

Supplement to

“Cloud droplet number enhancement from co-condensing NH₃, HNO₃, and organic vapours: sensitivity study”

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10

Contents

	S1. Derivation of the volatility basis set.....	2
15	S2. Selection of model compounds for organic fractions.....	4
	S3. Inorganic ions balance for AIOMFAC model.....	9
	S4. Calculation of activity coefficients for organic fractions and water.....	10
	Figure S3	13
	Figure S4	14
20	Table S8.....	15
	References	16

S1. Derivation of the volatility basis set

25 To set up the two different VBS displayed in Fig. 2, we combined information from the experimental and modelling studies summarised in Table S1. Stirnweis et al. (2017) derived a VBS with bins $\log(C^*) = -1$ to 2 evaluating smog chamber organic yields for photo-oxidation of alpha-pinene at various NO_x/VOC ratios and RH from 23 to 69 % with $T = 21\text{--}26^\circ\text{C}$. They used a Monte Carlo simulation to parameterise the smog chamber SOA yields as a function of the condensed phase absorptive mass. This approach enabled the determination of VBS for eleven experiments. Table S1 lists the average normalised volatility bin distribution from these experiments. Stirnweis et al. (2017) refrained from determining the loading of the bin $\log(C^*) = 3$ as it could not be reliably extracted from the experimental data. Therefore, the mass that would be present in this bin was assigned to the bin $\log(C^*) = 2$, which may explain the large mass fraction assigned to it.

Lane et al. (2008) proposed a SOA parameterisation based on the SPARC99 chemistry mechanism for the oxidation of biogenic and anthropogenic precursors, which they lumped into different VOC classes of alkanes, olefins, aromatic compounds, isoprene, terpenes and sesquiterpenes. They determined normalised aerosol mass fractions in the volatility bins from $\log(C^*) = 0$ to 3 at 300 K. Table S1 lists averaged and renormalised volatility distribution factors for the base case and high-yields. Tsimpidi et al. (2010) simulated organic aerosol formation in the Mexico City metropolitan area for different types of precursors following a similar approach to derive a SOA parameterisation based on the SPARC99 mechanism at 298 K for the same VOC classes but divided into high and low NO_x . Table S1 gives averaged normalised contributions to the basis sets for “high NO_x ”, “low NO_x ” and “all NO_x ” conditions. Koo et al. (2014) developed a hybrid VBS that includes POA volatility distributions for different emission sources, including biomass burning based on chamber studies performed by May et al. (2013).

Cappa and Jimenez (2010) derived various VBS based on thermodenuder-aerosol mass spectrometer measurements and kinetic modelling with $\log(C^*)$ bins from -6 to 3. In Table S1, their VBS “total OA” is given with a lumped mass fraction for the bins with $\log(C^*) \leq -1$ and in normalised mass fractions.

The compilation of VBS in Table 1 shows that the aerosol mass fraction per bin increases with the volatility of the bin for SOA. POA shows a similar trend, except for the lowest volatility bin, which represents the species that can be considered as non-partitioning to the gas phase and therefore require no further discrimination with respect to saturation concentration. Its lumped character may explain why it contradicts the trend of increasing mass fraction per bin with increasing volatility.

50 Due to the similar aerosol mass distribution of the different VBS, we decided to set up two generic VBS that are representative of both LO-OOA and MO-OOA. The first one is based on SOA yields from Tsimpidi et al. (2010) and POA from Koo et al. (2014) to include primary and secondary contributions. Assuming that SOA dominates, we combine the averaged factors for all NO_x conditions from Tsimpidi et al. (2010) with the BBOA primary emissions from Koo et al. (2014) at a ratio of 2:1. As no information concerning the mass fraction in $\log(C^*) = 4$ could be found, we derived it by extrapolating the aerosol mass fractions in the $\log(C^*) = 0\text{--}3$ bins (see Figure S1) to obtain the final VBS by normalizing the summed mass from all bins to

one and renormalized thereafter (Table S1, “VBS 1”). The second generic VBS corresponds with “total OA” from Cappa and Jimenez (2010), which was extrapolated to obtain a $\log(C^*) = 4$ bin and renormalised (Table S1, “VBS 2”).

Table S1. Published VBS from experimental and modelling studies together with two generic VBS derived from them.

References and conditions	Volatility bins $\log(C^*)$					
	-1	0	1	2	3	4
Stirnweis et al. (2017)	0.016	0.056	0.137	0.791	nd	
Lane et al. (2008), base case		0.052	0.119	0.303	0.525	
Lane et al. (2008), high-yield case		0.114	0.270	0.296	0.321	
Tsimpidi et al. (2010) high NO _x		0.018	0.163	0.315	0.503	
Tsimpidi et al. (2010) low NO _x		0.054	0.180	0.303	0.463	
Tsimpidi et al. (2010), all NO _x conditions		0.038	0.173	0.308	0.480	
Koo et al. (2014), POA BBOA emission factors	0.2*	0.100	0.100	0.200	0.400	
Cappa and Jimenez (2010), total OA	0.242*	0.054	0.086	0.176	0.441	
This study: VBS 1 (based on Tsimpidi all NO _x & Koo)	0.042*	0.036	0.090	0.162	0.269	0.401 [†]
This study: VBS 2 (based on Cappa and Jimenez, total OA)	0.121*	0.03	0.044	0.093	0.238	0.474 [†]

60 * Represents the sum of all bins with $\log(C^*) \leq -1$; [†] derived by extrapolation from $\log(C^*)$ bins 0 to 3, see Figure S1.

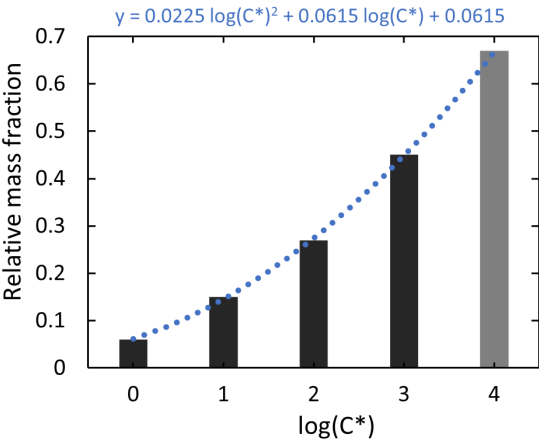


Figure S1. Extrapolation by second order polynomial to obtain a $\log(C^*) = 4$ bin for VBS 1.

S2. Selection of model compounds for organic fractions

65 To estimate the activity coefficients of LO-OOA and MO-OOA in aqueous solution with electrolytes, activity coefficients were pre-calculated with the group contribution model AIOMFAC (Zuend et al., 2008; Zuend et al., 2011) resulting in a look-up table for usage in the parcel model. For the calculation of activity coefficients with AIOMFAC, specific compounds are required as input, yet the exact composition of the organic fractions in Hyytiälä (LO-OOA and MO-OOA) is unknown. Therefore, a set of model compounds for LO-OOA and MO-OOA was created, which is presented in Tables S2 and S3, 70 respectively. Organic compounds were chosen according to the following criteria:

- **Vapor pressure** in the $\log(C^*)$ bin needs to be within ± 1 of the nominal value of the bin. So, for the $\log(C^*) = 1$ bin, the combination of vapor pressure p_{vap} and molecular weight MW must yield $C^* = 10^6 MW \gamma p_{vap} / (760 RT) = 1\text{--}100 \mu\text{g m}^{-3}$. Note that the assignment to volatility bins can only be approximate, as it also depends on the activity coefficients γ of the substances in the solution (set to 1 here), which again depend on the solution composition. The subcooled liquid vapor 75 pressure of each substance shown in the column “Est. p_{vap} ” in Tables S2 and S3 was estimated with the EVAPORATION method by Compernelle et al. (2011). In the subsequent modelling, vapor pressures of the substances were set to an artificial value to exactly match the $\log(C^*)$ of the bin (see column Set p_{vap} in Tables S2 and S3). By doing so, consistency of the pre-calculations with the parcel model was ensured, where the volatility of the organic bins is derived directly from the $\log(C^*)$ value.
- 80 • **O:C ratio** needs to be close to the estimated average O:C ratio of the bin. While the exact degree of non-ideality of a mixture depends on the specific functional groups of each mixture constituent, the O:C ratio has proved to provide a good proxy for non-ideality (Bertram et al., 2011). Ciarelli et al. (2017) determined average O:C ratios of a VBS from aged wood burning emissions for two different sets of SOA compounds (set 2: oxidation productions from semi-volatile organic compounds from primary emissions, set 3: oxidation products from non-traditional volatile organic compounds). Here, 85 we use the O:C ratios of SOA set 2 and SOA set 3 to estimate the O:C ratios of LO-OOA and MO-OOA, respectively (see Table S4). Substances were chosen to have an O:C ratio very close (± 0.13) to these O:C ratios.
- **Molecular weight (MW)** needs to be close to the estimated average MW of the bin. Similar to the O:C ratio, the MW was aimed to be close to the values determined by Ciarelli et al. (2017) (see Table S4).
- **Atmospherically relevant** compound are preferred. If possible, compounds of atmospheric relevance (e.g., alpha-pinene 90 oxidation products) were chosen (see column “Ref.” and footnotes of Tables S2 and S3).

Table S2. Model compounds for LO-OOA: Volatility bin $\log(C^*)$, substance name and SMILES string, molecular weight in g/mol, O:C elemental ratio, vapor pressure at 25°C estimated with EVAPORATION (Compernelle et al., 2011) in Pa, set vapor pressure calculated from $\log(C^*)$ at 25°C with the MW of the substance and $\gamma = 1$ (artificially set vapor pressure), dry mass fraction in the bin in total (sum of gas and particle phase, w^{tot}) and dry mass fraction in the particle phase at RH = 20 % (w^p), references for atmospheric compounds, and chemical structure.

bin	Substance name, SMILES	MW g mol ⁻¹	O:C	Est. p_{vap} Pa	Set p_{vap} Pa	w^{tot}	w^p	Ref.*	Structure
<0	Carminic acid <chem>CC1=C2C(=CC(=C1C(=O)O)O)C(=O)C3=C(C2=O)C(=C(C(=C3O)O)C4C(C(C(C(O4)CO)O)O)O</chem>	492	0.59	3.30e-33	5.04e-7	0.3	0.172		
	Val9_C10O6H18 <chem>CC(=O)C(O)CC(CC(=O)O)C(C)(C)OO</chem>	234	0.60	1.36e-7	1.06e-6	0.7	0.828	[1]	
0	Pinic acid <chem>CC1(C)C(CC(=O)O)CC1C(=O)O</chem>	186	0.44	4.74e-5	1.33e-5	0.4	0.460	[2, 3,4]	
	Sebacic acid <chem>C(CCCCCCCCC(=O)O)(=O)O</chem>	202	0.40	3.91e-6	1.23e-5	0.3	0.238	[5]	
	Azelaic acid <chem>C(CCCCCCCC(=O)O)(=O)O</chem>	188	0.44	1.20e-5	1.32e-5	0.3	0.302	[4,5]	
1	10-hydroxy pinonic acid <chem>CC1(C)C(CC(=O)O)CC1C(=O)CO</chem>	200	0.40	7.65e-5	1.24e-4	0.4	0.546	[2,3,6,7]	
	10-hydroxydecanoic acid <chem>OCCCCCCCCC(=O)O</chem>	188	0.30	8.42e-5	1.32e-4	0.5	0.389		
	3-hydroxydecanoic acid <chem>OC(CC(=O)O)CCCCCCC</chem>	188	0.30	3.49e-4	1.32e-4	0.1	0.065		
2	Pinolic acid <chem>CC(O)C1CC(CC(=O)O)C1(C)C</chem>	186	0.30	1.38e-3	1.33e-3	0.7	0.817	[7]	
	3-oxodecanoic acid <chem>O=C(CC(=O)O)CCCCCCC</chem>	186	0.30	3.88e-3	1.33e-3	0.3	0.183		
3	Ketopinic acid <chem>CC1(C)C2CCC1(C(=O)O)C(=O)C2</chem>	182	0.30	7.68e-3	1.36e-2	0.4	0.334	[3]	
	Pinonic acid <chem>CC(=O)C1CC(CC(=O)O)C1(C)C</chem>	184	0.30	1.53e-2	1.35e-2	0.5	0.482	[2,3,6,7]	
	1,3-octanediol <chem>C(CC(CCCCC)O)O</chem>	146	0.25	7.02e-2	1.70e-2	0.1	0.185		
4	10-hydroxypinonaldehyde <chem>O=CCC1CC(C(=O)CO)C1(C)C</chem>	184	0.30	1.76e-1	1.35e-1	0.5	0.605	[3]	
	3-pinalic acid <chem>CC1(C)C(CC(=O)O)CC1C(=O)O</chem>	144	0.25	5.66e-1	1.72e-1	0.5	0.395	[6]	

*References: [1] Valorso et al. (2011), Table 4, Substance 9, [2] Eddingsaas et al. (2012), [3] Jaoui and Kamens (2001), [4] Graham et al. (2003), [5] Sempéré and Kawamura (1996), [6] Rindelaub et al. (2016), [7] Fukuyama and Sekimoto (2024)

100 **Table S3.** Model compounds for MO-OOA: Volatility bin ($\log C^*$), substance name and SMILES string, molecular weight in g/mol, O:C elemental ratio, vapor pressure at 25°C estimated with EVAPORATION (Compernelle et al., 2011) in Pa, vapor pressure calculated from $\log(C^*)$ at 25°C with the MW of the substance and $\gamma = 1$ in Pa, dry mass fraction in the bin in total (sum of gas and particle phase, w^{tot}) and dry mass fraction in the particle phase at RH = 20 % (w^p), reference for atmospheric compounds, and chemical structure.

bin	Substance name, SMILES	MW g mol ⁻¹	O:C	Est. p_{vap} Pa	Set p_{vap} Pa	w^{tot}	w^p	Ref.*	Structure
<0	Citric acid <chem>C(CC(O)(C(=O)O)CC(=O)O)(=O)O</chem>	192	1.17	4.98e-5	1.29e-6	0.55	0.685		
	Sucrose <chem>O([CH]1[CH](O)[CH](O)[CH](O)[CH](O1)CO)[C]1(CO)[CH](O)[CH](O)[CH](O1)CO</chem>	342	0.92	2.81e-20	7.24e-7	0.45	0.315	[4,8]	
0	Levogluconan <chem>[CH]12[CH]([CH]([CH]([CH](OC1O2)O)O)O</chem>	162	0.83	2.71e-4	1.53e-5	0.5	0.531	[4,8]	
	Xylose <chem>O=C[CH](O)[CH](O)[CH](O)CO</chem>	150	1.00	9.14e-5	1.65e-5	0.5	0.469	[8]	
1	Adipic acid <chem>C(CCCCC(=O)O)(=O)O</chem>	146	0.67	3.41e-4	1.70e-4	0.3	0.252	[4,5]	
	2-methyltetrol <chem>CC(O)(CO)C(O)CO</chem>	136	0.80	6.18e-4	1.82e-4	0.4	0.390	[9]	
	Glutaric acid <chem>O=C(O)CCCC(=O)O</chem>	132	0.80	1.04e-3	1.88e-4	0.3	0.358	[4,5]	
2	2-methylglutaric acid <chem>CC(C(=O)O)CCC(=O)O</chem>	146	0.67	5.40e-4	1.70e-3	0.25	0.424	[10]	
	2-hydroxyterpenylic acid <chem>CC1(C)OC(=O)CC1C(O)C(=O)O</chem>	188	0.63	3.66e-4	1.32e-3	0.75	0.576	[11]	
3	6-hydroxyhexanoic acid <chem>OCCCCC(=O)O</chem>	132	0.50	7.34e-3	1.88e-2	0.5	0.514		
	Terpenylic acid <chem>CC1(C)OC(=O)CC1CC(=O)O</chem>	172	0.50	4.75e-2	1.44e-2	0.1	0.023	[11]	
	Dihydroxy heptanoic acid <chem>OC(C(=O)O)(CCCC)O</chem>	162	0.57	2.07e-3	1.53e-2	0.4	0.0463		
4	6-oxoheptanoic acid <chem>O=C(CCCCC(=O)O)C</chem>	144	0.43	1.11e-1	1.72e-1	0.1	0.063		
	3-oxohexanoic acid <chem>O=C(CC(=O)O)CCC</chem>	130	0.50	3.38e-1	1.91e-1	0.8	0.896		
	Norpinalic acid <chem>CC1(C)C(C(=O)CC1C(=O)O</chem>	156	0.38	1.43e-1	1.59e-1	0.1	0.041	[6]	

105 *References: [4] Graham et al. (2003), [5] Sempéré and Kawamura (1996), [6] Rindelaub et al. (2016), [8] Simoneit et al. (2004), [9] Claeys et al. (2004), [10] Li et al. (2015), [11] Kahnt et al. (2014)

Table S4. Average O:C elemental ratio and molecular weight of the volatility bins: two SOA sets from Ciarelli et al. (2017) “(Ciarelli)” identified with LO-OOA (set 2) and MO-OOA (3) compared with the values from this study corresponding to the average value in the condensed phase under dry conditions (RH = 20 %).

Volatility bin log(C*)	O:C elemental ratio				MW (g mol ⁻¹)			
	SOA set 2 (Ciarelli)	LO-OOA (this study)	SOA set 3 (Ciarelli)	MO-OOA (this study)	SOA set 2 (Ciarelli)	LO-OOA (this study)	SOA set 3 (Ciarelli)	MO-OOA (this study)
<0	0.5*	0.6	1.05*	1.05	194*	278	149*	239
0	0.43	0.43	0.90	0.9	189	191	144	156
1	0.36	0.36	0.76	0.76	184	193	140	138
2	0.29	0.30	0.63	0.64	179	186	135	175
3	N.A.	0.29	0.52	0.54	N.A.	179	131	146
4	N.A.	0.28	N.A.	0.49	N.A.	162	N.A.	134

* The values in the volatility bin log(C*) < 0 from Ciarelli et al. (2017) correspond to log(C*) = -1, while for LO-OOA and MO-OOA this bin includes species with lower volatility.

The O:C and MW values of each bin reported by Ciarelli et al. (2017) were not only aimed to be met by each individual model compound (values in Tables S2 and S3) but also by the average O:C and MW of each bin (Table S4, columns labelled “this study”). The values in Ciarelli et al. (2017) are based on measurements of aerosol particles under dry conditions and therefore the average bin values in this study also had to be determined for the condensed mass at dry conditions. Note that the average O:C of the total organic mass (gas and particle phase) in a bin can be different to the average O:C of the condensed mass (particle phase) of the same bin. This results if the compounds in the bin have different O:C ratios, which, depending on their functional groups, can result in a different gas-particle partitioning. The same applies for the average MW values. Thus, to determine the average O:C and MW values in the condensed phase, gas-particle partitioning (GPP) calculations under dry conditions (RH = 20 %) had to be performed as described in the next paragraph. By setting the mass fractions of the model compounds within each bin to the values shown in column w^{tot} in Tables S2 and S3, the mass fractions in the condensed phase at RH = 20 % (column w^p in Tables S2 and S3) and the average O:C and MW of each bin shown in Table S4 (“this study”) were optimized to be close to the average values.

GPP calculations were performed for internally mixed organic-inorganic particles with AIOMFAC (Zuend and Seinfeld, 2012; Zuend and Seinfeld, 2013) using the VBS 1 (see Sect. S1) for the organic fractions. The condensed mass concentration at RH = 20 % of LO-OOA (0.48 $\mu\text{g}/\text{m}^3$) and MO-OOA (1.28 $\mu\text{g}/\text{m}^3$) and the inorganic mass concentrations were forced to match the Q-ACSM measurements at Hyytiälä (see Tables 1 in main article and Sectc. S2). This is achieved for a total (gas + particle) mass concentration of LO-OOA and MO-OOA of 7.9 and 11.9 $\mu\text{g}/\text{m}^3$, respectively. Scaling the VBS to these amounts yields the absolute mass concentrations in the organic bins shown in Table S5.

Table S5. Volatility bases set in absolute amounts used for the pre-calculations (based on VBS 1) together with the relative mass fractions replicated from Table S1.

Volatility bin	Relative mass fraction	LO-OOA Total concentration (µg/m³)	MO-OOA Total concentration (µg/m³)	LO-OOA Hvap (kJ/mol)	MO-OOA Hvap (kJ/mol)
<0	0.042	0.3318	0.4998	0	0
0	0.036	0.2844	0.4284	124.73	125.62
1	0.09	0.7110	1.0710	114.09	115.56
2	0.162	1.2798	1.9278	105.03	105.3
3	0.269	2.1251	3.2011	94.14	95.03
4	0.401	3.1679	4.7719	83.66	84.49
Sum	1	7.9	11.9		
Yield in particle phase (at RH = 20 %, T=25 °C):		0.48	1.28		

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Note that three different cases were tested beforehand: i) equilibrium case allowing liquid-liquid phase separation if thermodynamically favoured (“eq-LLPS”), ii) always complete LLPS, i.e. organics in a separate phase from inorganics (“CLLPS”), and iii) allowing only one aerosol phase (“forced 1-ph”). It was found that at 20% RH, the condensed organic mass of the forced 1-ph case is close to the one in the eq-LLPS case (best estimate), whereas assuming CLLPS leads to a lower condensed organic mass. We therefore chose to calculate throughout with forced 1-ph since this is closely representing the eq-LLPS case and is easier to implement in the parcel model, which does not consider LLPS.

The resulting mass concentration per bin for VBS 1 and the partitioning depending on RH is shown in Fig. S2 for LO-OOA and MO-OOA. Based on this set of model compounds and VBS, activity coefficients were determined as a function of RH to set up a look up table for the parcel model calculations as described in the following section. Note that we performed these GPP calculations just for VBS 1 but apply the resulting look-up table to both VBS for consistency.

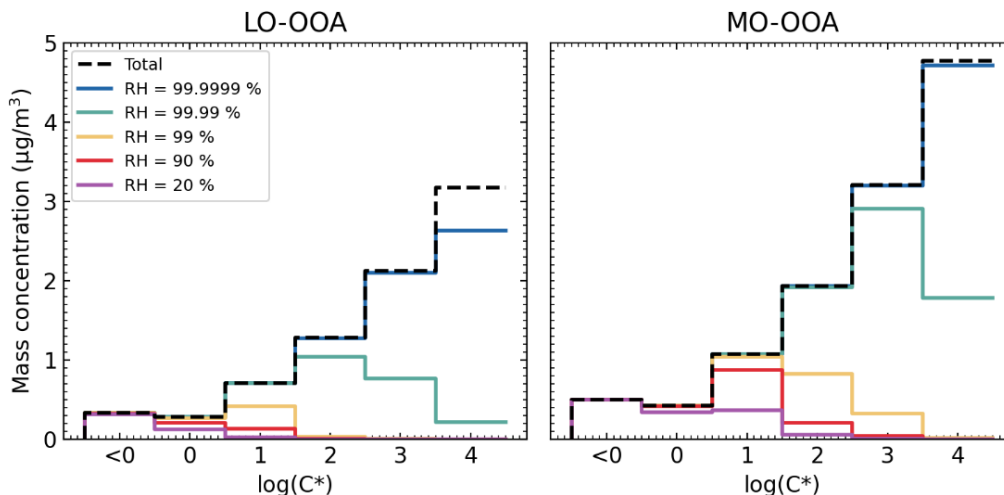


Figure S2. Gas-particle partitioning into mixed organic-inorganic aerosol particles of LO-OOA (left panel) and MO-OOA (right panel) per volatility bin from the pre-calculation (using AIOMFAC). The black dashed lines correspond to the total mass in each bin; solid colored lines show condensed-phase concentrations with increasing humidity.

S3. Inorganic ions balance for AIOMFAC model

A simplified ion pairing method was applied to ensure ion balance of the input data for the group contribution model AIOMFAC (Zuend et al., 2008; Zuend et al., 2011). Briefly, we calculate the molar concentrations (mol m^{-3}) of ions measured by Q-ACSM (NH_4^+ , SO_4^{2-} , NO_3^- , Cl^-) and assume that the total of cations and anions fulfil ion balance. The NH_4^+ ions are first assigned to Cl^- and then to NO_3^- , providing ammonium chloride (NH_4Cl) and ammonium nitrate (NH_4NO_3) because these are semi-volatile and require neutralization to partition into the condensed phase. The excess NH_4^+ ions are assigned to SO_4^{2-} to provide ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$), ammonium bisulfate (NH_4HSO_4), and sulfuric acid (H_2SO_4), depending on the ratio of excess NH_4^+ and SO_4^{2-} as described below.

$$(\text{NH}_4)_2\text{SO}_4, \quad \text{if } \frac{\text{excess } \text{NH}_4^+}{\text{SO}_4^{2-}} \geq 2$$

$$(\text{NH}_4)_2\text{SO}_4 + \text{NH}_4\text{HSO}_4, \text{ if } 1 < \frac{\text{excess } \text{NH}_4^+}{\text{SO}_4^{2-}} < 2$$

$$\text{NH}_4\text{HSO}_4, \text{ if } \frac{\text{excess } \text{NH}_4^+}{\text{SO}_4^{2-}} = 1$$

$$\text{NH}_4\text{HSO}_4 + \text{H}_2\text{SO}_4, \text{ if } 0.5 < \frac{\text{excess } \text{NH}_4^+}{\text{SO}_4^{2-}} < 1$$

$$\text{H}_2\text{SO}_4, \quad \text{if } \frac{\text{excess } \text{NH}_4^+}{\text{SO}_4^{2-}} \leq 0.5$$

The thus calculated concentrations of the inorganic salts for model input are given in Table S6. In this study, we have $1 < \frac{\text{excess } \text{NH}_4^+}{\text{SO}_4^{2-}} < 2$, so NH_4HSO_4 and $(\text{NH}_4)_2\text{SO}_4$ co-existed in the system.

165 Table S6: Input of inorganic compounds for AIOMFAC and the parcel model:

	Molar concentration (mol m ⁻³)	MW (g mol ⁻¹)	Mass concentration (µg m ⁻³)
NH ₄ Cl	2.82E-10	53.491	1.51E-02
NH ₄ NO ₃	3.06E-09	80.043	2.45E-01
(NH ₄) ₂ SO ₄	2.94E-09	132.14	3.88E-01
NH ₄ HSO ₄	1.33E-09	115.11	1.53E-01

S4. Calculation of activity coefficients for organic fractions and water

To include solution non-ideality in the parcel model, activity coefficients of water and the organic volatility bins were pre-computed from the gas-particle partitioning calculations with AIOMFAC (Zuend and Seinfeld, 2012; Zuend and Seinfeld, 2013) of the model system (see Sect. S2) as a function of RH (under the setting “forced 1-ph”, see above). An activity coefficient for each volatility bin was then calculated as a weighted average of the activity coefficients from the individual model compounds as:

$$\gamma_{bin}(RH) = \sum_i w_i \gamma_i(RH)$$

where w_i and γ_i are the mass fraction and the activity coefficient of compound i in the bin, respectively. The resulting activity coefficients for each organic bin and for water as a function of RH are shown in Table S7. This look-up table was used in the parcel model for simulations with VBS 1 and VBS 2.

Table S7: Look-up table for activity coefficients: Activity coefficient of water (gamma_w) and of the log(C*) bins of LO-OOA and MO-OOA as a function of water activity (a_w) and water mole fraction (x_w).

Water			LO-OOA							MO-OOA					
a_w	x_w	gamma_w	bin <0	bin 0	bin 1	bin 2	bin 3	bin 4	bin <0	bin 0	bin 1	bin 2	bin 3	bin 4	
0.999999	0.9999991	0.9999999	19.693	70.980	207.182	414.034	294.459	475.079	0.064	1.136	1.815	12.909	20.119	23.646	
0.999979	0.9999811	0.9999979	19.673	70.911	206.943	413.598	294.180	474.629	0.064	1.135	1.814	12.900	20.103	23.631	
0.999959	0.9999634	0.9999956	19.654	70.846	206.720	413.194	293.922	474.209	0.064	1.134	1.814	12.891	20.088	23.616	
0.999939	0.9999459	0.9999931	19.637	70.784	206.508	412.809	293.677	473.809	0.064	1.133	1.813	12.882	20.073	23.602	
0.999919	0.9999285	0.9999905	19.620	70.725	206.303	412.439	293.443	473.423	0.063	1.133	1.812	12.874	20.059	23.588	
0.999899	0.9999113	0.9999877	19.604	70.667	206.104	412.080	293.215	473.049	0.063	1.132	1.811	12.866	20.046	23.575	
0.999799	0.9998262	0.9999728	19.527	70.396	205.171	410.401	292.155	471.291	0.063	1.129	1.807	12.828	19.982	23.514	
0.999699	0.9997419	0.9999571	19.456	70.143	204.299	408.840	291.171	469.651	0.063	1.126	1.804	12.793	19.922	23.456	
0.999599	0.9996581	0.9999408	19.388	69.901	203.468	407.355	290.237	468.087	0.063	1.124	1.800	12.760	19.864	23.401	
0.999499	0.9995747	0.9999243	19.324	69.668	202.668	405.927	289.341	466.580	0.063	1.121	1.797	12.727	19.809	23.348	
0.999399	0.9994915	0.9999075	19.261	69.441	201.891	404.544	288.474	465.119	0.063	1.118	1.794	12.696	19.755	23.297	
0.999299	0.9994085	0.9998905	19.200	69.221	201.136	403.199	287.632	463.695	0.063	1.116	1.790	12.665	19.703	23.247	
0.999199	0.9993256	0.9998733	19.140	69.006	200.398	401.887	286.811	462.305	0.063	1.114	1.787	12.636	19.651	23.198	
0.999099	0.9992429	0.9998559	19.082	68.795	199.675	400.604	286.009	460.944	0.063	1.111	1.784	12.606	19.601	23.149	
0.998999	0.9991604	0.9998385	19.025	68.588	198.967	399.347	285.224	459.608	0.063	1.109	1.781	12.577	19.551	23.102	
0.998799	0.9989955	0.9998033	18.913	68.184	197.587	396.901	283.698	457.005	0.062	1.104	1.775	12.521	19.454	23.010	
0.998599	0.9988311	0.9997677	18.806	67.793	196.250	394.534	282.225	454.483	0.062	1.100	1.770	12.466	19.360	22.921	
0.998399	0.9986669	0.9997318	18.701	67.412	194.951	392.238	280.797	452.030	0.062	1.096	1.764	12.413	19.269	22.834	
0.998199	0.9985029	0.9996956	18.600	67.041	193.687	390.003	279.409	449.640	0.062	1.091	1.758	12.361	19.179	22.749	
0.997999	0.9983391	0.9996593	18.501	66.679	192.453	387.825	278.058	447.306	0.062	1.087	1.753	12.310	19.091	22.666	
0.997599	0.9980121	0.9995861	18.309	65.978	190.067	383.618	275.453	442.789	0.061	1.079	1.743	12.212	18.922	22.505	
0.997199	0.9976855	0.9995124	18.126	65.306	187.779	379.588	272.962	438.449	0.061	1.072	1.732	12.117	18.758	22.350	
0.996799	0.9973592	0.9994383	17.950	64.657	185.576	375.712	270.571	434.264	0.061	1.065	1.723	12.025	18.600	22.200	
0.996399	0.9970331	0.9993640	17.780	64.031	183.450	371.972	268.268	430.216	0.060	1.058	1.713	11.936	18.446	22.056	
0.995999	0.9967070	0.9992896	17.616	63.424	181.392	368.354	266.042	426.291	0.060	1.051	1.704	11.850	18.297	21.915	
0.995599	0.9963810	0.9992151	17.457	62.835	179.396	364.847	263.887	422.478	0.060	1.044	1.695	11.767	18.152	21.778	
0.995199	0.9960550	0.9991407	17.303	62.262	177.457	361.439	261.796	418.766	0.060	1.038	1.686	11.685	18.011	21.645	
0.994799	0.9957288	0.9990662	17.153	61.705	175.571	358.123	259.762	415.147	0.059	1.032	1.677	11.605	17.873	21.515	
0.992799	0.9940951	0.9986962	16.456	59.106	166.805	342.694	250.313	398.219	0.058	1.004	1.637	11.232	17.227	20.906	
0.990799	0.9924546	0.9983319	15.829	56.759	158.928	328.771	241.790	382.832	0.057	0.979	1.599	10.893	16.637	20.349	
0.99	0.99180	0.99819	15.595	55.878	155.982	323.544	238.587	377.030	0.057	0.970	1.585	10.765	16.415	20.138	
0.985	0.98765	0.99732	14.281	50.933	139.576	294.140	220.500	344.168	0.054	0.921	1.505	10.039	15.155	18.937	
0.98	0.98346	0.99649	13.168	46.737	125.872	269.083	204.925	315.893	0.053	0.882	1.436	9.412	14.071	17.887	
0.97	0.97496	0.99491	11.359	39.939	104.196	228.202	179.023	269.294	0.050	0.824	1.322	8.371	12.283	16.115	
0.96	0.96641	0.99337	9.956	34.696	88.021	196.446	158.350	232.711	0.048	0.784	1.231	7.540	10.876	14.673	
0.95	0.95788	0.99177	8.847	30.582	75.723	171.427	141.633	203.644	0.047	0.757	1.158	6.867	9.751	13.484	
0.94	0.94943	0.99007	7.961	27.310	66.223	151.489	127.988	180.313	0.046	0.737	1.098	6.315	8.841	12.496	
0.93	0.94109	0.98822	7.242	24.674	58.768	135.417	116.748	161.388	0.046	0.724	1.049	5.858	8.096	11.667	
0.92	0.93287	0.98620	6.653	22.524	52.830	122.310	107.402	145.869	0.046	0.715	1.009	5.475	7.481	10.965	
0.91	0.92480	0.98400	6.165	20.749	48.032	111.501	99.559	133.005	0.046	0.710	0.975	5.151	6.966	10.367	
0.9	0.91686	0.98161	5.756	19.267	44.102	102.489	92.918	122.229	0.046	0.707	0.946	4.875	6.532	9.852	
0.89	0.90905	0.97904	5.410	18.017	40.846	94.899	87.248	113.114	0.046	0.706	0.922	4.637	6.161	9.406	
0.88	0.90137	0.97629	5.115	16.953	38.117	88.445	82.367	105.332	0.046	0.707	0.902	4.430	5.843	9.016	
0.87	0.89381	0.97336	4.860	16.039	35.807	82.909	78.135	98.631	0.047	0.709	0.885	4.250	5.566	8.673	
0.86	0.88636	0.97026	4.640	15.247	33.833	78.121	74.441	92.815	0.047	0.713	0.870	4.091	5.325	8.370	
0.85	0.87900	0.96700	4.447	14.556	32.133	73.950	71.195	87.730	0.048	0.717	0.857	3.950	5.113	8.100	
0.84	0.87174	0.96359	4.278	13.949	30.658	70.291	68.326	83.253	0.048	0.722	0.846	3.824	4.925	7.858	
0.83	0.86456	0.96003	4.128	13.412	29.370	67.060	65.778	79.288	0.049	0.727	0.836	3.711	4.758	7.641	
0.82	0.85745	0.95632	3.995	12.935	28.236	64.191	63.501	75.756	0.050	0.733	0.828	3.609	4.608	7.445	
0.81	0.85041	0.95248	3.876	12.509	27.234	61.628	61.459	72.592	0.051	0.739	0.821	3.517	4.473	7.266	
0.8	0.84343	0.94851	3.769	12.125	26.344	59.328	59.619	69.743	0.051	0.746	0.814	3.433	4.352	7.103	
0.79	0.83650	0.94441	3.673	11.779	25.548	57.254	57.954	67.167	0.052	0.753	0.809	3.357	4.241	6.954	
0.78	0.82961	0.94020	3.585	11.465	24.834	55.375	56.441	64.827	0.053	0.760	0.804	3.286	4.140	6.817	
0.77	0.82276	0.93587	3.505	11.180	24.190	53.665	55.062	62.692	0.054	0.767	0.800	3.221	4.047	6.691	
0.76	0.81595	0.93143	3.432	10.918	23.609	52.104	53.800	60.738	0.055	0.775	0.796	3.161	3.962	6.573	
0.75	0.80916	0.92689	3.366	10.678	23.080	50.673	52.642	58.942	0.055	0.782	0.793	3.104	3.883	6.464	

Table S7 (continued): Look-up table for activity coefficients: Activity coefficient of water (gamma_w) and of the log(C*) bins of LO-OOA and MO-OOA as a function of water activity (a_w) and water mole fraction (x_w).

Water			LO-OOA						MO-OOA					
a_w	x_w	gamma_w	bin <0	bin 0	bin 1	bin 2	bin 3	bin 4	bin <0	bin 0	bin 1	bin 2	bin 3	bin 4
0.74	0.80239	0.92225	3.304	10.457	22.599	49.356	51.576	57.286	0.056	0.790	0.790	3.052	3.810	6.363
0.73	0.79563	0.91751	3.247	10.253	22.159	48.141	50.592	55.754	0.057	0.798	0.788	3.003	3.743	6.268
0.72	0.78888	0.91268	3.195	10.064	21.756	47.016	49.681	54.333	0.058	0.806	0.785	2.957	3.680	6.179
0.71	0.78214	0.90776	3.146	9.888	21.385	45.970	48.834	53.011	0.059	0.814	0.784	2.914	3.620	6.095
0.7	0.77540	0.90275	3.100	9.723	21.043	44.997	48.046	51.777	0.060	0.821	0.782	2.872	3.565	6.016
0.69	0.76866	0.89766	3.057	9.569	20.726	44.087	47.311	50.622	0.061	0.829	0.780	2.833	3.513	5.941
0.68	0.76191	0.89249	3.017	9.424	20.433	43.235	46.623	49.539	0.061	0.837	0.779	2.796	3.463	5.870
0.67	0.75515	0.88725	2.980	9.288	20.160	42.434	45.977	48.520	0.062	0.845	0.778	2.761	3.417	5.802
0.66	0.74837	0.88192	2.944	9.160	19.906	41.681	45.369	47.560	0.063	0.853	0.776	2.727	3.372	5.738
0.65	0.74156	0.87653	2.910	9.038	19.668	40.969	44.797	46.654	0.064	0.861	0.775	2.694	3.330	5.676
0.64	0.73474	0.87106	2.879	8.922	19.446	40.296	44.256	45.795	0.065	0.868	0.774	2.663	3.290	5.617
0.63	0.72788	0.86552	2.848	8.812	19.237	39.657	43.744	44.981	0.066	0.876	0.774	2.633	3.252	5.560
0.62	0.72100	0.85992	2.819	8.707	19.040	39.050	43.257	44.206	0.067	0.884	0.773	2.603	3.215	5.505
0.61	0.71408	0.85425	2.792	8.606	18.854	38.471	42.794	43.469	0.068	0.891	0.772	2.575	3.179	5.452
0.6	0.70712	0.84852	2.766	8.510	18.678	37.919	42.353	42.765	0.069	0.899	0.771	2.547	3.145	5.401
0.59	0.70011	0.84272	2.741	8.417	18.511	37.390	41.930	42.091	0.069	0.906	0.770	2.520	3.112	5.351
0.58	0.69306	0.83687	2.717	8.328	18.352	36.883	41.526	41.446	0.070	0.913	0.769	2.494	3.080	5.303
0.57	0.68596	0.83095	2.694	8.241	18.201	36.396	41.137	40.827	0.071	0.920	0.769	2.468	3.049	5.256
0.56	0.67880	0.82498	2.671	8.157	18.056	35.926	40.763	40.232	0.072	0.928	0.768	2.443	3.019	5.211
0.55	0.67159	0.81895	2.650	8.076	17.917	35.473	40.403	39.659	0.073	0.934	0.767	2.419	2.990	5.166
0.54	0.66431	0.81287	2.630	7.997	17.784	35.035	40.054	39.106	0.074	0.941	0.766	2.394	2.961	5.122
0.53	0.65697	0.80673	2.610	7.920	17.655	34.611	39.716	38.572	0.075	0.948	0.765	2.370	2.933	5.079
0.52	0.64956	0.80054	2.591	7.844	17.531	34.200	39.387	38.056	0.075	0.955	0.764	2.347	2.906	5.037
0.51	0.64208	0.79430	2.573	7.770	17.410	33.799	39.067	37.555	0.076	0.961	0.763	2.324	2.879	4.995
0.5	0.63451	0.78800	2.555	7.698	17.294	33.410	38.756	37.069	0.077	0.967	0.762	2.301	2.853	4.954
0.49	0.62687	0.78166	2.538	7.627	17.180	33.029	38.451	36.597	0.078	0.974	0.761	2.278	2.827	4.913
0.48	0.61914	0.77526	2.522	7.556	17.068	32.658	38.152	36.137	0.079	0.980	0.760	2.255	2.801	4.873
0.47	0.61133	0.76882	2.507	7.487	16.959	32.294	37.859	35.689	0.080	0.985	0.758	2.233	2.776	4.834
0.46	0.60342	0.76233	2.493	7.419	16.853	31.938	37.571	35.251	0.080	0.991	0.757	2.210	2.751	4.794
0.45	0.59541	0.75579	2.479	7.351	16.747	31.587	37.286	34.824	0.081	0.997	0.756	2.188	2.727	4.755
0.44	0.58729	0.74920	2.466	7.284	16.644	31.243	37.006	34.406	0.082	1.002	0.754	2.166	2.702	4.716
0.43	0.57907	0.74256	2.454	7.218	16.541	30.904	36.728	33.996	0.083	1.007	0.753	2.144	2.678	4.678
0.42	0.57074	0.73588	2.443	7.152	16.440	30.570	36.453	33.595	0.084	1.013	0.751	2.122	2.654	4.639
0.41	0.56229	0.72916	2.433	7.086	16.339	30.240	36.181	33.200	0.084	1.018	0.749	2.100	2.630	4.601
0.4	0.55372	0.72239	2.424	7.021	16.239	29.913	35.909	32.813	0.085	1.022	0.747	2.078	2.606	4.563
0.39	0.54502	0.71557	2.416	6.955	16.139	29.590	35.639	32.431	0.086	1.027	0.746	2.056	2.583	4.524
0.38	0.53618	0.70871	2.409	6.890	16.039	29.270	35.370	32.056	0.087	1.031	0.743	2.034	2.559	4.486
0.37	0.52721	0.70181	2.404	6.825	15.939	28.953	35.101	31.685	0.087	1.036	0.741	2.013	2.535	4.448
0.36	0.51809	0.69486	2.400	6.760	15.839	28.637	34.833	31.320	0.088	1.040	0.739	1.991	2.512	4.409
0.35	0.50881	0.68787	2.398	6.694	15.739	28.324	34.564	30.959	0.089	1.044	0.737	1.969	2.488	4.371
0.34	0.49938	0.68084	2.398	6.629	15.638	28.012	34.294	30.602	0.090	1.047	0.734	1.947	2.465	4.332
0.33	0.48979	0.67376	2.399	6.563	15.537	27.702	34.024	30.250	0.090	1.051	0.732	1.925	2.441	4.294
0.32	0.48001	0.66665	2.403	6.497	15.435	27.392	33.753	29.900	0.091	1.054	0.729	1.903	2.418	4.255
0.31	0.47006	0.65948	2.409	6.431	15.332	27.084	33.480	29.554	0.092	1.057	0.727	1.880	2.394	4.216
0.3	0.45992	0.65228	2.417	6.365	15.228	26.775	33.206	29.212	0.092	1.060	0.724	1.858	2.370	4.177
0.29	0.44959	0.64504	2.428	6.298	15.123	26.468	32.930	28.871	0.093	1.063	0.721	1.836	2.347	4.137
0.28	0.43905	0.63775	2.442	6.231	15.016	26.160	32.652	28.534	0.094	1.065	0.718	1.814	2.323	4.098
0.27	0.42829	0.63042	2.460	6.164	14.909	25.853	32.372	28.199	0.094	1.068	0.714	1.791	2.299	4.058
0.26	0.41731	0.62305	2.481	6.096	14.800	25.545	32.089	27.866	0.095	1.070	0.711	1.769	2.275	4.018
0.25	0.40609	0.61563	2.506	6.027	14.689	25.237	31.804	27.535	0.095	1.071	0.708	1.746	2.251	3.977
0.24	0.39462	0.60818	2.536	5.958	14.577	24.929	31.517	27.206	0.096	1.073	0.704	1.723	2.227	3.936
0.23	0.38290	0.60068	2.570	5.889	14.464	24.620	31.226	26.879	0.097	1.075	0.700	1.700	2.202	3.895
0.22	0.37091	0.59314	2.610	5.819	14.349	24.311	30.933	26.554	0.097	1.076	0.697	1.677	2.178	3.854
0.21	0.35863	0.58556	2.656	5.748	14.232	24.001	30.637	26.231	0.098	1.077	0.693	1.654	2.153	3.813
0.2	0.34606	0.57794	2.708	5.677	14.113	23.690	30.338	25.909	0.098	1.078	0.689	1.631	2.129	3.771

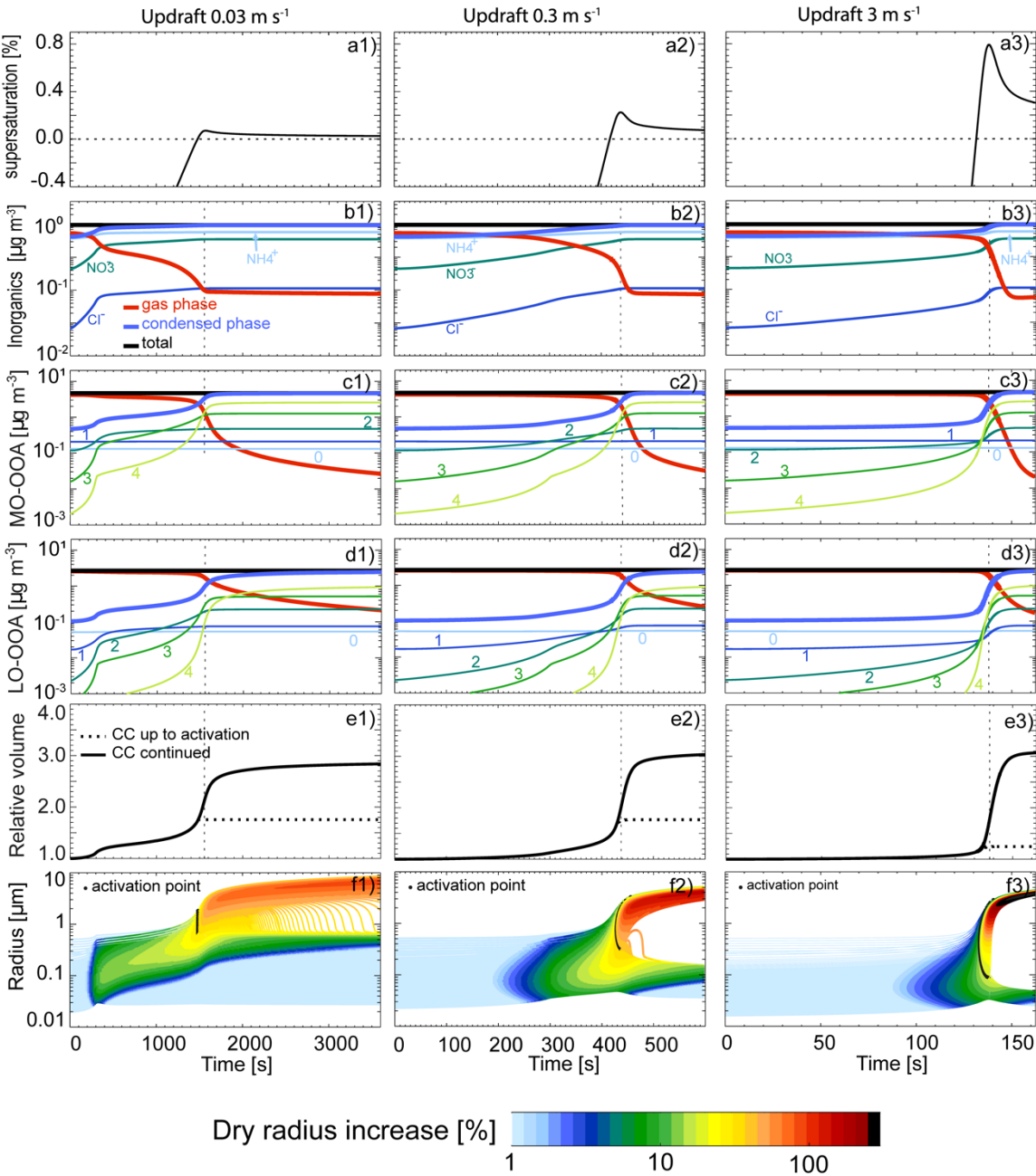


Figure S3. Time series of a) supersaturation, b-d) inorganic and organic species, e) droplet dry volume relative to the initial state of 80% RH, f) wet radius, at updraft of 0.03 (left), 0.3 (middle), and 3 m s⁻¹ (right panel) for VBS 1.

Figure S4

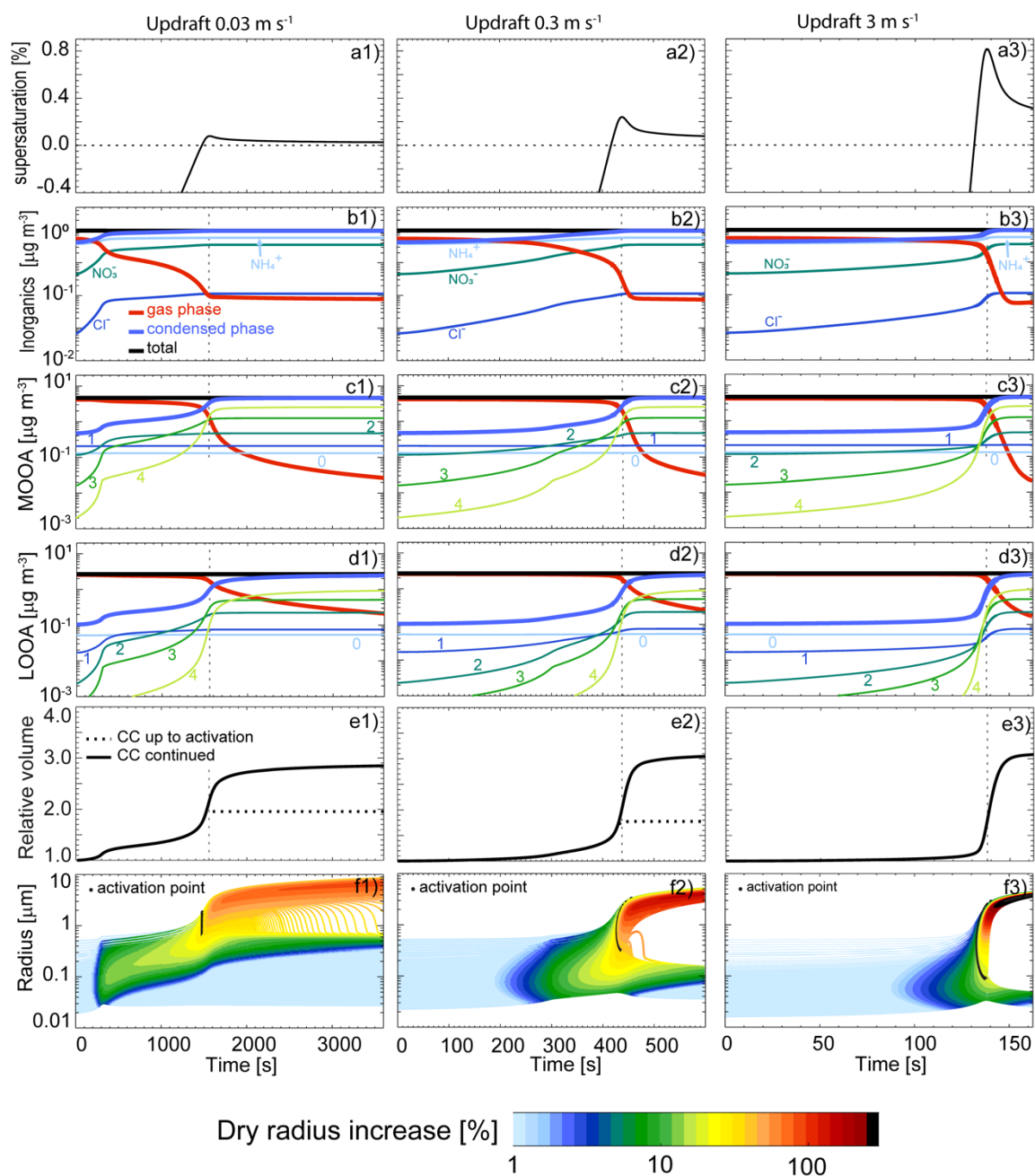


Figure S4. Time series of a) supersaturation, b-d) inorganic and organic species, e) droplet dry volume relative to the initial state 80% RH, f) wet radius, at updraft of 0.03 (left), 0.3 (middle), and 3 m s⁻¹ (right panel) for VBS 2.

Table S8

195 Table S8. Condensed-phase composition of soluble species for parcel model initialisation.

	Molality (mol per kg H ₂ O)		molar mass (g mol ⁻¹)	molar volume (cm ³ mol ⁻¹)
	VBS 1	VBS 2		
SO ₄ ²⁻	3.79E+00	1.74E+00	9.61E+01	3.81E+01
HSO ₄ ⁻	1.93E+01	2.30E+01	9.71E+01	4.00E+01
NH ₄ ⁺	2.25E+01	2.17E+01	1.80E+01	1.48E+01
NH ₃	2.90E-09	7.34E-10	1.70E+01	1.38E+01
NO ₃ ⁻	2.14E-01	2.03E-01	6.20E+01	2.84E+01
Na ⁺	4.62E+00	4.95E+00	2.30E+01	1.12E+01
Cl ⁻	6.32E-04	4.89E-04	3.55E+01	1.72E+01
H ⁺	3.58E-03	5.94E-03	1.01E+00	9.98E-01
OC bin < 0	1.19E+01	2.68E+01	2.52E+02	1.55E+02
MO-OOA bin 0	9.04E+00	7.17E+00	1.56E+02	1.03E+02
MO-OOA bin 1	2.38E+01	1.11E+01	1.38E+02	9.93E+01
MO-OOA bin 2	3.13E+00	1.59E+00	1.75E+02	1.36E+02
MO-OOA bin 3	5.24E-02	4.23E-02	1.46E+02	1.18E+02
MO-OOA bin 4	6.60E-03	7.11E-03	1.34E+02	1.12E+02
LO-OOA bin 0	5.30E+00	2.52E+00	1.91E+02	1.45E+02
LO-OOA bin 1	8.81E-01	2.28E-01	1.93E+02	1.58E+02
LO-OOA bin 2	1.27E-02	3.94E-03	1.86E+02	1.62E+02
LO-OOA bin 3	2.95E-03	1.42E-03	1.79E+02	1.63E+02
LO-OOA bin 4	2.76E-04	1.76E-04	1.62E+02	1.54E+02
Black Carbon	2.31E+02	2.48E+02	1.20E+01	5.31E+00

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