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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.

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Interactive comment on "An ecosystem-scale perspective of the net land methanol ïňĆux: synthesis of micrometeorological ïňĆux measurements" by G. Wohlfahrt et al. Anonymous Referee #1 Received and published: 26 March 2015

The authors present a comprehensive synthesis of methanol ïňĆux 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

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Discussion Paper

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 ïňĆux 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 ïňĆux 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 ïňĆux, 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 ïňĆuxes uncoupled from GPP?

Reply: Achieving temporal representativeness requires that the community moves away from campaign-style to year-round and multi-annual measurements. We believe that this is already partially the case (3 out of the 8 study sites in this analysis had multi-year data) and also that this analysis shows the potential of seasonal/multi-year measurements for better understanding the temporal variability of the ecosystem-atmosphere exchange. The need for longer-term measurements is addressed on p.

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2593 l. 18-21 of the ACPD paper. In terms of spatial representativeness, site selection presently is entirely bottom-up and thus lacks the degree of representativeness, which would be required at global scale. Improving on this is likely to require a co-ordinated effort by the community and thus appropriate international funding. The need for studies in presently under-sampled regions is addressed on p. 2593 l. 13-17 of the ACPD paper. Constraining the uncertainty of fluxes which have never been measured, in our opinion, falls into the category of "known unknowns". Statistically speaking, our sample of study sites is known not to be representative of the population and thus we cannot answer this question. The uncertainties associated with this sampling bias are addressed on p. 2587 l. 2-5 of the ACPD paper.

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 ïňĆuxes 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?

Reply: The need for multi-year and more spatially representative flux measurements is discussed already on p. 2593 l. 13-21 of the ACPD paper. Following the reviewer comment we will add further discussion on the need for multi-disciplinary and multi-scale measurements to the conclusion section.

3) The paper suggests that controls behind biosphere-atmosphere methanol exchange seem to be largely site-speciïňĄc. 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 ïňĆuxes, but are they alone sufïňĄcient to understand the controls? While focusing on the EC ïňĆuxes why not also to combine with the full array of available tools including remote sensing, and look

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at scales from molecular through leaf, branch, tree, ecosystem, regional, to global?

Reply: Ecosystem-scale measurements alone are clearly not sufficient to move forward, as our study has shown that more detailed measurements are needed to understand the net exchange of methanol at the ecosystem scale. We will expand the conclusion section to emphasise this issue.

4) Because methanol is a relatively non-speciïňĄc 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)?

Reply: We agree with the reviewer that improving on the rough approximation of the current version of MEGAN will be challenging. We expect that algorithms will be needed to represent both the stress and non-stress components of methanol emissions, as well as other sources and sinks. Methanol is the not the only BVOC to be generated by more than one process but the large flux and the different controlling process warrant treating methanol differently than other compounds.

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

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as a result of defense mechanisms. Every year an unimaginable number of catepillars molt into butterïňĆies 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?

Reply: Extreme events are indeed an issue which is poorly represented in ecosystem-scale data (but see e.g. Bamberger et al. 2011) and thus poorly constrained in models. Performing multi-year measurements increases the likelihood of observing extreme events, such as insect attacks. We will expand the conclusion section to emphasise this point. The need to better understand the role of microbes inhabiting plants and soils is already discussed on p. 2592 l. 14-19 of the ACPD paper.

6) In terms of methanol deposition, seems like the biggest problem is that we are lacking the measurements around the globe and as authors nicely point out the deposition largely depends on speciïňĄc site. Deposition of methanol in a situation when anthropogenic methanol (e.g. from a landïňĄll) 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?

Reply: We agree in that for some of the processes emerging at ecosystem scale there is the need to go back to the laboratory for conducting targeted experiments under controlled conditions in order to disentangle the processes involved. However, there is also the need for detailed studies in the field in order to make laboratory study results transferable to real-world conditions. We will add a short paragraph to the paper outlining this perspective.

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

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methanol exchange?

Reply: Definitely – doing so will require better data on surface wetness in order to be able to address condensation and drying after wetting, processes which are not captured by rain gauge data – a corresponding paragraph will be added to the paper.

SpeciïňĄc 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?).

Reply: Global budgets are closed to be consistent with atmospheric measurements – the variability in atmospheric inventories is thus typically small between studies (e.g. 3.4-4 Tg; Jacob et al. 2005). These similar atmospheric inventories are achieved with widely diverging sources (122-350 Tg/y) and sinks (50-270 Tg/y), however (Jacob et al. 2005). This issue is dealt in more detail throughout the main text of the paper and for the abstract just mentioned briefly.

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

Reply: done as suggested (including reference to Seco et al. 2011)

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?

Reply: Added a reference emphasising the role of dairy farming for regional methanol budgets (Gentner et al. 2014)

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 ïňĄrst step to standardizing these protocols for future methanol EC measurements which hopefully will be done more routinely in the future.

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Reply: Standardizing measurements is a totally different paper (likely in a different journal, e.g. AMT) with an entirely technical focus and thus out of scope of the present study.

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.

Reply: Generally the methanol flux calculated from the covariance between vertical wind and m/z 33 is not impacted due to internal background (e.g. the isotopes O18(O+), O2H+ and DNOH+) as demonstrated by Müller et al. (AMT, 2010), because it would only contribute random noise to the signal. All reported methanol concentrations were background corrected by bypassing air through a catalytical converter and substracting the residual from ambient measurements.

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

Reply: “so-called” was removed

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 2577, 2015.