

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 3:

„Is the technique improved over existing technology? The authors do not show data for $D < 300$ nm, the authors do not quantify the total number concentration of drops or typical droplet size produced, the authors do not quantify the composition of particles produced, the range of solvents that can be used (I guess some organic solvents might be problematic), the degree to which mixed particles (e.g. ammonium sulfate + organic compounds and preserving the ratio in the atomized particles) can be generated from aqueous stock solutions, or the minimum aerosol diameter that can be generated, which is determined by cleanliness of the solvent and drop size, the maximum time the instrument can run unattended, the degree of drying that is needed, and the range of pressure and flow rates at which the atomizer produces particles. All of these are critical to evaluate if such a device is suitable for application in laboratory research, including for instrument calibration. Thus, the answer to the question is no.“

Due to the OPS detection limit of $0.3 \mu\text{m}$ we were not able to show data $D < 300\text{nm}$.

PROTeGE can run as long as there is solution to be dispensable and an air flow is present to disperse the droplets. The maximum time is limited by the

lifetime of the dryer downstream to ensure that the generated droplets are dried correctly.

The flow rate of PROTeGE is stated in the text.

„Experiments including an SMPS to measure the full size distribution should be included. Experiments should systematically characterize the output for a much wider range of inputs (solvent, composition, solute weight percent) and analyze the results to infer drop number size and concentration. Ideally composition measurements of mixed particles are included to test for artifacts such as dissolution of the plastic and faithful representation of stock solution (e.g. adsorption of organics while the liquid passes through the atomizer).“

We regret that this is beyond the scope of this study. Certain solvents are not suitable for 3D printed parts. Since those are material specific the end user needs to make sure based on the MSDS to ensure proper functionality of the printed part.

As suggested, we performed a measurement with a SEMS to obtain particle number size distributions from 10 nm to 1000 nm for the generated ammonium sulphate solution of 0.6 g/L. We added the following paragraph to the manuscript followed by plot showing the average particle number size distribution of the generated ammonium sulfate particles (Fig. 4d).

“For the 0.6 g L⁻¹ solution an additional experiment using the SEMS instrument was performed. The size range was scanned from 10 nm to 1000 nm with a resolution of 60 bins and a sampling rate of 1 second per bin. The maximum particle number concentration was found at ~50 nm with ~40,000 cm⁻³. The average PNSD for a 420 second sampling period is shown in Fig. 4d. During the experiment the generated size distributions did not change over time. Combining the obtained size distributions from SEMS and OPS shows that PROTeGE is capable of generating particles as small as 10 nm up to 2.4 μm based on the dispersed ammonium sulfate solution.”

„Does the work increase accessibility of the technology? The paper states that the authors were able to build this device, which is nice. However, there is no benefit to the community if it is not widely shared on how to do that. The authors state that STL files are “available upon request”. This is insufficient. In my experience, share requests are often conveniently ignored or come with strings attached by the sharer. They present an unnecessary barrier. Thus, the answer to the question is no.“

Please see related comments by Referee #2: The .stl files are on a public repository for free download: <https://www.thingiverse.com/thing:4444498>

On the repository there are also pictures of PROTeGE and post processing details, this is now stated in the paper. Since the discussion paper was posted on AMTD we had more than 10 requests for the .stl file and already 16 downloads of the files from the repository.

The download statistics of the .stl file can also be found in the repository.

„If the authors want this instrument to be a low cost, self-manufacture replacement, the authors should provide the STL files as a supplement or make them available in an archived repository. The paper should include an itemized list what people need to purchase, including part numbers and cost estimates. A photo of the instrument would be a good addition to the paper. The printing could be performed by a 3D printing service and ordered with a couple of clicks. Quotes can be generated from online vendors within minutes (e.g. sculpteo) by uploading the STL file. Assembly instruction should be provided. Comments about alternative print materials should be made and the precision that is needed for printing (is 100 micron the limit?). All of the designed parts should be made available using open licenses, e.g. the CERN open hardware license (<https://www.ohwr.org/cernohl>). Such a device would be very welcome and provide a platform where anyone could build, try, and characterize the output for themselves. In this case, the likely performance limitations and/or deficits in characterization raised earlier are less critical“

Please see the answers above, this information is now provided and has been used by >10 readers of the discussion paper.

„Irrespective the route the authors wish to pursue, the authors need to comment on the technical limitations above in the revised paper. The authors should also compare cost and performance to other techniques. For example, the TSI atomizer is \$3k and very stable, and very well characterized. Small medical nebulizers (pressure and ultrasonic) can be obtained for < \$30 and are more than sufficient to generate good aerosol for shorter duration (5-15 min). It might be useful to juxtapose data from these side-by-side and discuss use cases for the printed design.“

Please see the response to referee #2 first comment on the production cost for a PROTeGE generator. We did run an exhaust time experiment with an 80ml ammonium sulphate solution where after ~10 hours the nitrogen supply did run

out even before the solution did run out. Therefore, we conclude that PROTeGE is also capable of long-term production of aerosol. We believe we have detailed the production and instrument performance within the paper so that potential users can compare this to other options. We in no way suggest PROTeGE should replace either high-end TSI atomizers nor nebulizers but, as has been demonstrated by multiple groups now using this technology, some researchers will find it the best solution for their needs.