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Atmospheric Chemistry and Physics Discussions

ACPD

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Interactive Comment

Interactive comment on "Trace gas and particle emissions from open biomass burning in Mexico" *by* R. J. Yokelson et al.

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The authors thank both Referees for excellent comments that will strengthen the paper. Our responses are inserted as point by point replies to the comments below.

Anonymous Referee #1

This manuscript presents previously unpublished data for several types of open burning made during the 2006 airborne campaign in Mexico and completes the presentation of the emission factor (EF) measurements made during the MILAGRO campaign. The study improves the existing knowledge of the biomass burning emissions in the Northern tropics, by providing EFs integrated by various fire types relevant for Mexico. This work also provides some interesting insights into factors other than vegetation types that may control EFs of NOx and possibly other species. By combining EFs with estimates of fuel consumption, the authors estimate the typical annual emissions from biomass burning, garbage burning and biofuel use in Mexico. They find that these sources combined can be comparable to or exceed emissions from fossil fuel combustion for a number of species, thus having a great impact on the atmospheric composition in the tropics. This is a valuable study, well researched and presented in a clear manner. The covered material is relevant to Atmospheric Chemistry and Physics and can be of great interest to its readers. I certainly recommend this manuscript for publication. A few comments and questions are listed below.

R1.1. Page 7331, Lines 13 - Page 7332, Line 15: These two paragraphs describe the derivation of average EFs relevant for all types of fires, not just crop residue burning, therefore I would suggest moving them to the "Experimental details" section.

A1.1. We liked this idea and explored moving the text to section 2.2; partly because of its scope and also because a reader skipping the crop residue section might overlook the text (although the same reader might also skip section 2.2). However, if this text is moved to section 2.2, it has a few undesired "side effects." For instance, Table 2 would come before Table 1 leaving the tables in sort of odd order after renumbering - with a fuel type, followed by all the fire locations, and then back to fuel types. Also, this is a novel approach to getting average EF so it's probably best to develop it as a case-study/discussion-item rather than being packaged as a routine procedure. Thus, we propose another slightly different option that may be more workable. We can rename section 3.1.1 "Crop residue fires and description of a new method for deriving average emission factors." We can also add a brief new section before section 3.1.1 that highlights two general issues regarding emission factors in this study (see also next Referee comment). Finally, to make the general nature of the procedure more clear, on P7332, L13-14, we can change "In summary, we recommend using the EF computed at the average MCE . . ." to "In general, we recommend using the EF computed at the average MCE for each vegetation fire type . . ."

R1.2. Page 7332: The last paragraph in Section 3.1.1 seems to be out of place here. I suggest moving it to the "Experimental details" section.

A1.2. This is also an idea that we liked and planned to implement that turned out to be tricky. Originally there didn't appear to be an ideal spot to put this discussion, so we put it at the point in the text where it first becomes relevant. That was not ideal, but it is also true that some of the "black carbon" methods we discuss in paper are from other work, and not part of our experimental procedure. Thus, in the spirit of the Referee's suggestion, it seems to make sense to add an introductory sentence and insert this paragraph in the new text (proposed above) between the section 3.1 section 3.1.1 headers (i.e. Page 7330, between lines 17&18). Then we can conclude this paragraph with a new sentence to highlight the derivation of a new averaging method in section 3.1.1 Approximate proposed new text to insert on page 7330 after line 17: "We begin this section by noting two issues that affect the discussion of the emissions from all the fire types addressed in this study (1) the terminology for light-absorbing carbon, and (2) the calculation of average emission factors for a fire type. . . (move/insert last paragraph in section 3.1.1 that discusses light-absorbing carbon here). . . In this paper we also use a new procedure for calculating the average emission factors for each fire type. The rationale for this new method and the details of its implementation are developed in case study format in the next section."

R1.3. Page 7335, lines 14-16: Please clarify the possible link between the surface windspeed and emission factors of NOx. (Is it because windspeed affects the type of combustion?)

A1.3. The possible link between windspeed and NOx emissions is speculation at this point and so far we can also only speculate about a mechanism for this. One possibility is that wind could promote or enable flaming combustion of vegetation elements that are known to have high levels of N, but may be too moist too burn in the absence of wind. This could include freshly-growing foliage that is likely to be present in the springtime. We will mention these possibilities, clearly identified as speculation, in the

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revised text.

R1.4. Page 7365, Table 7: Please correct syntax in the footnote to the Table 7, e.g., "In this work PM2.5 was estimated.."

A1.4. Thank you, we changed: "1This work PM2.5 estimated from 1.2 times the sum of species measured on Quartz filters (Christian et al., 2010)." to "1PM2.5 for the 2007 fires in Mexico was estimated as 1.2 times the sum of species measured on quartz filters as described by (Christian et al., 2010)."

R1.5. Page 7366, Table 8: EF for NMOC for biofuel and the resulting NMOC emissions in Table 7 of Christian et al. 2010 are 12 times higher than the numbers in Table 8 of this work (e.g., EF of 54 vs. 4.34 g/kg). Why is this difference?

A1.5. Due to overlap in the author list we are familiar with the procedure used in Christian et al. We ultimately agree with their work, but the approach taken in this paper is more detailed and developed in step-wise fashion. In Christian et al., (2010), the goal was to quickly compare ("developing tropical country") probable total emissions from just cooking fires to probable total fossil fuel (FF) emissions for two case studies: Zambia (mostly rural) and Mexico (mostly urban). In the less detailed approach taken in Christian et al., (2010), Table 7 contained the final most likely value for EFNMOC for cooking fires, which was derived as follows. Bertschi et al., (2003) measured 27 g/kg of total NMOC emitted by cooking fires in Zambia using FTIR alone (a high-sensitivity apparatus), which was doubled to account for unmeasured species as described in Yokelson et al., (2008) to arrive at 54 g/kg. This number was then recycled in Christian et al., (2010). In the current study Table 8 showed the most conservative value for EFNMOC for cooking fires (4.34 g/kg from a less sensitive FTIR system, which was deployed on more efficient fires (in Mexico)) as just one component of the total non-FF contribution. However, our current text contains a discussion (page 7345, lines 10-14) that explains why the EFNMOCs for all the fire types in Table 8 are likely far too low and probably closer to 60 g/kg for all the fire types. In fact, the main point, that non-fossil fuel sources have great importance is well served with either the low or high estimate and both estimates have high uncertainty as indicated in the text.

The Referee's comment did bring two more issues to our attention. (1) The text as written failed to clearly specify the origin of all the biofuel EF so we have changed P7344, L17-24 from:

"In Table 8, for each type of open burning sampled in this study, we have coupled the FINNv1 national fuel consumption estimates with our field-measured EF to estimate the total national emissions for several key species. We also show the emissions from national biofuel use and the national urban emissions; both taken from Table 7 of Christian et al. (2010). The latter were based mostly on the MILAGRO 2007 measurements and by scaling the 2004 MCMA emissions inventory (http://www.sma.df.gob.mx/sma/index.php?opcion=26\&id=392) to the total urban population of Mexico (∼75 million) as described in detail therein."

To:

"In Table 8, for each type of open burning sampled in this study, we have coupled the FINNv1 national fuel consumption estimates with our field-measured EFs to estimate the total national emissions from various types of open burning for several key species. We also show the national biofuel consumption and the national urban emissions taken from Table 7 of Christian et al. (2010). We coupled the national biofuel consumption with Mexican cooking fire EFs taken from Table 3 of Christian et al., (2010) and NOx EF for cooking fires taken from Table 1 of Bertschi et al., (2003). The national urban emissions were derived by scaling the 2004 MCMA emissions inventory (http://www.sma.df.gob.mx/sma/index.php?opcion=26\&id=392) to the total urban population of Mexico (∼75 million) as described in detail by Christian et al., (2010)." (2) In addition, there was a typo in Table 8. "4.34" should have been "4.64." We double checked the other Table 8 entries and they are fine. So we will correct Table 8 and make the appropriate small changes throughout the text.

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Bertschi, I. T., Yokelson, R. J., Ward, D. E., Christian, T. J., and Hao, W. M.: Trace gas emissions from the production and use of domestic biofuels in Zambia measured by open-path Fourier transform infrared spectroscopy, J. Geophys. Res., 108, D13, 8469, doi:10.1029/2002JD002158, 2003.

Anonymous Referee #2

The authors present a new compilation of emission factors for the most important fire types occurring in Mexico. It includes a large number of airborne, in terms of sampling and measurement techniques largely consistent measurements, a large fraction of which is first published in this work. The manuscript is very well structured and written. The results are presented and discussed in a very concise manner. This work is a very valuable contribution to an improved understanding of factors influencing emission production (e.g. the relation between emission factors and MCE) and to more accurate emission estimates. I therefore recommend this manuscript for publication in ACP. I only have minor comments:

General comments:

R2.1. In section 3.1.3, emission factors for mixed crop residue and tropical dry forest fires are presented. Emission factors of this category are not used in the estimate of primary emissions from open burning in Table 8. What is the recommended usage of this category?

A2.1. The EF for the mixed fire types may not have an immediate conventional application. They were presented separately to document the fact that these fires were different and to help point out the possibility of other driving factors influencing EF such as windspeed. It is also of general interest that the emissions from a mixed fuel type were not a linear combination of the base fuel types as might be assumed.

R2.2. In section 3.2 the dependency between MCE and the contribution of soot (and OA) particles to the total number of biomass burning particles in individual samples is discussed. The dependency in Figure 6 is visible; however, only 35 and 39% of the variations in the contribution can be explained by variations in the MCE. Possibly, it would be helpful to add a sentence that the contribution of soot and OA particles is apparently also determined by other influencing factors.

A2.2. The scatter in the plots is expected for a number of reasons. The soot production (essentially BC/CO2) for turbulent diffusion flames (the type of flames in BB) has long been known to be extremely variable (e.g. factor of eight in: Shaddix et al., 1994; review of topic in: Kennedy, 1997) with the degree of turbulence being a major controlling factor for soot yield. In similar fashion, OA/CO also varies over a large range (> 4 in de Gouw and Jimenez, 2009). We will add concise text to acknowledge this.

Shaddix, C. R., Harrington, J. E., and Smyth, K. C.: Quantitative measurements of enhanced soot production in a flickering methane/air diffusion flame, Combustion and Flame, 99, 723-732, 1994.

Kennedy, I. M.: Models of soot formation and oxidation, Prog. Energy Combust. Sci., 23, 95-132, 1997.

de Gouw, J. and Jimenez, J. L.: Organic aerosols in the Earth's atmosphere, Environ. Sci. Technol., 43, 7614–7618, doi:10.1021/Es9006004, 2009.

R2.3. In Section 3.3 you estimate total biomass burning emissions for Mexico using biomass burnt estimates from the FINN inventory. Adding the corresponding GFED inventory estimates (van der Werf et al., 2010) for comparison would possibly be of great interest to many readers.

A2.3. This comparison is of interest, but not totally straightforward at the level of detail we present in section 3.3. From FINNv1 we have, specifically, the total for Mexico for open biomass burning broken out by vegetation/fuel type. GFED3 is produced with 0.5 degree resolution and then derives open biomass burning totals by region, by fuel type globally, or by country summed over all fuel types. The link for the GFED3 country level

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totals is: http://www.falw.vu/∼gwerf/GFED/GFED3/tables/countries/C_absolute.txt Regarding uncertainties, we note that the GFED3 country totals are accompanied by a disclaimer: "These country-level estimates are for indicative use only, they are not suitable for official reporting due to large uncertainties and potential for missing key regional aspects in the global approach used." The FINNv1 also uses a global approach and the uncertainty is estimated at a factor of two. GFED3 assumes the biomass is 45% carbon and their total carbon values imply that 33 Tg biomass burned in open biomass fires in Mexico in both 2006 and 2003. 2003 had almost twice as many hotspots as 2006. This shows that the total biomass consumption is sensitive to the assumed vegetation type and assumed fuel loadings, which we don't have access to broken out at the national level for the GFED3 product. The sum of the biomass consumption for all the open burning types in Mexico (excluding garbage burning) in FINNv1 was 96.3 Tg - so the GFED3 total is 35% of the FINNv1 total. The FINNv1 estimate may be larger partly because it is based on active fire detection, which may "see" the small fires that are typical of Mexico more efficiently than burned area detection, which drives GFED3. Neither FINNv1 nor GFED3 include fuel consumption by cooking fires or garbage burning. Using GFED3 for the open vegetation burning contribution to total BB instead of FINN v1 brings the total for BB+GB to 102.4 Tg instead of 175 Tg or about a 40% reduction. We have elected to note this discrepancy, but not discuss it at length in the revised paper or add a column with a single entry to Table 8 for several reasons. (1) The estimates are very useful and important, but difficult and understood to have high uncertainty, which is discussed at length in the FINNv1 paper and which we have already strongly acknowledged in this paper. (2) Both products underestimate crop residue fires, which could be a major component of the total open burning based on our visual observations from the aircraft. (3) Several recent studies suggest that many current emissions inventories underestimate the emissions from biomass burning in the tropics (e.g. Kopacz et al. 2010; Liu et al., 2011) or boreal region (Yurganov et al., 2011). Our main point; that all forms of biomass burning summed with garbage burning is comparable to FF doesn't change. For instance, the

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primary fine PM emissions from BB+GB would drop from 47 times FF to 28 times FF.

We propose to add the following text in section 3.3 at P7342 L5 or in section 3.5 at P7345 L2 or L20: "There is high uncertainty in the open burning fuel consumption estimates and considerable variation between available estimates as discussed in detail by Wiedinmyer et al., (2010). For instance, if we use GFED3 (van der Werf et al., 2010) instead of FINNv1 to estimate the fuel consumption by open BB then the total fuel consumption by biomass burning plus garbage burning would go down by about 40%. However, our basic conclusions about the major importance of BB+GB do not depend on choosing a specific inventory. Thus more BB measurements and more work on BB inventories are needed."

Kopacz, M., Jacob, D. J., Fisher, J. A., Logan, J. A., Zhang, L., Megretskaia, I. A., Yantosca, R. M., Singh, K., Henze, D. K., Burrows, J. P., Buchwitz, M., Khlystova, I., McMillan, W. W., Gille, J. C., Edwards, D. P., Eldering, A., Thouret, V., and Nedelec, P.: Global estimates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES), Atmos. Chem. Phys., 10, 855-876, 2010.

Liu, C., Beirle, S., Butler, T., Liu, J., Hoor, P., Jöckel, P., Penning de Vries, M., Pozzer, A., Frankenberg, C., Lawrence, M. G., Lelieveld, J., Platt, U., and Wagner, T.: Application of SCIAMACHY and MOPITT CO total column measurements to evaluate model results over biomass burning regions and Eastern China, Atmos. Chem. Phys. Discuss., 11, 1265-1331, doi:10.5194/acpd-11-1265-2011, 2011.

Yurganov, L., Rakitin, V., Dzhola, A., August, T., Fokeeva, E., Gorchakov, G., Grechko, E., Hannon, S., Karpov, A., Ott, L., Semutnikova, E., Shumsky, R., and Strow, L.: Satellite- and ground-based CO total column observations over 2010 Russian fires: accuracy of top-down estimates based on thermal IR satellite data., Atmos. Chem. Phys. Discuss., 11, 12207-12250, doi:10.5194/acpd-11-12207-2011, 2011.

Referee #2 technical comments:

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R2.4. I sometimes encountered a semicolon in sentences where I doubt it is correct, e.g. page 7343 line 13, page 7344 line 6, page 7345 lines 11 and 17. Please check if a comma is more appropriate in these cases.

A2.4. 7343/13: the semicolon sets off a clause that contains a comma. 7344/6: changed the semicolon to a comma. 7345/11: changed the semicolon to a comma. 7345/17: the semicolon sets off a clause that contains a comma, but it seemed OK to change the semicolon to a comma anyway.

R2.5. Page 7341 line 4: Table 8 includes values from Tables 2,3,5,6,7.

A2.5. Good point. We did not consider the fuel consumption in mixed-vegetation fire types (Table 4) in Table 8. We also derive the garbage burning fuel consumption estimate in Table 8 at a point further into the text (GB EF appear in Table 7) so we changed "Tables 2-6" to "Tables 2, 3, 5, and 6"

R2.6. Tables 2 to 7: Would it be possible to add a label why values are not listed (e.g. measured but below the detection limit, not measured)?

A2.6. We will add "nm" for "not measured" and "bdl" for "below detection limit." Some of the bdl occurred because the fire was small and located in mountainous terrain where we could only access the very dilute plume.

R2.7. Figure 3: Not using abbreviations for Africa and Mexico in the legend would probably look nicer.

A2.7. Done, thanks.

R2.8. Figure 4: You may consider exchanging "high MCE point" with "high MCE points".

A2.8. Thanks, we changed "high MCE point" to "the highest MCE point" since that is the single airborne point.

Additional voluntary changes

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Update Adachi and Buseck, Akagi, and other references and add references cited in response when used in revised text.

P7332, L24 (part of the text to be moved): change “mass” to “amount” since we report number fraction and not mass for soot.

P7332, L27 (part of the text to be moved): to acknowledge the increasing use of SP2-based BC measurements, add: “or calibrated incandescence measurements (Moteki and Kondo, 2007)” after “efficiency”

Moteki, N., and Kondo, Y.: Effects of mixing state of black carbon measurements by laser-induced incandescence, Aerosol Sci. Tech., 41, 4, 398-417, 2007.

P7347, L11&L12: change “accounts” to “accounted”.

P7348, L15: Add, “We thank Holly Eissinger for production of Figure 1 and Supplement Figure 1.”

Fig 2 caption: change to “The solid black circles show all 6 EF measurements on crop residue fires for 1,3-butadiene and the open red square shows the average of those measurements (0.114 g kg-1). The filled red square is the EF for 1,3-butadiene (0.151 g kg-1) computed from the fit at the average MCE for all 14 crop residue fires sampled in Mexico. The value from the fit is 32% larger, more consistent with the average MCE, and likely better reflects the average EF (see text).”

Fig 5: We will reduce the line thickness on the axes to be more consistent with other figures.

Fig 6: We will match the plot sizes better.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 7321, 2011.

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