

We thank the reviewers for the useful comments. We address them below, with replies in blue.

Reviewer 1

The current manuscript provides a comprehensive model for simulating aerosol dynamics and signal response in the evaporative light scattering detector (ELSD). The overall quality of the manuscript is very good and scientifically sound. The experimental and simulation results are in quite good agreement. Nevertheless, some scientific issues have to be addressed:

- What is the background behind Eq. (3), which seems to be an empirical correlation? How were required parameters obtained?

We thank the reviewer for the question. We have modified the relevant section to more clearly point to the experimental study, and to clarify the reason why the selected correlation is used, namely to accommodate the use of liquids of different properties used in the original measurements, compared to the solvents used in the actual ELSD experiment.

- Details of the ELSD measurements are missing or only very briefly described: How is the scattering geometry? Which scattering angles are used? What is the detection scheme (homodyne or heterodyne)? Figure 2 gives a very small sketch, but more details need to be mentioned in the text. In addition, I cannot see Eq. 47 which seems to be related to the ELSD method.

Details of the optical geometry are proprietary to the manufacturer, but the scattering is homodyne, with illumination across the flow axis, and collection into the detector at an angle of 60 degrees off the same. The particle concentration is assumed to be constant across the section. The comparisons of experiments and models are performed on an overall normalised basis regarding the total signal, so the absolute scattering signal obtained (from a larger or smaller collection angle) should not be material to the comparisons. A corresponding statement has been added around line 263.

We reproduced Eq. 37 (not 47) below in case something in the pdf document was not appropriately typeset. The equation represents total signal reaching the detector at the relevant angle, for all particle sizes in the distribution.

$$S(\theta) = N \int_0^{\infty} I(\theta, d_p) p(d_p) d(d_p)$$

- Are multiple scattering effects relevant for the measurements?

The mixture is fairly dilute (particle concentrations of $10^6/\text{cm}^3$, corresponding to particle volume fractions of 0.001, see Figs. 9-10, so multiple scattering was not considered.

- The authors stress out the relevance of temperature. However, it is not clear for me how the temperature of the droplets can be controlled or measured experimentally. A comment on this issue would help.

Thank you for the opportunity for clarification. The evaporator section and incoming gas is temperature controlled, but not the spray generation section. The temperatures referred on Fig. 12 are evaporator gas and wall temperatures in the full detector. The spray generator section run at ambient temperatures, and the main particle losses are assumed to be due to impingement rather than evaporation. Evaporation takes place in the evaporator tube, with a long residence time of the order of 0.5 seconds, and controlled temperatures. The droplet temperatures are not controlled. The model assumes that evaporation is controlled by the equations listed in the evaporator section 3.4.1, which details in Eq. 16 how the temperature of the droplet varies in time. The temperature of the gas T_g is set and controlled; the temperature of the droplet evolves in time. Eq. 26 shows how the vapour pressure of the droplet is calculated, where the temperature T_d of the droplet evolves quickly to that of the surrounding gas.

Equations 26 and 27 have been edited to clarify that T is the temperature of the surface of the (uniform temperature) droplet.

- An uncertainty statement or consideration for the experimental results is lacking so far. Such an evaluation is recommended to be performed for the property of interest.

We thank the reviewer for the comments. The experimental paper details the estimated uncertainties in concentration and size measurement: uncertainties in the mode of the distributions measured are small, of the order of 5 nanometer variability for the mode and less than 5% variability for the concentrations measured with AAC and SMPS. PDPA measurements of number vs diameter distributions showed variability of the curves of the order of 1-2% for 3 repeats. An additional statement has been added around line 313.

Furthermore, some typos or technical errors are given in the manuscript. For example, some numbers are missing. The figures are also partly not fully readable or understandable (see, e.g., Figures 6 and 7).

We thank the reviewer for the comments. Without specifics, we cannot check what numbers may be missing, but we have re-read the manuscript to look possible omissions. Regarding the figures: we do agree that the aspect ratio in Fig. 7 is not attractive. Nevertheless, the figure can be zoomed in with full resolution pdf. We have attempted to further improve the explanations (especially with ref. to Fig. 9), so the reader can follow.

Reviewer 2

This paper presents a computational fluid dynamics (CFD) simulation of an Evaporative Light Scattering Detector (ELSD). Although the study is well motivated and well written, several important issues should be addressed:

- It would be valuable to include a mesh convergence study, clearly specifying the parameters used.

We thank the reviewer for the question. As mentioned in the manuscript, the mesh size was iteratively reduced until further reductions no longer affected the convergence residuals of the final simulation. More specifically, a mesh

convergence study using 5 meshes in decreasing size was performed, and the first refinement beyond convergence was used.

- Since the authors employ the $k-\epsilon$ turbulence model, what is the maximum y^+ value obtained?

The particular y^+ parameter was not logged, but the boundary layer at the walls was sufficiently resolved for the current purposes of obtaining mean velocities (in the longer tubes), and spray impingement losses (in the atomising section).

- The $k-\epsilon$ model performs well away from walls, however, given the narrow geometry of the channels, wall effects may significantly influence the flow and particle behavior. The use of an alternative turbulence model, such as SST $k-\omega$, is recommended for comparison.

Alternatives are possible; the current model was chosen as computationally less expensive. Note that differences in k -epsilon and k -omega affect primarily the details of the rms of the flow, rather than the overall residence time characteristics. In the absence of flow validation, we would not be able to tell which model provides better accuracy.

- What are the R^2 values associated with the fitted curves shown in Fig. 8?

For the diffuser curves, the calculated R^2 for the whole curve is 0.72, and 0.95 for values where the efficiency (and Stokes) is larger than 0.5. For the Y-piece, the correlations are not as favourable, with R^2 for both for the whole curve and the values of $Stk > 0.5$ also around 0.51. This is a result of the difficulties in getting convergence of the statistics of droplets: a much larger number of simulations is required for full convergence.

- Was a dispersion model used for the Discrete Phase Model (DPM)?

No dispersion model was used; here the physics is governed by the high momentum and impingement of the droplets, rather than diffusive dispersion.

- Line 336: the Appendix citation is missing.

This has been removed.

- Line 378: the Figure citation is missing.

The figure references have been corrected.