Determining optimal sampling conditions in the TSI Nanometer Aerosol Sampler 3089

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Responses to reviewer comments on the article: *"Determining optimal sampling conditions in the TSI Nanometer Aerosol Sampler 3089."*

We thank the Journal of Atmospheric Measurement Techniques and editorial team for considering our article for review. It was great to receive feedback, and the comments were constructive. Consequently, we believe the manuscript to be substantially improved. All comments were responded to, and some related portions of the manuscript were revised. Please refer to the tables below for specific details about the amendments made to the manuscript. <u>The line and page numbering refer to the revised version, rather than the track-changes file.</u>

Comment	**Response**
Anonymous Referee #1 comments	
1. I question the use of PBS aerosols as a surrogate for marine aerosols. Why not use NaCl particles instead? As a test aerosol, it may not be necessary to specify the type of surrogate they represent.	*We appreciate the reviewer for his/her valuable time in reviewing our manuscript and providing us with useful comments; they have certainly improved our article.* **Answer**: We aimed to create a model that is not only applicable to marine aerosols but can also be used to collect aerosols from other sources. PBS was chosen because it not only contains sodium and chlorine, which are primary components of marine aerosols but also includes potassium and phosphate. These additional components align with the chemical complexity of marine aerosols, where phosphate ions are present due to dissolution processes in marine environments and potassium is often found as a minor constituent. While NaCl is indeed a simpler and more traditional surrogate for marine aerosols, the inclusion of phosphate and potassium in PBS provides a closer approximation of the diverse ionic composition of real aerosols. Moreover, using PBS allowed us to explore the collection efficiency and deposition behavior of aerosols with a composition that is not limited to a single ionic type, enhancing the broader applicability of our findings. **Action:** To justify our choice of PBS, we

Comment	Response
	have added the following sentence and additional references on page 4 lines 99-100: "Consequently, PBS was chosen to represent the complexity in the ionic composition of not only the marine aerosols [1,2] but also in general other aerosol." [1] Nenes, Athanasios, et al. "Atmospheric acidification of mineral aerosols: a source of bioavailable phosphorus for the oceans." *Atmospheric Chemistry and Physics* 11.13 (2011): 6265-6272. [2] Baker, A. R., et al. "Trends in the solubility of iron, aluminium, manganese and phosphorus in aerosol collected over the Atlantic Ocean." *Marine Chemistry* 98.1 (2006): 43-58.
2. What are the flow rate and pressure of the air passing through the nebulizer?	**Answer**: The nebulizer used in our study was a vibrating mesh nebulizer, which operates without requiring external air pressure. Instead, it aerosolizes the medium by vibrating a mesh plate, and no air or pressure source is needed for its operation. Regarding the flow rate, we adjusted the nebulization time for different experiments to achieve the desired particle number concentrations. Specifically: • For low deposition experiments, we nebulized 1.5 mL of the solution for 3 seconds. • For medium deposition experiments, we nebulized 1.5 mL of the solution for 9 seconds. • For high deposition experiments, we nebulized 1.5 mL of the solution for 20 seconds. The duration of nebulization allowed us to control the number concentration of the particles effectively, ensuring relatively consistent deposition densities under varied experimental conditions. **Action:** To address the comment, we have added the following sentence on page 5, lines 129-132: "The desired input particle number concentrations for each experiment were

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	achieved by modifying the nebulization time "of the VMNs. Specifically, 1.5 mL of the solution was nebulized for 3 seconds in low deposition experiments, 9 seconds for medium deposition experiments, and 20 seconds for high deposition experiments." [1] Niazi, Sadegh, et al. "Utility of three nebulizers in investigating the infectivity of airborne viruses." Applied and Environmental Microbiology 87.16 (2021): e00497-21.
3. The NAS was operated at a flow rate of 1 L·min^{-1} and a voltage of -9 kV. Is this the recommended setting for the NAS? If not, why were these particular flow rate and voltage settings used?	**Answer**: The NAS was operated at a flow rate of 1 L·min^{-1} and a voltage of -9 kV in our study, which aligns with conditions demonstrated to achieve high collection efficiency. Based on the study by Chengjue Li et al. (2010), the collection efficiency of the NAS was highest at 1 L·min^{-1} and 9.3 kV, especially for ultrafine particles. These settings were selected to maximize particle deposition efficiency while maintaining uniformity and stability in the collection process. The flow rate of 1 L·min^{-1} was specifically chosen to optimize residence time in the electrostatic field, which directly influences particle collection. **Action:** This justification is included in the manuscript on page 4, lines 116-117, where we state: "The NAS was operated at a flow rate and voltage of 1 L·min^{-1} and -9 kV, respectively which has been shown to have the highest collection efficiency [1]." [1] Li, Chengjue, Shusen Liu, and Yifang Zhu. "Determining ultrafine particle collection efficiency in a nanometer aerosol sampler." Aerosol science and technology 44.11 (2010): 1027-1041.
4. Including a schematic diagram of the NAS would be beneficial for reader comprehension. Illustrating the airflow streamlines, aerosol deposition trajectory, and the electrical field within the NAS would enhance understanding.	**Answer**: To address this, we have included the schematic diagram provided by the manufacturer in the supplementary materials (Section S1). **Action:** We have included a sentence in the manuscript on page 4, lines 117-118: "A schematic diagram of the NAS is presented in Fig. S1 to highlight airflow

Comment	Response
	patterns, aerosol deposition, and electrical field dynamics.”
5. Why was the NAS positioned after the Kr-85? Is the Kr-85 an essential component for the NAS under real sampling conditions?	**Answer**: The NAS was positioned after the Kr-85 neutralizer to ensure that the aerosols entering the NAS had a well-defined and uniform charge distribution. The neutralizer helps bring the particles to a near Boltzmann equilibrium charge state, which is critical for reproducible and accurate sampling. This step minimizes variations in particle charge that could otherwise affect collection efficiency and introduces inconsistencies in the experimental results. Under real sampling conditions, the Kr-85 neutralizer is not always an essential component for the NAS. Its necessity depends on the application and the characteristics of the aerosol being studied. For laboratory-based or controlled experiments, where precise charge control is crucial, the neutralizer is highly beneficial. However, for field sampling of environmental aerosols, where charge distributions may vary naturally, the use of a neutralizer might not be required unless specific conditions demand it. By including the neutralizer in our experimental setup, we aimed to control the charging conditions and focus on evaluating the performance of the NAS itself, independent of variations in aerosol charge. [1] Johnson, Tyler J., et al. "Measuring the bipolar charge distribution of nanoparticles: Review of methodologies and development using the Aerodynamic Aerosol Classifier." Journal of Aerosol Science 143 (2020): 105526. [2] Li, Chengjue, Shusen Liu, and Yifang Zhu. "Determining ultrafine particle collection efficiency in a nanometer aerosol sampler." Aerosol science and technology 44.11 (2010): 1027-1041.
6. Figure 4 compares the normalized size distributions from NAS sampling and SMPS. Could the authors also provide a comparison of the absolute number concentrations obtained from NAS sampling and SMPS? It seems possible to estimate aerosol number concentration based on TEM images, flow rate, and deposition time.	**Answer**: We have added particle size distribution using absolute numbers rather than normalised values to the supplementary materials (Section S2) to allow for a more direct comparison between the NAS sampling and SMPS results. **Action:** We have included a sentence in the

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	manuscript on page 8, line 182: "Particle size distributions with absolute concentrations are provided in Fig. S2."
7. Did the authors convert the electrical mobility diameter from SMPS to a geometric diameter (e.g., volume equivalent diameter) to ensure comparability with the area-equivalent diameter obtained from TEM images?	**Answer**: In this study, we did not perform a conversion from electrical mobility diameter to geometric diameter. This decision was based on the shared assumption between both techniques that the particles are spherical. Under this assumption, the shape factor correction is unnecessary, as the electrical mobility diameter and the area-equivalent diameter are expected to closely align for spherical particles. We recognise that for non-spherical particles, such conversions and corrections might be needed to account for shape effects. However, given that the aerosols studied here were assumed to have spherical geometries, a direct comparison between the two techniques without additional corrections is valid. **Action:** To address the comment, we added the following explanation in the manuscript on page 7, lines 174-176: "Spherical particle geometry was assumed for consistency between the measurement techniques. This allowed the electrical mobility diameter from SMPS to be compared with the area-equivalent diameter from SEM without requiring shape factor corrections."
Anonymous Referee #2 comments	
1. The authors state that the aerosol sampler "has prominently been used" (Abstract, line 9). In my view, this particular sampler is one in a comparatively broad spectrum of sampling devices. It would be helpful if the author specify more precisely why they think that it is justified to dedicate an entire technical paper on the characterization of only this particular device (and not others). This could be complemented for example by a more comprehensive list of references that have worked with the NAS 3089.	*We appreciate the reviewer for his/her valuable time in reviewing our manuscript and providing us with useful comments; they have certainly improved our article.* **Action**: To address your concern, we have compiled a comprehensive list of studies that have utilized the TSI 3089 Nanometer Aerosol Sampler. These studies span a wide range of applications and are published in high-quality journals, predominantly in Q1 and Q2 categories. The table, included at the

Comment	Response
	end of this response file, is sorted by publication year (most recent first) and includes citation counts to highlight the widespread and impactful use of this instrument in the scientific community. It is important to note that this list was developed based on a preliminary search, and many additional studies using the TSI 3089 NAS are likely available in the literature. However, only a selection of these papers is cited in our work (page 2, line 54), as the focus was on studies most relevant to our specific research field and objectives. The consistent application of the TSI 3089 NAS across diverse, high-impact studies demonstrates its utility and relevance as a reliable tool for aerosol sampling and characterization. We believe this underscores the justification for dedicating this paper to the characterization of the TSI 3089 NAS.
2. General comment: I am sceptical whether, for systematic sampling of ultrafine particles of a given aerosol, scientists would really rely on the linear regression model developed here from three specific test aerosols, or whether they would generally carry out systematic test sampling runs and microscopic checks to optimise the sampling conditions for the given aerosol properties. In other words, is careful pre-sampling not always an essential part of any systematic sampling experiment and do the results of this study really allow this to be omitted? Could the authors comment on this further?	**Answer**: We agree that careful pre-sampling and microscopic checks remain critical for ensuring optimal sampling conditions, especially when working with aerosols with unique properties. However, the linear regression model developed in this study is designed to provide researchers with a reliable starting point, significantly reducing the time and effort required for trial-and-error adjustments. By providing a reliable estimate of the sampling time based on known input concentrations, the model allows researchers to avoid common challenges encountered during aerosol sampling: 1. **Insufficient particle collection**, which can result in extended time spent in the electron microscopy facility to gather sufficient data. 2. **Excessive particle collection**, which complicates single-particle characterization and introduces challenges in distinguishing individual particles in densely

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	packed samples. The model offers a robust starting point, enabling researchers to bypass the trial-and-error process often required to identify optimal sampling conditions. This capability is particularly advantageous as it ensures the collected samples are already suitable for further analysis while allowing for fine-tuning if adjustments are needed due to specific experimental setups or unique aerosol properties. In this regard, the model complements systematic sampling practices by reducing the time and effort required for preliminary tests, thereby enhancing overall experimental efficiency and ensuring high-quality data acquisition.
3. The section on page 2, lines 32 to 47 appears to be a bit lengthy for this particular paper as it summarizes various details on SEM and TEM that are not of particular relevance for this study. I recommend some shortening here.	**Answer**: We have revised this section to retain only the details that directly support the relevance of microscopy techniques to this study. **Action:** we improved the paragraph by removing/ editing sentences on page 2, lines 32-42.
4. On page 4, line 126 the authors state that “An AeroTrak Handheld Particle Counter 9306 (OPC) was used to ensure the experimental system was clean and free from any residual particles prior to aerosol injection.” Does this device to the particle detection range of 300 nm upwards really ensure that the experimental setup was free of ultrafine particles? Please justify.	**Answer**: Our flushing procedure was designed to ensure the removal of all residual particles, including ultrafine particles, to the greatest extent possible. The experimental setup has been used in other research conducted in our laboratory. We have previously confirmed that when the OPC shows zero particle concentrations, it aligns with the SMPS, where the particle concentrations in channels below 300 nm are approximately zero. As part of our protocol, we monitored the particle counts across all detectable bins of the OPC and ensured that all bins consistently showed zero counts for at least five minutes. To further validate the effectiveness of the flushing method, we conducted additional checks using a CPC at multiple random intervals. The CPC consistently recorded

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	zero particle counts after flushing, confirming the efficacy of our procedure in removing ultrafine particles. Moreover, the system was operated under controlled laboratory conditions with a continuous HEPA-filtered air supply, further minimizing the risk of residual UFP contamination. This combination of a flushing process, verification using the CPC, and clean air operation provides a high level of confidence that the experimental setup was effectively free of ultrafine particles. [1] Johnson, Graham R., et al. "A novel method and its application to measuring pathogen decay in bioaerosols from patients with respiratory disease." *PLoS One* 11.7 (2016): e0158763. [2] Niazi, Sadegh, et al. "Dynamics and viability of airborne respiratory syncytial virus under various indoor air conditions." *Environmental Science & Technology* 57.51 (2023): 21558-21569. [3] Niazi, Sadegh, et al. "Humidity-dependent survival of an airborne influenza a virus: practical implications for controlling airborne viruses." *Environmental Science & Technology Letters* 8.5 (2021): 412-418. [4] Niazi, Sadegh, et al. "Susceptibility of an airborne common cold virus to relative humidity." *Environmental Science & Technology* 55.1 (2020): 499-508.
5. For Figures 2, 3 and 4, I recommend to enhance the contrast of the images further since structures in the background are hardly visible.	**Action**: The contrast of Figures 2, 3, and 4 has been enhanced as recommended
6. In page 6, line 155 the authors state that "The deposition density of the collected particles was calculated by dividing the number of detected particles in each grid square by the total area of that grid square." In this context, it is not clear if the 'puddle' (= drying residue) around some particles has been included in the area of the deposited particles or not. Please clarify.	**Answer**: In our analysis, the drying residue surrounding particles was not included in the calculated area of deposited particles. This distinction was made possible by utilizing SEM images, where particles and drying residues typically exhibit different contrasts and contours. This makes boundaries of individual particles identifiable, allowing us to exclude the drying residue when counting and determining the area of the deposited particles.
7. In Fig. 4: what does "normalized dN/dlogD" exactly mean here? Please specify, ideally directly in the figure caption.	**Answer**: In this study, "normalized dN/dlogD" refers to the particle size distribution, where the particle number concentration (dN) is scaled relative to the

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	maximum concentration within the distribution. This normalization facilitates direct comparison between the size distributions obtained from the SMPS and SEM data by removing differences in absolute concentrations. **Action:** Particle size distribution with absolute values is added in the supplementary materials for readers interested in the raw data. We revised Fig. 4 caption to clarify this explanation. The updated caption will read: "SEM images and particle size distributions of (a) phosphate buffered saline (PBS) (b) Dulbecco's Modified Eagle Medium (DMEM), and (c) human saliva. Particle size distributions were normalized to the maximum concentration for easier comparison between SEM and SMPS. Particle size distributions with absolute concentrations are provided in Fig. S2."
8. Page 9, lines 197 to 199: It seems that the R/ R^2 values could be rounded to two digits after the decimal point.	**Action**: On page 9, line 200, the multiple R-value is rounded to 0.99, and the R^2 value is rounded to 0.98.

Detailed responses for Referee #2 comment #1

Paper	Journal ranking	Citation
Rissler, Jenny, et al. "Zinc speciation in fly ash from MSWI using XAS-novel insights and implications." Journal of Hazardous Materials 477 (2024): 135203	Q1	-
Lyu, Yezhe, et al. "Tribology and airborne particle emissions from grey cast iron and WC reinforced laser cladded brake discs." Wear 556 (2024): 205512.	Q1	2
He, Qingyan, Yuxin Zhou, and Xiaoqing You. "Effect of ferric chloride addition on soot formation during ethylene pyrolysis in a laminar flow reactor." Proceedings of the Combustion Institute 40.1-4 (2024): 105677.	Q1	-
Zhou, Yuxin, et al. "Effects of ferrocene addition on soot formation characteristics in laminar premixed burner-stabilized stagnation ethylene flames." Journal of Aerosol Science 175 (2024): 106265.	Q1	3
Stoll, Daniel, et al. "Suitability of Low-Cost Sensors for Submicron Aerosol Particle Measurement." Applied system innovation 6.4 (2023): 69.	Q2	2

Kang, Shipeng, et al. "Design and evaluation of a thermal precipitation aerosol electrometer (TPAE)." *Atmospheric Measurement Techniques* 16.12 (2023): 3245-3255.	Q1	-
Li, Li, et al. "Nanoparticle growth in thermally diffusive sublimation-condensation systems with low vapor pressure solids." Journal of Aerosol Science 173 (2023): 106225.	Q1	2
Lehotska Mikusova, Miroslava, et al. "Titanium dioxide nanoparticles modulate systemic immune response and increase levels of reduced glutathione in mice after seven-week inhalation." Nanomaterials 13.4 (2023): 767.	Q2	8
Bauer, Paulus Salomon, et al. "In-situ aerosol nanoparticle characterization by small angle X-ray scattering at ultra-low volume fraction." Nature Communications 10.1 (2019): 1122.	Q1	30
Weiss, Victor U., et al. "Native nano-electrospray differential mobility analyzer (nES GEMMA) enables size selection of liposomal nanocarriers combined with subsequent direct spectroscopic analysis." Analytical chemistry 91.6 (2019): 3860-3868.	Q1	25
Buckley, Alison, et al. "Slow lung clearance and limited translocation of four sizes of inhaled iridium nanoparticles." Particle and fibre toxicology 14 (2017): 1-15.	Q1	55
Fonseca, Ana Sofia, et al. "Intercomparison of a portable and two stationary mobility particle sizers for nanoscale aerosol measurements." *Aerosol Science and Technology* 50.7 (2016): 653-668.	Q2	34
Kaminski, Heinz, et al. "Measurements of nanoscale TiO2 and Al2O3 in industrial workplace environments-methodology and results." Aerosol and Air Quality Research 15.1 (2015): 129-141.	Q2	37
Albuquerque, Paula Cristina, et al. "Assessment and control of nanoparticles exposure in welding operations by use of a Control Banding Tool." *Journal of Cleaner Production* 89 (2015): 296-300.	Q1	29
Schlagenhauf, Lukas, et al. "Weathering of a carbon nanotube/epoxy nanocomposite under UV light and in water bath: impact on abraded particles." *Nanoscale* 7.44 (2015): 18524-18536.	Q1	41
Bekker, Cindy, et al. "Airborne manufactured nano-objects released from commercially available spray products: temporal and spatial influences." Journal of Exposure Science & Environmental Epidemiology 24.1 (2014): 74-81.	Q1	42
Liati, Anthi, et al. "Electron microscopic study of soot particulate matter emissions from aircraft turbine engines." *Environmental science & technology* 48.18 (2014): 10975-10983.	Q1	85
Wierzbicka, Aneta, et al. "Detailed diesel exhaust characteristics including particle surface area and lung deposited dose for better understanding of health effects in human chamber exposure studies." Atmospheric Environment 86	Q1	101

(2014): 212-219.		
Wasisto, Hutomo Suryo, et al. "Airborne engineered nanoparticle mass sensor based on a silicon resonant cantilever." Sensors and Actuators B: Chemical 180 (2013): 77-89.	Q1	180
Wasisto, Hutomo Suryo, et al. "Silicon resonant nanopillar sensors for airborne titanium dioxide engineered nanoparticle mass detection." Sensors and Actuators B: Chemical 189 (2013): 146-156.	Q1	81
Kumar, Ajay, et al. "Formation of nanodiamonds at near-ambient conditions via microplasma dissociation of ethanol vapour." Nature communications 4.1 (2013): 2618.	Q1	194
Albuquerque, Paula Cristina, João F. Gomes, and J. C. Bordado. "Assessment of exposure to airborne ultrafine particles in the urban environment of Lisbon, Portugal." Journal of the Air & Waste Management Association 62.4 (2012): 373-380.	Q2	52
Schlagenhauf, Lukas, et al. "Release of carbon nanotubes from an epoxy-based nanocomposite during an abrasion process." Environmental science & technology 46.13 (2012): 7366-7372.	Q1	148
Buonanno, Giorgio, et al. "Chemical, dimensional and morphological ultrafine particle characterization from a waste-to-energy plant." Waste Management 31.11 (2011): 2253-2262.	Q1	81
Jung, Jae Hee, et al. "Preparation of airborne Ag/CNT hybrid nanoparticles using an aerosol process and their application to antimicrobial air filtration." Langmuir 27.16 (2011): 10256-10264.	Q1	184
Avino, Pasquale, et al. "Deep investigation of ultrafine particles in urban air." Aerosol and Air Quality Research 11.6 (2011): 654-663.	Q2	53
Buonanno, G., A. A. Lall, and L. Stabile. "Temporal size distribution and concentration of particles near a major highway." Atmospheric Environment 43.5 (2009): 1100-1105.	Q1	99
Barone, Teresa L., and Yifang Zhu. "The morphology of ultrafine particles on and near major freeways." Atmospheric Environment 42.28 (2008): 6749-6758.	Q1	51