

## Responses to reviewer #1 comments and changes made according to the suggestions

### General comments

Kuss and collaborators present high-resolution measurements of Hg<sup>0</sup> in seawater and air-sea fluxes in the Baltic Sea. High-resolution measurements make an important scientific contribution to the field. Ocean emission are large global source of Hg to the atmosphere and, as pointed out by the authors, there is considerable uncertainty in air-sea fluxes and so I'm very glad to see the authors working in this area.

I recommend the manuscript of publication after revisions. The manuscript is clearly written and logically organized. The greatest area for improvement is Section 3. Section 3 currently reads as a dense report-out on results and is a bit too light on the discussion. It would greatly improve the manuscript to add more insight and context to Section (i.e., tell the reader why the results matter, how the results change or add to existing knowledge, and the implications).

- In Section 3 we present a follow up from the "surface water and atmospheric concentrations of Hg<sup>0</sup>" to the resulting flux driving gradients "Variability of the Hg<sup>0</sup> sea-air concentration difference", considering local peculiarities by discussing "the contribution of coastal upwelling". Then we provide an estimate of the emitted amounts of mercury by calculating "the average seasonal mercury emissions of the Baltic Sea" by using climatological wind speed data. Actual fluxes during our campaigns are subsequently compared to the observations in other marginal sea areas ("Hg<sup>0</sup> emission fluxes of the Baltic Sea and other marginal seas"). Thereby we emphasize the different methods in measurement and calculation. Finally, we provide an "emission budget of the Baltic Sea". We discuss the impact of different k-parameterizations as well as of other controlling parameters and quantify the emission according to current knowledge. It is clear to report things that are supported by data and it was not the aim to go too far beyond the determined Hg<sup>0</sup> emission fluxes. However, we explained some observations in more detail in the revised manuscript: Upwelling (Page 9, Lines 23-28), atmospheric contribution (Page 10, Lines 16-18).

### Specific comments

Page 1, line 16: The use of "major" is ambiguous. Major in what context? A major source in the Baltic region? In the global context, it's small (<1% of global ocean emissions). Consider deleting "major", replacing with a quantitative statement, or clarifying the context in which it's a major source.

- It was aimed as a general remark, however it appeared misleading. We modified the sentence to distinguish between local findings and the global meaning (Page 1, Line 16).

Page 1, lines 18-23: "A membrane equilibrators enabled continuous... Hg<sup>0</sup> wat could also be characterized in deeper water layers." This level of details seems more appropriate the Methods section than the Abstract.

- It is aimed as a brief summary of the applied methods. Since the methods reflect an important part in the paper, it appears adequate to give this summary in the abstract. No changes were made (Page 1, Lines 18-23).

Page 8, lines 10-17: This paragraph is especially dense with numbers. Consider summarizing in a table instead of the main text.

- We summarized the data of fitted and averaged atmospheric  $\text{Hg}^0$  measurements in a table, now Table 1. A brief introduction to Table 1 is given (Page 8, Lines 10-15).

Page 9, lines 25-27: "Upwelled water affects areas.... We conclude that upwelling contributes significantly to  $\text{Hg}^0$  emissions." This seems like an important result and merits further elaboration. Why does this matter? How does it change or add to the current understanding of what's going on in the Baltic or other marginal seas?

- The coastal upwelling itself is a challenging subject. Its spatial and temporal variability make it difficult - even by using sophisticated modelling - to quantify the contribution, so we decided to further explain the phenomenon and to include a reference that shows episodic upwelling areas in the Baltic Sea (Lehmann and Myrberg, 2008) (Page 9, Lines 23-28).

Page 11, lines 21-22: A 60% difference is substantial. If Nightingale 2000 and Weiss 2007 yield such different results, what's the implication for current global budgets of ocean emissions?

- It is really a serious problem for sea-air flux calculations. However, as discussed in the paper, the Nightingale approach appeared a suitable compromise. The also often used parameterization of Wanninkhof (Wanninkhof, 1992) was later re-calculated by using an extended data base (Sweeney et al., 2007). It revealed a "new Wanninkhof" which was clearly close to the Nightingale parameterization. Hence, there is some confidence that the relationship between  $k$  and  $u$  is constrained somewhere around Nightingale's parameterization. Thus, the uncertainty is expected to be reduced for most environmental conditions by using it. No changes were made (Page 11, Lines 20-22).

Section 3.6: What the relationship between the emission budget presented for the Baltic Sea and the trends stated in the introduction (decline since 1990s, relatively flat since 2006)?

- Based on Figure 4 by looking at the summer month data, a slight declining trend might be deduced in the sea-air  $\text{Hg}^0$  concentration difference. However, the variability is large and the Baltic Sea hydrography is complicated by frequent upwelling and inflow events in time intervals of several years. Thus considering spatial variability and almost decadal changes a trend is not trustworthy unless the consequences of the different processes on  $\text{Hg}^0$  emission are basically understood – which seems currently not the case. Therefore, we didn't feel ready to make conclusion on this point. (No changes were made).

Data availability: I strongly encourage the authors to make the un-averaged data available, in addition to the averaged data. Un-averaged data will be of greatest interest to modelers want to compare simulated and measured values.

- The data are now available on request from the IOW data base (details are given on Page 12, Lines 4-5).

Figure 3: It's really hard to distinguish the symbols for  $\text{Hg}^0_{\text{wat}}(1)$  and  $\text{Hg}^0_{\text{wat}}(2)$ . I'd suggest using two colors with greater contrast.

- We modified the symbols of  $\text{Hg}^0_{\text{wat}}(1)$  to a lighter grey to better distinguish both data sets.

### References used for the answers

Lehmann, A., and Myrberg, K.: Upwelling in the Baltic Sea — A review, *J. Mar. Syst.*, 74, S3-S12, 10.1016/j.jmarsys.2008.02.010, 2008.

Sweeney, C., Gloor, E., Jacobson, A. R., Key, R. M., McKinley, G., Sarmiento, J. L., and Wanninkhof, R.: Constraining global air-sea gas exchange for CO<sub>2</sub> with recent bomb <sup>14</sup>C measurements, *Global Biogeochem. Cycles*, 21, GB2015, doi:10.1029/2006GB002784, 2007.

Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean, *J. Geophys. Res.*, 97, 7373-7382, 10.1029/92JC00188, 1992.