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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 #2

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In their manuscript "Single particle measurements of bouncing particles and in-situ collection efficiency from an airborne aerosol mass spectrometer (AMS) with light scattering detection", Liao and coworkers use results from airborne measurements with an AMS with implemented LS detection to investigate effects associated with vaporization of individual particles. From these measurements the authors draw conclusions about particle bounce and vaporization in the AMS and how this affects collection efficiency correction factors or measurements of particle size distributions. With this approach and focus the manuscript is well suited for Atmospheric Measurement Techniques and I recommend publication of the manuscript AMT after the following issues were resolved.

Generally, the manuscript is well written. However, at places the authors should select wording that is scientifically more correct (see examples below). Furthermore, some of the conclusions regarding the processes within the ionizer chamber after bouncing of particles off the vaporizer are rather speculative and should be supported by additional evidence or rephrased. Most of the comments below, however, are more associated with clarity or correctness of presentation than with the content of the work.

Detailed comments:

P1L15-16: Not only particles larger than $\sim$250 nm scatter light, but also smaller ones. Rephrase.

P1L24: Delayed particles are detected later than "expected". I suggest rewording to something like "delayed particles are detected later than appropriate for their size" or "...later than expected from their measured velocity".

P1L26-27: "higher null fractions and corresponding lower CE for this study may have been related to the lower sensitivity of the AMS during SENEX". While the lower sensitivity of the AMS probably causes some of the single particles to produce insufficient signal for single particle identification, all the aerosol signal is probably measured in MS mode where mass spectra are averaged over large time intervals. While the missing of single particle counts using the LSSP mode results in smaller CE factors, the real CE factor should probably higher, thus this should result in a CE factor biased low. Is this correct? How does the "lower sensitivity" affect LSSP and MS mode measurements? How does this potentially affect m/z signals with low relative intensity?

P2L5: aerosol inhomogeneity is blamed on "short atmospheric lifetimes, compared to greenhouse gases". While it is correct that the lifetime of aerosol particles is short compared to that of greenhouse gases, this is irrelevant here. In this context it is more important that aerosol lifetimes are short compared to timescales of mixing processes.

P2L17: Here and in several other places in the manuscript CE is described as the

C2

fraction of mass or number of particles detected by AMS. Traditionally CE is defined as the fraction of mass of particles detected by the AMS. As shown in Figure 3, the bouncing fraction of particles is a function of particle size. Therefore using only the fraction of number of particles that are detected will likely introduce a bias.

P2L24: "bouncing on the vaporizer" should read "bouncing off the vaporizer".

P3L16-18: The sentence starting with "Many airborne ..." contains information that is not relevant here and should be removed.

P3L23: Section 2 ("Experimental") already includes a large number of results. Either the section should be renamed or the results moved into the results section.

P4L11 (and many other places): the unit "bits-ns" should read "bits*ns" since it is the product of the hight (bits) and the width (ns) of the peaks in the ToF mass spectra.

P4L25: Between "as" and "instruments" a word is missing, either "of" or "for".

P4L28: The LS-AMS is not capable of measuring "single particle mass" but either "single particle composition" or "non-refractory single particle mass".

P5L2-3: What does "LS-triggered particle scattered light signal" mean? Rephrase.

P5L12: "the corresponding scattered light signal" should probably be "the corresponding light scattering signal".

P5L13-14: Is there any background subtraction for the mass spectra applied for single particle information?

P5L16-17: "The cases ... are called coincident particles." This does not make sense. Rephrase.

P5L27: Replace "the third measurement of ..." with "the third type of measurement of ..."

P6L5: "... an additional brief amount of time ...". Can you provide typical numbers for

this?

P6L7: What do you mean with "different signal paths"?

P6L8: "the slowest of these processes and largest uncertainty . . .". Can you provide an estimate on how long this is and how large uncertainty is?

P6L11-30: In these paragraphs there are several statements that should be phrased more correctly, e.g. "The second method . . . is the vacuum aerodynamic diameter . . .", ". . . calibrated with known particles . . .", ". . . where the calculated particle velocity uses . . .", or ". . . that the laser not only counts . . . but also counts . . ." (the laser does not count, it can be used to count).

P6L22: How can the maximum of the Gaussian distribution be before Toffset, which is defined as the mean of the distribution?

P6L26-28: Why is dva-LS calculated from the laboratory calibration of dva and not calibrated individually?

P7L1: What do you mean with "compared to MS mode"? Reword.

P7L11: The IE for nitrate is claimed to be 7E-7 ions/g. This unit is wrong – or the IE of this instrument is orders of magnitude lower compared to other AMSs.

P8L4 (and many other locations in the text): A clear differentiation between "signal" and "signal intensity" (i.e. the magnitude of a signal) should be made throughout the text. Similarly, wording like "maximum light scattering intensity" (P8L10) should be replaced by something like "maximum light scattering signal intensity" since not the intensity of the scattered light but of the signal from the measurement of the scattered light is meant.

P8L12: "criteria" should be "criterium"

P8L14-15: The light scattering intensities do not vary over a significant range because the laser beam is broad, but because the beam intensity has a shape (likely Gaussian

shape) across its cross section.

P8L24: "axis" should be "axes". What is a "mass spectrum arrival time"?

P8L31-P9L6: The "delayed particles" were explained by particles that bounce off the vaporizer and vaporize at other, colder surfaces. To explain the time delays observed in the measurements the particles have to travel with very low velocities after bouncing off the vaporizer. This explanation is very speculative. Do the authors have any evidence for this? I could imagine if the particles bounce of the vaporizer in such a way that they have to loose most of their energy, they must be strongly deformed during the bounce process which I could imagine results in good contact with the vaporizer. Therefore I would expect that they stick to the vaporizer like ammonium nitrate and other liquid particles do. What do the authors suggest as the process that causes a loss of the vast majority of kinetic energy of the particles and at the same time avoid sticking of the particles to the vaporizer?

P9L8-15: This paragraph is very speculative. Do the authors have any evidence for this behavior of the particles?

P10L1-2: "The single particle mass is near the chemical detection limit . . .". For which size of particles is the mass near the LOD?

P10L1-12: According to this discussion appearance of a particle as a "null" particle is not a proof or hint of particle bounce but for a large fraction of the particles (most of the mass measured with an AMS is typically in the 100-400 nm range) could as well simply be due to low particle signal. This should be discussed when discussing bounce.

P10L26: Add "for" between "as" and "the SENEX".

P10L30-P11L8: Without more information on how detector sensitivity affects data acquisition and without any interpretation or discussion of the differences in the mass spectra observed at different detector sensitivities, this paragraph does not provide very helpful information.

P11L15: "The location where the delayed particles impact and vaporize is likely further away from the vaporizer center . . .". Do the authors mean that delayed particles impact near the edge of the vaporizer? In this case a shift towards more delayed particles should be observed when scanning the particle beam across the vaporizer. Furthermore, not every location further away of the vaporizer is colder than the vaporizer. The hottest location in the ion source is the filament. Be more clear here.

P11L10-23: Here the different chemical signal (e.g. less fragmentation) of the delayed particles is explained by vaporization of these particles off cooler surfaces. Where do the authors assume that these surfaces are? In the AMS ionizer the hottest point is the filament, not the vaporizer. Therefore some of the other surfaces might be even hotter than the vaporizer. If particles vaporize from other colder surfaces one would expect longer vaporization times for the individual particles. The authors report that this is not observed and explain this by the large range of vaporization temperatures where the vaporization time is not extended (400-800 °C). Which vaporization temperature would be expected to obtain the observed changes in the fragmentation pattern? Is this consistent with this range of vaporization temperatures for quick vaporization?

P11L25: "mass spectral peak widths" sounds like the width of the peaks in the mass spectra while the duration of particle vaporization is meant. Be clearer.

P12L8-12: The AMS ionization efficiency strongly depends on the exact location of the vaporizer within the ion source. If the vaporizer (i.e. the location from where the evolved vapor originates) is moved, IE drops strongly. Therefore I would expect strongly reduced ion signals (i.e. measured particle mass) from particles vaporizing at a completely different location within the ion source. This would not be in agreement with the observations made in this work and the suggested processes occurring in the ion source. Furthermore, in section 3.1.4 I miss a discussion on the reasons for the missing mass in the delayed particles. Is there a different composition for the delayed and prompt particles? In addition: how certain is it that the null particles are also of the same composition as the measured ones? Is the loss in particle mass different for

different instruments or what influence does the large difference in the criterion for MS detection (see Table 1) have?

P12L14-27: With about 10% black carbon content in the fine aerosol observed in Mexico City it is hard to believe that the much lower measured delayed particle mass (less than half of the prompt ones) observed in this study is caused by larger black carbon content during that measurement. What was the BC content during this study?

P12L28: The title of section 3.2 is not very well chosen. Most of the findings presented here are not observations using the AMS LSSP mode but are still characterization of the LSSP and PTOF measurements.

P13L16: Since here general ways to generate mass distributions from AMS instruments with LS module are described it is not appropriate to use an effective particle density of 1.55 g/cm3. This is valid for this data set only.

P13L25: Define "UHSAS".

P14L8: The ratio between the AMS PTOF and the other distributions for large particle sizes is not constantly four as claimed here, but varies strongly.

P14L12: The delayed particles do not only create a bias towards the larger size end of the mass distribution, but as shown in Figure 1 can also leak into the next chopper cycle where they contribute to the lower size end of the distribution and increase the measured background, i.e. lower the effectively measured distribution.

P14L24ff: In section 3.3.2 I miss a discussion on the potential influence of the mixture state of the particles and size-dependent composition on determined CE factors. While the traditional CE factor is defined on a mass basis, using the LS a number-based CE factor (which is partially transformed into a mass-based CE) is used. This relies on the assumption that all particles (the bounced and the detected ones) have the same composition – even for particles smaller than the lower particle size cut-off of the LSP. Externally mixed aerosol should result in different fractions of bouncing particles (e.g.

pure ammonium nitrate and other liquid particles would not bounce while the majority of pure ammonium sulfate particles would bounce). This should at least be discussed.

P15L10: In this context not the PTOF but the MS mode is relevant.

P15L6-24: In Figure 8 the mass and number-based LSSP CE factors sometimes differ by much more than their given uncertainty. What could be the reason for this? Is the assumed uncertainty too small?

P16L7: The authors claim that "particle bouncing physics" was investigated. What kind of particle bouncing physics was measured in this work?

P16L14-16: Please provide clear information on the limitations of the method.

Figure 1/2/3/5/7/8: Present the units as recommended by IUPAP etc.: e.g. "Maximum Scattered Light Signal (V)" should be "Maximum Scattered Light Signal / V".

Figure 1: For the ammonium sulfate particles almost no time dependence of the frequency of delayed particles occurs. How can this be explained? Is this potentially an artefact? What is the "mixed composition"?

Figure 2/4: The labels "a)" and "b)" are very hard to see.

Figure 2, caption: make clear that not the scattered light signal but the intensity of the scattered light signal is shown. It should read "above 0.04 V (red points)", i.e. add "V".

Figure 8 (and Figure S2): Since the altitude at which the measurements were taken is never mentioned or discussed in the manuscript it could be removed from the graph to make it a bit less crowded.

Supplement P1L19: I suggest using the term "dead time" instead of "extra time".

P1L27: "multiply by chopper" should read "multiplied by the chopper".

P2, Table S1: Some of the coefficients change by almost a factor of 2. How large is the uncertainty of determination of these coefficients. Is it expected that they change over

the course of a flight? Would not an average number be the better choice?

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