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

comments from RefereesïijŽForest is an important ecosystem on the earth. Characterizing the role of forest in the global biogeochemistry cycling of Hg is an important research topic in global Hg cycling studies. This study investigated the GEM gradient at two typical forests in subtropical zone of China. GEM concentrations and GEM fluxes measured are valuable for the atmospheric Hg studies in regional and global scale. I don't see major errors in the sampling techniques and method, and the discussions, for

most cases, are sound throughout the whole manuscript. I would suggest a publication after addressing the following questions.

Comments 1: Major points: The result from this study found the forest in subtropical zone of China is a net source of atmospheric GEM, and I have no doubt for this result. My question is, why the net emissions were observed in the study areas, soil emissions or foliar emissions? The authors did discuss this scientific issue in section 3.4, but we still don't know the exact causes. I think this is critical for atmospheric science. As we know, previous field observations on foliage/atmospheric Hg fluxes mostly revealed a net sink of GEM. If forest canopy is a net source in the study area, this would be an important finding. If the net emissions of GEM from forest were caused by elevated soil Hg emissions (soil Hg concentrations were elevated), then future studies regarding the mass balance of GEM in forest using litterfall approach approaches should also consider the soil emissions or reemissions. The net emissions observed in this study might be also due to many other factors including the specific tree species (evergreen tree species generally have higher uptake capabilities of GEM relative to deciduous tree species), lower leaf area index, reemission of dew water and transpiration stream, which should be also assessed in the manuscript.

Response 1: 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 showed the soil also 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 respectivley; unpublished data). The above 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.

Comments 2: Minor points: Line 79: why did the authors select the two sampling heights at QYZ and HT? Are you any pervious studies to support your setting; Will the chose of different sampling height affect the flux result? Line 81: 'half canopy height'? The lower sampling heights were 25 and 22.5 m agl, which were much higher than the canopy height.

Response 2:The result measured by AGM represent a mean value of GEM flux in

regional area, i.e, footprints area of tower, which is related to the measuring height and meteorological conditions (Fritsche et al., 2008b). Previous study estimated that the footprint of intake at 40 m height on the flux tower was 100 - 400 m (Zhao et al., 2005). Therefore, the footprints of the intakes located at different height in our study may be similar due to the relatively uniform distribution of pinus massoniana or cunninghamia lanceolata forest within 500 m around the flux towers in our research. Considering the extremely large disturbance of temperature and wind speed over forest canopy, especially close to the canopy, the lower air intake should be set at least half canopy height (Table 1) above the canopy to ensure the stability of the results (Lindberg et al., 1998). Thus, the lower sampling heights were about 1.5 times of the canopy height.

Comments 3: Section 3.1: I think the annual variations of GEM gradient at QYZ and HT are also important and should be presented in Figures. The authors should also show the annual, seasonal and diurnal trend of GEM gradient.

Response 3: The GEM gradient and the turbulent transfer coefficient (K) are both critical for estimating GEM flux, and the annual, seasonal and diurnal variation in different season were presented in SI.

Comments 4: Line 155: the authors should note the sampling height of the annual mean GEM concentration.

Response 4: The atmospheric GEM concentrations presented in manuscript were the average GEM concentrations at two heights, which was clarified in the revised manuscript.

Comments 5: Line 156: the global and northern hemisphere background should be referred to GMOS studies.

Response 5: A reference for GMOS studies was added.

Comments 6: Line 159: References are needed here

Response 6: Two references were added.

Comments 7: Line 161: Do you have any evidence for this hypothesis? I strongly suggest the authors analyze the source-receptor relationships at the sampling sites? Measurement of GEM in the atmosphere is also an negligible part of this study.

Response 7: The sentence was removed. The source-receptor relationships at QYZ site was analyzed by using the Hg content of precipitation and throughfall, and the fluxes of soil/atmosphere, foliage/atmosphere and forest/atmosphere, the discussion was added in Section 3.4.

Comments 8: Line 190: should be 'have positive values at QYZ'.

Response 8: Revised

Comments 9: Line 196: References are needed here.

Response 9: "See section 3.3" was added because there were many related references and discussion in section 3.3.

Comments 10: Line 227: the source from WS mercury mining area? Any evidence?

Response 10: The WS mercury mining area is located in the northwest of the HT site about 100 km away. The sudden rise of GEM concentration not only on May 14 presented in Figure 7, but also on January 17, September 5, October 10 and November 17 24 at HT site, corresponded to northwest wind prevailed according to the wind direction records. Thus, we believe that the sudden rise of GEM concentration might be caused by the approach of a high-mercury-content air mass from WS Mercury Mine leading by northwest wind.

Comments 11: Line 297: the study only reveal the whole forest is a net source, but not vegetation, you did not measure foliage/atmospheric Hg flux. Table-1: the relative abundance of major tree species should be listed.

Response 11:The study of foliage/atmosphere Hg fluxes was also conducted at QYZ site by using the DFC method. And the results were published in 2016, and could

be used to explain the contribution of foliage to the whole forest GEM emission. The relative abundance of major tree species was listed in Table 1.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-349/acp-2017-349-AC3-supplement.zip

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