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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Drivers of atmospheric deposition of polycyclic aromatic hydrocarbons at European high altitude sites" *by* Lourdes Arellano et al.

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Reviewer's Comment

Dear Editor, I am mostly satisfied with the Author's replies. I can truly understand that they have done the best measurement design they could. I have only one additional remark concerning the Air-Water exchange process: Namely in many studies (Tsapakis et al., ES&T 2006, Giglioti et al. Environmental Toxicology and Chemistry 2002, and othersâAËŸC for the sea; Lohmann et al. Environ. Sci. Technol., 2015 for lakes (Lake ′ Superior) it was shown that the absorptive air-water flux dominates atmospheric loadings (e.g. wet, dry particle) to water for PAHs of molecular weight < 234, which also are the main members found within the lakes in the present study. I believe it would be important for the paper to add a brief discussion and mention that the mixed wet and dry deposition is only part of the inputs.

Authors' response

We agree with the referee that air-water exchange is an important process to take into account in the context of atmospheric inputs to aquatic ecosystems, mainly for low molecular weight PAH, which dominate the composition of the atmospheric gas phase. However, air-water exchange is a dynamic process that includes not only inputs, but also losses from the water column to air. The net air-water fluxes depend on the PAH concentrations in the atmospheric gas phase and water dissolved phase, among other factors such as wind speed or temperature. However, these processes can represent inputs or losses of pollutants from the water column. Thus, Tsapakis et al 2006 (Environ. Sci. Technol., 40, 4922-4927) observed a net absorption of low molecular weight PAH in the eastern Mediterranean Sea, more important than the atmospheric loads by wet and dry deposition, which, in contrast, dominated the inputs of high molecular weight PAHs. Ruge et al, 2015 (Environ. Sci. Technol., 49, 13777-13786), obtained similar results for Lake Superior, although differences in flux direction were observed between sites and seasons. Net inputs to the water were determined in areas near urban and industrialized sites, whereas net volatilization was observed in open lake sites far from point sources.

Authors' changes in manuscript

A brief discussion about these processes and their relative importance has been introduced in section 3.5. Atmospheric PAH deposition and lacustrine sedimentary fluxes, as follows: Comparison of the PAH atmospheric deposition and lacustrine sedimentary fluxes showed much higher values in sediments, i.e. 24-100 μg m-2 yr-1 and 120-3000 μg m-2 yr-1, respectively (Table 6). All lakes considered in this comparison are located in high mountain areas and their hydrological regime is determined by atmospheric precipitation into the watershed. The strong difference in flux values of the direct PAH atmospheric and sedimentary measurements may respond to processes such as the sediment focusing (Rowan et al., 1995) or lake sediment concentration of these hydrocarbons falling into the surface of the lake catchment. Studies at low altitude (seawater) have reported that air-water exchange is the most important process for low molecular weight PAH inputs into aquatic systems, exceeding the wet and dry deposition (Tsapakis et al., 2006; Ruge et al., 2015). In these high mountain lakes, the average concentrations of volatile PAHs are lower than in low altitude aquatic systems , e.g. air and water concentrations of phenanthrene 0.99 ng m-3 (Fernandez et al., 2003; van Drooge et al., 2010) and 180 ng m-3 (Vilanova et al., 2001), respectively, vs. 3.3-16 ng m-3 and 450-5600 ng m-3, in marine systems (Gigliotti et al., 2002; Tsapakis et al., 2006). The smaller concentrations in high mountains should involve lower gas-water transfer gradients (Nelson et al., 1998). Irrespectively of these values, flux calculations in some of these high mountain lakes for compounds with properties similar to those of PAH showed that the main transfer essentially occurs from water to air (Meijer et al., 2009). The overall mass balance involved pollutant incorporation into the lake waters due to atmospheric precipitation and a substantial degassing to the atmosphere. Thus, in these lakes the air-water transfer processes cannot explain the higher sedimentation fluxes in comparison to atmospheric precipitation. Regarding to the contributions of PAH inputs from the watershed, an estimated sediment flux was calculated considering that the total amount of atmospheric PAHs deposited in the lake and its catchment area were accumulated in the lake sediments (Table 6)...

References added:

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