



1 **Co-condensation and co-evaporation of levoglucosan onto and from deliquesced**  
2 **ammonium sulfate particles – influence of relative humidity, particle mass and**  
3 **size, and presence of a surfactant**

4

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18 **Abstract**

19

Co-condensation is the process by which condensable vapors condense alongside water vapor onto  
20 growing aerosols, and it can significantly alter the properties of clouds. Semi-volatile species  
21 partition dynamically between the gas and the condensed phases, and their co-condensation together  
22 with water vapor may increase the condensed mass and amplify water uptake. Although this process  
23 is based on thermodynamics and has been simulated in models, it has been scarcely investigated  
24 experimentally. In this study, the ability of levoglucosan to co-condense and co-evaporate from  
25 inorganic particles together with water was investigated by monitoring, under ambient-like  
26 conditions, its gas-particle partitioning on ammonium sulfate (AS) monodispersed particles in the  
27 Experimental Multiphase Atmospheric Simulation Chamber (CESAM). The net evaporation flux of  
28 levoglucosan depending on particle size and the presence of a surfactant was explored for relative  
29 humidities (RH) from 100% to dry conditions as particle concentration in the chamber was reduced.  
30 Due to the high deliquescence point of AS, wet experiments were initialized at RH above 80%. Co-  
31 condensation of levoglucosan was observed when RH increased up to 100%, while co-evaporation  
32 of levoglucosan occurred when RH decreased. It was shown that gas-particle partitioning of  
33 levoglucosan was sensitive to all the investigated parameters, but the main drivers were the  
34 levoglucosan-to-sulfate mass ratios and RH. The effect of the levoglucosan-to-sulfate mass ratio was  
35 interpreted as salting-out, and the significant influence of RH provides experimental proof of co-  
36 evaporation or co-condensation of levoglucosan from or onto AS particles.

37



## 38 1. Introduction

39

40 Co-condensation is the process by which condensable vapors condense alongside water vapor onto  
41 growing aerosols (Kulmala et al., 1993; Topping et al., 2013). This can significantly alter the  
42 properties of clouds (Kulmala et al., 1997). Indeed, the co-condensation of gas-phase semi-volatile  
43 organic substances together with water vapor may increase the condensed mass and amplify water  
44 uptake. Semi-volatile species partition dynamically between the gas and the condensed phases (Ijaz  
45 et al., 2025; Thompson et al., 2017; Lopez et al., 2025; Donahue et al., 2006). As relative humidity  
46 (RH) increases, water condenses on particles, increasing the condensed volume and enhancing the  
47 condensation of semi-volatile species. Conversely, when RH decreases, water evaporates from the  
48 particles, along with semi-volatile species. Although this process is based on thermodynamics and  
49 has been simulated in models, few studies have investigated it experimentally. Several field studies  
50 coupled to models have shown that this effect is potentially large for secondary organic aerosol (SOA)  
51 containing a large fraction of semi-volatile species (Wang et al., 2025; Heikkinen et al., 2024; Rastak  
52 et al., 2017; Topping et al., 2013; Topping and McFiggans, 2012; Topping et al., 2011). In the same  
53 way, a new “diagonal” volatility basis set has been developed to assess the condensation of organic  
54 vapors onto particles and was tested in several CLOUD chamber experiments on  $\alpha$ -pinene SOA  
55 (Lopez et al., 2025). However, this process has never been experimentally investigated on a single  
56 semi-volatile compound under various controlled conditions.

57 In the present study, we have investigated experimentally the ability of a single semi-volatile organic  
58 compound to co-condense and co-evaporate from inorganic particles under various conditions.  
59 Levoglucosan (1,6-anhydro- $\beta$ -D-glucopyranose,  $C_6H_{10}O_5$ ) was chosen because it is a semi-volatile  
60 compound (Zhang et al., 2024) that is miscible with water. The molecule bears 3 hydroxyl groups  
61 that can provide strong H-bonds with water molecules and ions (Qin et al., 2021; Pye et al., 2018).  
62 Furthermore, levoglucosan is a widely used molecular marker of biomass burning in the atmosphere,  
63 it has high emission factors and is almost exclusively emitted by the pyrolysis of cellulose and  
64 hemicellulose. It has been measured in particles at various sites all over the world, and overall, 90%  
65 of levoglucosan mass concentrations were found in particles with aerodynamic diameters of less than  
66 2  $\mu\text{m}$  (Bhattacharai et al., 2019). However, levoglucosan is semi-volatile and water-soluble.  
67 Consequently, its mass fraction in particles is influenced by long-range transport, as well as by  
68 particle-phase mixing, evaporation, co-condensation, and multiphase chemical (and biological)  
69 processes (Bhattacharai et al., 2019; Yazdani et al., 2023).

70 To investigate the ability of a semi-volatile water-soluble organic compound to co-condense and co-  
71 evaporate from inorganic particles, the gas-particle partitioning of levoglucosan was monitored under  
72 ambient-like concentrations on ammonium sulfate (AS) monodispersed particles in the CESAM  
73 simulation chamber (French acronym for Experimental Multiphase Atmospheric Simulation  
74 Chamber). Various conditions were explored, including relative humidity (from dry to 100% RH),  
75 dilution, particle mass and size, and the presence of a surfactant on the particles. The net evaporation  
76 flux of levoglucosan was quantified under each condition.

77

## 78 2. Methods

79 To investigate the partitioning behavior of levoglucosan onto AS particles under dry and high  
80 RH conditions, fifteen experiments were performed under controlled conditions, as shown in Table



81 1. Except for those performed under dry conditions, all the experiments were started at RH above the  
 82 deliquescence point of AS particles. This was done to focus the study on the effects of levoglucosan  
 83 (partitioning into an aqueous AS particle) on particle growth or shrinking as a function of RH.

84 Two control experiments (1a, 1b) were performed under high RH without levoglucosan, using  
 85 AS particles of two different selected dry sizes (electrical mobility diameters of ~100 and ~200 nm,  
 86 respectively). Note that OA contributed to 10 and 8% of the total particle mass for Exp 1a and 1b,  
 87 respectively, in good agreement with (Wu et al., 2022).

88 The partitioning of levoglucosan onto AS particles was investigated under constant RH  
 89 conditions: three experiments under dry conditions (exp. 2a, 2b, 2c with selected electrical mobility  
 90 diameters of ~200, ~200 and ~100 nm, respectively) and three under constant high RH conditions  
 91 (exp. 3a, 3b, 3c with selected dry electrical mobility diameters of ~100, ~100 and ~200 nm).

92 The impact of varying RH on levoglucosan partitioning was examined in three experiments with  
 93 a selected dry electrical mobility diameter of ~200 nm (exp. 3d, 3e, 3f), and in two experiments with  
 94 a selected dry electrical mobility diameter of ~100 nm (exp. 3g, 3h).

95 Finally, two experiments (4a, 4b) were conducted to investigate the effect of a surfactant on  
 96 levoglucosan partitioning. These two experiments were conducted under the same conditions as  
 97 experiments 3f and 3c, respectively, but sodium dodecyl sulfate (SDS) was added to the solution to  
 98 generate particles. SDS was chosen as one of the most well-characterized water-soluble surfactants.  
 99 It is also slightly soluble in AS aqueous solutions, which were nebulized under the same conditions  
 100 as in all the other experiments (Faust and Abbatt, 2019).

101 **Table 1.** Summary of the experimental conditions. NA = non available;  $k_{net}$  is the net evaporation  
 102 rate of organic matter from AS particles. All numbers, except for T and  $k_{net}$  are given at time 0, when  
 103 the introduction of particles stopped. T are the temperature average values, and the  $k_{net}$  values are  
 104 determined from the exponential fitting of the time profiles of organic-to-sulfate mass ratio  $f_{o/s}$  (see  
 105 Sect. 2.6).

Exp no.	Aerosol type	T (°C) ±0.1	RH (%) ±0.8	[number] (10 <sup>4</sup> #/cm <sup>3</sup> ) ±5%	D <sub>ve</sub> dry* (nm) ± 7 %	[mass]* (µg.m <sup>-3</sup> ) ± 26%	OA/sulfate ( $f_{o/s}$ )	$k_{net}$ (× 10 <sup>-5</sup> s <sup>-1</sup> )	Exp. date yy/mm/dd
1a	AS	24.0	89.4	2.33	90.1	15.7	0.14± 0.01	NA	24/03/18
1b	AS	24.1	94.1	1.35	146	38.6	0.091± 0.001	NA	24/03/19
2a	AS/Levo	21.9	0	1.01	164	32.6	1.79± 0.01	1.66 ± 0.01	24/03/01
2b	AS/Levo	21.7	0	1.09	174	38.8	2.10± 0.01	1.64 ± 0.02	24/03/04
2c	AS/Levo	23.0	0	4.37	98.7	23.8	1.66± 0.01	5.41 ± 0.05	24/03/05
3a	AS/Levo	22.4	88.7	1.89	NA	NA	2.29± 0.02	6.00 ± 0.09	24/03/06
3b	AS/Levo	22.4	88.1	3.55	93.7	18.8	2.13± 0.02	4.36 ± 0.06	24/03/07
3c	AS/Levo	22.4	85.5	0.969	174	32.3	2.33± 0.01	3.36 ± 0.05	24/03/08
3d	AS/Levo	23.1	97.8	0.715	166	29.7	2.08± 0.01	-1.15 ± 0.10	24/03/11
		23.2	99.8	0.446	169	19.4	2.13± 0.02	0.91± 0.09	
		23.4	96.0	0.331	169	14.6	1.99± 0.02	4.44 ± 0.14	
3e	AS/Levo	23.8	86.0	1.02	175	49.8	2.38± 0.01	3.26 ± 0.06	24/03/12
		23.7	76.0	0.664	160	24.7	1.76± 0.03	11.5± 0.3	
3f	AS/Levo	24.4	85.1	0.918	177	45.4	2.39± 0.01	4.21 ± 0.07	24/03/13
		24.5	90.0	0.605	174	28.6	1.88± 0.02	11.6 ± 0.2	
		24.8	65.1	0.405	159	14.8	0.82± 0.02	29.3 ± 0.8	
3g	AS/Levo	23.8	85.7	1.77	93.9	13.7	1.54± 0.02	15.9 ± 0.3	24/03/14



3h	AS/Levo	24.4	89.2	2.92	79.8	13.3	2.25± 0.04	14.2 ± 0.3	24/03/15
		24.5	90.1	1.73	78.0	7.2	1.11± 0.03	29.3 ± 1.0	
4a	AS/Lev/SDS	24.5	88.5	NA	NA	NA	3.45± 0.02	6.34 ± 0.18	24/03/20
		24.7	87.2	0.742	186	37.6	3.30± 0.03	14.7 ± 0.6	
4b	AS/Lev/SDS	25.6	84.8	0.879	180	41.6	3.28± 0.02	9.14 ± 0.33	24/03/21

\* Corrected data (see Sect. 2.5.2)

106

## 107 2.1 CESAM chamber and experimental design

108 The CESAM chamber has been used extensively in recent years for the study of atmospheric  
109 multiphase processes and is described in detail in Wang et al (Wang et al., 2011). The chamber is  
110 made of stainless steel, with a volume of 4.2 m<sup>3</sup> and a surface-to-volume ratio of 4.3 m<sup>-1</sup>. Purified  
111 humidified (or dry) air inlet flow is employed to offset sampling flows and maintain automatically  
112 the requested pressure. A mixing time of approximately one minute within the chamber is ensured  
113 by a 50 cm diameter stainless steel fan. The chamber is equipped with a wide range of analytical  
114 instruments to measure physical parameters such as pressure (Baratron MKS), temperature (Type T  
115 Thermocouple - 321 Stainless Steel Sheath (400°C), TC-Direct), RH (Chilled Mirror, Michell  
116 Optidew model 501). Special care was taken with RH control and measurements, particularly for  
117 experiments performed at high RH. The saturation water vapor pressure was derived from the dew  
118 point measured by the chilled mirror using the Buck equation (Buck, 1981), and the water vapor  
119 pressure was derived from the same equation, but using the average temperature provided by three  
120 thermocouples T sensors suspended in the air inside CESAM. RH was calculated as the ratio between  
121 these two vapor pressures.

122 The chamber also provides additional connections for other dedicated instruments that were  
123 required (described in Sect. 2.5). All these instruments were operated online, leading to a constant  
124 air flow out of the chamber, which was continuously compensated by an inflow of humid N<sub>2</sub>,  
125 inducing continuous dilution of the mixture inside the chamber. The humidification of N<sub>2</sub> was done  
126 using the HUMANS set up (described in Sect. 2.2), which allowed a precise control of RH throughout  
127 each experiment. To minimize contamination from external air, the chamber was operated at a  
128 pressure of 5 mbar above the prevailing ambient one. In addition, the chamber was daily vacuumed  
129 after each experiment and remained under vacuum (i.e. 1×10<sup>-3</sup> mbar) overnight with its walls heated  
130 at 55 °C, and the inner walls of the chamber were manually cleaned prior to each new series of  
131 experiments.

132

## 133 2.2 HUMidificAtioN System (HUMANS) to regulate the relative humidity at high values in the 134 chamber

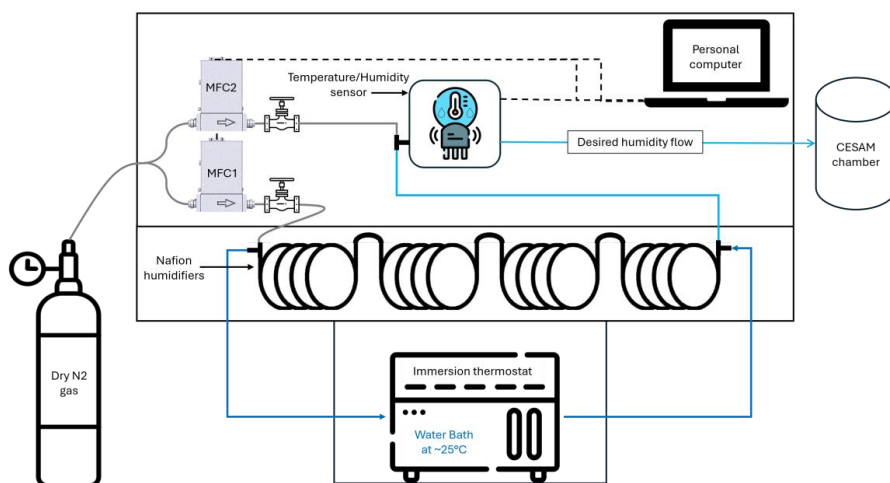
135 At the start of each experiment, a known quantity of water (92 g on average) was introduced into  
136 CESAM under vacuum, and then dry nitrogen was added to reach the desired pressure (5 mbar above  
137 ambient). The amount of water introduced was calculated to achieve a relative humidity (RH) of  
138 ~90% when the desired pressure was reached. Because RH was difficult to maintain at high RH, the  
139 HUMidificAtioN System (HUMANS) was then connected to CESAM to regulate RH precisely by  
140 adjusting the RH of the nitrogen input, which had an initial RH of less than 10%. The HUMANS  
141 system was employed to precisely maintain or decrease the desired RH within CESAM throughout



142 the experimental period. HUMANS can generate a total flow of up to  $40 \text{ L min}^{-1}$ , but was operated  
143 at a constant total flow rate of  $10 \text{ L min}^{-1}$  in this study. It utilizes two mass flow controllers (MFCs;  
144 Alicat MC-20SLPM), referred to as MFC1 and MFC2 (see Fig. 1). The dry  $\text{N}_2$  flow through MFC1  
145 is routed through four Nafion humidifiers (Perma Pure MH-110) connected in series (as shown in  
146 Fig. 1). These tubes are surrounded by Milli-Q water, which is circulated via a thermostatically  
147 controlled water bath (Lauda E306) maintained at  $\sim 25^\circ\text{C}$ . Water vapor diffuses through the Nafion  
148 membranes, producing a humidified  $\text{N}_2$  stream (sky blue colour line in Fig. 1). Simultaneously, a  
149 separate dry  $\text{N}_2$  stream from MFC2 bypasses the Nafion tubes. The humidified and dry streams are  
150 mixed downstream, enabling precise RH control through dynamic adjustment of the individual MFC  
151 flow rates.

152 The final RH of the flow was typically set to match the target RH of the chamber. To do so, the  
153 CESAM compensating flow (which is  $\geq 10 \text{ L min}^{-1}$ ) was divided into two: i) a constant  $10 \text{ L min}^{-1}$   
154 flow passed through HUMANS, and ii) the rest of the passed through a heated water bubbler. For  
155 instance, to maintain a chamber RH of 65%,  $6.5 \text{ L min}^{-1}$  was directed through MFC1 and  $3.5 \text{ L min}^{-1}$   
156 through MFC2. For near-saturation conditions ( $\sim 90\%$  RH), the full  $10 \text{ L min}^{-1}$  flew through MFC1,  
157 while for dry conditions ( $<30\%$ ), MFC1 was shut off and only dry  $\text{N}_2$  from MFC2 was supplied to  
158 the chamber. The final RH of the mixed flow was monitored in real time using a calibrated  
159 temperature and humidity sensor (Sensirion SHT75). Sensor feedback was integrated into a Python-  
160 based control interface, which dynamically adjusted the flow rates to maintain the target RH with  
161 high accuracy and stability. To prevent contamination and ensure reproducibility, the Milli-Q water  
162 used in the humidification bath was replaced daily prior each experimental run.

163



164 **Figure 1.** Schematic of HUMANS (HUMidificAtioN System) for regulating relative humidity (RH) within the CESAM  
165 chamber. MFC1 and MFC2 represent the mass flow controllers, where MFC1 directs dry nitrogen through the Nafion  
166 humidifiers, while MFC2 controls the dry nitrogen flow for mixing. The dark blue line indicates the circulating water  
167 from the thermostatically controlled water bath. The grey and light blue lines represent the dry nitrogen stream (RH  
168  $<10\%$ ) and the humidified nitrogen stream, respectively. The dashed black line denotes the system components  
169 monitoring and control via the computer interface.



## 170 2.3 Chemical reagents

171 Levoglucosan (purity: 99%), ammonium sulfate (99.9999% Suprapur) and sodium dodecyl  
172 sulfate (SDS; purity:  $\geq 99.0\%$ ) were purchased from Sigma-Aldrich. LC-MS grade water (Thermo  
173 Fisher Scientific Inc.) has been used for preparing the solutions to be nebulized in the chamber and  
174 for maintaining the desired RH in the tank. The particles were generated by nebulization of 50 mL  
175 solutions containing  $15 \text{ g L}^{-1}$  of AS and  $15 \text{ g L}^{-1}$  of levoglucosan, freshly prepared on the day of each  
176 experiment. For SDS experiments, the solution composition was  $15 \text{ g L}^{-1}$  of AS,  $15 \text{ g L}^{-1}$  of  
177 levoglucosan and  $4.6 \text{ g L}^{-1}$  of SDS. The latter concentration was chosen after pre-experiments of  
178 nebulization using AS + SDS, which confirmed the feasibility of nebulizing SDS + AS solutions to  
179 generate monodispersed particles of the desired diameter, and the necessity of using a large impactor  
180 between the atomizer and the Nafion dryer (see Supplementary Information SI-1).

## 181 2.4 Introduction of monodisperse particles

182 An atomizer (TSI Model 3076) was used to produce polydisperse aerosols. The particles then  
183 passed through a dryer and an Aerodynamic Aerosol Classifier (AAC, Cambustion). In the AAC, the  
184 particle aerodynamic diameters were selected at 170 or 300 nm, corresponding to particle mobility  
185 diameters of approximately 100 or 200 nm, respectively. The conversion from aerodynamic to  
186 mobility diameter was further confirmed using the shape factor and the mixed density, using the  
187 AMS data (DeCarlo et al., 2004). Before introducing the particles into CESAM, the size-selection  
188 performance of the AAC was verified by measuring the monodisperse size distribution using a TSI  
189 SMPS placed at the AAC outlet. Once this monodisperse size distribution was Gaussian, aerosols  
190 from the AAC outlet were introduced into the chamber. The aerosol flow rate through the AAC and  
191 into the chamber was maintained within  $1\text{-}2 \text{ L min}^{-1}$ , with aerosol concentrations of approximately  
192  $4 \times 10^5 \text{ particles cm}^{-3}$  for 100 nm and  $3 \times 10^5 \text{ particles cm}^{-3}$  for 200 nm, respectively. One to two hours  
193 of injection were required to achieve target number concentrations of at least  $10\,000 \text{ particles cm}^{-3}$   
194 of 100 nm and  $8000 \text{ particles cm}^{-3}$  of 200 nm particles in CESAM. These values were targeted to  
195 ensure sufficient particles to be detected after several hours of experiment while avoiding coagulation.  
196 At the same time, a flow of humid nitrogen was introduced into the chamber to adjust the relative  
197 humidity, which was controlled by the HUMANS system.

## 198 2.5 Particle characterization in CESAM

### 199 2.5.1 Characterization of chemical composition of particles

200 The chemical composition of particles was measured with the Aerodyne high-resolution time-  
201 of-flight aerosol mass spectrometer (AMS) (Drewnick et al., 2005; DeCarlo et al., 2006). Standard  
202 calibrations were performed using 300 nm size-selected dried ammonium nitrate and ammonium  
203 sulfate particles at the beginning and the end of the campaign. The data were analyzed using the ToF-  
204 AMS HR Analysis Toolkit v1.25, which includes Pika version 1.25F and Squirrel version 1.65F. To  
205 verify that the organic signal detected by the AMS corresponds to levoglucosan, we used the  
206 characteristic levoglucosan marker  $m/z$  60.02, as described in previous studies (Lee et al.,  
207 2010; Alfarra et al., 2007), corresponding to the fragment  $\text{C}_2\text{H}_4\text{O}_2$  in HR MS. The contribution of the  
208 signal at  $m/z$  60.02 to the total organic aerosol signal remained constant throughout each experiment.  
209 In experiments 4a and 4b, involving the addition of SDS, we used  $m/z$  168.19 to identify the presence  
210 of SDS. This  $m/z$  corresponds to  $\text{C}_{12}\text{H}_{24}$  as reported by (Faust and Abbatt, 2019). It was chosen due  
211 to its high signal-to-noise ratio, characterized by a low background concentration and a clear signal



212 upon SDS addition, as we had validated by preliminary specific experiments (SI-1). The detailed  
213 quantification of SDS and levoglucosan during Exp 4a and 4b can be found in SI-2.

214 The AMS provided the mass concentration of sulfate ( $m_{sulfate}$ ) and the total mass of organic  
215 compounds ( $m_{org}$ ). The organic to sulfate mass ratio ( $f_{o/s} = m_{org}/m_{sulfate}$ ) was used to track the  
216 partitioning of levoglucosan during the experiments. Using this ratio also enabled to correct  
217 internally any changes in particle density and collection efficiency during the experiment (provided  
218 that the aerosol was internally mixed). Assuming that the organic matter represents levoglucosan  
219 with a density of  $\rho_{org} = 1.69 \text{ g cm}^{-3}$ , and taking the ammonium sulfate density ( $\rho_{AS} = 1.77 \text{ g cm}^{-3}$ ),  
220 the density of the mixed particles  $\rho(f_{o/s})$  is a function of  $f_{o/s}$  given by Eq. 1:

$$221 \quad \rho(f_{o/s}) = \frac{\frac{M_{AS}}{M_{sulfate}} + f_{o/s}}{\frac{M_{AS}}{\rho_{AS} M_{sulfate}} + \frac{f_{o/s}}{\rho_{org}}} \quad (1)$$

222 where  $M_{AS}$  is the molar mass of AS;  $M_{sulfate}$  is the molar mass of sulfate.

223 In addition, particles were collected on quartz fiber filters during each experiment. SDS was  
224 quantified using the methodology developed in (Grisillon et al., 2026). Briefly, the filter was  
225 extracted in 10 mL of ultrapure water, and the resulting solution was filtered using a pre-cleaned  
226 polyethylene frit, following the protocol described in (Grisillon et al. 2026). The determination of  
227 SDS quantities in the filtrate was performed by colorimetry with ortho toluidine blue, as described  
228 in (Grisillon et al. 2026). More details are provided in SI-2.

### 229 2.5.2 Size and mass of particles

230 A Scanning Mobility Particle Sizer (SMPS, TSI, 3081) equipped with X-ray neutralizer (TSI  
231 model 3088) and coupled with a Condensation Particle Counter (CPC, TSI, 3772), was connected  
232 directly to the CESAM chamber, to measure the size of the wet (deliquesced) particles without pre-  
233 drying. The sample flow rate and sheath flow rate were set to  $1 \text{ L min}^{-1}$  and  $10 \text{ L min}^{-1}$ , respectively,  
234 resulting in a size detection range of 7.64–289 nm for wet particles during the 100 nm dry injected  
235 size experiment days. During the 200 nm dry injected size experiment days, a sheath flow of  $5 \text{ L}$   
236  $\text{min}^{-1}$  and an aerosol flow of  $1 \text{ L min}^{-1}$  were used, yielding a scanned size range of 11.3–461.4 nm.

237 To monitor the size distribution of dry particles, a TROPOS-style Mobility Particle Size  
238 Spectrometer (MPSS) system equipped with a  $^{63}\text{Ni}$  neutralizer and coupled with a Nafion dryer was  
239 connected to CESAM. The MPSS consisted of two TROPOS-modified Hauke-type Differential  
240 Mobility Analyzers (DMA) (Wiedensohler et al., 2012; Winklmayr et al., 1991). A short DMA was  
241 used for detecting particles in the small size range (5.1 - 40 nm), with sheath air flow and sample air  
242 flow rates set at 15 and  $1.5 \text{ L min}^{-1}$ , respectively. A long DMA measured particles in the larger size  
243 range (20 - 800 nm), with corresponding sheath air flow and sample air flow rates of 5 and  $1 \text{ L min}^{-1}$ ,  
244 respectively. An ultrafine condensation particle counter (CPC, TSI 3750) was employed to count  
245 aerosols downstream of the short DMA, while a standard condensation particle counter (CPC, TSI  
246 3772) was used for particles from the long DMA. The particle number size distributions across the  
247 two size ranges were calculated and merged using the TROPOS inversion routines to produce the  
248 final particle number size distribution. All flow rates were continuously monitored and recorded with  
249 the same time resolution as the particle size distribution measurements. Further details on the MPSS  
250 can be found elsewhere (Birmili et al., 1999; Aalto et al., 2001; Wiedensohler et al., 2012). The SMPS  
251 and MPSS were intercalibrated with a CPC (TSI, 3750) in ambient air. The MPSS and the CPC gave



252 a good agreement on the number concentration, and this number concentration was 1.2 times higher  
 253 than that of the SMPS. The MPSS signal was taken as the reference in all experiments.

254 It was expected that after the Nafion dryer, the RH in the MPSS would be low enough for all  
 255 particles to dry completely before entering the instrument (which is 40% RH from the ACTRIS  
 256 recommendations). However, due to technical issues with the dryer, the RH sensor integrated into  
 257 the sheath flow of the MPSS indicated values ranging from below 15 to 50%, depending on the RH  
 258 in CESAM and, as a result, the measured particle size was dependent on the internal RH% (as shown  
 259 in Fig. 2 for Exp 1a). It was thus necessary to adopt a methodology of particle size correction to  
 260 obtain dry particle size and mass concentrations, even though it provided additional uncertainties, as  
 261 shown below. The following correction procedure was applied:

262 1/ based on the study from (Lei et al., 2018) who investigated the water uptake between 15%  
 263 and 95%RH of AS and levoglucosan mixed particles experimentally and theoretically, we have built  
 264 a two dimensional matrix of growth factors  $g(RH_{MPSS}, f_{o/s})$  by interpolating their experimental data,  
 265 where RH varied from 15% to 95% with a precision of 1% and  $f_{o/s}$  varied between 0.45 and 4.12  
 266 with a precision of 0.02. More details on the matrix of growth factors  $g(RH_{MPSS}, f_{o/s})$  can be found  
 267 in the SI-3.

268 2/ The growth factor  $g(RH_{MPSS}, f_{o/s})$  was then used to correct the particle size and total mass  
 269 concentrations only when RH in the MPSS was higher than 15%. Below 15% RH, no correction was  
 270 applied.

271 3/ Assuming that the growth factor did not depend on particle size, the dry particle size and  
 272 particle mass concentration were calculated according to eq. 2.

$$273 \quad \begin{cases} D_{ve} = \sqrt[3]{\frac{\sum_{i=1}^k n_i \cdot (d_i/f_g)^3}{\sum_{i=1}^k n_i}} = \frac{D_{ve\_raw}}{f_g} \\ m_{total} = \rho(f_{o/s}) \frac{\pi}{6} \sum_{i=1}^k n_i \cdot (d_i/f_g)^3 \end{cases} \quad (2)$$

$$274 \quad \text{with } \begin{cases} f_g = 1, \text{ if } RH_{MPSS} < 15\% \\ f_g = g(RH_{MPSS}, f_{o/s}), \text{ if } 15\% \leq RH_{MPSS} \leq 95\% \end{cases}$$

275 where  $D_{ve}$  is the volume equivalent diameter under dry conditions in  $nm$ ;  $D_{ve\_raw}$  is the  
 276 volume equivalent diameter given by the MPSS in  $nm$ ;  $k$  is the bin number of the size distribution  
 277 given by the MPSS;  $d_i$  is the average diameter of each bin in  $nm$ ;  $n_i$  is the number concentration of  
 278 each bin in  $cm^{-3}$ ;  $m_{total}$  is the total mass concentration of particles under dry conditions in  $\mu g m^{-3}$ ;  
 279  $\rho(f_{o/s})$  is the density of particles obtained from Eq. 1; and  $f_g$  is the correction factor. The 5%  
 280 uncertainty on  $D_{ve\_raw}$  (MPSS default) and the 2% uncertainty on  $f_g$  (Lei et al., 2018) result in 7%  
 281 uncertainty on  $D_{ve}$  and 26% on  $m_{total}$ . The RH% limits for  $f_g$  were chosen based on Fig. S4, which  
 282 shows that, to limit the bias between the uncorrected and corrected dry mass of particles to below  
 283 10%, the RH inside the MPSS must be  $\leq 15\%$ .

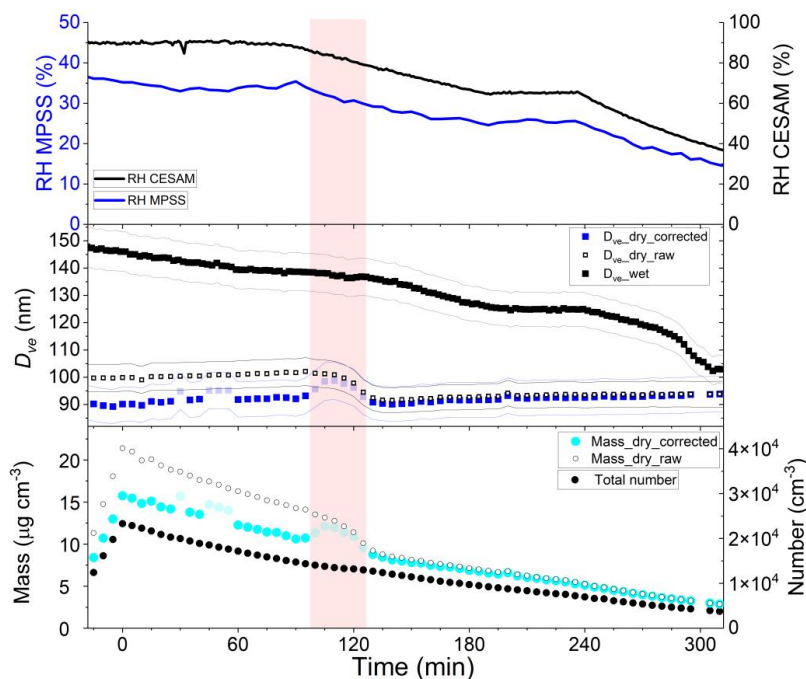
284 4/ The mass concentration of ammonium sulfate ( $m_{AS}$ ) and organic matter ( $m_{org}$ ) were  
 285 finally calculated based on  $m_{total}$  using the organic to sulfate ratio ( $f_{o/s}$ ) determined by the AMS.

286 Figure 2 shows the effect of this correction procedure for Exp 1a, performed with pure AS  
 287 particles. The RH in CESAM decreased from 85% to 38% in two steps (first decrease starting at



288 13:45 and the second one at 16:15). The RH in the MPSS clearly followed the same time-profile,  
289 decreasing from 35% to 16%. This shows that the efficiency of the Nafion dryer was not sufficient  
290 to dry the particles completely. This is confirmed by the comparison between the time profiles of  
291  $D_{ve\_wet}$  and  $D_{ve\_dry\_raw}$  (Fig. 2, middle panel). Although  $D_{ve\_wet}$  logically follows the decrease  
292 of RH in the chamber,  $D_{ve\_dry\_raw}$  shows a sudden decrease from 100 nm to 90 nm at 14h,  
293 corresponding to the efflorescence point of the AS particles. The same behavior happens with the  
294 total mass concentrations (Fig. 2, bottom panel).

295 After applying the procedure, the corrected dry volume-equivalent diameter  
296 ( $D_{ve\_dry\_corrected}$ , Fig. 2, middle panel) remains stable at approximately 90 nm throughout the  
297 experiment. The corrected dry mass (Mass\_dry\_corrected, Fig. 2, bottom panel) shows a smooth  
298 exponential decrease throughout the experiment, due to the decrease of the particle number  
299 concentration. Several data points (pink shaded area in Fig. 2) represent the efflorescence transition  
300 when RH in the MPSS ranges from 30 to 33%, in good agreement with the water uptake studies of  
301 pure AS particles (Zardini et al., 2008; Vlasenko et al., 2017; Lei et al., 2020; Lei et al., 2018; Zhang  
302 et al., 2023). This transition period is removed from the final corrected data. Note that, due to the  
303 RH% in the MPSS, four points between 20 and 60 minutes were already in this transition period,  
304 they were also removed from the final corrected data.



305

306 **Figure 2** : Comparison between corrected and uncorrected data of dry AS particles under decreasing  
307 RH conditions (Exp 1a). Time profiles of RH in CESAM and in the MPSS instrument (top panel),  
308 raw (uncorrected) volume equivalent diameter ( $D_{ve\_dry\_raw}$ ) and corrected volume equivalent  
309 diameter under dry condition ( $D_{ve\_dry\_corrected}$ ), and volume equivalent diameter under wet  
310 condition ( $D_{ve\_wet}$ ) (middle panel), raw (uncorrected) total mass concentration (Mass\_dry\_raw),  
311 corrected total mass concentration (Mass\_dry\_corrected), and total number concentration (bottom



312 panel). The period of efflorescence is shaded pink, and open blue symbols denote the excluded data  
313 points

314 These results show the importance of considering the applied correction for the dry size of the  
315 particles. Although this correction procedure generated larger uncertainties than direct measurements  
316 on the particles mass (26%, Tab. 1), these corrections did not affect  $f_{o/s}$  which was used to calculate  
317 the net evaporation of levoglucosan from AS particles.

318

319 2.6 Calculation of the net evaporation of levoglucosan from AS particles

320 The net evaporation rate of levoglucosan from the AS particles was determined by measuring  
321 the mass concentrations of sulfate and levoglucosan using the AMS. Due to dilution, wall loss  
322 processes, and gas-particle partitioning, both concentrations decreased with time, but at different  
323 rates:

$$324 \quad [S]_t = [S]_0 \exp(-k_1 t) \quad (3)$$

$$325 \quad [OA]_t = [OA]_0 \exp(-k_2 t) \quad (4)$$

326 where  $[S]_t$  is the sulfate mass concentration at time  $t$ , and  $[OA]_t$  the particle-phase levoglucosan mass  
327 concentration at time  $t$  (time 0 is the time when the introduction procedure was finished);  $k_1$  is the  
328 decay rate constant including wall loss processes and dilution of sulfate, and  $k_2$  the combined decay  
329 rate constant of wall loss processes and dilution, as well as gas-particle partitioning of levoglucosan.  
330 Combining (1) and (2) gives:

$$331 \quad [OA]_t/[S]_t = [OA]_0/[S]_0 \exp(-k_{net} t) \quad (5)$$

332 where  $k_{net}$  is the resulting rate constant:  $k_{net} = k_2 - k_1$ . Assuming an internal mixture of the particles  
333 one can state that the dilution and wall loss processes are the same for both sulfate and particle-phase  
334 levoglucosan. As a result,  $k_{net}$  is the rate constant representing the net evaporation of levoglucosan  
335 from the particles, and its values are positive when levoglucosan evaporates from the particles,  
336 whereas they are negative when levoglucosan condenses onto the particles.

337 Absolute uncertainties of  $k_{net}$  values (Tab 2) were calculated from their relative uncertainties which  
338 was the sum of the relative uncertainties of  $f_{o/s}$  and those given by the linear regression of the  
339  $\text{Log}(f_{o/s})$  versus time.

### 340 3. Results and discussions

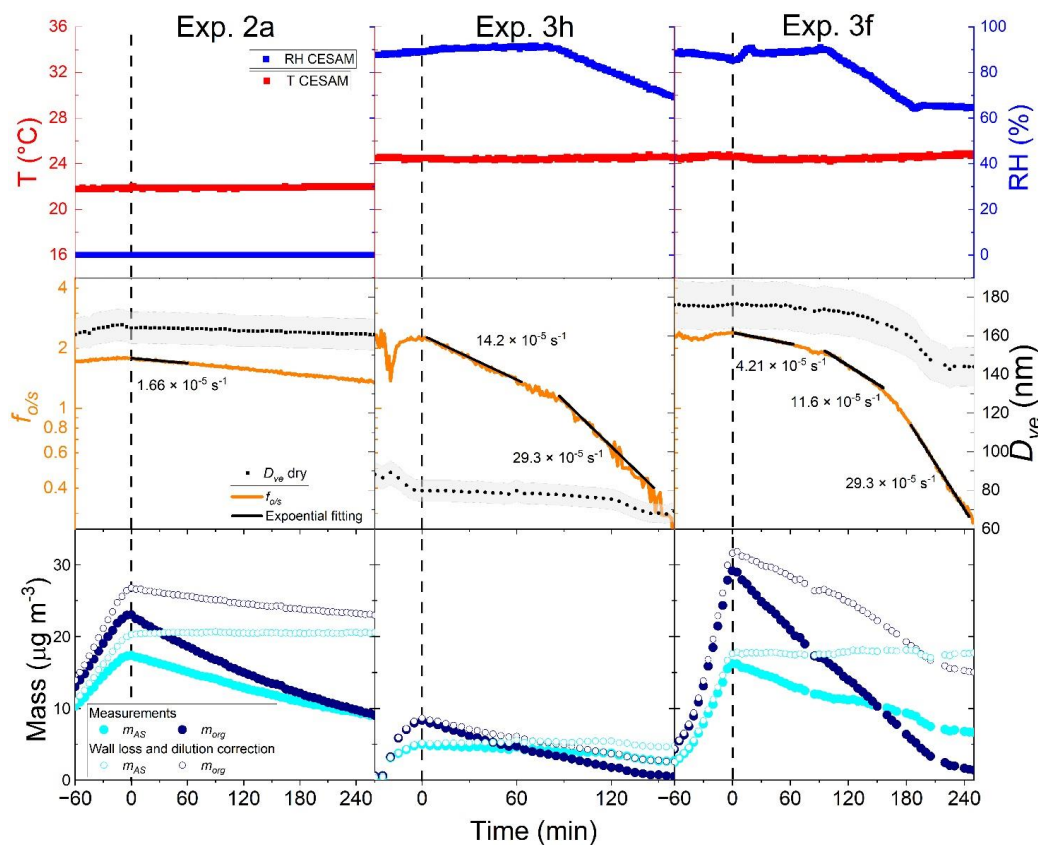
#### 341 3.1 Observation of levoglucosan evaporation under various RH%

342 In each experiment, once the particles are introduced, they stabilize and undergo continuous  
343 dilution and wall loss, leading to a steady decrease in the mass concentration of each component.  
344 Figure 3 shows three experiments performed under different conditions: constant dry 0% RH with  
345 ~200 nm injected dry size particles (left panels); decreasing RH starting from > 90% with ~100 nm  
346 injected dry size particles (middle panels), and with ~200 nm injected dry size (right panels). The  
347 vertical dashed lines correspond to time  $t = 0$ . Note that the initial dry size ( $D_{ve}$ ) at time  $t = 0$  is  
348 always below the targeted size (100 or 200 nm) and can vary from one experiment to another due to



349 the variable time taken for particle injection (from 1 to 2 hours), during which evaporation already  
 350 occurred.

351



352

353 **Figure 3.** Evaporation of levoglucosan during three experiments performed under different RH and  
 354 particle monodispersed size conditions: dry 0% RH at ~200 nm (Exp 2a, left column), decreasing  
 355 RH at ~100 nm (Exp 3h, middle column), and decreasing RH at ~200 nm (Exp 3f, right column).  
 356 Time series of T, RH (top row), volume equivalent dry particle size ( $D_{ve}$ ), organic to sulfate  
 357 mass ratio ( $f_{o/s}$ ) (middle row), masses of AS ( $m_{AS}$ ) and total organic ( $m_{org}$ , attributed to levoglucosan) with  
 358 and without wall loss and dilution corrections (bottom row). The grey shades show the uncertainty  
 359 of  $D_{ve}$  after correction (see SI-3). The black lines indicate the exponential fitting curves (during 60  
 360 min) to determine  $k_{net}$  (according to Eq. 5), which values are shown. The vertical dashed line marks  
 361 time 0, at the end of particle introduction into CESAM.

362 In all experiments, although T was constant throughout each experiment (Fig. 3, top row),  
 363 particle shrinking was observed versus time (Fig. 3, middle row, black dots). Initially, when RH was  
 364 kept constant (dry (Exp 2a) or wet (Exp 3h and 3f just after time 0)), the shrinking was only slight  
 365 but became significant once RH started to decrease. For example, according to the variation of  $D_{ve}$   
 366 in Exp 3h (middle panel), a total particle volume of 6.8% was lost in the first 70 min when RH was  
 367 constant at  $89.1 \pm 0.6\%$ , and this value increased to 33% in the next 70 min when RH decreased down



368 to 70%. In Exp 3f (right panel), the total volume loss was 5.4% in the first 97 min (RH was constant  
369 at  $88.9 \pm 1.1$  %) and increased to 32% in the next 97 min (RH decreased down to 65%).

370 Figure 3 (bottom rows) shows that, after dilution and wall loss corrections (Doussin et al., 2023),  
371 the mass concentrations of AS remained almost constant over time whereas that of levoglucosan  
372 (represented by OA) decreased continuously. Thus, the observed particle shrinking versus time was  
373 due to the evaporation of levoglucosan. This is consistent with the non-volatility of AS and the  
374 significant volatility of levoglucosan. This observation becomes even more obvious in the decrease  
375 of the organic to sulfate mass ratio versus time (Fig. 3, middle rows). The organic to sulfate mass  
376 ratio constantly decreases versus time under constant RH, and it decreases at a significantly higher  
377 rate when RH decreases. This is reflected by the net evaporation rate ( $k_{net}$ ) values, calculated from  
378 the decrease of the organic to sulfate mass ratio. For each experiment,  $k_{net}$  shows higher values when  
379 RH decreases than when it is constant. For instance, at  $\sim 100$  nm dry size (Exp 3h, middle column),  
380  $k_{net}$  increases by a factor of 2.1 (from  $14 \times 10^{-5} \text{ s}^{-1}$  to  $29 \times 10^{-5} \text{ s}^{-1}$ ) when RH decreases from 92% to  
381 70%; at  $\sim 200$  nm dry size (Exp 3f, right column),  $k_{net}$  increases by a factor of 2.8 (from  $4.2 \times 10^{-5} \text{ s}^{-1}$   
382 to  $12 \times 10^{-5} \text{ s}^{-1}$ ) when RH decreases from 90% to 65%. The factors influencing this net evaporation  
383 rate will be discussed in detail in section 3.3.

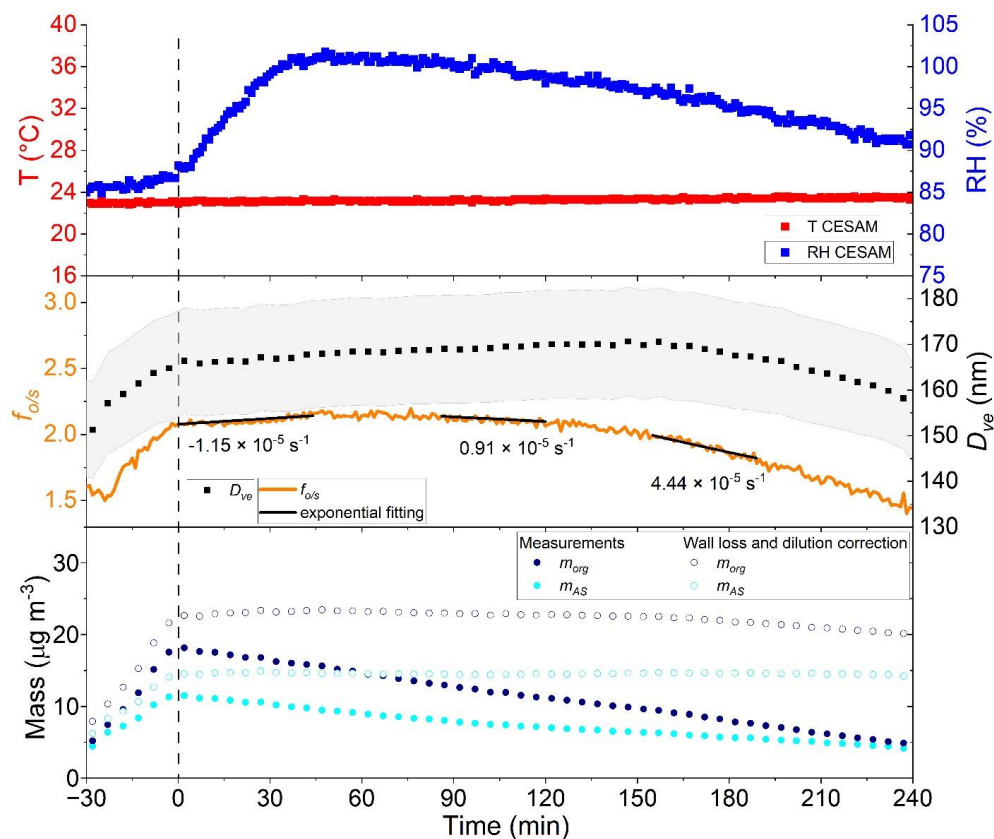
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### 385 3.2 Co-condensation of levoglucosan

386 Due to the high deliquescence RH of AS, all the experiments conducted under humid  
387 conditions were initialized at RH above 80%, then, RH either remained constant or decreased over  
388 time (Table 1). However, in one experiment (3d), we were successful in increasing the relative  
389 humidity to 100% after the particle injection. Note that, although the very high relative humidity, no  
390 cloud formation was observed in CESAM, as shown by the absence of particle washout (Giorio et  
391 al., 2017). This appeared to impact the trend in the organic-to-sulfate mass ratio. As shown in Fig. 4,  
392 an increase in RH from 88% to 100% coincided with a rise in the organic to sulfate mass ratio from  
393 2.1 to 2.2, suggesting that levoglucosan was co-condensing with water vapor. This effect is further  
394 supported by dry MPSS measurements, which show a slight increase in the corrected dry  $D_{ve}$  during  
395 RH increase. Note however that this increase continues even when the RH stabilizes. This disagrees  
396 with the change in  $f_{o/s}$  evolution after 60 minutes in Fig. 4. It is probably due to the uncertainties  
397 generated by the dry  $D_{ve}$  corrections, which however do not affect  $f_{o/s}$  values (see section 2.5.2).

398 Using Eq. 5 to calculate  $k_{net}$  (see Sect. 2.6), a negative value of  $-1.15 (\pm 0.11) \times 10^{-5} \text{ s}^{-1}$  was  
399 determined by fitting the organic to sulfate mass ratios ( $f_{o/s}$  in Fig. 4, middle row) during the RH  
400 increase. This indicates condensation of levoglucosan, as evidenced by the negative sign of the rate  
401 constant and the fact that its magnitude significantly exceeds the uncertainty. Furthermore, the  
402 absolute value of this co-condensation rate constant is in the range of the fitted net evaporation rate  
403 constants obtained in other experiments which span from  $0.91 \times 10^{-5}$  up to  $29 \times 10^{-5} \text{ s}^{-1}$  (Table 1).  
404 However, given the likely significant competitive uptake of gas-phase levoglucosan onto the  
405 chamber walls (Lamkaddam et al., 2017), the calculated co-condensation rate constant is likely  
406 influenced by the substantial wall losses, the full quantification of which is out of the scope of the  
407 present study.

408



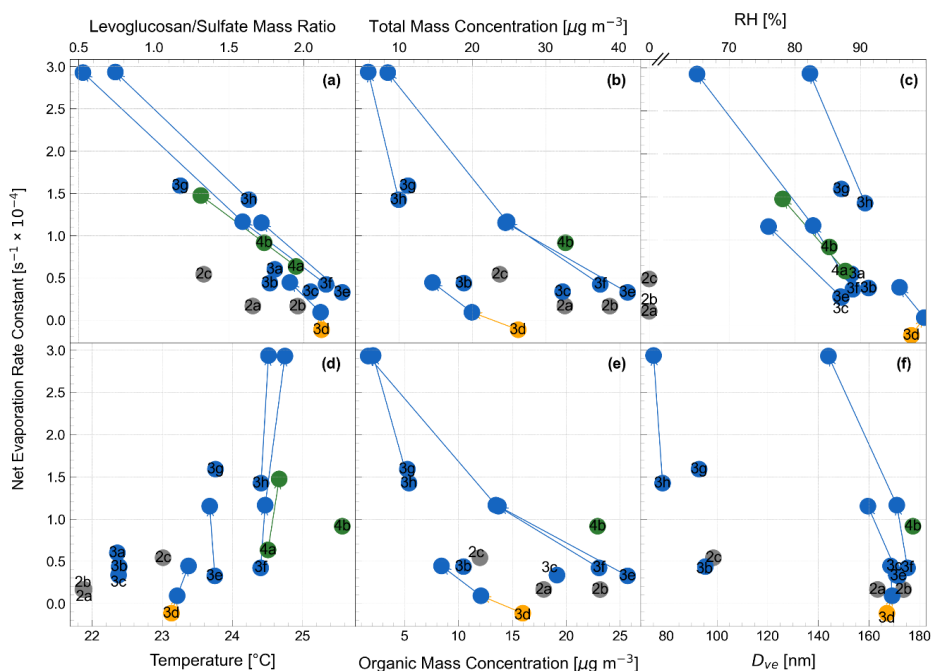
409

410 **Figure 4.** Co-condensation, then evaporation of levoglucosan during experiment 3d, performed  
411 under varying RH. Time series of T, RH% (top panel), volume equivalent dry particle size ( $D_{ve}$ ),  
412 organic to sulfate mass ratio ( $f_{o/s}$ ) (middle panel), ammonium sulfate and total organic (attributed to  
413 levoglucosan) masses (with and without wall loss and dilution corrections) (bottom panel). The grey  
414 shades show the uncertainty of  $D_{ve}$  after correction (see SI-3). The three black lines indicate the  
415 exponential fitted curve to determine  $k_{net}$  values (shown) during 45 min when RH increases, and 40  
416 min when RH stays constant, and decreases, respectively. The vertical dashed line marks time 0,  
417 at the end of particle introduction in CESAM.

418

### 419 3.3 Factors driving the net evaporation rate constant of levoglucosan

420 Figs. 3 and 4 clearly demonstrate the evaporation and condensation of levoglucosan from and onto  
421 AS particles for several experiments. Yet, it seems that the extent of these phenomena is linked to  
422 multiple factors, not only the variations in RH. In this section, we discuss the influence of the  
423 measured parameters on the partitioning of levoglucosan. Figure 5 shows the values of  $k_{net}$  versus  
424 each of the investigated parameters across all experiments.



425

426 **Figure 5.** Net levoglucosan evaporation rate constant  $k_{net}$  as a function of levoglucosan to sulfate  
427 mass ratio (a), total mass concentration (b), RH (c), temperature (d), organic mass concentration (e)  
428 and particle dry diameter (f). Exp 2a, 2b, 2c (gray dots) were performed under dry conditions (RH =  
429 0%). Exp 4a and 4b (green dots) involve SDS. Blue dots represent experiments performed at high  
430 initial RH either constant or decreasing. The orange dots correspond to the 1<sup>st</sup> part of Exp 3d, when  
431 RH increased from 88 to 100% (Sect. 3.2). The arrows connect experiments performed under varying  
432 RH conditions, their arrowheads pointing towards increasing time.  $k_{net}$  are plotted at the middle value  
433 for parameters that change during the evaluation period (i.e. all parameters except from T).

434 Overall, Fig. 5 shows that  $k_{net}$  values are largely influenced by the mass ratio of levoglucosan  
435 to sulfate (Fig. 5a). A general trend is observed where lower levoglucosan-to-sulfate ratios  
436 correspond to higher  $k_{net}$  values, potentially due to a salting-out effect. As the experiment progresses  
437 and levoglucosan volatilizes, the proportion of AS in the particles increases, which likely enhances  
438 the salting-out effect, thereby accelerating the volatilization rate. AS has been reported to exhibit a  
439 salting-out effect for polyol/water mixtures (Marcolli and Krieger, 2006). This effect, which depends  
440 on the oxygen-to-carbon ratio (Song et al., 2012), the molecular structure of the organic molecules,  
441 and the organic-to-sulfate mass ratio (Bertram et al., 2011) likely enhances the evaporation (Angle  
442 et al., 2023).

443 In addition to the levoglucosan-to-sulfate mass ratio, RH (Fig. 5c), particle size (Fig. 5f) and  
444 temperature (Fig. 5d) also influence the  $k_{net}$  values. Under dry conditions (Exp 2a and 2b), the  $k_{net}$   
445 values are among the lowest positive values compared to particles of similar size under higher RH  
446 (Fig. 5f). This can be explained by the fact that levoglucosan effloresces at very dry conditions in the  
447 presence of crystalline AS (Parsons et al., 2004), and, as a crystalline solid it has a lower vapor  
448 pressure compared with the supersaturated solutions (Booth et al., 2011). Moreover, it is possible  
449 that levoglucosan forms an external mixture with AS in Exp 2a, 2b and 2c, this would explain why

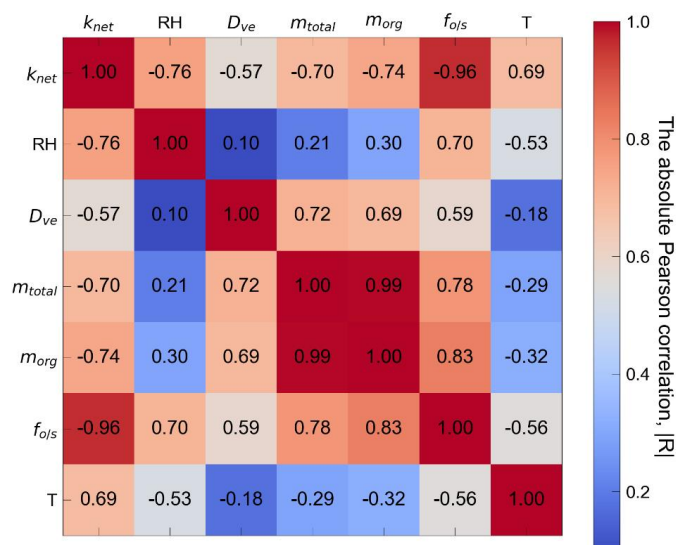


450  $k_{net}$  of these experiments do not align well with the others in Fig. 5a. However, Exp 2a and 2b were  
 451 performed under the lowest temperature, which also influences  $k_{net}$  values: Fig. 5d shows that  
 452 increasing T provides higher  $k_{net}$  values. For wet particles, Fig. 5c shows that decreasing RH  
 453 systematically results in higher  $k_{net}$  values, thus potentially indicating co-evaporation of levoglucosan.  
 454 Also, larger particles are associated with slower volatilization rates (Fig. 5f) due to their decreasing  
 455 surface/volume ratios with increasing size. In the same way, lower total mass (Fig. 5b) is associated  
 456 with higher evaporation, consistently with evaporation being exacerbated by N<sub>2</sub> dilution in the  
 457 chamber.

458 Overall, the lowest positive  $k_{net}$  values (with a corresponding half-life of approximately 12  
 459 hours) are observed when the levoglucosan-to-sulfate mass ratio is highest, and either RH is high (>  
 460 80 %) or the temperature is low. The highest  $k_{net}$  values (with a corresponding half-life of  
 461 approximately 1 hour) occur when the levoglucosan-to-sulfate mass ratio is lowest, and either RH is  
 462 low or temperature is high.

463 The analysis above demonstrates physical trends, but it is difficult to isolate the individual  
 464 effects of each parameter, as they are not completely independent. To elucidate their combined  
 465 influence on the  $k_{net}$  values and investigate their interrelationships, we constructed a Pearson pairwise  
 466 linear correlation matrix derived from the original datasets shown in Fig. 5 for Exp 3b to 3h (see SI-  
 467 4 for the details). Exp 3a was excluded from this matrix due to the lack of data for  $D_{ve}$ ,  $m_{org}$  and  $m_{total}$ .  
 468 Furthermore, Exp 4a and 4b were done under different conditions (with the presence of SDS) and  
 469 are treated separately in the next section. The resulting matrix is shown in Figure 6.

470



471

472 **Figure 6.** Correlation matrix showing pairwise Pearson correlation coefficients among the  
 473 investigated parameters and the net evaporation rate  $k_{net}$  for Exp 3b to 3h.  $D_{ve}$  is the dry volume  
 474 equivalent particle size;  $m_{total}$  is the total mass concentration;  $m_{org}$  is the organic mass concentration,  
 475  $f_{ols}$  is organic-to-sulfate mass ratio; T is the temperature.



476

477 The resulting matrix (Figure 6) clearly reveals that, under our experimental conditions, the  
478 levoglucosan-to-sulfate mass ratio serves as the dominant factor governing the volatilization rate of  
479 levoglucosan. RH emerges as the second most influential parameter, closely followed by the organic  
480 mass and the total mass, demonstrating their importance. The dry particle size and temperature  
481 appear to be the least influential parameters on  $k_{net}$  values, but their absolute Pearson correlations are  
482 still higher than 0.5.

483 As mentioned above, the strong influence of the levoglucosan-to-sulfate mass ratio can be  
484 explained by the salting-out effect that is strengthened when the proportion of salt increases while  
485 levoglucosan evaporates from the particles. Remarkably, RH is independent of all parameters except  
486 the two linked parameters,  $f_{o/s}$  and  $k_{net}$ . This demonstrates that levoglucosan co-evaporates (or co-  
487 condenses) with water in these experiments. In Fig 6, Exp 4a and 4b were excluded as they were  
488 performed under different conditions (SDS presence), they are discussed below.

### 489 3.4 Influence of the presence of SDS

490 Two experiments, Exp 4a and 4b, were performed with addition of SDS, to investigate the impact of  
491 a surfactant on levoglucosan partitioning. As a surfactant, SDS is expected to form a layer on the  
492 surface of the particle, potentially affecting the partitioning of both water and levoglucosan. However,  
493 as seen in Fig. 5, the presence of SDS does not seem to impact the  $k_{net}$  values. This suggests that the  
494 presence of SDS does not affect the net evaporation of levoglucosan. Contrasting results have been  
495 reported in previous studies on the effect of SDS on the kinetics of water evaporation. Experiments  
496 performed by (Taflin et al. 1988) and (Shulman et al. 1997) indicated that the presence of SDS slows  
497 down the evaporation of water. However, other studies suggested that SDS has no effect on the  
498 kinetics of water evaporation (Davies et al. 2012; Doganci et al. 2011; Lunkenheimer et Zembala  
499 1997). Davies et al. (2012) explain their results with the large, charged head group of SDS, which is  
500 unlikely to provide a densely packed monolayer of SDS at the surface. More generally, soluble  
501 surfactants such as SDS seem to have little effects on the evaporation of water (Lunkenheimer et  
502 Zembala 1997). According to (Doganci et al. 2011), two competing effects arise from the presence  
503 of SDS: i) surface tension reduction would promote water evaporation, and ii) the presence of SDS  
504 molecules at the surface would hinder molecules diffusion and thus impede water evaporation.  
505 However, to account for these effects, the surface tension and surface coverage of SDS must be  
506 estimated in Exp 4a and 4b.

507 In the particle, surfactant SDS molecules partition between the ‘interior’ of the particle – the bulk –  
508 and the surface. The bulk concentration of SDS and its surface coverage were estimated from the  
509 total SDS concentration in the particle and the geometry of the particle, following the methodology  
510 proposed by (Jacobs et al. 2024). This methodology accounts for the high surface area – to – volume  
511 ratios of small particles (see Table 2). The SDS concentration is calculated from both AMS data and  
512 the filters collected during the experiments, and using the wet particle  $D_{ve}$  obtained from the SMPS.  
513 More details on the calculations can be found in SI-2. The results are shown in Table 2 at three and  
514 two time-steps for Exp 4a and 4b, respectively.

515 The results presented in Table 2 for filters and AMS are approximate estimations, due to the  
516 uncertainty on AMS derived SDS concentration (see SI-2-1) and because the exact flow rate of filter  
517 sampling was not precisely controlled (see SI-2-2). For Exp 4b, there is close agreement between  
518 the values obtained from AMS data and filter data. However, there is a significant difference between



519 the results from the AMS and the filter for Exp 4a, with a factor 5-7 for the estimated total  
 520 concentration in the particle. This leads to significant differences between the bulk concentrations,  
 521 surface coverages, and surface tension calculated from AMS and filter data, although these  
 522 differences can arise from the uncontrolled sampling flow rate or the uncertainty on AMS derived  
 523 SDS concentration (see SI-2-1). In particular, the bulk concentration is predicted to be below the  
 524 CMC during experiment 4a according to filter data, whereas AMS predicts a bulk concentration  
 525 above the CMC. This explains the discrepancies between AMS and filter data for surface coverage  
 526 and surface tension. Note that the surface tension data used for these estimations do not account for  
 527 the presence of ammonium sulfate in the particle, which can greatly impact bulk – surface  
 528 partitioning due to salting-out effects (El Haber et al., 2023). Therefore, the surface coverages and  
 529 surface tensions estimated in Table 2 can be considered as lower limits and upper limits of the true  
 530 values, respectively.

531

532 **Table 2.** SDS experiments (Exp 4a and 4b): estimated surface-to-volume (A/V) ratios, SDS  
 533 concentrations in the particle, surface coverages by SDS (detailed in SI-2). SDS concentration and  
 534 surface coverages were calculated from both AMS and filter data. A/V ratios and results calculated  
 535 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
$k_{net}$ ( $\text{min}^{-1}$ )		$3.8 \cdot 10^{-3}$	$3.8 \cdot 10^{-3}$	$8.8 \cdot 10^{-3}$	$5.5 \cdot 10^{-3}$	$5.5 \cdot 10^{-3}$
A/V ratio ( $\times 10^6 \text{ m}^{-1}$ )		23.1	23.3	26.9	26.9	26.9
Wet $D_{ve}$ (nm)		260	257	223	248	261
SDS total concentration ( $\times 10 \text{ mmol L}^{-1}$ )	AMS	39	55	88	93	61
	Filter	8.5	8.8	13	114	69
SDS bulk concentration ( $\times 10 \text{ mmol L}^{-1}$ )	AMS	14	29	58	62	36
	Filter	0.16	0.16	0.25	84	44
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	98	99	99	100	99
	Filter	32	33	43	99	99
Surface tension ( $\text{mN m}^{-1}$ )	AMS	35	35	35	35	35
	Filter	61	61	56	35	35

536

537 Overall, the estimations suggest that particles were indeed coated with SDS, and that surface tension  
 538 was reduced compared to pure water. Nevertheless, the net evaporation of levoglucosan seems to be  
 539 unaffected by this coating and surface tension reduction, as shown in Fig. 5. This suggests that these  
 540 two effects counteract, as suggested by (Doganci et al. 2011), or that even at high surface coverages,  
 541 SDS is not tightly packed enough to prevent water molecules diffusion. In addition, note that the  
 542 bulk SDS concentrations are smaller than the levoglucosan and AS concentrations, suggesting that  
 543 bulk SDS has a small impact on bulk properties and thus on  $k_{net}$ . Moreover, although our calculations  
 544 predict full coverage of SDS, it is possible that surfactants are not evenly distributed across the  
 545 surface, i.e. that surface concentration is not homogeneous. (Griffith et al. 2012) showed that



546 surfactants can exhibit heterogeneity at the surface, with regions of 3D hydrophobic aggregates,  
547 condensed monolayer, or exposed hydrophilic “holes”, where the solute is at the surface in place of  
548 surfactant molecules. Although the surfactant used in the latter study was not SDS, it is still possible  
549 that the particle surface presents such hydrophilic holes due to the presence of levoglucosan, leading  
550 to unaffected levoglucosan evaporation.

#### 551 4. Conclusions and atmospheric implications

552 The ability of a semi-volatile water-soluble organic compound to co-condense and co-  
553 evaporate onto or from inorganic particles was investigated. The gas-particle partitioning of  
554 levoglucosan was monitored under ambient-like conditions on ammonium sulfate (AS)  
555 monodispersed particles in the CESAM simulation chamber. Various conditions were explored,  
556 including RH (from dry to 100%), particle mass and size, and the presence of a surfactant on the  
557 particles. The net evaporation flux of levoglucosan was quantified under each condition.

558 Due to the high deliquescence point of AS, wet experiments were initialized at RH above 80%.  
559 In these experiments, special care was taken with RH control and measurements. To maintain high  
560 RH values in the chamber while preventing liquid water from depositing on the walls, the  
561 HUMidificAtioN System (HUMANS) was connected to CESAM to regulate RH precisely. The  
562 saturation water vapor pressure and the water vapor pressure were derived from the Buck equation,  
563 using the dew point (measured by a chilled mirror) and the average temperature (provided by three  
564 thermocouples Type T sensors suspended in the air inside CESAM), respectively. RH was calculated  
565 as the ratio between these two vapor pressures. All these precautions made it possible to perform  
566 experiments under constant high RH for minutes to hours (and as high as 100%), with a precise  
567 control of RH variations. It was shown that these long-time high RH conditions impacted the drying  
568 process required to determine the dry volume equivalent diameter of the particles. To achieve this  
569 under our experimental conditions, the RH limit value was set at 15% for all experiments.

570 The results showed that the net evaporation rate of levoglucosan was sensitive to all the  
571 investigated parameters, but the main drivers under our experimental conditions were the  
572 levoglucosan-to-sulfate mass ratios and RH. The effect of the levoglucosan-to-sulfate mass ratios  
573 was interpreted as the increasing salting-out effect of the organic fraction while decreasing the  
574 particle water content. The influence of RH provides experimental proof of co-evaporation or co-  
575 condensation of levoglucosan from or onto AS particles.

576 To investigate the impact of a surfactant on levoglucosan partitioning, out of the fifteen  
577 experiments, two were performed with addition of sodium dodecyl sulfate (SDS) under wet  
578 conditions. As a surfactant, SDS was expected to form a layer on the surface of the particle,  
579 potentially affecting the partitioning of both water and levoglucosan. The quantification of SDS in  
580 the particles made it possible to estimate that particles were indeed coated with SDS, and that surface  
581 tension was reduced compared to pure water. Nevertheless, compared to the thirteen other  
582 experiments, the net evaporation of levoglucosan seemed to be unaffected by this coating and surface  
583 tension reduction. This result was interpreted as the counteracting effects of coating and surface  
584 tension reduction, and/or as the potential heterogeneity of SDS molecules at the particle surface,  
585 inducing, in some places, a lack of packing tightness of the SDS layer to inhibit diffusion across it.  
586 These showed that more investigations should be carried out on the effect of surfactants on the



587 partitioning of semi-volatile organic compounds, using various types of surfactants and investigating  
588 observations of the surface properties.

589 Overall, the results showed that the net evaporation flux of levoglucosan for particle-phase  
590 levoglucosan corresponds to atmospheric lifetimes of 1 to 12 hours, which is comparable to the  
591 photochemical degradation rates of the particle phase (approximately 17 hours) at similar  
592 temperatures. This result highlights the importance of considering the phase partitioning of semi-  
593 volatile compounds when assessing their atmospheric multiphase reactivity.

594 These findings should be followed up with consideration of the co-evaporation or co-  
595 condensation of organic, semi-volatile, water-soluble species when determining the CCN ability of  
596 particles. A modeling study of these data is also required.

597

#### 598 **Author Contributions**

599 JX, JW and AM wrote the article. JX, BTR, Julien K, JG, MGS, SD, SH, EP, and KZA participated  
600 in the CESAM chamber campaign that was coordinated by JW, BPV, MC, and AM. JX processed  
601 and interpreted the data with support from JW, BTR, Judith K, NB and supervised by AM. JG and  
602 FRP analyzed and interpreted the surfactants data. KZA, CM, and AM secured funding for the  
603 ORACLE project. All co-authors reviewed the article.

#### 604 **Data availability**

605 The data that support the findings of this study are openly available in ACTRIS DC ASC Unit (also called EUROCHAMP  
606 DC) <https://www.eurochamp.org/>.

#### 607 **Notes**

608 The authors declare no conflict of interest.

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