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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico" *by* T. J. Christian et al.

Anonymous Referee #2

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Review of Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico by Christian et al for Atmos. Chem. Phys. Discuss.

Gaseous and aerosol measurements were made for 5 little studied but important emission sources: wood cooking fires, garbage burning, brick and charcoal making kilns, and crop residue burning. By combining the emission observations per unit fuel burned with activity estimates, one gets an estimate of emissions from biomass burning which can then be compared to normal urban emissions. The conclusion is that in the whole of Mexico, cooking fires are the dominate source of PM2.5 and NMOCs and important

contributors to CO, NOx, CH4 and NH3. I have mulled this over at the end of this review (in conjunction with Table 7) and come to the conclusion that the very high PM2.5 emission rates reported here may be in conflict with HOA observations and would require that most of the OA attributed to ageing be reassigned as primary emissions. I do not dismiss the later possibility.

Based on measured CO to NOx ratios it would be difficult to quantify the effects of emissions from cooking fires.

Table 7, according to its title provides only the emissions from cooking fires and from urban sources. Fire emission estimates are very high. I would like to see what the other burning emission sources contribute. Also, I would like to know if the cooking fires are such that emitted particles make it into the ambient atmosphere.

I would like to see the authors follow the implications of their emission estimates on observable quantities as I have started to do in this review.

This paper should provoke a lot of thought and hopefully more measurements. Most everyone expected that Mexico City would be dominated by the usual variety of urban emissions but this may not be the case. Or pending further analysis it still might be. So it goes. I look forward to seeing this study published with minor revisions as the authors see fit.

Comments p 10102, line 5-6. It would help the reader to identify the biofuel estimates as global as is done on line 16 for garbage.

p 10103 line 2 thin margins. meaning?

p 10105, lines 12-13. Are these figures dried weight or carbon?

p 10107, lines 4-5. indoor air pollution is the largest factor causing mortality in children under five globally (Dherani et al., 2008) This is an astounding figure. I looked at Wikpidi and UNICEF publications for non-professionals. The single largest cause of mortality after the neonatal period is pneumonia (about 20%). Poor nutrition and sani-

tary conditions are mentioned as root causes. At this level of inquiry, I found no mention of indoor air pollution. I did read Dherani et al but I could not understand most of the article. In particular, I could not tell how influenza entered their considerations. No doubt, indoor air pollution is dangerous. For a variety of reasons, mostly preventable, childhood is a dangerous place.

p 10112, line 3-5. A 20-30% accuracy for determining PM2.5 from bscat is very optimistic. This is a respectable accuracy, if you knew the size distribution and composition.

p 10116, lines 5-8. The improved stove has a chimney which could scavenge reactive compounds. How open are the open cooking fires? Are they in buildings with 4 walls and a roof? If so, I would expect reactive species from the open fires to also be efficiently scavenged. I guess compounds like CO and C2H2 make it to the outside because there is nothing else that can happen to them.

p10117, lines 9-14. Is the formation of CH3CN in combustion sufficiently similar to HCN, that one can predict low concentrations in cooking fires?

Table 7. I recommend providing lines for the biofuel emissions from Mexico City and for the sum of urban and biofuel emissions. In so far as one can assess the accuracy of the emission estimates in this paper it will be done with MCMA data as that is where there is a very rich data set. I have determined Total MCMA emissions = Annual MCMA emissions + emission factor times 68 times 0.2. I have converted CO (MW=28) and NOx (MW=46) into moles. I have used the conversion factor 1ug/m3 CO = 0.8 ppb/m3 at STP (0C, 1 atm). Correct?

First, a minor point. To 2 significant digits, PM2.5 for National urban emissions (Tg) should be 0.025. The ratio National biofuel/national urban is correctly calculated without the round off.

The MCMA 2004 inventory yields a CO to NOx molar ratio of 16.3. Adding in biofuel emissions, the CO increases by 44% and the NOx by 15%. The ratio increases by 25%

to 20.4. The 2006 MCMA inventory (Fast et al., 2009) gives a ratio of 17.1. An increase of 25% to account for cooking fires yields 21.4. Measurements made in 2006 show a ratio of about 20 (this should be checked for the different aircraft and surface sites). It appears that the ratio CO/NOx does not vary enough to quantify the contribution of cooking emissions. Burning gives an MCMA increase of CO by 44%. If you really trust a model, you might be able to distinguish a burning from non-burning scenario by comparison with observations.

MCMA NMOC emissions without cooking fires are 0.53 Tg/year. With cooking fires, NMOC emissions increase by a factor of 2.38 to 1.26 Tg/y. I see that the NMOC emission factor is from a different study. Perhaps there is large perturbation due to biomass burning in a single compound or group of compounds that can be used in absolute value or in a ratio with CO to test the biomass emission rate.

The really big change is in PM2.5. MCMA burning is 0.092 Tg. Ordinary urban sources are 0.0066Tg, a factor of 13.9 less. Model calculations of HOA (primary emissions) by Fast et al (2009) show reasonable agreement with aircraft and surface measurements. If one were to multiply the HOA calculations by 13.9, one would get total OA that is much greater than observed total OA.

One can look at the ratio of PM2.5 to CO. Without a biomass source for PM2.5 or CO, this ratio is 4.6 ug/m3 (STP) per ppm CO. With a biomass source for PM2.5 and CO, this ratio is 47.5 ug/m3 (STP) per ppm CO. Ratios of 10 or less have been observed where primary emissions are expected to predominate. Ratios higher than 50 have been observed in downwind areas where SOA production is expected. It is possible that there are a range of primary emission rates, yielding low values for PM/CO in the city center and near roads and yielding high values outside the most urbanized areas. In that case, the increases in OA/CO that have been attributed to aging have been misidentified which is a possibility. Evidence to the contrary would be changes in the OOA to HOA ratio and in the O to C ratio attributed to ageing. Also, the work of Volkarmer et al (2006) is based solely on urban observations.

The emission estimates from cooking reported here yield a modern carbon content of more than 90% (neglecting fossil SOA), higher than the surprisingly high values observed by Marley et al (2008). As modern carbon in urban areas has been somewhat of a mystery, additional sources are of great interest.

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