

Reviewer 3 Response – Woo et al., “Concept for an electrostatic focusing device for continuous ambient pressure aerosol concentration.”

General comments

“This manuscript presents a concept for the application of in situ electrostatic focusing to isothermally concentrate a continuously flowing aerosol stream of submicron particles, at ambient pressure. The authors demonstrated proof-of-concept, through theoretical calculation and laboratory measurement using a prototype. This system may have potential implications in aerosol measurements under low particle concentration. I recommend publication of this manuscript with minor revision.”

We thank the reviewer for their comments and feedback. Responses to specific comments are addressed below.

Introduction

“It would be better to include more studies that requiring size-selected by a DMA, which should be more related to the current study.”

Several studies employ DMA-based selection to explore size-dependent properties on ambient (Mei et al., 2013; Thalman et al., 2017) or laboratory-generated (Ahern et al., 2016; Petters et al., 2006; Vaden et al., 2011) organic particles. In size-selecting a narrow cross-section of a polydisperse aerosol distribution, resulting aerosol concentration is subsequently low, and would benefit from in-situ concentration enrichment. Discussion of these applications will be added to the Introduction section.

Results and Discussion

“Line 3: “The observed enrichment is summarized in Figure... As shown in Figure”, please indicate the specific figure number.”

These lines refer to Figure 6; we will update the text to reflect this.

“From Figure 3, it can be seen that the enrichment factors are linearly related to the applied voltage for particles in the size range of 75 to 200 nm. I am afraid this size range does not cover the sub-micron particles in ambient.”

The range of tested aerosol diameters is consistent with a number of laboratory-based chamber experiments that utilize monodisperse organic aerosol. (Frosch et al., 2011; Vaden et al., 2011) Field campaigns in polluted urban environments, have also observed ambient mean particle diameter within the upper range of our tested diameters. (Klejnowski et al., 2013; Xu et al., 2016) As such, we believe that the sizes considered in this study are both relevant and appropriate for a wide range of applications.

While smaller particle diameters were not experimentally assessed, we estimate values of applied voltage that will satisfy the $\kappa \geq 1$ conditions necessary to achieve concentration for smaller-mobility aerosol; the results of these calculations for 30nm particles are demonstrated in Figure 2. For larger, coarse-mode aerosol, inertial forces are likely to dominate beyond the point where electrostatic deflection will achieve any meaningful concentration enhancement effects (i.e. $\kappa < 1$.) However, our proposed inlet geometry applies an electric field to a virtual impactor geometry; as a result, under such conditions where inertial forces govern aerosol motion, this system would hypothetically still be able to achieve concentration enhancement.

“I wonder if other factors such as relative humidity and temperature in the system affect the enrichment.”

Under the temperature ranges where the inlet system is generally expected to be implemented (i.e. standard or near-standard temperatures and pressures,) relative humidity and temperature are not expected to significantly affect the electric field resulting from the electrical potential differences in the inlet system, or aerosol charging behavior. However, while humidity is not expected to significantly affect the charging properties of low-hygroscopicity aerosols, such as those described in this work, it does play a factor in the corona discharge behavior of the unipolar charger used to generate the positively charged aerosol to be deflected; at higher RH values (>80%,) corona onset voltages decrease and discharge currents increase, implying lower efficiencies in charging aerosol.(Yawootti et al., 2015) As a result, the maximum potential enrichment may be reduced, though the exact dependences on RH are outside the scope of this work.

“Given such a low enrichment factor observed in Figure 6, how could the authors extend the implications of this system[?]”

As the reviewer correctly notes, the inlet system is presented as a proof-of-concept rather than as a standalone technology. However, the observed concentration enhancement demonstrated by our prototype understate the theoretical extents of enrichment possible using electrostatic deflection (Figure 3); our estimations for concentration enhancement reach as high as 65% within our explored ranges of aerosol sizes and applied voltages. As seen in Figure 6, the observed reduction in enrichment did not uniformly apply across different aerosol diameters, leading to opposite trends at different applied voltages. This nonuniformity implies that several competing factors are contributing to these losses.

As discussed in our Results and Discussion, we attribute a significant amount of our losses to electric field distortions within the inlet system. Beyond the assumptions already described in the manuscript regarding interactions between the immersion lens and other solid surfaces in the inlet system, the immersion lens geometry assumes negligible edge field effects and that the two cylindrical tubes comprising the lens are identically coaxial. In practice, the implementation of the two tubes may have introduced translational and angular asymmetries not represented in the idealized immersion lens geometry, leading to spherical field aberration effects (e.g. coma, astigmatism). (Heddle, 2000) The resulting radial and angular field asymmetry may cause deflected particles to impact the inside of the collection probe, rather than into the minor flow. Furthermore, any physical irregularity in the surface of the lens tubing (i.e. machining imperfections, surface roughness, etc.) would lead to further reduced focusing effects as their resultant electric field deviates from idealized circumstances. As enrichment was still measurable in the inlet system without accounting for these electric field nonidealities, we believe that the inherent premise of using electrostatic deflection to concentrate aerosol streams is viable, especially as future iterations improve upon these distortion effects.

Discussion of the aforementioned additional sources of charge field distortion will be added to the Results and Discussion section of the manuscript.

References

Ahern, A. T., Subramanian, R., Saliba, G., Lipsky, E. M., Donahue, N. M. and Sullivan, R. C.: Effect of secondary organic aerosol coating thickness on the real-time detection and characterization of biomass-burning soot by two particle mass spectrometers, *Atmos. Meas. Tech.*, 9, 6117–6137, doi:10.5194/amt-9-

6117-2016, 2016.

Frosch, M., Bilde, M., DeCarlo, P. F., Jurányi, Z., Tritscher, T., Dommen, J., Donahue, N. M., Gysel, M., Weingartner, E. and Baltensperger, U.: Relating cloud condensation nuclei activity and oxidation level of α -pinene secondary organic aerosols, *J. Geophys. Res. Atmos.*, 116(D22), D22212, doi:10.1029/2011JD016401, 2011.

Heddle, D. W. O.: *Electrostatic Lens Systems*. Institute of Physics Publishing, London, 2000.

Klejnowski, K., Krasa, A., Rogula-Kozłowska, W. and Błaszczak, B.: Number Size Distribution of Ambient Particles in a Typical Urban Site: The First Polish Assessment Based on Long-Term (9 Months) Measurements, *Sci. World J.*, 2013, 1–13, doi:10.1155/2013/539568, 2013.

Mei, F., Setyan, A., Zhang, Q. and Wang, J.: CCN activity of organic aerosols observed downwind of urban emissions during CARES, *Atmos. Chem. Phys. Discuss.*, 13(4), 9355–9399, doi:10.5194/acpd-13-9355-2013, 2013.

Petters, M. D., Kreidenweis, S. M., Snider, J. R., Koehler, K. A., Wang, Q., Prenni, A. J. and Demott, P. J.: Cloud droplet activation of polymerized organic aerosol, *Tellus B Chem. Phys. Meteorol.*, 58(3), 196–205, doi:10.1111/j.1600-0889.2006.00181.x, 2006.

Thalman, R., De Sá, S. S., Palm, B. B., Barbosa, H. M. J., Pöhlker, M. L., Alexander, M. L., Brito, J., Carbone, S., Castillo, P., Day, D. A., Kuang, C., Manzi, A., Ng, N. L., Iii, A. J. S., Souza, R., Springston, S., Watson, T., Pöhlker, C., Pöschl, U., Andreae, M. O., Artaxo, P., Jimenez, J. L., Martin, S. T. and Wang, J.: CCN activity and organic hygroscopicity of aerosols downwind of an urban region in central Amazonia: seasonal and diel variations and impact of anthropogenic emissions, *Atmos. Chem. Phys.*, 17, 11779–11801, doi:10.5194/acp-17-11779-2017, 2017.

Vaden, T. D., Imre, D., Beránek, J., Shrivastava, M. and Zelenyuk, A.: Evaporation kinetics and phase of laboratory and ambient secondary organic aerosol., *Proc. Natl. Acad. Sci. U. S. A.*, 108(6), 2190–5, doi:10.1073/pnas.1013391108, 2011.

Xu, X., Zhao, W., Zhang, Q., Wang, S., Fang, B., Chen, W., Venables, D. S., Wang, X., Pu, W., Wang, X., Gao, X. and Zhang, W.: Optical properties of atmospheric fine particles near Beijing during the HOPE-J3A campaign, *Atmos. Chem. Phys.*, 16(10), 6421–6439, doi:10.5194/acp-16-6421-2016, 2016.

Yawootti, A., Intra, P., Tippayawong, N. and Rattanadecho, P.: An experimental study of relative humidity and air flow effects on positive and negative corona discharges in a corona-needle charger, *J. Electrostat.*, 77, 116–122, doi:10.1016/J.ELSTAT.2015.07.011, 2015.