

Response to reviewers

AR-2026-14

We sincerely thank the reviewers for their careful reading of our manuscript and for their constructive comments. We have done our best to provide a thoughtful point-by-point response below, shown in *italic*. Where changes have been made in the manuscript, we indicate the specific sections and lines and we will highlight **the revised text in cyan** accordingly.

We hope that the clarifications and revision provided satisfactorily address the concerns raised, and we thank the reviewers and the editor once again for their valuable input.

Reviewer #1 :

This manuscript presents measurements of levoglucosan condensation and evaporation to and from aqueous droplets containing inorganic salts, collected with a laboratory chamber setup. While the results are potentially an interesting addition to the relevant literature, the authors should do a better job in relating this work to literature and state-of-the-art knowledge. They should also add additional quantitative analysis to raise to the level where this work can fit the scope Aerosol Research being "focused on studies with general implications for any of the subject areas rather than investigations that are primarily of local interest or of descriptive nature. " At present, I do not think that the authors use what is already known about aerosol thermodynamics and kinetics to say something novel enough about the present system. Furthermore, I have an important concern regarding the appropriateness of the experimental approach for measuring the phenomenon that the authors aim to target - i.e. the use of a dry instead of humidified DMA measurement and the associated evaporation of some of the key species (please see specific comments #8 below). Here are some suggestions on how to improve the manuscript:

Response : *We thank the reviewer for their constructive suggestions, which helped us improve the manuscript. Crucially, we have made it clearer that this manuscript is the experimental part of a two-paper series, where the second manuscript is dedicated to model calculations by changing the title to:*

“Co-condensation and co-evaporation of levoglucosan onto and from deliquesced ammonium sulfate particles – Part 1: Chamber experiments”

Concerning the question of state of the art knowledge, indeed, evaporation of single (S)VOC from ammonium sulfate particles has been widely studied. Yli-Juuti et al. (2013) reported the evaporation of succinic acid in a flow tube at various values of relative humidities (RH) maintained constant during each experiment. Choczynski et al. (2024) studied the evaporation of Triethylene glycol in a levitation system under different RH. Both applied thermodynamic models to compare with experimental results. However, these works were not performed in a large simulation chamber, which allows for monitoring slow kinetic processes, under extended range of conditions including variations of RH.

The kinetics differ significantly between simulation chamber and flow-tube experiments, with much longer experimental timescales in chambers allowing for the observations of variations of many parameters under more relevant atmospheric conditions (Doussin et al., 2023). The surface-to-volume ratios are much larger in simulation chambers, inducing longer lifetimes for volatile species. Furthermore, the aerosol lifetimes can reach several days in CESAM, versus a few minutes in flow-tubes.

To our knowledge, volatility of secondary organic aerosols is widely studied in large simulation chambers such as AIDA with varied RH (Jonsson et al. 2007) and CESAM by increasing RH (Giorio et al. 2017). They both focused on complex chemical systems consisting of secondary organic aerosol (SOA). Levoglucosan has also been studied in complex chemical systems: Bertrand et al. (2018) studied a wood-smoke mixture in the PSI atmospheric simulation chamber. Gong et al. (2018) demonstrated RH-dependent partitioning of levoglucosan onto ammonium sulfate particles over 10 - 95% RH, but their experiments were performed in a small reaction flask and investigated uptake and partitioning rather than RH-driven co-condensation and co-evaporation.

In summary, several studies have addressed related aspects, but none combines the specific elements of the present work. The present study therefore bridges these previous approaches by combining a single well-defined SVOC (levoglucosan), inorganic ammonium sulfate particles and controlled RH changes in both directions in a large atmospheric simulation chamber, with the aim of discussing the complex interrelation between particle size, total mass concentration, relative humidity, levoglucosan/ammonium sulfate ratio, presence of a surfactant, and temperature.

We have modified the text in the introduction to account for the previous work in more detail. It now reads (new text in blue):

“Co-condensation is the process by which condensable vapors condense alongside water vapor onto growing aerosols (Kulmala et al., 1993; Topping et al., 2013). This can significantly alter the properties of clouds (Kulmala et al., 1997). Indeed, the co-condensation of gas-phase semi-volatile organic substances together with water vapor may increase the condensed mass and amplify water uptake. Semi-volatile species partition dynamically between the gas and the condensed phases (Ijaz et al., 2025; Thompson et al., 2017; Lopez et al., 2025; Donahue et al., 2006). As relative humidity (RH) increases, water condenses on particles, increasing the condensed volume and enhancing the condensation of semi-volatile species. Conversely, when RH decreases, water evaporates from the particles, along with semi-volatile species. Although this process is based on thermodynamics and has been simulated in models (Topping et al., 2013; Wang et al., 2026), few studies have investigated it experimentally. Field studies coupled to models have shown that this effect is potentially large for secondary organic aerosol (SOA) containing a large fraction of semi-volatile species (Wang et al., 2025; Heikkinen et al., 2024; Rastak et al., 2017; Topping et al., 2013; Topping and McFiggans, 2012; Topping et al., 2011). Intermediate between field and laboratory studies, simulation chamber studies have been performed to investigate the volatility of SOA under varying RH (Jonsson et al. 2007; Giorio et al., 2017). Levoglucosan has also been studied in complex chemical systems: Bertrand et al. (2018) investigated a wood-smoke mixture in a large atmospheric simulation chamber. In

smaller scale experiments, Zardini et al. (2010) and Yli-Juuti et al. (2013) observed succinic acid evaporation from a flow of deliquesced ammonium sulfate and NaCl particles, respectively, at constant RH between 60 and 80 %. They found a salting-out effect that depended on the organic-to-inorganic ratio and noticed enhanced evaporation of succinic acid at lower RH. Therefore, large scale experiments investigated complex chemical mixtures, while smaller scale experiments investigated simpler chemical systems, under a restricted range of parameters. None of the previous studies observed a simple chemical system under various conditions in a large simulation chamber representing atmospheric conditions.

We present a two part-study (part 1: experimental, and part 2: modeling) bridging the previous experimental approaches by examining the ability of a single semi-volatile organic compound to co-condense and co-evaporate from inorganic particles over the entire subsaturated RH range (0–100 %), and under various conditions of particle mass and size, and the presence of a surfactant on the particles. Levoglucosan (1,6-anhydro- β -D-glucopyranose, $C_6H_{10}O_5$) was chosen because it is a semi-volatile compound with a saturation concentration of $\log(C^) = 1.1 \mu\text{g.m}^{-3}$ at 298 K (Zhang et al., 2024; Booth et al., 2011). It has an aqueous solubility of 61–63 wt% (Lienhard et al., 2012; Osmanbegovic et al., 2023) and its supercooled liquid is miscible with water (Parsons et al., 2004). The molecule bears 3 hydroxyl groups that can provide strong H-bonds with water molecules and ions (Qin et al., 2021; Pye et al., 2018). Furthermore, levoglucosan is a widely used molecular marker of biomass burning in the atmosphere, it is almost exclusively emitted by the pyrolysis of cellulose and hemicellulose, it represents 52 to 290 mg.g^{-1} of organic carbon (OC) in biomass combustion, depending on the type of wood burnt and the combustion conditions (Bhattacharai et al., 2019). It has been measured in particles at various sites all over the world, and overall, 90% of levoglucosan mass concentrations were found in particles with aerodynamic diameters of less than 2 μm (Bhattacharai et al., 2019). However, due to its physical and chemical properties, the mass fraction of levoglucosan in particles is influenced by long-range transport, as well as by particle-phase mixing, evaporation, co-condensation, and multiphase chemical (and biological) processes (Bhattacharai et al., 2019; Yazdani et al., 2023).*

The gas-particle partitioning of levoglucosan was monitored under ambient-like concentrations on ammonium sulfate (AS) monodispersed particles in the CESAM simulation chamber (French acronym for Experimental Multiphase Atmospheric Simulation Chamber). Various conditions were explored, including relative humidity (from dry to 100% RH), dilution, particle mass and size, and the presence of a surfactant on the particles. The net evaporation flux of levoglucosan was quantified under each condition.”

General comments:

Comment 1.

The results should be interpreted more quantitatively to provide useful and specific constraints for understanding the relevant atmospheric processes. The theoretical frameworks for both the relevant thermodynamics (e.g. E-AIM or similar thermodynamic models) as well as the kinetics (condensation and evaporation) exist, and the authors should at least attempt to use these frameworks to quantitatively answer the following questions: Can the behavior of levoglucosan be explained with the present knowledge on thermodynamics of the relevant systems? If not,

what do the results imply about the activities of the relevant species in the condensed phase? For some inspiration (as well as potentially interesting older literature) on such quantitative analysis, please look at e.g. the work by Yli-Juuti et al. 2013.

Response : *A simulation of the chamber experiments was indeed planned from the beginning when the study was conceived. Yet, the modelling of the experiments proved more complex than anticipated because both, the AIOMFAC and the E-AIM models did not describe the ammonium sulfate/levoglucosan system with sufficient accuracy. We therefore decided to develop our own Pitzer model based on additional new experimental data that we acquired in collaboration with Ulrich Krieger (see responses below to comment #3). The model study (part 2) should be ready for submission by the end of this or the beginning of next year.*

Comment 2.

The authors motivate the study by the impact that aqueous phase thermodynamics have on cloud droplet activation in supersaturation conditions. This is indeed also the conditions to which the term "co-condensation" usually refers to in the literature. The results presented in the manuscript have, however, been collected in water activities up to 100% RH, without observing the actual cloud droplet activation but rather staying in the metastable hydrated regime. This needs to be clarified throughout the manuscript to avoid misunderstandings, as the water content and condensation kinetics close to the activation point can be rather different from those at lower levels of water vapor saturation.

Response : *We thank the reviewer for highlighting this potential source of misinterpretation. We confirm that our chamber experiments remain below water saturation. Yet, we are not in a metastable hydrated regime but in the thermodynamically stable regime of hygroscopic growth and shrinkage. We have clarified this aspect in the revised introduction by replacing "under various conditions" by "over the entire subsaturated RH range (0–100 %), and under various conditions of particle mass and size, and the presence of a surfactant on the particles".*

Comment 3.

Along the lines of comment #1 above, I encourage the authors to investigate the sensitivity of the theoretical evaporation and condensation rates to e.g. a) the RH in the chamber; b) the concentration of the particles (which links to the gas-phase saturation, but also coagulation); c) assumptions about the molecular properties of levoglucosan and its interactions with water and AS. Such sensitivity analyses would help you to understand what really governs the behavior of your system and constrain key parameters associated with it.

Response : *These suggestions are indeed the subject of the model study that is in preparation (i.e. part 2). In the model study, we investigate whether a thermodynamic equilibrium between the gas and the particle phase is established within the chamber and how important gas-phase deposition of levoglucosan to the chamber walls is. Moreover, we will account for the non-ideality of aqueous levoglucosan and ammonium sulfate solutions. As this system is not well described by AIOMFAC, which predicts a liquid-liquid phase separation that is not observed in experiments (e.g. Parson et al., 2004), we had evaporation rates of levoglucosan from mixed ammonium sulfate/levoglucosan particles measured by Ulrich Krieger in his electrodynamic balance (setup as described in Lienhard et al., 2012) and used the derived activities together with data from Lienhard et al. (2012) to develop a Pitzer model for this system.*

The aim of the present experimental study is to present the chamber data and to discuss the complex interrelation between particle size, total mass concentration, relative humidity, levoglucosan/ammonium sulfate ratio, presence of a surfactant, and temperature. We are confident that splitting up the study into two papers is the best solution as the manuscript would become too lengthy when the experiments and the simulations were described in the same paper.

We have modified the text in the introduction (see our response to comment 5 of this reviewer) and conclusions (see our responses to comments 9 and 10 of this reviewer) to make these elements clearer.

Specific comments:

Comment 4.

Title: As mentioned above, I believe the term "co-condensation" is often used in the context of cloud droplet activation (i.e. water vapor supersaturations above the critical supersaturation), while the results presented in this manuscript are obtained from lower water vapor activities. Please consider revising the title to better reflect the content of the manuscript.

Response : *While co-condensation is indeed more often discussed in the context of cloud droplet activation, this term is equally applicable to haze development under subsaturated conditions (Gunthe et al., 2021; Chen et al., 2022). Co-condensation describes the enhanced water uptake driven by the simultaneous condensation of intermediate and semi-volatile species alongside water vapor. Consequently, the same mathematical and physical principles apply across both subsaturated and supersaturated regimes. Indeed, cloud parcel models describe co-condensation below and above water saturation as the same physical process (Topping et al., 2013; Wang et al., 2026).*

Comment 5.

Introduction, lines 55-56: The authors state "this process has never been experimentally investigated on a single semi-volatile compound under various controlled conditions." I do not think this statement is true. In minimum, the article by Yli-Juuti et al. and the references therein is evidence of that, and I strongly encourage the authors to do a thorough review of the literature and modify this paragraph accordingly. Secondly, the authors themselves do not directly observe the co-condensation of organic species upon cloud droplet activation (to which many of the references in this paragraph refer to). Please therefore revise this paragraph to better reflect the content of the study and the scientific literature.

Response : *Thank you for pointing out the article of Yli-Juuti et al. We further searched the literature and found a second paper from the same research group (Zardini et al., 2010). Both investigated the evaporation of succinic acid either from ammonium sulfate (Yli-Juuti et al. 2013) or NaCl particles (Zardini et al., 2010) at RH between 60 and 80 %. While these studies emphasized the relevance of organic-to-inorganic ratio for the evaporation rate, they indeed also discussed the role of RH. In the revised manuscript, we therefore acknowledge the findings of these studies by replacing the sentence: "However, this process has never been experimentally investigated on a single semi-volatile compound under various controlled conditions." by:*

“In smaller scale experiments, Zardini et al. (2010) and Yli-Juuti et al. (2013) observed succinic acid evaporation from a flow of deliquesced ammonium sulfate and NaCl particles, respectively, at constant RH between 60 and 80 %. They found a salting-out effect that depended on the organic-to-inorganic ratio and noticed enhanced evaporation of succinic acid at lower RH. Therefore, large scale experiments investigated complex chemical mixtures, while smaller scale experiments investigated simpler chemical systems, under a restricted range of parameters. None of the previous studies observed a simple chemical system under various conditions in a large simulation chamber representing atmospheric conditions.”

Furthermore, we modify the sentence on line 64 to:

“We present a two part-study (part 1: experimental, and part 2: modeling) bridging the previous experimental approaches by examining the ability of a single semi-volatile organic compound to co-condense and co-evaporate from inorganic particles over the entire subsaturated RH range (0–100 %), and under various conditions of particle mass and size, and the presence of a surfactant on the particles.”

Comment 6.

Introduction, lines 59-60 (and later in the manuscript). Please quantify what you mean by "semi-volatile" and "soluble", i.e. give the relevant values of saturation vapor pressures, aqueous-phase activities and water solubilities available in the literature. You will later on need these to make quantitative estimates on the relevant condensation and evaporation rates.

Response : To follow this suggestion, we modify the sentence starting on line 68 to:

“Levoglucosan (1,6-anhydro-β-D-glucopyranose, C₆H₁₀O₅) was chosen because it is a semi-volatile compound with a saturation concentration of $\log(C^) = 1.1 \mu\text{g}/\text{m}^3$ at 298 K (Zhang et al., 2024; Booth et al., 2011). It has an aqueous solubility of 61–63 wt% (Lienhard et al., 2012; Osmanbegovic et al., 2023) and its supercooled liquid is miscible with water (Parsons et al., 2004).”*

Comment 7.

Table 1 and the associated discussion: The number concentrations of particles in the chamber are rather high. What does this mean for the importance of a) wall-losses; b) coagulation; c) gas-phase saturation wrt levoglucosan in interpreting your results? Please add a discussion of this.

Responses :

a) The wall-losses of particles mainly depend on their size (Alfarra et al., 2023). In CESAM, due to the non-electrostatic (stainless steel) wall surface material and its low surface-to-volume ratio, the 1st order decay of particles of 100-200 nm is of the order of $5 \cdot 10^{-5} \text{s}^{-1}$, corresponding to a lifetime of 5h30 (see for details our response to reviewer #2, comment 16f).

b) coagulation: We can rule out the influence of particle coagulation because the particle size distribution remained stable. Specifically, we observed no significant increase in the mode/mean diameter after the injection: In Figure 2 (Exp 1a. Pure AS) with initial particle number concentration

of $2.33 \times 10^4 \text{ cm}^{-3}$, no significant coagulation effect has been observed, i.e., a mode size of $90 \pm 5 \text{ nm}$ to $92 \pm 5 \text{ nm}$ during 5 hours.

c) gas-phase saturation wrt levoglucosan : This is a good question and it will be part of the model study (part 2 paper). Preliminary model simulations have indeed shown that gas-phase saturation wrt levoglucosan is not reached because the gas-phase wall loss is too substantial. Based on Alfara et al. (2023) the gas-phase wall loss of levoglucosan follows a 1st order rate constant $k_{\text{gas levo}}$ of the order of 4.10^{-4} s^{-1} corresponding to a lifetime of $1/k_{\text{gas levo}} = 42 \text{ min}$ as mentioned in more detail in our response to reviewer #2, comment 16.

Comment 8.

Methods, lines 237-240: Why did you dry the particles prior to the measurement - isn't there a risk of losing some of the semi-volatile material? Why did you not use a humidified DMA system? As is pointed out in the manuscript, the humidity control is a critical element for measuring what the authors intend to measure. Unless I have severely misunderstood something, I would strongly encourage the authors to consider repeating the experiments with a better RH control throughout the measurements, including the sizing. This is in my view a critical concern for the validity of the results.

Response : Actually, both humidified DMA and dry DMA systems were used (section 2.5.2): Figure 2 shows the comparison of AS particle size ($D_{\text{ve_wet}}$) obtained by the wet SMPS system and that ($D_{\text{ve_dry_raw}}$) obtained by the dry MPSS system.

To better estimate the mass concentration of levoglucosan and ammonium sulfate in the particle phase, total dry mass measured by the MPSS and the mass ratio given by the AMS have been associated (section 2.5.2). So that the drying process has been applied to avoid the overestimation of total mass concentration. The residence time of aerosols in the MPSS system is usually a few seconds. Compared to our observations, i.e., evident levoglucosan condensation or evaporation in hours, the modification of particle size in the dry MPSS is negligible.

According to our observations, if we take the maximum of the net evaporation rate of 29.3 s^{-1} (Exp 3f and Exp 3h), for a particle size of 200 nm and 100 nm, the mass loss, in both cases, during the dry MPSS (10 s) is around 0.2% and the modification of the particle size is 0.07% and can be neglected.

The humidity control is a critical element, and we have done our best to control it throughout all experiments.

Comment 9.

Conclusions, lines 570-572: The authors state "The results showed that the net evaporation rate of levoglucosan was sensitive to all the investigated parameters, but the main drivers under our experimental conditions were the levoglucosan-to-sulfate mass ratios and RH." Unfortunately, I find this statement rather uninformative, given the potential of the data collected (especially if the RH control is taken care of). However, with the appropriate quantitative frameworks and potential further experimental analysis with a more careful humidity control, I believe the authors can do much better than this to find clearer and more impactful contributions to the

present knowledge about atmospheric aerosols and their thermodynamics. I hope that the suggestions given above will help with this task.

Response : *We have replaced the sentence “The results showed that the net evaporation rate of levoglucosan was sensitive to all the investigated parameters, but the main drivers under our experimental conditions were the levoglucosan-to-sulfate mass ratios and RH.”*

By:

“This experimental chamber study allowed us to discuss the complex interrelation between particle size, total mass concentration, relative humidity, levoglucosan/ammonium sulfate ratio, presence of a surfactant, and temperature. The results showed that levoglucosan-to-sulfate mass ratios and RH were the main drivers of the net evaporation rate of levoglucosan from AS particles.”

Comment 10.

Conclusions, line 596: I find that the outlook statement about the "modeling study" does not add much without some further elaboration. What kind of modeling do the authors mean, exactly? Please clarify.

Response : *We improve the conclusions by removing the last sentence of the manuscript and replacing it by a more detailed description starting on line 585 (after: “from or onto AS particles.”):*

“To disentangle the influence of RH, levoglucosan-to-sulfate mass ratio, total particle number, and temperature on the levoglucosan evaporation and condensation rates, we performed a theoretical study using a box model of these experiments that takes gas- and particle-phase wall losses and solution non-ideality into account, see the Part 2 study, Luo et al., in prep.”

References:

Taina Yli-Juuti, Alessandro A. Zardini, Axel C. Eriksson, Anne Maria K. Hansen, Joakim H. Pagels, Erik Swietlicki, Birgitta Svenningsson, Marianne Glasius, Douglas R. Worsnop, Ilona Riipinen, and Merete Bilde

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Reviewer #2 :

This manuscript describes a set of experiments conducted in the CESAM chamber to study evaporation and condensation of levoglucosan to/from aqueous solutions droplets under varying conditions of relative humidity and initial dry particle size.

The topic is relevant and the experimental idea interesting. However, I find however that the manuscript in its current form is very difficult to read and that it needs to be reconsidered after clarifications and major revision.

First, I find that the description of the experiments and the corrections need clarification, see further comments and some suggestions for improvement below.

Second, in some places, statements are given without literature references, clear description of assumptions, or without support by thermodynamic calculations which could be done using online-tools (for example using AIOMFAC or E-AIM). The authors could attempt to constrain or model droplet and gas phase concentrations of levoglucosan from known properties. I suggest it is better explained in the context of existing knowledge how the experiments contribute new insights.

Response : *We fully agree that for a full rationalization of the experimental data, thermodynamic calculations are required. The semi-quantitative analysis performed in this study shows the complex interrelation between total mass concentration, RH, levoglucosan/ammonium sulfate ratio, presence of a surfactant, and temperature, and it requires a simulation study to disentangle these interrelations.*

AIOMFAC is not suited to describe the non-ideality of the ammonium sulfate/levoglucosan/water system as it predicts a phase separation that was not observed in the experiments (Parsons et al., 2004). We remeasured aqueous ammonium sulfate/levoglucosan droplets in collaboration with Ulrich Krieger (ETH Zurich) to optimize a Pitzer model for this system. The description of these measurements would make the present manuscript too lengthy. We therefore decided to describe the model and the simulations in a separate manuscript that is in preparation. To connect the experimental and simulation studies more closely, we intend now to publish them as Part 1 and Part 2. See the response to the general comment of reviewer #1.

Third, I find that the authors make quite firm conclusions, and in the current form of the manuscript it is hard to see that they are all fully justified. For example, as I understand, the conclusion on co-condensation is based entirely on one experiment (3d) and significant corrections were done to arrive at Dve due to technical issues with an RH sensor. In the conclusion section, the authors provide numbers on atmospheric lifetimes which I do not see are explained. Some suggestions for improvement are given below.

Response : *We had to concentrate on co-evaporation in this study because ammonium sulfate is an efflorescing substance with a deliquescence RH of 80 %. To obtain liquid particles, the experiments had to start above this RH and then be decreased. Given the large chamber volume,*

RH changes are slow. Thus, particle concentrations became too low to be increased again within the same experiment. Moreover, from a thermodynamic viewpoint, co-condensation and co-evaporation are equivalent. Thus, from the observation of co-evaporation, co-condensation can be implied for increasing RH.

Methods

Comment 1.

Introduction of dry monodisperse particles into the chamber: If I understand correctly, dry particle diameter was selected by AAC and converted to electrical mobility diameter. This was checked by SMPS – how well did the two methods correspond -what were actual numbers in terms of e.g. geometric mean diameters obtained? Most places it says “~” when a diameter is given – what were the actual values measured?

Response : When an aerodynamic diameter (d_a) of 300 nm and an effective density of 1.77 g cm^{-3} were set on the AAC, the expected mobility diameter (d_m) was 207.7 nm, as outlined in the AAC manual (Section A.4, Mobility Diameter Conversion). The mobility diameter (d_m) actually measured by the SMPS was 209.1 nm, as shown below (from Experiment 3d).

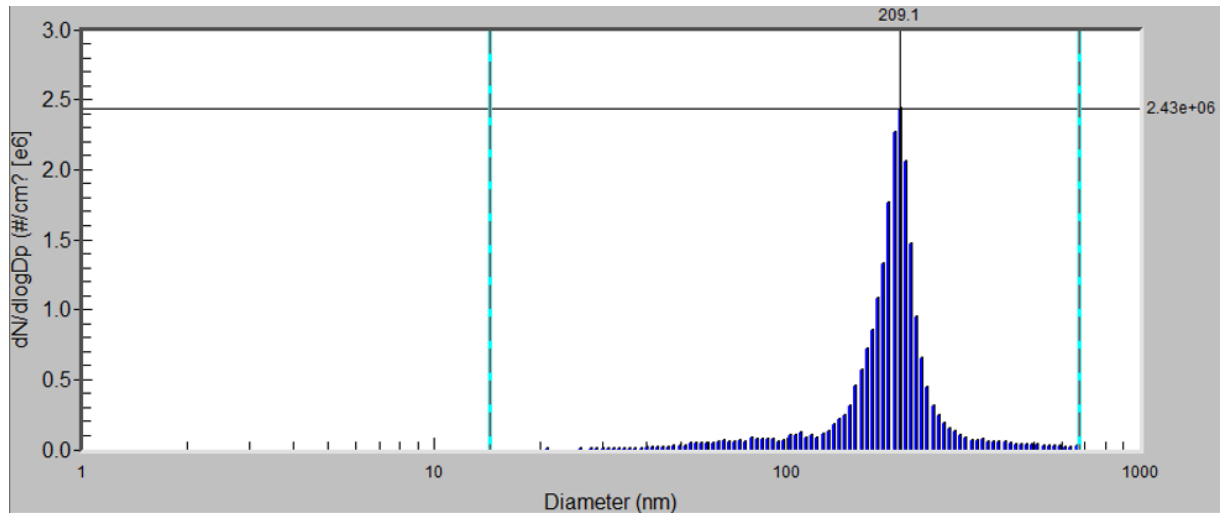


Fig A : SMPS size distribution checked during exp 3d prior to aerosol introduction in the CESAM chamber.

The procedures is given below:

According to DeCarlo et al. (2004), the conversion from d_a to d_m can be calculated by:

$$d_a = d_m \sqrt{\left(\frac{\rho_{eff}}{\rho_0}\right) \cdot \left(\frac{C_c(d_m)}{C_c(d_a)}\right)}$$

where d_a is the aerodynamic diameter; d_m is the electrical mobility diameter; ρ_{eff} is the effective density of the particle; ρ_0 is the standard reference density; $C_c(d)$ is the Cunningham slip correction factor for diameter d . The Cunningham slip correction factor $C_c(d)$ for a particle of diameter d is given by:

$$C_c(d) = 1 + \frac{2\lambda}{d} [\alpha + \beta \cdot \exp(-\frac{\gamma \cdot d}{2\lambda})]$$

where λ is the mean free path of air molecules (approx. 66.5 nm); d is the particle diameter; $\alpha = 1.142$; $\beta = 0.558$; and $\gamma = 0.999$. Because the Cunningham slip correction factor $C_c(d_m)$ depends nonlinearly on the unknown $d_{m,0}$ a fixed-point iteration scheme was implemented. Starting with an initial estimate

$$d_{m,0} = d_a \sqrt{\frac{\rho_0}{\rho_{eff}}}$$

$C_c(d_m)$ was iteratively updated until d_m converged to within 0.01 nm. The computed d_m was then compared to the experimental mode mobility diameter obtained from the SMPS size distribution.

Comment 2.

The dry size distribution was measured using the MPSS system. Due to a problem in the drying system, a correction was used to get the dry size distribution. The description of the correction and the related uncertainties is substantial, but difficult to follow and could be explained more clearly. I miss figures showing the actual and corrected particle size distributions.

Response : *Indeed, the detailed description of the correction procedure might look more complex than it is. In principle, we just subtracted the water content predicted at the RH measured in the MPSS from the particles based on the growth factors measured by Lei et al. (2018) for mixed AS/levoglucosan particles. To make this clearer, we did the following changes:*

- We added the following sentence on lines 279-282 (changes in blue):

“It was thus necessary to adopt a methodology of particle size correction to obtain dry particle size and mass concentrations (even though it provided additional uncertainties), developed in SI 5. To obtain particle diameters that represent completely dried particles, the increase in diameter predicted based on the hygroscopic growth at the RH measured in the MPSS was subtracted to obtain dry diameters based on the growth factors measured by Lei et al. (2018) for mixed AS/levoglucosan particles. The correction procedure applied is described in detail in SI 5-4.”

Figure 2 shows the comparison between uncorrected and corrected particle size for Exp 1a, performed with pure AS particles.”

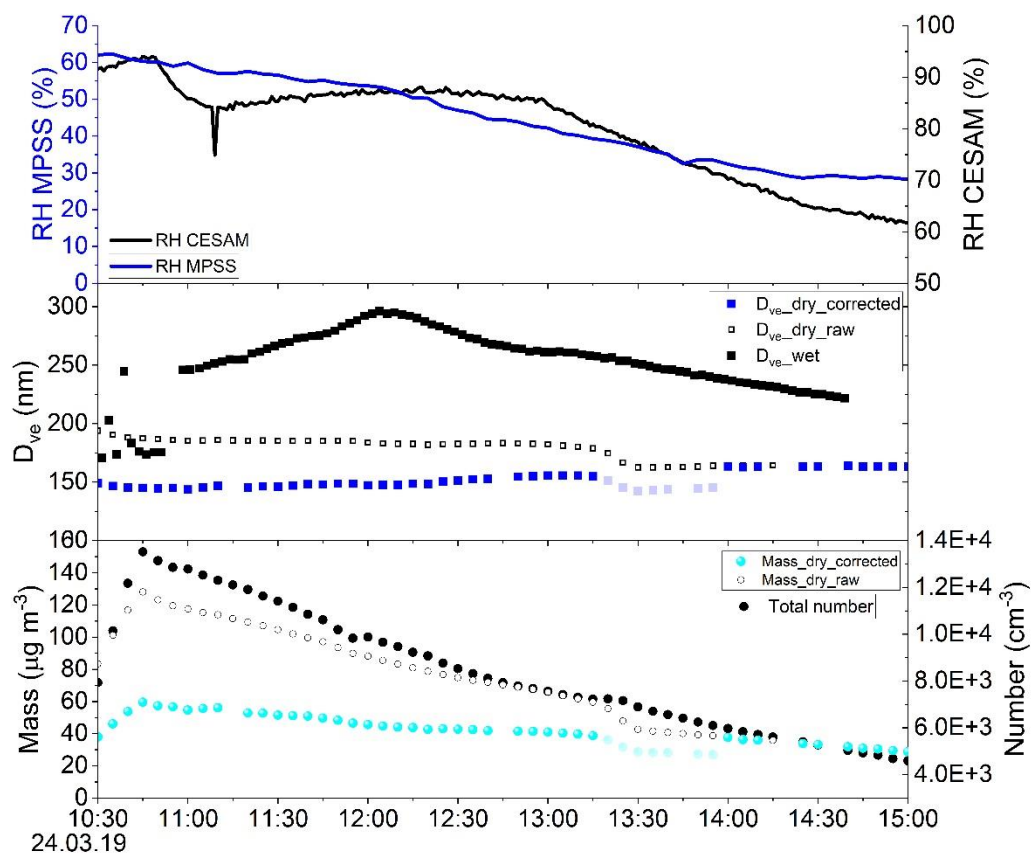
- We sent the correction procedure in the supplementary information SI 5-4
- We added the figures showing the comparisons between uncorrected and corrected particle size for all experiments (other than Exp 1a) in section SI 5-5 of the supplementary information, as shown below:

“SI 5-5 Comparison between raw particle size and corrected particle size

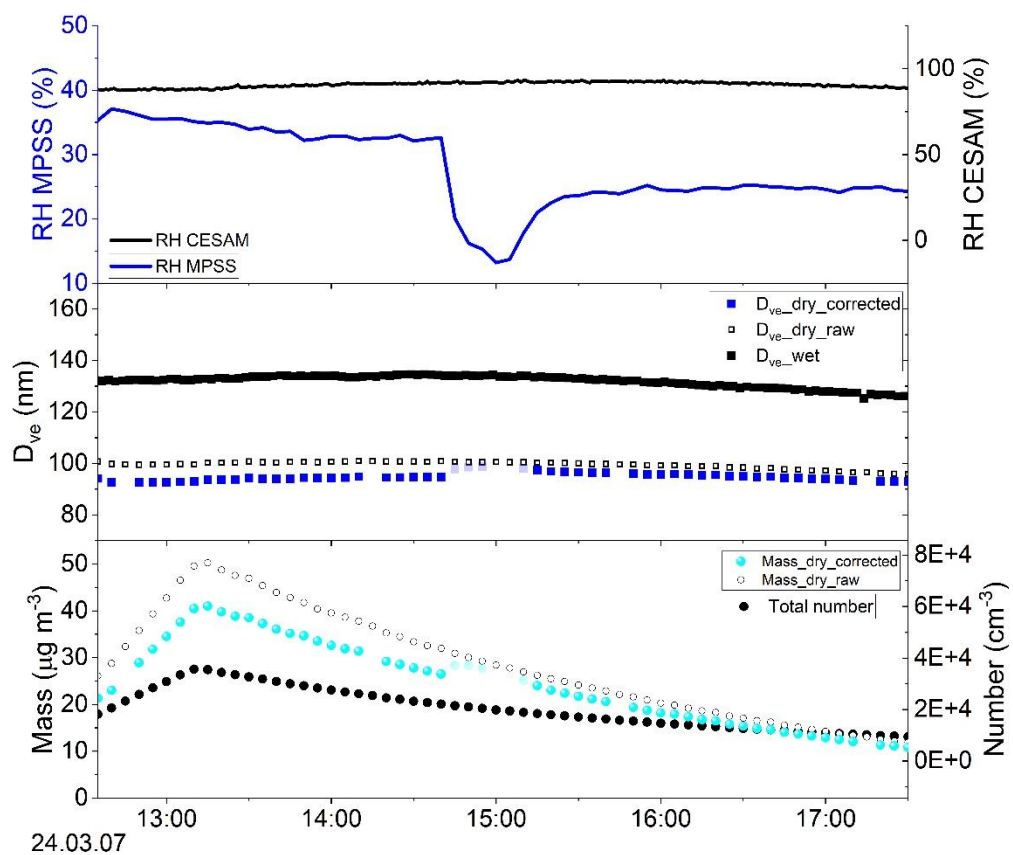
Table S 5-2: Comparison between raw particle size and corrected particle size

Exp.	Figures
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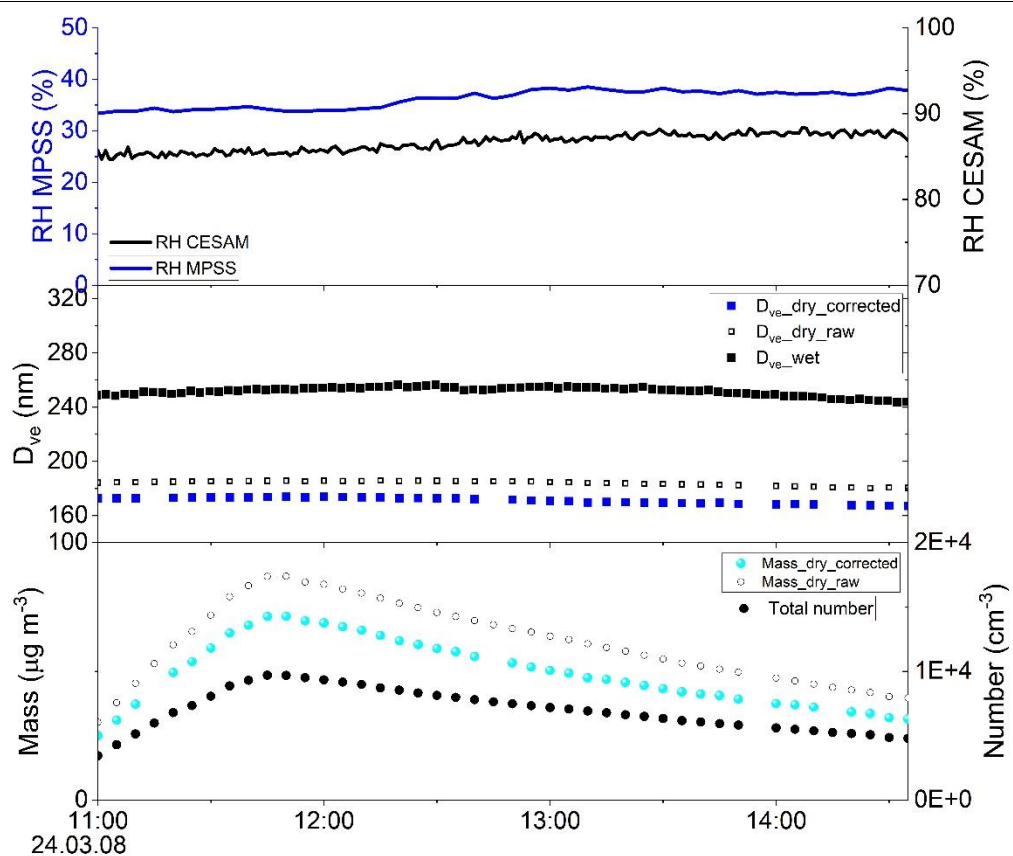
1b



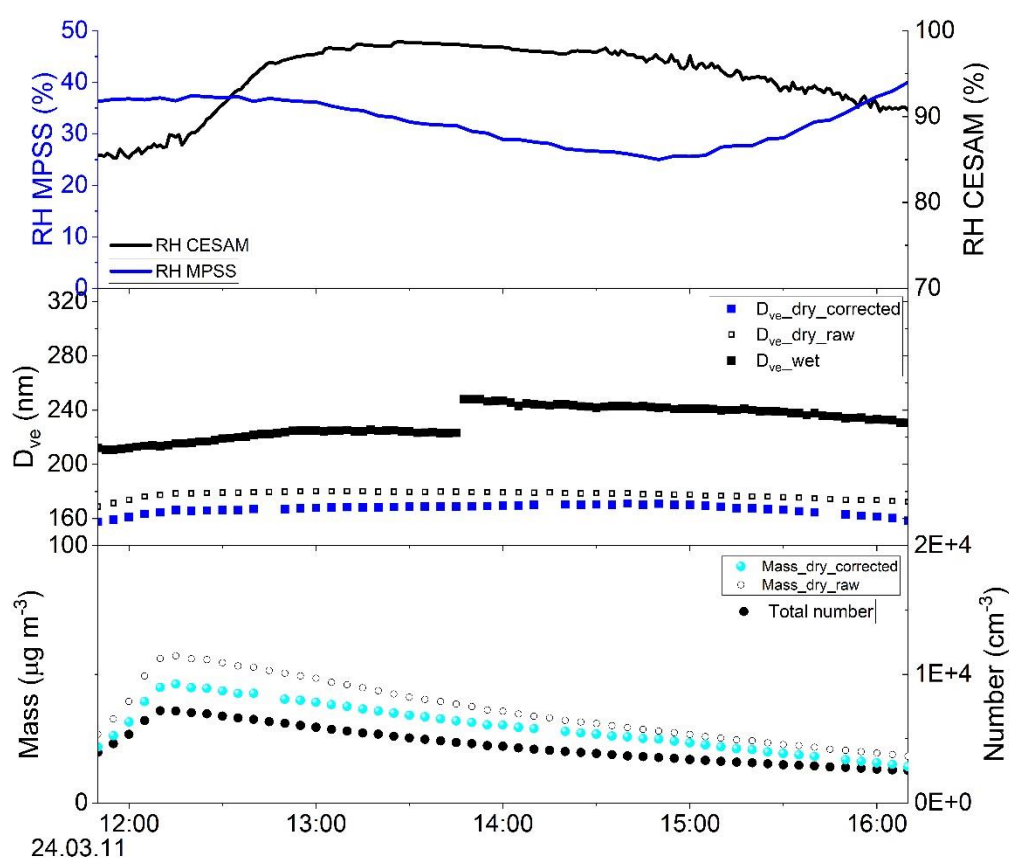
3b



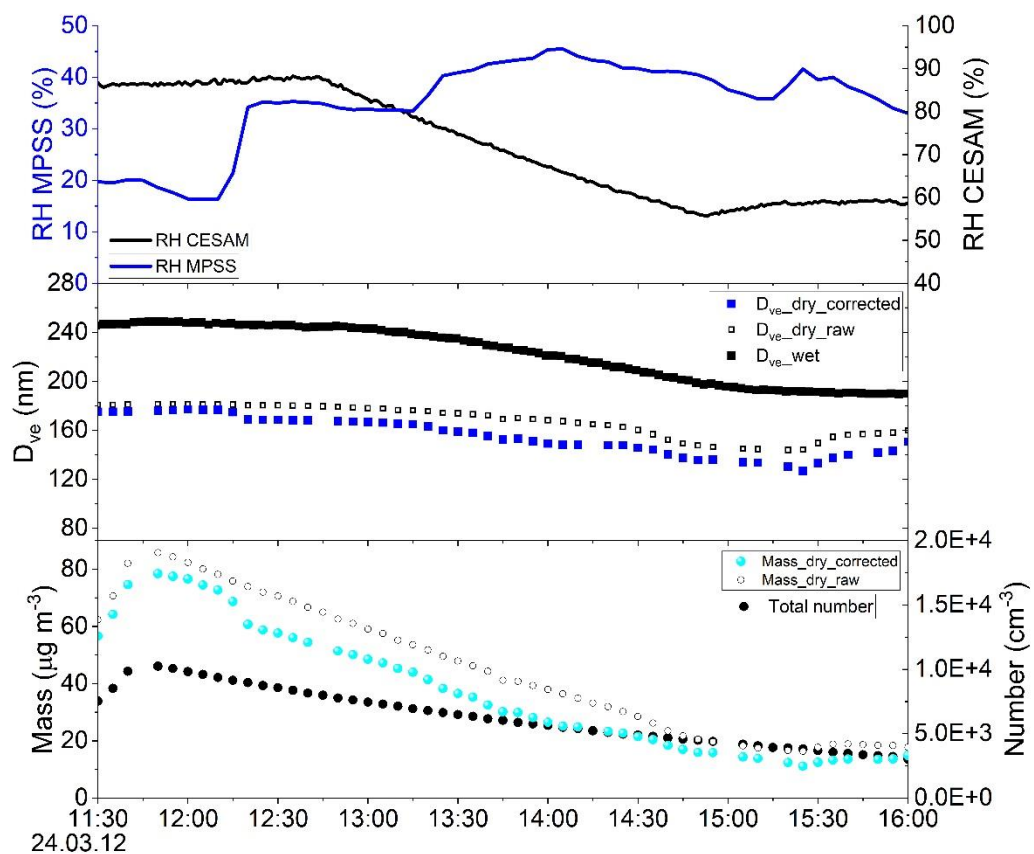
3c



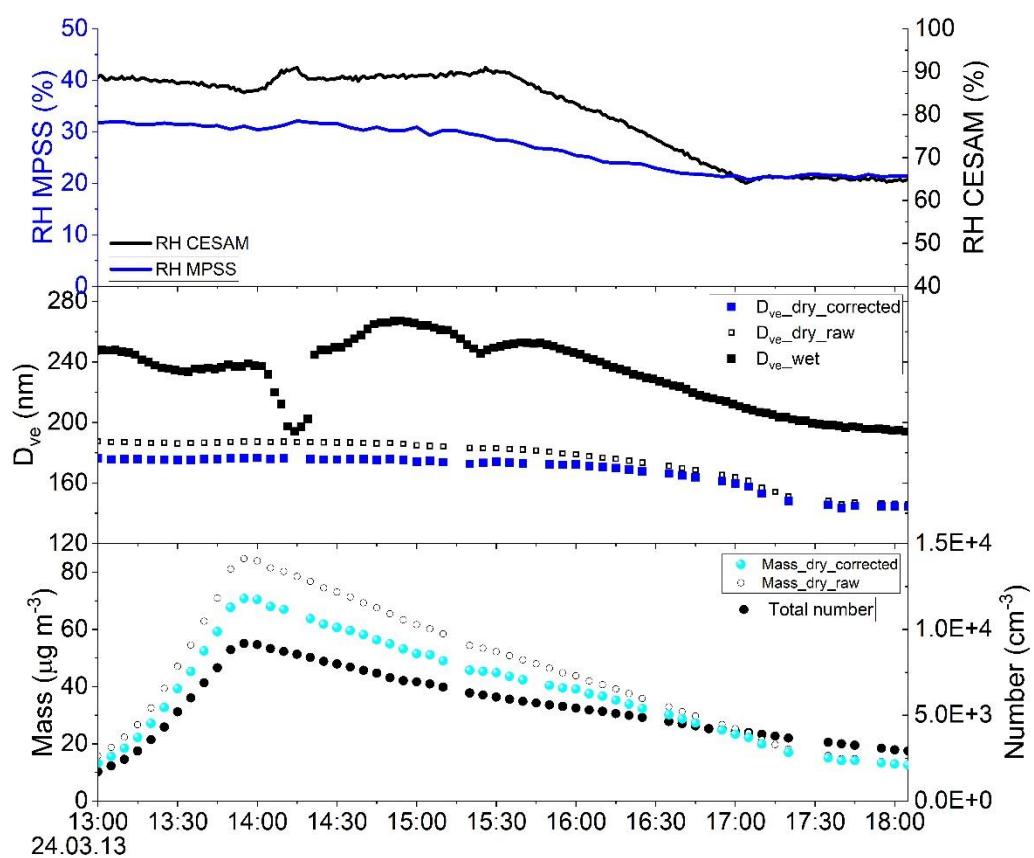
3d



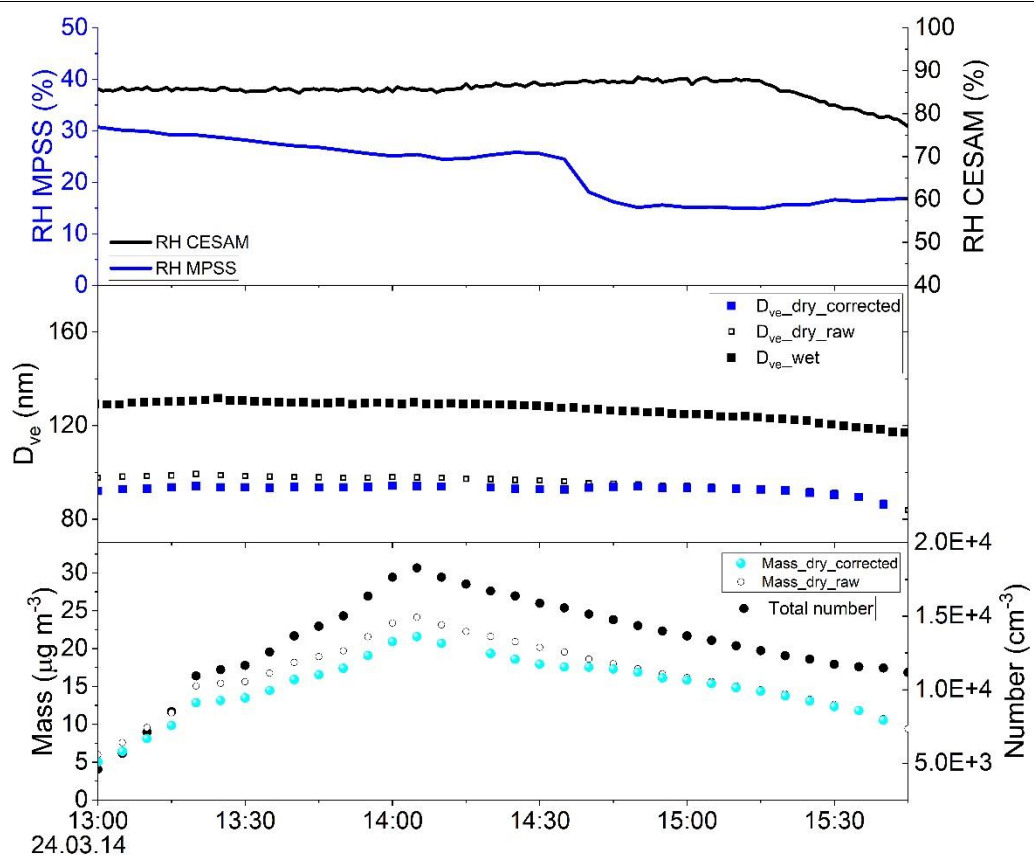
3e



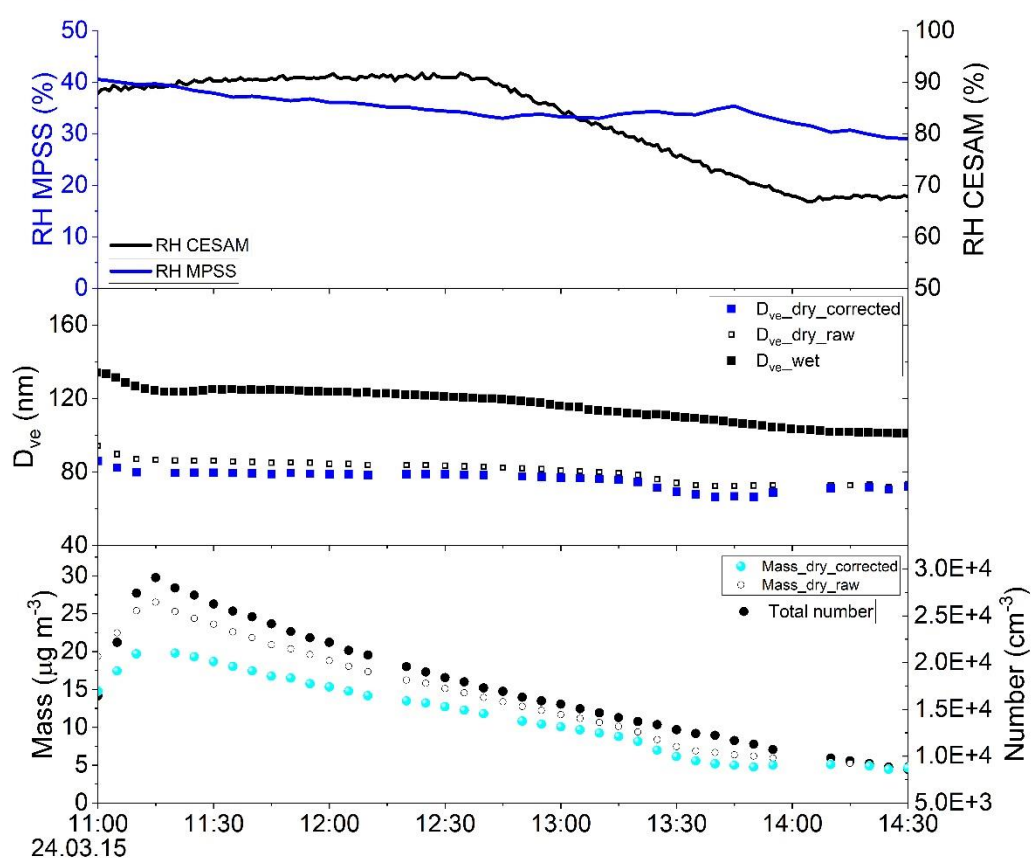
3f

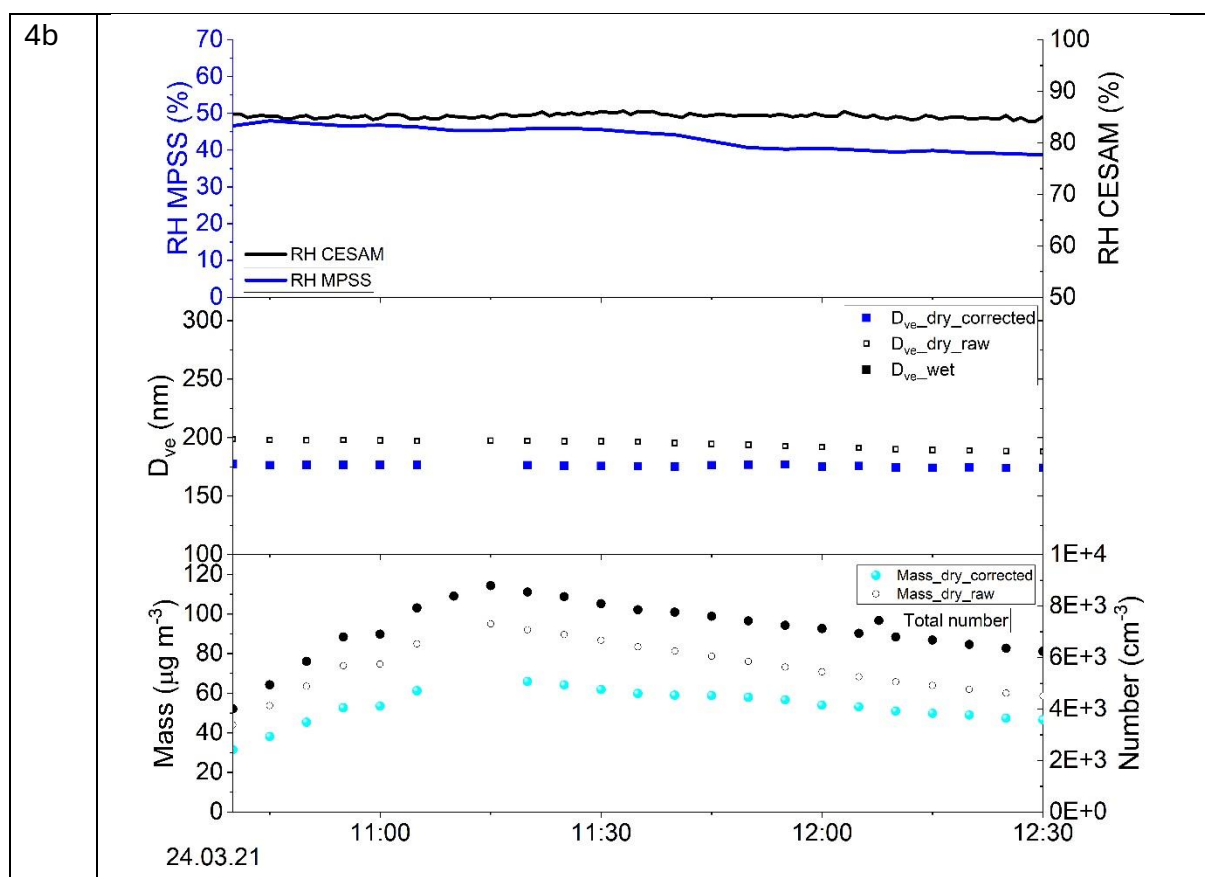


3g



3h





Comment 3.

If the authors have the possibility to repeat some of the experiments with correct drying (in particular exp. 3d) and thus avoid the corrections it would significantly strengthen the discussion and conclusions that can be made.

Response : Unfortunately, it is not possible to repeat this large experimental work that required a large collaborative team, gathering all the instruments thanks to dedicated funding.

Comment 4.

Wet particle size during experiments were monitored using an SMPS system. Was the relative humidity of the sheath air controlled or could it be different than the relative humidity in the aerosol flow coming in ? If so – what uncertainties does that introduce on derived growth factors?

Response : The relative humidity (RH) of the sheath air was not controlled, resulting in a difference between the sample flow RH (considered as same as in the CESAM chamber) and the sheath flow RH (monitored). This difference occurred due to two factors: 1) the SMPS was operated in internal circulation mode, and 2) the sheath-to-sample flow ratio was 10:1. Consequently, there was a delay in the response of the sheath flow RH relative to the sample flow RH. RH in the CESAM chamber and RH in the sheath flow are shown in Figure 2 (top panel) with Exp 1a taken as an example.

Comment 5.

I suggest a schematic (could be in supporting material) showing all the instruments and where and when they were connected with flow rates given as well as indications for RH in different parts of the system and where it was measured.

Response : To follow this suggestion, we added the following Figure and Table in SI 1:

“SI -1: Schematic of the CESAM chamber connected to the various instruments

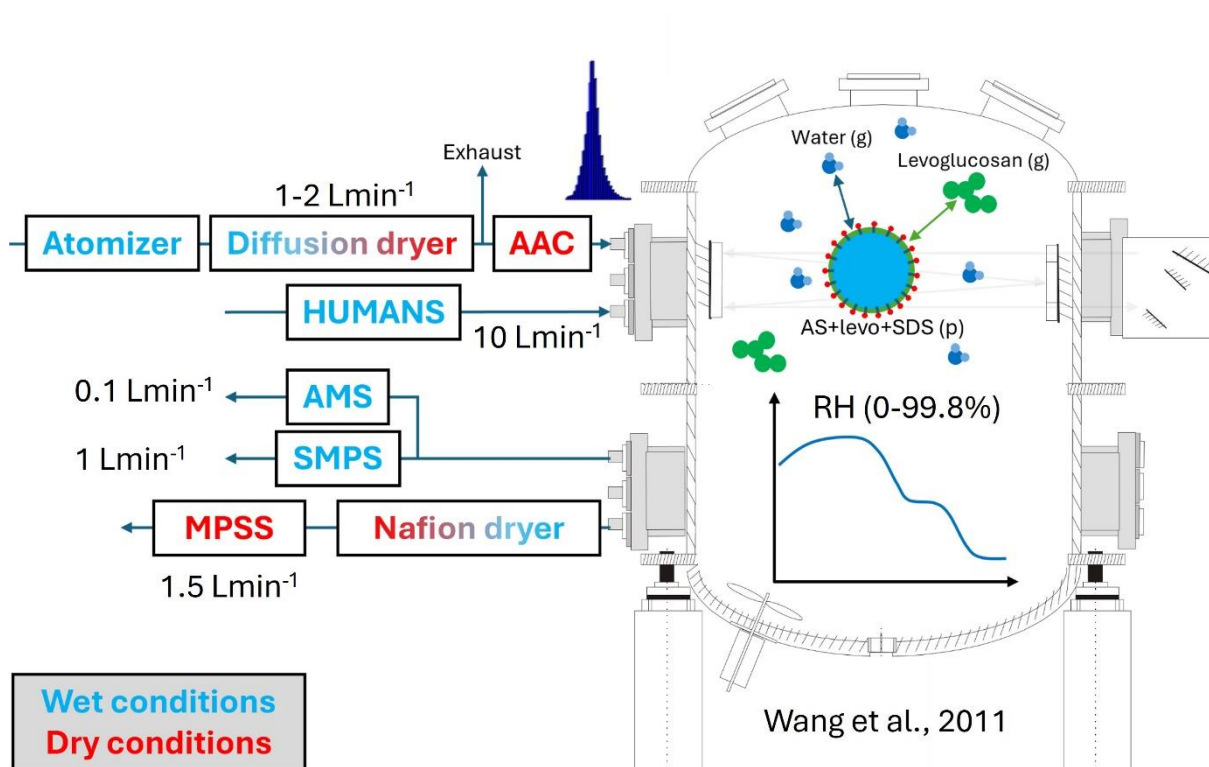


Figure SI 1-1: schematic of the CESAM chamber connected to the various instruments used for this study.

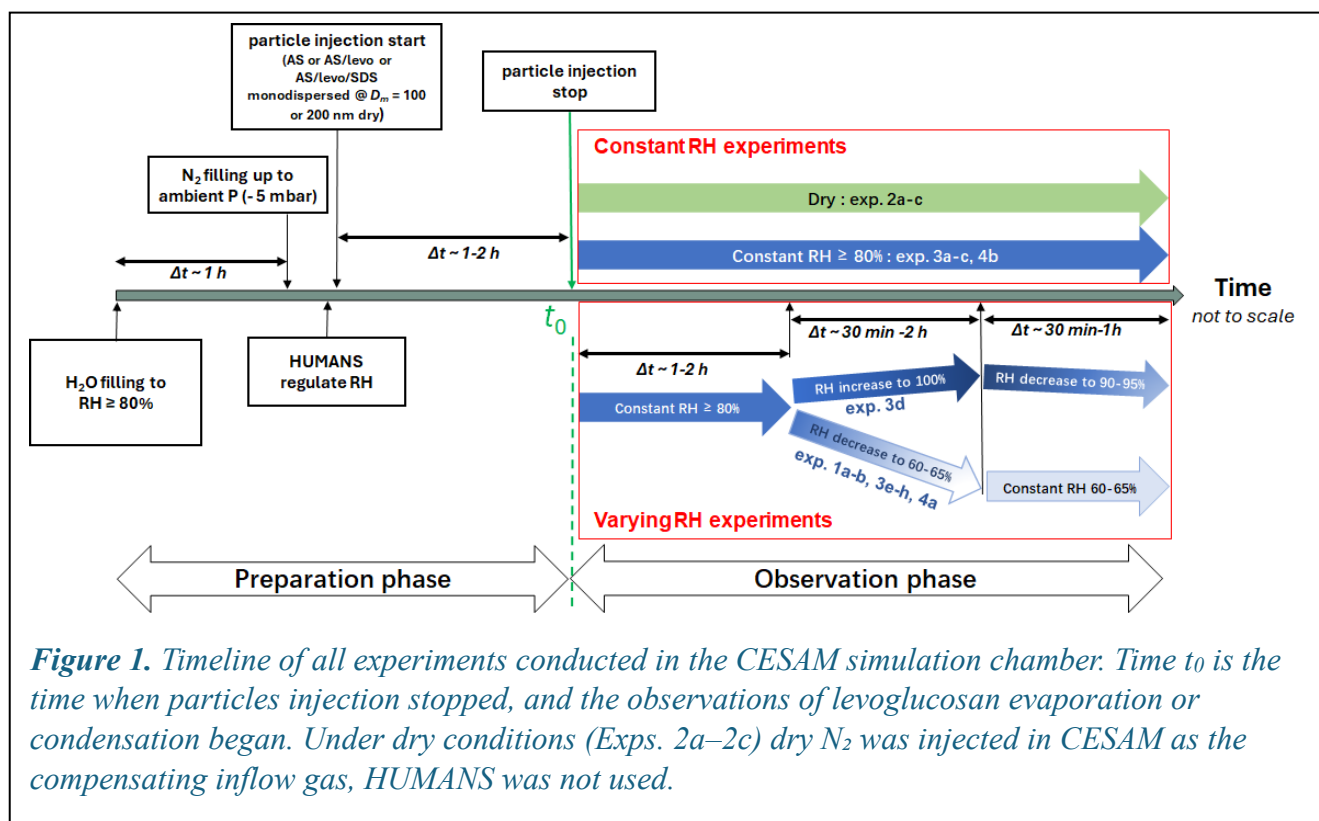
Table SI 1-1 : Instruments connected to CESAM with their flow rates:

	Instrument	Company/Model	Flow (L/min)	Comments
Flow out	CPC	TSI	1.5	0.3 or 1.5 (starting from 6/3/2024)
	SMPS (wet)	TSI	1	
	SMPS (dry)	DMPS	1	
	AMS	Aerodyne	0.1	HR-ToF-AMS
	Others		7.4	not used for the present study
Flow in	AAC	Cambustion	1	~ 1 L/min goes into CESAM
	CESAM	home-built		1 mbar P_{diff} during injection, 5 mbar otherwise
	HUMANS	home-built	10	~ 10 L/min to maintain high RH

Comment 6.

I miss a clear timeline for each type of experiment. What was the conditions during exp 1a,b? at what times was the RH varied and how? Similar, please also give an explanation of the timeline in other experiments where RH was varied.

Response : We have added a timeline of all experiments (see below), and we have placed it as Figure 1 instead of the description of HUMANS, which we have decided to move to the SI (following the comment #14 of this reviewer)



Comment 7.

I suggest table 1 includes : 1) the size aerodynamic size selected by the AAC?, 2) the corresponding calculated mobility size, and the volume equivalent size. Please include a column stating whether RH was constant or varied during a given experiment (and if it was varied, give the ranges).

Response : The size selected by AAC was always the same for each series of experiments (either 170 or 300 nm). The corresponding mobility size is explained in the text, but is different from the mobility size measured in CESAM, which is the focus of Table 1. Concerning RH, Table 1 was modified according to the reviewer's comment. It now looks as below (changes are highlighted in cyan in the Table).

Table 1. Summary of the experimental conditions. NA = non available; k_{net} is the net evaporation rate of organic matter from AS particles, determined during 35 to 75 min (= "effective duration") from the exponential fitting of the time profiles of organic-to-sulfate mass ratio $f_{o/s}$ (see Sect. 2.6). Number concentration, dry particle size, total mass concentration and organic to sulfate ratio are given at time

0, when the introduction of particles stopped. *T* is the average temperature during the effective duration. For experiments conducted at constant RH, the average RH value is given, for the others, the initial → final values are given.

Exp no.	Aerosol type	T (°C) ±0.1	RH (%)	[number] (10 ⁴ #/cm ³) ±5%	D _{ve} dry* (nm) ± 7 %	[mass]* (μg.m ⁻³) ± 26%	OA/sulfate (f _{o/s})	k _{net} (× 10 ⁻⁵ s ⁻¹)	Effective duration (min)	Exp. date yy/mm/dd
1a	AS	24.0	89.4 → 40.1	2.33	90.1	15.7	0.14 ± 0.01	NA	75	24/03/18
1b	AS	24.1	94.1 → 61.8	1.35	146	38.6	0.091 ± 0.001	NA	75	24/03/19
2a	AS/Levo	21.9	0	1.01	164	32.6	1.79 ± 0.01	1.66 ± 0.01	75	24/03/01
2b	AS/Levo	21.7	0	1.09	174	38.8	2.10 ± 0.01	1.64 ± 0.02	75	24/03/04
2c	AS/Levo	23.0	0	4.37	98.7	23.8	1.66 ± 0.01	5.41 ± 0.05	75	24/03/05
3a	AS/Levo	22.4	88.7	1.89	NA	NA	2.29 ± 0.02	6.00 ± 0.09	75	24/03/06
3b	AS/Levo	22.4	90.2 ± 1.0	3.55	93.7	18.8	2.13 ± 0.02	4.36 ± 0.06	75	24/03/07
3c	AS/Levo	22.4	86.4 ± 0.6	0.969	174	32.3	2.33 ± 0.01	3.36 ± 0.05	75	24/03/08
3d	AS/Levo	23.1	88.2 → 100.1	0.715	166	29.7	2.08 ± 0.01	-1.15 ± 0.10	35	24/03/11
		23.2	100.7 → 98.8	0.446	169	19.4	2.13 ± 0.02	0.91 ± 0.09	34.5	
		23.4	97.1 → 94.9	0.331	169	14.6	1.99 ± 0.02	4.44 ± 0.14	34	
3e	AS/Levo	23.8	86.7 ± 0.4	1.02	175	49.8	2.38 ± 0.01	3.26 ± 0.06	30	24/03/12
		23.7	87.4 → 74.6	0.664	160	24.7	1.76 ± 0.03	11.5 ± 0.3	35	
3f	AS/Levo	24.4	88.2 ± 1.5	0.918	177	45.4	2.39 ± 0.01	4.21 ± 0.07	45	24/03/13
		24.5	90.0 → 82.6	0.605	174	28.6	1.88 ± 0.02	11.6 ± 0.2	30	
		24.8	65.2 ± 0.4	0.405	159	14.8	0.82 ± 0.02	29.3 ± 0.8	30	
3g	AS/Levo	23.8	86.3 ± 0.6	1.77	93.9	13.7	1.54 ± 0.02	15.9 ± 0.3	30	24/03/14
3h	AS/Levo	24.4	90.4 ± 0.5	2.92	79.8	13.3	2.25 ± 0.04	14.2 ± 0.3	40	24/03/15
		24.5	90.1 → 75.9	1.73	78.0	7.2	1.11 ± 0.03	29.3 ± 1.0	50	
4a	AS/Lev/S DS	24.5	88.5	NA	NA	NA	3.45 ± 0.02	6.34 ± 0.18	45	24/03/20
		24.7	87.2	0.742	186	37.6	3.30 ± 0.03	14.7 ± 0.6	30	
4b	AS/Lev/S DS	25.6	85.5 ± 0.3	0.879	180	41.6	3.28 ± 0.02	9.14 ± 0.33	35	24/03/21

* Corrected data (see Sect. SI 3-4 in the supplementary information)

Section 2.6.

Comment 8.

The assumptions behind equations (3) and (4) should be stated.

Response : The paragraph has been changed to explain in detail the expected (and observed) 1st order kinetics behavior of the compounds.

It now reads:

“The net evaporation rate of levoglucosan from the AS particles was determined by measuring the mass concentrations of sulfate and levoglucosan using the AMS. All experiments were performed in the dark without any reactants, and thus, once introduced in the chamber in the particle phase (at time 0), both compounds were not removed by chemical reactions, but only 1st order kinetics phenomena induced their concentration decreases versus time, i.e. dilution, wall loss processes, and gas-particle partitioning (Seakins et al., 2023). Due to their different physical properties, these phenomena occurred at different rates for each compound. And thus, sulfate and levoglucosan concentrations decreased with time with 1st order kinetics, at different rates:

$$[S]_t = [S]_0 \exp(-k_1 t) \quad (3)$$

$$[OA]_t = [OA]_0 \exp(-k_2 t) \quad (4)$$

where $[S]_t$ is the sulfate mass concentration at time t , and $[OA]_t$ the particle-phase levoglucosan mass concentration at time t (time 0 is the time when the chamber filling with aerosol was finished); k_1 is the decay rate constant including wall loss processes and dilution of sulfate, and k_2 the combined decay rate constant of wall loss processes and dilution, as well as gas-particle partitioning of levoglucosan.”

An example of the exponential fittings is shown below for EXP 2a (Figure 3 left panel). However, We did not want to show these fittings in Figure 3 and 4 of the manuscript for the sake of clarity of the figures.

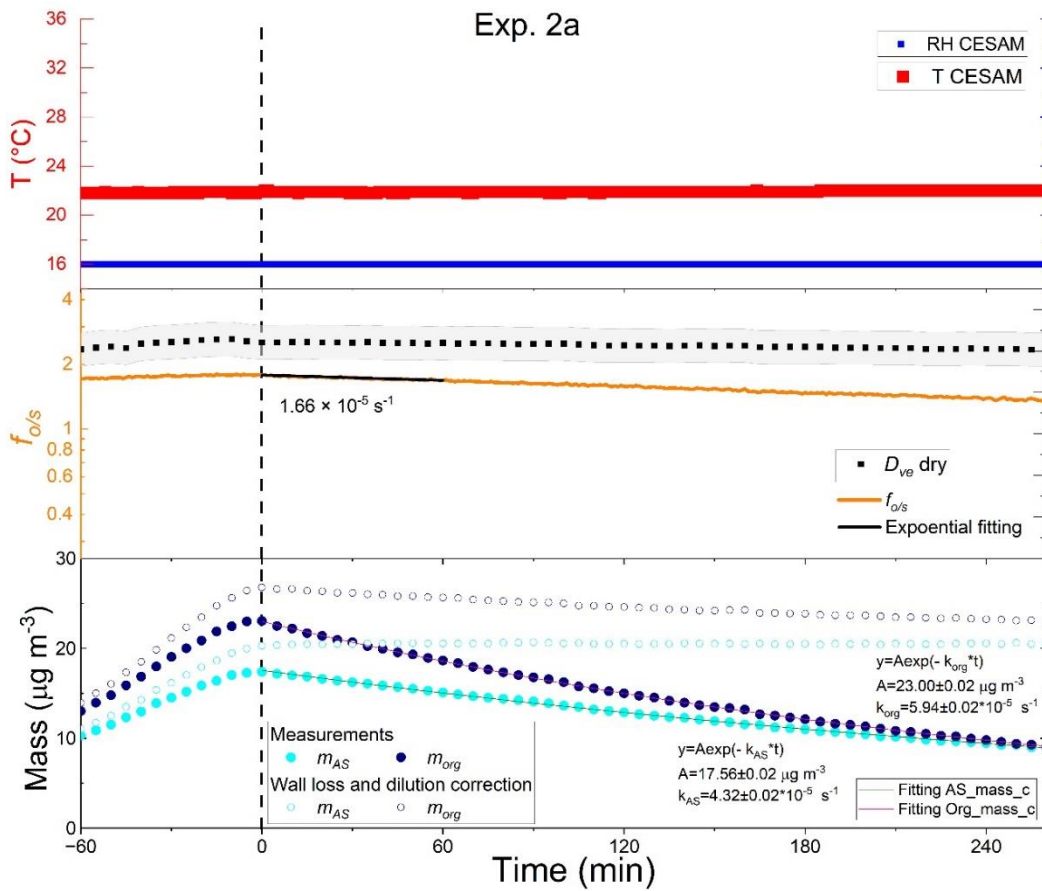


Figure 3 (left panel): Evaporation of levoglucosan during Exp 2a. Time series of T , RH (top row), volume equivalent dry particle size (D_{ve}), organic to sulfate mass ratio ($f_{o/s}$) (middle row), masses of AS (m_{AS}) and total organic (m_{org} , attributed to levoglucosan) with and without wall loss and dilution corrections (bottom row). The grey shades show the uncertainty of D_{ve} after correction (see SI 5). The black lines indicate the exponential fitting curves (during 60 min) to determine k_{net} (according to Eq. 4), which values are shown. The vertical dashed line marks time 0, at the end of particle introduction into CESAM. Exponential decays of the mass concentrations are added : the exponential decay of the total organic mass (levoglucosan) is $5.94 \pm 0.02 \times 10^{-5} \text{ s}^{-1}$, and that of ammonium sulfate is $4.32 \pm 0.02 \times 10^{-5} \text{ s}^{-1}$)

Comment 9.

I miss the explanation for how to come from the change in particle mass with time due to evaporation or condensation for a particle in the transition regime to a first order loss equation. What are the assumptions, is the Kelvin effect ignored?

Response : *To investigate the potential importance of the Kelvin effect, we have calculated the Kelvin factor under the experimental conditions :*

$$\text{Kelvin factor } K = \exp\left(\frac{4\sigma M}{\rho R T d}\right)$$

For $M=162.14 \text{ g mol}^{-1}$, $\rho = 1.6 \text{ g.cm}^{-3}$, $\sigma = 0.072 \text{ N.m}^{-1}$, $T=298\text{K}$ and d varying from 180 nm to 220 nm, one obtains the following values:

$d=180 \text{ nm}$, $K=1.029$

$d=200 \text{ nm}$, $K=1.026$

$d=220 \text{ nm}$, $K=1.023$

So, a difference of 10% on particle size induces a difference of 0.29% on the Kelvin factor

Comment 10.

Regarding figure 2 – in the text the steps of increasing RH are stated as 13.45 and 16.15, but the timescale in the figure has been normalized and it is not clear what time zero is.

Response : *We thank the reviewer for pointing this out. The text has been revised as “first decrease starting at 88 s and the second at 237 s.”*

Comment 11.

It says that the mass shows an exponential decay – this could be supported by showing an exponential fit.

Response : *See our answer to comment 8 of this reviewer, where Figure of EXP 2a is shown as an example with the exponential fits of total organic mass (levoglucosan) and that of AS. We did not want to show them in Figure 3 and 4 of the manuscript for the sake of clarity of the figures.*

Comment 12.

Why were some data removed from the figures and what is “the final corrected data” ?

Response : *In Figure 2, the blue solid dots represent the corrected dry Dve (middle panel) and the cyan solid dots represent the corrected total dry mass (bottom panel), using the correction protocol described in section 2.5.2. The efflorescence RH of pure AS is around 34%. During this period (shaded pink in Figure 2), a part of the particles has already effloresced while a part is still deliquesced. This decreases the accuracy of the correction in this RH range. The semi-transparent dots represent an under-correction for particles that have not yet deliquesced and these have therefore been excluded. So, the final corrected data are the solid dots (Blue for dry Dve and Cyan for dry total mass).*

The text has been changed and now reads (changes in blue):

“It was thus necessary to adopt a methodology of particle size correction to obtain dry particle size and mass concentrations (even though it provided additional uncertainties), developed in SI 5., (see SI 5). To obtain particle diameters that represent completely dried particles, the increase in diameter predicted based on the hygroscopic growth at the RH measured in the MPSS was subtracted to obtain dry diameters based on the growth factors measured by Lei et al. (2018) for mixed AS/levoglucosan particles. The correction procedure applied is described in detail in SI 5-4.

Figure 2 shows the comparison between uncorrected and corrected particle size for Exp 1a, performed with pure AS particles”

Comment 13.

Page 6, line 186 – which shape factor was used? Is there a reference?

Response : A dynamic shape factor (X) was not explicitly applied in the AAC parameterization. Instead, the effect of particle morphology is intrinsically incorporated into the effective density (ρ_{eff}). As detailed in Section A.4 (Mobility Diameter Conversion) of the AAC manual, the conversion between aerodynamic diameter (d_a) and mobility diameter (d_m) relies on the effective density, which bridges the two equivalent diameters without requiring an a priori assumption of the shape factor. Therefore, the shape factor is inherently accounted for within our effective density calculations. In the manuscript, we have replaced the sentence by:

“The mobility diameter was further confirmed using the effective density (ρ_{eff}) derived from the AMS data (DeCarlo et al., 2004)”

Comment 14.

Is the HUMANS system new – or why is it described in so much detail?

Response : The HUMAN system is new. It has been designed for the demanding control of RH in a large volume chamber. This is why it is detailed in this manner. However, we decided to move the figure to SI and to replace it with a schematic of the total setup following comment # 5 of this reviewer.

Comment 15.

Line 152: “typically” – which experiments were “typical” and which not?

There were several errors in the sentence. We have corrected the text, it now reads :

“The final RH of the flow was set to match the target RH of the chamber. To do so, the CESAM compensating flow (which is $\geq 10 \text{ L min}^{-1}$) was divided into two: i) a constant 10 L min^{-1} flow passed through HUMANS, and ii) the rest of the flow passed through a heated water bubbler.”

Comments 16.

Concentrations of levoglucosan in particle, droplet and gas phase:

- **Comment 16.a.** I miss some even simple calculations to constrain the budgets of gas phase and particle/droplet phase levoglucosan. For example, knowing the number

concentration of particles one could estimate the maximum gas phase concentration of levoglucosan in the chamber if all evaporated and at different stages on the way to this point.

Response : *We agree that the levoglucosan partitioning between the particle and the gas phase would be highly interesting for the various conditions studied. However, the total amount of levoglucosan introduced is not known precisely enough (see our response to comment 16.b) and the wall loss of gaseous levoglucosan is an order of magnitude faster than that of the particles (see our response to comment 16.f). This is the reason why we have focused the study on the net evaporation rates of levoglucosan. The gas-phase concentration of levoglucosan and its wall loss will be an important subject of the model study (Part 2 paper), as these quantities can be assessed based on a theoretical model of the chamber experiments.*

- **Comment 16.b.** In the atomizer bottle the mass concentrations of ammonium sulfate and levoglucosan are 1:1. Do the authors assume that the composition of the particles coming out of the atomizer are also 1:1? If so, what is the initial value of $f_{o/s}$? (based on line 215 I get $f_{o/s} \sim 1.19$ initially). If the value of 1.19 is correct for the initial composition, how is it possible that $f_{o/s}$ is higher than 1.19 at the start of the experiments if levoglucosan evaporated from the particles during the injection? (which would mean that $f_{o/s}$ should go down)?

Response : *The solution in the atomizer bottle contains 15.0 g/L of levoglucosan and 15.0 g/L of AS $(\text{NH}_4)_2\text{SO}_4$, thus 10.9 g/L of sulfate. Assuming that levoglucosan represents the organic matter, the mass fraction $f_{o/s}$ in the solution is thus $15.0/10.9 = 1.38$. We do not assume any $f_{o/s}$ based on the composition in the atomizer, we only rely on the values measured in the chamber at time t_0 , which is the time when the injection was completed, and the observations of evaporation (or condensation) started. Due to the length of the injection step in the chamber (see the new Fig. 1) evaporation and/or condensation do occur prior time t_0 . Due to its properties, levoglucosan partitions between the gas, the particles and the walls. At the start of the injection step, it is possible that levoglucosan evaporation is enhanced by wall loss that occurs at a higher rate than that of the particles (see our response to comment 16.f). Then, more and more particles (containing both AS and levoglucosan) are injected, providing more surface for levoglucosan to condense, altering the partitioning between gas/particle/walls. This may explain the increase of $f_{o/s}$ in the chamber above the theoretical value of 1.38 during the chamber filling (up to time t_0 ; see the middle panel of Fig. 4). However, we focus the study on the observation phase when the injection step is over (time $> t_0$).*

To clarify these points, we have modified the text accordingly and we replaced Figure 1 by a timeline of the experiments conducted in the CESAM chamber. The figure is followed by a brief note: “Note that, for the experiments conducted with AS and levoglucosan, the $f_{o/s}$ values range from 1.11 to 2.39 at time t_0 (Table 1), which differs from the value of 1.38 in the nebulizing solution. This phenomenon can be explained by the variable duration of the injection phase into the chamber (see the new Figure 1), which affects the distribution of levoglucosan among the gas, the particles, and the chamber walls up to time t_0 ”

- **Comment 16.c.** Above figure 3 it says that the initial particle size varied between experiments due to the different times of injection and evaporation during this time. How come that before time zero (time of injection) particle diameter D_{ve} goes up and $f_{o/s}$ goes up? If particles were evaporating, I would expect the diameter to decrease and also the mass of organics? (if levoglucosan is evaporating)?

See the answer to the previous comment

- **Comment 16.d.** Similarly, why in figure 4 is $f_{o/s}$ going up in the period of injection when RH is constant at around 85%? This would suggest that levoglucosan condenses during the injection period?

Response : In Figure 4, $f_{o/s}$ is slightly going up during the injection step (prior time t_0) in the same manner as in the other experiments (note that the scale is zoomed in Fig. 4 compared to Fig. 3). As mentioned above, this observation can be tentatively explained by the fact that during the injection step, the particle number concentrations increased, thus providing more surface for levoglucosan to condense. The difference between experiments is due to the fact that the injection flow had to be manually adjusted according to the pressure difference between the AAC and the chamber. It is likely that the flow impacted the $f_{o/s}$, and this is why we focus the study only on the observation phase, after time t_0 .

- **Comment 16.e.** In figure 3 panel 3h the fraction of organics in the particles is changing from ~ 2 to ~ 0.4 (a factor of five) -- what change in volume (D_{ve}) does that correspond to based on simple estimations using mass and density of AMS and LG? Does it correspond to the measured change in D_{ve} and thus particle volume?

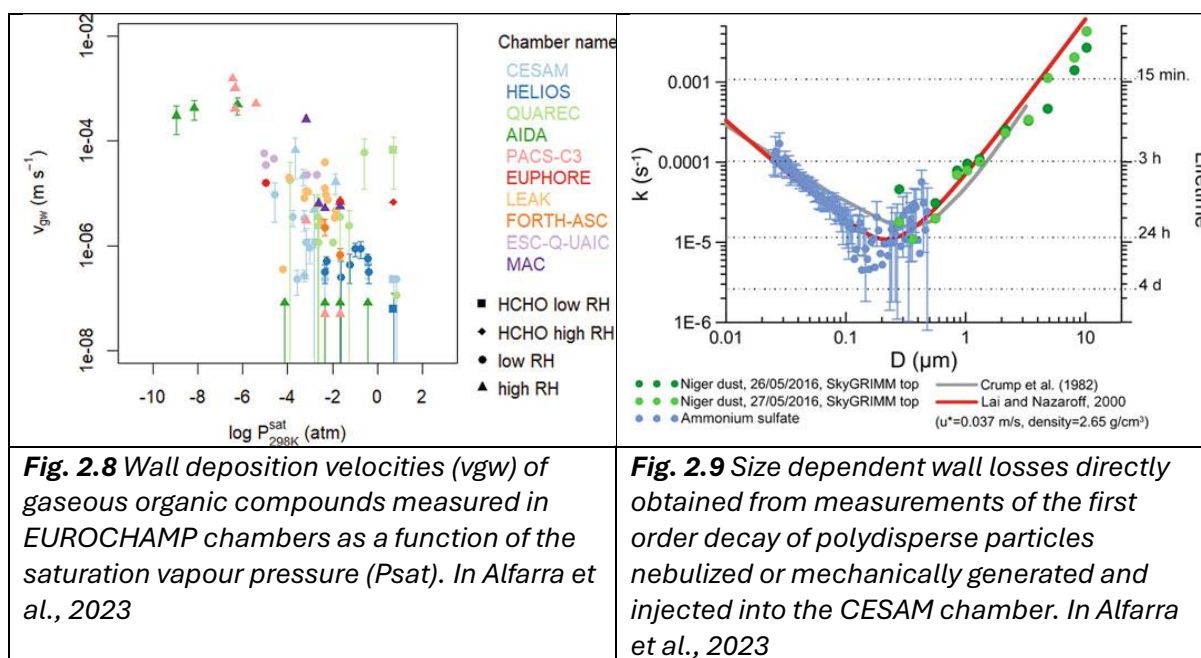
Response : We calculated D_{ve} for panel 3h as $f_{o/s}$ decreased from 2.2 to 0.4, assuming that ammonium sulfate is non-volatile ($\rho_{AS} = 1.77 \text{ g.cm}^{-3}$) and levoglucosan evaporates ($\rho_{levo} = 1.69 \text{ g.cm}^{-3}$). With an initial observed D_{ve} of 80 nm, the calculated final D_{ve} was 63 nm, compared to the SMPS-measured D_{ve} of 68 nm. The difference is 7.3%, which is consistent with the measurement uncertainty of the D_{ve} (7%).

- **Comment 16.f.** What is assumed about the wall loss rate of levoglucosan in the gas phase relative to wall loss rate of the particles? Is it used to explain some of the observed trends?

Response : Using the data compiled by Alfarra et al., 2023, Fig. 2.8 (below) shows the wall deposition velocities of gaseous organics. For levoglucosan, taking $\log(P_{sat}) = -5.8$ (Booth et al., 2011), one obtains a wall deposition rate of the order of 10^{-4} m.s^{-1} in CESAM. Using the S/V of CESAM (4.3 m^{-1} Wang et al., 2011), this leads to a 1st order rate constant $k_{gas levo} = v_{gas levo} * (S/V)$ of the order of 4.10^{-4} s^{-1} for gaseous levoglucosan (corresponding to a lifetime of $1/k_{gas levo} = 42 \text{ min}$).

On the other hand, in the same reference (Alfarra et al., 2023), Fig. 2.9 shows that the 1st order decay of particles of 100-200 nm is of the order of 5.10^{-5} s^{-1} (corresponding to a lifetime of 5h30).

Therefore, based on this study, the wall loss deposition of gaseous levoglucosan is an order of magnitude faster than that of the particles (at time t_0).



General

Comment 17.

I find that the title should revised - co-condensation is not studied in the presence of a surfactant for example.

Response : It is true that, in the presence of a surfactant, only co-evaporation was studied. Nevertheless, we would like to keep co-condensation in the title. In the new title proposed (mentioned earlier) we do not mention surfactant:

“Co-condensation and co-evaporation of levoglucosan onto and from deliquesced ammonium sulfate particles – Part 1: Chamber experiments”

Comment 18.

Please give the values used in various calculations and discussions (e.g. values for ERH and DRF points etc. with literature references.

Response : We presume the reviewer is referring to ERH and DRH. The reference for bare ammonium sulfate (Onasch et al., 1999) has been added to the revised manuscript.

Comment 19.

The word co-condensation was originally for condensation of soluble gases alongside water onto growing cloud droplet at supersaturated conditions. The current study is for RH<100%. I suggest to explicitly state that the word is used here before activation into a cloud droplet.

Response : See the responses to Comments 4 and 5 of Reviewer #1.

Comment 20.

Line 48: “Although this process is based on thermodynamics and has been simulated in models” – please add references for this statement.

Response : *We added the following references after “models”: (Topping et al., 2013; Wang et al., 2026).*

Comment 21.

As already pointed out by reviewer 1, the gas-particle partitioning of organics to/from aqueous solution droplet containing salt has been previously investigated and it would be natural to use the results from that work as perspective (Yli-Juuti et al. EST 2013, DOI: 10.1021/es401233c).

Response : *We have now mentioned this study in the introduction. See response to Comment 5 of reviewer #1.*

Comment 22.

Page 2 line 55. The cloud experiments referred to was conducted at low temperature (243 or 263 K) (Lopez et al. 2025) – how does it relate to the conditions in the current experiments? What is the importance of the diagonal basis-set for the current work?

Response : *We agree with the reviewer that this basis set paper is not relevant, we have deleted in the revised version.*

Comment 23.

Line 60: Levoglucosan is a solid at room temperature – the term miscible applies to mixtures of liquids? Should it perhaps say "soluble" instead?

Response : *Following a comment of reviewer #1, we now discriminate between solubility of the solid and miscibility of the subcooled liquid. The revised sentence reads:*

“Levoglucosan (1,6-anhydro- β -D-glucopyranose, $C_6H_{10}O_5$) was chosen because it is a semi-volatile compound with a saturation concentration of $\log(C^) = 1.1 \mu\text{g}/\text{m}^3$ at 298 K (Zhang et al., 2024; Booth et al., 2011). It has an aqueous solubility of 61–63 wt% (Lienhard et al., 2012; Osmanbegovic et al., 2023) and its supercooled liquid is miscible with water (Parsons et al., 2004).”*

Comment 24.

Later it says: semi-volatile and water-soluble – please provide values and references for water solubility and vapor pressure.

Response : *See our previous response (comment 23 of this reviewer)*

Comment 25.

Pag2 line 63 “high emission factors” – please be quantitative and provide references

Response : *We have changed the text, it now reads:*

“Furthermore, levoglucosan is a widely used molecular marker of biomass burning in the atmosphere, as it is almost exclusively emitted by the pyrolysis of cellulose and hemicellulose, it represents 52 to 290 mg.g⁻¹ of organic carbon (OC) in biomass combustion, depending on the type of wood burnt and the combustion conditions (Bhattacharai et al., 2019). It has been measured in particles at various sites all over the world, and overall, 90% of levoglucosan mass concentrations were found in particles with aerodynamic diameters of less than 2 µm (Bhattacharai et al., 2019).”

Comment 26.

Figure 4: 60 minutes is highlighted in the text – it is difficult to see in the figure what happens at 60 minutes – the second fit is started later. It says that RH stabilizes after 60 minutes but from panel a RH seems to decrease steadily after 60 min?

Response : *From panel a, RH increases from 88 to 100% within 45 minutes, when the fo/s increases, then RH is stabilized for the next 40 minutes at $100.7 \pm 0.4\%$, when fo/s is constant, and finally RH decreases with time, and fo/s also decreases.*

We have revised the text as: “Note however that this increase continues even when the RH stabilizes at $100.7 \pm 0.4\%$ (between 45 and 85 minutes) and starts to decrease (after 85 min). This disagrees with the change in fo/s evolution after 45 min in Fig. 4. It is probably due to the uncertainties generated by the dry Dve corrections, which however do not affect fo/s values (see section 2.5.2).”

Comment 27.

Regarding the SDS experiments: I miss figures with data (evaporation rates, relative humidities etc.) for the SDS experiments, and the table values do not include uncertainty estimates – I suggest these elements are included and discussed in a revised manuscript.

Response : *A figure has been added in SI-5-5 for Exp 4b. The uncertainties are indeed valuable to correctly draw conclusions from presented data, and are often missing from modeling works, as they are not always straightforward to estimate. Estimations of uncertainties have been added to Table 2, along with the explanation on the method used, in section SI 4-5 of the supplementary information. Note that in Table 2 the values related to AMS measurements of SDS concentrations have been changed, as we identified an error in our previous estimations, that have been corrected in the supplementary information SI 4-1.*

- *The manuscript has been changed in lines 518-525, it now reads (changes in blue):*

“The results presented in Table 2 for filters and AMS are approximate estimations, due to the uncertainties on AMS and filter derived SDS concentrations (see SI 4-1 and SI 4-2), as highlighted by the high uncertainties in Table 2, and because the exact flow rate of filter sampling was not precisely controlled (see SI 4-2). The particularly high uncertainties of SDS

bulk concentrations derived from the filter quantification come from the lognormal distribution assumed for the bulk concentration (see SI 4-5) and the large uncertainties of SDS total concentrations derived from the filters. The methodology for uncertainties estimations are presented in section SI 4-5 of the supplementary information.”

- Table 2 now reads (changes highlighted in cyan):

Table 2. SDS experiments (Exp 4a and 4b): estimated surface-to-volume (A/V) ratios, SDS concentrations in the particle, surface coverages by SDS (detailed in SI 4). SDS concentration and surface coverages were calculated from both AMS and filter data. A/V ratios and results calculated from AMS data were averaged for the first 10 min of experiments after each indicated time.

Exp no.		4a			4b	
Time (min)		19	93	153	0	49
$k_{net} (min^{-1})$		$3.8 \cdot 10^{-3}$	$3.8 \cdot 10^{-3}$	$8.8 \cdot 10^{-3}$	$5.5 \cdot 10^{-3}$	$5.5 \cdot 10^{-3}$
A/V ratio ($\times 10^6 m^{-1}$)		23.1 ± 3.8	23.3 ± 3.9	26.9 ± 4.5	23.8 ± 3.9	23.0 ± 3.8
Wet D_{ve} (nm)		260 ± 42	257 ± 41	223 ± 36	252 ± 40	261 ± 42
SDS total concentration ($\times 10 \text{ mmol L}^{-1}$)	AMS	59 ± 20	83 ± 20	132 ± 37	83 ± 19	92 ± 30
	Filter	8.5 ± 5.3	8.8 ± 5.5	13 ± 8	77 ± 48	69 ± 43
SDS bulk concentration ($\times 10 \text{ mmol L}^{-1}$)	AMS	33 ± 13	56 ± 12	101 ± 27	56 ± 10	66 ± 20
	Filter	0.15 ± 0.19	0.16 ± 0.20	0.25 ± 0.37	50 ± 40	43 ± 37
Levoglucosan concentration (mol/L)		1.1	1.1	1.2	2	1.3
AS concentration (mol/L)		1.4	1.8	3.1	2.5	2.1
SDS surface coverage (%)	AMS	99 ± 0	99 ± 0	100 ± 0	99 ± 0	100 ± 0
	Filter	31 ± 15	32 ± 14	43 ± 10	99 ± 0	99 ± 0
Surface tension ($mN m^{-1}$)	AMS	35 ± 0	35 ± 0	35 ± 0	35 ± 0	35 ± 0
	Filter	61 ± 1	61 ± 1	56 ± 2	35 ± 0	35 ± 0

- The error on the SDS concentrations has been corrected in SI 4-1, that now reads (changes in cyan):

“S4-1 AMS-derived SDS concentration in the particle

The SDS concentration is derived from AMS measurement using Eq. S 4-1, where $c_{SDS,j}^{air}$ is the concentration of SDS at time j (in $\mu g m^{-3}$), $c_{org,j}^{air}$ is the total organic mass concentration measured by the AMS at time j (in $\mu g m^{-3}$), $c_{levo,j}^{air}$ is the levoglucosan mass concentration measured by the AMS at time j (in $\mu g m^{-3}$), M_{SDS} is the molar mass of SDS with $M_{SDS} = 288.38 \text{ g mol}^{-1}$, and $M_{Sulfate}$ is the molar mass of a sulfate ion with $M_{Sulfate} = 96.06 \text{ g mol}^{-1}$. Note that because of the presence of impurities (Wu et al. 2022), the obtained SDS concentration is a slight overestimation of the real SDS concentration.

$$c_{SDS,j}^{air} = \frac{c_{org,j}^{air} - c_{levo,j}^{air}}{M_{SDS} - M_{Sulfate}} M_{SDS} \cdot \quad \text{Eq. S 4-1}$$

The obtained SDS concentration in the air is then converted into a concentration in the particle $c_{SDS,j}^{AMS}$ at time j with Eq. S 4-2, where $n_{j,part}$ is the particle number concentration measured by the SMPS at time j , and $V_{part,j} = \frac{\pi}{6} D_{ve,j}^3$ is the volume of the wet particle obtained from the volume equivalent diameter $D_{ve,j}$ measured by the SMPS at time j . Note that the volume of the wet particle is underestimated due to the lower RH% encountered in the SMPS compared to the CESAM chamber, especially at high RH.

$$c_{SDS,j}^{AMS} = \frac{c_{SDS,j}^{air}}{n_{j,part} V_{part,j}} = \frac{c_{SDS,j}^{air}}{n_{j,part} \frac{\pi}{6} D_{ve,j}^3} \cdot M_{SDS} \quad \text{Eq. S 4-2''}$$

- The calculation of uncertainties is now detailed in the supplementary information, S4-5, it reads:

“SI 4-5 Uncertainties calculations

The uncertainties were calculated using a Monte-Carlo approach. The uncertainties on the input parameters for each calculation are provided in Table SI4-1. For the calculations of SDS total concentration, SDS bulk concentration, SDS surface coverage, and surface tension uncertainties, the equations S4-2 and S4-4, S4-6, S4-5, and S4-7, respectively, were used with input parameters calculated randomly, assuming a normal distribution of their values centered on the average values and with a standard deviation equal to the uncertainty associated with that input parameter. The uncertainties on input parameters are presented in Table SI4-1. The process was repeated 100.000 times providing 100.000 values for the calculated property (SDS total concentration, SDS bulk concentration, SDS surface coverage, or surface tension). The value of 100.000 repetitions was chosen to limit the differences between two separate runs and thus provided a stable standard deviation. The shape of the obtained distribution was checked graphically, to fit an appropriate statistical distribution in order to obtain the associated standard deviation. The appropriate statistical distributions for each parameter presented in Table 2 in the main text are presented in Table SI4-2. The standard deviation was then obtained using the fit method of the appropriate distribution with the `scipy.stats` package from Python. The uncertainty was then calculated as the product between the standard deviation and the average value provided in Table 2 in the main text.

Table SI4-1: Table of uncertainties in input parameters used to calculate uncertainties of the properties presented in Table 2 in the main text.

Input parameter	Associated uncertainty
D_{ve}	16%
n_{part}	10%
c_{SDS}^{air}	35%
$n_{SDS}^{colorimetry}$	$0.25 \cdot 10^{-9} \text{ mol (exp 4a)}$
	$0.62 \cdot 10^{-9} \text{ mol (exp 4b)}$
K_{ads}	$0 \text{ m}^3 \text{ mol}^{-1}$
Γ_{SDS}^{max}	$8.2 \cdot 10^{-7} \text{ mol m}^{-2}$

Table SI4- 2: Table of statistical distributions used to fit to output parameters calculated from the input parameters presented in Table SI4-1, assuming they were following a normal distribution.

Output Parameter		Appropriate statistical distribution
<i>A/V ratio</i>		<i>Lognormal</i>
<i>SDS total concentration</i>		<i>Lognormal</i>
<i>SDS bulk concentration</i>		<i>Lognormal</i>
<i>SDS surface coverage</i>	<i>From filter data</i>	<i>Lognormal</i>
	<i>From AMS data</i>	<i>Johnson SU</i>
<i>Surface tension</i>	<i>From filter data</i>	<i>Johnson SU</i>
	<i>From AMS data</i>	<i>Lognormal</i>

Comment 28.

Regarding the photochemical degradation rates of the particle phase of approximately 17 hours (line 592) – where does this number come from? It would depend on the OH concentration in the droplets? How is the atmospheric lifetime calculated?

Response : We agree with the reviewer that the paragraph was not precise enough. We have changed it as follows (line 602): “Overall, the results showed that the net evaporation flux of levoglucosan for particle-phase levoglucosan corresponds to atmospheric lifetimes of 1 to 12 hours. This is faster than the photochemical degradation rates of levoglucosan studied in the particle phase which have shown that levoglucosan has an atmospheric lifetime of 17h to 3.9 days when biomass burning particles are exposed to $1 - 1.5 \times 10^6$ molecules cm^{-3} of gaseous OH radical (Hennigan et al., 2010; Lai et al., 2014). This result highlights the importance of considering the phase partitioning of semi-volatile compounds when assessing their atmospheric multiphase reactivity.”

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