

Autor response to Reviewer comments 4, the reviewers comments are in normal black font and the autor response are in blue bold font.

This study combines Hg fluxes and Hg isotopes in atmospheric Hg(0) and Hg(0) from snow air and soil air to investigate the fate of Hg(0) in Arctic tundra terrestrial environment. This study is part of a larger, systematic study, which I think is well planned and well carried out. The data on Hg isotopes in soil air and snow air is very novel, and they indeed provide new insight in Hg cycle in Arctic and support the conclusions from many previous studies about terrestrial-air Hg cycle that were based on experiments and field observations. The paper is also well written and the data is clearly and properly analyzed and interpreted. Overall, I recommend the publication of this paper with minor revisions: **We thank reviewer 4 for this constructive and positive assessment.**

General comment:

The observation of opposite Hg isotope signals between snow air and soil air is indeed interesting. I think it would add some value to this paper if the authors can give a more thorough thoughts on this, especially regarding the mechanism of Hg isotope signals in snow air. The current interpretation relying on lichen uptake does not seem to be very convincing. The source and process of Hg in lichen and snow air could be very different. Are there any redox processes within snow that could produce the isotope pattern in snow air and what is the possible mechanism?

In the revised manuscript we will provide a more nuanced discussion on Hg stable isotope systematics in soil and snow air.

Specific comment:

1. P6, L1: it is a little confusing for the word “complementary”. Does lichen and snow air represent two complementary pools of Hg? Lichen represents a long term accumulation of atmospheric Hg(0) throughout the year, whereas snow air is a more temporary pool of Hg. The source and process of Hg in lichen and snow air could be very different.

We agree and have revised the respective paragraph in order to provide a more accurate explanation

2. P6, L26-31: This section is about AMDE season, but why suddenly you switch to the discussion about May, which is after the three AMDE events? What about the isotope signals in snow air during AMDE? Should this be mentioned?

We agree that this was confusing and renamed the chapter to “spring”, which includes the AMDE events and snowmelt. During AMDE’s we saw a shortterm increase in the Hg(0) concentration in snow air. However, we were not able to sample specifically for Hg(0) snow air during AMDE’s for Hg stable isotope analysis. The time periods were too short and with a very low sampling rate in the snow air they would not be isotopically resolvable.

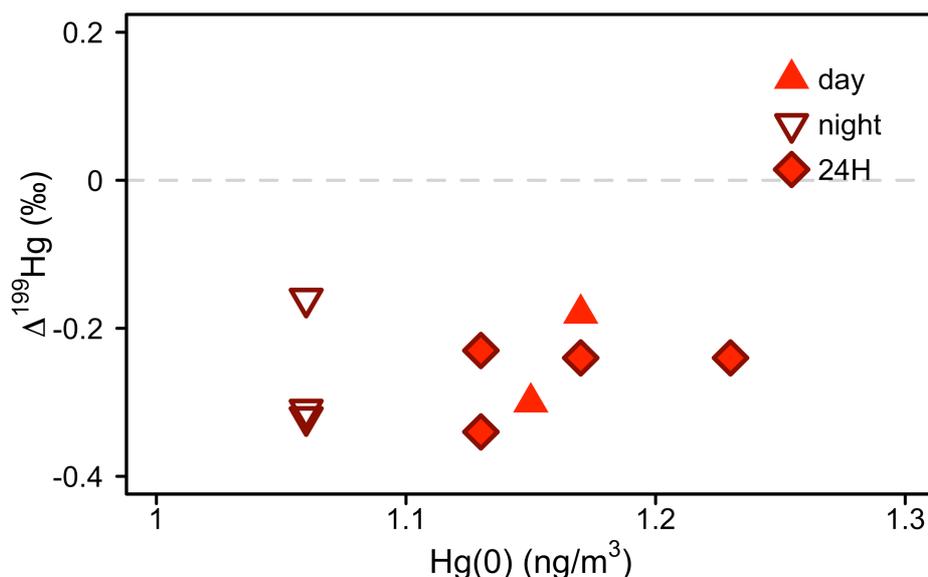
3. P6, L35: Figure 6 seems to be a wrong figure, it is not about even MIF, do you mean figure 4? **Yes, thanks for spotting this, error was corrected**

4. Figure 4: the x axis should be explained. **The x axis is explained as: Mass dependent fractionation ($\delta^{202}\text{Hg}$), which is defined in equation 1. In our view no further explanations are needed, no changes were made**

5. P8, L1: “do not only reflect”, delete “do” **changed as suggested**

6. P8, L27: Could you show the change of MIF with Hg(0) concentration?

In the manuscript we write that nighttime and daytime $\Delta^{199}\text{Hg}$ values are similar, as a consequence of which there is no significant variation of $\Delta^{199}\text{Hg}$ with Hg(0) concentration. For the courtesy of the reviewer we show a corresponding figure below.



7. P8, L8-9: I agree, but this does not explain your high enrichment factor, which does not distinguish between pure foliar uptake and the net effect of uptake and re-emission. Re-emission would indeed affect the d202Hg and you certainly need to discuss this scenario, but re-emission likely occurs in all situations and would not cause the difference between yours and other studies. Furthermore, re-emission is accompanied by MIF, but your data shows no change of MIF between day and night.

We disagree with the suggestion that Hg re-emission occurs at all time. There are several papers showing that net foliar Hg re-emission only occurs during daytime and during nighttime a net uptake was observed (e.g. Fu, X.; Zhu, W.; Zhang, H.; Sommar, J.; Yu, B.; Yang, X.; Wang, X.; Lin, C. J.; Feng, X., Depletion of atmospheric gaseous elemental mercury by plant uptake at Mt. Changbai, Northeast China. *Atmos. Chem. Phys.* 2016, 16, (20), 12861-12873. or Yuan, W.; Sommar, J.; Lin, C.-J.; Wang, X.; Li, K.; Liu, Y.; Zhang, H.; Lu, Z.; Wu, C.; Feng, X., Stable isotope evidence shows re-emission of elemental mercury vapor occurring after reductive loss from foliage. *Environmental Science & Technology* 2018.). Furthermore, the odd-mass Hg isotope signatures provide a strong indication for photochemical processes, suggestion that photoreduction is the dominant process causing foliar Hg re-emission. Therefore, we are confident in our assumption that Hg(0) re-emission predominantly occurs during daytime. As explained in the manuscript daytime deposition/re-emission processes are not expected to be traceable in atmospheric Hg concentration of stable isotope signature due to strong mixing with tropospheric air. No changes were made to the manuscript.

8. P8, L11-15: I agree with this interpretation and I believe this is a more likely interpretation than the re-emission scenario. The d202Hg in atmospheric Hg(0) is not only affected by foliar uptake. Mixing with other Hg sources should be considered in the first place. The Rayleigh model

shown in Figure 6S is based on the assumption that the change of $\delta^{202}\text{Hg}$ was completely caused by processes, which should be clarified.

We agree that the calculation of a fractionation factor for foliar $\text{Hg}(0)$ uptake from the diurnal variation observed is only possible based on the assumption that foliar uptake is the dominant factor for the variation and that there are no other major processes or sources. We will clarify this assumption in the revised manuscript. We would like to re-emphasize that the study was conducted in the arctic tundra, hundreds of km away from the next anthropogenic Hg source. As discussed in the manuscript, the diurnal patterns in Hg concentration in relation to CO_2 concentration and boundary layer stability provide strong evidence that the variation observed is indeed from vegetation uptake of $\text{Hg}(0)$ and not from different sources.

9. P8, L32: How did you estimate the <5% of total Hg deposition flux? Can you elaborate a little? The concentration of $\text{Hg}(0)$ in soil air is almost lowered by half compared to atmospheric $\text{Hg}(0)$. This seems to be a significant sink. **We compared the soil uptake flux estimated by Obrist et al. 2014 with the net ecosystem exchange flux measured at Toolik field station. We clarified this in the revised manuscript.**

10. P8, L34-35: I agree that the difference between soil air and atmosphere is caused by uptake of $\text{Hg}(0)$ by soil because the isotope signals are very consistent with the experimental work. However, the opposite Hg isotope signals between soil air and snow air do not directly support that argument that soil $\text{Hg}(0)$ sink is minor, because the isotope signals of $\text{Hg}(0)$ in snow air is likely controlled by other mechanisms, which I believe is not clearly identified. **See discussion to comment 1 above.**