Atmos. Chem. Phys. Discuss., 12, C7333–C7339, 2012
www.atmos-chem-phys-discuss.net/12/C7333/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The influence of boreal forest fires on the global distribution of non-methane hydrocarbons" *by* A. C. Lewis et al.

Anonymous Referee #1

Received and published: 24 September 2012

This paper reports measurements of the ratio of gas-phase non-methane organic compounds (NMOC) to CO in some boreal forest fire plumes in eastern Canada. The ratios are also used in a global chemistry-transport model (GEOS-Chem) to estimate the relative contribution of biomass burning (BB) and anthropogenic sources to the surface-level abundance of four hydrocarbons, globally, at the model spatial resolution. An important point is the widespread scale at which benzene regulations may need to be adjusted to realistically account for BB emissions, which are generally more difficult to control than anthropogenic emissions. Another useful result is that the BB and anthropogenic contributions are estimated specifically for several global atmospheric watch (GAW) stations. Benefits from the GAW estimates are (1) Demonstrating that interpreting future GAW data will require both better global BB inventories and better anthropogenic inventories (that reflect recent advances from pollution controls). (2)

Perhaps more important and perhaps too obvious to be mentioned by the authors is the study demonstrates that an enhanced suite of GAW NMOC measurements may help constrain anthropogenic and BB source strengths. The latter is highly significant because biomass burning is an enormous poorly-constrained source of emissions to the troposphere.

The paper reads very well, but it does have some gaps in its context and methodology. It may not be practical to re-run the model to address the gaps, but the gaps should be acknowledged in the introduction and the discussion at least in concise form for context and also used to inform a realistic, expanded discussion of uncertainty. The gaps are summarized next.

First, there is a huge previous literature estimating the relative source strength of anthropogenic and biomass burning emissions that is not compared too or even acknowledged in this paper. These estimates date back at least to Crutzen's 1979 Nature paper. For many years thereafter it was SOP for atmospheric chemists to switch open canisters all over the planet, measure a ratio to CO, multiply by a global CO estimate in Tg, take the result as a global estimate for the numerator species in Tg, and compare it to the global anthropogenic source in Tg. This tradition continued thru Crutzen and Andreae (Science, 1990) till about 1996 (Hao et al., JGR, 23577, 1996) before dying out. The old estimates had issues, the smoke age was often unknown for reactive species, many species could not be measured then, and they generally only compared source strengths and not "tagged" abundances. Thus, this work provides significant advances such as GAW-specific comparisons, but selected, earlier work pointing to BB as a significant global source could be cited for perspective.

Second, only one other biomass burning paper is compared too and the global model is fed by the authors boreal forest BB ratios even though BB emission ratios have also been measured directly in the tropics where most fires occur and from where the authors model output primarily originates. For instance, Sinha et al. (2003) used essentially the same methodology as the authors to measure NMOC/CO ratios in southern

ACPD
12, C7333–C7339, 2012

Africa. Previous work in the boreal region includes papers from the ABLE campaigns, Nance et al. (1993), Goode et al. (2000), etc, which are easily located via citations in the Simpson et al. (2011) paper the authors do cite. The extensive biomass burning emissions literature has been reviewed thoroughly from two perspectives by Andrea and Merlet (2001) and Akagi et al., (2011). Both papers can be used as a shortcut to determine if e.g. the tropical emission ratios may differ from the boreal ones the authors used in a significant way, or at least estimate the variability in these ratios. That would provide a more realistic idea of uncertainty than comparing to just one other study that was conducted from the same type of platform in the same forest type as the authors work.

Third, within the boreal region a (or the) major source of fire emissions is ignored. Many papers, have shown that much or most of the fuel consumption in boreal forest fires is from smoldering that produces initially-unlofted emissions that are chemically different (Yokelson et al., 2012) from the lofted emissions and not amenable to airborne sampling. Recent examples include: Yurganov et al. (2011), Turetsky et al. (2011), Hyde et al. (2011), etc. An initial framework to estimate the impact of initially-unlofted emissions on total emissions dates back to Bertschi et al. (2003) and is incorporated into the ecosystem-specific BB estimates of Akagi et al. (2011). The poorly lofted smoldering emissions may be difficult to deal with in a global model, but they are real and important to acknowledge. They occur in other global ecosystems as well such as Amazonian pasture fires (Kauffman et al., 1998) and peat fires in SE Asia (Rein, 2009).

Akagi et al. (2011) http://www.atmos-chem-phys-discuss.net/10/27523/2010/acpd-10-27523-2010.html

Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cycles, 15(4), 955-966, doi: 10.1029/2000GB001382, 2001.

ACPD

12, C7333–C7339, 2012

Yurganov et al. (2011) http://www.atmos-chem-phys.net/11/7925/2011/acp-11-7925-2011.html

Turetsky, M. R., Kane, E. S., Harden, J. W., Ottmar, R. D., Manies, K. L., Hoy E., and

Kasischke, E. S.: Recent acceleration of biomass burning and carbon losses in Alaskan forests and peatlands, Nature Geoscience, 4, 27–31, doi:10.1038/ngeo10272011, 2011.

Hyde, J. C., Smith, A. M. S., Ottmar, R. D., Alvarado, E. C., and Morgan, P.: The combustion of sound and rotten coarse woody debris: a review, Int. J. Wildland Fire, 20(2), 163-174, 2011.

Yokelson et al. (2012) http://www.atmos-chem-phys-discuss.net/12/21517/2012/acpd-12-21517-2012.html

Bertschi, I. T., Yokelson, R. J., Ward, D. E., Babbitt, R. E., Susott, R. A., Goode, J. G., and Hao, W. M.: Trace gas and particle emissions from fires in large diameter and belowground biomass fuels, J. Geophys. Res. 108, D13, 8472, doi:10.1029/2002JD002100, 2003.

Kauffman, J. B., Cummings, D. L. and Ward, D. E.: Fire in the Brazilian Amazon 2. Biomass, nutrient pools and losses in cattle pastures, Oecologia, 113, 415-427, 1998.

Rein G (2009) Smouldering combustion phenomena in science and technology, International Review of Chemical Engineering, 1, 3-18.

Title: Strictly speaking methanol and acetone and some of the other major species measured are not hydrocarbons and thus not NMHC. A more general term is non-methane organic compounds (NMOC) which can be specified as gas or condensed phase. I have also recently seen “NMOG” as a compact term. In addition, the paper is about global fire emissions of NMOC so the title should probably be changed to "The influence of biomass burning on the global distribution of selected non-methane organic compounds."

ACPD

12, C7333–C7339, 2012

Introduction: The introduction is smoothly written but should be revised to concisely reflect additional measurements in the boreal region, other global estimates, and the complication of poorly lofted smoldering emissions in the boreal region. Regarding the unlofted emissions, it's probably too late to change the model, but it's a limitation that should be recognized at the outset for context.

Comments on specific text by location.

P23438, L13: define FAAM

Hereafter the format is XX, YY: where XX is the last two page numbers and YY is the line number.

39, 24: "in"?

40, 13: "emission ratio" no "s"

40, 13-16: good discussion of these issues in Akagi et al., 2011

40, 15-25: Hobbs et al. (JGR, 2003) give nice plots of NMOC decays with time in African smoke plumes. In this work, the OH may have been lower. Akagi et al., (2012) show C2H4 decay in a BB plume at lower OH. Also the ratio plots may have slopes close to slope values measured in fresh smoke by Simpson et al. (2011) if the high mixing ratios, which tend to be measured in fresher plumes dominate the slope.

Akagi et al., (2012)http://www.atmos-chem-phys.net/12/1397/2012/acp-12-1397-2012.html

41, 9: The work compared to is also the same type of (airborne) platform in the same forest type, which could be an important factor in the degree of agreement observed.

41, 11-12: The authors can check their propene/CO also against papers by Nance et al and Radke et al tabulated in Table S2 of Akagi et al. (2011).

41, 17: Isoprene is also a combustion product as can be seen from the high isoprene

ACPD

12, C7333–C7339, 2012

emissions from peat measured by Christian et al. (2003).

41, 21 eliminate “the”

42, 22: It’s a bit of an exaggeration to call these four species anthropogenic tracers since their emission from fires has been well-known for a long time. Papers by Don Blake’s group identify halogenated hydrocarbons that are more rigorously restricted to anthropogenic sources. It is however valid that the control of these species will not be achieved through control of anthropogenic emissions alone so it is valuable to estimate the BB contribution.

43, 12: “factors” should be “ratios” - - - Also„ at this point in the text it is really clear that ER measurements in other global ecosystems may have been more relevant and it should at least be recognized that they exist (Akagi et al 2011) and used to help assess the uncertainty in the modeled impacts.

43, 20 & 25: The NCAR fire emission inventory and other inventories are compared to GFED-3 by Wiedinmyer et al. (2011) and this could help the authors assess the uncertainty in global BB-CO. The uncertainty in global BB-CO is large and is coupled with the uncertainty in BB-ER. This should be acknowledged for context and a more realistic error estimate.

Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, Geosci. Model Dev., 4, 625-641, doi:10.5194/gmd-4-625-2011, 2011.

44, 5-6: ??????

44, 11: It may be worth noting that high OH has been measured in tropical BB plumes by Hobbs et al. (JGR, 2003) and Yokelson et al. (ACP, 2009). There is also OH data available for boreal plumes in the ARCTAS archives.

45, 5-6: One wonders why the authors prove that the RETRO anthropogenic emissions

ACPD

12, C7333–C7339, 2012

of benzene are wrong (needs 2/3 reduction to agree) at one station and then use the unaltered RETRO emissions anyway? It could be briefly explained. Could the RETRO benzene data be evaluated at more sites relatively unaffected by BB?

45, 12: Using estimated year 2000 emissions in a model with 2009 meteorology and the adjusted emissions are labeled “2010” – maybe clarify.

45, 17-21: make more explicit? Again Sinha et al. (2003) used same WAS approach as Simpson et al. (2011) and measured benzene/CO for African fires and African fires dominate the authors model emissions. Christian et al. (2003) measured benzene/CO for peat fires, which could apply in SE Asia. Perhaps at least show the range of benzene /CO measurements in the refereed literature.

45, L19-20: is there a better way to say this?

46, 5-11: In the 1970s thru 1990s hundreds of papers were published that measured ER for BB and often then compared global anthropogenic and BB sources as noted above. Compare to a few of these estimates here?

46, 19: eliminate “and tightening”

47, 12: Clarify if different model years but all with 2009 meteorology?

47, 14: eliminate “these”

47, 15: eliminate “a proportion of”

47, 16 add “relative” before “impact”

48: capitalizing “Global” everywhere?

49, 10-11: The authors need to revisit this statement after conducting a realistic error analysis as outlined in comments above and that considers much other published high quality work.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 23433, 2012.