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Interactive Comment

Interactive comment on "Summertime elemental mercury exchange of temperate grasslands on an ecosystem-scale" *by* J. Fritsche et al.

J. Fritsche et al.

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Response to interactive comment of anonymous Referee #2

The referee has reviewed the manuscript very carefully and merits our work as a valuable contribution to the assessment of the mercury exchange in different environmental compartments. The presented study not only provides information for mercury transport models, but also endeavours to apply micrometeorological methods under challenging conditions at concentration levels of a few ng per m3. However, the referee has raised some important concerns, which the authors will address in the following. While in some cases these concerns must be attributed to imprecise descriptions in the manuscript, others require re-evaluation of some data, and some measurements may need to be omitted. The authors therefore propose to submit a revised manuscript that takes the aspects noted by the referee into account. These aspects and the proposed

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Interactive Discussion

Discussion Paper

changes to the manuscript are as follows:

1) Site description: the referee disapproves the description of the footprint at the study sites at Fruebuel and Oensingen and raises several questions about the size of the footprint, the uniformity of the vegetation cover and the sensor heights.

It is right, that the present description of the footprint at Fruebuel may lead to the assumption that the site might not have an adequate fetch. More details will be provided in the revised manuscript. It is furthermore correct, that the fetch length at the Oensingen site is smaller than the sensor height for the GEM gradient measurements would require. However, this inadequacy was accepted for the benefit of a well characterised grassland site and higher vertical gradients (the measured gradients are already close to the detection limit and a lower sensor height would produce gradients that could not be resolved with the applied methods).

The authors propose the following corrections to the manuscript:

• Site description Oensingen: add the EC sensor height (1.2 m); remark on insufficient fetch for GEM gradient measurements (as noted above); footprint contributions were calculated using the model of Kormann and Meixner (2001).

• Site description Fruebuel: "The predominant wind direction at Fruebuel is SW to SSW, showing a distinct channelled flow as a result of the local, undulating, sub-alpine topography. The footprint of the EC flux measurements has been determined by using the footprint model of Kljun et al. (2004). For the EC sensor height of 2.55 m this resulted in a footprint coverage of >80% within a radius of 60 m. Within approximately 200 m of the predominant wind direction vegetation is homogenous and the calculated footprint area covers the intensively managed part of the grassland."

2) Data coverage: the referee remarks that our effort to determine the bias of the applied method was unsatisfactory, as we performed the determination of the detection limit (MRG) only once at one site (for a measurement period of three days).

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The authors used cleaned Teflon tubing at each site and measured mercury free air at the beginning and end of each campaign as quality assurance steps (measurements of mercury free yielded values below the detection limit). The exact determination of the MRG is very time consuming considering the short measurement periods available, and the authors argue that the MRG is system-specific. They consider the applied quality assurance steps as sufficient for the estimation of the mercury exchange fluxes and their spatial variability. Moreover, the authors have conducted the MRG test at several occasions before this study and found that the detection limit changed insignificantly.

The authors propose the following corrections to the manuscript:

• Extension of the measurement setup section and the description of the MRG determination: mercury free air was measured at beginning and end of each measurement period; MRG was determined only once at one site; tubing was renewed at each site; studies at each site were performed separately.

3) CO2 and GEM fluxes: the referee is concerned about the discussion of the observed GEM (gaseous elemental mercury) gradients and fluxes. At the study site in Oensingen we observed a period with enhanced nocturnal GEM gradients that followed a rain event; these enhanced gradients were not visible during the day. The referee questions our explanation that atmospheric ozone would oxidise the emitted mercury – and thus obliterate a day-time gradient – but rather attributes the observed pattern to advection effects related to an insufficient fetch.

During the mentioned period we observed a marked relationship between the night-time gradients and the soil moisture content. To show this effect we did not exclude flux data determined under low windspeeds by the aerodynamic method (Fig. 3; this was done for the MBR-GEM fluxes and the EC-CO2 fluxes). However, under such conditions, when turbulence beaks down, advection is in fact an issue and the affected data may need to be discarded.

The authors propose the following corrections to the manuscript:

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• Exclude those cases of the aerodynamic GEM-flux and CO2-flux data for which breakdown of turbulence occurred (as was done for the EC data); correct the discussion accordingly (update Tab. 1 and Fig. 3; replace the discussion of the GEM-soil moisture relationship with a discussion of the updated data).

The referee also points out that our conclusion "Both micrometeorological methods were consistent regarding the sign of the average fluxes..." is redundant, as it was the GEM gradient that determined the sign.

This is correct and we will remove this statement from the manuscript.

4) Discussion section: The authors smoothed the GEM flux data, which seems to have resulted in an inconsistent sign of the two applied methods during some phases. The referee hence proposes to reduce the length of the smoothing window.

The authors propose the following corrections to the manuscript:

• Reduce the applied moving average filter as suggested by the referee from a 9-point to a 7 or 8-point moving average. Smoothing of this length will result in a more noisy representation of the signal and the authors suggest to apply a different filter method (e.g. locally weighted regression [lowess], rectangular filter) if this will improve the result.

The referee also questions our evaluation of the two applied micrometeorological methods and states that we report some of our data as being valid, although certain assumptions of these methods were not met.

In the manuscript the authors discuss the sources of uncertainty and explain discrepancies of the aerodynamic and modified Bowen ratio (MBR) methods for the estimation of GEM exchange rates. The authors applied the two different methods in order to evaluate their suitability and show the limitations of each for GEM flux estimations. It first appeared, that the MBR method would be more robust and hence the method of choice. But, looking at the results and considering the assumptions for the MBR

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method (equal sources and sinks of GEM and CO_2; similar spatial variability of GEM and CO_2), it seems that the aerodynamic method yields a better estimation of the GEM flux. To discuss this finding, it is necessary to report the results as shown in the manuscript.

The authors propose the following corrections to the manuscript:

• Discussion of the MBR method: "Due to complex vertical distributions of sources and sinks of trace gases within terrestrial ecosystems, the theoretical basis for the MBR method may be generally questioned. Nevertheless, there are several examples in literature (e.g. Doskey et al., 2004; Muller et al., 1993; Walker et al., 2006) where it could be shown that despite this shortcoming, the MBR yield sensible, unbiased flux estimates. It is therefore legitimate to test the applicability of the MBR method for measuring GEM fluxes. The comparison with the aerodynamic method presented in our manuscript shows that the latter yields more reliable GEM fluxes than the MBR method. This may actually be due to the fact that during daytime conditions the net flux of CO_2 is dominated by plant photosynthesis (uptake of CO_2 from the atmosphere), obscuring the release of CO_2 from the soil surface. As the soil is thought to represent the major source of GEM this would clearly invalidate the theoretical basis of the MBR method."

• Introduction: Emphasize the objective of this study by adding " ... we test the potential and limitations of the two methods and attempt to capture... "

Finally, the referee questions our discussion of the oxidative effect of ozone that seems to suppress GEM emission from the soil surface. While the authors argue, that the day-time reaction of ozone with GEM would reduce GEM emission, the referee thinks that emission would be enhanced.

As during our measurement campaign ozone was in excess the authors think that any mercury emitted from the soil surface is readily oxidised (maybe by a heterogeneous reaction at the soil and plant surfaces) and hence not detected, even at the lowest

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sampling line.

The authors propose the following corrections to the manuscript:

• Remove the sections that address a possible relationship between ozone and GEM-flux, as this might be too speculative and not absolutely necessary for the scope of this paper.

Technical/Editorial comments of the referee:

ad 1) will be changed as suggested

ad 2) we will replace atmospheric turbulence by friction velocity as suggested

ad 3) the unit ng/m3/m is correct; GEM concentration is measured in ng/m3 and the gradient is scaled to a height of one meter

ad 4) the reference indeed pertains to a remote, but contaminated site; will be corrected

ad 5) will be corrected as suggested

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1951, 2008.

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