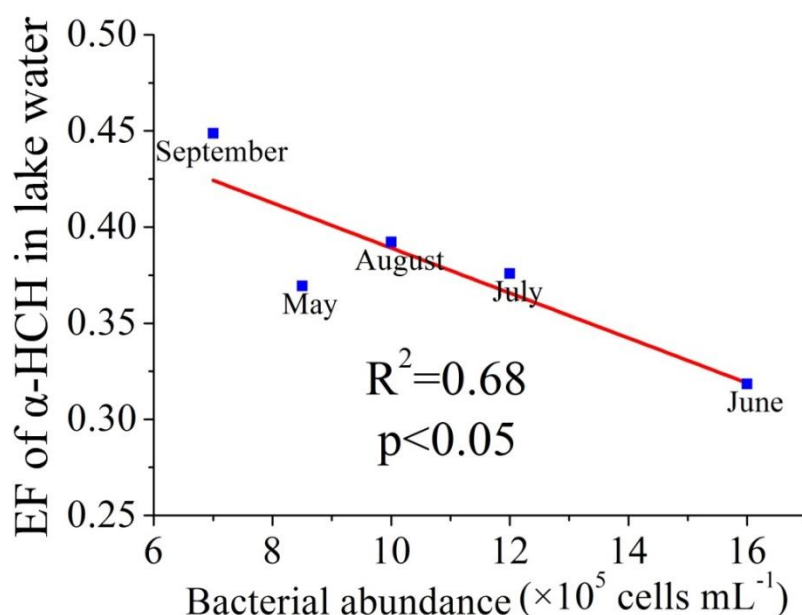
Figure. The correlation between EF of α-HCH in the lake water and seasonal bacterial abundance in Nam Co Lake.

12. 481. Carbon cycling. The authors extrapolate 8.7 tons (tonnes? metric?) of carbon to the Tibetan lakes, based on their atmospheric deposition estimates of PAHs to Nam Co. The paragraph goes on to claim this is a "significant allochthonous carbon flux". The Conclusions (lines 503-504) also mention a "substantial carbon source for this oligotrophic lake". This discussion is presented without putting the PAH deposition into context with other carbon fluxes. There must be additional carbon sources – soot, organic compounds other than PAHs, delivery of DOC by snowmelt. Perhaps the authors could provide some perspective by examining the carbon budgets for other small and medium-sized lakes?

Response: The unit of extrapolated carbon deposition to the Tibetan lakes should be tonnes. We corrected it in the revision, please see Page 21, Line 587.

We agree with the reviewer's concern, there must be other carbon sources to the lake in addition to the measured PAHs in our study. These sources include soot, dissolved organic carbon (DOC) in the water, and other anthropogenic organic compounds, which will drive a much larger deposition of carbon to the lake. We discussed this in the revision, please see Page 21, Line 588-590.

In the future work, we will focus on small-sized lakes to examine carbon budgets from different sources and compare their relative contributions.

Reference:

Gonzalez-Gaya, B., Fernandez-Pinos, M. C., Morales, L., Mejanelle, L., Abad, E., Pina, B., Duarte, C. M., Jimenez, B., and Dachs, J.: High atmosphere-ocean exchange of semivolatile aromatic hydrocarbons, Nature Geoscience, 9, 438-442, 2016.

Reviewer #2:

1. In the abstract and rest of the text, indicate how many PAHs. It is not the same to report the sum of concentrations of 16, 32 or 64 PAHs.

Response: In the revision, $\sum_{15}$PAHs was indicated. Please see Page 2, Line 42.

2. PAHs are not strictly POPs, maybe refer to OPs instead of POPs.

Response: We revised this, please see Page 3, Line 46.

3. Lines 53-54. Rephrase and maybe cite Ma et al. Nature climate change 2011 and/or Cabrerizo et al. EST 2013 or Galbán-Malagon et al. Atm Environ. 2013.

Response: We rephrased this statement: "melting ice and glaciers also have released OPs back into the atmosphere, which buffers the decline of OP levels in the polar regions" and cited Ma et al. (Nature climate change, 2011) as supporting reference. Please see Page 3, Line 61-63.

Reference:

Ma, J. M., Hung, H. L., Tian, C., and Kallenborn, R.: Revolatilization of persistent organic pollutants in the Arctic induced by climate change, Nature Climate Change, 1, 255-260, 2011.

4. Note that low MW PAHs originate not only from fossil fuels and biomass burning, but could also be originating from biodegradation of some organic compounds, such as diterpenes. There are several works suggesting this for soils as well.

Response: There are indeed biogenic sources of low MW PAHs such as naphthalene, perylene, phenanthrene, but they are primarily produced in organisms, soils and sediments (Chen et al., 1998; Wilcke et al., 2000; Cabrerizo et al., 2011). The focus of our study is atmospheric processes of PAHs, thus the source contribution from biodegradation is minimal.

References:

Chen, J., Henderson, G., Grimm, C. C., Lloyd, S. W., and Laine, R. A.: Termites fumigate their nests with naphthalene, Nature, 392, 558-559, 1998.

Wilcke, W., Amelung, W., Martius, C., Garcia, M. V. B., and Zech, W.: Biological sources of polycyclic aromatic hydrocarbons (PAHs) in the Amazonian Rain Forest, Journal of Plant Nutrition and Soil Science, 163, 27-30, 2000.

Cabrerizo, A., Dachs, J., Moeckel, C., Ojeda, M. J., Caballero, G., Barcelo, D., and Jones, K. C.: Ubiquitous Net Volatilization of Polycyclic Aromatic Hydrocarbons from Soils and Parameters Influencing Their Soil-Air Partitioning, Environ. Sci. Technol., 45, 4740-4747, 2011.

5. The sampling times (deployment times) are long. This may be problematic for 2-4 ring PAHs, which could be degraded during sampling. This would indicate that the given PAH concentrations are a lower-end estimate, even though at low temperature this artifact is minimized.

Response: Yes, the long sampling time may induce the degradation of 2-4 ring PAHs,

resulting the underestimation of their concentrations. However, the air temperature in our study region is very low, which has an annual average of 0 ℃, therefore the influence of degradation is minimized.

6. Lines 166. . .. I suggest to estimate the uncertainty due to propagation error for the fugacity ratios and air water exchange. This can be done as described in the supplementary material of Bigot et al. 2016, for example. The uncertainty for the fugacity ratio is significantly lower than the factor of three assumed by the authors.

Response: In the revision, we re-evaluated the uncertainty according to the propagation error method described in Bigot et al. (2016). The propagated uncertainty in fugacity ratio (f_w/f_a, FR) can be estimated as follows:

$$RSD^2_{FR} = RSD^2_{Cw} + RSD^2_{CG} + RSD^2_H$$

in which the relative standard deviations (RSD) for C_w and C_a are 35% and 30% in our study, while RSD for Henry's law constants (H) is estimated to be 20% (Sahsuvar et al., 2003), thus resulting in a 50% uncertainty for FR. This indicates that the uncertainty range of air-water equilibrium should be 0.5-1.5. Based on this, we revised Figure 6 (please see Page 36) and the related discussion, please see Page 8, Line 198-201.

References:

Bigot, M., Muir, D. C. G., Hawker, D. W., Cropp, R., Dachs, J., Teixeira, C. F., and Nash, S. B.: Air-Seawater Exchange of Organochlorine Pesticides in the Southern Ocean between Australia and Antarctica, Environ. Sci. Technol., 50, 8001-8009, 2016.

Sahsuvar, L., Helm, P. A., Jantunen, L. M., and Bidleman, T. F.: Henry's law constants for alpha-, beta-, and gamma-hexachlorocyclohexanes (HCHs) as a function of temperature and revised estimates of gas exchange in Arctic regions, Atmos. Environ., 37, 983-992, 2003.

7. Equations 6-7. This works well for estimating precipitation of gas phase compounds by rain, but not by snow. Justify how snow deposition was estimated. There are several published snow washout ratios for PAHs and PCBs (Franz and Eisenreich EST 1998, or Zhang et al. ACP 2015).

Response: In the original manuscript, we used the snow washout ratios reported in Franz and Eisenreich (1998) to estimate snow deposition fluxes of PAHs. To make this more clearly, we revised the statement (please see Page 9, Line 234-237) and listed the adopted values as Table S7 in supporting information.

Reference:

Franz, T. P., and Eisenreich, S. J.: Snow scavenging of polychlorinated biphenyls and polycyclic aromatic hydrocarbons in Minnesota, Environ. Sci. Technol., 32, 1998.

8. Lines 251-253. I like Figure 3 and the correlation between the Monsson index and

POP concentrations. Is it possible to block the influence of monsoon and then test the influence of temperature?

Response: In our study, the influence of temperature on POPs concentrations was assessed by using the Clausius-Clapeyron (C-C) equation. Please see Page 13, Line 340-354.

9. Lines 280-281. Maybe true, but I find difficult to believe. Is there any alternative source?

Response: Generally, the sources of HCB include pesticide application, manufacturing emissions, incomplete combustion and secondary emissions (volatilization of old HCB from past contamination) (Bailey, 2001). First, HCB has never been used directly as a pesticide in China (Wang et al., 2010). Second, our study region is a remote and high-altitude lake (4700 m), local agricultural and industrial activities are very scarce. Given the high volatility of HCB, we also tested the contribution of re-volatilization from local surface. However, the atmospheric concentrations of HCB in our study showed a negative linear correlation with the temperature (please see the figure below), which demonstrated that the local re-volatilization of HCB is very limited. Therefore, the sources mentioned above are all ruled out. Based on the field survey and literature research, we found that the local residents in Nam Co use large amounts of local biomass (mostly yak dung) for cooking and heating (Xiao et al., 2015). Such biomass combustion will certainly produce HCB (Bailey, 2001). Combined with the higher level of HCB in the winter, which may be caused by enhanced combustion activities in the colder conditions, we finally attributed the HCB to the local biomass combustion.
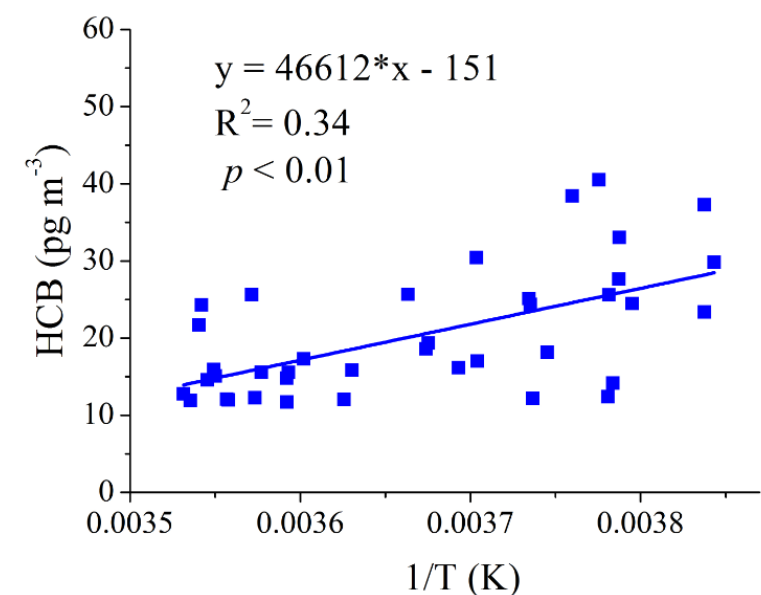
Figure. The relationship of atmospheric HCB with the reciprocal of temperature (T)

References:

Bailey, R. E.: Global hexachlorobenzene emissions, Chemosphere, 43, 167–182, 2001.

Wang, G., Lu, Y. L., Han, J. Y., Luo, W., Shi, Y. J., Wang, T. Y., and Sun, Y. M.:

Hexachlorobenzene sources, levels and human exposure in the environment of China, Environ. Int., 36, 122-130, 2010.

Xiao, Q. Y., Saikawa, E., Yokelson, R. J., Chen, P. F., Li, C. L., and Kang, S. C.: Indoor air pollution from burning yak dung as a household fuel in Tibet, Atmos. Environ., 102, 406-412, 2015.

10. I like the section related to the estimates of depositional fluxes. However, somewhere should say that only 15 PAHs were considered. There are other PAHs with higher abundances than phenanthrene (for example alkylated phenanthrenes), in addition to thousands of other anthropogenic organic compounds.

Response: We agree with the reviewer's concern. In the revision, we added some discussion about this: "In addition to the 15 PAHs considered here, there are other PAHs with higher abundances than Phe, for example alkylated phenanthrenes, which will drive a much larger depositional fluxes to the lake". Please see Page 20, Line 557-559.

11. Rewrite the section on the uncertainty of fluxes after a proper estimation of these uncertainties (see above).

Response: Actually, we did use propagation error method to estimate the uncertainties of air-water gas exchange fluxes in the manuscript, please see Page 21, Line 565-567. However, due to the unavailable data about the relative errors of atmospheric degradation rate (K_{OH}) and scavenging rates by wet deposition (W_G and W_P), the uncertainties for degradation and wet deposition fluxes are difficult to estimate quantitatively. We discussed this in the revision, please see Page 21, Line 568-571

12. In section 3.5, maybe cite Gonzalez-Gaya et al. Nature Geoscience 2016, who were the first to discuss the inputs of aromatic hydrocarbons in a context of carbon inputs. I agree with the authors that atmospheric inputs of semivolatile organic compounds, but not only PAHs, may be supporting the microbial communities in the Plateau's lakes. I suggest that future work should follow this line of research.

Response: We cited Gonzalez-Gaya et al. (2016) in this section, please see Page 21, Line 587-588. Moreover, we agree with the reviewer's concern and will follow this line of research in the future work.

Reference:

Gonzalez-Gaya, B., Fernandez-Pinos, M. C., Morales, L., Mejanelle, L., Abad, E., Pina, B., Duarte, C. M., Jimenez, B., and Dachs, J.: High atmosphere-ocean exchange of semivolatile aromatic hydrocarbons, Nature Geoscience, 9, 438-442, 2016.

Atmospheric processes of organic pollutants over a remote lake of the central Tibetan Plateau: Implications for regional cycling

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Abstract

Atmospheric processes (air-surface exchange, and atmospheric deposition and degradation) are crucial for understanding the global cycling and fate of organic pollutants (OPs). However, such assessment over the Tibetan Plateau (TP) remains uncertain. More than 50% of the Chinese lakes is located on the TP, which exerts a remarkable influence on the regional water, energy, and chemical cycling. In this study, air and water samples were simultaneously collected in Nam Co, a large lake on the TP, to test whether the lake is a "secondary source" or "sink" of OPs. Lower concentrations of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were observed in the atmosphere and lake water of Nam Co, while the levels of polycyclic aromatic hydrocarbons (PAHs) were relatively higher. Results of fugacity ratios and chiral signatures both suggest that the lake acted as the net sink of atmospheric hexachlorocyclohexanes (HCHs), following their long-range transport driven by the Indian Monsoon. Different behaviors were observed in the PAHs, which primarily originated from local biomass burning. Acenaphthylene, acenaphthene, and fluorene showed volatilization from the lake to the atmosphere; while other PAHs were deposited into the lake due to the integrated deposition process (wet/dry and air-water gas deposition) and limited atmospheric degradation. As the dominant PAH compound, phenanthrene exhibited a seasonal reversal of air-water gas exchange, which was likely related to the melting of the lake ice in May. The annual input of HCHs from air to the entire lake area (2015 km^2) was estimated as 1.9 kg year^{-1}, while those estimated for $\sum_{15}$PAHs can potentially reach up to 550 kg year^{-1}. This study highlights the significance of PAH deposition on the regional carbon cycling in the oligotrophic lakes of the TP.

1. Introduction

Since the past century, large quantities of organic pollutants (OPs), such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs), have been discharged into the global environment. Soils, water bodies, and snow/ice are generally considered as reservoirs or sinks of these pollutants (Dalla Valle et al., 2005; Froescheis et al., 2000; Guglielmo et al., 2012). However, due to the influence of global warming (Komprda et al., 2013; Noyes et al., 2009), growing evidence indicates that OPs previously stored in reservoirs can be re-released back to the environment (Ma et al., 2011). For example, air-soil exchange of OCPs has showed the re-emission of OCPs from past contaminated soils in Europe (Ruzickova et al., 2008), North America (Kurt-Karakus et al., 2006), and India (Chakraborty et al., 2015). Modeling results have suggested that large parts of the global ocean have been losing dichlorodiphenyltrichloroethane (DDT) via volatilization (Stemmler and Lammel, 2009). In addition, air-sea exchange of PAHs has revealed that the seawater in the Mediterranean has turned into a temporary secondary source of PAHs, which is related to biomass burning in that region (Mulder et al., 2014). Moreover, melting ice and glaciers also have released OPs back into the atmosphere, which buffers the decline of OP levels in the polar regions (Ma et al., 2011; Geisz et al., 2008).

Similar to the polar regions, the Tibetan Plateau (TP) has been regarded as a "convergence" of OPs (Wang et al., 2016). Due to the continuous use of OPs in the surrounding countries and the "cold trapping" by the TP, the enrichment of OPs in the TP environment has been reported (Sheng et al., 2013; Wang et al., 2015). However, the TP has experienced great warming (Liu and Chen, 2000), and results of the air-soil exchange of OPs have indicated that the Tibetan soils are acting as a sink of DDT and higher molecular weight PAHs, but are a potential secondary source for hexachlorobenzene (HCB) and hexachlorocyclohexanes (HCHs) (Wang et al., 2014; Wang et al., 2012). This shows that the cold temperature over the TP might not be sufficient to trap volatile OPs. More studies on the air-surface exchange of OPs over

74   the TP are therefore needed to test the role of the terrestrial and aquatic ecosystems of

75   the TP in the regional cycling of OPs.

76   Known as "Asia's water power", the TP contains the headwaters of many major rivers

77   in Asia, which provide water sources for about one-sixth of the world's population

78   (Yao et al., 2012). The TP also has large numbers of remote lakes that are important

79   components of water bodies. Low temperature, oligotrophic conditions, and the long

80   duration of ice-cover are distinct features of these lakes. Based on the higher

81   atmospheric concentrations of α-HCH in summer, Xiao et al. (2010) deduced that

82   these enhanced concentrations may be caused by the thawing of lake ice, which

83   promotes the re-evaporation of α-HCH. However, the study did not include

84   measurements of HCH levels in lake water or the corresponding air-water exchange

85   analysis (Xiao et al., 2010). Therefore, it is still unclear whether the lake water of the

86   TP is the secondary source of a large number of OPs. Furthermore, biomass burning is

87   a widespread activity over the TP (Hu et al., 2015). A recent study demonstrated that

88   the locally sourced biomass combustion particles contributed substantially to the

89   black carbon (BC) loading of the TP glacier (Li et al., 2016). Given that PAHs and BC

90   both mainly originate from incomplete combustion of biomass, regional air-water

91   exchange of PAHs would also contribute to the overall air-surface exchange of

92   carbon.

93   We therefore conducted air and water sampling in a remote lake on the TP, and

94   assessed the air-water gas exchange, and the dry and wet deposition processes of

95   OCPs, PCBs, and PAHs. The aims of this study were to ascertain whether the Tibetan

96   lake represents a secondary source of OPs, to investigate the influence of seasonal

97   lake ice melting on the gas exchange of different OPs, and to estimate the contribution

98   of PAH exchange to the lake carbon budget.

2. Materials and methods

2.1 Site description

101   Nam Co Lake (30°30′–30°56′N, 90°16′–91°01′E, 4718 m) is located in the north of

the Nyainqentanglha Mountains, on the central TP (Figure 1). It is the second largest lake in Tibet with an area of 2015 km^2 and a maximum depth exceeding 90 m (Wang et al., 2009). The lake is mainly supplied by precipitation and glacier meltwater. Annual riverine delivery of water to the lake is approximately 1.3×10^9 m^3 year^{-1}, while there is no outflow (Wu et al., 2014). The lake water is alkaline (pH=9.21) and slightly saline (Wang et al., 2009). The climate of Nam Co is relatively cold and windy with an annual average temperature of ~0 °C and an annual wind speed of ~4 m/s. The regional climate also has large seasonal variation: the Indian Monsoon dominates summer (May to September) and the westerlies control winter climate (October to April) [see the Supplement (S), Figure S1]. High temperatures and precipitation are usually observed in summer (Figure S2), and the lake begins to thaw from the beginning of May and melts completely by the end of May, which coincides with the onset of the Indian Monsoon. During the winter, the lake is covered by ice due to the subzero temperatures (Figure S2) and maximum instantaneous wind speeds reaching up to 9.9 m/s.

The dominant land cover in Nam Co is alpine steppe and meadow, and the local residents herd yak and sheep that graze around the lake. Biomass burning occurs for heating, cooking, transport, and religious reasons. Near the southeastern shore, the Nam Co Monitoring and Research Station for Multisphere Interactions (NCMORS) is operated by the Chinese Academy of Sciences (Figure 1b). This station not only facilitates the consecutive collection of field samples used in the current study, but also provides local meteorological parameters for flux calculations.

2.2 Air and water sampling

An active air sampler (AAS) was deployed on the roof of NCMORS (Figure 1b) and the air monitoring was conducted for two consecutive years from September 2012 to September 2014. The flow rate of AAS was 60 L min^{-1} and the air samples were collected every 2 weeks with a volume of approximately 600 m^3 for each sample. The air stream passes first through glass fiber filters (GFFs 0.45 μm, Whatman) to collect the total suspended particles (TSP) and then through polyurethane foam (PUF, 7.5 ×6

131   cm diameter) to retain the OPs in gas phase. In total, 47 air samples were collected.

132   Details regarding the sampling period, average air temperature, and wind speed are

133   given in Table S1. All harvested PUF and GFFs were stored at -20 ℃ until extraction.

134   To determine the OP levels in water, two sampling programs were conducted. First,

135   15 sites around the Nam Co Lake (surface lake water, 0–1 m depth) were selected to

136   obtain the spatial distribution of OPs in lake water (Figure 1b), which provides a

137   direct over-view of OP contamination over the lake. Second, monthly water samples

138   were collected at a site close to NCMORS (Figure 1b) from May to September, 2014

139   (water samples were not obtained during winter due to the ice cover). This provided

140   information regarding temporal variations in OP levels, isomer ratios, and the

141   enantiomeric fraction in lake water. Furthermore, coupled with the monthly average

142   air concentrations of individual OPs obtained, this allowed us to investigate the

143   air-water gas exchange of OPs (direction, flux, and monthly variations).

144   Water samples (200 L) were filtered with GFFs (0.7 μm, Whatman) to obtain the total

145   suspended particulate matter (SPM), and then pumped through an XAD-2 resin

146   column to collect the dissolved phase compounds. For each sampling month, triplicate

147   samples were collected. In total, 15 samples for the spatial study and 15 samples for

148   the temporal study were collected. XAD columns were kept at 4 ℃ until extraction.

149   The lake water properties (temperature, pH and salinity) are provided in Table S2.

150   **2.3 Sample extraction and analysis**

151   The chemical extraction and cleanup methods are detailed in Text S1 for each sample

152   type [air (PUF plug), TSP, water (XAD column), and SPM]. OPs were analyzed on a

153   gas chromatograph with an ion-trap mass spectrometer (GC-MS, Finnigan Trace

154   GC/PolarisQ) operating under MS–MS mode. More information on the

155   chromatographic conditions is given in Text S2. The target compounds are as follows:

156   HCHs (including α-HCH, β-HCH, and γ-HCH), HCB, DDTs (*o,p'*-DDE, *p,p'*-DDE,

157   *o,p'*-DDT, and *p,p'*-DDT), PCBs (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153,

158   and PCB 180), and 15 priority PAHs listed by the United States Environment

159 Protection Agency (USEPA, without naphthalene), including acenaphthylene (Acel),

160 acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant),

161 fluoranthene (Fla), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr),

162 benzo[b]fluoranthene (Bbf), benzo[k]fluoranthene (Bkf), benzo[a]pyrene (BaP),

163 dibenz[a, h]anthracene (DahA), benzo[g, h, i]perylene (BghiP), and

164 indeno[1,2,3-cd]pyrene (IcdP). Enantiomers of α-HCH were determined with a

165 BGB-172 chiral column (see Text S2 for details). The chiral signature of α-HCH is

166 expressed using the enantiomeric fraction (EF), which is equal to the ratio of peak

167 areas of the $(+)/[(+) + (-)]$ (Harner et al., 2000).

2.4 Quality assurance/quality control (QA/QC)

169 All analytical procedures were monitored using strict QA/QC measures. Prior to

170 sampling, PUF and XAD resin were pre-cleaned using dichloromethane (DCM) for

171 16 h and GFFs were baked at 450 ℃ for 4 h. Six PUF field blanks, three XAD field

172 blanks, and six procedural blanks were prepared; HCB, Phe, Ant, Fla, and Pyr were

173 detected in the field blanks (Table S3). The definitions of the method detection limits

174 (MDLs) are described in Text S3, and the derived MDLs are given in Table S4. The

175 breakthrough of PUF plugs was checked in eleven split PUFs, and the results show

176 that the individual OPs in the second half varied from 8% to 23% (Table S5),

177 indicating good retention capacity. Certified surrogate standards (from Dr.

178 Ehrenstorfer GmbH, Germany) were added to each sample before extraction and

179 analysis. The recoveries ranged from 71% to 94% for PCB 30, 79% to 105% for

180 Mirex, and 65% to 92% for perylene-D12. The reported concentrations were

181 subtracted by mean blanks but not corrected for recoveries. To check the

182 reproducibility of the chiral analysis, the racemic standard of α-HCH was injected

183 repeatedly and its average EF value was 0.499 ± 0.001 (n = 5).

2.5 Calculations of air-water gas exchange

185 Concurrent air and water samples were used to assess the status of air-water gas

186 exchange in Nam Co Lake. The gas exchange direction can be determined by the ratio

187 of fugacity in water (f_w) and air (f_a), giving the fugacity ratio (f_w/f_a) (Jantunen et al.,

188   2015):

189    $$f_a = C_G R T_a \qquad (1)$$

190    $$f_w = C_w H \qquad (2)$$

191   where C_G and C_w are the gaseous and dissolved concentrations of target compounds in

192   air and water (mol m^{-3}), respectively, R is the gas constant (8.314 Pa m^3 mol^{-1} K^{-1}), T_a

193   (K) is the air temperature, and H (Pa m^3 mol^{-1}) is the Henry's law constant. Due to the

194   possible sorption by dissolved organic carbon (DOC), OPs retained by XAD were

195   DOC-corrected to derive the truly freely dissolved concentrations in water (Text S4)

196   (Gonzalez-Gaya et al., 2016). H values were adjusted for the real water temperature

197   and salinity of Nam Co by the procedure described in Text S5 (Cetin et al., 2006; Ma

198   et al., 2010). The uncertainty involved in f_w/f_a was estimated by propagating the errors

199   in C_a (30%), C_w (35%), and H (20%), which was 50%. Accordingly, ratios of f_w/f_a

200   between 0.5 and 1.5 are assumed as air-water equilibrium, while $f_w/f_a > 1.5$ and <0.5

201   indicate net volatilization and deposition, respectively.

202   Net fluxes of air-water gas exchange (F_{AW}, ng m^{-2} day^{-1}) were quantified using the

203   Whitman two-film model, which has been used in many previous studies (Iwata et al.,

204   1993; Khairy et al., 2014):

205    $$F_{AW} = K_{ol}\left(C_w - C_G R T_a / H\right) \qquad (3)$$

206   where K_{ol} (m s^{-1}) is the overall mass transfer coefficient, which contains contributions

207   from the mass transfer coefficients of the water and air layers, K_w and K_a, respectively.

208   They are related to the wind speed and compound-specific molecular diffusivity; a

209   detailed calculation is presented in Text S6. Positive flux values indicate net

210   volatilization, and negative values indicate net deposition.

211   **2.6 Estimation of dry and wet deposition fluxes**

212   In addition to the gas exchange, dry and wet deposition are also important processes

213   that control the input of OPs from air to lake. Dry deposition fluxes (F_{DD}, ng m^{-2} day^{-1})

214   of atmospheric particulate-phase OPs can be calculated using (Gonzalez-Gaya et al.,

215   2016) the following equation:

$$F_{DD}=0.864V_{D}C_{P} \tag{4}$$

where V_D (cm s^{-1}) is the compound specific deposition velocity, C_P is the measured OPs concentrations in TSP (pg m^{-3}), and 0.864 is a unit conversion factor. V_D for each sampling period and compound was estimated using an empirical equation derived by Gonzalez-Gaya et al. (2014):

$$\log(V_{D})=-0.261\log(P_{L})+0.387U_{10}Chl_{s}-3.082 \tag{5}$$

where P_L (Pa) is subcooled liquid vapor pressure of chemicals that was corrected to the local temperature using the equations given in Table S6, U_{10} (m s^{-1}) is wind speed at 10 m height converted from the field-measured wind speed at 1.5 m (Table S1), and Chls is the surface chlorophyll concentration (mg m^{-3}, Liu et al., 2010).

Wet deposition fluxes by rain (F_{WD}, ng m^{-2} day^{-1}) were estimated using the method established by Jurado et al. (2005):

$$F_{WD}= P(W_{G}C_{G} + W_{P}C_{P}) \tag{6}$$

where P is precipitation depth per day (m d^{-1}) derived from the data recorded in the NCMORS, and W_G and W_P are the gas and particle washout ratios, respectively. Assuming that equilibrium is attained between the gas phase and the dissolved phase in a raindrop, W_G was estimated by (Wania et al., 1998a):

$$W_{G}=RT_{a}/H \tag{7}$$

The recommended value of W_P in Jurado et al. (2005) was used to consider the particle scavenging by rain.

For snow deposition, the fluxes were calculated by adopting the washout ratios reported in Franz and Eisenreich (1998) (Table S7).

3. Results and discussion

We determined the OP concentrations in air, TSP, water, and SPM separately; the full data sets are listed in Tables S8-S11. OCPs and PCBs were rarely detected in TSP (Table S9), and were therefore not considered in further discussion. Comparisons between the data from this study and previously published values for the TP and other remote regions are presented in Tables S12-S15.

The concentrations of OPs in the atmosphere and TSP in Nam Co are summarized in Figure 2 using box-and-whisker plots. Among the OCPs, HCB was the dominant chemical with an average concentration of 20 pg m^{-3} (Figure 2a), which was two times higher than that reported for southeastern TP (Sheng et al., 2013) and Mt. Everest (Li et al., 2006) (Table S12), but lower than the values in the Rocky Mountains (42 pg m^{-3}, Wilkinson et al., 2005) and the Arctic (64 pg m^{-3}, Su et al., 2006). The α-HCH (average 4.0 pg m^{-3}) and γ-HCH (2.1 pg m^{-3}) values in this study were much lower than those measured using a flow-through sampler (FTS) during 2006 to 2008 (48.7 and 7.9 pg m^{-3}, respectively) (Xiao et al., 2010). The DDT concentrations in the current study (0.8-46.4 pg m^{-3}) were lower than those observed for Lulang in southeastern TP (Table S12) (Sheng et al., 2013), which is the entrance of the Indian Monsoon. In spite of this, the levels of DDTs were still one order of magnitude higher than those for the Arctic (Table S12) (Su et al., 2008). Lower concentrations of $\sum_6$PCBs were also detected in air with an average value of 2.5 pg m^{-3}.

The sum concentrations of $\sum_{15}$PAHs in the atmosphere ranged from 0.5–13 and 0.1–3.4 ng m^{-3} in the gaseous and particulate phases, with averages of 2.2 and 0.6 ng m^{-3}, respectively. The 3- and 4-ring PAHs were predominant in both phases, including Phe, Flu, Fla, and Pyr (Figure 2b and 2c). The PAH levels in Nam Co were one order of magnitude lower than those reported for Lhasa (35.7 ng m^{-3}, Table S13), which is the capital city of Tibet with a large population, extensive tourism, and abundant religious activities (Gong et al., 2011). Compared with background levels in other regions of the world (Table S13), the PAHs in this study were comparable to the levels in Arctic air (Ding et al., 2007), but significantly higher than those from European mountainous regions (Fernandez et al., 2002).

In the lake water, the average dissolved concentrations of α-HCH, β-HCH, γ-HCH, HCB, and PCB 28 were 9.9, 85.2, 7.0, 7.6, and 1.9 pg L^{-1}, respectively; while DDT-related compounds were below MDLs in most cases for both dissolved and

273   SPM phases (Table S10 and S11). The current measured HCH concentrations were

274   approximately two orders of magnitude lower than values reported for the Yamdrok

275   and Co Ngoin Lake in 2002 (Table S14) (Zhang et al., 2003). Two possible reasons

276   for this discrepancy are: i) the inter-annual variation of chemicals, i.e., the

277   concentrations declined rapidly since 2002; and ii) the uncertainties caused by

278   analytical and instrumental method (electron capture detector in Zhang et al. (2003)

279   study and the MS detector in the current study). From a global perspective, the HCH

280   concentrations obtained by this study were overall lower than those in European

281   mountain lakes (Table S14). DDT class chemicals were rarely detected and only PCB

282   28 could be quantified in the Nam Co lake water. These features combined with the

283   low levels of HCHs suggest that the OP levels in Nam Co lake water were close to the

284   values reported for ocean waters, such as, the North Atlantic and Arctic oceans (with

285   DDTs and PCBs mostly below the detection level, or <1 pg L^{-1}) (Gioia et al., 2008;

286   Lohmann et al., 2009). By contrast, high levels of PAHs were found in the Nam Co

287   lake water, ranging from 6.9 to 83.6 and 1.7 to 28 ng L^{-1} for the dissolved and SPM

288   phases, respectively. The dissolved $\sum_{15}$PAH levels were one order of magnitude

289   higher than those reported for Himalayan high-altitude lakes in Nepal (Table S15)

290   (Guzzella et al., 2011) and the Great Lakes (Table S15) (Venier et al., 2014), and two

291   orders of magnitude greater than values for open oceans (Table S15) (Ma et al., 2013)

292   and European mountain lakes (Table S15) (Vilanova et al., 2001).

293   **3.2 Possible sources**

294   Long range atmospheric transport (LRAT) is considered an important source

295   contributing to the occurrence of OPs in remote environments (Dalla Valle et al.,

296   2005). Considering that the prevailing climate system operating over Nam Co in

297   summer is the Indian Monsoon, if the seasonal pattern of a chemical is similar to that

298   of the monsoon, monsoon transport may therefore be the source of OPs in Nam Co air.

299   Thus, the interrelationship between monsoon intensity (Indian Monsoon Index, IMI,

300   W m^{-2}) and OP concentrations was investigated (Figure 3). Figure 3 shows that

301   α-HCH and *o,p'*-DDT displayed synchronous seasonal variation with the IMI. This

302 suggests that monsoon transport was the principal reason for the occurrence of OCPs

303 in the Nam Co atmosphere. In addition, isomer ratios can provide insight on the

304 source and fate of the OPs. In this study, we found that the isomer ratios of *p,p'*-DDT

305 to *p,p'*-DDE were broadly in agreement with those found for the source regions of

306 India and the Bay of Bengal (Table S16) (Gioia et al., 2012; Zhang et al., 2008).

307 Similar to other remote regions, such as, the Arctic (Hung et al., 2010), Antarctic

308 (Baek et al., 2011), Rocky Mountains (Daly et al., 2007), and southeastern TP (Sheng

309 et al., 2013), in which LRAT is the primary transport mode of OPs, the dominance of

310 α- to γ-HCH was observed in the Nam Co atmosphere (Table S16). Results of isomer

311 ratios associated with the seasonal variations supported the interpretation that OCPs in

312 the Nam Co atmosphere had undergone LRAT. In contrast to OCPs, neither the

313 gaseous nor the particulate phases of PAHs showed a clear and consistent seasonal

314 variation during the two years of air monitoring (Figure S3), which is likely because

315 there were primary emissions of PAHs surrounding the Nam Co region.

316 Apart from the seasonal trends, spatial distribution patterns can also provide valuable

317 information on OP sources. The spatial distributions of OPs in the surface water

318 across the Nam Co Lake are presented in Figure 4. First, HCHs showed a uniform

319 distribution (Figure 4) without significant differences among the different regions of

320 the lake (Table S17). The even distribution of HCHs in the water was most likely

321 caused by the LRAT origins and relatively higher water solubility. Second, relatively

322 high levels of HCB and PAHs occurred in water from the northwestern and eastern

323 parts of the lake (Table S17 and Figure 4). The elevated HCB and PAHs in these

324 regions were likely related to anthropogenic activity in the vicinity. As shown in

325 Figure 1b, two townships (Baoji and Namco), which have the highest population

326 around Nam Co Lake, are located in the northwestern and eastern corners of the lake.

327 Following a traditional lifestyle, the residents use large amounts of local biomass

328 (mostly yak dung) for cooking and heating (Xiao et al., 2015). High PAH

329 concentrations have been reported in local Tibetan tents that were emitted mainly

330 from burning yak dung (Li et al., 2012). A ratio of BaA/(BaA+Chr) = 0.33 was

331 recommended as a specific diagnostic fingerprint for yak dung combustion (Li et al.,

332 2012). The BaA/(BaA+Chr) ratios observed in our study (0.27±0.08 for air and

333 0.24±0.10 for water) were in good agreement with this diagnostic ratio. This suggests

334 that local combustion emission is likely the source of PAHs in Nam Co. With the

335 exception of PAHs, biomass combustion can also produce HCB (Bailey, 2001), which

336 may be the reason for the higher HCB concentrations occurring around the townships.

337 The spatial distribution of OPs in the Nam Co lake water highlights the important

338 contribution of local sources for PAHs and HCB.

339 **3.3 LRAT versus re-volatilization**

340 From the above results, we found that LRAT is a key factor that determines the

341 seasonality of the atmospheric HCHs and DDTs in Nam Co (higher concentrations

342 occurred in summer). However, high temperatures generally occur during summer,

343 which may promote the evaporation of chemicals from local surfaces (e.g., soils and

344 water bodies). To what extent does this re-evaporation contribute to the atmospheric

345 OPs? The Clausius-Clapeyron (C.C.) equation can be used to assess this probability

346 (Wania et al., 1998b). If a strong relationship is found between the partial pressure of

347 atmospheric OCPs and the air temperature, this indicates that volatilization may occur.

348 Otherwise, low temperature dependence will occur in the case of LRAT. In the

349 present study, the results of the C.C. equation are summarized in Table S18. The

350 correlation with temperature ($p>0.05$, Table S18) for most chemicals was not

351 significant, except for α-HCH, which displayed a relatively lower correlation

352 coefficient (R^2=0.29, $p<0.05$, Table S18). This indicates that weak volatilization of

353 α-HCH from local surfaces at Nam Co may exist, while the re-evaporation of other

354 chemicals is limited.

355 Enantiomers of chiral OPs have been used to distinguish the contribution of LRAT

356 and re-volatilization of OPs from surfaces (Bidleman et al., 2012). For example,

357 technical HCH contains the (+)- and (-)-α-HCH enantiomers in a racemic proportion

358 (EF=0.5). Abiotic processes (transport, hydrolysis, and photolysis) do not favor either

359 enantiomer, while only biological processes, such as, microbial degradation in soils

360 and water, show enantioselectivity and will alter the EFs of α-HCH (Ridal et al.,

361 1997). Therefore, nearly racemic signatures usually indicate input from LRAT, while

362 nonracemic signatures represent the influence of local microbial degradation. In the

363 present study, both the enantiomeric signatures of α-HCH in air and water were

364 measured simultaneously from May to September. As shown in Figure 5a, all the lake

365 water samples showed a selective depletion of (+)-α-HCH, with EFs ranging from

366 0.318 to 0.449. This has previously been reported for other cold oligotrophic water

367 systems, such as the Arctic lakes (EFs: 0.359−0.432) (Law et al., 2001). From Figure

368 5a, we found that extensive enantioselective degradation occurred in June and July,

369 which coincided with the bacterial bloom period (Figure 5b) (Liu et al., 2013). This

370 negative correlation between the EF of α-HCH in lake water and bacterial abundance

371 is presented in Figure S4. Law et al. (2001) suggested that under low nutrient

372 conditions, oligotrophic bacteria are able to use xenobiotic carbon sources, such as

373 α-HCH. This implies that the Tibetan lake microbes can also metabolize, or

374 cometabolize, α-HCH.

375 If high temperatures favor the evaporation of α-HCH from the lake water, depletion of

376 (+)-α-HCH should be observed for air. However, the EFs of the air samples were

377 overall racemic. This is similar to the racemic composition observed in the

378 atmosphere over Indian regions (Huang et al., 2013), which is the potential source

379 region of HCHs in Nam Co. With respect to the enantiomeric signature in air samples

380 from June and July, only some (+) α-HCH depletion was observed in air (Figure 5a),

381 indicating weak evaporation of α-HCH from the lake water. Combined with the EF

382 values in air and water, the fraction of the contribution from lake water volatilization

383 (*f*) can be quantified by (Huang et al., 2013):

$$f = (EF_a\text{-}EF_b)/(EF_w\text{-}EF_b) \tag{8}$$

385 where EF_a and EF_w are the EF values in air and water, respectively; and EF_b is the

386 background EF value in air, which was assumed to be the average EF of the standard.

387 The estimated results show that only 19% and 17% of atmospheric α-HCH came from

388 water volatilization in June and July, respectively, demonstrating that LRAT is indeed

389 the major source (more than 80%) of α-HCH. This result is in contrast with the

390 conclusion of Xiao et al. (2010) that evaporation from Nam Co Lake largely

391 contributed to the atmospheric α-HCH concentration. In that study, both levels and

392 enantiomeric signatures of α-HCH in Nam Co lake water were absent.

393 **3.4 Atmospheric processes**

394 **3.4.1 Air-water gas exchange**

395 Although some α-HCH evaporation was recorded in June and July, the air-water

396 exchange process during the entire ablation period is of great concern as this is the

397 main season transferring pollutants between air and water. Fugacity ratios (f_w/f_a) and

398 net exchange fluxes (F_{AW}, ng m^{-2} day^{-1}) were quantified using paired air-water

399 samples collected from May to September in 2014. The average exchange status

400 (average of f_w/f_a) for HCHs, HCB, PCB 28, and PAHs during the ablation period is

401 illustrated in Figure 6. Because of DDTs, Ant and Fla were not quantified in the lake

402 water (Table S10) and were therefore excluded from the discussion. α- and γ-HCH

403 had low f_w/f_a values ranging from 0.08 to 0.15, and 0.02 to 0.08, respectively (Figure

404 6a). The low f_w/f_a ratio suggests that α- and γ-HCH were overall prone to deposition

405 from air to water during the ablation period. The deposition fluxes were -1.6 ± 0.4 ng

406 m^{-2} day^{-1} for α-HCH and -1.0 ± 0.2 ng m^{-2} day^{-1} for γ-HCH, which are within the

407 same order of magnitude as those reported for the Great Lakes (Khairy et al., 2014).

408 Connected to the source of HCHs discussed above, this result implies that the

409 following LRAT-deposition event of HCHs occurred in the ablation period of Nam Co

410 Lake. In terms of β-HCH, HCB, and PCB 28, their f_w/f_a ratios were either overlapping

411 with the equilibrium range (0.5-1.5) or on the edge of deposition threshold. Therefore,

412 low deposition fluxes for β-HCH (-0.2 ng m^{-2} day^{-1}) and PCB 28 (-0.1 ng m^{-2} day^{-1}),

413 and large variability for HCB (-1.0 ± 0.6 ng m^{-2} day^{-1}) were observed (Figure 7a).

414 The results of the air-water gas exchange for PAHs are presented in Figure 6b. The

415 fugacity ratios of thirteen PAHs varied depending on their molecular weight and

416 volatility (Figure 6b). Acel, Ace, and Flu showed f_w/f_a values significantly higher than

417 1.5 (Figure 6b), indicating that the lake acted as a secondary source for these volatile

418   chemicals. The f_w/f_a values for Phe covered a large range (from 0.3 to 3), showing a

419   shift between volatilization and deposition (Figure 6b). Other high molecular weight

420   (MW > 202) PAHs, including Pyr, BaA, Chr, Bbf, Bkf, Bap, IcdP, DahA, and BghiP,

421   favored net deposition with f_w/f_a values lower than 0.5 (Figure 6b). Greater

422   volatilization fluxes were observed for Acel, Ace, and Flu (3-ring), which could reach

423   up to 203 ng m^{-2} day^{-1} (Figure 7b). Whereas, the gaseous deposition fluxes for high

424   molecular weight PAHs were two orders of magnitudes lower and only varied from

425   -1.0 to -4.6 ng m^{-2} day^{-1} (Figure 7c). Although average deposition fluxes of 339 ng

426   m^{-2} day^{-1} were calculated for Phe, the deposition fluxes showed large variability ($\pm$604

427   ng m^{-2} day^{-1}). This result is broadly consistent with the exchange direction revealed by

428   the f_w/f_a values, implying that the exchange of Phe between air and water may be

429   reversed during the entire ablation period.

430   **3.4.2 Reversal of the air-water exchange of Phe**

431   In section 3.4.1, we observed that both the air-water exchange direction and the flux

432   of Phe showed a large range of values and uncertainties. This raises a question about

433   what drives this variation. The monthly calculated f_w/f_a and F_{AW} of Phe during the

434   ablation period showed that the volatilization of Phe occurred during May and June,

435   but deposition begun in July, which represents a reversal (Figure 8). Given that lake

436   ice begins melting during May, the melted ice may discharge large amounts of

437   accumulated PAHs into the lake, causing the relative enrichment (high fugacity) of

438   Phe in the water, and triggering the secondary emission of Phe from the water to the

439   atmosphere. This was confirmed by the increased water concentration of Phe found

440   during May and June (Table S10). This is also the reason why a large uncertainty of

441   F_{AW} was observed for Phe during the ablation period. Linked to the source of PAHs

442   discussed above, the final exchange status of PAHs is the combined effects of the

443   depositional input caused by biomass burning, the properties of the chemical, and the

444   melting of lake ice.

445   Seasonal ice cover is an important feature of water bodies in cold regions. In the

446   Arctic region, Jantunen et al. (2008) and Wong et al. (2011) both observed the

447   occurrence of volatilization of OCPs from seawater coincident with the breaking up of

448   ice cover in the summer. The Nam Co Lake also undergoes long periods of ice-cover

449   (Liu et al., 2013). During the winter, the lake surface is covered by ice and gas

450   exchange is restricted, meanwhile dry and wet deposition exerts a significant

451   influence on the input inventory of PAHs to the lake. Both of these two deposition

452   processes are one-way (no volatilization), which keeps the contaminants being

453   accumulated. As summer arrives, the lake ice begins to thaw and air-water gas

454   exchange begins to dominate. On the one hand, after the higher accumulation of

455   deposition, supersaturation of PAHs in the lake may occur. On the other hand, the

456   fugacity capacity of ice is much higher than that of water, and therefore the decrease

457   of the fugacity capacity during melting will increase the fugacity of the PAHs (Wania

458   et al., 1998c), which also promotes their re-emission from the water. Although the

459   seasonal ice cover did not show any obvious influence on the fate of OCPs and other

460   PAHs, it played an important role in the fate of Phe, which was a dominant compound

461   in the Nam Co atmosphere. The lake therefore acted as a secondary source of Phe in

462   May and June, and shifted to a net sink during other months, which is likely driven by

463   the seasonal freeze-thaw cycle of lake ice (Figure 8).

464   **3.4.3 Atmospheric degradation**

465   Reactions with the hydroxyl radical (OH) are an important removal process of

466   gaseous OPs from the atmosphere. The resulting degradation fluxes (F_{deg}, ng m^{-2} day^{-1})

467   are dependent on the concentration of OH radicals in the air (Spivakovsky et al., 2000)

468   and the compound-specific degradation rate constant (K_{OH}, cm^3 molecules^{-1} day^{-1}).

469   The K_{OH} values of gaseous OCPs and PAHs are from Brubaker and Hites (1998) and

470   Keyte et al. (2013), respectively. Due to the lack of information on K_{OH} for β-HCH

471   and BaA, their degradation fluxes (F_{deg}) were not considered in this study. The

472   calculated F_{deg} values were averaged for individual OPs and are presented in Figure

473   S5. The degradation fluxes for α-, γ-HCH, HCB, and PCB 28 ranged between 0.3 and

474   0.9 pg m^{-2} day^{-1} (Figure S5), 3 orders of magnitude lower than their F_{AW}. This

475   indicates that the contribution of atmospheric degradation to their total inventory in

476   the environment is negligible.

477   In contrast to the OCPs, the PAHs are more susceptible to photodegradation

478   (Lohmann et al., 2009). In our study, lower molecular weight PAHs showed higher

479   degradation fluxes, such as 4–184 ng m^{-2} day^{-1} for Phe, and 1–160 ng m^{-2} day^{-1} for

480   Ant (Figure S5). These values are similar to those reported for F_{deg} in the remote

481   atmosphere of the Atlantic Ocean (i.e., 7–120 and 9–50 ng m^{-2} day^{-1} for Phe and Ant,

482   respectively) (Nizzetto et al., 2008). We observed relatively low F_{deg} values for 5- and

483   6-ring PAHs, ranging from 0.01 to 0.18 ng m^{-2} day^{-1} (Figure S5). Generally, the F_{deg}

484   of all PAH compounds was one order of magnitude lower than their F_{AW}. OH

485   depletion is the primary process that removes atmospheric PAHs, presumably causing

486   the continuous volatilization of low molecular weight PAHs from the water. This

487   raised questions about other processes that may have supplied PAHs into the lake

488   water. On the other hand, OH degradation also decreases the input of high molecular

489   weight PAHs into the water and it is unclear to what extent this degradation

490   counteracts other deposition processes.

491   **3.4.4 Atmospheric deposition**

492   In addition to the gas exchange, dry and wet deposition are also important processes

493   that influence the input of OPs from air to the lake. Dry (F_{DD}) and wet (F_{WD})

494   deposition fluxes were estimated using the method described above (section 2.6).

495   With respect to HCHs, HCB, and PCB 28, their dry deposition fluxes (F_{DD}) were

496   negligible due to their low detection frequency in the particulate phase (Table S9).

497   However, the average F_{WD} for α-HCH, β-HCH, and γ-HCH was -0.3, -0.9, and -0.4 ng

498   m^{-2} day^{-1}, respectively, which is comparable to their F_{AW} levels. F_{WD} for HCB (-0.02

499   ng m^{-2} day^{-1}) and PCB 28 (-0.002 ng m^{-2} day^{-1}) was two magnitudes lower than their

500   F_{AW}. In general, precipitation scavenging is most efficient in HCHs compared with

501   the other chemicals (Carrera et al., 2002). Greater wet deposition fluxes of HCHs

502   occurred in August (Figure S6), coinciding with the highest amount of precipitation in

503   Nam Co. Combining the F_{AW} and F_{WD} of HCHs, the estimated annual input of HCHs

504   from air into the whole lake (2015 km^2) was 1.9 kg year^{-1}. This result highlights the

input of HCHs by the LRAT-deposition process during the ablation period (open water season). Snow scavenging of HCHs has been reported as an important clearing process in mountain regions (Kang et al., 2009). However, the transport of HCHs in winter is very limited due to the unfavorable air circulation patterns (westerly wind), ruling out the significant contribution of input of HCHs by snow scavenging.

Compared with OCPs, the close association between PAHs and the particulate phase accounted for their relatively higher deposition fluxes. The estimated dry and wet deposition fluxes for individual PAHs during the ablation and frozen periods, respectively, are provided in Table 1. We found that the F_{DD} values of PAHs for the ablation period are, in general, lower than those for the frozen period. For example, the F_{DD} of total $\sum_{15}$PAHs increased by one order of magnitude from the ablation period (4.5 ng m^{-2} day^{-1}) to the frozen period (38 ng m^{-2} day^{-1}; Table 1). Two factors may lead to the increase of F_{DD} in winter: the increased wind speed during the winter season and the growing particulate-PAH concentrations due to the enhanced combustion activities in winter. Compared with other studies, the estimated F_{DD} for the total $\sum_{15}$PAHs (4.5–38 ng m^{-2} day^{-1}, this study) is broadly within the range reported for global oceans (8.3–52.4 ng m^{-2} day^{-1}) (Gonzalez-Gaya et al., 2014).

Wet deposition was found to be the dominant deposition process for the input of PAHs into Nam Co (Table 1). This was expected because precipitation scavenging of organic chemicals underlies the accumulation of pollutants in mountain regions (Tremolada et al., 2008). In addition, there was an obvious difference between the values of F_{WD} during the ablation and frozen periods. For the total 15 PAHs, the F_{WD} in the frozen period (702 ng m^{-2} day^{-1}) was approximately 5 times higher than that for the ablation period (161 ng m^{-2} day^{-1}), which may be due to the different precipitation types between these two periods (snow *vs.* rain). Snow has been suggested to be more efficient than rain for scavenging particulate-PAHs, which had a high concentration during winter in Nam Co (Table S9). Thus, although the precipitation of Nam Co in winter is low (less than 30 mm, Figure S2), the strong scavenging ratio of snow to PAHs combined with the relatively high particulate-PAHs concentration in winter

534   caused the enhanced PAHs deposition in winter. The frozen season coincided with the

535   period of high emission and high deposition of PAHs, implying a significant

536   contribution of this season in the input of PAHs into the lake.

537   To calculate the comprehensive contribution of all above-mentioned processes, three

538   groups of PAHs were classified in Table 1 based on their fate during the air-water

539   exchange processes. PAHs (Acel, Ace, and Flu) showing volatilization behavior were

540   placed into one group, PAHs with large F_{AW} variability between the status of

541   volatilization and deposition were in the second group; and the remaining PAHs

542   displaying deposition behavior were placed into the third group (Table 1). In this

543   classification, although the air-water exchange direction and fluxes of Ant and Fla

544   cannot be estimated, we still placed them into the second group because of their

545   similarity to Phe in their physicochemical properties. For the volatilization group, the

546   total outgassing from the lake was estimated to be approximately 126 kg per year,

547   which cannot alone be supplied by their total deposition flux (sum of F_{WD} and F_{DD}).

548   This suggests that there may be additional natural sources of PAHs in the lake, such as,

549   degradation of pigments carrying aromatic structure and turnover of organic matter

550   (Nizzetto et al., 2008). Regarding the deposition group (Pyr, BaA, Chr, Bbf, Bkf, Bap,

551   IcdP, DahA, and BghiP), their total deposition flux ($F_{AW}+F_{DD}+F_{WD}$) will roughly

552   cause the annual input of 208 kg high molecular weight PAHs into the lake. Although

553   the F_{AW} of Phe was reversed and the total volatilization of Phe was estimated at

554   around 26 kg year^{-1}, this loss will be complemented by the continuous deposition of

555   Phe (~373 kg year^{-1}) from July to April. This indicates that the annual net input of Phe

556   will be above 340 kg, which suggests that Phe is the most dominant contributor in the

557   total PAHs deposition. In addition to the 15 PAHs considered here, there are other

558   PAHs with high abundances, for example alkylated phenanthrenes, which will drive a

559   much larger depositional fluxes to the lake.

3.4.5 Uncertainties in flux estimation

561   Several factors were involved in the uncertainties of the flux estimation: (i) loss

562   during sample extraction and clean-up; (ii) measurement errors; and (iii) accuracy of

563   the parameters in meteorology and physicochemical properties. The air-water gas

564   exchange flux (F_{AW}) is the most important contributor to the total inventory of PAHs

565   into the lake. The uncertainty involved in F_{AW} was estimated by propagating the

566   errors in C_a (30%), C_w (35%), K_{ol} (40%), and H (20%), which was 64%. This small

567   error demonstrates that the estimate of the gas exchange fluxes was relatively robust.

568   By contrast, the uncertainties in other fluxes were higher. The uncertainty of F_{DD} was

569   estimated as a factor of 3 (Gonzalez-Gaya et al., 2014). However, the uncertainties of

570   F_{deg} and F_{WD} are difficult to quantify due to unavailable data on the relative errors of

571   K_{OH}, W_G, and W_P. For example, the scavenging rates of PAHs by wet deposition were

572   highly variable, which was caused by the complexity of the size distribution of

573   aerosols, meteorological conditions, and the scavenging process (Jurado et al., 2005).

574   Considering these aggregated uncertainties, the estimated total fluxes here are only

575   expected to capture the order of magnitude for the different processes. In addition,

576   other input processes into the lake, such as, glacier meltwater, river runoff, and soil

577   erosion may also occur in this study region, which will lead to an underestimation of

578   the total input flux.

579   **3.5 Implication for the regional carbon cycling**

580   Lakes are increasingly recognized as an important component of the terrestrial carbon

581   cycle (Tranvik et al., 2009). Nearly 50% of the area of Chinese lakes is located on the

582   TP, with general oligotrophic conditions and a total lake area of >43000 km^2 (Zhang

583   et al., 2014). Compared with other components, such as grassland and forest, organic

584   carbon burial in Tibetan lakes has been largely ignored. Although our study only

585   focused on one of these lakes (Nam Co, area = 2015 km^2), we can extrapolate the

586   annual atmospheric deposition of $\sum_{15}$PAHs into the remaining Tibetan lakes, and

587   estimate it at 8.7 tonnes C, when expressed as carbon fluxes (Gonzalez-Gaya et al.,

588   2016). In addition to these 15 PAHs, there are other carbon sources such as soot, DOC,

589   PAHs derivatives, and other anthropogenic organic compounds, which would become

590   a significant allochthonous carbon source for the oligotrophic lakes in TP. Because the

591   Tibetan lakes are low in nutrients, bacteria in the lake have adapted to using a wide

range of organic compounds and growing under starvation conditions (Liu et al., 2009). Recently, bacteria from the genus *Sphingomonas* were detected in Nam Co lake water and various glacier snows of the TP (Liu et al., 2013; Liu et al., 2009), and they were reported to have the ability to degrade PAHs (Leys et al., 2005). The presence of these bacteria in Nam Co suggests that the atmospheric inputs of organic pollutants can act as a carbon source to support the survival of Tibetan microbial communities. Despite the natural PAH background in the environment, increasing biomass burning has led to the accumulation of PAHs in the lake sediments, especially during the past 50 years (Yang et al., 2016). Therefore, the continuous atmospheric deposition of various PAHs and their ecological impact deserve greater concern.

4. Conclusions

This study confirmed that the Nam Co Lake was still a net sink of HCHs, following the LRAT-deposition process, rather than a secondary source. By contrast, PAHs primarily originated from local biomass burning. Dominated by gas exchange and wet deposition, the air-water fluxes of $\sum_{15}$PAHs to the whole Nam Co Lake were estimated to be 550 kg year^{-1}, providing a substantial carbon source for the oligotrophic lake. Among the PAHs compounds, Phe showed a distinct behavior with monthly reversals of the air–water exchange, which was most likely driven by the seasonal melting of lake ice. This hypothesis requires further investigation, and a passive sampling technique is recommended as a viable alternative to enhance the spatial coverage of the investigation of air-water exchange in the TP.

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Table 1. Estimated fluxes (ng m^{-2} day^{-1}) of air-water gas exchange (F_{AW}), atmospheric degradation (F_{deg}), dry deposition (F_{DD}), and wet deposition (F_{WD}) for individual PAHs during the ablation period and frozen periods, respectively.

PAH	ablation period				PAH	frozen period		
	F_{AW}	F_{deg}	F_{DD}	F_{WD}		F_{deg}	F_{DD}	F_{WD}
Volatilization compounds								
Acel	80 ±49	6 ±5	NA	-0.2 ±0.1	Acel	1 ±0.7	-0.02 ±0.001	-0.03 ±0.1
Ace	51 ±19	4 ±4	-0.003 ±0.002	-0.4 ±0.3	Ace	0.9 ±0.5	-0.01 ±0.01	-6 ±5
Flu	203 ±162	11 ±8	-0.1 ±0.02	-9 ±7	Flu	1.8 ±0.9	-0.2 ±0.2	-43 ±30
sum	**335**	**21**	**-0.1**	**-9**	**sum**	**4**	**-0.2**	**-49**
Phe	-340 ±604	82 ±67	-0.5 ±0.1	-42 ±35	Phe	10 ±4	-2.2 ±1.2	-345 ±237
Ant	NA	60 ±63	-0.04 ±0.03	-5 ±5	Ant	4 ±2	-0.11 ±0.05	-16 ±14
Fla	NA	5 ±5	-0.5 ±0.1	-20 ±18	Fla	0.5 ±0.3	-4.6 ±2.8	-93 ±64
Deposition compounds								
Pyr	-145 ±154	20 ±21	-0.4 ±0.1	-18 ±17	Pyr	2 ±1	-3 ±1.5	-128 ±83
BaA	-19 ±23	NA	-0.1 ±0.1	-3 ±4	BaA	NA	-1.1 ±0.5	-15 ±10
Chr	-54 ±62	7 ±8	-0.5 ±0.3	-47 ±56	Chr	0.2 ±0.1	-4.7 ±2.3	-19 ±13
Bbf	-5 ±3	0.2 ±0.1	-0.6 ±0.5	-6 ±5	Bbf	0.02 ±0.01	-2.2 ±3.2	-4 ±8
Bkf	-2 ±1	0.2 ±0.1	-0.4 ±0.4	-2 ±1	Bkf	0.1 ±0.04	-3.8 ±1.9	-8 ±5
Bap	-2 ±1	0.2 ±0.2	-0.3 ±0.5	-3 ±1	Bap	0.04 ±0.03	-4.7 ±2.3	-16 ±10
IcdP	-2 ±1	0.7 ±0.5	NA	-2 ±2	IcdP	0.1 ±0.1	NA	-3 ±6
DahA	-1 ±0.7	0.1 ±0.1	NA	-0.1 ±0.2	DahA	0.01 ±0.01	NA	-0.6 ±1
BghiP	-2 ±0.4	0.02 ±0.01	-1 ±1	-3 ±1	BghiP	0.01 ±0.01	-12 ±6	-6 ±3
sum	**-231**	**28**	**-3**	**-85**	**sum**	**2**	**-31**	**-199**
Total PAHs	\	**196**	**-4.5**	**-161**	**Total PAHs**	**20**	**-38**	**-702**

NA: not available; For F_{AW}, F_{DD} and F_{WD}, positive values indicate volatilization, and negative values indicate net deposition.

(a)

885 (b)

886

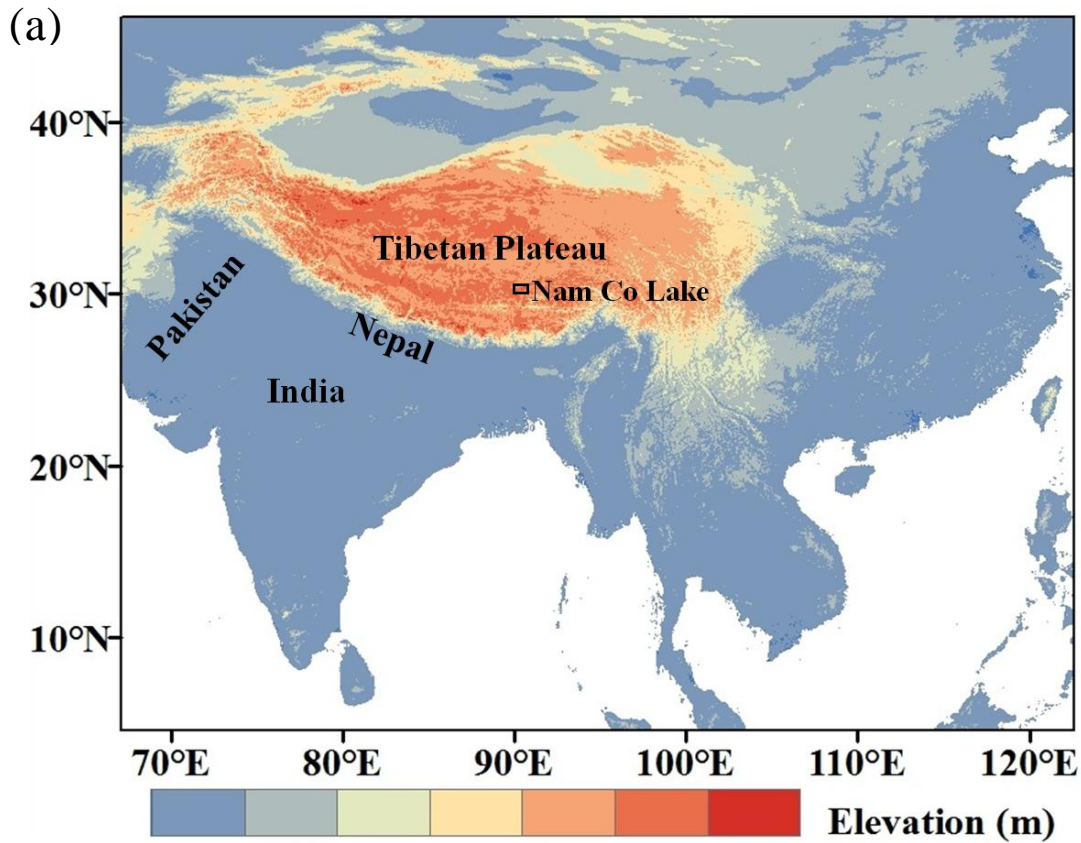
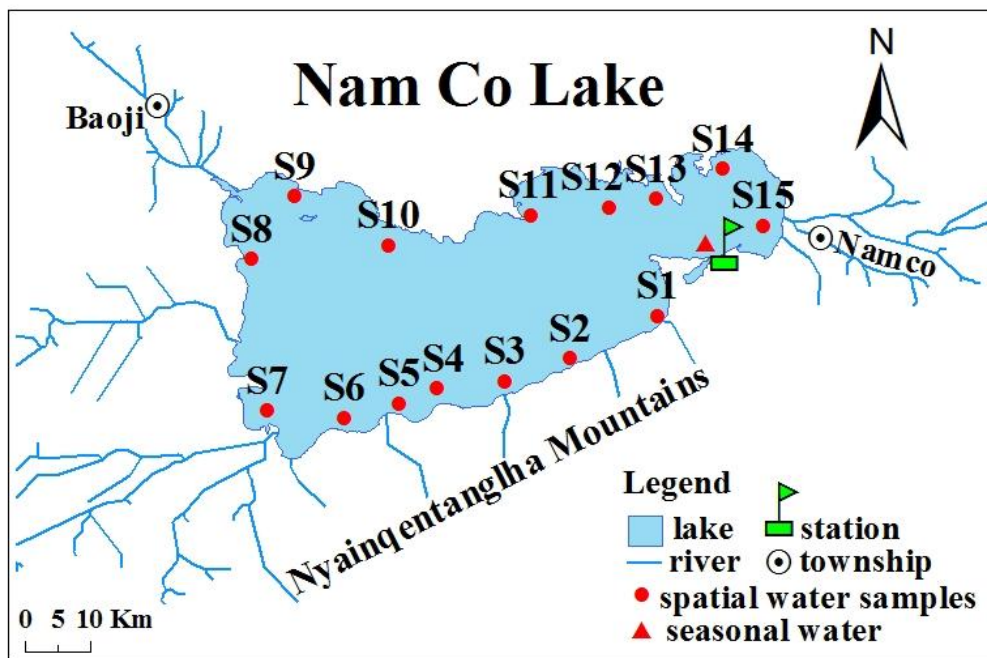
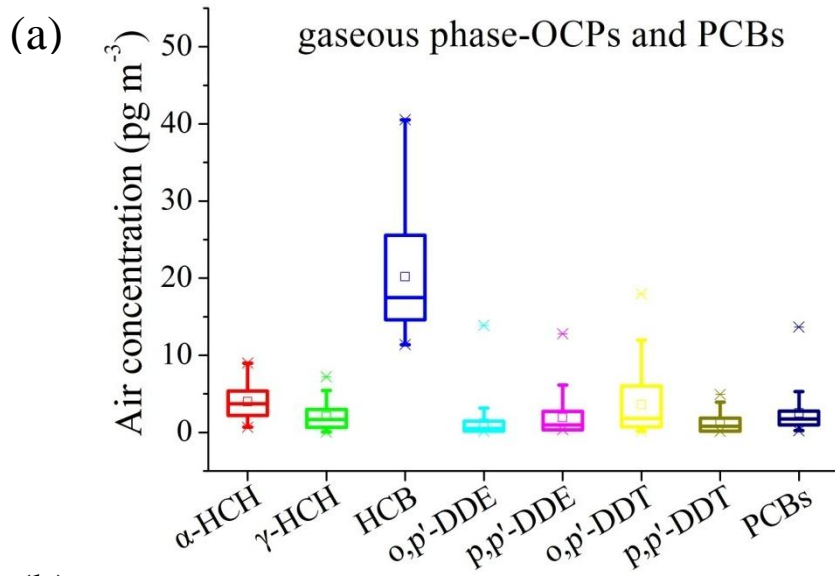



Figure 1. Location of Nam Co Lake on the Tibetan Plateau (a) and the sampling
sites for air and lake water (b). The station refers to the Nam Co Monitoring and
Research Station, and it is also the air sampling site; S01 to S15 represent the 15
sampling sites of surface water around the lake; the red triangle represents the
sampling site of seasonal water from May to September.

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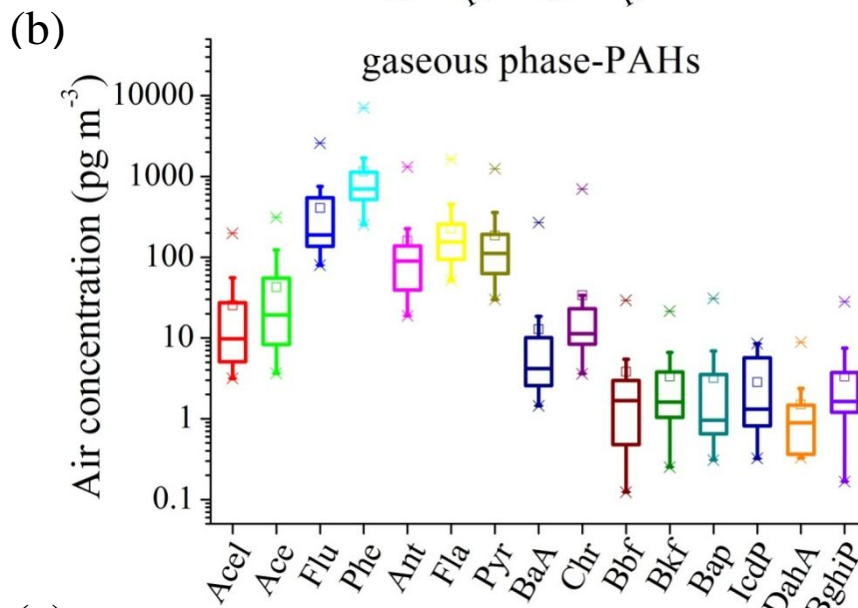
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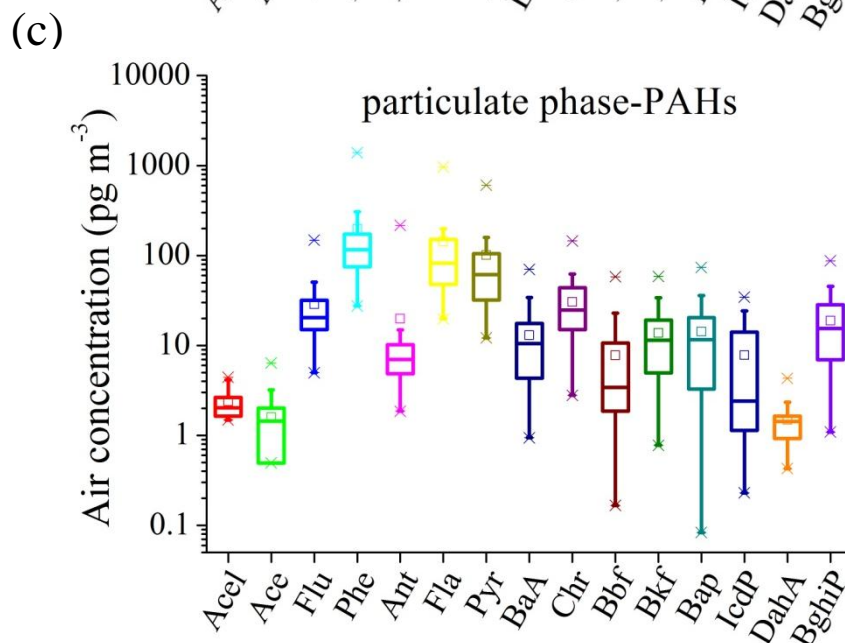

894

895 **Figure 2. Air concentrations of gaseous OCPs and PCBs (a), gaseous PAHs (b),**
896 **and particulate phase PAHs (c) in Nam Co. The boxes are defined by the 25th**

and 75th percentiles; whiskers mark the 10th and 90th percentiles; the median is represented by a horizontal line; the mean by a square; and outliers with an asterisk.

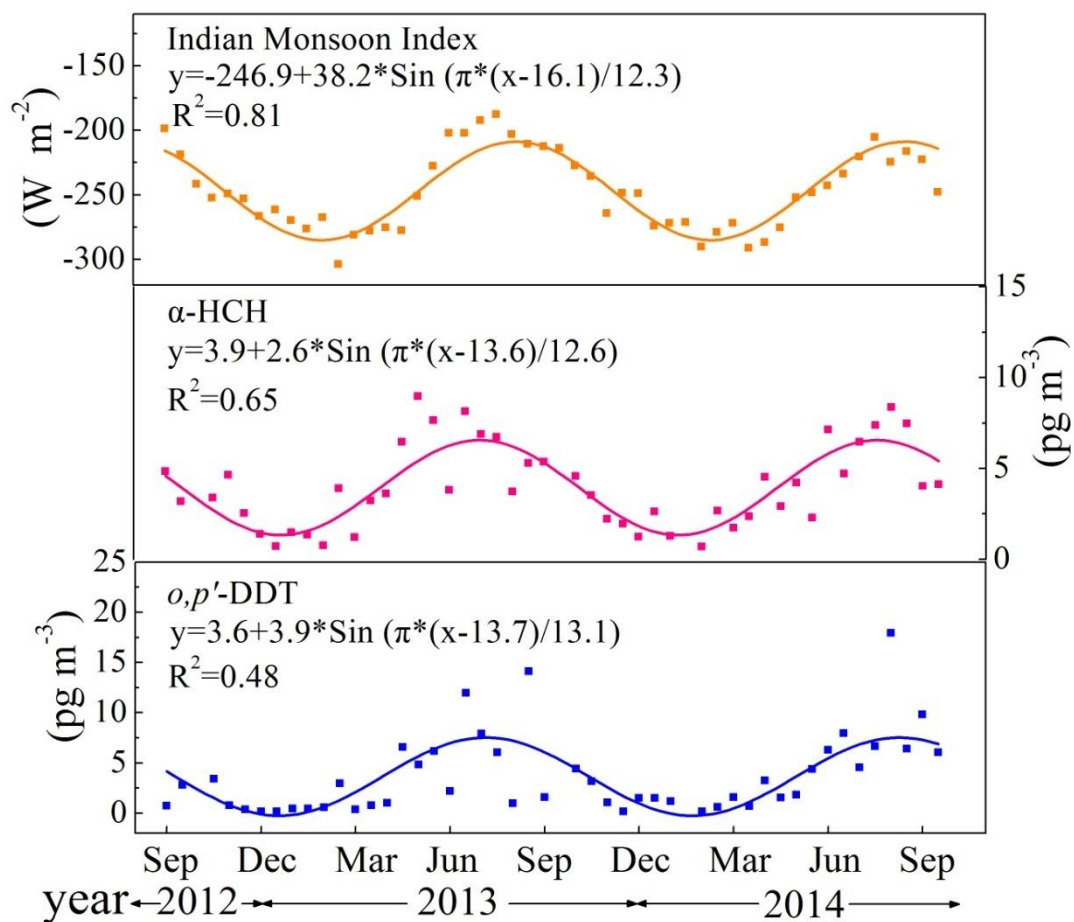
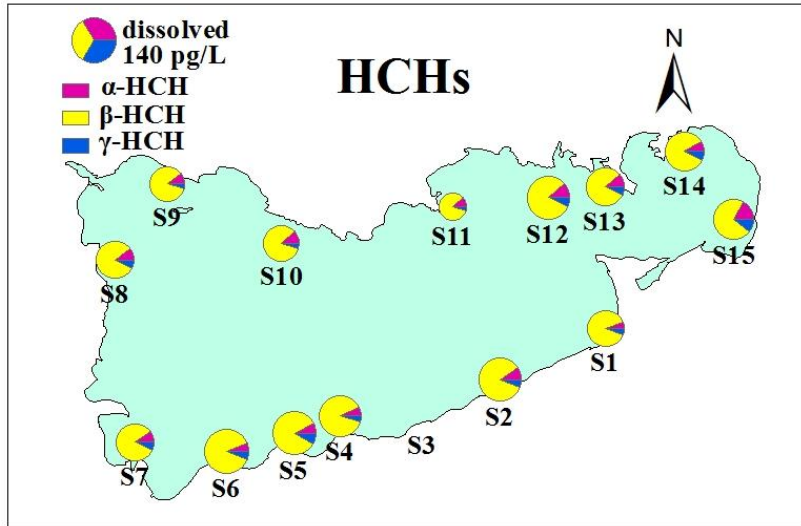



Figure 3. Seasonal patterns of Indian Monsoon Index, the atmospheric concentrations of α-HCH and *o,p′* -DDT.

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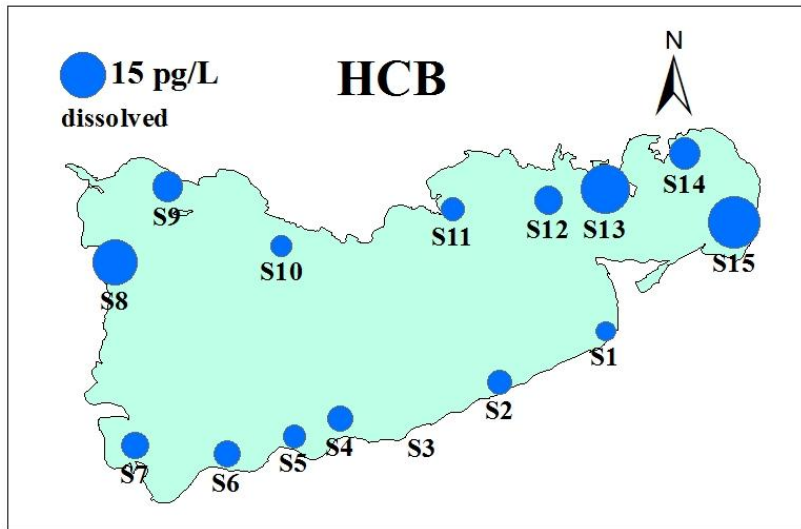
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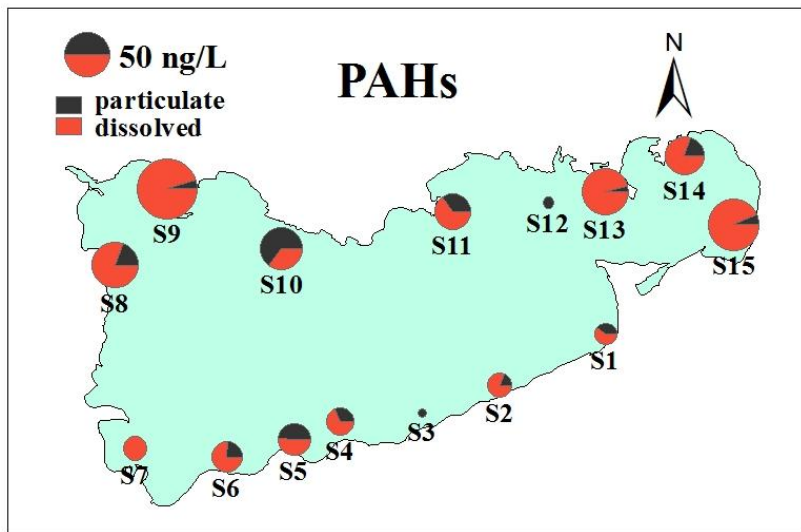
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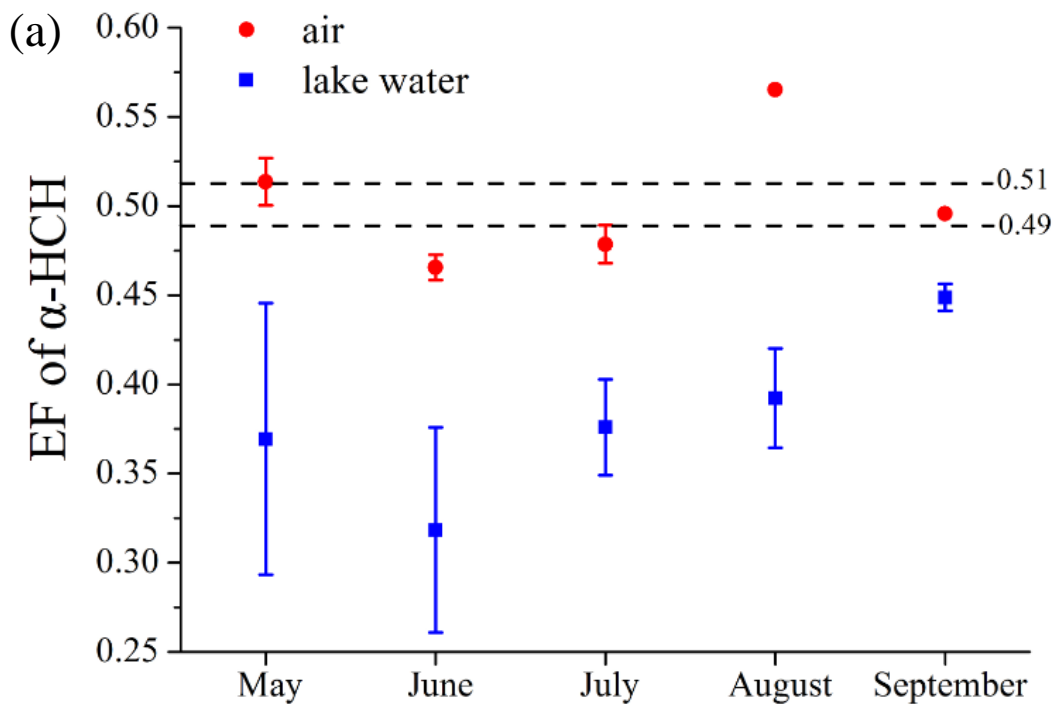
906 **Figure 4. Spatial distribution pattern of HCHs, HCB, and PAHs in the surface**

907 **water of Nam Co Lake.**

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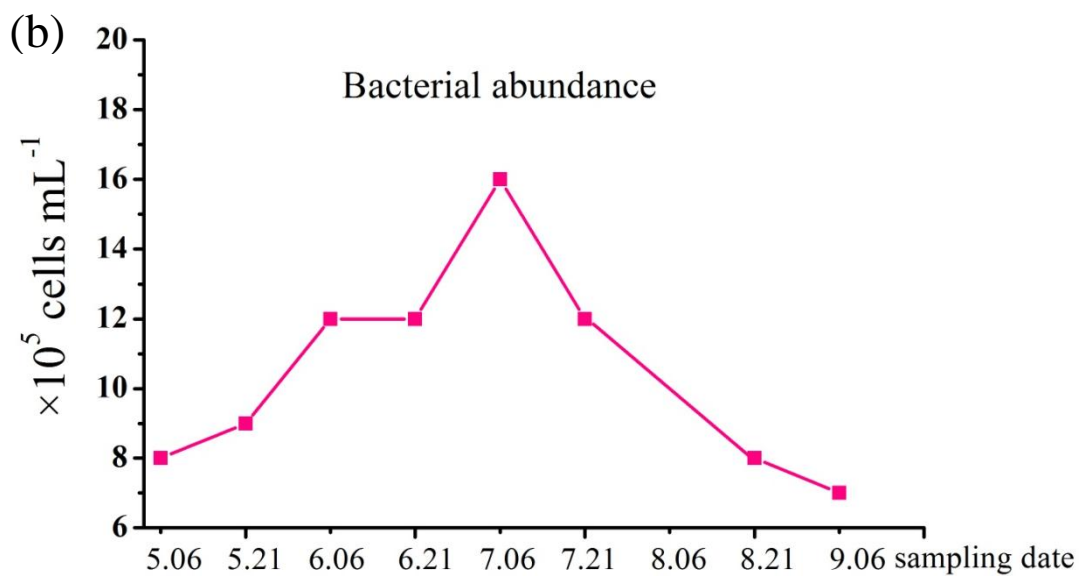
909

910 **Figure 5. Enantiomer fraction (EF) of α-HCH in the air and surface water from**

911 **May to September (a), and the seasonal bacterial abundance in Nam Co Lake**

912 **water (b). The data of bacterial abundance was derived from Liu et al. (2013),**

913 **which represents the total bacteria in the lake surface water.**

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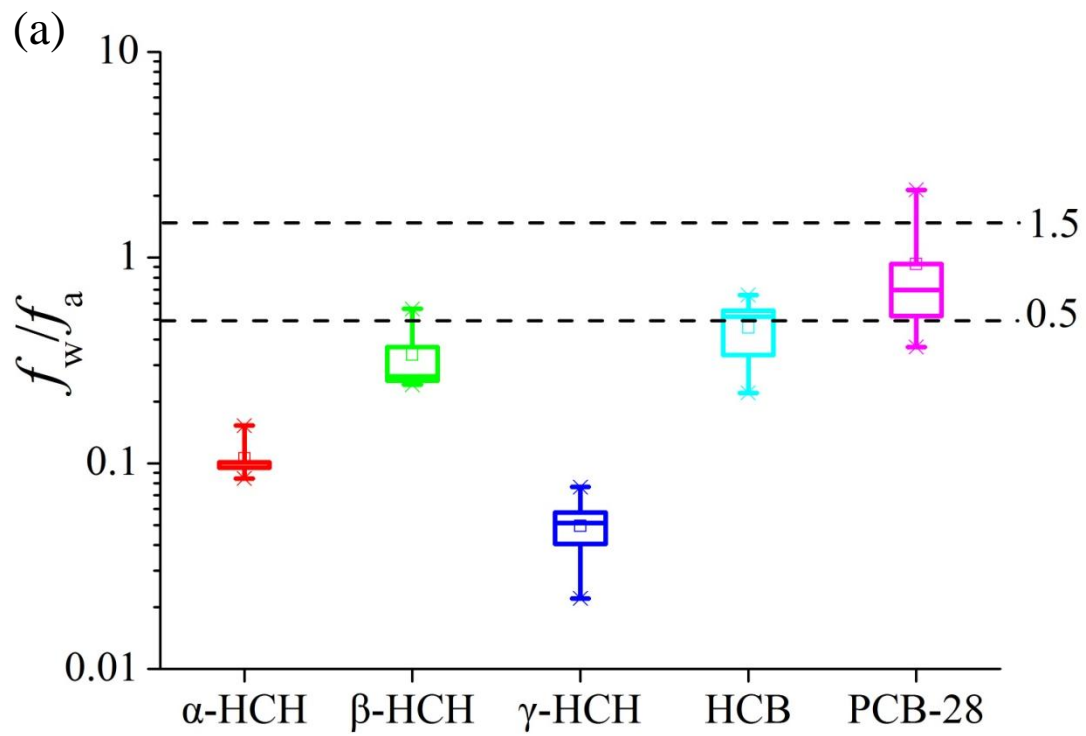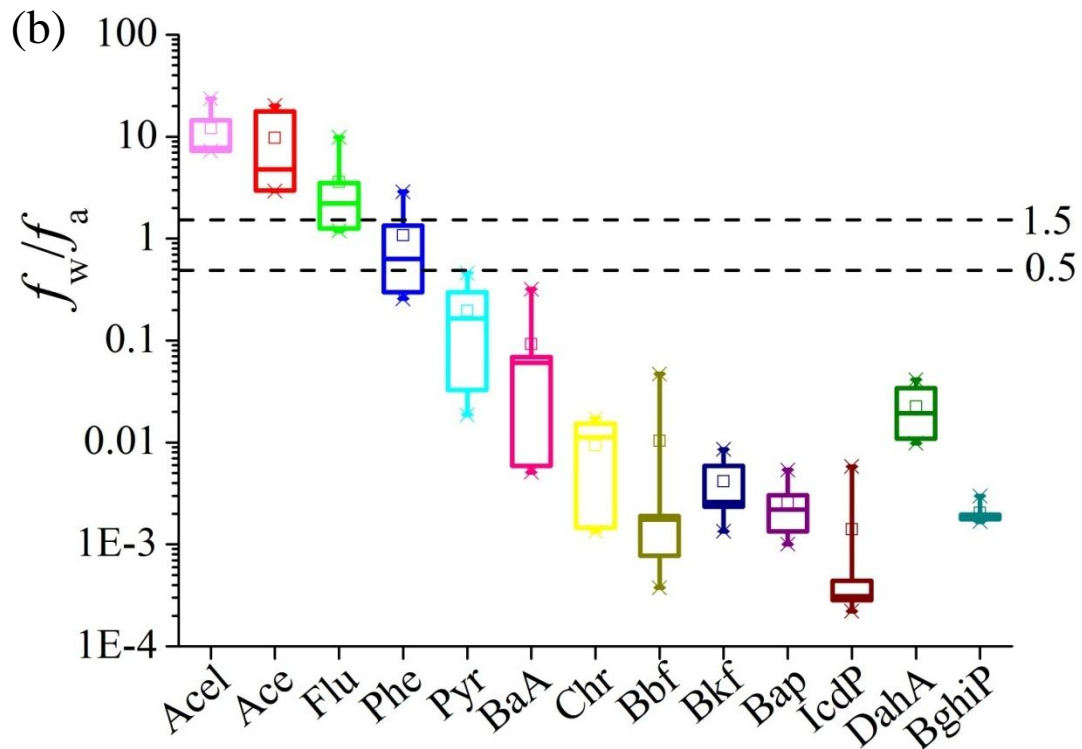
Figure 6. Water/air fugacity ratios (f_w/f_a) for OCPs and PCB 28 (a), and individual PAHs (b) in Nam Co Lake. The horizontal lines represent the uncertainty range, 0.5-1.5 was considered as equilibrium.

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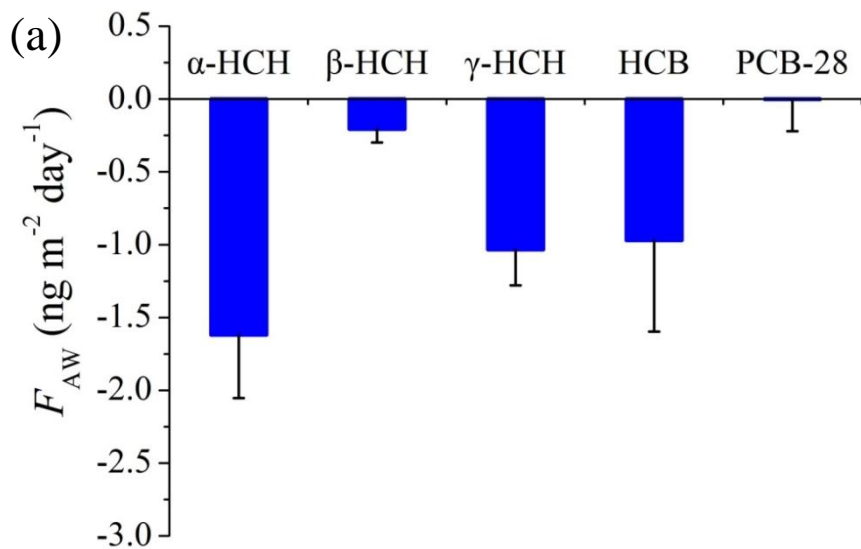
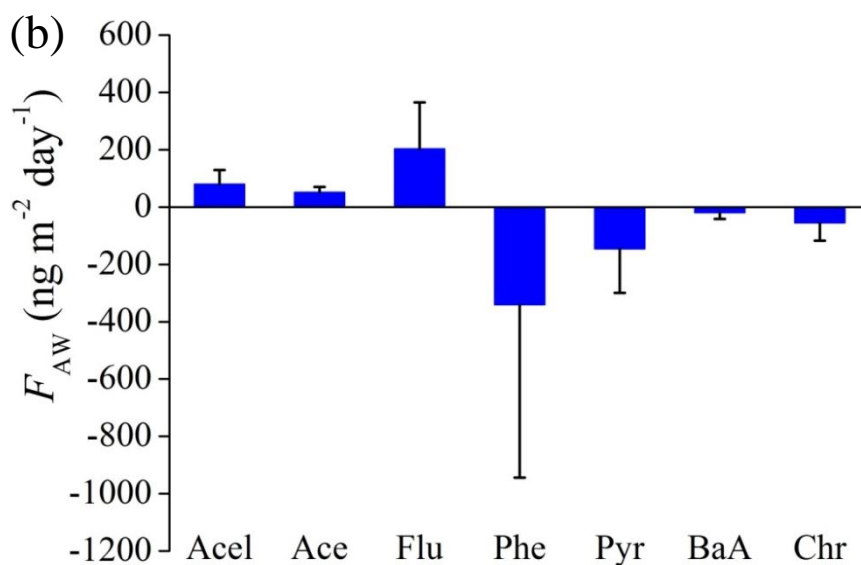
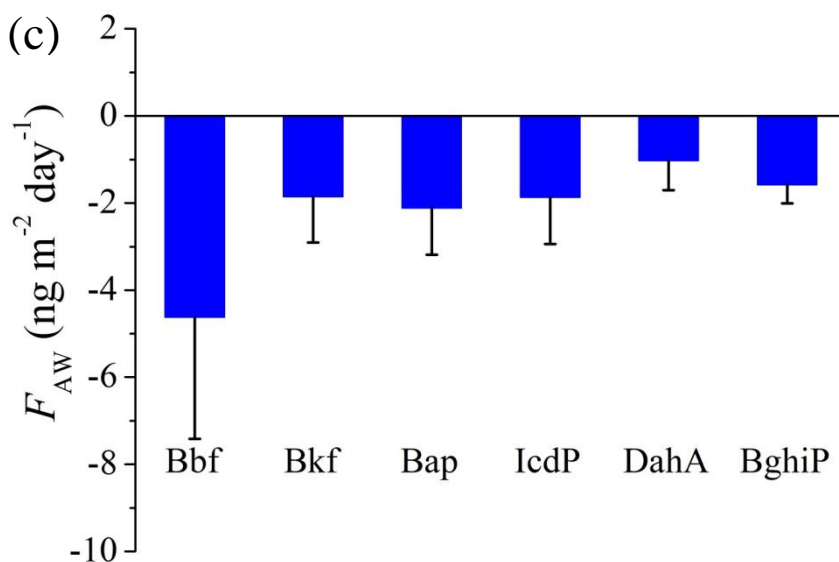
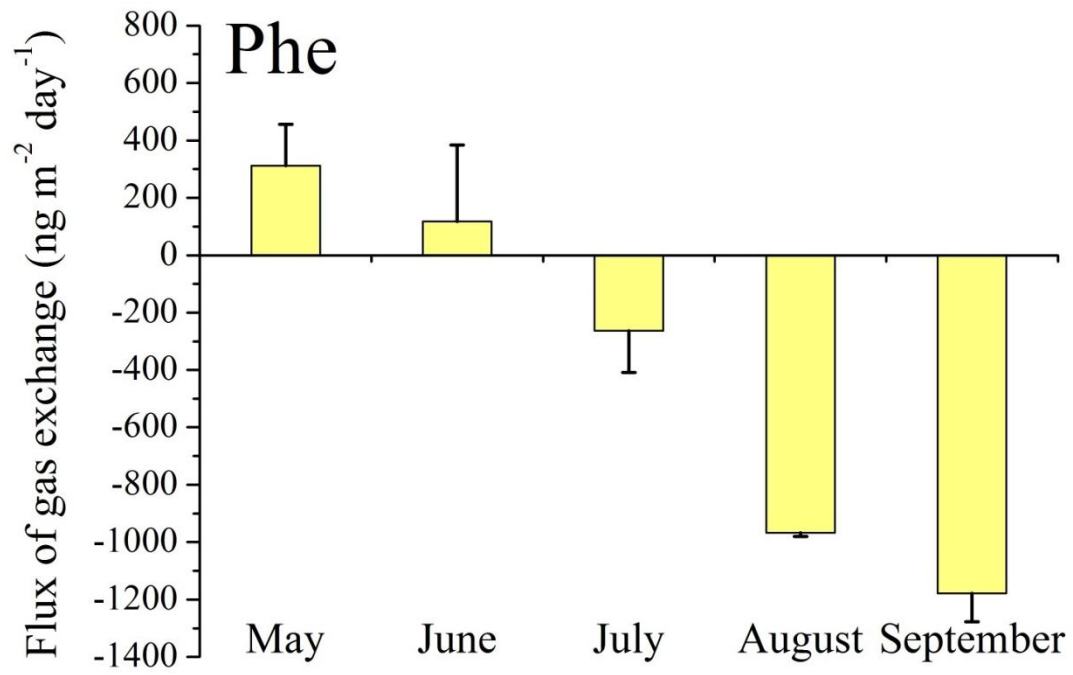


Figure 7. Average air-water gas exchange fluxes (F_{AW}) for individual OCPs, PCB 28 (a), and PAHs (b, c) in Nam Co Lake. Positive values indicate net volatilization, and negative values indicate net deposition.

926
927 **Figure 8. Reversal of the air-water gas exchange for Phe from May to September.**