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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Emissions from forest fires near Mexico City" *by* R. Yokelson et al.

R. Yokelson et al.

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Response to comments by Reviewer # 2 on "Emissions from forest fires near Mexico City"

On behalf of all authors by R. Yokelson.

Comments by Anonymous Referee #2 R2.1. This manuscript describes the results of airborne measurements of forest fire emissions near Mexico City. The most significant finding, to me at least, is that NOx and HCN emissions were relatively high in the forest fire plumes, possibly as a result of nitrate deposition to the vegetation surrounding the city. The authors use the emission factors for HCN and PM1 to estimate the contribution of biomass burning to the outflow of aerosol from the Mexico City Metropolitan Area (MCMA) and this is where the manuscript runs into some very serious problems as outlined below. I suggest a very major revision before the manuscript is acceptable for publication in ACP.

Au2.1. We are not sure that the “problems” in the paper were “very serious” or required “very major revision,” but we do thank reviewer 2 for several good comments as will be seen below. In particular, we thank both reviewer #2 and P. DeCarlo (the attributed comment) for raising the important issue of secondary aerosol formation. Our paper attempts a preliminary source apportionment for the MC area plume downwind from the city and since secondary formation of aerosol can occur within hours to days; it is important to consider. We have added a discussion of this in section 3.5 as discussed above and it is a useful addition to the paper.

R2.2. A. The authors derive the biomass burning contribution to the aerosol in the MCMA by attributing the difference in measured PM1/CO ratios downwind from MCMA and the PM10/CO ratio in the emission inventory entirely to regional biomass burningĚ..

Au2.2. Actually this is not how we estimated the biomass burning contribution to the MC-area plume. We used 2 different procedures, both of which are clearly described in the paper and neither of which are accurately described by the reviewer’s text above. We are not sure how reviewer 2 came up with the above idea, but we continueĚ

R2.3. This assumes that (1) the PM10 emission inventory is accurate, Ě

Au2.3. One source of uncertainty in our bottom-up source apportionment is the accuracy of the PM10 in the MCMA EI. One source of uncertainty in our tracer-based source apportionment is the accuracy of the PM10/CO ratio in the MCMA EI. West et al argued that the CO in the EI was underestimated by a factor of two. If both CO and PM were underestimated by a factor of two, the ratio would be unchanged and our second estimate (consistent with our first estimate) would not change. In any case, the accuracy of the EI is already mentioned in the paper as a source of uncertainty.

R.2.4. and (2) that there are no other sources of aerosol apart from direct emissions. The first assumption is not tested in the current manuscript, and the second assumption is clearly not true.Ě.

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Au2.4. As shown above, the revised paper now addresses secondary processes. We do not test the accuracy of the EI in this paper as that is not the topic of our research. However, as already mentioned, we do clearly list the EI as a potential source of error.

R2.5. Secondary formation of nitrate and sulfate are significant in the MCMA plume and also, several authors have shown that secondary organic aerosol in urban plumes can exceed primary sources by an order of magnitude (summarized in Volkamer, GRL 2006). In addition, mineral dust was shown to be an important aerosol component during Milagro (for example from the airborne Lidars onboard the NASA King Air and the DC-8. Mineral dust contributes mostly in the super-micron size range of course, but also below 1 micron (Moffet, ACPD 2007).

Au2.5. Secondary aerosol formation is important in both the industrial emissions and fire emissions. The Volkammer paper could represent an upper limit for urban secondary organic aerosol as it is based entirely on one potentially anomalous day of a month-long campaign. The base-day used (April 9, 2003) had the highest Zn levels of the MCMA-2003 campaign (Johnson et al., 2006) and that could indicate that high levels of garbage burning (or incineration) contributed significant amounts of OC. Volkammer et al also assumed that the contribution of biomass burning to OC was minimal on April 9, 2003 due to the low number of MODIS hotspots. However, the MODIS hotspot count was likely too low due to cloud cover that would obscure hotspots in the MC-area that day (http://www.conabio.gob.mx/mapaservidor/incendios/modis/imagenes2003/abril/diurnas/terra/paso2/ql_t1.030409.1748.pn
We use the total secondary aerosol formation estimates of P. DeCarlo's attributed comment as typical in our added discussion. Mineral dust will contribute below 1 micron, but we are already being conservative in our estimate of the fire contribution since we are not including the fire contribution above the 1 micron size level.

R2.6. B. A wealth of data was presented at the Milagro workshop in Mexico City from May 16-19, which indicated that the BB contribution to the MCMA plume estimated in this manuscript is at the high end: i. Measurements of levoglucosan (T0 and T1),

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acetonitrile (T0, T1, the Aerodyne mobile lab and the C-130), and a BB component derived from the AMS spectra (T0, Aerodyne mobile lab and C-130) all indicated lower BB contributions in the MCMA outflow. In addition, the ground-based measurements of these BB tracers were the highest in the early morning when local emissions built up in a shallow mixing layer under low wind conditions, indicating a significant local contribution to the BB source, i.e. no the forest fires discussed in this manuscript. ii. Measurements of potassium in single particle mass spectra at T0 indicated a high percentage of particles attributed to biomass burning (Moffet, ACPD 2007). However, the diurnal variation of the BB particles was quite different from the other BB indicators (acetonitrile, levoglucosan, BB factor from the AMS). iii. Carbon 14 data indicated a relatively high fraction of modern carbon, which can of course also be attributed to secondary formation from biogenic VOCs (although this is a less likely explanation in the MCMA).

Au2.6. The above comments are interesting, but its not appropriate to use the unpublished data (UPD) discussed above in our current paper: (1) The UPD have limited relevance to our paper for obvious geometric reasons (expanded below). (2) The UPD are un-reviewed and unpublished and therefore still have higher potential for error or misinterpretation as we also show below.

(1) Our paper discusses the emissions from a pronounced “ring of fire” surrounding MC at much higher elevation than the city itself. On average about half or more of these emissions will originate downwind of downtown and they will mix with the outflow, but not be transported to downtown MC. Even the fraction of fire emissions that originate in the angular fetch of a point in downtown MC (some 20-30%) will be emitted at much higher elevation and have some tendency to pass well above ground-based sites. This is confirmed by Wennberg/Crounse data showing that the fraction of biomass burning products increases with altitude in vertical profiles above downtown MC (in preparation). Thus one expects that the real mountain fire contribution to the downtown MC aerosol is smaller than the mountain fire contribution to the regional outflow. This is

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already clearly explained in our introduction (lines 1-4, page 6689), shown in Figure 2 (which shows fires mixing with the outflow that are, by definition, downwind of MC), and in lines 24-28 of page 6706. In fact, an estimate of a 20-30% fire PM fraction in downtown MC is not evidence that our outflow source apportionment is wrong. It is actually strong evidence for a much larger fraction in the regional outflow: similar to our estimates. A less obvious point related to study geometry is that a morning peak in biomass burning indicators could have a contribution from the mountain forest fires we sampled. These fires can smolder in the evening and night and the emissions can be collected in the downslope flow and build up in the MC basin.

(2) There are numerous reasons why simple, preliminary conclusions from the biomass burning indicator data discussed above may not be valid. The MC acetonitrile data cannot be used for rigorous source apportionment of the fire contribution because there were no measurements of the initial emissions of acetonitrile from MC area fires. Nor is the source region for the acetonitrile detected in downtown MC clear.

Next we go in to some detail on levoglucosan - not to single it out, but to illustrate the real complexity and uncertainty associated with most biomass burning "markers." Using levoglucosan (lg) as a biomass burning (bb) tracer involves several significant uncertainties or assumptions, which we list here: a) The lg emissions per unit mass of paper burned can be much higher than those from burning some types of biomass (Simoneit et al., 2003). In developing countries (including MCMA), much lg could be from paper burning at dumps. b) The lg emissions per unit mass of biomass burned are extremely variable (Simoneit et al 2003; Mazzoleni et al., 2007). In the absence of paper burning, lg may not peak locally when total bb peaks locally, since the peak in burning could involve consumption of a low lg emitter. c) Depending on how lg is measured, and what it is co-emitted with (which can vary), chemical interference could be a significant problem. d) Low-temperature pyrolysis (similar to smoldering combustion) of cellulose favors formation of levoglucosan, but high-temperature pyrolysis of cellulose (more like flaming combustion) favors production of glycolaldehyde instead (Essig

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et al., 1989). Thus, lg is probably formed mostly by smoldering combustion. Most of the fuel consumption in most global burning is due to flaming combustion. The two types of combustion also can have very different post-emission transport. Thus, "tracers" or "markers" from different combustion processes may not peak locally or remotely at the same time. e) Acid washing of cellulose removes the alkali metals, such as K, and also increases the pyrolysis yield of lg by a factor of 10 or more. (One of many examples in the literature can be seen in Table 2 of Radlein et al. (1991).) Thus, in nature, species with high K content may be low lg emitters and vice versa. In addition, the emission factor for K for flaming combustion is about 5-10 times higher than it is for smoldering combustion (e.g. Figure 6 in Ward and Hardy, 1991), which likely produces the lg. Thus, there many reasons why lg and K could be measured accurately, but still not correlate well. Therefore, an anti-correlation between these tracers at a marginally relevant location is not a reason to change our estimates. In contrast to the above complexities, we note that the EFHCN were found to relatively independent of the mix of flaming and smoldering combustion in Africa (Yokelson et al., 2003), Brazil (Yokelson et al., 2007), and in this work. We added a sentence to the revised paper noting this. Finally, the peak in any bb tracer (including acetonitrile) at a given location does not necessarily reflect a local source and the true time/location that the detected emissions were produced can be highly uncertain. Sampling networks or back trajectory calculations can help, but cannot completely eliminate this uncertainty. In particular, valleys tend to collect the overnight smoldering emissions from a large surrounding mountainous area, leading to a morning peak in urban locations that reflects remote smoldering production followed by downslope transport and not a local source We don't know what the "AMS BB factor" is, but it should be thoroughly tested and described in a publication. Until then, we cannot use it for source apportionment in our paper. The carbon 14 data should be very useful as the PM and condensable organics from vegetation and fires should contain carbon 14. We look forward to seeing the published results. However, the "relatively high contribution" mentioned by the reviewer is not quantitative information that we can use at this time, but, it does highlight that the data referred

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to by reviewer 2 may not yet have a consistent interpretation. We do add a brief discussion of the papers by Moffet et al and Stone et al, which are now published on the ACPD website. These papers clearly support our estimates. To do this we changed the title of section 3.7 from “Relevance to earlier measurements in Mexico City urban area” to “Relevance to ground-based measurements in the Mexico City basin” Then we add a paragraph to the end of this section that reads as follows: “As part of the March 2006 MILAGRO campaign, at least two ground-based studies focused on source apportionment for particles, in the MC basin, during roughly the same time period as our airborne measurements. Stone et al (2007) estimated that biomass burning accounted for 5-50% of the organic carbon in particles collected on filters in the Mexico City basin. At a downtown MC site during MILAGRO, Moffet et al. (2007) found that particles with a biomass-burning core accounted for most of the submicron, ambient particles by number. The results of both of these studies would have been partially affected by types of biomass burning other than the mountain fires that we directly sampled from the air and discuss in this paper. Nevertheless, as explained above, both of these estimates are consistent with our preliminary source apportionment for the outflow.”

R2.7. In summary, different measurement results disagreed on the BB contribution to the MCMA outflow. Few of these results have been published, thus far, so in principle the authors of this manuscript are not strictly required to take them into account. Nevertheless, it is important for them to understand that there are conflicting pieces of evidence, which they may want to consider in publishing these results. It seems to me that the authors could choose between 2 possibilities: they could significantly shorten the manuscript by focusing on the determination of emission factors (and give more details on those). Or they could strengthen their arguments about the BB contribution to the outflow from the MCMA by taking into account additional evidence that should be readily available to them: the AMS data for BB organic aerosol (Jimenez) and the PTR-MS and TOGA data for acetonitrile (Karl, Apel) from the C-130, as well as the findings from the other platforms and ground sites.

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Au2.7. We do understand that a lot of important data was collected, but we don't know yet if the data is in conflict and we disagree with reviewer 2's characterization of our choices. The workshop data described above should be scrutinized and published as appropriate. We hope there will be a second round of MILAGRO papers focused on synthesis of the initial results that would hopefully establish conclusions that are consistent with all the published data. A valid synthesis of the urban data would be very useful for estimating how much modern smoke management techniques (e.g. careful timing of wildland burning) could improve air quality in Mexico City. However, a different, important problem is the contribution of area fires to the area outflow. Our paper is about the second issue thus it would be illogical to change or eliminate our conclusions based on unpublished data that is only marginally related to our issue. All of the data eventually published from the campaign should be discussed in some future overview paper, but our current paper is necessarily more narrowly focused.

R2.8. Further comments: 1. The authors use MODIS hot spots to estimate BB emissions in the vicinity of the MCMA. I wonder if this can lead to overestimates of BB emissions. Grass fires, for example, can be observed by MODIS, but the burnt biomass is very different from forest fires. This may be a problem in the MCMA where open fires to burn agricultural waste and weeds along roads are quite commonĚ.

Au2.8. The reviewers concern about some hot spots being grass fires is understandable since, globally, many forest fires do burn much more biomass per unit area than grass fires. We have two main points to make in this regard: (1) We first point out that in our 7 flights to the MC area we saw many pine-savanna understory fires from the air, just two grass fires (one very small one by the landing strip in Toluca), and no land-clearing, agricultural, or weed fires. This suggests that a similar mix of fires should be detected in this area from space, which we have partially confirmed by overlaying the vegetation with the hotspots. (2) In addition, the pine savanna fires near MC are not like the slash and burn fires in the Amazon or the destructive, stand-replacing crown fires in the boreal forest. The pine-savanna fires are understory fires that burn mostly

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grass (which is usually less dense under a partial canopy), and some shrubs and litter. Therefore, the fuel consumption per unit area is not that much different than for grass fires. In fact, on the planned fire, the fuel consumption in the open areas of grass was 5.36 Mg/ha, which is close to the literature average for grass fires. The fuel consumption in the canopy areas was always 7.9 Mg/ha or more. We assumed that the average planned fire fuel consumption of 6.54 Mg/ha was appropriate for the "average fire" in the MC area and this value is both conservative and not much higher than the grass value. Finally, it is well-known that the small, understory forest fires that dominate this region can be very difficult to detect from space because the canopy temperature is rarely elevated. Thus the hotspots are likely to underestimate the number of forest fires at overpass time. In this paper we do not attempt hotspot validation, but just use the hotspots as a starting point for a preliminary, reasonable, conservative estimate of biomass burned. Hotspot validation is being undertaken by the Mexican government and more quantitative estimates may result from that in the future. We have revised the section on the ground-based fire measurements to clarify these issues and the concerns of reviewer 1. The new section reads:

"All but a few of the many fires observed from the MILAGRO aircraft in our MC study area were understory fires in forests dominated by pine. However, overlapping the March 2006 MODIS hotspots (http://maps.geog.umd.edu; Justice et al., 2002) with a vegetation map (Loveland et al., 2000) indicates that a sizeable fraction of the fires occurred in grassland (Figure 1). This may be partly due to the fact that fires under a forest canopy can be difficult to detect from space (Brown et al., 2006). In addition, at least two of the forest fires we actually sampled (and photographed) are in non-forest areas according to multiple vegetation maps (INEGI and INE, 1998; CONABIO, 1999; Loveland et al., 2000). In any case, to ensure both airborne and ground-based sampling of a representative, well-characterized fire; a planned fire was carried out by CONAFOR (Mexican Federal Forest Service) and the Department of Ecology of the state of Morelos. Prescribed fires are conducted every year before the fire season to reduce fuels and wildfires in the region. However, CONAFOR agreed to burn one

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of the areas in March to allow us to measure fire behavior, fuel consumption, and smoke emissions representative of the fire season. The fire included adjacent units of grassland and pine savanna and was located in the Corredor Biológico Chichinautzin, National Park, which has one of the highest incidences of forest fires in central Mexico. The planned fire was ignited around noon local time on 17 March and burned for about one hour. The fuel consumption was measured by the difference between pre- and post-fire fuel loading as determined by the linear intersect method (Brown, 1974). In the grassland areas the fuel consumption was 5.36 Mg/ha. In the forested section, the fuel was mainly grass plus some litter, shrubs, and small woody debris and the fuel consumption was 7.9 Mg/ha. The area burned by the fire and the total fuel consumption are shown in Table 1.

Some of the other pine-understory fires sampled by the Twin Otter were later located by the ground-based crew. The burned area was measured and the fuel consumption was measured by comparing adjacent burned and unburned areas (Table 1). In addition, fires that were not sampled by the MILAGRO aircraft were also located by the ground-based crew and the burned area and fuel consumption measurements obtained are also shown in Table 1."

R2.9. The authors could strengthen their case considerably by overlaying the MODIS hot spots with land use and vegetation type around the MCMA to estimate how many of the hot spots were actually forest fires. Wiedinmyer at NCAR has been working on this problem for North America and may have some relevant data for Milagro.

Au2.9. Figure 1 has been updated to include land cover based on the 1-km2 resolution North American Seasonal Land Cover Database Version 2.0 (SLCV2.0) from the USGS Land Cover Characterization Database (http://edcsns17.cr.usgs.gov/glcc/glcc.html ; Loveland et al., 2000). The original land cover categories of SLCV2.0 were aggregated to produce the simplified land cover classification used in Figure 1. As the updated Figure 1 shows, during the month of March 2006, the MODIS active fire detections in the study region occurred predomi-

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nantly on land belonging to a forest or woodland classification. Specifically, the MODIS active fire detections during the study period may be summarized as 50 % forest (mixed forest (mostly pine-oak), tropical forest, conifer), 41% grassland, 6% agriculture, and 3% shrubland. Given the above explanation that forest fires dominated the visual observation from the air and that their fuel consumption was not that different from grass fires, we don't think any adjustment in our crude estimate of the amount of biomass burned in the study area is necessary.

R2.10. 2. Another problem arises upon examination of Figure 1B: one of the hot spots seems to be on the top of Popocatepetl. The summit of this volcano is at 5400 meters and is well above tree line. I would guess that the hot spot detected by MODIS is the crater of Popocatepetl in this case. I am not an expert in this area, but a quick literature search shows that thermal volcanic features are also detected using MODIS. 3. Many of the other hot spots to the north of Popocatepetl also seem to be high up on the volcanoes. Are these hot spots above or below tree line? Ě..

Au2.10. MODIS detected 260 hotspots in March 2006 in our study area. Forty-two of those hotspots were classified as "zero confidence level" that they were fires. This is often because the hotspot is permanent and thus is probably due to a smokestack, large metal roof, or a volcanic feature. If one of the 218 remaining hotspots was mistakenly the crater of Popocatepetl, this is not a significant error in our crude estimate: i.e 1/218 is less than one-half of one percent. Keep in mind there are a lot of high elevation forests and that tree-line is about 13-14,000 feet at this latitude.

R2.11. The higher up the volcanoes the forest fires are, the less likely it is that the emissions are mixed down to the surface.

Au2.11. This is why the outflow contribution and the urban contribution are not the same issue as discussed above.

R2.12. Also, are there any volcanic/geothermal features that could be observed by MODIS?

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Au2.12. We doubt this is a significant source of error since the data are screened as explained above.

R2.13. 4. The first author of this paper has a long and outstanding track record in the determination of BB emission factors in the laboratory. It seems obvious to add a section to this manuscript to determine the HCN and NOx emission factors for vegetation in the MCMA, which could make the conclusions about the influence of pollution on HCN and NOx emission factors a lot stronger.

Au2.13. We are not sure we understand this comment. Sections 2.2.1 and 2.2.2 already describe the procedure for determining the emission factors. If the suggestion is to add lab data to this paper, we have none to add.

OTHER CHANGES

1. Corrected affiliation of CU authors.

2. Added to the acknowledgements: "We also thank the fire staff of Mexico's Comision Nacional Forestal in the states of Morelos, Mexico, Distrito Federal, and the Guadalajara Headquarters for the execution of the prescribed fire in the state of Morelos and for logistical support to conduct post-fire assessment of several fires of opportunity flown by the Twin Otter aircraft."

3. We moved the description of the ground-based fire/fuel measurements (now 2.1.6) so that it follows the description of the other data acquired. Then the airborne sampling protocol (now 2.1.7) follows.

4. The caption for the revised Figure 1 is: "The location of the MODIS hotspots and the fires sampled by MILAGRO aircraft in the study area. The land cover map in Figure 1 is based on the 1-km2 resolution North American Seasonal Land Cover Database Version 2.0 (SLCV2.0) from the USGS Land Cover Characterization Database (http://edcsns17.cr.usgs.gov/glcc/glcc.html ; Loveland et al., 2000). The original land cover categories of SLCV2.0 were aggregated to produce the simplified land

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cover classification used in Figure 1. The mixed forest category consists mostly of pine mixed with other conifers or oak."

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