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Interactive comment on "A new aerosol collector for on-line analysis of particulate organic matter: the Aerosol Collection Module (ACM)" by T. Hohaus et al.

Anonymous Referee #4

Received and published: 26 May 2010

General Comments: The manuscript by Hohaus et al. entitled "A new aerosol collector for on-line analysis of particulate organic matter: the Aerosol Collection Module (ACM)" describes a newly developed instrument and related initial characterization. The general concept of the instrument is certainly good, and will be useful for investigation of ambient and laboratory aerosol. Several aspects of the manuscript are either confusing or somewhat poorly worded. The authors need to clarify several aspects of the discussion accordingly. I discuss below some points that I think should be addressed before publication, some of which have also been addressed by the other reviewers as well. After these points have been clarified, I recommend publication of the manuscript in AMT.

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Specific Comments:

(1) The manuscript bills the technique in several places as 'continuous' and 'on-line.' The system is clearly limited to certain lower limit of collection period, however, and this time is long enough to make me hesitant to agree to the terms the authors apply. For example, the collection time for the beta-pinene experiment was 60-minutes, and 6-hours for the ambient study. I understand this reasoning has inherent flaws, since every instrument has dead time associated with detection. Having collection periods with timescales on the order of hours, however, strikes me as much beyond the appropriate (albeit arbitrary) threshold to be fully 'online'. I agree with the term "quasi-online," used only once (in the abstract) and suggest that this terminology be used through-out the text and in the title as well.

(2) The authors discuss the ACM as a new aerosol collector capable of being applied to a number of different types of detectors, but then exclusively apply it to the GC-MS/FID detector. I think it would be more straight-forward to discuss the technique from the beginning as the ACM-GC-MS, and then say as a part of future work that the front-end ACM part may be able to be combined with other detectors. As it is written, however, the manuscript seems to promise more universality than it tries to demonstrate.

(3) Several places within the manuscript lack sufficient references and comparison to previous work. For example, there is no discussion of other particle collection and analysis systems that utilize a cooled plate that is later heated for aerosol desorption (e.g. TD-PB-MS, Tobias et al., 2000). While the obvious difference is that the ACM discussed here can theoretically be applied to a number of different detectors, it would still be generally useful to discuss the inherent advantages and disadvantages that it offers with respect to similar techniques that utilize cold aerosol collection for quasionline detection (with appropriate citations).

(4) The ACM relies on the fact that the collected aerosol is: (a) un-perturbed by the collection process, (b) collected efficiently, and (c) volatile enough to desorb at the

temperatures supplied. These assumptions may be appropriate in many cases, but will not always be the case. I have associated questions with these assumptions that are not discussed in the manuscript. For example:

(4a) How much will semi-volatile aerosol evaporate in the reduced pressure of the instrument before collection, and therefore bias the resultant measurements?

(4b) Will the particle beam always be reliably focused such that all particle will hit the collection area (how does the set-up differ from the AMS geometry, for example)? How likely are particles to avoid collection due to their ability to bounce off the surface (this has been a topic of discussion with the AMS instrument)? What other factors significantly influence the fraction of particles that are collected?

(4c) The particle material adsorbed to the collector will only adsorb if it has sufficient volatility at the desorption temperature (e.g. 225oC). How much material will remain undetected in this case? It will clearly depend on the aerosol composition, but this will introduce a significant bias into the measured results.

While this initial manuscript need not exhaustively answer all these questions, the fact that these (and clearly other) assumptions exist should at least be mentioned in passing, and reference to other groups that have looked into some of these issues should be given. This will aid the reader in understanding how to interpret data from the ACM-GC-MS, and will also aid future development of the instrument.

(5) The manuscript describes the instrument as discusses as "capable of linear, quantitative measurements of ambient aerosol mass loadings." This seems to be an overstatement if at least some mass must be missing due to the fact that it will not desorb at the temperatures used, and because the GC-MS is able speciate far less than 100% of the compounds introduced. One proof given for this statement is the Figure 9 comparison with the AMS, but in this figure the integrated ACM-GC-MS peak area is given in arbitrary units. I do not doubt that in many circumstances the ACM-GC-MS will scale with the AMS linearly, but this does not mean that the instrument discussed here is

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able to quantitatively report mass loadings – and certainly not without being rigorously "calibrated" with another instrument such as the AMS (which would have a number of uncertainties that would need to be mention in a forthcoming manner). Furthermore, any chemical aerosol detector misses certain types of particles (AMS included: low volatility organics and other refractory material) and this always must be taken into account.

(6) The discussion of the use of FID and MS as detectors was somewhat confusing to me. I realize that the FID detector was also connected within the GC-MS system, but the authors should use more clarity in going back and forth between the two. This adds a bit to the confusion of the fact that the ACM is already de-coupled from the detector.

(7) Minor points/questions

(7a) Page 1365: The description of the ACM particle collection system describes what seems to be narrow cone where the particles are trapped. Is this the appropriate mental image of the collection surface? A small diagram inset in the instrument schematic, or in the supplementary online material, might help clarify this.

(7b) Page 1366, Line 18: What is 'helium 6.0'?

(7c) Page 1372, Line 24: What is Linde, 4.8? Does the 4.8 refer to the company part number? If nothing else the references to the Linde gas products should be consistent (with or without registered trademark sign; with or without comma between 'Linde' and number; etc.).

(7d) Page 1372, Line 27: The authors state that the concentrations of aerosol precursor and ozone were "orders of magnitude above ambient levels." They justify why this is, but in the next paragraph. For clarity of reading I think it would be helpful to move part of the justification to immediately after this initial sentence.

(7e) Page 1373, Line 14: Define CPC

(7f) Page 1373, Line 17: Define SMPS

(7g) Page 1373, Line 25: Add "to prevent re-condensation of gases onto valve and lines."

(7h) Page 1378: What types of air masses does Billerica typically see? The authors state that "Similar chromatograms from urban samplings were also observed ...". Does this imply that the Billerica sampling location is a 'typical urban environment'?

(7i) Places within the manuscript lack consistent use of grammatical tense (present and past). Please be careful to refer to the instrument and experiments consistently.

Technical Corrections:

- (1) Page 1368, Line 4: Line-break should be "cryo-focusing" not "cry-ofocusing".
- (2) Page 1372, Line 21: Remove "which is"
- (3) Page 1372, Line 23: "content" should be "contents"
- (4) Page 1372, Line 27: Change "are" to "were"
- (5) Page 1373, Line 17: No comma after TSI in parentheses
- (6) Page 1374, Line 17: Move "filter" to after "(HEPA)"
- (7) Page 1377, Line 8: Change "taken" to "collected"
- (8) Page 1378, Line 7: Change "is" to "was"

(9) Figure 2: Arrows in figure are too small, especially the directional arrows indicating direction of flow from blue/green box. I would suggest adding the caption of mode state (e.g. 'Standy/backflush') to the figure itself. This would make glancing at the figure much more meaningful. Also, 'particle' is misspelled in the last sentence of the caption.

(10) Figure 4: Shift fit coefficients in last two lines of figure legend one tab to right so that the equal signs line up vertically. Remove "the different" from last sentence of caption.

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(11) Figure 5: Remove '26.11.08 11:18' from legend unless it is crucial for the reader to know the time/date the experiment was performed. Also add "expected" to retention time of alkanes so that the reader knows that this was not measured by the authors.

(12) Figures 6, 7: Time should start with zero and be in minutes. This would make reading the graph much more useful (again, it does not matter that the experiment took place on 28.11.2008).

(13) In general, please be consistent with font size and whether a box around legend is show. Figure 8, for example, has no box, while all the other figures do.

References:

Tobias, H. J., Kooiman, P. M., Docherty, K. S. and Ziemann, P. J.: Real-time chemical analysis of organic aerosols using a thermal desorption particle beam mass spectrometer, Aerosol Science and Technology, 33, 170-190, 2000.

Interactive comment on Atmos. Meas. Tech. Discuss., 3, 1361, 2010.