Atmospheric
Measurement
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Discussions

Interactive comment on "Single particle measurements of bouncing particles and in-situ collection efficiency from an airborne aerosol mass spectrometer (AMS) with light scattering detection" *by* Jin Liao et al.

Anonymous Referee #1

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This work represents an important and unique data set that provides a deeper insight into AMS collection efficiency. The authors use airborne measurements made with a light scattering time-of-flight aerosol mass spectrometer (LS-AMS) to draw conclusions about particle bounce and how differences in vaporization processes affect collection efficiency and AMS particle size distributions. This analysis is very valuable to the community of AMS users who frequently must deal with issues related to collection efficiency. Overall the manuscript is of good quality, but the following comments should be addressed before publication.

General Comments:

(1) If the lowered sensitivity of the CToF-AMS during Senex was significant enough to impact the CE (i.e., a substantial fraction of low signals were missed) how do the results of this work, in terms of low CE (e.g., < 0.5), relate to AMS measurements in general? Can we expected that CE is often below 0.5 in other measurements or is this largely an artefact of the low instrument sensitivity?

(2) The filament is hotter than the vaporizer, and is very close to the ion chamber. Do we expect that some areas of the ion chamber are actually hotter than the vaporizer? If so, how does this impact the authors hypothesis that differences in prompt and delayed spectra arise from particles vaporizing at lower temperature or fewer collisions of gas-phase molecules with hot surfaces.

(3) Wording in this manuscript is generally confusing. Problems with the structure of this paper and difficulties with grammar tend to obscures meaning in the manuscript. The Results and Discussion section in particular would benefit from the use of focused topic sentences to guide the reader through the sections and paragraphs. At present it is very difficult for the reader to discern where the discussion is going. More descriptive subsection titles might help in this effort.

(4) The notation used to indicate the different d_{va} types is somewhat confusing. Could d_{va-MS} and d_{va-LS} be used instead?

(5) Various acronyms are used to describe the instrument throughout the paper (LS, LSSP, LS-AMS, LSSP-AMS etc.). The authors would gain some clarity by choosing only one acronym, or by explicitly stating the reasons for using different ones.

Specific Comments:

(1) Abstract:

P1 L18-20: "The individual particle mass from the spectra is proportional to the mass derived from the vacuum aerodynamic diameter determined by the light scattering signals (d_{va-LS}) rather than the traditional particle time-of-flight (PToF) size (d_{va})." Do the authors mean that the particle mass from single particle spectra is proportional to the LS d_{va}, but not to the pToF d_{va}? It is not clear.
P1 L20: "The delayed particles capture about 80% of the total chemical mass compared to prompt ones." Do the authors mean that delayed particles capture 80% of the mass while prompt ones capture 20P1 L24-25: Change to "..especially at larger sizes."
P1 L27-28: Can this be clarified? The measured and calculated CE seemed to agree well when CE > 0.5, but when the calculated (assumed) CE was 0.5 the measured CE was often lower.

(2) Introduction:

P2 L9: The AMS has also been used to quantify sea-salt.
P2 L9-10: I doubt that mass accuracy (usually referring to the accuracy of a particular mass-to-charge ratio) is what is meant here. Please re-phrase for clarity.
P2 L15: " Not all particles introduced to the inlet are vaporized and ionized." Since the particles are not ionized, rather the resulting gas-phase molecules are ionized, I suggest this is re-phrased.
P2 L16: Do the authors wish to suggest here that the CE is the largest source of uncertainty in AMS measurements? This should be referenced and discussed with

more depth.
P2 L18: It could be a bit misleading to simply state the range in CE as 0.3-1. A discussion of the actual distribution of values presented in Middlebrook 2012 would be more informative. How frequently did Middlebrook 2012 see CE <0.5? > 0.5? A more accurate discussion of the magnitude of likely uncertainty is needed here.
P2 L25-28: The paragraph would benefit from an explicit discussion of why an in situ CE is better than comparing to other instruments like the PILS-IC and UHSAS.
P2 L31-32: The description of the LS-derived CE is made needlessly confusing here. Please re-phrase.
P3 L1: Are these the only studies that have applied the LS module to measure CE? "…was about…" Are these the mean values reported by these authors? If so, please state this.
P3 L3: delete "there"
P3 L4-5: change to "…the capability of the LS-AMS to capture CE variations."
P3 L14: Be consistent on the use of LS-AMS and LS-ToF-AMS. LS-ToF-AMS has not been introduced up to this point.
P3 L19: CE, not "The CE…"
P3 L20: change to "particles bouncing on the vaporizer"
P3 L21-22: "delayed" particles have not been introduced yet. It would be more clear to say something like "inefficiently collected particles" or "particles with less then unity collection efficiency." Also, it is not clear what "traditional" refers to.
P3 L22: (and other instances) Is "size resolved mass distribution" the best way to describe pToF data? It appears redundant.

(3) Methods:

P4 L24: Can the authors describe the implications of this non-linearity in sensitivity to small signals?

P5 L14: Where there frequently more than once mass spectrum per particle? Could the authors include representative or averaged particle event profiles in terms of summed signal over time?

P6 L9: How is this known?

P7 L6-17: Were these frag table corrections applied to LSSP mass spectra? Or, were these applied to MS mode only, in order to calculate CE using the CDCE algorithm?

(4) Results and Discussion:

P8-9/S3.1.1: The point of this section seems to be that delayed particles are likely the result of bouncing in the vaporizer. This is important for the reader to understand and it would really help the reader if the authors stated the major conclusion of this section at some point, either at the beginning or the end of the section. At present it is buried in this middle.

P8 L13: "...as expected.." It would help the reader to state why this is expected.

P8 L17: perhaps state which measure of d_{va} is referred to here

P8 L18-20 (Figure 2): Plots of summed MS signal (minus air) versus d_{va} would fit well in figure 2 and with the discussion of detection limit for a single particle, and don't seem to be in the paper at all. This plot should be coloured in a similar manner for Figure2a, showing the particle events above and below the MS detection limit

P8 L20: "about 36 ions" seems vague, is there a measured uncertainty in the single ion signal that can be converted into a range of ions per spectra here?

P8 L32: change to "subsequently vaporize off..."

P8 L33: should this read (NH4)2SO4 vaporizes in less than 50us?

P9 L1: rephrase to: "...,with no measurable bounce, does not produce delayed particle events"

P9 L1: Except for the reference to the figure, it is not clear here that the authors refer to their own laboratory data. This is a grammar issue.

P9 L 1-5: What are the velocities of particles over the size range we expect? Much larger than this, but not obvious to the reader. What fraction of particles would have this velocity?

P9 L17-18: This is a confusing sentence. Consider re-phrasing or breaking into two sentences. Reference to Figure2b would be useful here, too.

P9 L20-22: This sentence seems to repeat the information given in L17-18. Also, it is not clear what is meant by "...which are also defined by the time differences in the Figure1 histograms." What is being defined here? As far as I understand, Figure2B is used to identify prompt and delayed particles.

P9 L25: What exactly are these improvements and how do these definitions differ? This needs some explanation, rather than just referring to the table. It would help the reader to state exactly why the definition of Cross 2009 would result in more delayed particles.

P9 L30: It would be useful the remind the reader here that the "prompt + delayed fraction" represents the fraction of particles that are detected by the mass spectrometer. This quantity could be affected by (1) particle bounce or (2) below detection limit number of ions. (1) and (2) are both impacted by particle composition and phase, as well as instrument sensitivity, so it is unclear to me why we should expect very similar fractions between studies. The various factors affecting prompt+delayed fractions need to be explained more clearly than is currently done in P8 L31-32 and P10 L1-13.

P10 L9: kinetic momentum or kinetic energy?

P10 18-19: What might this mechanism be? It would be more helpful to the reader if the authors summarized their hypothesis here before embarking on a long discussion.

P10 L21: Different physical properties that resulted in different bouncing characteristics?

P10 L23-25: This sentence is very difficult to follow.

P10 L23-28: Do the individual particle event profiles (i.e., ions in an individual mass spectrum versus time) collected from this laboratory data (and/or field data?) show longer evaporation times for delayed particles? Could the authors collect laboratory

data at a higher frequency to allow for this analysis?

P11 L 7-8: Is this because delayed particles are evaporating at a lower temperature? This section could be re-organized to make the conclusion more clear

P11 L11: "processes for" is really unclear. Do the authors mean "processes during detection" or "processes at the vaporizer"?

P11 L11-12: Since the filament is hotter than the vaporizer, what does this tell us about the vaporization of delayed particles? do delayed particles generally show lower total ion signals?

P11 L15: Canagaratna et al., JPCA, 2015 (DOI: 10.1021/jp510711u) could provide some supporting discussion here about vaporization temperature and fragmentation. At present this discussion seems to lack references.

P12 L1-5: Is a simplified UMR fragmentation table being applied to LSSP data to derive total sulphate and organic masses?

P13/S 3.2.1: Despite being observed previously, that the correlation in Figure 6 shows the LSSP data to be representative of the bulk relative composition is notable since all sizes are not detected with the same efficiency in LSSP mode.

P13 L13: What is meant by "slightly independent" is ambiguous

P13 L26: define UHSAS here

P13 L27: Did the authors measure transmission in their lens? Lens transmission as illustrated in Liu et al., may not be representative (or expected) for most AMS lenses. See this User's Meeting presentation: http://cires1.colorado.edu/jimenez-group/UsrMtgs/UsersMtg16/CampuzanoJostLensAndePToF-AMSUsersMeeting2015.pdf

P14 L6: Please be specific about what "these two" are

P14 L7: "Uncertainty by including..." should be "Uncertainty introduced by including..."?

P14 L9: please refer to a specific section

P14 L28: It might be helpful to briefly define mass-based CE here with a brief reference to the size distributions

P15 L3: Was this the case during SENEX?

P15 S3.3.2: What causes CE <0.5? Other factors beyond RH and acidity? Did chemical composition or other parameters vary more significantly with CE on the July 6, 2013 flight relative to other flights?

(5) Conclusions:

P16 L7: change to: ..."and uses these measurements to investigate the AMS CE in situ."

P16 L11: What does "reduce" mean in the context?

P 16 L23-24: Under what conditions? Specifically low CE?

(6) Figures:

Figure 2: This figure would benefit from the addition of a total MS signal (or total non-air ions) versus d_{va}, coloured to indicate which particle events are below the detection limit. Prompt and delayed particles should also be indicated in this plot. Part (b): Please colour data in this figure to indicate which particle events are classified as prompt and delayed. I assume that null particles are not included in this figure, this could be explicitly stated.

Figure 7: This figure is missing labels (i.e., a, b, and c). That the difference between the d_{va-LS} and d_{va} size distributions arises because delayed particles are excluded from the d_{va-LS} distribution and not because d_{va-LS} somehow accounts for the delay time should be make clear here and in the text

(7) Supplement:

Table S1: Make it clear that these corrections are (presumably) applied to single particle data? What frag table/assumptions were applied to single particle data? How can you have a negative CO_2 correction?

Figure S2: There are times of significant discrepancy between the calculated CE and measured LS CE. Is there any more information that can explain this? What do the particle size distributions look like? How acidic are these particles? Is their mixing state changing substantially? Could the authors attempt to understand these differences?