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Interactive comment on “Assessing the trends and effects of environmental parameters on the behavior of mercury in the lower atmosphere over cropped land over four seasons” by A. P. Baya and B. van Heyst

Anonymous Referee #1

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Review of Assessing the trends and effects of environmental parameters on the behavior of mercury in the lower atmosphere over cropped land over four seasons

General comments

This manuscript addresses the important question of terrestrial surfaces as a source and/or a sink for atmospheric mercury. The authors' study follows the previous work of several other studies but it remains unclear to me why their study is useful to the scientific community and what is the problematic. The data presented here are of good

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quality though the use of the different abbreviation TGM, Hg(0) or RGM is to my opinion not satisfactory. This should be clearly defined in the experimental section. Due to instrumental failure, there are data missing which I can understand. But I encourage the authors to clarify the period when data are missing. There are several contradictions throughout the manuscript. Moreover it seems to be not convincing to discuss about fall data without a full data set. Overall, the method section is too weak.

The overall impression is that a significant work has to be made on discussing the most important facts and to avoid on long discussion on common admitted facts such as sources. A stronger discussion should be addressed on emission processes. Then, authors should try to better compare their data with others. No attempt is made to discuss the Hg speciation data, thus I wonder if there are useful for the present study. If not, they should be removed. Finally, a very important effort has to be made on the figures which are not of suitable quality for several aspects such as readability, scales, printing quality, etc. Some figures are also to my opinion useless and an effort should be made to combine other figures.

I feel that this manuscript could be synthesized a lot by combining different figures, results and discussion and that it should be completely revised.

Specific comments:

Abstract

line 10 : a “one year study” : it is wrong

Introduction

The introduction should introduce much more in detail the state-of the art of Hg fluxes knowledge over soils in particular over agricultural ground covers, snow covers.

p2551

line 7. It is commonly admitted today that the residence time of Hg⁰ is around 1 year

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(e.g. (Lindberg et al., 2007)

line 22. Do the authors mean RGM and HgP at background sites?

line 25. This sentence is quite evasive and the references are not up to date. Do you mean that 95% of atmospheric mercury accumulates in the soil? “Accumulate” is not the proper word. There are recent model that suggest that a global flux of 33 Mmol/y is deposited on earth with 55% deposited on terrestrial surfaces (Mason and Sheu, 2002). There is confusion between soil and terrestrial ecosystem (that include vegetations that could play a role on the deposition fluxes).

p2552

line14. “one short-coming...” . I did not go through all the references, but Obrist et al.’study has been conducted over 1 year! The author’s statement is therefore wrong. What about your table 3, it seems that there are many other annual studies.

Methodology

p2553

Reading the first line, I realise that the study has started on November 1st, while the abstract says that it started on July! Then November 1st to august 13th cannot be considered as a full year. Almost $\frac{1}{4}$ of the year is missing. I am concerned about your definition of the fall season that will be use in the next section. This season, probably defined for the period sept 21st to dec 20th, is reduced to 1 month and the half. The measurements period and missing data have to be clearly defined.

The authors do not mention the influence/presence of agricultural work on their station that may disturb their measurements: mechanical tools, important soils disturbance, watering, etc.

line 15 : “the top soil average (...)” . An average value is presented. I assume that it is a total mercury concentration (it should be mentioned). How is this average calculated?

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have several samples been collected ? Where, when? Or do the authors assume that the THg content is constant throughout the year ?

line 16: Acid digestion is not enough to analyse a mercury concentration.

line16: How is calculated ' 0.006 '? What does it represent? You cannot have a better precision for you error (0.006) that for your mean value (0.05). This is not consistent. In that case, you should write 0.05 ± 0.01 or if applicable with your method 0.050 ± 0.006 . There are several mistakes throughout the manuscript, and I encourage the authors to check all the values (including their tables)

p 2554

I have several problems to understand the instrumental set up and I want to make sure that a reader can understand. TGM total gaseous mercury comprises Hg° plus a portion of gaseous $\text{Hg}(\text{II})$. Usually, if a filter is used (and/or a sodalime trap) at the inlet sample port, $\text{Hg}(\text{II})$ would probably not pass the filter and thus the 2537A unit determines the GEM concentration only. The setup is not described enough but I assume that a Teflon or equivalent filters is used at the sample inlet port (or perhaps two as it is usually prescribed: one for the sample inlet and one outside). Unless the authors have done some tests, I think that TGM should be replaced by GEM or Hg° .

Are the micromet data available on the tower? Are measurements carried out at a height of 8.5 m?

A method detection limit for RGM and PHg has to be shown/evaluated to discuss the data presented later. You should evaluate the precision too.

Why did you use such a high flow rate whereas the typical flowrate of 2537 is 1.5 lpm? Where is exactly located the pump? Is it contamination-checked?

Why didn't you try to measure RGM fluxes ?

line 25 why don't you use GEM data from the speciation unit???

p2556

line 21 I am not sure that the mean GEM value can be really compared to other sites since the other studies show seasonal means, or multiyear averages. Why do the author not include all the data cited in the introduction in their table 3 (Edward 2005; obrist, Engle?)

line 24 “the behaviour of TGM (...) displayed diurnal patterns with highest concentrations recorded at midday”. From your figure1, this conclusion cannot be drawn. Winter values are: at midday, gem is 0.95, around 0.98 at 9 am, around 0.93 at midnight. You should put error bars. Is it statistically higher?

p2557

line 3 What is an inverse trend?

line5 What kind of regression analysis? What is the statistical tests used in this manuscript?

line 9 “after June (...)” the deviation is not that clear for august and authors do not have any data for September and October. How can they say that TGM is elevated in the fall in compared to the net radiation level? Is it only based on measurement carried out in November? What is the purpose of this paragraph?

p2558

line 6 what about the winter? Is there any industrial influence?

p2559

Line 24 : now it is a multiple regression? Please clarify in the methodology section the statistical test that were applied.

p 2560

Section 3.3.1 Why is there no data in December-January? Why is there no data in

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April?

from figure 7, I cannot see any evidence of a more pronounced diurnal pattern (how can you derive a diurnal pattern from this graph?)

line 19: high value of 130 is not visible on the figure

line 24 To which extent the soil moisture affect the flux? Does the moisture at 30 cm deep is likely to influence Hg° fluxes?

line 25 where do you see high TGM fluxes during the winter?

The facts that radiation affects the Hg° flux seem to be obvious to the authors throughout the manuscript. They should mention what are the mechanisms involved in that irradiation-mediated emission? Is it photochemistry? Is it an effect of increased temperature, and indirect effect of increase microbiological activity?

p2561

line 3 that mercury is liberated from one water molecule is physically impossible.

Reading Cobbett and van Heyst, I do not see this hypothesis. That the solubility of Hg (which oxidizing state?) is lower in the ice than in the water has to be supported by a reference.

line 13 I do not see how the soil air may promote aqueous conversion of bound mercury to elemental mercury. Do you mean that Hg° is adsorbed on the soil matter (what kind of molecules) and then desorbed? or is there a chemical reduction of Hg(II) to Hg° . This part is quite unclear.

line 21. "A spike of TGM occurred following a major precipitation". How do you explain this phenomenon?

p2562

The snow might be considered as a source of Hg° too ((Lalonde et al., 2002; Lalonde

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et al., 2003). Why is it not visible? What about the peaks around days 60, 70.

line 5 When does the snowmelt occurs ? The snowmelt is known to liberate solutes in a sort of ionic pulses (Bales et al., 1993). This might be applicable to mercury(II).

Table 1

define the seasons is this table relevant to the article? correct win by wind

Table 2 the first row is incorrect, please revise. why is there no data in march for rgm and pm

Figure 1 and figure 3 there are no error bars Are the figure 4 and 5 really useful?

Figure 7 to 10

the horizontal scale is not acceptable.

Figure 9 is of really poor quality and thus difficult to read

Some typos. 2251 line11. "states" p2552 line2 "compartments. " Instead of "compartments" line 24 : these

Bales, R. C., Davis, R. E., and Williams, M. W.: Tracer release in melting snow: diurnal and seasonal patterns, *Hydrolog. Process.*, 7, 389–401, 1993.

Lalonde, J. D., Poulain, A. J., and Amyot, M.: The role of mercury redox reactions in snow on snow-to-air mercury transfer, *Environ. Sci. Technol.*, 36, 174–178, 2002.

Lalonde, J. D., Amyot, M., Doyon, M. R., and Auclair, J. C.: Photo-induced Hg(II) reduction in snow from the remote and temperate Experimental Lakes Area (Ontario, Canada), *J. Geophys. Res.*, 108, 2003.

Lindberg, S., Bullock, R., Ebinghaus, R., Engstrom, D., Feng, X. B., Fitzgerald, W., Pirrone, N., Prestbo, E., and Seigneur, C.: A synthesis of progress and uncertainties in attributing the sources of mercury in deposition, *Ambio*, 36, 19–32, 2007.

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Mason, R. P., and Sheu, G. R.: Role of the ocean in the global mercury cycle, *Global Biogeochemical Cycles*, 16, 2002.

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10, C473–C480, 2010

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