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Interactive comment on "Processing of soot in an urban environment: case study from the Mexico City Metropolitan Area" *by* K. S. Johnson et al.

Anonymous Referee #2

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Referee Report on "Processing of soot in an urban environment: case study from the Mexico City Metropolitain Area" by K. S. Johnson et al. Submitted to Atmospheric Chemistry and Physics Discussion

This paper addresses the significant question on the mixing state of soot aerosol generated in a heavily polluted urban environment and the associated question of the atmospheric aerosol lifetime. The present findings are undoubtedly relevant for an understanding of the radiative forcing of free tropospheric carbon-containing aerosol, whatever its origin, which means that it is either emitted in the boundary layer or directly injected into the troposphere by aviation traffic. The study has been conducted in a careful manner involving three different sampling sites and several complemen-

tary analytical and imaging techniques that were used to study thousands of individual particles. The statistical basis of this work appears very solid although it is not entirely clear how the authors arrive at an average particle diameter and associated narrow uncertainty (pages 5593 and 5594) for such disparate particles as displayed in the Figures. I recommend that this study be published with minor modifications and additions once the authors have had a chance to consider the following questions:

Ţ There is very little information about the storage of samples from the moment of sampling to analysis unless on-line techniques were used. It would be informative to learn about the storage of such samples and their potential changes during storage.

Ţ On page 5591, line 10, a distinction between sampled carbonaceous particles and soot is made. Soot contains an amorphous carbon core with up to a 50% by weight of an unspecified organic phase. What were the criteria of distinction between the two types of particles?

Ţ The “sulfate” part of the fresh particulate emissions (pg. 5592, lines 20 ff.) must be H2SO4, and cannot be (NH4)2SO4 or NH4HSO4 because of limited access of NH3 to the freshly emitted particles. Are the authors able to distinguish between H2SO4 adsorbed on primary emitted particles and H2SO4 that is deposited as an aerosol onto the soot particles during sampling? In other words, did the authors check for a sampling artifact in the presence of H2SO4 aerosol that is always emitted in much larger numbers compared to H2SO4 that is already adsorbed on soot? Total sulfur comes out of a combustion engine overwhelmingly as SO2, a few % of sulfur is emitted as (nucleation mode) H2SO4 aerosol, and a fraction of this sulfuric acid aerosol ends up as H2SO4 adsorbed on the soot. This is true for fuel sulfur contents of up to 500 ppm, but depends somewhat on combustion conditions (rich vs. lean flame).

Ţ Is the liquid organic coating of soot particles really from lubrication (engine) oil (pg. 5596, line 5 ff), or could it be a partially oxygenated organic phase of low vapor pressure? Glycols and poly-ols could well be formed from partial oxidation of higher hydro-

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carbons such as undecane, an often-used surrogate for “Diesel” fuel. How could you tell? Oxygen content? It is known that the odour from Diesel combustion comes from polycyclic phenols that have a relatively low vapor pressure (semivolatiles).

Ţ Pg. 5598, line8 ff: what kind of “meter” for the hygroscopic properties of soot are the authors thinking of? From their results it becomes apparent that a simple bulk measurement of hygroscopicity using for instance the contact angle of a sessile drop becomes problematic in the wake of such heterogeneous particles. Part of the particle will be hydrophilic, and other parts will remain hydrophobic for a longer period of time.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5585, 2005.