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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aging fingerprints in combustion particles" *by* V. Zelenay et al.

Anonymous Referee #2

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Introduction and verdict:

This study presents results examining the influence of photochemical aging on the chemical composition and water uptake of soot particles emitted from a wood stove, a EURO 2 car (not fitted with a catalytic converter) and a EURO 3 car (equipped with a diesel oxidation catalyst). Analysis of single fresh and aged soot particles was performed using x-ray absorption spectroscopy and scanning electron microscopy. In addition, ensemble, online measurements of composition and hygroscopicity were also presented and compared to the single particle results. This ,in principle, is a good manuscript describing interesting experiments and providing good observations for an important and timely research topic. However, I have a strong reservation on the interpretation of certain aspects of the presented results, which affect the major scientific conclusion of the work. I recommend publication of the manuscript in ACP, only after the authors address the following comments in a satisfactory manner.

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Discussion Paper

Main Comments:

My main concern is with the authors' message that "we show that the soot particles take up water in humid environments and that their water uptake capacity increases with photochemical aging". Although this might be true for some datasets, I strongly do not believe that the provided data in this manuscript can be generalised to support this conclusion. Using the offline technique, water uptake was only clearly shown for the wood sample and not much uptake was shown for the two cars. On the other hand, the HTDMA data indicated that the photochemical aging had hardly any influence on hygroscopic growth factors of all three soot samples (wood, EURO2 and EURO3). The authors should discuss the resolution of the GF measured by the HTDMA?. The largest reported change was between 1.02 to 1.04 (or 1.06) in the EURO2 case. In my experience, the resolution of the GF reported by most HTDMAs is about 0.05, which makes the reported changes insignificant. The authors should quantify the errors associated with this data and show if there was really any water uptake as a function of photochemical aging. References to this should be revised accordingly throughout the manuscript.

Other Comments:

Title: the manuscript does not really provide aging "fingerprints". It does, however, provide characterisation or investigation of aging effects on the properties of soot particles.

Abstract: It is mostly qualitative in nature and I suggest that it should include the appropriate quantitative results as discussed in the manuscript. The final statement of the manuscript should be revised in accordance with the outcome of the main comments above.

Page 14459, L12 – 13: O3 was added to the mix before lights on. Was this before or after the collection of the fresh soot sample for the offline analysis? If before, could ozonolysis reactions have altered the chemical composition of the collected fresh sam-

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ples?

Page 14460, L1: Figure 10 does not really show changes in GF after 2 hours. The "definition" of aged sample should be revised.

Page 14463, L16 -18: I disagree with this statement. On their own, SEM size distributions do not really show an obvious change with aging. This might be due to evaporation of semi-volatile material in the high vacuum of the SEM? Based on the provided results, I doubt that the technique is suitable for probing the size evolution, unlike the SMPS.

Page 14464, L 29 – Page 14465, L1: If that was the case, why did it not feature in the other samples?

Page 14465, L12 – 13: I am not sure how does this rule out any potential size dependant composition? Please explain.

Page 14467, L2: is it possible to provide an error estimates associated with these numbers? Is the change from 0.84 to 0.80 significant? Similarly on line 6: provide an error estimate of the m/z 44 fraction to show whether the change from 10.1 to 10.3% is meaningful or not.

Page 14467: Relating m/z 44 fraction to the offline carboxyl measurements deserves a brief discussion. Note that the wood sample showed the highest fraction of m/z 44 at 10% and yet the EURO2 sample appeared to have the biggest change in the carboxyl measurement despite being much lower.

Page 14469, L16: the errors associate with the AMS O:C ratios should be provided.

Page 14471, L2 -5: Could the authors provide any explanation of the lack of potassium signal, where it was expected to feature?

Page 14473, L12 -13: Based on the discussion above, I think the phrase "in agreement with the HTDMA measurements" should be omitted.

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Page 14473, L19 -21: The increase was observed in the SMPS data and only for the EURO3 SEM data. The statement should be revised.

Page 14474, L4 – 5: I am not sure where this conclusion came from. Please provide the appropriate discussion in the relevant section.

Page 14474, L13 – 15: I do not believe that this conclusion is supported by the presented results. This should be revised.

Minor comments:

Page 14459, L17: The acronym STXM-NEXAFS has not been introduced in the manuscript and should be.

Page 14459, L20: should be organic 'loading' not 'compounds'.

Page 14459, L28 – 29: how long was the fresh sample collected for?

Page 14465, L15 – 17: Figure 4 is introduced but not explained. A brief introduction of absorption maps and how they are utilised in this manuscript is appropriate here.

Page 14467, L21: 'shows' not 'show'

I suggest the x-axis of figures 1 -3 are shown with the same size range for ease of visual comparison.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14455, 2011.