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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Distribution and air-sea exchange of mercury (Hg) in the Yellow Sea" *by* Z. J. Ci et al.

Z. J. Ci et al.

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We appreciate the detailed and helpful comments from the reviewer. These comments and suggestions have helped us improve the manuscript. Please find our point-by-point responses to the reviewer's comments below in bold.

General comments:

The paper "Distribution and air-sea exchange of mercury (Hg) in the Yellow Sea" presents data on atmospheric Hg0, and aquatic total Hg, reactive Hg and DGM from the atmospheric outflow region of Asia. In the paper calculation and analysis of air-

sea fluxes is also made. The paper is mostly well written and the data is an import contribution for the understanding of the spatial distribution of atmospheric and aquatic mercury.

Response: We appreciate the reviewer's recognition of the merits of this work.

I am wondering if there are any ancillary atmospheric data on CO, O3 or other species? This could greatly strengthen some of the interpretations on the influence from anthropogenic sources.

Response: Unfortunately, there were not other trace gaseous measurements available in this cruise.

When comparing fluxes it is important to remember that others papers might have used different methods for the flux calculation. Including a table in the supporting information with ancillary data including e.g. wind speeds and temperatures for the flux calculations as well as some words on this would help the reader understand the importance in the different air-sea flux estimates better.

Response: We totally agree with the reviewer that the calculation of Hg(0) flux is significantly influenced by the choice of methods. As we discussed in the present manuscript [P. 1527, L. 13 - P. 1528, L. 12], for the given data (DGM, GEM, Water temperature, Wind speed), the calculation of Hg(0) flux will principally depend on the choice of the diffusion coefficient of Hg(0) [*D*] and gas transfer parameterization of wind-induced mass transfer coefficient [*K*]. In the revised manuscript, we have added a new table (Table 1), which includes all relevant data required to calculate the Hg(0) flux at the air-sea interface (e.g., DGM, GEM, Water temperature and Wind speed). This data will help reader evaluate the magnitude of relevant factors on the influence of the Hg(0) flux calculation. [In the revised manuscript, we have recalculated the Hg(0) flux using the diffusion coefficient

of Hg(0) developed by Kuss et al. (2009) as suggested by Dr. Joachim Kuss in last comment (www.atmos-chem-phys-discuss.net/11/C15/2011/). For more information about the Hg(0) flux recalculation, see the response to P. 1519, L. 7/8 of Specific comment by Dr. Joachim Kuss].

Is there any possibility to make some statistic comparison between this cruise and some of the other cruises presented in Figure 2?

Response: We have seriously considered the insightful suggestion by the reviewer. However, at present it is very difficult to obtain the enough detailed data reported by other researchers to make some statistic comparison.

I would recommend removing words like "clearly" and "significantly" from the text when referring to comparisons of different results. "Clearly" is a very subjective word when considering the large standard deviations on most results. "Significantly" should be followed by an indication of the significance level for the statement (e.g. ($P<0.05$)) otherwise I would suggest only stating that averages are higher or lower than each. When no standard deviations are given on some of the averages you compare to you should be even more careful with the choice of words. If you have made a statistical analysis please state this clearly and give the test and the significance level of the result.

Response: We have followed the reviewer's suggestion to remove most of "clearly" and "significantly". Only several "significant" or "significantly" are kept in the revised manuscript as the significance level is reported.

Specific comments:

1512 line 21: Can the three fractions of mercury measured in the waters be referred to as species? Total Hg is certainly not.

Response: We have changed "*all three species of Hg*" to "*DGM, RHg and THg*". Correspondingly, the titles of Section 2.4 [*Hg species (DGM, THg and RHg) measurements in the water*] and Section 3.4 [*Vertical distribution of Hg species in the water column*] also have been changed to [*DGM, THg and RHg measurements in the water*] and [*Vertical distribution of DGM, RHg and THg in the water column*] in the revised manuscript, respectively.

1514 line 7-9: I am a little bit unsure about what you want to say here but I think (as a number for the contribution of evasion compared to deposition) the Strode et al. paper only report 60% re-evasion. In any case you might want to reference the updated version of the GEOS-Chem slab ocean model by Soerensen et al. (2010, "An Improved Global Model for Air-sea exchange of Mercury: High concentrations over the North Atlantic."). They found re-evasion to account for 80% of depositions.

Response: We have followed the reviewer's suggestion to reference the result of Soerensen et al. (2010). Correspondingly, the ratio of evasion/deposition has been changed to 80%.

[Reference:]

Soerensen, A.L., Sunderland, E.M., Holmes, C.D., Jacob, D.J., Yantosca, R.M., Skov, H., Christensen, J.H., Strode, S.A. and Mason, R.P.: An improved global model for air-sea exchange of mercury: high concentrations over the North Atlantic, *Environ. Sci. Technol.*, 44, 8574-8580, 2010.

1517 line 4: "Due to the low depth of the"-I am not sure why the low depth of the Yellow Sea is a cause for selecting three station and make vertical profiles? Can these not be

made if the water is deeper? Maybe change the formulation?

Response: We have followed the reviewer's suggestion to rephrase this sentence. This sentence "*Due to the low depth of the Yellow Sea, three stations (water depth of 50-80 m) were selected for studying the vertical distribution of Hg.*" has been changed to "*Three stations were selected in the deep region of the Yellow Sea (water depth of 50-80 m) for studying the vertical distribution of Hg.*"

1520 line 4: Is the mean and the median concentrations the same?

Response: We have checked the data again and confirm that the mean and the median number is same.

1521 line 4 and 23: Maybe a reference to Lindberg et al (2007) or Sprovieri et al (2010,"Worldwide atmospheric mercury measurements: a review and synthesis of spatial and temporal trends") would be more general and updated references for background concentrations in the northern hemisphere?

Response: We have followed the reviewer's suggestion to reference the available ACP paper of Sprovieri et al. (2010).

[Reference:]

Sprovieri, F., Pirrone, N., Ebinghaus, R., Kock, H., and Dommergue, A.: A review of worldwide atmospheric mercury measurements, *Atmos. Chem. Phys.*, 10, 8245-8265, doi:10.5194/acp-10-8245-2010, 2010.

1521 line 13-14: Please state what you compare to when you say that concentrations are elevated near the coast and decreased in the open ocean. Maybe say they are elevated at the coast compared to the open ocean?

Response: We have followed the reviewer's suggestion to rephrase this sentence. The sentence "*As shown in Fig. 3, the spatial distribution of GEM was generally characterized by elevated levels near the coast of China and decreased levels in the open ocean.*" has been changed to read "*As shown in Fig. 3, the GEM concentrations were generally elevated at the coast of China compared to the open ocean.*"

Page 1523 line 4-23: Why compare to the cruise mean when many of the other samples are also made close to land and might be affected by smaller rivers etc. Why not compare the CJ-2 to CJ-7 stations with samples taken a certain distance from land? Use for an example a t-test to see if those samples are statistically significant different from the ones near the river mouth? This would give a much better idea of the possible influence from the river. You could also test if there is a trend from the CJ-2 station closest to the coast and as you get further away.

Response: We thank the reviewer for this suggestion. We have followed the reviewer's suggestion to compare the THg concentration of water samples taken within 100 km from land and other stations by the *t*-test method. The *t*-test method also is applied to compare the THg concentrations of water samples taken from the Stations CJ-2 to CJ-7 and other stations.

Page 1523 line 26: This sentence is strange. RHg is not large at the coast because concentrations in the open ocean of THg are small. Reformulate.

**Response: We have followed the reviewer's suggestion to rephrase this sentence. The sentence "*Because of the low THg levels in open ocean, the RHg concentrations in the Yellow Sea were generally higher than many open oceans ..... Cossa and Gobeil, 2000).*" has been rephrased to read "*The RHg concentrations in the Yellow Sea were generally higher than many other open oceans (e.g.,*

Atlantic Ocean: 0.34 ng l^{-1}, Mason and Sullivan, 1999; Mediterranean Sea: 0.09 ng l^{-1}, Horvat et al., 2003) and were higher or comparable to those of near-shore environments (e.g., Long Island Sound: 0.26-0.90 ng l^{-1}, Rolfhus and Fitzgerald, 2001; Lower St. Lawrence Estuary: $<$ 0.04-0.22 ng l^{-1}, Cossa and Gobeil, 2000).**".**

Page 1524 line 11: Does the RHg/THg fraction give any indication of the dynamic cycling? If the part of the THg fraction that is not RHg is thought to be mostly inactive then it is only the actual RHg level that tells us anything about how dynamic the system is?

Response: In this paragraph (P. 1523, L. 24 - P. 1524 L. 12 in the present manuscript), we intend to express the opinion that the high RHg/THg ratio in the Yellow Sea may correspond to a short turnover time of oceanic Hg and further indicate more dynamic cycling of Hg in this marine system. To improve the clarity of our opinion, we have changed "***....., the high RHg levels and RHg/THg ratios indicated that the Hg cycling in the Yellow Sea may be more dynamic than other marine systems.*****" to read "*****....., the high RHg levels and RHg/THg ratios suggest that the turnover time of Hg in the Yellow Sea may be shorter than other marine systems and potentially indicate the more dynamic cycling of Hg in this marine system.*****"**

Page 1525 line 13-29: Maybe elaborate a little on the fact that, if there is an extensive amount of algae in the water, light will not penetrate very far into the water column. In this case I would expect that photoreduction (and oxidation) would be of less importance in the water column even if the insolation is high.

Response: We totally agree with the reviewer that the algae bloom can decrease the light penetration and subsequently reduce the importance of photochemistry on the Hg redox reaction in the water column. As suggested by many

studies (see Introduction in the present manuscript), the solar radiation is generally considered as the primary factor on the redox transformations of Hg in aquatic environment. But other factors, such as microbe activity and sea water chemistry, also can not be ignored. However, based on our existing data, it is difficult to evaluate which factor is more important on the Hg redox reaction in the Yellow Sea. As we have addressed in the present manuscript, more integrated research strategies are needed in the future.

Page 1527 line 27-29: Should this reference be to the Soerensen et al. 2010 paper (An Improved Global Model for Air-sea exchange of Mercury: High concentrations over the North Atlantic.)? They treat the D suggested by Kuss et al (2009) in the Supporting Information of their paper. I don't recall it being treated in Strode et al (2007) paper.

Response: Yes, just as pointed out by the reviewer, in the modeling study of Strode et al. (2007), the authors estimated the global oceanic Hg(0) fluxes using the Hg(0) diffusion coefficient [*D*] suggested by Poissont et al. (2000). In the last part of this paragraph (P. 1527 L. 25 - P. 1528 L. 2 in the present manuscript), we attempt to discuss the choice of *D* on the impact of the Hg(0) flux calculation. In the study of Kuss et al. (2009), the authors used the modeling work of Strode et al. (2007) as example to show that using different *D* (one is suggested by Kuss et al. (2009) and another is suggested by Poissont et al. (2000)) will obtain the different strength of global oceanic Hg(0) flux. Therefore, we choose the study of Kuss et al. (2009) to support our discussion. In the revised manuscript, we have modified the text to further clarify it.
[Reference:]

Strode, S.A., Jaeglé, L., Selin, N.E., Jacob, D.J., Park, R.J., Yantosca, R.M., Mason, R.P. and Slemr, F.: Air-sea exchange in the global mercury cycle, *Global Biogeochem. Cycles*, 21, 21, GB1017, doi:10.1029/2006GB002766, 2007.

Poissant, L., Amyot, M., Pilote, M. and Lean, D.: Mercury water-air exchange over the Upper St. Lawrence River and Lake Ontario, *Environ. Sci. Technol.*, 34, 3069-3078, 2000.

Kuss, J., Holzmann, J. and Ludwig, R.: An elemental mercury diffusion coefficient for natural waters determined by molecular dynamics simulation, *Environ. Sci. Technol.*, 43, 3183-3186, 2009.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C595/2011/acpd-11-C595-2011-supplement.pdf

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