

Review of: Jiao Ren et al., Atmospheric processes of persistent organic pollutants over a remote lake of the central Tibetan Plateau: Implications for regional cycling (MS No.: Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-989, 2016).

This is an excellent study of atmospheric processes for PAHs and chlorinated compounds in Lake Nam Co. Considerable thought and effort was put into the sampling design, analysis and interpretation of results. Procedures and data are well summarized in the extensive set of tables and figures of the Supporting Information. The paper is very well written, figures and tables are clear and informative.

I am giving the paper a “accept after minor revisions” recommendation, because there are some points that require attention. My comments below are by line numbers in the ACPD paper.

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.

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 ($\text{m}^3 \text{y}^{-1}$)? 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.

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 .

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

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 than 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?

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?

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).

159. Please give units of C_G and C_W . They should be mol m^{-3} as used in Eq. 1 and 2.

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 (C_W)”. 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.

$$C_W = C_{XAD}/(1 + K_{DOC}C_{DOC})$$

Doing this correction implies that C_{XAD} includes BOTH the free POPs and POPs associated with DOC, in contradiction to the earlier statement (see above, “According to Ruge...”).

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?

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)?

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?