Authors' response

Dear Referees,

Thank you for your comments to improve the manuscript. We have addressed your points and made the following changes to the manuscript. We believe that the manuscript is greatly improved and is now ready for publication in AMT.

Thank you.

Best regards,

Michael Rösch and Dan Cziczo

Referee 2:

"The manuscript keeps insinuating a "low cost" nebulizer (e.g. L44).. but does not provide any cost reference neither for the competition nor for the actual set-up. In terms of competition, depending on the particle range of the aerosols that are actually being generated, medical nebulizers are «100\$ with the actual disposable nebulizer being 3\$. Your 3D printed ones are not that cheap as they 1) require a 5K printer 2) the raw plastic material and 3)in your set-up a brass nozzle at 10\$.. Therefore it would be critical to have a more nuanced discussion. Also the commercial (expensive) devices might have a much better performance (see next point)."

We added the material price to print one PROTeGE plus the price for the brass nozzle to the manuscript for reference. In Addition, there are online print services that can 3D print parts and most universities now have fabrication labs or in-house 3D printing capabilities.

"The cost to produce one PROTeGE is around ~\$2.50 depending on the type of resin and the percentage of scaffolding used. The commercial brass nozzle costs <\$10, for a total PROTeGE cost under \$15. For users with no access to a 3D printer, it is also possible to upload the .stl file for PROTeGE (provided at no cost at https://www.thingiverse.com/thing:4444498) to an online print service. Pictures of

PROTeGE and post processing details for the instrument are also provided on the data repository."

"The data analysis of the particles generated is very superficial. Only focus is on number concentrations generated (again context of commercial and alternative systems is inexistent). There is no significant discussion in the manuscript on the actual distributions generated and their stability. Number concentrations is one aspect but what about distributions. Also the "heatmaps" provided the distributions are very confined to a small area and no effort was made to quantitatively analyze that data... e.g. how does the mode of the distribution changes over time.. or does not? What is the broadness of the distribution? any quantitative distribution metric and how this relates to commercial systems or applications."

Thank you for your detailed comment.

Both aspects particle number concentrations and the corresponding size distributions are described in the manuscript through the heat maps. One could also derive the broadness of the size distribution and how it changes over time from there. We performed an additional experiment using a SEMS instrument to detect the generated PNSD in the size range of 10 nm to 1000 nm. A comparison to other commercial systems is beyond the scope of this study as we are limited by instrumentation.

We changed the paragraph in the manuscript as follows:

"A time-series measurement of 420 seconds was performed for each of the four PSL samples. The obtained PNSDs showed particle number concentrations of ~10,000 cm⁻³ for 0.75 μ m PSL particles, ~1000 cm⁻³ for 1.5 μ m PSL particles, ~800 cm⁻³ for 2.0 μ m PSL particles to ~100 cm⁻³ for 5.0 μ m PSL particles (Fig. 3). All four investigated PSL samples showed a narrow PNSD except the 5.0 μ m PSL sample where a fraction of sub-micrometer particles was detected (Fig. 3d). This fraction of particles likely originates from the solution matrix in which the PSLs are suspended. Overall the generated PNSDs were stable over their measured period of time while only the 1.5 μ m PSL sample showed a slight decrease. These data show that the curved design of the chamber enables PROTeGE to disperse PSL particles with diameter up to 5.0 micrometers."

"Related the PSL "calibrations" seem disconnected to what can be achieved with the salt solutions?"

The PSL measurements/calibrations are essential for most end users. Our measurements show that PROTeGE can be used for multiple types of particles including the generation of PSL particles.

"All experiments except the comparison were done with the brass nozzle? This id stated a little bit as an aside given that the whole paper makes it sound as it whole nebulizer was 3D printed when in fact the most critical part (nozzle) was not.. but it was brass and purchased. One wonders why if the conclusion of figure 5 is that they are equivalent? Could you comment on this?"

Both nozzle types generate the same width of the size distribution with the only difference in their particle number concentration for certain bin sizes. Please see the changes we made in the manuscript based on your next comment.

"For the very least the discussion on figure 5 needs to be extended...Saying that they are similar is not true, there is a lot of difference (y axis is log) and it does matter."

We extended the discussion on Figure 5 and added the following paragraph to the manuscript:

"The generated total particle number concentration with the printed orifice was ~4,500 cm⁻³ versus ~2,400 cm⁻³ for the brass nozzle with minimal difference in PNSDs shape. The printed nozzle exhibited a broader shoulder from 0.3 μ m to 0.5 μ m with the higher particle number concentrations than the brass nozzle. The latter showed a decline with the highest particle number concentration at 0.3 μ m leveling out to 1.0 μ m. Overall, the width of both PNSDs ranged from 0.3 μ m to 1.0 μ m (Fig. 5)."

"Abstract: would be more informative to actually say what that the comparison with the brass orifice revealed rather than just say it was done. Results should be summarized in the abstract not just written what was done."

We added the following paragraphs to the abstract:

"A comparison of a 3D printed 0.5 mm orifice against a commercially available 0.5 mm brass orifice using the same ammonium sulfate solution was also performed. The particle number concentration generated with the printed orifice was higher, by $\sim 2x$, than the particle number concentration generated with the brass orifice."

"For the different concentrated ammonium sulfate solutions particle number concentrations from ~14,000 cm-3 for 0.1 g L^{-1} to 7,600 cm-3 for 5.0 g L^{-1} were measured."

"On the other hand the typical last paragraph of the introduction, where one typically says that will eb discussed in the manuscript is missing...Then again the first paragraph of the results (L112-115) is actually just that: saying what will be done...and this would belong as last paragraph of the intro. This is just an odd way of writing a manuscript."

We added a new paragraph to the end of the introduction:

"In the following two sections we describe how PROTeGE is designed and manufactured. The experimental setup and performance tests using different PSL and ammonium sulfate solutions is discussed. Three types of experiments were conducted to demonstrate the performance of PROTeGE: (1) an aerosol production experiment using four different sizes of PSL's, (Polysciences Inc., NIST traceable) ranging from 0.75 to 5.0 μ m, (2) experiments where different concentrations of ammonium sulfate solutions were dispersed and monitored over time with an optical particle sizer (OPS, Model 3330, TSI Inc.) and for the 0.6 g L⁻¹ also with a Scanning Electrical Mobility Spectrometer (SEMS, BMI Inc.), (3) an experiment comparing the performance of a printed 0.5 mm orifice to a 0.5 mm commercial brass orifice using the same ammonium sulfate solution of 0.6 g L⁻¹."

"L21 Please use comma for thousands to ease reading"

We changed all notations to comma for thousands for ease reading throughout the manuscript.

"Please be precise on the brass nozzle and diameter used. The cat number for McMaster-Carr shows orifices in inch (of various sizes)... Does the 0.5mm mean you used the 0.02 inch one?"

We provide information on the orifice in the manuscript and also on the part number to select the correct one as follows:

"The 0.5 mm orifice used in the comparison is a commercially available brass nozzle (Part Number 2943T887, McMaster-Carr) that is threaded into the pressurized air inlet of PROTeGE (Fig. 1d)."

"L122 use center dot as multiplication sign not a star"

We changed the multiplication sign to center dot throughout the manuscript.

"L120: please explain double distilled deionized.? Millipore systems do not distill? Where does tht DDI come from?"

Thank you for the indication. We corrected the term DDI in the manuscript according to the manufacturer as follows:

"Destilled De-ionized (DDI)"

"L149 "will be sufficient enough" can you be more quantitative.. what do you consider sufficient?"

We changed Line 149 to a more quantitative statement as follows:

" Overall, the generated particle number concentrations for the different tested ammonium sulfate solutions are high enough (> 1,000 cm⁻³) to operate particle size selection instruments downstream of PROTeGE, e.g. a differential mobility analyzer (DMA), assuming ~10% of the introduced particles are selected as monodisperse aerosol particles."

"Figure 1: what is the rationale behind the numbering.. why does (2) jump to pane d"

This was due to the numbering of the logical steps of the components of PROTeGE.

We changed the numbering in Fig. 1.

"Figure 4 there is no discussion at all why the time scale varies so much between panels a,b and c… Please discuss in the text what you want to show going from a) 1800sec to b) 450 sec to c) 20000 sec"

This was due to the fact that some measurements were performed followed by each other without creating a new logging file on the OPS.

We did now separate all measurements and took only the first 420 seconds of all measurements and created new figures (Fig. 3 and Fig. 4) with equal length timescales.

For the comparison experiment between the printed and the brass orifice the data was measured in a single file to show the differences. Therefore, we also created a new figure (Fig. 5).

"Figure 4 and 5. I suggest that the top panels with number concentrations should present the same extent of range… To have a visual meaning"

We adjusted the top panels of Fig. 4 and Fig. 5 to the same range.

"Right now the resolution is so poor in the figures that the legend of the top panels (fig4 and 5) are hardly readable."

We adjusted the resolution for all figures in the manuscript to be better readable.