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Can Semi-Volatile Organic Aerosols Lead to Less Cloud Particles?

Chloe Y. Gao[1,2], Susanne E. Bauer[2], and Kostas Tsigaridis[3,2]

[1]Department of Earth and Environmental Sciences, Columbia University, New York, NY, 10027, USA

[2]NASA Goddard Institute for Space Studies, New York, NY, 10025, USA

[3]Center for Climate Systems Research, Columbia University, New York, NY, 10025, USA

Correspondence to: Kostas Tsigaridis (kostas.tsigaridis@columbia.edu)

Abstract. The impact of condensing organic aerosols on activated cloud number concentration is examined in a new aerosol microphysics box model, MATRIX-VBS. The model includes the volatility-basis set (VBS) framework coupled with the aerosol microphysical scheme MATRIX (Multiconfiguration Aerosol TRacker of mIXing state) that resolves aerosol mass and number concentrations and aerosol mixing state. By including the condensation of organic aerosols, the new model produces less activated particles compared to the original model, which treats organic aerosols as non-volatile. Parameters such as aerosol chemical composition, mass and number concentrations, and particle sizes which affect activated cloud number concentration are thoroughly tested via a suite of Monte-Carlo simulations. Results show that by considering semi-volatile organics in MATRIX-VBS, there is lower activated particle number concentration, except in cases with low cloud updrafts, in clean environment at above freezing temperatures, and in polluted environments at high temperature (310K) and extremely low humidity conditions.

1 Introduction

Atmospheric aerosols influence climate mainly via two pathways: aerosol-radiation interactions (the aerosol direct effect; Charlson et al., 1992) which affect the Earth's radiative energy balance by absorbing and scattering terrestrial and solar radiation, and aerosol-cloud interactions (the aerosol indirect effect; Twomey, 1974; Albrecht, 1989) which affect cloud microphysics by activating and serving as seeds for cloud formation (Myhre et al., 2013; Seinfeld and Pandis, 2016). Aerosol activation as cloud condensation nuclei (CNN) is critical to the evolution and microphysics of clouds (Reutter et al., 2009). However, the relationship between aerosol mixing state and cloud microphysical properties remain a large uncertainty in aerosol-

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cloud interactions (Ghan et al., 1998; McFiggans et al., 2006; Ervens et al., 2007; Gibson et al., 2007; Medina et al., 2007; Cubison et al., 2008; Anttila, 2010).

Climate models calculate cloud droplet number concentration (CDNC) using aerosol activation schemes, whose main governing parameters include aerosol number, size, hygroscopicity, updraft velocity, as well as critical supersaturation. Physically-based aerosol activation schemes (e.g. Abdul-Razzak and Ghan, 2000; Fountoukis and Nenes, 2005; Ming et al., 2006; Shipway and Abel, 2010) are commonly used in global climate models for fast diagnostics of nucleation and to estimate the aerosol indirect effect in long-term climate simulations (Ghan, 2011). Several studies examined the relationship between the fore-mentioned parameters and how they play together to activate particles. Ghan et al. (1998) examined sea salt's influence on sulfate particle activation and introduced the competition effect. Since all CCN have to compete for available water vapor in order to activate, the competition limits the maximum supersaturation in in-cloud updrafts (Storelvmo et al., 2006). Ghan et al. (1998) concluded that activated number concentration increases with increasing sea salt when sulfate is low and updraft is strong, and it decreases when sulfate is high and updraft is weak, because maximum supersaturation is reduced. Another study (Reutter et al. 2009) explored how much CDNC depend on updraft velocity, size distribution and hygroscopicity. They found that size distribution played a greater role than particle hygroscopicity on CDNC and discovered different CCN activation and cloud droplet formation regimes, which are determined by aerosol number concentration and updraft velocity.

Semi-volatile organic aerosols contribute significantly to the growth of particles to CCN sizes (Yu, 2011). More notably, as aerosol size increases, the range of organic volatilities involved in aerosol growth increases (Pierce et al., 2011; Yu, 2011). The inclusion of semi-volatile organics in models modifies CCN formation rates (Petters et al., 2006, Riipenen et al., 2011; Scott et al., 2015) as well as hygroscopicity (Petters and Kreidenweis, 2007), in addition to bulk aerosol mass, size distribution and composition. By adding semi-volatile organic partitioning to our existing microphysics model MATRIX (Multiconfiguration Aerosol TRacker of mIXing state; Bauer et al., 2008), which resolves aerosol mixing state, we were able to examine how they change bulk aerosol mass, size distribution and composition. However, the effects of semi-volatile organic partitioning combined with aerosol mixing state on particle activation remain unexplored.

In our previous work, we demonstrated that including semi-volatile organics would lead to higher aerosol number concentration and smaller particles (Gao et al., 2017). As was the case for

the original aerosol microphysics model MATRIX, our further-developed box model MATRIX-VBS (Gao et al., 2017) follows the same multi-modal aerosol activation approach by Abdul-Razzak and Ghan (2000). The activation parameterization accounts for aerosol size distribution, composition, mixing state, and in-cloud updraft velocity. Curious about the change in activation with the newly-present semi-volatile organics and the governing parameters influencing it, we investigated the difference in activated number concentration in two box model set ups: MATRIX (Bauer et al., 2008) and MATRIX-VBS (Gao et al., 2017).

2 Methods

2.1 Model Description

MATRIX-VBS (Gao et al., 2017) is an aerosol microphysics model that includes organic aerosol volatility in its calculations. It was developed by implementing VBS (volatility-basis set; Donahue et al., 2006) in the aerosol microphysics model MATRIX (Bauer et al., 2008), which is a box model that is also used in the NASA GISS ModelE Earth System Model (Bauer et al., 2008, 2012; Schmidt et al., 2014). Since the publication of Gao et al., 2017, which included organic condensation on fine mode aerosols, we further developed the model which now allows semi-volatile organics in the system to condense on coarse mode dust and sea salt as well. We have also included nitrate radicals as an oxidant for organics in addition to the hydroxyl radical that was used in the original VBS scheme, even though it is a very minor oxidation pathway in the model (rate constant for the oxidation by $NO_3^{\bullet}$ is $1*10^{-13}$ cm^3 $molecules^{-1}$ s^{-1}; Atkinson, 1997). As previously stated, we use Abdul-Razzak and Ghan (2000) activation parameterization, which calculates the activated particle number concentration depending on chemically-resolved number concentrations using Köhler Theory. The hygroscopicity parameters κ for each aerosol species presented in Table 1 were calculated from their solubility fraction. For organics, we assumed a linear increase of solubility with decreasing volatility (Jimenez et al., 2009).

2.2 Simulations

A Monte-Carlo analysis with a range of chemical and meteorological conditions (Table 2) was performed, to pinpoint which processes affect organics and the mixed aerosol population in general the most. Since global models need to resolve a wide range of conditions, from very clean to very polluted and for a wealth of meteorological conditions, we simulated 630 possible

atmospheric scenarios on Earth across the whole parameter space, e.g. temperature, relative humidity, latitude, emissions levels and updraft velocity, for 120 hours (5 days) simulations with no deposition and dilution. Three types of environmental conditions were simulated: clean, moderate and polluted, as defined by different levels of emissions which were determined using a probability distribution of the gridded emission fields in GISS ModelE for January present-day conditions. During this development phase, biogenic secondary organic aerosols from terpenes oxidation in MATRIX-VBS are treated as nonvolatile, while only the anthropogenic aerosols are treated as semi-volatile.

3 Results and discussion

We found that activated number concentration is lower for most cases in the MATRIX-VBS model, which considers semi-volatile organic aerosols, as compared to the MATRIX model. However, under low updrafts, in clean environment at above freezing temperatures, and in polluted environments at high temperature (310K) and extremely low humidity conditions (0% RH) during aerosol formation, activated number concentration is higher in MATRIX-VBS than in MATRIX.

As an example, the activated number concentration for a case with temperature at 290˚K, relative humidity at 40%, medium emission levels and an updraft of 0.5 m/s at 30°N latitude is shown in Figure 1 for the two models. Mixing states of aerosols in MATRIX and MATRIX-VBS are represented as aerosol populations, which all contain SO_4, NO_3, NH_4 and H_2O, in addition to the species that define the populations (Bauer et al., 2008, 2013). The four most dominant aerosol populations for the activated number concentration in MATRIX are ACC (SO_4, NO_3, NH_4), OCS (organics, SO_4, NO_3, NH_4), BOC (black carbon, organic carbon, SO_4, NO_3, NH_4) and BCS (black carbon, SO_4, NO_3, NH_4). Only two dominant populations are calculated in MATRIX-VBS, OCS and BOC, as in Gao et al., 2017, since OCC evaporates and re-condenses on all particles, based on their calculated surface area and mass concentration. Since OCS and BOC have the largest surface area, they are calculated to have the strongest growth via organics condensation. Additionally, the competition between sulfate, organics and black carbon, determines the loss of ACC and the formation of BCS: OCC coagulates with ACC to form OCS, and this coagulation increases in MATRIX-VBS due to smaller OCC particles; therefore, there are less ACC particles left to coagulate with black carbon to form BCS. At the end of the 5-day simulation (Figure 1), MATRIX-

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VBS has approximately a total of 30 activated particles/cm^3, whereas MATRIX has approximately 60 activated particles/cm^3 under the same conditions.

Figure 2 shows a more comprehensive look across all temperature and relative humidity scenarios studied. The results show that for most scenarios, MATRIX-VBS has lower (red circles) activated number concentration compared to MATRIX. However, some rare cases show the opposite behavior. These are for above freezing temperatures in the low emission level under low updraft (top left) scenarios, high temperature (310K) and extremely low humidity (0% RH) in the medium emission level under low updraft (middle left) scenarios, as well as the high emission level under low (bottom left) and medium (bottom middle) updraft scenarios. Across all scenarios, the changes in activated number concentration between MATRIX-VBS and MATRIX range from a -56% to +31% (Table 3). The range of the difference becomes more significant as emission levels increase, yet less significant as updraft velocity increases. Within most emission level-updraft velocity scenarios, as temperature increases, the fractional change in activated number concentration between the two models decreases. Also within most emission level-updraft velocity scenarios (Figure 3, Table 4), as temperature increases, there are less activated particles in MATRIX. We also observed the same behavior in MATRIX-VBS, higher temperature, less activated particles.

In order to understand the cause of the difference in activation, we traced back to the key difference between the two models: partitioning of organics. The inclusion of organics partitioning leads to changes in aerosol mixing state and size distribution, as discussed in Gao et al. (2017). Therefore, the change in activated number concentration could only be caused by changes in mass concentration, number concentration and particle size. Since we use the Abdul-Razzak and Ghan (2000) parameterization, and the activated number concentration is only a function of number concentration and dry particle diameter.

As was the case in Gao et al., (2017), MATRIX-VBS has higher aerosol number concentration (Figure 4 left) but smaller particles (Figure 4 right) compared to MATRIX in the case presented in Figure 1. At first we expected that smaller particles would less likely activate, so we performed a simple sensitivity test to confirm it. By changing dry particle diameter of the particles in the activation scheme, the decreasing dry particle diameter indeed led to lower activated number concentration. However, a second sensitivity test with changing only number

concentration showed that higher number concentration would actually lead to lower activated number concentration as well.

In the Abdul-Razzak and Ghan (2000) scheme, increasing number concentration decreases critical supersaturation, and lower critical supersaturation leads to higher minimum dry particle radius that is able to activate. Therefore, activation is suppressed, since less particles exceed the threshold radius. The activated number concentration is calculated from the activation fraction and the number concentration. When the fraction is greater than the increase in number concentration, lower activated number concentration is achieved, as shown here.

As mentioned previously, within most of the scenarios, there is a decrease in fractional change as temperature increases, while both models experience decrease in activated number concentration with increased temperature. This means the decrease in activated number concentration for MATRIX-VBS is not as significant as that for MATRIX. There are two factors that contribute to such change. First, the heat and moisture diffusion term is dependent on temperature in the activation scheme (Abdul-Razzak and Ghan, 2000). Second, volatility of organics is temperature dependent. In MATRIX-VBS, when organic volatility is considered, the change is dampened. In other words, its number of activated particles is less sensitive to temperature change as compared to MATRIX, leading to what we see in the circle plots that the greater change at lower temperatures.

The length of day and season changes the duration and intensity of gas phase oxidation of semi-volatile gases, which is why we also looked at aerosol evolution driven by photochemistry at different latitudes. Since the model uses January emissions, different seasons are simulated at the different hemispheres, while different day lengths are simulated at higher latitudes of the southern hemisphere compared to tropical and high latitude northern hemisphere ones. As we inspected results across latitudes in the two hemispheres, we found varying activated number concentration in MATRIX-VBS compared to MATRIX and observed no evident trend. Such inconclusive and complex results may be due to gas-phase chemistry and photochemical ageing of semi-volatile organic vapors, which would require further examination in a separate dedicated study.

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4 Conclusions

With the inclusion of organic partitioning in an aerosol microphysics model, activated aerosol number concentration is decreased under most temperature and relative humidity conditions, except when under low updrafts, in clean environments at most temperatures and relative humidities, and in polluted environments at high temperatures and extremely low humidity conditions. Such changes are due to increased aerosol number concentration and smaller particles in the new model, as well as how number concentration and size are calculated in the chosen aerosol activation scheme, which determines how many particles are activated. Additionally, the temperature dependence of activated number concentration is decreased for most scenarios.

The simulations in this study, however comprehensive, are still highly idealized. In fact, Topping et al. (2013) showed that co-condensing organics lead to enhanced cloud droplet number concentration, which seems to contradict our results. However, it is important to note that our study is performed in a box model that does not resolve cloud physics. Activated number concentration is a precursor for CDNC, whose actual numbers will depend on the cloud microphysical calculation, which is not part of this study. We will investigate the effects of condensing organics in a global climate model in the future. The results presented here implicate that in the new model, most areas on Earth would experience less CCN on a typical day, but clean environments with above freezing temperatures, or polluted environments on an extremely dry and hot day, would form more CCN under low updraft velocity conditions, as compared to the old model. We expect that implementing the improved box model in the global scale that includes a two moment cloud microphysical scheme (Morrison and Gettelman, 2008; Gettelman and Morrison, 2015) would more accurately represent aerosol-cloud interactions, which will be our focus on a follow up study. Thus it would offer us valuable insights on how the addition of organic partitioning would change cloud activation in the global atmosphere and its implications for climate.

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207 The GISS ModelE Earth system model is publicly available. The box model code used here is
208 available upon request and will be publicly available in the future as part of GISS ModelE. The
209 data from all model simulations will be available upon request.

210 The authors declare that they have no conflict of interest.

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Table 1. Hygroscopicity κ used for each organic aerosol volatility bin.

	$\log_{10}C^*$ [μg m^{-3}]	soluble fraction [%]	κ
Sulfate	/	100	0.507
Black carbon	/	0	$5•10^{-7}$
Non-volatile organic carbon	/	78	0.141
Semi-volatile organic carbon	-2	100	0.180
	-1	87.5	0.158
	0	75	0.135
	1	62.5	0.113
	2	50	0.090
	3	37.5	0.068
	4	25	0.045
	5	12.5	0.023
	6	0	0.000
Dust	/	13	0.14
Sea salt	/	100	1.335

Table 2. Parameters used in the Monte-Carlo simulations.

Parameter			Range
T [K]			270, 280, 290, 300, 310
RH [%]			0.1, 20, 40, 60, 80, 100
Latitude			0, 30N/S, 60N/S, 90N/S
Updraft velocity [m/s]			0.5, 1, 2
Emissions of aerosols [**μ**g/m^3/s]	Sulfate (SO_2 in molecules/cm^3)		10^5, 10^6, $5•10^6$
	Primary organics		$5•10^{-6}$, $5•10^{-5}$, $5•10^{-4}$
	Nonvolatile biogenic organics from terpene source		$1•10^{-8}$, $5•10^{-6}$, $1•10^{-5}$
	Black Carbon		10^{-6}, 10^{-5}, 10^{-4}
Emissions of gases [molecules/cm^3]	VOCs (in sets)	Alkenes	$5•10^2$, $5•10^3$, $5•10^4$
		Paraffin	$5•10^3$, 10^4, $5•10^4$
		Terpenes	10^4, 10^5, 10^6
		Isoprene	10^4, 10^5, 50^6
	NO_x		10^5, 10^6, 10^7

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Table 3. Minimum and maximum of fractional change in average activated number concentration over the last 24 hours between the two models with low, medium and high level emissions at updraft velocities of 0.5, 1 and 2 m/s.

	Fractional change in activated number concentration					
Updraft velocity (m/s)	0.5		1		2	
	min	max	min	max	min	max
Low emission level	-9%	+21%	-16%	+2%	-14%	+5%
Medium emission level	-51%	+14%	-42%	-5%	-36%	-13%
High emission level	-56%	+31%	-48%	+9%	-43%	-9%

Table 4. Minimum and maximum of average activated number concentration over the last 24 hours of MATRIX and MATRIX-VBS with low, medium and high level emissions at updraft velocities of 0.5, 1 and 2 m/s.

		Activated number concentration					
Updraft velocity (m/s)		0.5		1		2	
		min	max	min	max	min	max
Low emission level	MATRIX	23	305	351	1160	963	2799
	MATRIX-VBS	24	283	338	1026	887	2473
Medium emission level	MATRIX	19	152	359	1233	1476	3711
	MATRIX-VBS	16	139	304	884	1021	2498
High emission level	MATRIX	3	60	199	1280	1925	5703
	MATRIX-VBS	3	63	185	1150	1677	4142

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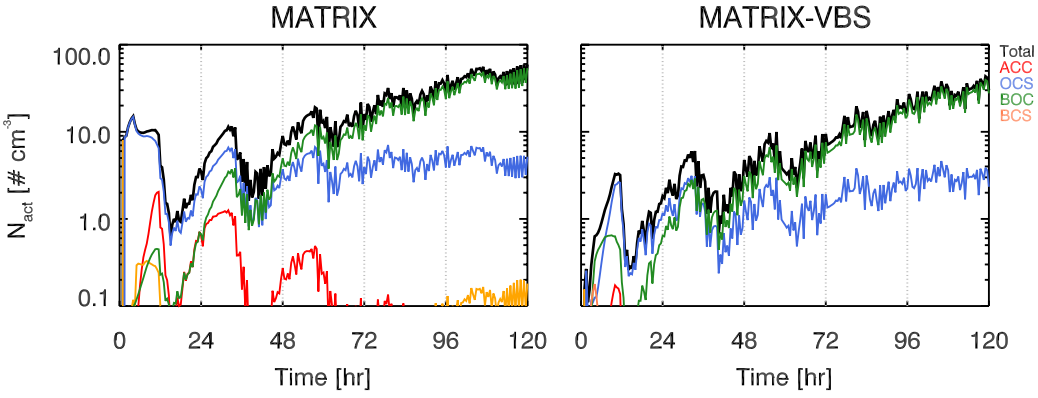

403

404 **Figure 1. Activated number concentration of aerosol populations (see main text for details)**
405 **for MATRIX (left) and MATRIX-VBS (right) for 290 K and 40% RH at 30°N latitude with**
406 **medium emission levels and 0.5 m/s updraft velocity.**

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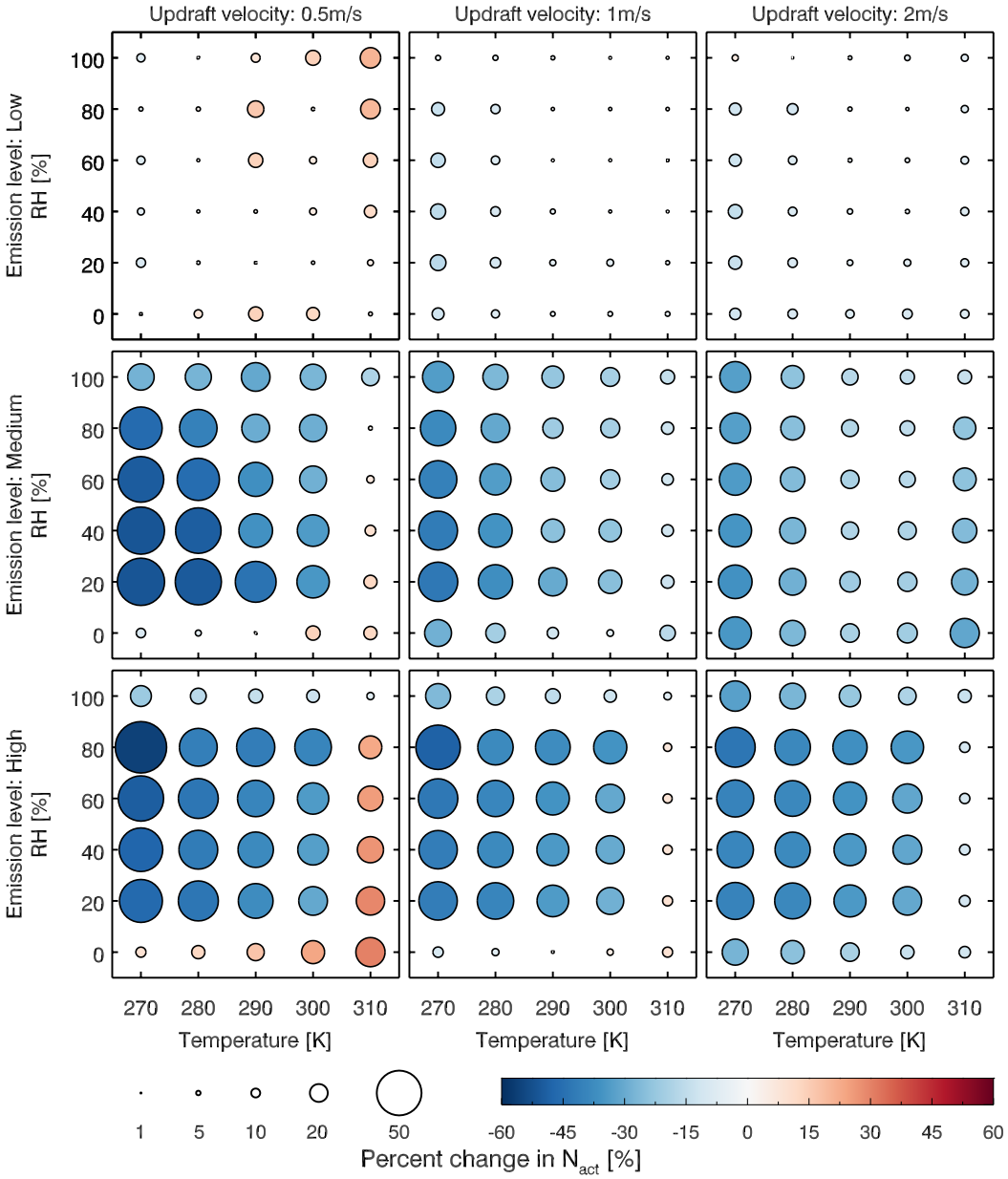

408

409 **Figure 2. Fractional change of average activated number concentration (size and color of the**
410 **circles) over the last 24 hours of a 5-day simulation between the two models with low (top**
411 **row), medium (middle row) and high (bottom row) level emissions at updraft velocities of 0.5**
412 **(left column), 1 (middle column) and 2 (right column) m/s.**

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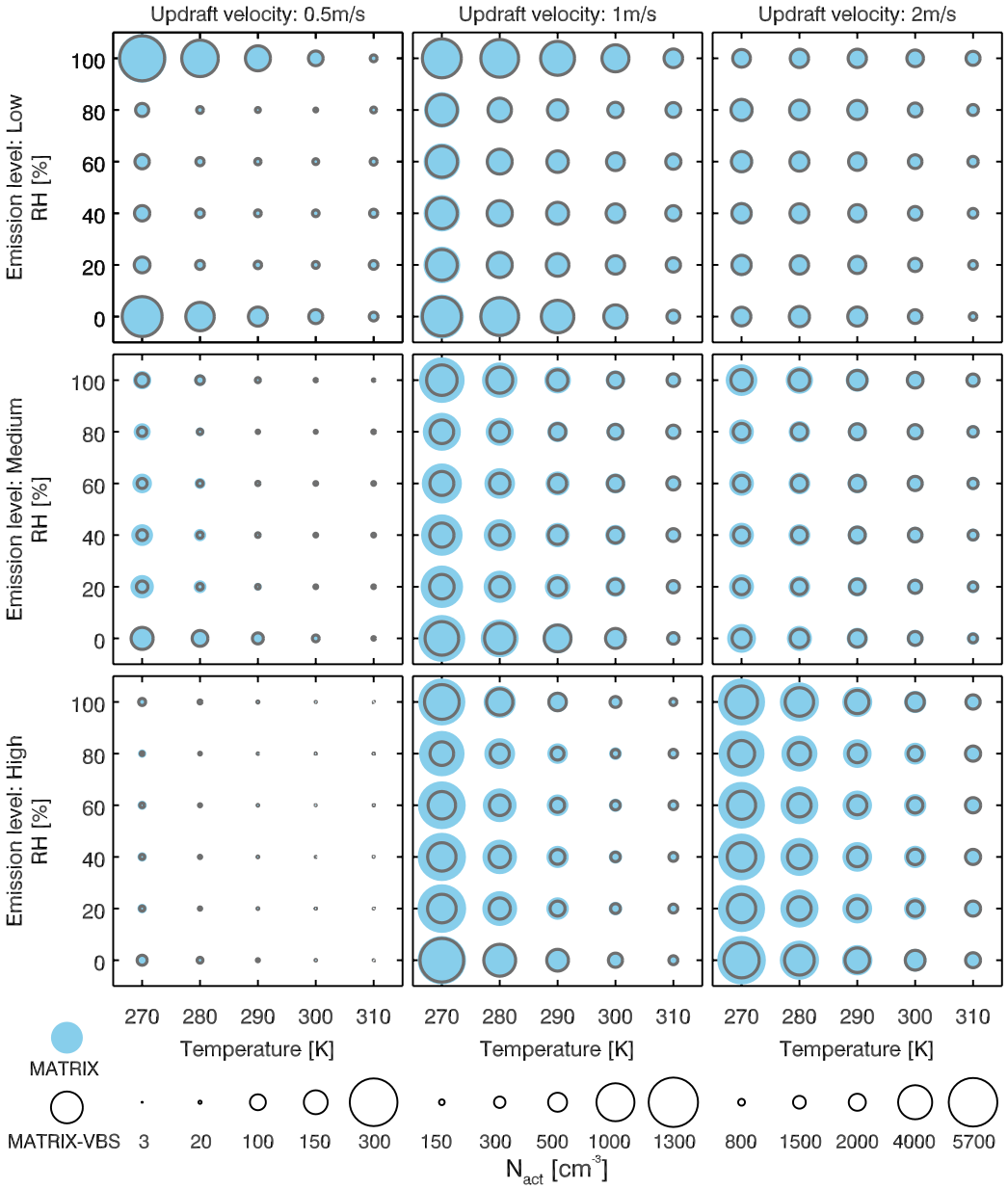

Figure 3. Average activated number concentration (circle size) during the last 24 hours of a 5-day simulation in MATRIX and MATRIX-VBS with low (top row), medium (middle row) and high (bottom row) emission levels at updraft velocities of 0.5 (left column), 1 (middle column) and 2 (right column) m/s. Note difference in scales per column.

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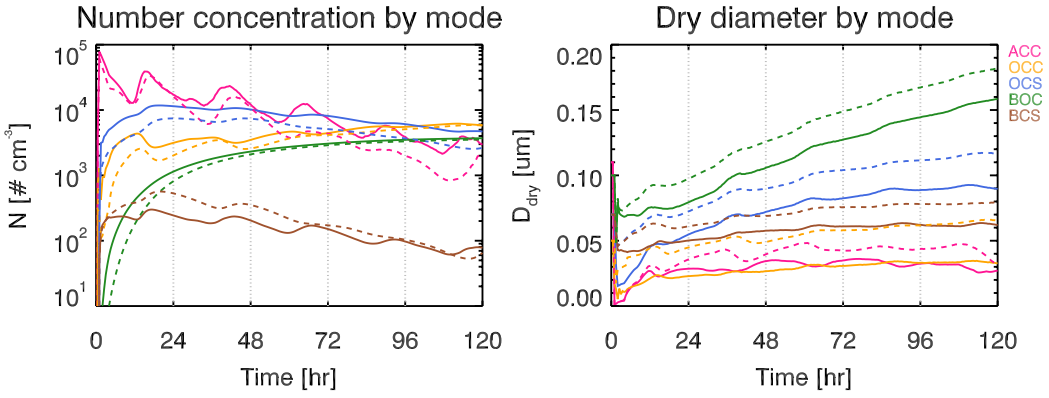

419

420 **Figure 4. Number concentration (left column) and dry particle diameter (right column) by**
421 **mode (color lines) for MATRIX (dashed lines) and MATRIX-VBS (solid lines) for the**
422 **experiments with the same conditions as Figure 1.**