

Interactive comment on “Microscopic characterization of carbonaceous aerosol particle aging in the outflow from Mexico City” by R. C. Moffet et al.

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Received and published: 1 December 2009

Response to reviewer 2

We thank the reviewer for their helpful comments. We have responded to each specific comment below in italics. We have included the reviewer's original comments in normal font.

General Please check to see if the acronyms for all analytical techniques have been defined.

Done

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Judging from Fig. 1, transport is not from T0 to T1 to T2. Just the mention of Lagrangian transport conveys that impression. It would be useful to refer to published CTM calculations and (if available) PMF analysis to identify March 22 as a day with low or high biomass burning impacts.

The manuscript notes that the transport model used was a Lagrangian transport model and referenced Doran et al, 2008 where Mar 22 was identified as a day of T0 ->T2 transport.

The text states (lines 457-459): “Also, several researchers detected high levoglucosan, a biomass burning marker, on this sampling day (Aiken et al., 2009; Stone et al., 2008).” Hence, we believe the text has already addressed this issue.

Are the particles with inorganic cores surrounded by carbon containing material due to transport from the Tula complex followed by condensation of OA? A more easily answered question is whether this morphology occurs only when there is transport from Tula to T0.

Particle morphology indicating inorganic and/or BC cores surrounded by OC material was typical for the T0 site, including many times when the transport was not from the Tula region. Many of the cores contained K and S, both of which can be attributed to emissions related to combustion of biofuels. In a separate publication (Moffet et al., 2008), found that these particles were very abundant at the T0 site starting around 12:00.

Can conjugated double bonds be differentiated from isolated double bonds? In the NEXAFS analysis, can anything be said about the distribution of locations of the “EC phase”?

In general, C=C bonds have a transition around 285 eV. In principle, the transition energy is shifted to lower energies upon increasing conjugation. As noted in Hopkins et al 2007b, quinone, a ring conjugated with a C=O group has the C=C transition at

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a slightly lower energy. The peak at 285 eV arises from many different overlapping peaks from different bonding environments. For pure substances, a shift depending upon conjugation may be observable. However, due to the complex composition of aerosol, we believe a differentiation between conjugated and isolated double bonds is unlikely.

Although not the focus of this manuscript, yes the distribution of the EC phase is observable using this technique.

Specific Page 17000, line 24-25. 500 um by 500 um spot resulting in 3 hour resolution. Is this the amount the TRAC sampler advances in 3 hours?

The TRAC sampler was not used for collection of the samples for SXRF. The DRUM sampler advanced 1 mm of the Teflon strip in 6 hrs. The time resolution of the SXRF measurement is determined by the X-ray beam spot size, which is 0.5 mm in diameter. Therefore, this corresponds to a temporal resolution of 3 hours resolution for the analysis of DRUM collected samples.

Page 17001, Sec 2.3.4 STXM/NEXAFS How many particles were analyzed? Is the number similar to 25-35K analyzed by SEM? If much smaller, are there concerns about sampling statistics?

A minimum of 250 particles for each sample were analyzed by STXM. However, single energy images of at least 5 to 10 x that number of particles were examined. While these images do not provide spectra of each particle, contrast and morphology are readily observed. In examining field collected aerosol samples this process does provide further information on how "representative" or even homogeneous particles are.

For the detailed analysis, particle statistics were kept high enough to avoid excessive statistical uncertainty. The most sparse particle classes carry a 33% error in their numbers as calculated by Poisson counting statistics. This was added to the discussion in lines 352-354.

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Page 17002, line 21-22. "PIXE and SXRF data confirmed that particles from the same source were sampled..." How so? A distinctive combination of trace elements or a more general urban signature?

On lines 234-235, the manuscript claimed that sampling devices at T0, T1, T2 captured the same plume (and not that the particles were coming from the same source). This assertion is supported by the spike in sulfur at each of the three sites. To further clarify this, "the same plume" has been changed to "regional aerosol plume".

Page 17003, line 5-6. " O_3 (32 ppb) and SO_2 (125 ppb) maximum concentration were not abnormally high on this day" The ozone concentration is implausibly low unless something very strange was happening locally. 125 ppb is a very high SO_2 concentration. A few percent oxidation would make sulfate the dominate aerosol species whereas in actuality it is about 10% with a wide spread regional distribution.

As noted by the reviewer, the quoted ozone concentration was incorrect. The O_3 concentration was 125 ppb and the SO_2 concentration was 32 ppb. We have corrected this in the text and are grateful to the reviewer for catching this. Updated text: line 243.

Page 17003, line 20-23 "highest carbon concentrations occur at the particle edges" This is a very interesting finding. Fig. 4 presents an x-y two dimensional view. In a three dimensional view, could the carbon in the middle (0.5 relative position) be explained by the carbon in the z direction above and under the core?

Yes

Page 17007, line 2-3. What is the possible connection between the increase in homogeneous OC particles and high sulfur concentrations?

The most probable reason for the increase in homogenous OC particles is the growth of nucleation mode particles. If organics and sulfate condensed simultaneously during this growth, they would be well-mixed and appear as homogenous particles, particularly if the aerosols have not undergone a deliquescence/efflorescence cycle. This

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discussion was added to the text in lines 357-361.

Page 17008, line 7 “five analyzed samples” It would help the reader to specify that “samples” refers to 1 at T0, 2 at T1, and 2 at T2.

The text was changed as suggested (line 397).

Fig. 5 I cannot distinguish blue from green on the on the STXM images (at ACPD resolution). Are the aerosol particles in the image subject to the criteria that they be bigger than $0.35 \mu\text{m}$ to be identified? The number distribution peaks at smaller size but I don't see evidence of it in the picture.

No, the particles in figure 5 were subject to the STXM lower size limit of $0.1 \mu\text{m}$. We have stated this more explicitly on p 10, lines 227-228.

Fig. 9 Trace species cannot be seen in figure at ACPD figure resolution.

This figure provides an overview of the particle types and mixing states. If we made it so single particles could be made out, the figure would be impractically large. The focus of this manuscript is the evolution of particle types rather than detailed particle speciation and morphology of a few isolated particles.

Fig. 11 Same question as for Fig. 5. Is the size spectra limited by a $0.33 \mu\text{m}$ cutoff for computer identification?

We believe the reviewer is referring to the size spectra in figure 10. There, the minimum x data point shown corresponds to a bin centered at $0.325 \mu\text{m}$, however, the data point at $0.325 \mu\text{m}$ has a $0.25 \mu\text{m}$ width, and therefore spans from 0.2 - $0.45 \mu\text{m}$. The same is true for the STXM data. This was clarified in the legend for figure 10.

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