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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Modeling the impacts of biomass burning on air quality in and around Mexico City" *by* W. Lei et al.

Anonymous Referee #2

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Review of Lei et al

The impact of biomass burning (BB) on tropical megacities is an important topic that is treated by a dedicated regional model in this study. This paper has a thorough introduction that reviews relevant past work. The description of the methodology confirms a strong focus on accuracy in the model. The model has useful applications including: (1) the year-round impact of BB on the Mexico City region, or annual trends, can be assessed for periods when no measurements are available - even if the model has limitations in assessing how much BB occurs. (2) the model can assess the representativeness of various monitoring stations. (3) the model can roughly estimate specific impacts from processes that cannot be directly measured. A few of the model limitations could be described a bit more clearly and closer to the beginning of the paper. For that reason my review is repetitious as it points out where a lack of clarification is an issue in multiple locations. But the clarification is only needed once. Some language issues occur here and there as noted in my specific comments below. Overall a strong paper.

Some general thoughts

Fires inject smoke at a range of altitudes. Often the same fire injects smoke low during ignition, near the top of the boundary layer or above the boundary layer during maximum intensity, and then at lower elevations again during smoldering. We also know that the majority of the fires in the tropics are small and do not get detected as hot spots or burn scars. Further, the T1 and T0 sites are excellent for monitoring the BB impact on Mexico City and environs, but not ideal for constraining total BB in the region since their valley floor location does not provide maximum sensitivity to the BB emissions, which mainly originate high above the city in the surrounding mountains. It seems that the same impact could occur at e.g. T0 if less smoke is injected in lower layers or more smoke is emitted in higher layers – although this may have been tested to an extent that is not clarified and depends on how fast vertical mixing is. In general, fires are a difficult problem for any model and it's important that aircraft measurements in the outflow offer an alternative perspective on how much BB occurs in the region that compelments the bottom-up inventory. Two different aircraft studies suggested that biomass burned accounted for a much higher percentage of the total regional CO source than indicated in the authors Table 2 (25-31% by aircraft/tracers and 10% in the Table 2 bottom-up inventory). The authors note the high uncertainty in both their BB inventory and the anthropogenic inventory in the text and the aircraft measurements also have significant uncertainty. Thus the values in Table 2 should come with uncertainties, or be flagged to be consistent with the discussion in the text. More generally, the uncertainties discussed above are relevant at several locations throughout the paper. Addressing these widely-relevant issues at one point early on in the paper would improve the flow of the paper.

ACPD

12, C7084–C7093, 2012

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Comments referred to specific locations in text:

P22894, L11 is "lay" better than "lied"

P22895, L4 "for about"

P22896, L22 should "will be" be "are"?

P22896, L25 should "will" be eliminated?

P22896, L29-P22897, L1 change "and the impacts" to "the impacts" and replace "as well as" with "and also"

P22898, L2-15: a bit tedious, but technically supposed to define acronyms at first use: e.g. QNSE-YSU and MYJ

P22899, L3-4: I think it's more useful if "(strictly speaking a portion of biomass burning emissions comes from human activities)" is replaced with "(note that in some studies biofuel use is included in anthropogenic emissions)" - - - Even though nearly all fires in the tropics are human-initiated only the biofuel use is sometimes lumped together with anthropogenic emissions to my knowledge (see e.g. Bond et al., 2004)

P22900, L4: The emission factors measured by Yokelson et al. (2007) cover only about one half of the actual emitted NMOC as discussed there-in and elsewhere (Yokelson et al 2009; 2012 (http://www.atmos-chem-phys-discuss.net/12/21517/2012/acpd-12-21517-2012.html)) and later in this paper. In other words there were unmeasured and unmeasureable species not included in the MILAGRO BB-EF. This is a bit related to the fact that the FF emissions were augmented by 7.5 times the POA to account for unspecified IVOCs and SVOCs. On P22900 L25-29 the authors state that the BB SVOC were scaled similar to the FF SVOC, but that no BB-IVOC were added. It's not clear why no BB-IVOC were added. It's also not clear how much the scaling counteracted the error due to unmeasured BB-VOC. It appears that the final BB-VOC may be underestimated since in Table 2 the VOC/CO ratio for FF is about one-third, but the VOC/CO ratio for BB is only about 10% when it should probably be closer to one-third

ACPD

12, C7084–C7093, 2012

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(Akagi et al., 2011; Yokelson et al., 2012). I think it's too late to re-run the model with more BB-VOC, but in an early discussion of uncertainties it may be good to clarify that the BB contribution to VOC precursors is probably too low. This is relevant later in the paper and the BB impact on O3 is probably higher than estimated here – although maybe still less than 10% and not a major issue in the region.

P22900, L16-17: the alternative diurnal profile for fire activity actually has a take-off point that agrees well with what Yokelson et al (2007) observed from their aircraft in Mexico. I'm not sure what is meant by "Note that the start time in the diurnal profiles for individual fires may shift sooner or later."

P22900, L21: change "where" to "when"

P22900, L25-29: For both BB and FF I am curious how the total NMOC changes once the POA evaporation is added in. Also, is this a fast step in the model after emission or are the evaporated VOC included in the VOC shown in Table 2?

P22901, L17: Table 2 is introduced here and the total emissions shown in Table 2 are of interest as noted above. The ratio of BBCO/FFCO is about 10% based on inventories that the authors carefully developed for this study. In contrast, aircraft measurements in the outflow using tracers provide a check on inventories. The aircraft measurements in MILAGRO have come up with higher values for the contribution of BB to total CO: e.g. 31+/-3% in Crounse et al. (2009) for nearly the same domain or ∼25% Yokelson et al. (2007) based on flights in the MCMA outflow for all of March 2006. A quote from the Crounse et al abstract is relevant: "We find that during the period of our measurements, fires contribute more than half of the organic aerosol mass and submicron aerosol scattering, and one third of the enhancement in benzene, reactive nitrogen, and carbon monoxide in the outflow from the plateau. The combination of biomass burning and anthropogenic emissions will affect ozone chemistry in the MC outflow." So this seems potentially inconsistent with BB-CO being 10% and BBVOC and BBNOX being only ∼3% of the total in the inventories? The BB O3 impact will depend on the scaling of

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12, C7084–C7093, 2012

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the BB source to some extent. This raises a few interesting questions: (1) is this a discrepancy between studies, a possible under-scaling of BB due to missed fires? (2) or is this a function of transport and are all these studies actually consistent within the stated uncertainty (e.g. factor of two in the bottom up BB emissions)? (3) if BB was increased in the model would the agreement with measurements get worse or could any additional biomass burning have been directed above or away from the ground-based measurement sites used here, but detected by the aircraft? (4) in general terms - is the uncertainty in the transport and in measuring BBOA in a complex environment at a limited number of sites low enough to dispute the airborne measurements? (5) would future model runs with BB emissions and the BB injection heights scaled upwards effect O3 and be of value? It's a very challenging problem and it's not clear to me how much a model/measurement comparison can constrain the BB source strength, but again the topic could be raised and treated concisely at some point early in the text.

One other comment on Table 2: Does the VOC column include IVOC and SVOC and if so, would it better to call these compounds "gas-phase NMOC" or "NMOG" throughout the text?

P22902, L8-11: The national total of biomass burned estimated by Yokelson et al seems irrelevant at this point and can be eliminated or replaced by estimates of national PM emissions from BB and FF.

P22904, L16: change "with" to "within"

P22904. L17-18: I can see the justification for shifting the diurnal profile temporally, but not sure why that would be coupled with releasing the emissions at a lower altitude except at night. It seems more physically appealing that the injection altitudes might distributed over all the model layers in the boundary layer for all fires during the day.

P22904, L25-28: I find it surprising that injection altitude would not matter beyond the sensitivity noted for the first 2-3 layers unless 3 layers covers the whole boundary layer or vertical mixing is really fast within the boundary layer. Perhaps the range of injection

ACPD

12, C7084–C7093, 2012

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altitudes tested was insufficient to see an effect if the top of the boundary layer was considered? If there is no sensitivity above the first three layers it might be explained if the fires were far away and there was good vertical mixing and wind direction did not vary with altitude. However, wind direction often varies strongly with altitude and other studies in other environments have found that injection altitude was important.

P22905, L1: Figure 6 comments: Shifting the onset of the emissions forward a few hours and injecting them in the lowest layer instead of layers 2 and 3 makes a dramatic difference and a big improvement – suggesting that injection altitude can matter. However, the two main BBOA peaks are still not accounted for in the model. The BBPOA measurement integrated over the 17th thru the 22cnd time period appears to be about twice the integrated BBPOA in the simulation. This seems to be consistent with the possibility that not all the actual BB emissions are represented in the model. This can happen easily since most of the fires in the tropics do not get detected as hotspots as noted in Table 1 of Yokelson et al., 2011. If there was a large amount of additional BB emissions not captured in the model it might bring the total BB-CO closer to the fraction of FF emissions that was suggested by the aircraft tracer experiments discussed above.

P22905, L24 and P22906, L19: It's probably fair to eliminate "small" in first instance as even large fires can be missed due to clouds, timing, lack of coverage, etc. The missed peaks at T0 in Figure 6 are pretty substantial and could be from a small fire that is very close or a much larger fire that is further away, but "missed" T1. This is a coupled problem, because the measurements indicate a fire hit T0 and missed T1, but the model has a high probability of transporting air from T0 to T1. Perhaps the transport is too fast in the model and the real air parcel experienced substantial dilution. As the authors conclude, a small fire very close to T0 seems most likely as it could dilute enough to have minimal impact downwind. Nevertheless, in the more general case even larger fires are sometimes missed by active fire detection.

P22906, L4: The word smoldering is included here and the data to support the idea of

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12, C7084–C7093, 2012

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a smoldering fire appears later in Fig 9 as a lack of EC enhancement (OK I guess).

P22907, L6: Not sure "at night" is needed.

P22907, L18-25: The overestimate of SOA at T0 on March 12-13 seems much larger than 10% and so maybe adding OH consumption to the model would not do the trick. It seems the POA is well simulated during this period, but not the SOA, especially after adding BB. Thus maybe the longer range transport from a more distant large fire that actually missed T0 is incorrectly impacting T0 with aged smoke in the model? As the vertical profile of BB-CO in Crounse et al shows, T0 is not really the idealized location to study fires from. Thus the beginning statement about "meteorological" conditions is probably right, but a bit vague.

P22908, L2: Fig 9 now shows that when the BBOA was dramatically underestimated at T0, the EC was not and that supports, in hindsight, the idea of a small smoldering fire near T0 introduced at P22906, L4.

P22908, L11: garbage burning possible EC contributor in addition to brick kilns?

P22909, L6-11: Change: "The BB VOC emissions are usually estimated by projected biomass burned and the emissions factors for VOC species detected by available instrumentation; however, only about 50% of VOCs are identified on the mass basis (Yokelson et al., 2008; Akagi et al., 2011), most of those unidentified are high mass reactive compounds, and not all identified species have measured EFs." To "The BB VOC emissions were generated by multiplying the estimated biomass burned times the emission factors (EF) for the VOC species that were measured by the available instrumentation. Only about 50% of the mass of VOCs emitted was measured (Yokelson et al., 2008; 2012; Akagi et al., 2011) and most of the unmeasured VOC are high-mass, reactive compounds that are missing in the model." What remains unclear is how much the missing VOC were addressed by the scaling of SVOC to POA.

P22909, L11-12: It makes sense for the BB contribution next to a megacity to be lower

ACPD

12, C7084–C7093, 2012

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than the global average because most BB occurs in remote areas.

P22909, L18-20: On the other hand if BB-CO was 30% of FF-CO as estimated by Crounse et al. (2009) and dO3/dCO was 30%, which could occur with removal of the NOx limitation via mixing, the BB-O3 starts to become significant (though not major) at 6 times 1.5%, so within the uncertainty some impact is still possible.

P22910, L5-8: The point about the early morning anthropogenic emissions being released in a shallow boundary layer, which concentrates them is important and one might also add that early AM anthropogenic pollutants have more time for SOA and O3 formation than afternoon BB emissions. But since the BB emissions are released in the lower layers do the modeled injection heights lead to less concentration mainly due to rapid post-emission vertical mixing in the model in the afternoon?

P22910, L8-10: Not sure the simulation really gives higher BB impacts than most observations. If you weight aircraft/tracer outflow observations equally to ground-based estimates as two independent approaches, then the model impacts might be a bit lower than measured impacts?

P22910, L14-16: This is important that the model can weigh in on which ground based sites are most representative of the MCMA as a whole. In Table 4, does "overall" indicate the average of the two high fire periods and NOT the whole month of March as I first understood it? If so maybe label that row as "average for high fire periods simulated" or something specific?

P22910, L20-25: Do you mean the eliminating the "simulated" impact or eliminating the "observed" impact has a small difference on the overall BB impact? It seems like the observed/missed peaks are a significant part of the measured total.

P22911, L6-7: On a quick read, when I saw the ∼30% I wondered if the BB impacts dropped a factor of 2 from earlier in the text? Also the BB contribution here is lower than on line 12 just below. It might be helpful to state somewhere that TOA and OA are

ACPD
12, C7084–C7093, 2012

Interactive Comment

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the same thing or chose one acronym and use it consistently. Also note somewhere early on that TOA is about twice as large on average as POA. With the above points in mind, it then makes good sense that during the high fire episode 60% of POA is BBPOA, but only 30% of OA (or TOA) is BBOA. E.g. SOA and POA are about equal halves of TOA and BBSOA is small compared to total SOA.

P22912: At this point it starts to feel like the paper has already covered all the comparisons and now we seem to be repeating some of them? There may be too many comparisons and percentages in the text. Maybe a few of the most important in a bar graph is enough? See my suggestion for a figure below.

P22912, L7: Maybe change: "We further estimate the BB partitioning on the MCMA-local and regional scales (Table 5b)." to "We further estimate the BB partitioning on the local (MCMA) and regional (domain wide) scales in Table 5b." At first I was not sure what was meant by "regional" in the text.

P22912, L8: change "contribution from" to "contribution to TOA from" – it's hard to follow all the interrelated comparisons – so it will help to be very specific. A bar chart showing TOA, POA, and SOA totals from all sources and then BBPOA, BBSOA, TBPOA, and TBSOA all on the same scale as sums or as fractions of TOA (which would have a bar height of 1) might be a concise, clear way to show the whole picture and clarify the extensive text discussion. One bar chart each at the T0, T1, MCMA, and the domain-wide scale would be useful. It could all be in one figure with parts a-d and possibly replace some tables; or some of the tables could move to supplementary material?

P22913, L2 & L4: Does "PEC" mean "primary EC" and wouldn't just "EC" as used elsewhere be OK?

P22913, L10-11: Is the 0.09 fraction of hotspots occurring in March calculated for Mexico as a whole or for the model domain? The fire activity in the MCMA probably usually loosely reflects the national trends, but in 2006 March was high in model-domain hotspots relative to April and May compared to the rest of the country, which had dra-

ACPD
12, C7084–C7093, 2012

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matically more fire activity in April and May than in March.

P22913, Sect 4.2: The biofuel use may be lower per capita in urban areas than in rural areas

P22914, L10: consumption (no “s”)

P22914, L13-15: change: “such as low frequency overpass and difficulties in detecting small size and short duration fires, as well as the meteorological factors” to “such as low overpass frequency, difficulty in detecting fires of small size or short duration, and clouds”

P22914, L23-4: Uncertainty alone doesn’t indicate the BB impacts are underestimated, but the higher BB/FF contribution from the airborne/tracer approach may be the best evidence of that along with the low ratios of BB-VOC/BB-CO; both mentioned above.

P22915, L15: It’s good to recommend burning during the day, but this is the only feasible time and standard practice already. (ref) Hardy, C. C., Ottmar, R. D., Peterson, J. L., Core, J. E., and Seamon, P.: Smoke management guide for prescribed and wildland fire; 2001 ed., PMS 420-2, National Wildfire Coordinating group, Boise, ID. 226 p., 2001.

Table 1 header “OA” to “aerosol” “neph to FTIR?

Fig 1 minor point – change labels of T0-T2 to another color?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22891, 2012.