

Responses to reviewer #2 comments and changes made according to the suggestions

Air-sea exchange is one of the major uncertainties in understanding the global mercury cycle. The presented study improves on previous work by Kuss et al. in the Baltic Sea. Based on high resolution measurements it gives novel insights into important processes and short term variability of air-sea exchange. This is a valuable contribution to research on mercury. Moreover, the paper is well written, the methodology robust and the measurements trustworthy.

I support publication of the manuscript after a few issues are addressed:

Major points:

Page 7 line 1-2: Please discuss the error introduced by the usage of average wind speeds as compared to high resolution (e.g. hourly) data. As wind speed is squared ($0.222 u^2 + 0.333u$) even using the median instead of the mean might have a large impact on the calculated fluxes. I think you need at least estimate the error due to the averaging. Especially in early autumn when Hg^0 concentrations are still high and storms are more common.

- As we pointed out in the respective sentence (now it is Page 6 Line 36 to Page 7 Line 1&2), we used mean wind speeds and mean square wind speeds. This reliably avoids the averaging bias. The mean square wind speed accounts for the original wind speed variance. Thus, it appears a feasible method to use climatological data sets without the averaging bias, if mean square wind speeds are available (no changes were made).

Page 11 line 26-31: This section needs to be clarified: The 1.73 Mg Hg^0 annual evasion is supposed to be the estimate for the whole Baltic Sea? So the Bothnian Sea, Bay of Bothnia, Bay of Finland, Bay of Riga have a combined Hg^0 flux of only 730 kg? How do you extrapolate the data to get to this conclusion? This leads to many unanswered questions and I would ask you to give more information on the extrapolation method and its uncertainty (e.g. Do you consider the effects of sea ice? Do you have measurement data for the Bay of Riga and Bay of Finland? What is the effect of average wind speeds?)

- We deduced 1.73 Mg for the whole Baltic Sea based on the areas given in Table 4 (now Table 5). The whole Baltic Sea represents an area of 412 560 km², the study area is 235 000 km² representing ~57% of the whole Baltic Sea. The extrapolation was based on the area ratio and was argued as likely realistic because of the sporadic measurements that were done in the Bothnian Sea and the Bay of Finland. Winter was not considered as an important season of emission, hence ice coverage was not explicitly mentioned. Potential accumulation of Hg^0 below ice would likely be released after cracking and melting of the ice coverage in spring. We don't think that this would change the estimate significantly. We added the term "according to the area ratio" (Page 11, Lines 27-28)

Minor points:

Page 1 line: I suggest that you also cite the HELCOM reports (2007 & 2011) on which the Soerenson et al. riverine influx estimate is based on.

- Yes, we included the "5th Baltic Sea pollution load compilation" as the important reference behind (Page 2, Line 1).

Page 2 line 24: “The aims are” instead of “The aim were” This clarifies that you are talking about the actual study and not a previous one.

- We made the change accordingly (Page 2, Line 24).

Page 5 line 13: Please clarify: “of $\pm 3\%$ only in a Hg₀ concentration range of 14–38 ng m⁻³” To me that means that the 3% error was only validated for concentrations in the range of 14-38ng/m³. If this is the case the question arises how large the error is outside this range. Otherwise I suggest to drop the word “only” which makes the sentence clearer.

- That’s right. “only” was introduced to emphasize that the deviation is small. However, it is obviously misleading and we deleted the word “only” (Page 5, Line 12-14).

Page 8: lines 5-6: This finding is based on average wind speeds. How well does this capture storm events?

- We couldn’t figure out to which number this is referred to, however, as commented on the first major point, we used mean and mean square winds that omits an averaging bias, i.e., it accounts for the distribution between very low and high wind speeds (No changes were made).

Page 9, lines 25-27: This is a great result. It would be interesting if you could estimate the impact of an upwelling event on the mercury flux that would normally occur without this event. This could also be a source for inter-annual variability due to shifts in wind fields.

- Upwelling is a complex spatial and temporal process {Lehmann, 2008 #2840}. We give some more information about upwelling in the revised version (Page 9, Lines 23-28). Unfortunately, based on our data set a detailed quantification of the impact on emission is not possible, it would have required a complete spatial and temporal coverage of the upwelling area and the adjacent area by measurements, which was not possible during our campaign.
Yes indeed, shifts in the wind fields certainly contribute to the inter-annual variability of the Baltic Sea Hg₀ emission. However, other meteorological parameters are important as well (solar radiation, cloudiness ...).

Page 11 lines 21-22: You identify a 60% difference in calculated air-sea flux due to differences in parametrizations. How important do you think the inter-annual variability is in comparison. And how large is the effect of averaging wind speeds in comparison?

- The inter-annual variability is basically reflected in Figure 7, where seasonal averages have been calculated. It appears to be about 30-50% in spring and summer. We think that the parameterization of Nightingale et al. is a good choice, probably less than 25% uncertainty. As commented on the first major point, we used mean and mean square winds that omits an averaging bias, i.e., it accounts for the distribution between very low and high wind speeds (No changes were made).

Page 11 line 26-31: I suggest that you compare the results of your extrapolation with modelling results which can be seen as a more sophisticated way of extrapolating measurement data (e.g. Soerenson et al., 2016; Bieser and Schrum et al., 2016).

- Yes indeed modelling appears the only way to account for the variable influences on the Hg⁰ gas exchange with a full area coverage. However, we are a bit cautious on this suggestion, as on our point of view modelling did not and could not account for all drivers operating on mercury emissions by the sea in a proper way. Current state of the art requires more measurements and process studies that would enable an improved modelling in the future (No changes were made).

Page 11 line 37: fluxes instead of flux. I agree with reviewer #1: "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 who want to compare simulated and measured values."

- We exchanged flux by fluxes (Page 11, Line 37).
- The data are now available on request from the IOW data base (details are given on Page 12, Lines 4-5).