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Comment

Interactive comment on “Mass spectrometry of refractory black carbon particles from six sources: carbon-cluster and oxygenated ions” by J. C. Corbin et al.

Anonymous Referee #1

Received and published: 17 December 2013

This paper presents the application of the new Soot Particle AMS to several black carbon (BC) standards and details measured differences between them, with particular attention to investigating the relative peak sizes of the carbon and carbon-oxygen ions produced by the instrument.

While BC is a highly relevant field of study and much of the recent advances in understanding have occurred through advances in instrumentation, this paper is very close to what I would consider to be having too much of a technical angle for ACP, as the main theme seems to be the characterisation of the SPAMS as an instrument, which would be more within AMT's scope. However, as detailed in the comments below, I think that

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with a modest change of emphasis and a slight expansion of the CO_x discussion, it can be brought more in-scope. There are other shortcomings, but these mainly relate to technical details. I therefore recommend that this be published subject to revisions. At this stage, I'm minded to classify them as 'major' revisions because of the potential scope of some of the modifications I envisage.

General comments:

As stated above, the paper could be brought more into ACP's scope if it included more regarding the relevance to atmospheric science. While there is some discussion of the atmospheric significance of the functionalization, this is effectively buried within the discussion at the bottom of page 27579. As far as I can tell, the biggest direct link between this work and atmospheric aerosols is the aircraft engine work and to a lesser extent, the CAST source (which while not being identical to atmospheric sources of soot, is at least a combustion source). Minus these, I would have considered recommending this be resubmitted to AMT, or at least qualified as a 'technical note'. But with these included, the paper potentially offers new insights into the composition of atmospheric rBC and can authoritatively comment on the relevance of a number of commonly-used analogues, so I could still consider it in-scope for ACP. All this said, I would still recommend that the atmospheric implications be emphasised more within the paper. Currently, the conclusions are entirely technical, the abstract has a single speculative sentence at the end regarding the atmosphere and the introduction does not really spell out the motivation for this work beyond the ongoing development of the SPAMS as a potential source apportionment tool. I would suggest that the relevance of this work to the atmosphere be more explicitly stated in all three places, detailing the new understanding gained.

Further to this point, I would consider one of the major atmospheric implications of this work to be the reported observations of functionalization of the particles. The results presented seem to be mainly from the RB particles and a systematic comparison of the CO and CO₂ content of the different soot sources relative to the C_x peaks seems

to be absent, which I would consider to be a major oversight, even if it could only be considered qualitative at this stage. The authors mention that it is present in the CAST soot without presenting any graphs and speculate that this could be extended to atmospheric soot particles, and yet don't bring the jet engine particles into the discussion. Given the interest in aviation particles from the IN perspective, this seems to be a bit of an omission. For the sake of making the paper more atmospherically relevant (and satisfying my own curiosity), I would strongly recommend that a comparison of the Cx/COx ratios is included in a manner similar to figure 4.

I do not see where the SP2, APM or DMA2 fit into the results presented in this manuscript. If these instruments were not required to produce the data used in this paper, there is no point even introducing them.

Specific comments:

Page 27566: No reference here is made in the introduction to the fullerene signals reported elsewhere in the literature. This should be mentioned here.

Page 27566, line 25: The authors should expand on what they mean by 'filter system'.

Page 27566, line 9: The custom-built DMA had an inadequate description, as the Widensohler reference isn't specific to an individual DMA geometry. I'm left to assume that it is of the Vienna design (on the grounds that most European home builds are), which if it is the case, the authors should cite an appropriate paper (e.g. Winklmayr).

Page 27566, line 15: I note that the flow ratio is 2.5:1, which is very far removed from the nominal 10:1 of both Vienna and TSI DMAs. Given that this is a departure from standard operating conditions, the authors should discuss what effect this has on the data.

Page 27572, line 15: The authors need to explain what they mean here better. Do they mean that the ions themselves are in the form of fullerenes, or that they originate from fullerenes in the particles, or both? Could graphitic material in the BC also be

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responsible for making fullerene ions at the point of vaporisation?

Page 27577: I can think of alternative explanations for the tail on the m/z 36 distribution. It could be that some of the particles are not completely vaporising, but enough chemical bonds within the BC are broken such that these clusters can be released after hitting the vaporiser surface, or that some particles that are not being vaporised are bouncing off the surface of the vaporiser and back into the laser beam. These could be tested by comparing with data with the vaporiser removed (I am assuming that there is an abundance of RB data both at ETH and Aerodyne).

Supplement: The first section of the supplementary material, while possibly useful to those not familiar to the instrument, really just paraphrases what is already in Onasch et al. and the main manuscript. I suggest that this is tightened up.

Technical corrections:

Line 27564, line 3: I would qualify the statement about combustion particles being the second 'strongest' climate forcing agent as 'in terms of instantaneous radiative forcing' and specify that the statement refers to the BC specifically. The latest IPCC report (amongst other sources) is quick to point out that combustion also produces OM, which can offset or reverse the warming effect of BC and as aerosols are very short-lived, the long-term forcing potential is not significant compared to other agents. I would also question the wording of the point on line 13 identifying combustion as 'ideal candidates for near-term climate mitigation'. Their large radiative forcing makes them ideal candidates for mitigation, but their short lifetime means that the benefit of mitigation will only be felt in the near term if CO₂ emissions continue to increase.

Page 27565, line 5 (and elsewhere): The word 'vaporize' and its derivatives should be either spelled 'vaporise' or 'vaporize'.

Page 27566, line 4: The SP2 strictly quantifies according to the amount of incandescent material, which just happens to the refractory, light-absorbing component in the

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case of rBC. It can also detect the non-BC component in its effect on the scattering cross section but admittedly does not measure the composition.

Page 27568, line 1: The AMS vapouriser cone should be specified to be an inverted cone

Page 27568, line 12: The laser should also be described as ‘active cavity’, as this is a key design feature.

Page 27568, line 29: The mass spectrometer mode should be referred to as ‘V mode’, as this is the commonly used term.

Page 27569: Please provide some references for CAST source, in terms of technical description and characterisation.

Page 27574, line 16: Given that Tim Onasch is a co-author on this paper, it seems inappropriate to cite a personal communication from him. If the measurements were performed at Aerodyne Research, this should be simply stated as such.

Page 27577, line 10: I would not agree that the m/z 36 distribution is bimodal. It certainly has a ‘tail’, but I see no second mode.

Figure 3: This figure would be clearer in colour.

Figure 6: The symbols and line styles should be included as a legend rather than described in the caption. If the authors are pushed for space, the arrows are surplus to requirements because the respective axes are indicated by the m/z referred to.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 27561, 2013.

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