

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.

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 °C, 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:

$$RSD^2_{FR}=RSD^2_{C_w}+ RSD^2_{C_g}+ RSD^2_H$$

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 Monsoon 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.

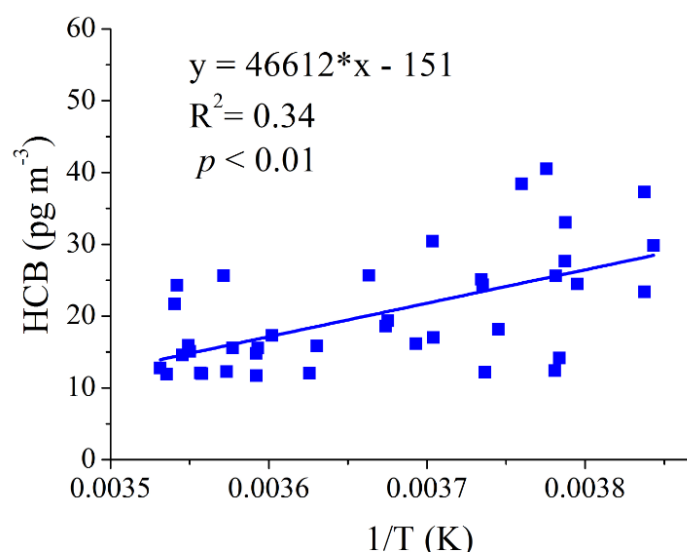


Figure. The relationship of atmospheric HCB with the reciprocal of temperature (T)

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.