**Atmospheric
Chemistry
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Discussions**

Interactive
Comment

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.

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(General comments were provided first)

Ref 1. 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.

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Auth1. We conclude that cooking fires produce more particles than urban sources on a national scale, but we did not discuss the possibility that open, landscape-scale fires may produce even more $PM_{2.5}$ than cooking fires, which we are addressing in a separate publication.

Ref 2. 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.

Auth 2. (i) Cooking fires may be a predominantly rural source and so we are not sure how much they contribute to MCMA emissions. This uncertainty is discussed more, later in this response. (ii) We suspect that other burning is likely the dominant national source of particles, but prefer to leave a careful analysis of that to a separate a paper in progress that also presents our emissions date for these fires.

Ref 3. Also, I would like to know if the cooking fires are such that emitted particles make it into the ambient atmosphere.

Auth 3. We address this later in our response where this question is raised again.

Ref 4. 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.

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Auth 4. We unfortunately did not include some important background information about cooking fires, which we will stress throughout the revised paper. That is that biofuel cooking fires are thought to occur mainly in rural areas where biofuel is readily available although some do occur in cities; perhaps mainly using transported charcoal (Bertschi et al). Natural gas may be relatively more important as a fuel in urban areas. Therefore, most of the quantification of biofuel cooking fires is based on surveys of the rural population and there are not good estimates of how much occurs specifically in urban regions. Nor is there a good tracer for cooking fires as we stress throughout the text. Thus, unfortunately, we cannot make a rigorous estimate of the potential impact of urban cooking fires and we would have to employ a regional model to assess the probable influence of rural regional fires on Mexico City (i.e. to account for transport from outlying areas around the city). To summarize: conventional wisdom suggests that the cooking fires may not contribute a lot to primary emissions in Mexico City, but that cannot be rigorously confirmed or disproved at this time.

(Specific comments were provided next)

Ref 5. 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.

Auth 5. In line 5 we changed "Biofuel" to "Global biofuel." We also added the word "globally" after the word "generated" in line 15 to indicate that that was a global estimate also.

Ref 6. p 10103 line 2 thin margins. meaning?

Auth 6. We changed "thin" to "very small profit"

Ref 7. p 10105, lines 12-13. Are these figures dried weight or carbon?

Auth 7. After the first fuel consumption estimate in paper (P10104, L14) we added "dry matter (dm)" and after the other values we added "dm"

Ref 8. 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 sanitary 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.

Auth 8. We oversimplified this statement. Indoor air pollution (IAP) is considered the largest single factor contributing to the largest single cause of child mortality. Specifically, pneumonia is the largest single cause as the Referee confirmed with some estimates of the percentage being higher than 20%. Metadata analyses of the causes of pneumonia attribute about 40% of pneumonia to IAP according to Dr. Kirk Smith at the Berkeley School of Public Health who is considered the leading authority on these matters. We also note that countless epidemiological studies have empirically found a 1-2% increase in mortality for every 10 ug/m^3 increase in ambient PM. With that in mind, and the fact that PM levels in homes with active cooking fires typically exceed 1000 ug/m^3, the link between IAP and mortality becomes apparent. We have rephrased the text in question and added the reference that derives the 40% estimate mentioned above.

Original:

"The chimney provides an approximate 70% reduction in indoor air pollution (Zuk et al., 2007), which is the largest single factor causing mortality in children under five globally (Dherani et al., 2008)."

Revised:

"The chimney provides an approximate 70% reduction in indoor air pollution (Zuk et al., 2007). Indoor air pollution is believed to be one of the major risk factors for pneumonia,

which is the largest single cause of mortality globally in children under five (Smith et al., 2004; Dherani et al., 2008)."

Smith, K. R., Mehta, S., and Maeusezahl-Feuz, M.: Indoor smoke from household solid fuels, in: Comparative Quantification of Health Risks: Global and Regional Burden of Disease due to Selected Major Risk Factors, edited by: Ezzati, M., Rodgers, A. D., Lopez, A. D., and Murray, C. J. L., World Health Organization, Geneva, Vol 2 pp. 1435-1493, 2004.

Ref 9. 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.

Auth 9. We base our accuracy estimate on what we term "gravimetric calibrations of" nephelometers. Specifically, we collect filters while simultaneously measuring b$_{scat}$ and then plot the two quantities versus each other for a range of particle mass concentrations. The slope is taken as the empirical conversion factor. Typically, for a given fuel type, for us and others, the 2σ uncertainty in the slope is about 5-7% of the slope (Yokelson et al., 2009; Nance et al., 1993). For the cooking fires, we instead found two studies by other groups that carried out gravimetric calibrations for light scattering using the same model nephelometer as us and for cooking fires in Mexico or Honduras. One of those studies also reported an EC/OC ratio, which was similar to ours (as noted). We used the average of the two conversion factors and estimated the uncertainty (in equation 1) as the range between the two conversion factors. In this case, the uncertainty is 13.6% of the conversion factor. Thus, we thought 20-30% uncertainty was a reasonable estimate, but will change it to 20-40%.

Nance, J. D., Hobbs, P. V., Radke, L. F., and Ward, D. E.: Airborne measurements of gases and particles from an Alaskan wildfire, J. Geophys. Res., 98, 14873-14882, 1993.

Ref 10. 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.

Auth 10. The focus of cooking fire research has been on indoor measurements because of the huge public health impacts noted above. There has not been a targeted measurement of scavenging efficiency that we know of though some must occur. We think metal chimneys might scavenge more effectively than wood or cement but we don't know. It's also relevant that the homes are often well ventilated (open doors and windows etc). The smoke can therefore escape without being forced through a narrow metal duct where turbulence may promote wall contact. Also indicative of the good ventilation is the observation that the smoke in these studies typically drops to background levels within minutes to about one-half hour after the cooking stops. A worthwhile component of future research would be to measure the ratio of reactive to stable species both indoors and outdoors in homes without chimneys. To acknowledge the uncertainty we will add a condensed version of the above discussion.

Ref 11. p10117, lines 9-14. Is the formation of CH3CN in combustion sufficiently similar to HCN, that one can predict low concentrations in cooking fires?

Auth 11. We suspect that both compounds could arise from pyrolysis of proteins, but do not know if this has been proven as the main source. We do know that the ratio of CH_3CN/HCN is about 0.3-0.5 and does not seem to vary greatly between a number of biomass burning studies (Yokelson et al 2009), which gives us an empirical basis for our conjecture.

Ref 12. 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?

Auth 12. We would like to add the requested table entries, but are not sure how we would generate those estimates. After carefully considering the above comment and the additional comments below, we believe that the Referee is assuming that since Mexico City has 20% of the national population that it would produce approximately 20% of the cooking fire emissions. Based on that assumption, quite a few interesting projections are made. We appreciate the Referees interest, effort and insight, and regret that our paper did not include the information that cooking fires are thought to be less common in urban areas. If the latter assumption is right, then we lack a strong basis for projections. Nonetheless, the Referee's analysis demonstrates that a survey of biofuel use in urban areas of Mexico would be of great value.

Ref 13. 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.

Auth 13. Thanks, we fixed that.

Ref 14. 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.

Auth 14. (i) We used an emission factor from our study in Africa only for NO_x since it is an important emission that we were unable to measure in the Mexican cooking fires. (ii) As explained earlier, we suspect that the calculations above may be based on assuming too large a contribution of urban cooking fires, but can't confirm that. (iii) The Referee mentions the possibility that there is a compound or group of compounds that could serve as a marker or tracer for biomass burning. While the suite of gas-phase NMOC and particle constituents does differ between cooking fires and urban emis-

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sions, the cooking fire species are also emitted by open biomass burning, industrial biofuel use, and garbage burning. For example burning paper emits levoglucosan. As another example, HCN which is emitted by open biomass burning appears to be emitted in much smaller amounts per unit fuel consumption in cooking fires. Thus, we have yet to identify a species (or a suite of species) that can be used to isolate the contribution from cooking fires or the contribution from cooking fires plus open large-scale fires. Crounse et al also discuss this in the context of measurements downwind of Mexico City. In summary, the Referee's exploratory calculations, our Figure 6, and the modern carbon issues discussed both in the paper and by the Referee offer intriguing hints that biofuel cooking fires may be more common in urban areas that has been conventionally assumed. Thus, a survey exploring the extent of biofuel use in urban areas would be valuable as would future research targeted at identifying tracers. In addition we have added a reference that suggests that food waste may be more prevalent in Mexican dumps than we thought. This would not affect the percent carbon and EF calculations, but does strengthen the case for garbage burning contributing modern carbon.

Crounse, J. D., DeCarlo, P. F., Blake, D. R., Emmons, L. K., Campos, T. L., Apel, E. C., Clarke, A. D., Weinheimer, A. J., McCabe, D. C., Yokelson, R. J., Jimenez, J. L., and Wennberg, P. O.: Biomass burning and urban air pollution over the Central Mexican Plateau, Atmos. Chem. Phys., 9, 4929-4944, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 10101, 2009.