Reviewer 1.

The manuscript presents particle and PAHs monthly deposition fluxes in the European mountain areas. This manuscript also addresses the number of sites, sampling frequency and period of study comprise the most comprehensive approach performed so far on PAH deposition in high mountain areas. This is a worthwhile manuscript and can be accepted for publication in the ACP after minor revision as per some comments given below:

In introduction section Please add some recent studies of (2016) and (2017) for including the sources, abundances/distributions and long-range transport of PAHs such as. . . (Singh et al. 2017; Distributions of Polycyclic Aromatic Hydrocarbons, Aromatic Ketones, Carboxylic Acids, and Trace Metals in Arctic Aerosols: Long-range Atmospheric Transport, Photochemical Degradation/ Production at Polar Sunrise Environ. Sci. Technol. 2017, 51, 8992−9004).

Authors’ response:

The recent study by Singh et al., 2017 indicated by the reviewer has been included in Line 97. In addition, some other studies of 2016-2017 and 2018 have been also included:

Kirchgeorg et al., 2016, Environ Poll, 218; 804-812 in Line 112.

Yang et al., 2016, Environ Poll, 214, 1-7 in Line 99

Feng et al., 2017, Environ Poll, 230, 639-647 in Line 95

Zhang et al., 2018, Atmos Environ, 173, 256-264 in Line 94

Reviewer comment:

Line-342. These emissions involve the release of large amounts of particulate matter containing PAH. . .. . .Please provide the reference.

Authors’ response:

In fact, this statement is based on the results observed in this study; however, this is confirmed by the results observed by van Drooge et al., 2010 (Environ Sci Pollut Res 17:1207-1216), therefore this reference has been included in Line 343.

Reviewer comment

Line- 345. Some PAHs, e.g. benzo[a]pyrene, benzo[ghi]perylene and coronene, also showed significant negative correlations with temperature (p < 0.05;). . .. . .Please also mention the R2 value here.

Authors’ response

The following information has been added: B[a]P (R2=0.585), B[ghj]Per (R2=0.516 at p<0.001) and Cor (R2=0.561).

Reviewer comment

Line-397. Air masses from central/east Europe are negatively correlated with PAH deposition fluxes, mainly for HMW-PAH. . .. Please provide the reference.

Authors’ response

In fact, this is a result of the present study. We have referred to Table 5 to clarify it (Line 399).

Reviewer comment

Line 434, 437 and Figure 4 . . .. . .Please keep the R2 value at the two decimals such as (r2 = 0.9958 . . .. It should be (R2 = 0.99).

Authors’ response

Done as suggested

Reviewer comment

Please recheck the complete manuscript where mentioned PAH and PAHs, it should be mentioned with grammar and suitability of sentences.

Authors’ response

We have checked manuscript and corrected it when suitable

Reviewer 2

Reviewer’s general comments and suggestions:

Is that wet deposition and dry deposition samples were not separately collected and studied.

Authors’ response: This is beyond the scope of our study. The collection of these two fractions separately had required much higher sampling manpower that was not available in the research projects supporting the study.

Reviewer comment

Air-water exchange was not calculated for lakes. (See Tsapakis et al. 2006).

Authors’ response:

This is beyond the scope of the study, since the objective was not to perform a complete mass balance of PAHs in the studied lakes, but to evaluate the magnitude of their atmospheric deposition in these remote sites and it relation with their accumulation in sediments. In the case of PAHs, the measured deposition fluxes in the lakes already provide the net flux to the sediments.

Reviewer comment

The authors use rather obsolete literature: e.g. P. 3, lines 90-95 (20 y old!). There many studies after 2005 that could be used a literature.

Authors’ response: We updated the literature, adding studies that are more recent

Singh et al., 2017, in Line 97

Kirchgeorg et al., 2016, Environ Poll, 218; 804-812 in Line 112.

Yang et al., 2016, Environ Poll, 214, 1-7 in Line 99

Feng et al., 2017, Environ Poll, 230, 639-647 in Line 95

Zhang et al., 2018, Atmos Environ, 173, 256-264 in Line 94

Reviewer comment

The authors, in the Experimental Section and Table 1, should report which of the 15 EPA list PAHs are considered as LMW and HMW compounds.

Authors’ response: We have specified the compounds included in the LMW and HMW PAH groups the first time these acronyms appears in the text (Lines 267 and 376). However, they are already specify in the footnote of Table 1.

Reviewer’s comment

Since they compare with previous studies in which more PAHs were determined. They should also indicate the number of PAHs in the sum (\_) (see Tables).

Authors’ response

In principle, we have tried, when possible, to sum the 15 PAH included in the EPA list (all excepting naphthalene) as it is indicated in the Table 2 (where the comparison is made). However, this was not always possible, since many studies do not indicate the number of PAHs considered or/and the concentrations of each individual compound that can be used to get comparable PAH sums.

Reviewer’s comment

The interpretation of the presence of different PAHs on the basis of their diagnostic concentration ratios should be given very cautiously, since the individual compounds (especially the more reactive ones) are subjects to important degradation mechanisms during their atmospheric transport and before deposition and reaching the lacustrine environment (P. 8-9; l. 285-305).

Authors’ response: We agree with the author. We already consider the effects of photooxidation considering insolation and sampling location in lines 286-309.

Reviewer’s comment

Discussing the deposition fluxes and comparing them with those determined in different environments it would useful for the readers to focus on specific compounds. E.g. Those with the highest abundance.

Authors’ response

We agree with the author. This is performed in Table 2 where only the 15 compounds selected by the EPA (which includes the most abundant parent compounds) and a specific PAH, benzo[a]pyrene usually taken as a reference compound, are considered.

Reviewer 2 Additional comments

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 othersâA˘C 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.

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)…

Related references added:

Nelson E.D., McConnell L.L., Baker J.E. Diffusive exchange of gaseous polycyclic aromatic hydrocarbons and polychlorinated biphenyls across the air-water interface of the Chesapeake Bay. Environ Sci Technol32, 912–919, 1998.

Meijer, S.N., Grimalt, J.O., Fernández, P. and Dach, J. Seasonal fluxes and temperature-dependent accumulation of persistent organic pollutants in lakes: The role of internal biogeochemical cycling. Environ. Poll*.* 157**,** 1815-1822, 2009

Ruge, Z., Muir, D., Helm, P., and Lohmann, R.: Concentrations, Trends, and Air–Water Exchange of PAHs and PBDEs Derived from Passive Samplers in Lake Superior in 2011, Environ. Sci. Technol., 49, 13777-13786, 10.1021/acs.est.5b02611, 2015