

Interactive comment on “Bi-directional air-sea exchange and accumulation of POPs (PAHs, PCBs, OCPs and PBDEs) in the nocturnal marine boundary layer” by Gerhard Lammel et al.

Anonymous Referee #3

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This paper described a study of the gas and particle deposition flux of POPs over the marine boundary layer based on vertical gradient measurements. The results indicate both upward and downward fluxes of POPs occurred, and the direction of the fluxes changed frequently for some POPs. Emissions of POPs were driven by volatilization from the sea during daytime and nighttime for some POPs. Long-term measurements are needed to validate the results and apply them to modeling studies. This paper is suitable for publication after addressing the comments below.

Line 34: This sentence is not necessarily specific to the marine atmospheric environment; continental atmospheres are also impacted by land sources and contaminated soil emissions of POPs. Please also correct the grammar error, “which are advected

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from sources on land, primary and secondary, such as volatilization from contaminated soils”. I suggest revising to, “which are advected from primary sources on land and secondary sources, such as volatilization from contaminated soils.”

Lines 42-43: What are the sources of POPs to surface waters that can lead to a buildup of concentrations? Does deposition alone lead to elevated POP concentrations in surface water?

Lines 80-81: Please explain why these two heights were chosen for determining the vertical gradient. How does height selection potentially affect the vertical concentration and flux gradients?

Line 146: All equations should be labeled with numbers as well as in the text chronologically.

Line 223: I suggest replacing with “universal gas constant R ($\text{Pa m}^3 \text{mol}^{-1} \text{K}^{-1}$), and both sea surface temperature (SST or T_w) and salinity corrected. . .”

Lines 277-283: Only gas-phase pollutants are volatilize from the sea surface. Please mention if the ratios are based on gas-phase concentrations or gas+particulate concentrations and include the range of these ratios over the sampling period in addition to the average.

Lines 283-284: What could be the reasons for the lower $C_{\text{day}}/C_{\text{night}}$ ratio for chlorinated compounds specifically?

Lines 302-304 and lines 329-336: Volatilization tends to occur during daytime. Please explain how this process contributes to the elevated nighttime concentrations. Could wind speed be another factor controlling volatilization of gases other than temperature?

Lines 336-341: The temperature difference between daytime and nighttime (0.5-1.5K) is too small to explain the differences in daytime and nighttime concentrations and fluxes. What would be the corresponding difference on sea surface temperatures?

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Lines 363-365: What are the implications of frequent changes in the direction of air-sea exchange in terms of estimating the deposition budget of POPs? If the fluctuations are so frequent, it would be difficult to obtain an accurate deposition budget.

Fig. 2: Label which series is the predicted and observed data.

Lines 382-384: The vertical flux gradient of gases is based on a height difference of 1.8 m. How representative is this flux estimate of the overall deposition flux?

Line 388: It is not clear which height the concentration corresponds to in this equation for determining the deposition velocity. Is it the average concentration of the two heights or just the ground-level concentration?

Line 422: New sentence is needed after “reason”

Line 432: It is not clear which height the concentration corresponds to in this equation for determining the particle deposition flux.

Lines 446-450: The lack of particle size distribution data is a major source of uncertainty in the dry deposition flux calculations, since lower molecular weight PAHs are likely to partition onto particles of various sizes. See the variation of aerosol dry deposition velocities with particle sizes in Petroff and Zhang (2010, Geoscientific Model Development).

Lines 470-473: Could the upward flux of particulate-phase POPs be related to sea-salt emissions from waves?

Lines 492-493: The meaning of the sentence, “Fluctuation of more substances might have been hidden by the method’s uncertainties.” is unclear. Did you mean insufficient number of POPs analyzed or not enough samples collected?

Title: The paper is lacking some discussion specifically on the accumulation of POPs in the nocturnal marine boundary layer, as stated in the title. I feel the paper discussed the diel variation (both daytime and nighttime) of POPs.

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Supplement: There are two Table S6 in the supplement.

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