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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of the marine atmospheric boundary layer on VSLS abundances in the eastern tropical and subtropical North Atlantic Ocean" *by* S. Fuhlbrügge et al.

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Response to Referee #2:

We would like to thank reviewer 2 for his helpful comments and valuable suggestions. Below you find our detailed answers to the specific points.

(1) I believe this paper can be shortened and more focused in general. For example, there are 8 pages of preliminary information before results directly relevant to the title of the paper are discussed (first sentence of Section 3.3), which seems excessive.

We tried to further shorten the sections as best as we can. Still we think there are some important aspects that have to be mentioned as background for our analyses. See also our detailed answers below.

(2) A little more information could be given in the abstract. Only the relationship of VSLS with the MABL is mentioned, whereas correlations with various different meteorological parameters have been calculated. It would be useful to add a sentence mentioning other parameters investigated, and then say that the strongest correlation seen was with the MABL. In addition, for the same reason it might be better to generalize the title.

Thanks for this suggestion, the abstract has been rewritten.

(3) The 'Meteorology and MABL height' section could be shortened as it contains detail that is not directly relevant to the conclusions of the paper.

Both sections have been shortened. For example unnecessary details and sentences as 'Higher average wind speeds were observed from June 6 to 14 2010 between 16 °N and 20 °N, nearly matching the mean wind speed of leg 3.' have been removed or 'Two different tropopause definitions are used to identify the transition between tropical and extra-tropical air masses. The cold point tropopause (CPT) is defined as the minimum temperature in the vertical temperature profile and is commonly used for the tropics (Highwood and Hoskins, 1998). For the tropopause definition in the extratropics the lapse rate tropopause (LRT) criteria is calculated, which is defined as the lowest level at which the lapse rate decreases to 2 K km-1 or less. In addition the lapse rate is requested not to exceed 2 K km-1 within 2 km above this level (WMO, 1957).' which was shortened to: 'Two different tropopause definitions are used to identify the transition between tropical and extra-tropical air masses: the cold point tropopause (CPT, Highwood and Hoskins, 1998) and the lapse rate tropopause (LRT, WMO, 1957).'

(4) Sections 3.1 to 3.2.1: Much of the detail described can be determined from looking at the Figures. This section should be shortened to just a summary of the main points.

We shortened these sections, for example: ‘Fig. 10 shows HySplit trajectories, started at the ground and at the top of the boundary layer. A band of trajectories were run backwards for 120 hours (5 days), starting each day of the cruise at 12 UTC.’ was completely removed.

(5) Sections 3.3 and 3.4: These sections present some useful data and important results, however, I feel that these sections could be written in a more focused way, highlighting the most significant results to the reader (more detail can be obtained from the tables and Figures).

Sections have been rewritten and shortened. For example ‘The vertical distribution of the relative humidity has been a good indicator for mixing in and thickness of the MABL (section 3.1.1), which may point to a co-correlation between the surface relative humidity and the VSLS abundances reflected by the high correlation for the whole cruise. However, over the upwelling areas this relationship does not hold anymore, because evaporation is also, besides the current amount of humidity in the air, depending on the air – and water temperature. Here, ΔT becomes positive, hence we derive a negative sensible heat flux, suppressing convection leading to a lower relative humidity, which is in contrast to the VSLS abundances. This would explain the reversed correlation above the upwelling. The VSLS abundances are significantly negative correlated with SAT and SST variations in the open ocean and positive correlated with SAT in the coastal upwelling. According to atmosphere – ocean interactions, including heat fluxes from or into the ocean we take the sensible heat flux, in our case reflected by the temperature difference ΔT (TSAT – TSST) into account. We calculate positive correlations of at least 0.7 for all trace gases during the whole cruise and at least 0.6 at the coastal stations. This means, that the combination of higher air – and lower water temperatures coincides with increased trace gas abundances and vice versa, although this is inappropriate for evaporation and shows that one cannot simply infer from the surface relative humidity on the VSLS abundances or even the MABL height.’

was rewritten to

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'The vertical distribution of the relative humidity has been a good indicator for mixing in and thickness of the MABL (section 3.1.1), which may point to a co-correlation between the surface relative humidity and the VSLS abundances reflected by the high correlation for the whole cruise. However, over the upwelling area this relationship does not hold anymore. The cold upwelling water creates a positive ΔT and a negative sensible heat flux, that suppresses convection and leads to a low relative humidity, which is in contrast to the VSLS abundances. This would explain the reversed correlation above the upwelling. The VSLS abundances are significantly negative correlated with SAT and SST variations in the open ocean and positive correlated with SAT in the coastal upwelling. The sensible heat flux, reflected by the temperature difference ΔT (TSAT – TSST) correlates with at least 0.7 for all trace gases during the whole cruise and at least 0.6 at the coastal stations. The combination of higher air – and lower water temperatures coincides with increased trace gas abundances and vice versa, although this is inappropriate for evaporation. This shows that one cannot simply infer from surface relative humidity on VSLS abundances or even the MABL height.'

In addition, section 3.4 was completely rewritten to:

The sea to air fluxes, which are calculated depending on wind speed and the concentration gradient Δc between sea water and air (Section 2.1), show significant positive correlations to wind speed and negative correlations to MABL height (Table 4). The inverse relationship of atmospheric VSLS to MABL height as described in section 3.3 should lead to lower sea to air fluxes, F, as lower MABL heights lead to higher atmospheric mixing ratios, decreasing the concentration gradient, Δc (Eq. 2). However, higher air- sea fluxes are observed in the lower MABL height areas for dibromomethane and bromoform with an according positive relationship of F to ΔT (Table 4). This is due to elevated sea water production of brominated VSLS in the cold waters, leading to a large increase of the concentration gradient, which masks the flux suppression by the higher atmospheric mixing ratios (Hepach et al., 2013). Also the elevated atmospheric mixing ratios of methyl iodide have no effect on the fluxes, because methyl iodide is

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strongly supersaturated in the sea surface water throughout the entire cruise (Hepach et al., 2013). On the other hand, the observed air- sea fluxes reveal correlations to the atmospheric VSLS abundances (Table 5), showing that MABL height and air–sea fluxes in combination add to the VSLS variations in the atmosphere. The detailed analysis of the air-sea fluxes, their driving factors, such as the sea water concentrations, and their influences on the atmospheric VSLS abundances are discussed in detail in Hepach et al. (2013).

(6) I agree with Reviewer 1, that it would be interesting to compare the correlation of VSLS fluxes with VSLS abundances, to the correlation of the MABL with VSLS abundances. Which is more important?

Both have an influence, as they both add to the VSLS abundance in the atmosphere. (VSLS in boundary layer $\sim$ Flux/MABL). Low MABL heights and large sea-to-air fluxes together contribute to an increase of boundary layer VSLS. We now include an overall correlation table for the sea-to-air fluxes and the atmospheric concentrations, which reveal an influence of the sea-to-air fluxes on the atmospheric concentrations. The detailed analysis of the sea-to-air fluxes, their driving factors as the sea concentrations and their influences on the atmosphere goes beyond the scope of this paper. There are certain combinations and thresholds of MABL heights and fluxes and also there are different driving factors for the sea-to-air fluxes which are discussed in detail in the paper "Diel and regional drivers of air-sea fluxes by the study of Hepach et al., which will be submitted in the upcoming weeks. See also our detailed answer to reviewer 1.

Technical Corrections P31207, line 5: I believe VSLS is already plural, change 'VSLSs' to 'VSLS,'.

Has been changed.

P31207, lines 7-8: change 'tropospheric oxidation processes can be altered' to 'they can alter tropospheric oxidation processes'

Has been changed.

P31207, line 9: change ‘lifetime’ to ‘lifetimes’

Has been changed.

P31208, line 13: change ‘die’ to ‘the’

Has been changed.

Section 2: the subheadings lack numbering.

Numbers added.

P31209, line 6: change to ‘hourly VSLS measurements were performed at six 24 h stations during leg P339/2’

Has been changed.

P31209, lines 9-10: change to ‘Mauritanian coast as well as the nutrient poor’

Has been changed.

P31209, line 13: change to ‘an additional 20 atmospheric samples’

Has been changed.

P31209, line 25: change ‘4:00’ to ‘4’.

Has been changed.

P31223, line 10: change to ‘Hourly VSLS measurements were conducted at six 24 h stations in these areas’

Sentence has been rewritten.

Table 3: Last line of caption – there are no bold coefficients in my copy.

Has been corrected.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 31205, 2012.

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