We would like to thank for the comments and suggestions. We have addressed all comments below and have indicated the corresponding modifications in the revised version of the manuscript. The line numbers mentioned in our responses refer to those in the ACPD paper.

Anonymous Referee #3

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.

True, but will be left unchanged, as the statement itself is correct and nothing is or is intended to be said about continental atmospheres.

Please also correct the grammar error, “which are advected 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.”

To be corrected by shortening. The new text will read: “The marine atmospheric environment is a receptor for persistent organic pollutants (POPs) which are advected from primary and secondary sources on land.”

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?

Will be specified. The new text will read: “…fed by riverine or atmospheric deposition input.”

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?

Will be explained. The new text will read: “According to the Monin-Obukhov similarity theory of turbulent transport in the surface layer (Monin and Obukhov, 1954; s. supplement, sect. S1.3), vertical concentration profiles are expected to be logarithmic, while – due to the vertically non-linear behaviour of the eddy diffusivity – the resulting vertical flux is *per definitionem* constant. For a given surface source (sink) of gases or particles corresponding negative (positive) vertical concentration gradients ($\partial c/\partial z$) will be maximal close to the source (sink). Therefore, over aerodynamically smooth surfaces, the lower inlet height should be preferably in the order of a few tenths of a meter, while the distance to the upper inlet height, should be maximized to yield large (and consequently statistically significant) vertical concentration differences. However, the choice for the upper inlet height is limited by the horizontal extension of the so-called "fetch", i.e. the up-wind surface homogeneity (in terms of orography, structure, and vegetation), which should be at least $100 \times z_2$ (e.g. Foken, 2008). In the case of our study, the choice of the lower inlet height (z_1=1.05 m) is just due to the design of the aerosol sampler which does not allow sampling below 1.05 m above ground, while the upper inlet height (z_2=2.80 m) accounts for the limited surface homogeneity (200 m) of the Selles Beach site."

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

Line 223: I suggest replacing with “universal gas constant R (Pa m3 mol-1 K-1), and both sea surface temperature (SST or Tw) and salinity corrected: : :”
Will be done

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.
Will be specified. The new text will read: “For PAHs the mean ratio of day/night concentrations (gaseous only), c_{day}/c_{night}, was 0.67 (0.54-0.85) for ACE, while c_{day}/c_{night} ranged wider, 0.25-3.35, for PHE, FLT and PYR, with mean values of c_{day}/c_{night} = 1.41-1.53.”

Lines 283-284: What could be the reasons for the lower Cday/Cnight ratio for chlorinated compounds specifically?
Thanks for pointing this out. The individual data indicate that there is no significant trend hidden. More in depth analysis for a sub-set of samples is presented later in the text (section 3.5). The new text will read: “For chlorinated substances, we find mean $c_{day}/c_{night} < 1$, namely 0.56-0.66 for HCH isomers, and 0.68-0.94 for PCBs and DDE, while the individual values range widely, 0.04-4.8.”

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?
Volatilization tends to occur during daytime driven by surface heating during the day and surface cooling during the night – which was not the case during the measurements reported here (SST_{day} - SST_{night} = 0.1-0.9 K, see Table S4). Other processes overcompensated for this small difference in SST, the most prominent being variation of mixing depth (low during the night, high during the day). This hypothesis is tested with the non-steady state 2-box model (presented in section 2.6).

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?
SST_{day} - SST_{night} = 0.1-0.9 K (see Table S4 and previous reply, above)

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.
True. The statement “Implicitly, dry deposition is difficult to budget.” will be added to the revised text. The same is/was expressed in the text (section 4, conclusions): “Both flux directions were observed (fluctuation) (…), not determined by the day-night cycle. Fluctuation of more substances might have been hidden by the method’s uncertainties. Hence, the mean flux direction on one hand side and observations during part of the time of the trace substances may differ. (…) In general, longer observations and across seasons of the flux is needed to assess the state of air-sea exchange of those anthropogenic trace substances, which have been approaching phase equilibrium historically (…) or seasonally (…).”
This text will be slightly extended in the revised text. “… In general, longer observations and across seasons of the flux is needed to assess dry deposition fluxes and the state of air-sea exchange …”

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

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?
According to the Monin-Obukhov similarity theory of turbulent transport in the surface layer (Monin and Obukhov, 1954; see section S1.3), vertical concentration profiles are expected to be logarithmic, while – due to the vertically non-linear behaviour of the eddy diffusivity – the resulting vertical flux is *per definitionem* constant. New text to be added (in section 2.2, after line 1149): "As long as the vertical concentration gradient is statistically significant, and the choice of the upper inlet height accounts for the homogeneity of the up-wind so-called fetch (see above), the resulting flux, calculated from the transfer velocity and the vertical concentration gradient (see eq. (1)), is the vertical turbulent net flux of the corresponding gas, representative for the fetch area and equal to the overall deposition flux (if $c(z_2) - c(z_1) > 0$)."

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?
Thanks. Will be clarified in the revised text: "based on measurement at $z_1 = 1.05$ m"

Line 422: New sentence is needed after "reason"
Will be done

Line 432: It is not clear which height the concentration corresponds to in this equation for determining the particle deposition flux.
Will be specified. Modified text will read: "$F_{p\ dep} = -v_{dep}\ c_p$ (equ. 4) with v_{dep} being determined by particle size and wind speed and c_p determined close to the sea surface."

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).
We agree; 3-4 ring PAHs are expected to re-distribute in the aerosol mass size distribution. However, as the difference between gaseous and particulate deposition fluxes was anyway huge (≈3 orders of magnitude), the estimate made still leads to the same qualitative statement. This discussion will be correspondingly extended in the revised version. (see above, reply to Reviewer #2)

Lines 470-473: Could the upward flux of particulate-phase POPs be related to sea-salt emissions from waves?
Thanks for pointing this out. The particle source sea spray will contain POPs, whenever these were accumulated in surface waters. Therefore, upward flux could be related to waves (sea spray). Explanation will be given in the new text: "Marine aerosol contains organic matter (OM), mostly in the accumulation mode, in particular over biological productive surface waters and under low wind speeds (Gantt et al., 2011; Albert et al., 2012). A considerable part of OM is water insoluble (O'Dowd et al., 2004; Facchini et al., 2008). Hence, the marine aerosol contains traces of POPs, which previously were in either dissolved form or associated with suspended OM. The upward flux of particulate-phase POPs could be caused by sea spray."

Albert, M.F.M.A., Schaap, M., Manders, A.M.M., Scannell, C., O'Dowd, C.D., and de Leeuw, G.: Uncertainties in the determination of global sub-micron marine organic matter emissions. Atmos. Environ., 57, 289-300, 2012.
Facchini, M.C., Rinaldi, M., Decesari, S., Carbone, C., Finessi, E., Mircea, M., Fuzzi, S., Ceburnis, D., Flanagan, R., Nilsson, E.D., de Leeuw, G., Martino, M., Woeltjen, J., and O'Dowd, C. D.: Primary submicron marine aerosol dominated by insoluble organic colloids and aggregates, Geophys. Res. Lett., 35, L17814, 2008.
Gantt, B., Meskhidze, N., Facchini, M.C., Rinaldi, M., Ceburnis, D., and O'Dowd, C.D., 2011. Wind speed dependent size-resolved parameterization for the organic mass fraction of sea spray aerosol. Atmospheric Chemistry and Physics, 11, 8777-8790, 2011.
O'Dowd, C.D., Facchini, M.C., Cavalli, F., Ceburnis, D., Mircea, M., Decesari, S., Fuzzi, S., Yoon, Y.J., and Putaud, J.P.: Biogenically driven organic contribution to marine aerosol. Nature, 431, 676-680, 2004.

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?
Will be clarified in the new text by indicating for both PAHs and chlorinated species (Fig. 1a and 1b) and by rephrasing: "Fluctuation of more substances addressed might have been hidden by gaps in the time series of fluxes (limited by the uncertainties of sampling and analysis) or the time resolution (limited by the sensitivity of the analytical method)."

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.
Findings of night-time maxima and accumulation were presented and discussed (sections 3.1 and 3.2, Fig. 1 and 2), also covered in the conclusions and abstract. But, yes, we agree, should be more emphasized. The revised text will read (in section 3.1, lines 288-289) "This results in night-time accumulation. Diel variation, apart from mixing, is related to …" and (in section 3.2, lines 299-302) "The modelling results support that (during advection of one air mass) the diel variation of contaminant concentrations in air and, particularly, the night-time accumulation was explained by the combination of volatilisation from the sea surface and atmospheric mixing depth." The visualized volatilization fluxes (Fig. S4) had not been referenced in the text. In the revised text this (lines 302-305) will read: "Volatilisation from the sea surface has significantly contributed to the night-time maxima of HCB (upward flux, Fig. 2a) and PCB28 (Fig. 2b), as well as of PYR during 1 night (Fig. S4a; Table S5). This, to our knowledge, had never been observed before." To this end, the corresponding flux plots (Fig. S4) will be moved to the main text (revised Fig. 2) and complemented by HCB (not shown in Fig. S4, but in revised Fig. 2). In the conclusions all substances for which observations (mentioned in lines 329-331) or modelling (lines 302-305) suggests that volatilization from the sea surface sustained night-time accumulation in the BL i.e., HCB, HCH, PCB28, PCB52 and PYR, will be mentioned (not just 1 of these as now, in line 481).

Supplement: There are two Table S6 in the supplement.
Thanks, will be corrected (Tables S6, S7).