

Interactive comment on “Evolution of anthropogenic pollution at the top of the regional mixed layer in the central Mexico plateau” by D. Baumgardner et al.

Anonymous Referee #3

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General comments:

This paper presents very interesting results from a unique mountain-top site in Mexico. The air masses observed from 3 directions other than Mexico City are described in detail. Much of the data and discussion is publication-worthy. Two main points are called out in the abstract and conclusions:

1) Mexico City is not the only source of pollutants in the area, and 2) biomass and wood burning are the main sources of organic particulate matter.

There are serious problems with both of these assertions.

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While I don't disagree with the 1st point and agree that the site certainly experienced polluted air masses from the non-Mexico City locations, it would certainly help to have shown data from air masses originating from Mexico City for comparison. If no such air masses were observed or if the origin of suspected air masses is uncertain, this should be stated. Why are only 3 days of data used? It is fine to focus on select days, but it would help if the reader were given a feel for the rest of the dataset. It seems a reach to state that

"This mixture rapidly erases the signature of a unique Mexico City "plume"; and suggests that the environmental impact of this region should be considered as one that stems from a large area source rather than a single megacity."

To make the above claim it would seem necessary to consider data that actually looked at the Mexico City plume, likely from various distances until the distinct "Mexico City signature" was no longer evident. The above statement is not well supported by 3 days of data in which the air originated from other locations. Consideration of emission inventories would also be helpful, as would more information on Puebla and Cuernavaca (e.g., population, etc)

The 2nd argument (about biomass and wood burning) is based on a comparison of the observed CO/OM ratios with literature values for diesel, biomass, and wood burning. This analysis appears flawed for the following reasons:

a) it ignores the fact that the CO/OM ratios from urban areas, diesel exhaust and from wood burning decrease with photochemical aging (Kleinman et al, 2008, Greishop et al, 2008, Robinson et al, 2007, deCarlo et al, 2008). This was already partially described in this manuscript, and yet ignored in this part of the analysis!

b) It ignores gasoline exhaust; which is the major source of CO in most urban areas and has a much higher CO/OM ratio than diesel exhaust.

The earlier discussion of OM and OM/CO, especially as compared to other locations

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(Salcedo et al, Kleinman et al, etc), is a strong point of this paper (along w/ figure 15), as is the description of the meteorology. There is other data presented in the paper (e.g., FTIR analysis of the filter samples) that is extremely interesting but only briefly discussed. For example, what could be the cause of the differences in the organic composition shown in figure 11? This manuscript would benefit greatly from more in-depth discussion of the data presented in figures 11, 13, 14, and 15. Figure 16 and the accompanying discussion should be removed from this paper (as described earlier). Figure 17 and the accompanying text (BC and CO) could be removed too.

More of the time series data (beyond the 3 days examined) would certainly be welcome and very appropriate for ACP.

Specific comments:

Abstract: "highest concentrations were from the east". This should be clarified in the abstract; is this from Mexico City or not? Explicitly stating that Altzomoni is southeast of Mexico City would be helpful. Is this statement about high concentrations from the east only based on the 3 days evaluated or the entire dataset?

Background section: I found the background interesting, but it does not prepare the reader for the rest of the manuscript well. It does not set the framework for the discussion of the relative importance of pollution sources (Mexico City vs non-Mexico city), nor does it discuss the relative importance of the various organic PM sources (anthropogenic, wood/biomass burning, etc) - a topic for which there is not currently a consensus among researchers. The revision, which will hopefully focus on the more concrete findings of the study, should have a background/intro section that places the findings in context.

Fig 1, though interesting, is not necessary and could be described solely in the text.

table 1: does the accuracy column refer to a 1 sigma or 2 sigma uncertainty?

How is OM measured? Is this organic aerosol (OA) from the AMS? This needs to be

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explicitly defined in the experimental section, since OM is an ambiguous term - does it include gases and PM? "Organic PM" or "Organic aerosol" are preferred terms

More information should be provided regarding the operation of the AMS - how were the 2 calibrations performed? Did they agree? Collection efficiency?.

pg 3279: that O₃ is lower in Mexico City compared to a hill side site is likely also due to the proximity and amount of NO emissions in Mexico City.

The discussion of OM/CO (and similar ratios) are a strong point of the paper. These ratios should be calculated after subtracting the background OM and CO values; it is not clear if this was done. These calculations need to be better defined.

pg 3281: line 13: "The mass concentrations of OM and inorganic compounds do not show a significant decrease in concentration as related to the possible origins of the air masses." - meaning is unclear.

pg 3281, lines 22 - 25: the comparison of the concentrations of O₃ and PM between Mexico City and Altzomoni is interesting, but the analysis fizzles with: "From this we conclude that O₃ and particles at the Altzomoni site are not only coming from the primary emissions, but there are additional sources that offset the dilution as the RML grows." It is well known that O₃ is only formed as a secondary pollutant. It is also well established that particulate matter has both primary and secondary sources. These facts should form the starting point (rather than the conclusions) for a discussion of the observations.

pg 3282: the conversion factor of 1.8 to convert from OM to Organic Carbon is based on previous results (Takegawa et al 2005) that might not be applicable to the air masses observed at Altzomoni. A discussion of whether this factor is applicable is needed. In fact, Takegawa et al (2005) quotes studies that have shown OM/OC ratios in the range 1.2 to 2.1. The recent ACPD publication by deGouw et al (Atmos. Chem. Phys. Discuss., 8, 21265 - 21312, 2008), which was not yet published when this manuscript

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was submitted may contain useful data (though this would not be necessary if this section is removed).

Technical corrections: O₃ is in ppb, CO in ppm; why not use ppb for both?.

Abstract, line 12: "25E-3"; needs units (both in abstract and later in the body). Shouldn't this be 25 x 10³? (without the minus sign)

Abstract line 15: Insert the word "aerosol" as indicated: "The mass concentration of OM in the RML was greater than 70% of the total aerosol mass, regardless of the air mass origin." Rewriting as "OM accounted for more than 70% of the total aerosol mass" would be better still.

Abstract, line 12: "maximum CO at " should instead read

" maximum CO concentration" or "maximum [CO]"

figure 3 caption has typos: it appears that Mexico City and Cuernavaca are northwest and southwest of Mexico City (not northeast and southwest as written in the caption)

pg 3271, line 7: should read "west of Puebla" (not east)

fig 2 shows very interesting data. It would be easier to examine if there weren't separate axes for the 2 locations. Just use one common axis.

pg 3280: line 29: need units for OM/CO

pg 3274: best to avoid contractions in technical writing ("does not" instead of "doesn't")

Grieshop et al., Laboratory investigation of photochemical oxidation of organic aerosol from wood fires Part 1: Measurement and simulation of organic aerosol evolution, *Atmos. Chem. Phys. Discuss.*, 8, 15699-15737, 2008.,

Robinson et al., Rethinking organic aerosols: Semivolatile emissions and photochemical aging, *Science*, 315, 1259-1262, 2007.

Kleinman et al., The time evolution of aerosol composition over the Mexico City plateau,

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Atmospheric Chemistry and Physics, 8, 1559-1575, 2008.

DeCarlo et al, Fast airborne aerosol size and chemistry measurements above Mexico City and Central Mexico during the MILAGRO campaign, Atmos. Chem. Phys., 8, 4027-4048, 2008.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 3265, 2009.

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9, S229–S234, 2009

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