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Interactive comment on "Atmospheric total gaseous mercury (TGM) concentrations and wet and dry deposition of mercury at a high-altitude mountain peak in south China" *by* X. W. Fu et al.

Anonymous Referee #2

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The paper reports on gaseous mercury measurement at the summit of Mt. Leigong in southern China over a period of one year. Seasonal and diurnal variations are described and the elevated concentrations are interpreted in terms of transport of polluted air masses from different regions of China. Mercury in rainfall, throughfall and litterfall was also determined and the data were used to calculate wet and dry deposition fluxes. Apart from several awkward or unclear sentences the paper is generally well written. However, the chapter on deposition fluxes seems to me to be flawed for reasons detailed below. Thus I recommend the publication of the paper with substantial modifications suggested below.

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Deposition fluxes: The annual dry deposition fluxes are calculated from the measured throughfall, wet deposition and litterfall. In the "Site description" the authors mention that misty weather prevails at the summit of Mt. Leigong with cloud periods exceeding 300 days per year. That means that a substantial part of mercury might be deposited by interception of cloud droplets which will not be included in wet deposition but might partly be included in throughfall. In fact, low THg concentrations in precipitation combined with high litterfall THg concentrations as discussed in the second paragraph on page 23478 just indicate that a substantial deposition might be connected with cloud interception at Mt. Leigong. Flux estimations under these conditions would probably need additional measurements of mercury in cloud water and the term "dry deposition" then does not apply under these conditions. Neither can the findings made under these very specific conditions be generalized for other areas less exposed to cloud contact. For all these reasons this section is highly speculative and needs to be rewritten or even deleted. Because of its speculative character I would also suggest to move it, in case it is not deleted, from the more prominent second position in "Results and discussion" to a less prominent place somewhere at the end and to put less emphasis on the flux measurements in the abstract and in the summary and conclusions.

Other factual remarks:

The description of the sampling of precipitation and throughfall on page 23471 needs more information. Were the precipitation and throughfall samples from individual events stored individually before combining them into weekly samples or were they accumulated in the collectors over the week? As described, throughfall was sampled simultaneously with precipitation. But interception of clouds might lead to "throughfall" even in absence of rain. Was this observed and were there any attempts to measure it?

On page 23470 the authors claim that they measure TGM, because GEM is dominating and RGM does not matter according to the stated references. But the situation at the summit of Mt. Leigong (2178 m) with upslope and downslope drafts is quite similar

to Mt. Bachelor (about 2700m, Swartzendruber et al., JGR, 2006 – this reference should be added to the reference list) where substantial RGM concentrations during the downslope transports of free tropospheric air were observed. The equation TGM = GEM is thus not valid at these conditions. In addition, the frequent cloud contact at Mt. Leigong would almost certainly scavenge all RGM and, according to the evidence in literature, RGM does not pass through the sampling tubing to the gold cartridges under conditions of high air humidity. It is thus almost certain that only GEM was measured and the measurements should be presented as such.

The average gaseous mercury concentrations are presented on page 23474 as geometric means because of a lognormal distribution of the data (as shown in Fig. 3). They are then compared with measurements at Mt. Gongga (geometric mean), Mt. Changbai (arithmetic mean) and other places (mostly arithmetic means). As switching between different means make a meaningful comparison difficult the authors might think of additional presentation of medians which are less influenced by extreme values. Number of measurements should also be given to make statistical tests for mean differences feasible. This applies also for Table 2.

In the second paragraph on the page 23475 possible reasons for differences of elevated gaseous mercury concentrations at Mts. Gonggba and Changbai are discussed in terms regional pollution. As neither the station at Mt. Gonggba nor the station at Mt. Changbai are summit stations the authors might also allow for meteorological reasons for the discussed differences. E.g. nighttime downslope transports of free tropospheric air at Mt. Leigong may push the average concentration down in comparison with the other two stations where this flow regime might be absent. Also Mt. Leigong is the southernmost of the three stations and the lowest mercury concentrations there might be a result of a general decreasing gradient from the northern latitudes of about 50oN to more southern latitudes.

On page 23482 the average diurnal variation of gaseous mercury concentrations is correctly interpreted in terms of downslope transports during the night and upslope

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transports during the day. The consequence is that the nighttime measurements are less influenced by nearby sources and provide thus a better representation of the regional or even hemispherical background concentrations than the averages presented in Section 3.1 and in Table 2. For this reason the separate investigation of the nighttime levels of gaseous mercury concentrations and their seasonal variation might provide a much better insight into the large scale distribution of atmospheric mercury and its seasonal variation. A comparison of nighttime averages with other measurements in the northern hemisphere might be revealing. The authors mention a separate evaluation of nightime concentrations at the end of Section 3.5.2 and in Section 4 but they discuss only annual mean values at Mt. Leigong. In Section 3.5.2 backward trajectories and long term transport are discussed in terms of clusters of origin but for the reasons mentioned here this makes sense if only nighttime measurements are discussed. The text does not mention any downslope/upslope selection of the data used for the cluster analysis. It might thus be useful to add a nighttime version of the Figure 8 and its discussion.

Some editorial remarks:

Page 23468, line 15, 20 and 25: "knowledge" instead "Knowledge", delete "China", and "limitations" probably better than "restrictions", respectively.

Page 23469, line 12: "elevation of about 1000m against the surrounding..." probably better.

Page23470, line 23: The term "was guaranteed" usually apply for sold goods and is too strong in this case. A more modest "was controlled" or "was ensured" is more appropriate.

Page 23472, line 1: The sentence starting with "Field blanks..." awkwardly long and not quite correct. Field blanks cannot ensure a zero contamination as this would mean a zero concentration. But they can ensure that the contamination is much lower than the concentrations typically found in the samples.

Page 23472, line 13: I am wondering about the packing of litterfall samples into paper bags? How clean in terms of mercury concentrations is the environment and the air in which litterfall was air-dried?

Page 23472, line 19: "Between grindings.. " is probably more appropriate.

Page23477, line 17: "at Mt. Leigong"

Page 23477, line 28: "small" instead of "restricted"

Page 23479, line 4: "bi-directional pattern"

Page 23482, line 21: "Mt. Leigong". The two last sentences in this paragraph need rewording. "the elevated TGM concentrations in the free troposphere were probably caused by updraft" is in contradiction with the downslope flow regime at Mt. Leigong. If there was an updraft, it must have been far away as mentioned by the second sentence.

Page 23486, line 11: The sentence "Diurnal variations ... were likely due to mountain valley breeze circulation" reads like a tentative conclusion. In fact, it is certainly so as observed at other mountain summit stations like Mt. Bachelor.

Why do the TGM concentrations in Fig. 2 start with a negative concentration of -1 ng/m3?

Fig. 9: The yellow trajectory is hardly recognizable.

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