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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fast airborne aerosol size and chemistry measurements with the high resolution aerosol mass spectrometer during the MILAGRO Campaign" *by* P. F. DeCarlo et al.

P. F. DeCarlo et al.

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Responses to Anonymous Referee 1

1.1

This manuscript reports aerosol and supporting trace gas measurements from the C130 aircraft during the MILAGRO field campaign. The centerpiece of this study is the high resolution AMS; with many findings of general interest reported here in the context of measurements in and downwind of Mexico City. Results on the changing O to C ratios could not be obtained without high mass resolution capability. Data quality as judged by comparisons between the AMS, nephelometer, and SMPS is excellent. The paper is well written and I recommend

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publication after the following comments are considered.

We thank reviewer 1 for the overall positive evaluation and his/her detailed and thoughtful comments on our manuscript.

1.2

General comments: The reader needs to know more about the flights and the data set used in this study. Does the data set include free tropospheric air. It looks like it might from Fig. 6. If so, how does this effect results such as the pie diagrams in Fig. 3.

The altitudes sampled are now described in more detail (see response to comment 2.4 from reviewer 2). The dataset does include some FT air. However, the criteria for defining what is FT air is not straightforward, and so we did not attempt this, and instead presented the data as measured. Additionally if the FT air we sampled was mostly devoid of aerosol species, the relationships between the different chemical species should not change, but the absolute average concentration would change in Figure 3.

1.3

Time of day is an important parameter, especially for the near city observations; less so for plumes that are more than 1 day old. Information on chemical conditions corresponding to the 3 C-130 pie charts (Fig. 3) would serve to put the aerosol observations in perspective.

This comment concerns only the pie chart for regional air (Fig. 2c) as the other pie charts contain all or much city data where time of the day is highly relevant. We are unclear about what the reviewer is referring to with "chemical conditions", but various gas-phase species etc. varied across regional air and it would be too cumbersome to present such ranges of variation here. Rather the co-evolution of aerosol with the most relevant gas-phase species (CO, HCN) is already discussed in the manuscript (e.g. Fig. 5, 9, 10, 11). The comparison of gas vs particle phase organics for this dataset and several others was already addressed by Heald et al. (ACP, 2008). Further

ACPD

7, S9821–S9826, 2008

analysis of the gas-phase measurements will be presented in separate publications by researchers performing those measurements. The data is also publicly available in the MILAGRO archive for researchers desiring to perform various analyses or comparisons to other studies.

1.4

Specific comments: Abstract line 5-6 need to be re-worded. "... the first aircraft deployment of the HR-ToF-AMS, in which the instrument performed very well ...", implies that there were other deployments in which it did not perform well

The text has been changed to avoid this potential misunderstanding.

1.5

p 18280, line 1 CE for AMS was assumed to be 0.5 Whether or not this value was assumed according to literature references, it would be useful to alert reader to the instrument comparisons in Fig. 2.

As suggested, the reader is now directed to see Figure 2 at this point.

1.6

p 18284 line 18. Primary organics are higher in the evenings, nights, and early mornings. Does this out weigh the higher SOA in photochemically active periods?

Figure 6. of Salcedo et al. (ACPD 2006) shows the diurnal cycle of AMS mass species during the MCMA-2003 campaign at the CENICA supersite. Organic mass loadings are actually highest in the morning likely due to the combination of primary OA emissions and the start of the SOA formation with a lower boundary layer. Since the C-130 only flew in the city during the afternoon to evening, we found it more appropriate to consider only these times for the aerosol composition comparison with the ground site.

1.7

p 18285, line 24 and Figure 5. The ratio of organic and nitrate aerosol to excess

ACPD

7, S9821–S9826, 2008

CO is presented. How is excess CO defined? How sensitive are results to the definition? At 600 km downwind of Mexico City is there sufficient "excess" CO to give a robust result?

The following text was added to address these points:

"Excess CO is defined as the CO concentration above background, with the background value for each flight given in Table 1. Additionally data with OA concentrations less than 2 μg m^{-3} STP or NO3 concentrations less than 0.2 μg m^{-3} STP were eliminated from the analysis to reduce the impact of noise and of uncertainty in the CO background."

These criteria limited the data with CO levels which were only slightly above background. 98 percent of the points used had a CO excess greater than 20 ppbv and 90 percent of the points used had CO excess less than 35 ppbv. A note about this point has been added to the manuscript.

1.8

p 18288 Discussion of vertical profiles downwind of volcano. Text says that profiles were made in the same area. Are there differences in exact location, wind speed, or trajectory that yield a difference in atmospheric processing time?

Figure SI-1 in the supplementary information has been added to clarify the locations of both spirals, and is referred to in the text at this point. Text has been added to clarify that the additional daylight processing time for the 2nd spiral is the likely reason for the changes in the size distribution.

1.9

p 18288 Figure 8 shows that O/C ratios increase with distance from Mexico City. The explanation of addition of SOA makes sense. But Fig. 5 shows that, if anything, OA/CO decreases with distance from the City. Can these observations be reconciled?

ACPD

7, S9821–S9826, 2008

The addition of SOA occurs on short length scales, due to its formation timescale of about a day. There is also influence from biomass burning which perturbs the OA/CO ratio when averaged over all flights (as Fig 5 does). The OA/CO for individual flights will be presented in a subsequent paper which is now in preparation. When analyzed for RF12, when the influence of biomass burning is small, this analysis indeed shows that OA/CO increases with photochemical age due to SOA formation.

Regarding a potential small decrease in OA/CO, we are not sure the decreases are significant due to uncertainties in e.g. the CO background, influences from other regional sources, etc.

The text in the paper has been modified to address these points as follows:

"The OA/CO ratio in the outflow near the city is about 80 μg m^{-3} STP ppm^{-1}. This is likely due to a combination of rapid SOA formation from urban emissions and mixing of biomass burning OA, and will be analyzed in more detail in a subsequent publication. This ratio is similar to the value found by Kleinman et al. (2008), for their study of the near outflow on the DOE G-1 aircraft. It is also much larger than values of 5-10 μg m^{-3} STP ppm^{-1} for urban POA (Aiken et al., 2007a; Zhang et al., 2005c), which highlights the dominance of SOA in the pollution outflow of the city, consistent with previous observations in Mexico City (Kleinman et al., 2008; Volkamer et al., 2006; Volkamer et al., 2007) and at several other locations (Zhang et al., 2007a). Both the asymptotic value of OA/CO and the timescale of SOA formation of approximately one day are similar to findings reported for the outflow of the Northeastern US (de Gouw et al., 2008; de Gouw et al., 2005; Kleinman et al., 2007; Peltier et al., 2007a), and of the Po Valley in Italy (Crosier et al., 2007). The fact that similar asymptotic values are observed despite lower biogenic emissions being added to anthropogenic pollution during MILAGRO versus the summer in New England could be due to the higher BB emissions in Mexico during this study (Yokelson et al., 2007; Stone et al., 2008)."

1.10

ACPD
7, S9821–S9826, 2008

p 18289 line 22 "formation" should specify what is being formed.

“SOA” has been added to clarify this.

1.11

p 18290 O/C ratio approximately 0.55 tied to late arrival of C-130 into the city. Reader needs to know altitudes and times.

The following text has been added to clarify:

“Lowest values of the O/C ratio were found at altitudes of approximately 2900 m during a transect through the Mexico City basin around 20:20 UTC (14:30 local), which was clearly dominated by pollution sources.”

1.12

p 18291 Discussion of research flight 2. A map would be useful. Is time on the axis in Fig. 11, UTC? It would be useful to identify time of day in text.

UTC has been added to the legend, and text has been added to indicate the start of sampling corresponded to 18:00 UTC or 12:00 local time. A map of this flight denoting the different airmasses and periods I-III has been added to supplementary information (Figure SI-2) and is introduced in the text at this point.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 18269, 2007.