Rebuttal to Reviewer 2

We thank the reviewer for the comments. The manuscript has been modified accordingly.

1. *Overall comments:*
   A: the work does not aim to clarify the behaviour of the atomiser with respect to flow rates. Instead, it produces data which can be compared to a model of the atomisation and evaporation process. We agree that the results may be of interest to spray drying, but the range of sizes (< 1 um) and operating conditions (<100 C) used is far from usual bulk spray drying processes.

2. *P2 - L27 – These references are not appropriate. There is a huge amount of literature out there on particle morphology development in hundreds of systems in the spray drying literature. This include organic solutes (l28,29 incorrect). However these tend to focus on larger particles than produce in a nebuliser. However there is significant work on the production of particles for inhalable drug delivery and this focusses on smaller particles, of a size which is similar to the nebuliser.*
   A: We thank the reviewer for the comments, as well as for the references further on. We added these references to the introduction, and particularised the study to the ELSD conditions

3. *P2 – L37. I don't think you present results at different temperatures.*
   A: The reference has been removed. Additional data and simulations as a function of temperature are in the companion paper – Part B.

4. *P2 -l44 – Bertrani reference, clarify – submitted or in preparation?*
   A: the papers have been submitted jointly for review ot the same journal, as Part A: experimental and Part B: model.

5. *P3 – Table 1 – define symbols*
   A: Symbols have been defined.

6. *P3 = L69 – Provide details of impingement diffuser.*
   A: The impingement diffuser is shown in Fig. 3. Further details can be found in the PhD thesis by Bertani (https://doi.org/10.17863/CAM.116176), and the reference has been added as (Bertani, 2024).

7. *ELSD – Please clarify: Is the whole thing heated to 25°C or just the evaporator section? What is the Y piece for? Is the total nitrogen flow into the Y-Piece -0.4 or 2l/min, if there is makeup nitrogen? How much nitrogen goes down each branch of Y? What is geometry of the nebuliser section?*
   A: The evaporator is kept at 25 C. The Y-piece provides for the impingement of larger droplets emerging from the nebuliser. The total nitrogen flow is 2 + 0.4 l/min, with make-up nitrogen injected at the 'dry gas injection point' as indicated in Fig. 3. All the gas flows through the outlet, as the lower leg of the Y is kept flooded. The description has been amended with these details.

8. *Fig 3 – thanks for this clear diagram, could you also add tube diameters.*
   A: details of the geometry are available in the companion paper (Bertani, 2025) and the PhD thesis (Bertani, 2024), but the geometric details of diameter and length of each section have been added to the description. A full CFD mesh is available in the PhD thesis.

9. *P6 - l13,14 please give details of liquid trap, what particle sizes are you aiming to remove?*
   A: The original design by the manufacturer did not have estimates of the cut-off sizes, but these have been determined from CFD simulations of droplet impingement to be of the order of 5 micrometers for the Y-piece and 1 micrometer for the diffuser. This is discussed in the companion modelling paper (Bertani et al, 2025).

10. *P7 – l138 , 'fl tends to 1' or equals 1 for dp > 2 um, in your analysis?*
    A: the losses tend asymptotically to 1. This has been edited.

11. *PDPA – can you give some details of the system and discuss the detection reliability and accuracy for the minimum particle size. Depends on wavelength, magnification, detection volume, beam angle, refractive index, focal distance etc. What is the minimum size that the system software reports?*
    A: The system was geometrically arranged to probe a volume of approximately 2 mm length and the smallest possible diameter corresponding to the index of refraction of the the fluids considered (water, DOS). This has been added to the text.

12. *P8- l162 The equation for the D32, how universal is this equation. Typically these equations are not that accurate.*
    A: Indeed, the authors have not been able to find an available equation in the literature for the range of conditions and nebulisers used, and we acknowledge that there may be inaccuracies, even though the nebuliser is of the same type. This remains an outstanding problem in the field. This has been added as a footnote.

13. *P9 equation (6) – looks like np,s is an absolute number distribution not a pdf per unit concentration of gas? Make this clear. Also can you discuss correction factor more.*
    A: n throughout the document represents a number concentration distribution per unit volume per unit size. The definition is made clear in the first occurrence, just below equation (1). The correction factor is simply to acknowledge that the different setups (spray, well-mixed PDPA) reflect the fact that the spray is not well mixed unlike the well mixed experiment, so that the local concentrations are different, even if the number distributions may be similar.

14. *P9_l181 replace 'detector' by 'ELSD' for clarity*
    A: We have replaced it with 'flow network'

15. *P9 – section 2.4 what was the expected size distribution? Is it cut off? How reproducible, why 90 seconds scans?*

A: The cut-off reflects the limits of the instrumentation for the SMPS, around 10 nm. We do not know a priori what the expected size distribution would be, and that is the whole point of the experiment. The measurements were very reproducible, both in different days and same day. The SMPS scanning experiments require 90 seconds to scan through the size distribution.

16. *Reproducibility – is the data presented in figure 6 a single experiment, or average of a number of experiments? (I don't think you say how long the experiment was done for, whether you see any drift over time or run to run, day to day variability in any of these setups. A reader would benefit from seeing some error estimates, to get a feel for the accuracy of the predictions you make. Graphs I believe the output of the PDPA will be in a histogram format, please plot points rather than a piecewise line. I expect it's the same for the AAC (just use small points if there is a lot of them). Plot fits as lines.*

    A: The measurements of size distribution (both SMPS and AAC) were each repeated 3 times and averaged, as stated. The peak concentration variability was negligible, even across multiple days; the averaging just reduced the noise of the data. Whereas the distributions out of the instruments are given as histograms, the fitted lines are much clearer when there are multiple lines on the plot.

    Error estimates of the PDPA are harder to estimate. Calibration measurements were made with a standard aerosoliser unit before deployment. Variability was neglibible across multiple days.

17. *I suggest changing your graphs from Excel as it does not look professional. Add the np.m etc in the legend.*
    A: Graphs were produced with Matlab. The y-axis correctly and clearly represents what is being shown.

18. *Fig 6. In figure title says 'normalised', what do you mean by this as they do not appear to be pdfs. Are they the straight data of have they been scaled at all? There is no discussion of the 'C' factor.*
    A: The data on the figure are nomalized by the peak of the concentrations measured with the well-mixed AAC. The correction factor C was obtained from the ratio in Eq. 1.

19. *Fig 7a is fl a continuous function or purely based on a division of the two curved at points? Clarify in text. And plot as points if it is, or equation that you have. Would it not be better to plot np,m calculated from nA,m by equation (1) so the fit can be seen rather than replotting the curves from Fig 6?*
    A: The factor f_L is determined from Eq. 1 as a function of diameter along with the factor C for the peak number. The factor f_L is shown on the same plot as the numbers for for clarity, but the numbers are the same as in Fig. 6.

20. *Fig 7b you have cut this off at 1 um, what range is ft defined over? Is the impact of ft on the mode taken into account? Comment on accuracy at small R. Adding error*

bars to PDPA results would be of value.
A: As discussed above, it is difficult to place error bars on PDPA measurements, but measurements were reproducible to within a few percent. However, measurements below 2 micrometers cannot be trusted, as the signal to noise ratio is too low. Therefore, measurements for f_T are only considered from 2 micrometers and above.

21. *The validity of equation 5 needs some discussion and its validity in these circumstances justified. It looks to be based on experiments on a commercial two fluid nozzle which produces droplets that are of a considerably larger size that produced by a nebuliser. The work of Kemp, see below, as I recall discusses the validity of the droplet-size correlation between sizes. I think they use the correlation by Thybo, which appears to be for smaller droplet sizes.*

    A: We thank the reviewer for the additional helpful references. Having examined these references, it is clear that they are generally relevant, but that the geometry of the nebuliser is quite different from the one used in this study. Therefore, we prefer to use the correlation generate in a study with an almost identical nebuliser.

22. *P12 l220 – bulk density? Do you mean absolute/true density of solid. Bulk density of powder is not important. And bulk density of liquid is essentially the same as water.*
    A: Yes, the bulk density means the solid density, so that equations (7) and (8) can be used after drying.

23. *Fig 9. Again what is accuracy of these measurements. Please give some metrics for these distributions, such as mean, median, d10, d90 etc.*
    A: The variability of the peak number measurements is within a less than one percent. The statistical properties are of limited interest for the present paper, compared to the full distribution, as these will be further used for comparison with a model of elastic scattering and signal in the companion paper.

24. *P13 l247 – the estimated modal sizes are for what concentrations, I suggest a table, comparing these, and a graph comparing a predicted log-normal distribution to the measured might be of value?*
    A: The distributions are not by log-normal, as can be seen in Fig. 9, RHS column, as the impingement changes the original droplet size and thus the final distribution. Only two analytes have well defined bulk densities in Table 1, (citric acid and caffeine), and the corresponding modes are cited on the text: “the mode diameters from the peaks of Fig. 9, yields final mode diameters of 46 nm and 52 nm for citric acid and caffeine respectively. The resulting values should be compared to the measured modes of 35 nm 65 nm, respectively according to values on Fig. Therefore, it does not make sense to compare the log normal means.

25. P13 l246-255 This argument needs development and refinement: You are giving one number when you have data for 4 concentrations, so why not present all the data?
    A: it is not clear if the reviewer means the peak for the initial spray droplet diameter, or the comparisons. In the case of the former, one needs to refer to the combined

PDPA/AAC measurements for the peak mode. For the measurements in Fig. 9, the modes cited are for concentrations of 1 g/L. Although a table for different concentrations could be produced, it is not very useful in this context; comparisons of measurements and simulations of the full distributions are available in the companion paper (Bertani, 2025).

26. *Let's have some error estimates for the size estimated from the droplet size distribution (assuming spherical particle with no porosity). Caffeine particles are not spherical, SMPS measurement makes this assumption so what size/diameter are we comparing, is there data on how shape changes SMPS measurements. Shape may also affect the losses in the ELSD comment on this? Absolute densities used are for what form of solid: polymorph, hydrate, amorphous? Include dextran data, estimate density if not measured, it does not have big impact on result as it is proportional to 1/cubic root density.*
    A: All particles are measured using the electrical mobility diameter as usual per SMPS. The SMPS is calibrated with spherical particles. Estimates of error are determined from repeatability, as discussed above. Estimates of wall losses through the detector past impingement section are negligible (see Bertani, 2024). The state in flight is not known, but it can be assumed that all particles are solid after drying, and the densities for the corresponding dried solid.

27. *Also you are assuming the particles are dry leaving the ELSD, but all evaporated water ends up in nitrogen stream so will increase humidity, please estimate this humidity.*
    A: The nitrogen dilution is sufficiently large that the total mass evaporated does not affect the concentration of water in the bulk mixture significantly. See the modelling paper (Bertani et al., 2025) and the PhD thesis (Bertani, 2024).

28. *P13 l254 I believe these dilute solutions will have surface tension very close to water, so unlikely to be important, why not look up and use in the equation?*
    A: Unclear which equation is being referred to, but the mode diameter in the section is only calculated from a mass balance, for which surface tension is irrelevant.

29. *P14 -15 It would be nice to see this discussion discuss crystallisation, or lack of, of these systems in a little more detail, and draw on the spray drying literature. Crystallisation in bulk solutions is a very different process, and can lead to a very different particle morphology. The references for citric for example are all solution crystallisation so we would expect their morphology to be quite different.*
    A: We thank the reviewer for the suggestion, but this is beyond the scope of the study for the operation of the ELSD.

30. *P15 l282 'Down- stream measurements reveal that analyte properties, particularly volatility and bulk density, significantly influence final particle size distributions.' This is really the solidification and crystallisation behaviour, which leads to different morphologies. You mention volatility of the solute, if this is important you should estimate how much of the if this is important you should estimate how much of the solute will evaporate and if this will change particle size estimate.*

A: We agree that the crystallisation process leads to different morphologies. Volatility is discussed in the companion modelling paper (Bertani et al., 2025), rather than the present paper where only the measurement results are presented.

31. *Throughout avoid 'ELSD detector' and just ELSD, after all the 'D' is detector.*
A: we have modified the manuscript accordingly.