

## **Author response related to the manuscript “Size-resolved condensation sink as an approach to understand pathways how gaseous emissions affect health and climate” by Lepistö et al.**

Dear editor,

Thank you for considering our manuscript to be published in the Atmospheric Chemistry and Physics. We also want to thank the reviewers for their comments on the manuscript. We certainly believe that the comments helped us to strengthen the analysis and discussion of our study. Our responses and changes in the manuscript are provided in this document. We hope that our responses and changes are satisfactory.

With best regards,

Teemu Lepistö,  
and co-authors

### **Reviewer #1:**

Teemu Lepistö et al. explores the concept of size-resolved condensation sink (CS size distribution) by calibrating an electrical low pressure impactor to measure CS and then utilize the method in urban aerosol measurements conducted in different areas in Finland, Germany, Czechia, and India, covering diverse sources (road traffic, airports, industrial sites). Results showed significant regional variations in CS size distributions. The study highlights the condensation sink (CS) size distribution in linking particle properties to health (e.g., toxic vapour transport) and climate (e.g., CCN activation) effects. It also notes limitations, such as uncertainties from particle effective density and simplified condensation assumptions, but concludes CS size distribution is a simple, effective tool for improving climate models and air quality assessments. The topic fits scope of ACP. The paper can be considered for publication after revisions that address the following concerns.

- Thank you for reading and reviewing our manuscript. We found your comments especially helpful when clarifying the methodological approaches of our study. Also, the comments were highly valuable in the discussion of the limitations related to our methodology. We hope that our responses are satisfactory.

### **Specific comments:**

1. Section 2.2.1 “Validation of the calibration” is the sole subsection under Section 2.2. Please revise its numbering to 2.3. Correspondingly, Section 2.3 “Measurement campaigns” should be renumbered to 2.4.

- We corrected the section numbering according to the comment.

2. Equation (3): What are the values of  $D_{x,vap}$  (vapour diffusion coefficient) and  $M_x$  (molecular weight of the condensing vapour)? Based on Lines 108–110, I infer that the authors used values corresponding to sulfuric acid. So, was the condensation sink (CS) calculation in this paper solely focused on sulfuric acid? Whether this is the case or not, I recommend that the authors clarify this point explicitly.

- The values are for sulphuric acid. This point has now been clarified in the revised text. It is also clarified that we calculate CS only based on sulphuric acid in this study. We further discuss this theme in the next comment.
- Changes: After Equation 3: “where  $D_x$  is the diffusion volume,  $P$  ambient pressure and  $M$  molecular weight (Poling et al. 2000). For air, molecular mass and diffusion volume were set as 28.965 g/mol ( $M_{air}$ ) and 19.7 ( $D_{x,air}$ ), respectively. For sulfuric acid, **for which the CS in this study was calculated**, the same values were 98.08 g/mol ( $M_{vap}$ ) and 51.66 ( $D_{x,vap}$ ) (Poling et al. 2000).”

3. Another question concerns why the authors chose sulfuric acid as the reference vapour rather than organic vapours—given that organic vapours may be more critical for condensational growth within the particle size range focused on in this study (>10 nm) (Riipinen et al., 2012; Sakurai et al., 2005)). I suggest including a discussion to address this choice.

- This is an important comment. It’s true that organic vapours are critical when considering the condensation especially onto the larger particles. We decided to use sulphuric acid in our calculations as it is, according to our understanding, the most used method to calculate CS in literature.
- We, however, acknowledge that especially total CS calculation can be significantly dependent on the chosen vapour. For example, Tuovinen et al. (2021) have shown over 100 % differences in the estimated CS with different vapour properties. However, in the case of CS size distribution, the main contributing attribute seems to be the size distribution of the existing aerosol particles. To demonstrate (see major comment by reviewer #2), we estimated CS with different input parameters for particle effective density, accommodation coefficient, vapour properties and weather conditions in three of the studied environments of this study (locations Tampere: Highway, Düsseldorf: Airport, Delhi-NCR: Urban). The examples showed that even though the total CS can considerably depend on the vapour properties, the CS size distribution remains rather similarly shaped with almost equal mean size. Thus, the result shows that sulphuric acid -based CS calculation should be a reasonably good approach to estimate the CS size distribution related to other vapours as well. See further discussion of this analysis in the major comment by reviewer #2.
- Changes:
  - o In 2.2 it is noted that CS calculation, in general, is strongly dependent on the chosen vapour: “**In general, it is important to note that the obtained CS value considerably depends on the chosen vapour in the calculation (Tuovinen et al. 2021).**”

- In discussion: *“It should also be noted that the CS and CS size distribution reported typically (also here) are a theoretical quantity computed assuming that the condensing vapour is sulphuric acid with an accommodation coefficient of 1.0; in reality, the condensing compound of interest can have strongly different properties, and the actual condensation sink can be only a fraction of the theoretical one (see e.g. Tuovinen et al, 2021). In general, organic compounds are considered to contribute more to condensation than sulphuric acid on particles larger than 10 nm (e.g., Riipinen et al. 2012, Yli-Juuti et al. 2013) which causes uncertainties in the estimation of total CS if the calculation is based on sulphuric acid. These effects may also have implications to the CS size distribution which are difficult to quantify. On the other hand, the CS size distribution seemed to be mostly dependent on the existing particle size distribution, hence, the different vapour properties should not as significantly affect the shape or mean size of the CS size distribution (Table 3, Fig. S9-14). Thus, the sulphuric acid-based calculation should be a reasonably good approach to estimate the size-dependent condensation of other vapours as well. Overall, it is fair to conclude that CS measurement is a rather crude approximation of a highly complex atmospheric phenomenon, but...”*

4. Section 2.2: Many variables in this section are not clearly described, for instance, kb. I recommend adding a nomenclature list that includes specific parameters, their corresponding definitions, and the values utilized in the calculations.

- Thank you for the suggestion. We checked that all the parameters have now been described in section 2.2. Also, we added a list of parameter descriptions and utilised values in new Table S1.

5. Section 2.2: In this section, what type of particle diameter is referred to (e.g., as mentioned in Line 105)—mobility diameter or aerodynamic diameter? Please clarify this explicitly in the related context.

- This is an important point. After Eq. 2 it is now clarified that we refer to particle mobility equivalent diameter: *“In this study,  $d_p$  is referred as the particle mobility equivalent diameter.”*

6. Table 1: In Table 1, what does dpMean in the "Stage" column refer to? Is it a mobility diameter converted from the aerodynamic diameter, or simply the aerodynamic diameter corresponding to each stage?

- It refers to the aerodynamic diameter, because the ELPI+ stages have only been calibrated as a function of the aerodynamic diameter. To clarify this, in the revised text, we use the subscript “a”, when we refer to the aerodynamic diameter, for example: dpMean<sub>a</sub>.

- Related to this and the previous comment: We checked throughout the manuscript that the referred diameter concept in each section, table, figure or equation is clearly stated.
7. Line 236-238: The authors state in Figure 4 that "the relative difference between the instruments was clearly the highest with particles around 100 nm or smaller". However, it is difficult to identify the particle size corresponding to the peak relative difference from Figure 4 alone. In fact, I even observe that the peak of the relative difference in the third subplot (LRT) of Figure 4 appears to occur at approximately 200 nm. If the authors intend to illustrate this trend, it is recommended to supplement a particle size-resolved distribution plot of the relative differences to provide clearer evidence and possible.
- Thank you for pointing this out. We realise now that this statement was inaccurate and unclear, so we decided to remove that sentence in the revised text.
8. Line 273: Why the elevated  $PM_{2.5}$  mass concentration relatively reduced the condensational growth of ultrafine particles. Please provide relevant discussions and supporting references.
- Thank you for this note. We now realise that the sentence at line 273 was oddly placed and without proper explanation. In the original manuscript, the idea behind this sentence was better explained earlier in the same paragraph: *"In locations, where ultrafine particle concentrations were high (road traffic sites, airport),  $GMD_{CS}$  sizes were considerably smaller compared to other sites, being below 100 nm near airport and road traffic in Finland.  $GMD_{CS}$  of below 100 nm indicates that vapour molecules condense onto ultrafine particles more likely than on accumulation mode particles (larger than 100 nm), which may be important e.g., in terms of particle lung deposition or in the formation of CCN nuclei. As  $PM_{2.5}$  concentrations increased, also the CS size distributions shifted to larger sizes in terms of both  $GMD_{CS}$  and CSD values."* So, in principle, if  $PM_{2.5}$  concentration increases, the CS contributed by larger accumulation mode particles increases, causing that smaller fraction of CS is contributed by ultrafine particles. So, with respect to the  $PM_{2.5}$  concentration, relatively lower fraction of vapours condenses on ultrafine particles.
  - Changes:
    - o We decided to remove the sentence in line 273.
    - o This theme was already included in the discussion of the original manuscript. We decided, however, to clarify this point more: *"As seen in Table 4, Fig. 6 and Fig. S16-22, the linkage of CS, and especially  $CS_{0.1}$ , with  $PM_{2.5}$  clearly varies depending on the urban environment and geographic region. Lower  $PM_{2.5}$  mass (i.e., **lower concentration of larger accumulation mode particles**) causes a shift of the CS size distribution to smaller particle sizes, i.e. to sizes that have more efficient deposition efficiency in the lung alveoli. **As  $PM_{2.5}$  concentration increases, the vapour condensation onto larger accumulation mode particles increases, relatively reducing the fraction of vapours condensing on ultrafine particles with respect to  $PM_{2.5}$  concentration.** Thus, it could even be possible*

*that cleaner ambient air in respect of particulate mass concentration would lead to higher efficiency for semi-volatile compounds to reach e.g. the alveolar parts of lung, which could affect the health effects associated with  $PM_{2.5}$  concentration...*

9. Figure 2, 4 and 5: Please number the subplots (e.g., (a), (b), (c)) consistently with Figure 1, and refine the caption.

- We numbered the subplots and corrected the captions accordingly.

## **Reviewer #2:**

This manuscript proposes and calibrates a method to measure size-resolved condensation sink (CS) using an ELPI+ (diffusion charger + cascade impactor) by deriving stage-specific conversion factors from measured electric current to CS. The method is then applied across multiple urban environments (Finland, Germany, Czechia, India), and the authors introduce CS<sub>0.1</sub> (CS attributable to particles <100 nm) as a metric potentially relevant for cloud activation and health (lung deposition) considerations. The paper addresses a relevant and timely gap, moving beyond total CS to size-resolved CS and demonstrating its practical measurement and interpretive value across diverse settings. However, methodological assumptions (notably effective density, condensing vapour choice, and accommodation coefficient), uncertainty quantification, and validation depth need to be strengthened before publication in ACP.

- Thank you for reviewing our manuscript. We have carefully considered your thoughts regarding the uncertainties and validation of our approach. These comments helped us to strengthen the analysis of the manuscript. We hope that our changes and responses are satisfactory.

## **Major comments**

The CS derivation relies on assumptions about effective density ( $\rho_{\text{eff}}$ ) for converting between mobility and aerodynamic diameters, condensing vapour properties (sulfuric acid), mass accommodation coefficient (assumed = 1), and ambient T/P conditions; all of these feed into Equations (2-7) and the stage-specific conversion factors (Table 1). While sensitivity to  $\rho_{\text{eff}}$  is qualitatively discussed and conversion factors are tabulated, the manuscript lacks a consolidated quantitative uncertainty budget (per stage and total), including contributions from (i)  $\rho_{\text{eff}}$  choice and its size-dependence, (ii) accommodation coefficient, (iii) vapour selection ( $H_2SO_4$  vs. semi-volatile organics), (iv) temperature/pressure deviations from the assumed NTP, and (v) ELPI+ charging model/flow tolerances. Please provide a formal error propagation and a sensitivity analysis. This is crucial for interpreting inter-site differences and the CS<sub>0.1</sub> metric.

- This is a very important note, thank you. In the original manuscript, the focus in the uncertainty analysis was mainly on the effective density, because we think that it is the most important source of uncertainty from the methodological point-of-view. It is true that vapour properties, accommodation coefficient as well as conditions are great sources of uncertainty in CS measurement. On the other hand, we want to point out that these uncertainties are not necessarily measurement methodology -related because these parameters are always needed to

be assumed and estimated when measuring CS with other approaches as well. However, we agree that uncertainty analysis considering these five points would be extremely important to better understand the uncertainties in the CS size distribution measurement. In general, we want to note that we estimated the CS for sulphuric acid with  $\alpha$  of 1.0 because, according to our understanding, it is the most utilised method to estimate CS in literature.

- To estimate the uncertainties related to effective density, accommodation coefficient, vapour properties and conditions, data from three studied locations (Section 2.3, Tampere: Highway, Düsseldorf: Airport, Delhi-NCR: Urban) were utilised. These locations were chosen because they had significantly varying particle characteristics: In Tampere, roughly half of CS was attributable to ultrafine particles, whereas, in Delhi-NCR, CS was almost completely attributable to larger accumulation mode particles. The Düsseldorf location represents a mixture of these two (clear contribution of both ultrafine particles and regional accumulation mode particles). For the data from the locations, the CS was calculated by changing values for effective density, accommodation coefficient, vapour molecular weight and diffusion volume, temperature and pressure. We want to note that, in general, it is challenging to do an uncertainty analysis for ambient CS measurement because there is no proper reference method available. Also, the uncertainty is always dependent on the situation, like the shape of the particle size distribution and condensing vapours. Hence, it is practically very difficult to define universal uncertainty limits for the CS measurement that would be appropriate in different circumstances. Due to these reasons, we chose the approach of investigating how the varying parameters affect the CS measurement in these three locations. We hope that this approach is understandable and satisfactory. Regarding the general uncertainty of the ELPI+, we refer to the study by Järvinen et al. (2014).
- Results of the mentioned uncertainty analysis are collected in new Table 3, which is also below:

Varying parameter	Parameters						Result compared to reference			
	$\rho_{\text{eff}}$ (g/cm <sup>3</sup> )	$\alpha$	$M_{\text{vap}}$ (g/mol)	$D_{\text{x,vap}}$ (cm <sup>3</sup> )	T (K)	P (atm)	CS	CS <sub>0.1</sub>	GMD <sub>CS</sub>	CS <sub>0.1</sub> /CS <sub>2.5</sub>
Reference	1.0	1.0	98.08	51.66	293.15	1.0	-	-	-	-
$\rho_{\text{eff}}$	<b>1.5</b>	1.0	98.08	51.66	293.15	1.0	0.77-0.84	0.74-0.75	1.05-1.07	0.89-0.96
	<b>0.8</b>	1.0	98.08	51.66	293.15	1.0	1.09-1.14	1.17	0.96-0.97	1.03-1.08
$\alpha$	1.0	<b>0.1</b>	98.08	51.66	293.15	1.0	0.12-0.15	0.10-0.11	1.17-1.22	0.72-0.89
	1.0	<b>0.5</b>	98.08	51.66	293.15	1.0	0.54-0.60	0.51	1.07-1.10	0.86-0.95
Vapour	1.0	1.0	<b>300</b>	51.66	293.15	1.0	0.61-0.67	0.58	1.06-1.08	0.88-0.96
	1.0	1.0	98.08	<b>300</b>	293.15	1.0	0.70-0.86	0.93-0.95	0.84-0.89	1.1-1.32
	1.0	1.0	<b>300</b>	<b>300</b>	293.15	1.0	0.51-0.54	0.56	0.94-0.96	1.03-1.10
T	1.0	1.0	98.08	51.66	<b>253.15</b>	1.0	0.86-0.90	0.92	0.96-0.98	1.02-1.06
	1.0	1.0	98.08	51.66	<b>273.15</b>	1.0	0.93-0.95	0.96	0.98-0.99	1.01-1.03
P	1.0	1.0	98.08	51.66	293.15	<b>0.9</b>	1.02-1.04	1-1.01	1.01-1.02	0.97-0.99
	1.0	1.0	98.08	51.66	293.15	<b>1.1</b>	0.96-0.99	0.99-1	0.98-0.99	1.01-1.03

In addition, the results of this analysis are in the supplementary (Fig. S9-14), showing also the effect on the size distribution. The table shows the relative differences of CS, CS<sub>0.1</sub>, GMD<sub>CS</sub>, and CS<sub>0.1</sub>/CS<sub>2.5</sub> compared to the reference with the studied three locations when some of the parameters were altered. The reference represents the values that were utilised in this study by

default. As seen already in the original manuscript, the effective density can affect the obtained total CS by roughly  $\pm 25\%$ , but the effect on the size distribution (and  $GMD_{CS}$  and  $CS_{0.1}/CS_{2.5}$ ) is less significant, within  $\pm 10\%$ . This principle holds also when considering different vapour properties and the accommodation coefficient. There can be multiple time differences in the obtained CS when altering these parameters (as shown e.g. by Tuovinen et al. 2021), but still the effect on size distribution is considerably less significant. According to Järvinen et al. (2024) the general uncertainty of ELPI+ is approx. 20% for particles smaller than 40 nm, and 12% for particles larger than that (with 95% confidence interval). The uncertainty of the determined cut-point sizes in the ELPI+ calibration is 2%. Hence, the general uncertainty of ELPI+ should not considerably affect the CS size distribution results but can affect the total CS roughly by 12% as the CS attributable to particles smaller than 40 nm is typically low (see 3.2). We believe that this analysis shows that despite the general and methodological challenges in CS measurement, the CS size distribution measurement can be considered to be reasonable accurate.

- In addition, we show the variation of the ELPI+ CS response coefficient with different input parameter values for  $\rho_{eff}$ ,  $\alpha$ ,  $M$ ,  $D_x$ ,  $T$  and  $P$  in new Fig. S1.
- Changes made:
  - In 2.2: *“In general, it is important to note that the obtained CS considerably depends on the chosen vapour in the calculation and the assumed  $\alpha$  (Tuovinen et al. 2021).  $K$  with different values for  $\rho_{eff}$ ,  $\alpha$ ,  $M_{vap}$ ,  $D_{x,vap}$ ,  $T$  and  $P$ , and the relative change compared to the default values (sulphuric acid in NTP-conditions with  $\alpha$  of 1.0 and  $\rho_{eff}$  of 1.0 g/cm<sup>3</sup>) are shown in Fig. S1.”*
  - In 2.3: *“Furthermore, as mentioned in Section 2.2., the obtained CS is strongly dependent on the parameters related to the condensing vapour molecule ( $M_{vap}$ ,  $D_{x,vap}$ ), accommodation ( $\alpha$ ) and ambient conditions ( $T$ ,  $P$ ). To better understand the effect of chosen parameters on CS measurement, data from three studied locations (Section 2.3, Tampere: Highway, Düsseldorf: Airport, Delhi-NCR: Urban) were utilised in uncertainty analysis where values for  $\rho_{eff}$ ,  $\alpha$ ,  $M$ ,  $D_x$ ,  $T$  and  $P$  were altered. In this analysis, the effect of the varying parameters was estimated for total CS, CS attributable to ultrafine particles ( $CS_{0.1}$ ), geometric mean diameter of CS size distribution ( $GMD_{CS}$ ) and the  $CS_{0.1}/CS_{2.5}$  fraction (i.e.  $CS_{0.1}$  fraction in total CS ( $CS_{2.5}$ , particles < 2.5  $\mu$ m)), all of which were also measured at the measurement campaigns included in this study (Section 2.4, 3.2). These locations were chosen for the comparison as they represented clearly different aerosol characteristics in terms of ultrafine and accumulation mode particles (see 3.2).”*
  - In 3.1: *“In Table 3, the effect of  $\alpha$ , vapour properties ( $M_{vap}$ ,  $D_{x,vap}$ ) and conditions ( $T$ ,  $P$ ) together with  $\rho_{eff}$  are shown on the ELPI+ CS measurement with data from the three chosen locations (Tampere: Highway, Düsseldorf: Airport, Delhi-NCR: Urban). The results with the other parameters were rather similar as with  $\rho_{eff}$ . Especially,  $\alpha$ ,  $M_{vap}$ ,  $D_{x,vap}$  can strongly affect the obtained total CS. For example, the results show that even 100% differences with total CS values can be obtained with different vapours (as shown by Tuovinen et al. 2021). Also, with  $\alpha$  of 0.1, the obtained CS is roughly 10 times lower. Still,*

the effect of the varying parameters on the shapes of CS size distributions were relatively low (see Fig. S9-14) which can be seen also with the relative changes in  $GMD_{CS}$  and  $CS_{0.1}/CS$ . With all the chosen values for the studied parameters, the  $GMD_{CS}$  varied from -16 % to +22 % with the studied data. When considering the estimated  $CS_{0.1}/CS_{2.5}$  -fraction, the differences were from -28 % to +32 % with the chosen data.”

- In 3.1: “Thus, regarding the suitability of ELPI+ in terms of CS measurement, it can be concluded that methodology-dependent challenges (i.e.  $p_{eff}$ ) do not considerably affect the size-resolved measurement, but, in terms of total CS, the result can vary roughly  $\pm 25$  % if  $p_{eff}$  cannot be accurately estimated. In addition, the general uncertainty of the ELPI+ measurement is important to acknowledge: According to Järvinen et al. (2024) the uncertainty is approx. 20 % for particles smaller than 40 nm, and 12 % for particles larger than that (with 95 % confidence interval). The uncertainty of the determined cut-point sizes in the ELPI+ calibration is 2 %. Hence, the general uncertainty of ELPI+ should not considerably affect the CS size distribution results but can affect the total CS roughly by 12 % as the CS attributable to particles smaller than 40 nm is typically low (see 3.2). When considering the difference of ELPI+ compared to particle mobility -based measurement, the mobility size distribution -based CS measurement seems to measure roughly 20–30 % lower CS (Fig. 4), but it should be noted that this ratio can be different in other environments. Overall, it is important to note the general uncertainties of CS measurement, e.g., related to vapour properties and accommodation as well as conditions. As seen, in Table 3, these uncertainties can cause multiple time differences in the obtained CS, as shown by Tuovinen et al. (2021). However, regarding the CS size distribution measurement, these uncertainties are not as significant, supporting the idea of CS size distribution measurement with ELPI+ and other methods as well.”
- In Discussion: “It should also be noted that the CS and CS size distribution reported typically (also here) are a theoretical quantity computed assuming that the condensing vapour is sulphuric acid with an accommodation coefficient of 1.0; in reality, the condensing compound of interest can have strongly different properties, and the actual condensation sink can be only a fraction of the theoretical one (see e.g. Tuovinen et al, 2021). **In general, organic compounds are considered to contribute more to condensation than sulphuric acid on particles larger than 10 nm (e.g., Riipinen et al. 2012, Yli-Juuti et al. 2013) which causes uncertainties in the estimation of total CS if the calculation is based on sulphuric acid.** These effects may also have implications to the CS size distribution which are difficult to quantify. **On the other hand, according to the results, the CS size distribution seemed to be mostly dependent on the existing particle size distribution, hence, the different vapour properties, accommodation coefficient or conditions should not as significantly affect the shape or mean size of the CS size distribution (Table 3, Fig. S9-14). Thus, the sulphuric acid-based calculation should be a reasonably good approach to estimate the size-dependent condensation of other vapours as well. However, when interpreting the results in Fig. 5-6 and Table 4, the methodological and general uncertainties of CS estimation (Section 3.1) should be acknowledged.”**

## Minor comments

\* The manuscript argues that size-resolved CS can improve climate/air-quality modelling. Can you specify why this would improve climate modelling and how it would improve climate direct or indirect effects?

- We think vapour condensation is very relevant in terms of aerosol climate effects. First, condensation of vapour molecules directly affects aerosol optical properties which is important when considering how the particles interact with light (e.g., the lensing effect). Second, cloud formation has been considered to have a net cooling effect on climate. Typically, recently formed particles are mostly in the sizes below 10 nm, hence, condensation processes are needed to increase the particles size to 50–100 nm so that they can act as cloud condensation nuclei. CS size distribution can be valuable when estimating both of these phenomena. Overall, to model the climatic impacts of particles accurately, it is important to understand how the atmospheric particle size distribution changes in different conditions and environments. Thus, we believe that adapting the CS size distribution in reporting observational data could be useful and rather simple method to potentially provide additional data related to condensation, hence, improving the accuracy of climate models.
  
- In the original manuscript, we have discussed these points in “Introduction” and “Discussion:
  - o Introduction: *“Climate-wise, condensation of atmospheric vapours directly affects aerosol optical properties and, thus, on how the particles interact with light. For example, light absorption of soot particles can be increased due to soot particle coating with organic and inorganic compounds, causing so-called lensing effect (Riemer et al. 2019). On the other hand, particles larger than 50–100 nm can act as cloud condensation nuclei (CCN) (Kerminen et al. 2012) contributing to cloud formation and, thus, to the net cooling effects of clouds (e.g. Fuzzi et al. 2015). Typically, recently formed particles, e.g., in exhaust (Rönkkö et al. 2017) or during new particle formation (NPF) events (Kontkanen et al. 2017), are mostly in the sizes below 10 nm, hence, condensation processes are needed to increase the particles size so that they can act as CCN.”*
  
  - o Discussion: *“In addition to the health effects, the observed differences in the CS size distribution are interesting in terms of the aerosol climate effects. For example, the vapour condensation on particles changes the coating of particles, thus, affecting how the particles interact with the incoming solar radiation (e.g. Riemer et al. 2019). Therefore, understanding the CS size distribution could help to estimate for which particle types and sizes condensation is the most important in certain location. Also, the activation of particles as CCN depends on particle size (50–100 nm, e.g., Kerminen et al. 2012). The differences in CS size distribution indicates that nanoparticle growth into sizes where they could act as CCN can depend on the environment and existing particle concentrations. Better characterization of CS size distribution could help to better understand CCN formation in different environments. In order to model the climatic impacts of particles accurately, it is important to characterise the factors affecting formation and change in the atmospheric particle size distribution in different conditions and environments. Accurate modelling of the particle size distribution on a*

*global scale is notoriously difficult and resource-consuming; adapting the CS size distribution in reporting observational data could be useful and rather simple method to potentially provide additional constraining data for the important process of condensation, improving the accuracy of climate models. “*

\* Figures do not include error bars or shaded envelopes, could you somehow estimate errors of the results?

- This is a good note. In this case, we however find the use of error bars rather challenging. As discussed in the earlier major comment, CS measurement has fundamental uncertainties related e.g. to the vapour properties and the accommodation factor. So, in principle, if we take all the potential sources of uncertainty into account, we believe that the result could be blurry and potentially draw the attention away from the main thoughts of the manuscript. In the revised text, we have now more carefully analysed the effect of the potential sources of uncertainty (see the response to the major comment), so we hope that the reader can better interpret the results also regarding potential sources of error.
- On the other hand, in the revised manuscript, we decided to add shaded envelopes to show the observation range of the CS size distributions (Fig. S15). We hope that this figure helps the reader to see the variability of the measured aerosol in the studied locations. Overall, it is good to note that the measurements in each location were relatively short. In this paper, the main idea is, however, to explore the CS size distribution concept and show the potential of CS size distribution in atmospheric aerosol studies, so we do not consider this as a major issue in the context of this study. Still, we believe that long-term measurements of CS size distributions would be beneficial and necessary for detailed comparison of different environments.
- Changes made:
  - o Figure S15 added in the supplement. Also, in Section 3.2.: *“In Fig. 5, the average CS size distributions measured at all the studied environments (except the ELPI+ and DMPS comparison campaign) are **shown (see Fig. S15 for the observation ranges of the CS size distributions).**”*
  - o We clarified that the results should not be considered as long-term averages in Section 2.4: *“Therefore, the presented results should not be considered to represent long-term aerosol characteristics in the studied location.”*
  - o In Discussion: *“However, when interpreting the results in Fig. 5-6 and Table 4, the methodological and general uncertainties of CS estimation (Section 3.1) should be acknowledged. Also, it is important to note that the measurements at the studied locations were relatively short and, thus, the results should not be considered as long-term characteristics of aerosol at the studied locations. Hence, the observation ranges of the CS size distribution should be noted (Fig S.15). In general, the results related to CS size distribution,  $CS_{0.1}$ ,  $GMD_{CS}$  and CSD can be different during other conditions and in long-term measurements. Still, the results show clearly that size-resolved CS can be significantly location-dependent, which is valuable to understand when considering the potential health and climate effects of aerosols.”*

*“Long-term CS size distribution measurements in various locations and environments could be beneficial to better understand its relevance and impact in aerosol health and climate effects.”*

L56-58 – NPF events have also been observed in polluted environments (e.g. Kulmala et al., 2017) or particle loadings (e.g. Casquero-Vera et al., 2023), so the CS is expected to decrease NPF occurrence but depending on the CS efficiency (e.g. Tuovinen et al., 2021). I think those ideas are also important to include, since it shows the current lack of knowledge on the effect of CS in NPF events occurrence.

- Thank you for the suggestion. These findings are now mentioned in the introduction.
- Changes made: *“Low CS, on the other hand, may result in NPF-events as there is not high enough concentration of existing aerosol particles for all vapour molecules to condensate, causing more likely nucleation. Many studies have reported that stronger NPF events tend to occur during low CS periods (e.g., Boy and Kulmala 2002, Hamed et al. 2007, Zaidan et al. 2018). Therefore, CS can be a highly useful parameter in estimating and modelling the effects aerosol aging on the air quality and climate. **On the other hand, NPF-events are still commonly observed also when CS is high, e.g., in high polluted environments and during pollution events, showing the need for deeper understanding on the connection between CS and NPF-events (e.g., Kulmala et al 2017, Casquero-Vera et al., 2023).**”*

L59-69 – Following the major comment, here the text suggests that diffusion-charger-based instruments (ELPI+) better capture surface area without requiring shape/density assumptions, unlike DMPS. However, ELPI sizing still involves effective-density assumptions and sticking efficiency. Could the authors clarify whether ELPI-based CS measurements (total and size-resolved) avoid or still rely on particle density/shape assumptions, and how these uncertainties compare to DMPS? It is not clear this point when later the authors use sulfuric acid as the reference vapor.

- We agree that ELPI+ still has methodological challenges related to the effective density. As discussed in the earlier major comment, we have evaluated and clarified the challenges of ELPI+ CS measurement in the revised manuscript. Regarding the mentioned lines 59-69, we did not however make any changes, as the point of this paragraph is only to introduce the potential of diffusion charger -based CS measurement (for example ELPI+ as an instrument is not mentioned in this paragraph).
- In general, the particle shape -related underestimation of mobility particle sizer instruments in terms of particle surface area related -measurements has been observed in earlier studies e.g. regarding particle lung deposited surface area (LDSA) measurement.
- Changes made:
  - o In 3.1: *“The (density corrected) ELPI+ measured 1.31, 1.25 and 1.20 times higher total CS than the DMPS during the low background, inversion and LRT episodes, respectively. The difference is most likely related to the fractal structure of particles, which can be better observed with the electric current measurement by the ELPI+. **Similar results of about 5–25 % difference between diffusion-charger based***

***instruments and mobility particle sizers have been reported also in the measurement of particle lung deposited surface area (LDSA, e.g., Chang et al. 2023, Lepistö et al. 2024)."***

- In discussion: "*However, when interpreting the results in Fig. 5-6 and Table 4, the methodological and general uncertainties of CS estimation (Section 3.1) should be acknowledged.*"