Dear Editor,

We received your comments and suggestions for our discussion paper "Can Semi-Volatile Organic Aerosols Lead to Less Cloud Particles?" on ACPD. We thank you very much for your efforts in evaluating our submission.

Of the final referee's comment, we have modified this line in the Conclusions section, "*We would like to emphasize that our results do not imply that the Earth has less CCN than currently thought. Instead, they imply that if in a model semi-volatile organics will be simulated together with aerosol microphysics, a general decrease is to be expected*.", which now reads, "*We would like to emphasize that our results do not imply that the Earth has less CCN than currently thought. Instead, they imply that if in a model semi-volatile organics will be simulated together with aerosol microphysics, a general decrease is to be expected, assuming our model captures all relevant contributory processes.*" This change is highlighted in the updated revised manuscript.

We hereby resubmit the revised discussion paper to be considered for publication in *Atmospheric Chemistry and Physics.* We confirm that all authors listed on the manuscript concur with submission in its revised form. Should you have any remaining questions, we will be happy to address them.

Sincerely,
C. Y. Gao, S. E. Bauer and K. Tsigaridis

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

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9

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

22   **1 Introduction**

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

31   cloud interactions (Ghan et al., 1998; McFiggans et al., 2006; Ervens et al., 2007; Gibson et al.,

32   2007; Medina et al., 2007; Cubison et al., 2008; Anttila, 2010).

33   Climate models calculate cloud droplet number concentration (CDNC) using aerosol

34   activation schemes, whose main governing parameters include aerosol number, size,

35   hygroscopicity, updraft velocity, as well as critical supersaturation. Physically-based aerosol

36   activation schemes (e.g. Abdul-Razzak and Ghan, 2000; Fountoukis and Nenes, 2005; Ming et al.,

37   2006; Shipway and Abel, 2010) are commonly used in global climate models for fast diagnostics

38   of nucleation and to estimate the aerosol indirect effect in long-term climate simulations (Ghan,

39   2011). Several studies examined the relationship between the fore-mentioned parameters and how

40   they play together to activate particles. Ghan et al. (1998) examined sea salt's influence on sulfate

41   particle activation and introduced the competition effect. Since all CCN have to compete for

42   available water vapor in order to activate, the competition limits the maximum supersaturation in

43   in-cloud updrafts (Storelvmo et al., 2006). Ghan et al. (1998) concluded that activated number

44   concentration increases with increasing sea salt when sulfate is low and updraft is strong, and it

45   decreases when sulfate is high and updraft is weak, because maximum supersaturation is reduced.

46   Another study (Reutter et al. 2009) explored how much CDNC depend on updraft velocity, size

47   distribution and hygroscopicity. They found that size distribution played a greater role than particle

48   hygroscopicity on CDNC and discovered different CCN activation and cloud droplet formation

49   regimes, which are determined by aerosol number concentration and updraft velocity.

50   Semi-volatile organic aerosols contribute significantly to the growth of particles to CCN

51   sizes (Yu, 2011). More notably, as aerosol size increases, the range of organic volatilities involved

52   in aerosol growth increases (Pierce et al., 2011; Yu, 2011). The inclusion of semi-volatile organics

53   in models modifies CCN formation rates (Petters et al., 2006, Riipenen et al., 2011; Scott et al.,

54   2015) as well as hygroscopicity (Petters and Kreidenweis, 2007), in addition to bulk aerosol mass,

55   size distribution and composition. By adding semi-volatile organic partitioning to our existing

56   microphysics model MATRIX (Multiconfiguration Aerosol TRacker of mIXing state; Bauer et al.,

57   2008), which resolves aerosol mixing state, we were able to examine how they change bulk aerosol

58   mass, size distribution and composition. However, the effects of semi-volatile organic partitioning

59   combined with aerosol mixing state on particle activation remain unexplored.

60   In our previous work, we demonstrated that including semi-volatile organics would lead to

61   higher aerosol number concentration and smaller particles (Gao et al., 2017). As was the case for

62 the original aerosol microphysics model MATRIX, our further-developed box model MATRIX-

63 VBS (Gao et al., 2017) follows the same multi-modal aerosol activation approach by Abdul-

64 Razzak and Ghan (2000). The activation parameterization accounts for aerosol size distribution,

65 composition, mixing state, and in-cloud updraft velocity. Curious about the change in activation

66 with the newly-present semi-volatile organics and the governing parameters influencing it, we

67 investigated the difference in activated number concentration in two box model set ups: MATRIX

68 (Bauer et al., 2008) and MATRIX-VBS (Gao et al., 2017).

69 **2 Methods**

70 2.1 Model Description

71   MATRIX-VBS (Gao et al., 2017) is an aerosol microphysics model that includes organic

72 aerosol volatility in its calculations. It was developed by implementing VBS (volatility-basis set;

73 Donahue et al., 2006) in the aerosol microphysics model MATRIX (Bauer et al., 2008), which is

74 a box model that is also used in the NASA GISS ModelE Earth System Model (Bauer et al., 2008,

75 2012; Schmidt et al., 2014). Since the publication of Gao et al., 2017, which included organic

76 condensation on fine mode aerosols, we further developed the model which now allows semi-

77 volatile organics in the system to condense on coarse mode dust and sea salt as well. We have also

78 included nitrate radicals as an oxidant for organics in addition to the hydroxyl radical that was used

79 in the original VBS scheme, even though it is a very minor oxidation pathway in the model (rate

80 constant for the oxidation by $NO_3^{\bullet}$ is $1*10^{-13}$ cm^3 $molecules^{-1}$ s^{-1}; Atkinson, 1997). As previously

81 stated, we use Abdul-Razzak and Ghan (2000) activation parameterization, which calculates the

82 activated particle number concentration depending on chemically-resolved number concentrations

83 using Köhler Theory. The hygroscopicity parameters κ for each aerosol species presented in Table

84 1 were calculated from their solubility fraction. For organics, we assumed a linear increase of

85 solubility with decreasing volatility (Jimenez et al., 2009). Since we use Pankow type partitioning

86 (Pankow, 1994), water is not considered in the partitioning process. In addition, we do not use

87 different kappa/RH relationships per organic species, which was found to be important for

88 biogenic SOA (Rastak et al., 2017).

89

90   2.2 Simulations

91   A Monte-Carlo analysis with a range of chemical and meteorological conditions (Table 2)

92   was performed, to pinpoint which processes affect organics and the mixed aerosol population in

93   general the most. Since global models need to resolve a wide range of conditions, from very clean

94   to very polluted and for a wealth of meteorological conditions, we simulated 630 possible

95   atmospheric scenarios on Earth across the whole parameter space, e.g. temperature, relative

96   humidity, latitude, emissions levels and updraft velocity, for 120 hours (5 days) simulations with

97   no deposition and dilution. Three types of environmental conditions were simulated: clean,

98   moderate and polluted, as defined by different levels of emissions which were determined using a

99   probability distribution of the gridded emission fields in GISS ModelE for January present-day

100   conditions. During this development phase, biogenic secondary organic aerosols from terpenes

101   oxidation in MATRIX-VBS are treated as nonvolatile, while only the anthropogenic aerosols are

102   treated as semi-volatile.

103   **3 Results and discussion**

104   We found that activated number concentration is lower for most cases in the MATRIX-

105   VBS model, which considers semi-volatile organic aerosols, as compared to the MATRIX model.

106   However, under low updrafts, in clean environment at above freezing temperatures, and in polluted

107   environments at high temperature (310K) and extremely low humidity conditions (0% RH) during

108   aerosol formation, activated number concentration is higher in MATRIX-VBS than in MATRIX.

109   As an example, the activated number concentration for a case with temperature at 290˚K,

110   relative humidity at 40%, medium emission levels and an updraft of 0.5 m/s at 30°N latitude is

111   shown in Figure 1 for the two models. Mixing states of aerosols in MATRIX and MATRIX-VBS

112   are represented as aerosol populations, which all contain SO_4, NO_3, NH_4 and H_2O, in addition to

113   the species that define the populations (Bauer et al., 2008, 2013). The four most dominant aerosol

114   populations for the activated number concentration in MATRIX are ACC (SO_4, NO_3, NH_4), OCS

115   (organics, SO_4, NO_3, NH_4), BOC (black carbon, organic carbon, SO_4, NO_3, NH_4) and BCS (black

116   carbon, SO_4, NO_3, NH_4). Only two dominant populations are calculated in MATRIX-VBS, OCS

117   and BOC, as in Gao et al., 2017, since OCC evaporates and re-condenses on all particles, based on

118   their calculated surface area and mass concentration. Since OCS and BOC have the largest surface

119   area, they are calculated to have the strongest growth via organics condensation. Additionally, the

4

120   competition between sulfate, organics and black carbon, determines the loss of ACC and the

121   formation of BCS: OCC coagulates with ACC to form OCS, and this coagulation increases in

122   MATRIX-VBS due to smaller OCC particles; therefore, there are less ACC particles left to

123   coagulate with black carbon to form BCS. At the end of the 5-day simulation (Figure 1), MATRIX-

124   VBS has approximately a total of 30 activated particles/cm^3, whereas MATRIX has approximately

125   60 activated particles/cm^3 under the same conditions.

126   Figure 2 shows a more comprehensive look across all temperature and relative humidity

127   scenarios studied. The results show that for most scenarios, MATRIX-VBS has lower (red circles)

128   activated number concentration compared to MATRIX. However, some rare cases show the

129   opposite behavior. These are for above freezing temperatures in the low emission level under low

130   updraft (top left) scenarios, high temperature (310K) and extremely low humidity (0% RH) in the

131   medium emission level under low updraft (middle left) scenarios, as well as the high emission

132   level under low (bottom left) and medium (bottom middle) updraft scenarios. Note that low RH

133   values do not mean that these correspond to cloud conditions. Aerosols form outside of clouds in

134   our model, where RH can be very low. Activation though will occur after aerosol formation, when

135   an air parcel starts rising with a given updraft velocity, in which air parcel supersaturation will

136   develop and will cause aerosol activation.

137   Across all scenarios, the changes in activated number concentration between MATRIX-

138   VBS and MATRIX range from a -56% to +31% (Table 3). The range of the difference becomes

139   more significant as emission levels increase, yet less significant as updraft velocity increases.

140   Within most emission level-updraft velocity scenarios, as temperature increases, the fractional

141   change in activated number concentration between the two models decreases. Also within most

142   emission level-updraft velocity scenarios (Figure 3, Table 4), as temperature increases, there are

143   less activated particles in MATRIX. We also observed the same behavior in MATRIX-VBS,

144   higher temperature, less activated particles.

145   In order to understand the cause of the difference in activation, we traced back to the key

146   difference between the two models: partitioning of organics. The inclusion of organics partitioning

147   leads to changes in aerosol mixing state and size distribution, as discussed in Gao et al. (2017).

148   Therefore, the change in activated number concentration could only be caused by changes in mass

149   concentration, number concentration and particle size. Since we use the Abdul-Razzak and Ghan

150   (2000) parameterization, the activated number concentration is mainly a function of number

151   concentration and dry particle diameter in our model. The parameterization is also a function of

152   geometric standard deviation, which is constant per population in our model as it did in MATRIX

153   (Bauer et al., 2008), as well as a function of aerosol composition and hygroscopicity, as mentioned

154   in the model description, for which we assume a linear increase of solubility with decreasing

155   volatility. The hygroscopicity of the aerosol populations changes with time, as the internal mixing

156   of aerosol populations is altered by aerosol microphysics.

157   As was the case in Gao et al., (2017), MATRIX-VBS has higher aerosol number

158   concentration (Figure 4 left) but smaller particles (Figure 4 right) compared to MATRIX in the

159   case presented in Figure 1. At first we expected that smaller particles would less likely activate, so

160   we performed a simple sensitivity test to confirm it. By changing dry particle diameter of the

161   particles in the activation scheme, the decreasing dry particle diameter indeed led to lower

162   activated number concentration. However, a second sensitivity test with changing only number

163   concentration showed that higher number concentration would actually lead to lower activated

164   number concentration as well.

165   In the Abdul-Razzak and Ghan (2000) scheme, increasing number concentration decreases

166   critical supersaturation, and lower critical supersaturation leads to higher minimum dry particle

167   radius that is able to activate. Therefore, activation is suppressed, since less particles exceed the

168   threshold radius. The activated number concentration is calculated from the activation fraction and

169   the number concentration. When the fraction is greater than the increase in number concentration,

170   lower activated number concentration is achieved, as shown here.

171   As mentioned previously, within most of the scenarios, there is a decrease in fractional

172   change as temperature increases, while both models experience decrease in activated number

173   concentration with increased temperature. This means the decrease in activated number

174   concentration for MATRIX-VBS is not as significant as that for MATRIX. There are two factors

175   that contribute to such change. First, the heat and moisture diffusion term is dependent on

176   temperature in the activation scheme (Abdul-Razzak and Ghan, 2000). Second, volatility of

177   organics is temperature dependent. In MATRIX-VBS, when organic volatility is considered, the

178   change is dampened. In other words, its number of activated particles is less sensitive to

179   temperature change as compared to MATRIX, leading to what we see in the circle plots that the

180   greater change at lower temperatures.

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

191 **4 Conclusions**

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

200   Our conclusion that fewer particles are activated at higher updrafts is in contrast to
201 Connolly et al. (2014), who found that fewer particles activated at low updrafts, using a different
202 geometric standard deviation in the same parameterization of aerosol activation as the one we use.
203 Such a difference can be due to the fact that the Abdul-Razzak and Ghan (2000) activation
204 parameterization produces a different response when multiple modes are used, as shown by
205 Connolly et al. (2014b) and Simpson et al. (2014). Additionally, in our study, the geometric
206 standard deviation remained constant per aerosol population. However, it is worth exploring in the
207 future to use reduced geometric standard deviation in our calculations to directly compare with
208 values used by Connolly et al. (2014) and Crooks et al. (2017). In fact, in a comparison study,
209 Ghan et al. (2011) found that the Abdul-Razzak and Ghan (2000) scheme tend to have lower

210   activation fractions and droplet concentrations compared to the Fountoukis and Nenes (2005)
211   activation scheme.

212   Topping et al. (2013) showed that co-condensing organics lead to enhanced cloud droplet
213   number concentration, which seems to contradict our results. However, it is important to note that
214   contrary to Topping et al. (2013), our study is performed in a box model that does not resolve
215   cloud droplet growth as the air mass rises and cools, which leads to additional condensation of
216   organic vapors and water due to the temperature decline, and contributes to cloud droplet growth
217   due to additional water uptake. The simulations in this study, however comprehensive, are still
218   highly idealized.

219   We would like to emphasize that our results do not imply that the Earth has less CCN than
220   currently thought. Instead, they imply that if in a model semi-volatile organics will be simulated
221   together with aerosol microphysics, a general decrease is to be expected, assuming our model
222   captures all relevant contributory processes. We will investigate the effects of condensing organics
223   in a global climate model in the future. The results presented here implicate that in the new model,
224   most areas on Earth would experience less CCN on a typical day, but clean environments with
225   above freezing temperatures, or polluted environments on an extremely dry and hot day, would
226   form more CCN under low updraft velocity conditions, as compared to the old model. We expect
227   that implementing the improved box model in the global scale that includes a two moment cloud
228   microphysical scheme (Morrison and Gettelman, 2008; Gettelman and Morrison, 2015) would
229   more accurately represent aerosol-cloud interactions, which will be our focus on a follow up study.
230   Thus it would offer us valuable insights on how the addition of process level phenomena in aerosol
231   microphysics, as applied here for the organics partitioning, would affect cloud microphysics in the
232   global atmosphere and its implications for climate.

233

239   The GISS ModelE Earth system model is publicly available. The box model code used here is

240   available upon request and will be publicly available in the future as part of GISS ModelE. The

241   data from all model simulations will be available upon request.

242   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	$5\cdot10^{-7}$
Non-volatile organic carbon	/	78	0.141
	-2	100	0.180
	-1	87.5	0.158
	0	75	0.135
Semi-volatile organic carbon	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 [μg/m^3/s]	Sulfate (SO$_2$ in molecules/cm^3)		10^5, 10^6, $5\cdot10^6$
	Primary organics		$5\cdot10^{-6}$, $5\cdot10^{-5}$, $5\cdot10^{-4}$
	Nonvolatile biogenic organics from terpene source		$1\cdot10^{-8}$, $5\cdot10^{-6}$, $1\cdot10^{-5}$
	Black Carbon		10^{-6}, 10^{-5}, 10^{-4}
Emissions of gases [molecules/cm^3]	VOCs (in sets)	Alkenes	$5\cdot10^2$, $5\cdot10^3$, $5\cdot10^4$
		Paraffin	$5\cdot10^3$, 10^4, $5\cdot10^4$
		Terpenes	10^4, 10^5, 10^6
		Isoprene	10^4, 10^5, 50^6
	NO$_x$		10^5, 10^6, 10^7

438 **Table 3. Minimum and maximum of fractional change in average activated number**
439 **concentration over the last 24 hours between the two models with low, medium and high**
440 **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%

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443 **Table 4. Minimum and maximum of average activated number concentration over the last**
444 **24 hours of MATRIX and MATRIX-VBS with low, medium and high level emissions at**
445 **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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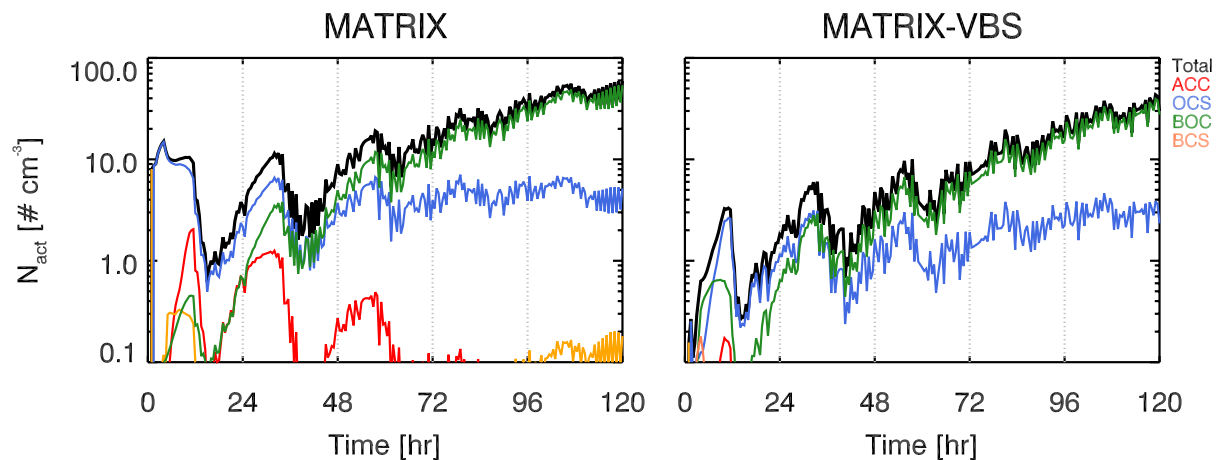
Figure 1. Activated number concentration of aerosol populations (see main text for details) for MATRIX (left) and MATRIX-VBS (right) for 290 K and 40% RH at 30°N latitude with medium emission levels and 0.5 m/s updraft velocity.
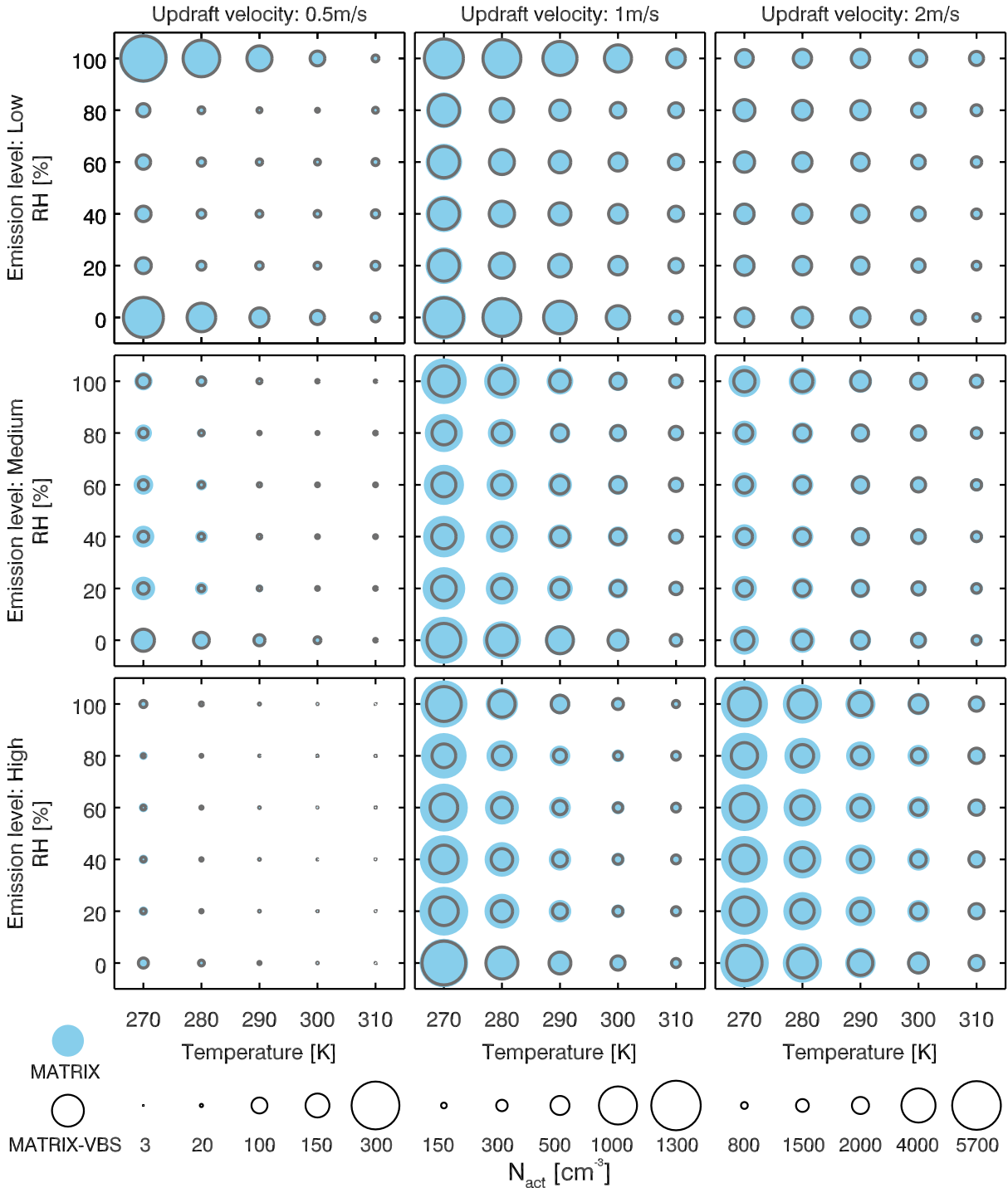
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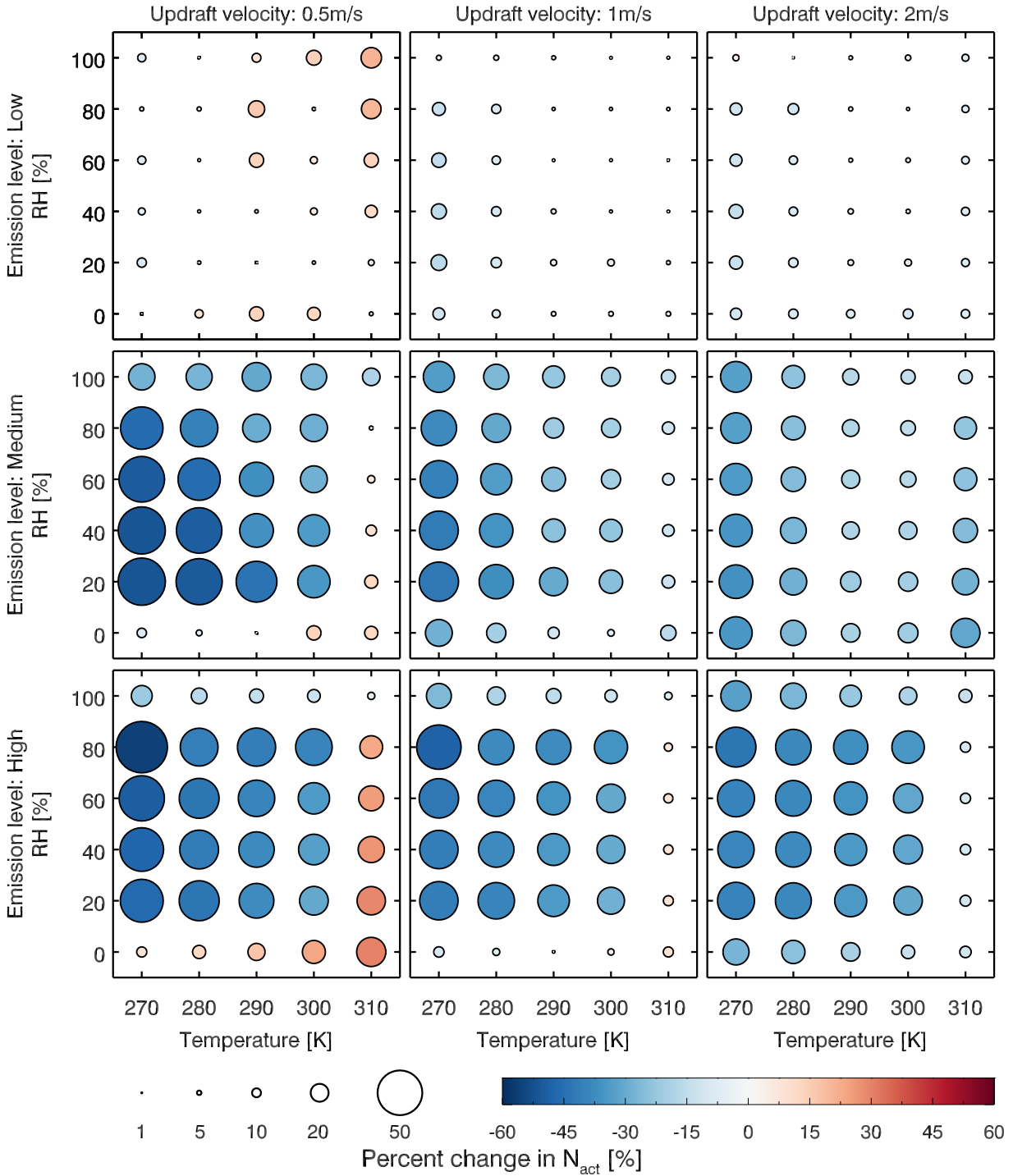
Figure 2. Fractional change of average activated number concentration (size and color of the circles) over the last 24 hours of a 5-day simulation between the two models with low (top row), medium (middle row) and high (bottom row) level emissions at updraft velocities of 0.5 (left column), 1 (middle 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.

Number concentration by mode

Dry diameter by mode

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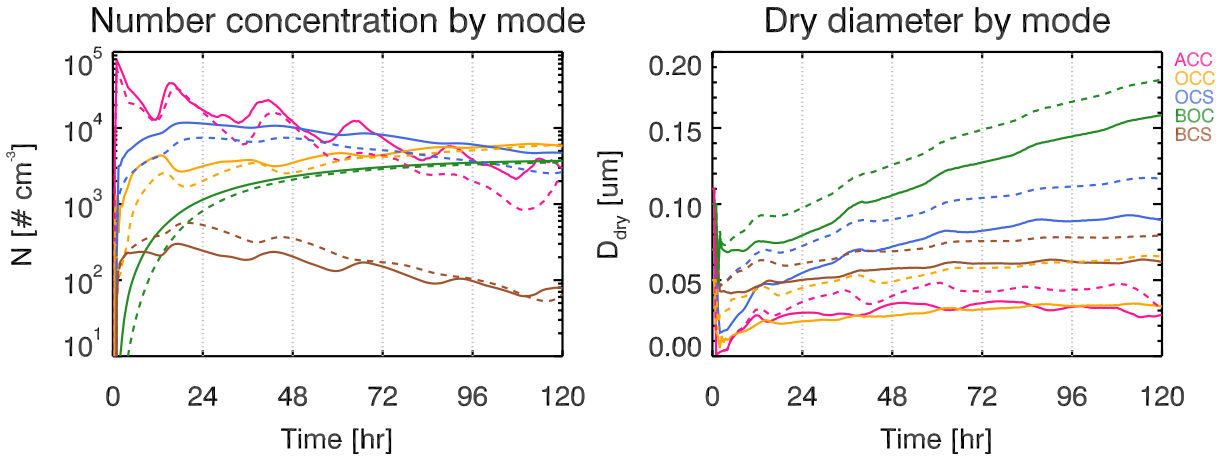
Figure 4. Number concentration (left column) and dry particle diameter (right column) by mode (color lines) for MATRIX (dashed lines) and MATRIX-VBS (solid lines) for the experiments with the same conditions as Figure 1.