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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An ecosystem-scale perspective of the net land methanol flux: synthesis of micrometeorological flux measurements" *by* G. Wohlfahrt et al.

Anonymous Referee #1

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The authors present a comprehensive synthesis of methanol flux measurements at ecosystem scale which seems particularly interesting in the context of GPP and ecosystem respiration. Ecosystem scale measurements are critical to understanding complexity of sources, fates and sinks of atmospheric methanol. One of the emphases is on the emerging observations of methanol deposition and on characterization of controls behind the bidirectional exchange of methanol. This kind of synthesis/compilation papers is increasingly needed for taking a bigger-picture perspective, something which individual contributions on their own could not fully achieve. Overall, the story aligns nicely with the scope of ACP and will be a useful reference of atmospheric methanol

ecosystem perspective. Below are just a few ideas for further discussions and relatively minor suggestions.

General

1) Globally plants are thought to be primary source of methanol and the relationships with GPP are typically clear for vegetative sites (e.g. Figure 5). Bearing in mind the challenges behind methanol measurements (e.g. the use of right materials in the sampling line, characterizing instrumental backgrounds, etc.), the compiled ecosystem flux dataset (altogether from 28 measurement sites) is impressive and it is encouraging to see the consistency of the net land methanol budgets with grand mean ecosystem flux measurements, even though currently most represented in measurements are temperate climates. The question is how to achieve the temporal and spatial representativeness in all different kinds of ecosystems including (and maybe focusing on) the tropics where the densest biomass is located. Consequently, another question is if we can constrain the overall uncertainty from environments which have never been sampled from? If relying on scaling from GPP data to infer methanol flux, how can we be certain that the part of unexplained variance is not disproportionally substantial in other sites in terms of possibly completely different magnitudes of methanol fluxes uncoupled from GPP?

2) I think that "future direction" element of the paper could be further enhanced. It is clear in the text that it makes sense to consider deposition and make use of micromet measurements but how can we address the enormous variability of fluxes during stress, different management practices, phenological/seasonal cycles, and massive herbivore infestations in the future global estimates? Perhaps the solution could be more routine long-term ecosystem tower networks (e.g. FLUXNET) which could include methanol measurements, and the constraints from mobile platforms and remote sensing?

3) The paper suggests that controls behind biosphere-atmosphere methanol exchange

seem to be largely site-specific. Thus, not only spatial representativeness of sites is needed, but also long-term character to characterize seasonal variations. Ecosystem scale measurements are excellent to characterize the net fluxes, but are they alone sufficient to understand the controls? While focusing on the EC fluxes why not also to combine with the full array of available tools including remote sensing, and look at scales from molecular through leaf, branch, tree, ecosystem, regional, to global?

4) Because methanol is a relatively non-specific volatile tracer in the atmosphere having numerous different sources and sinks, modeling methanol bidirectional exchange must necessarily be challenging. It is thus quite impressive that MEGAN seems to be doing a reasonably good job for modeling vegetative methanol but there could certainly be sources it cannot capture. The latest MEGAN version description (Guenther et al., 2012) suggested deposition estimate which I guess can be one approach when we simply do not have measurements or information on controls. One other question is how we can represent stress in the model and how to deal with the compound which can be both stress related and stress unrelated? Should methanol be classed as a special complex case of a BVOC or maybe we need to step back and look holistically using interdisciplinary approaches to understand atmospheric methanol better and then come up with the holistic (perhaps chemometric) modeling approach, embracing all sources, sinks and controls (see comment below)?

5) Are we missing any critical methanol sources? For example, large methanol emissions can be triggered by herbivores (e.g. von Dahl et al., 2006). These emissions are not just a result of wounding of a leaf but are the sustained stress-elicited responses as a result of defense mechanisms. Every year an unimaginable number of catepillars molt into butterflies devouring portion of foliage. Has anyone looked at their life cycles that typically coincide in spring when the vegetation is growing? What is the role of up to 10 million epiphytic microbes per cm2 of foliar surface (Lindow and Brandl, 2003) for methanol emission/uptake?

6) In terms of methanol deposition, seems like the biggest problem is that we are lack-

ing the measurements around the globe and as authors nicely point out the deposition largely depends on specific site. Deposition of methanol in a situation when anthropogenic methanol (e.g. from a landfill) is advected and deposited onto vegetation/soil can be different from modeling deposition of methanol emitted by elongating conifers and taken up by soil microbes within the canopy. Could it be worthwhile to return to laboratory for dedicated fumigation experiments to understand and characterize stomatal and non-stomatal uptakes of methanol?

7) Given low Henry's Law constants, I wonder if it would be interesting to look more closely at how rain frequency, fog, surface wetness affects global net estimates of methanol exchange?

Specific

8) In terms of the take-home message from this excellent synthesis (last sentence of the abstract), I agree that modeling separately deposition could be opposing the errors and this is important to mention but how do these errors compare to errors in overall uncertainty of global estimates (should not be mentioned?).

9) Introduction first paragraph: Could also add that methanol is often the most abundant in various places (example references).

10) Introduction: second paragraph. When talking about primary and secondary sources, is it not relevant to include emissions from dairies, for regional atmospheres at least?

11) P2583 L21 "little effort has been made . . . to standardize measurement protocols". Why not to make this effort here? This paper looks like a great opportunity to standardize or make the first step to standardizing these protocols for future methanol EC measurements which hopefully will be done more routinely in the future.

12) I like the idea of summarizing methodologies in a table so I found table S1 particularly useful. The setups of different PTR instruments indeed seem impressively

consistent. It would also be nice to include if the correction for isotopic 17O16O+ oxygen was done or not in each PTR-MS study and what was the relative percentage of O2+ relative to primary ions.

Technical

13) P2582 L26 “so-called” may be unnecessary.

References

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., & Wang, X. (2012). The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2. 1): an extended and updated framework for modeling biogenic emissions.

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