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Interactive comment on "Gaseous elemental mercury (GEM) fluxes over canopy of two typical subtropical forests in south China" *by* Qian Yu et al.

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Responses to comments of Anonymous Referee 1

Thanks to the reviewer for giving us very useful comments to improve the manuscript. Detailed responses to the comments are list bellow (underlined).

The manuscript reports flux data measured by the aerodynamic gradient (AGM) method over the canopy at two low-land forest sites in South China over a one-year period. Although the context of this study does not introduce new science beyond what has been known, such long-term flux datasets in forest ecosystem are rare and deserve

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Discussion paper

consideration for publication. The manuscript is organized, read well and carefully drafted given the data that it presents. The data reporting sections (Section 3.1 – Section 3.4) are appropriately supported by data and the depth of conclusions/implication can benefit from incorporating the recent findings obtained from mass balance study and stable mercury isotope investigation in forest ecosystems. Overall, I recommend publication of the manuscript and have the following minor comments.

1. The LAI and other descriptive characterization (e.g., other predominant vegetative species, forest floor coverage, radiation transfer under canopy) of the two forest sites should be provided for better assessment of the forest site.

The LAI and other descriptive characterization (other predominant vegetative species, Canopy density, radiation transfer under canopy) of the two sites were added in table 1. And the characterization was used as an evidence for the source/sink discussion of Section 3.4

2. Appropriately determining the turbulent transfer coefficient (K) is critical for estimating AGM flux, yet it is not clear in the manuscript how K varies. It will be useful to report the estimated K values and its diurnal variation in different in different season for evaluation of the reasonableness of estimated K values.

We agree that the turbulent transfer coefficient (K) is critical for estimating GEM flux, and the diurnal variation in different season was provided in SI.

3. The quality control statement of the AGM flux measurement is limited only to the detection of Hg vapor but did not consider other sources of bias (the long tubing, flow synchronization, intermittent sampling at the two level, etc.) that might introduce uncertainty to the flux measurement. Would it be a possibility that these variables can also be assessed to better represent the measurement uncertainty?

The blank experiments to measure the monitoring system error were conducted before the installation by placing the air intakes in the zero mercury gas (Zero Air Tank, Tekran

Inc.) for 48h. There were almost no adsorption/emission from the monitoring system (including of the long Teflon tube, the soda-lime tank, and the electromagnetic valves) with the measurement results less than the detection limit of the instrument (0.1 ng m-3). The manual calibration by placing the air intakes in certain Hg concentration (Tekran 2505, Tekran Inc.) for 24h were done once every one month. The recovery rates were between 95 to 105Since the automatic GEM analyser switched between two gold tubes and gets a value every 5 min, the two concentrations were averaged in each 10 min (matched to the single height sampling interval by adjusting the time relay) to avoid possible bias caused by different gold tubes. The 20min variations of GEM concentration at certain height were between -2

4. The characterization of the two sites (clean and contaminated) could cause confusion. Given the mean annual TGM concentrations (3.64 and 5.93 ng m-3), both locations should have been considered under the influence of regional Hg emission plumes. I suggest using “mildly polluted” and “moderately polluted” to avoid the confusion.

Revised

5. It would have been extremely useful if Hg flux was also measured over the forest floor under the canopy. Such data will help enhance the source/sink discussion of Section 3.4.

The GEM fluxes were also measured over forest floor under the canopy at QYZ site, and the results showed the soil manifest as net GEM sources at all the seasons (0.13 $\pm$ 0.43, 1.54 $\pm$ 1.78, 4.76 $\pm$ 1.86 and 2.07 $\pm$ 1.73 ng m-2 h-1 in winter, spring, summer and fall respectively; unpublished data) were added in SI, the discussion was added in Section 3.4.

6. Recent characterization of stable mercury isotopes in foliage of various ages and litter, along with the quantification of litterfall production and Hg deposition through litterfall all indicate that forest ecosystem is a net sink for Hg in remote regions. Such

data are also supported by the vertical gradient measurement of TGM concentration from forest floor through well above forest canopy, showing increasing TGM concentration with sampling height. The primary conclusions of this study appear to contradict these recent findings, even given the elevated TGM concentration in air. Since the flux data above forest floor are not reported in the manuscript, it is not possible to determine if it is the re-emission of contaminated soil that leads to the overall Hg source term. An in-depth discussion regarding these discrepancy will significantly enhance the scientific value of the manuscript.

There are ongoing debates regarding whether or not forest is a sink or a source of GEM because the forest/air exchange flux is the sum of vegetation and soil exchange flux, depending on not only atmospheric concentration and meteorological conditions, but also plant community composition and soil structure (Bash and Miller, 2009; Converse et al., 2010) over shorter or longer periods. The study of foliage/atmosphere mercury exchange at QYZ indicated that the vegetation presented as a net GEM source with a value of 1.32 ng m-2 h-1 (2.19, 0.32 2.51, -0.01, 1.32 ng m-2 h-1 in winter, spring, summer, and fall respectively) caused by high rates of photoreduction and plant transpiration due to high temperature and radiation, relatively large leaf surface area and elevated mercury deposition (Luo et al., 2016). In addition, the study of the mercury exchange between atmosphere and soil under the forest canopy at QYZ through the DFC methods also showed the soil manifest as net GEM sources at all the seasons (0.13 $\pm$ 0.43, 1.54 $\pm$ 1.78, 4.76 $\pm$ 1.86 and 2.07 $\pm$ 1.73 ng m-2 h-1 in winter, spring, summer and fall; unpublished data), and the results were added in SI. Thus, the net emissions observed at QYZ were contributed by both soil and foliar emissions. The GEM fluxes over forest canopy (8.09 ng m-2 h-1) in this study were almost similar to the sum (7.27 ng m-2 h-1) of emission fluxes from foliage and soil in summer, but had lager values in other seasons. It might be because of the underestimation of the GEM fluxes from soil due to the decreased turbulence in chamber using the DFC method, and the lack of GEM fluxes from the undergrowth vegetation. Although there were net GEM emissions (58.5 μg m-2 yr-1) from forest in this study at QYZ

site based on the measurement of the GEM fluxes over forest canopy, on account of extremely large Hg deposition (wet deposition:14.4 μg m-2 yr-1; dry deposition: 52.5 μg m-2 yr-1; Luo et al., 2016), the forest presented as a Hg source, overall. It is a pity that foliage/atmosphere and soil/atmosphere mercury exchange at HT have not been measured, respectively. However, the comparison of Hg content of current-year foliage and soil between two sites might indicate that there were larger GEM emission fluxes from soil but much larger GEM adsorption by foliage. In addition to the lager GEM concentration and soil Hg content, the in-depth discussion of discrepancy caused by many other factors including the flux quantification method, specific tree species (evergreen tree species generally have higher exchange capabilities of GEM relative to deciduous tree species), reemission from photoreduction and transpiration was added in Section 3.4.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-349/acp-2017-349-AC1-supplement.zip

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