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. 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 $1.3 \times 10^9$ $m^3$ $year^{-1}$.

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.

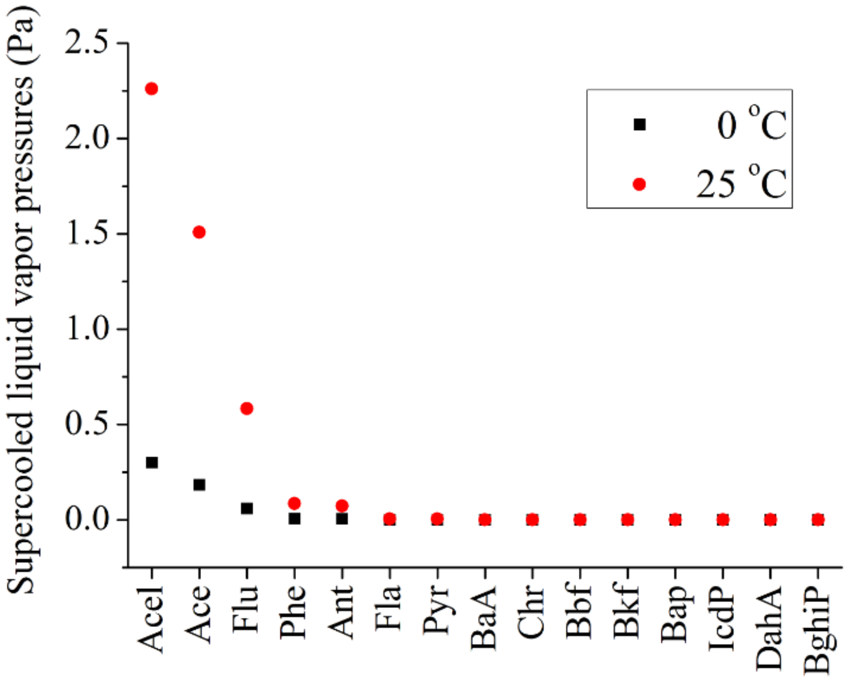

Figure. Supercooled liquid vapor pressures ($P_L$) of PAHs under different temperatures

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.

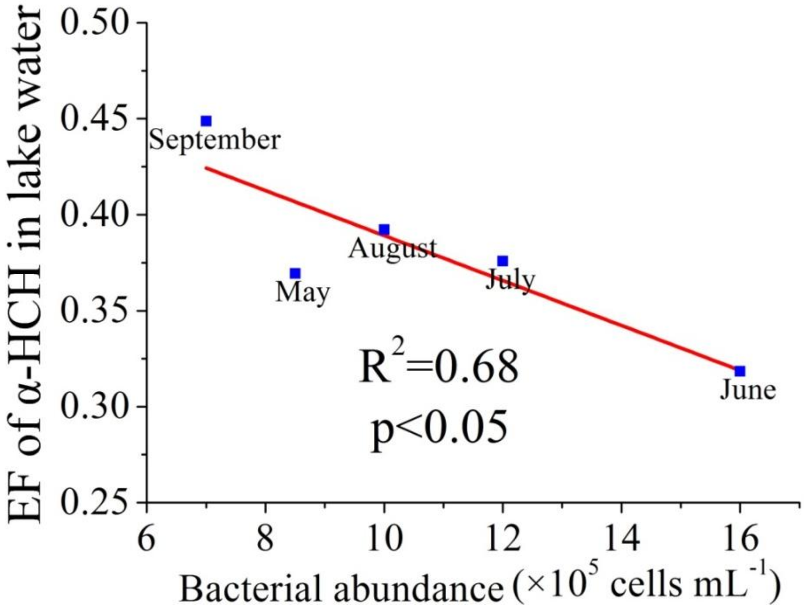

Figure. The correlation between EF of α-HCH in the lake water and seasonal bacterial abundance in Nam Co Lake.

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.