Reply to the review of "Can Semi-Volatile Organic Aerosols Lead to Less Cloud Particles?"

We would like to thank the reviewer for their careful reading and constructive comments. Please find below our answers to all points raised. The original reviewer's comments are in black font, and our replies are in blue. Changes in the text are *italicized*.

1. This paper investigates the sensitivity of cloud droplet activation, using the Abdul-Razzak et al. (2000) parameterisation, to the aerosol chemical composition, mass and number concentrations, and particle size. The main finding is that the simulations suggest that semi-volatile compounds almost always lead to fewer cloud droplets than without semi-volatiles. Whilst I agree this area is worthy of further investigations, I believe the main finding is an artefact of the parameterisation used – the Abdul-Razzak et al. (2000) scheme and perhaps also in the way it is implemented in the model.

The Abdul-Razzak and Ghan (2000) scheme is widely used in global climate models and in MATRIX (Bauer et al., 2008), which is the original version of the model that we compare the new model against. We are not aware of an artefact being reported in the literature. We also checked the implementation, and found no obvious problems with it. As a matter of fact, the Connolly et al. (2014) and Crooks et al. (2017) papers mentioned by the reviewer as more accurate in terms of aerosol activation calculations, use that same ARG scheme, only with different input parameters, appropriate for each individual study.

2. There are a few statements that lead me to conclude the authors might not be treating activation in the correct way. For example, on line 141 the authors write "the activated number concentration is only a function of number concentration and dry particle diameter". This is not true: in addition to environmental parameters such as temperature and pressure, activation is also a function of the geometric standard deviation (see equation 15 of ARG, 2000), and the aerosol chemistry (see equation 3 of ARG, 2000).

Thanks for pointing this out. This was worded imprecisely and may have delivered the wrong message, the activated number concentration is not "only" a function of number concentration and dry particle diameter in the ARG scheme. We used "only" because in our model the geometric standard deviation is constant for each aerosol population. Aerosol composition, which is calculated explicitly per population by the aerosol microphysics of the model, and hygroscopicity, are indeed taken into account in our model. We mention hygroscopicity in the submitted manuscript in lines 80-85: "*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)." Therefore, we considered them not changing variables per aerosol component, but due to size distribution evolution the hygroscopicity of aerosol populations evolves with time and is taken into account in our calculations.

To make this point clearer in the manuscript, this sentence has been revised as follows: "*Since we use the Abdul-Razzak and Ghan (2000) parameterization, the activated number concentration is mainly a function of number concentration and dry particle diameter in our model setting. The parameterization is also a function of geometric standard deviation, which is constant per population in our model as it did in MATRIX (Bauer et al., 2008), as well as a function of aerosol composition and hygroscopicity, as mentioned in the model description, for which we assume a linear increase of solubility with decreasing volatility. The hygroscopicity of the aerosol populations changes with time, as the internal mixing of aerosol populations is altered by aerosol microphysics.* "

3. Of the parameters above the geometric standard deviation is an important parameter for cloud drop activation. If the distribution is narrow (small geometric standard deviation) then the competition effect will be small and more particles will activate at once. If the aerosol size distribution is broad / geometric standard deviation is large one tends to find that fewer particles activate. Connolly et al. (2014) showed that for single aerosol modes it was necessary to shift the geometric standard deviation to smaller values in order to predict activated fractions more accurately – see their equation 15. Crooks et al. (2018) have now extended this result to multiple modes.

Please clarify whether this shifting done in the current manuscript.

We have not implemented geometric standard deviation shifting, and it remains constant both in MATRIX and other aerosol microphysics modules used by other models. MATRIX is based on the quadrature methods of moments and in a future version we might be able to treat higher moments of the size distribution, but at this point this version is not developed. But we will keep in mind the sensitivity of activation towards the standard deviation, and will perform future sensitivity experiments for that quantity.

We have included the following in our conclusions section:
"*In our study, the geometric standard deviation remained constant per aerosol population. However, it is worth exploring in the future to use reduced geometric standard deviation in our calculations to directly compare with values used by Connolly et al. (2014) and Crooks et al. (2017).*"

4. On line 152 the authors mention that, in the Abdul-Razzak et al. (2000) scheme, increasing number concentration decreases the ambient supersaturation, which reduces the number activated, therefore suppressing activation. This argument is slightly circular though since to reduce the ambient supersaturation more particles must have been activated.

It would have been circular if we were saying "increasing <u>activated number concentration</u> decreases ambient supersaturation" but we are saying "increasing <u>number concentration</u> decreases the ambient supersaturation."

5. We know that the Abdul-Razzak et al. parameterisation does not always predict the correct response to inputs. Connolly et al. (2014b) showed a comparison between the Abdul-Razzak et al. (2000) and the Fountoukis and Nenes (2005) parameterisations. Their Figure 3(a-d) is reproduced in Figure 1, below. Figure 1a shows how increasing the total aerosol mass (by increasing the aerosol particle number concentration, the x-axis) eventually leads to less particles being activated in the Abdul-Razzak et al. parameterisation. Such a reduction is not seen in the Fountoukis and Nenes (2005) parameterisation (see Figure 1d).

Figure 1. shows the activated fraction when adding NaCl particles to the aerosol population with a total mass loading indicated by the x-axis. Colours refer to different modal diameters of the NaCl particle size distribution (see Connolly et al, 2014b for full details). (a) is for the Abdul-Razzak et al. parameterisation; (b) is for the Fountoukis and Nenes parameterisation.

As mentioned by the reviewer, "*Figure 1a shows how increasing the total aerosol mass (by increasing the aerosol particle number concentration, the x-axis) eventually leads to less particles being activated in the Abdul-Razzak et al. parameterisation.*" This is essentially what we have also found.
We included the following into our conclusions: " *In fact, in a comparison study, Ghan et al. (2011) found that the Abdul-Razzak and Ghan (2000) scheme tend to have lower activation fractions and droplet concentrations compared to the Fountoukis and Nenes (2005) activation scheme.*"

Connolly et al. (2014b)'s result that the Fountoukis and Nenes parameterization does not give the same result as the ARG parameterization is sound, but not within the scope of our study, since it is not in our goal to perform an activation schemes comparison, but to compare two models with the same activation scheme but different organic aerosol volatility treatment. The results of our paper are based on the changes volatile organics cause in aerosol microphysical properties, changes in mass, number, size, composition, and how this affects activation. We are not testing the activation schemes themselves, as we want to be consistent with parameterizations within the GISS GCM, the model this new scheme is currently being implemented in.

6. The results in the presented manuscript, that fewer particles are activated with semi-volatiles for higher updrafts, are also in contrast to Connolly et al. (2014), which found ARG at low updraft speeds activated fewer particles with semivolatiles switched on; (see Figure 6 of Connolly et al, 2014). I suspect the reason for this contrast between the two studies is that the Abdul-Razzak et al. parameterisation gives a different response when multiple aerosol modes are used, as has been shown by Connolly et al. (2014b). Indeed results by Simpson et al (2014), which are reproduced in Figure 2, indicate that this is the case. The ARG parameterisation results are indicated by the '+' symbols and it is shown that ARG is further below the 1 : 1 line when the updrafts are high (red) vs when the updraft is low (blue).

Figure 2. Results from Simpson et al. (2014). Using a bimodal aerosol size distribution. Symbols coloured by updraft velocity (m s-1).

We have included these differences in our conclusions section as follows:
"*Our conclusion that fewer particles are activated at higher updrafts is in contrast to Connolly et al. (2014), who found that fewer particles activated at low updrafts, using a different geometric standard deviation in the same parameterization of aerosol activation as the one we use. Such a difference can be due to the fact that the Abdul-Razzak and Ghan (2000) activation parameterization produces a different response when multiple modes are used, as shown by Connolly et al. (2014b) and Simpson et al. (2014).*"

7. On line 126 there is mention of simulations looking at activation at extremely low humidity. How relevant are these simulations, given that activation would not occur at low RH anyway?

Admittedly the explanation in the manuscript was too brief regarding the RH values below 100%. We calculate aerosol formation and growth via MATRIX-VBS microphysics outside of clouds, where RH can have any value, even significantly below 100%. Then we calculate aerosol activation, assuming that the air mass containing those aerosols started rising with a given updraft velocity, and supersaturation conditions developed, emulating cloud formation.

We added a sentence following the line referred by the reviewer to explain this: "*Note that low RH values do not mean that these correspond to cloud conditions. Aerosols form outside of clouds in our model, where RH can be very low. Activation though will occur after aerosol formation, when an air parcel starts rising with a given updraft velocity, in which air parcel supersaturation will develop and will cause aerosol activation.*"

8. The statement on line 188 about the discrepancy between the results presented and those of Topping et al. (2013) seems to indicate that the differences are because Topping et al. (2013) resolved more physics than the cloud drop activation process. In fact this is not really true. The Topping et al. study only considered condensation until the point of cloud drop activation.

Topping et al. (2013) uses an adiabatic cloud parcel model to study cloud droplet formation, which does resolve some cloud physics, contrary to our box model, which does not. This is clearly stated in the abstract and throughout the Topping et al. (2013) paper, that describes cloud droplet formation and growth as an air parcel rises, which goes beyond just activation. That study, however, same as ours, does not handle autoconversion, but it does track the air mass as it rises and cools, which leads to additional condensation of organic vapors and water due to the temperature decline, leading to cloud droplet growth due to additional water uptake, which our model does not resolve. What our model can provide is the initial activation of an aerosol population when it first enters a supersaturated air mass with a given updraft velocity, and not the evolution of cloud droplets inside the rising plume.

We modified our statement which now reads: "*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 contrary to Topping et al. (2013), our study is performed in a box model that does not resolve cloud droplet growth as the air mass rises and cools, which leads to additional condensation of organic vapors and water due to the temperature decline, and contributes to both cloud*

droplet growth due to additional water uptake and enhanced activation during convection. ”

9. Unfortunately, because of these shortcomings I feel like the conclusions drawn about most areas on earth experiencing less CCN that currently thought, except the more polluted & dry areas, are all dependent on the parameterisation used and its implementation.

We would like to reiterate that our goal is to compare two versions of an aerosol microphysics model with different treatment of organic aerosols and the same activation scheme. Our results do not imply that the Earth has less CCN than currently thought, as the reviewer states; 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.

We included this statement in our conclusion:
“*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 during initial cloud formation.* ”

As mentioned before, we added semi-volatile organics and they changed number and size of particles, which affected the activated number concentration via the parameterization. See lines 183-185 of the submitted manuscript: “*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.*”

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Reply to the 2nd review of "Can Semi-Volatile Organic Aerosols Lead to Less Cloud Particles?"

We would like to thank the reviewer for their efforts in evaluating our submission and providing constructive comments. Please find below our answers to all points raised. The original reviewer's comments are in black font, and our replies are in blue. Changes in the text are *italicized*.

In this paper the authors discuss results from a box-model sensitivity study comparing changes in parameterized cloud droplet numbers to a representation of semi-volatile organic partitioning. Before consideration for publication, there are a number of issues that need addressing that are raised below.

1. Having read the original paper on the MATRIX-VBS model, I couldn't tell whether, since you are prescribing aqueous solubility, water is explicitly included in the organic partitioning simulations? If not, there is an inconsistency between prescribing a 'kappa' value [which will not stay constant unless 100% solubility is assumed] and assuming completely 'dry' partitioning unless all organics are thus actually assumed to have zero aqueous solubility. If so, please describe how you have accounted for varying solubility, and presumably molecular weight, in the new partitioning simulations. With varying solubility per bin, how would you then account for the influence of one VBS bin on the other in-line with mixing thermodynamics? If you do include water in the partitioning simulations, how might you account for this in equilibrium partitioning at 100%RH? How does the fixed linear change with solubility map to the VBS source VOCs used in a host model? For example, the use of experimentally determined RH variation in Kappa from isoprene and monoterpene SOA experiments has been shown to have significant impacts on two state-of-the-art climate model forcing estimates [Microphysical explanation of the RH dependent water affinity of biogenic organic aerosol and its importance for climate N. Rastak et al. https://doi.org/10.1002/2017GL073056]

All aerosol populations in the simulations (including organic-containing aerosol populations) include water as a component, however, water is not considered in the partitioning of organics. They are also separate processes: one is to get organics on the aerosols due to partitioning, another is to grow aerosols with water. We do not take any Henry solubility into account when calculating partitioning, we use the Pankow parameterization (Pankow, 1994) which does not take into account water. For the cloud parameterization we calculate kappa from the chemical composition of the populations. This is not 100% consistent with each other, but it is the state of the art in global climate models. This would certainly be an interesting question to pursue, but is beyond the

scope of this study. As for kappa per VBS bin, we have a constant kappa per VBS bin listed in Table 1 of the manuscript, and the molecular weight of the VBS species are considered constant.

To include this detail, we have included the following in line 85 of our text: "*....we assumed a linear increase of solubility with decreasing volatility (Jimenez et al., 2009). Since we use Pankow type partitioning (Pankow, 1994), water is not considered in the partitioning process. In addition, we do not use different kappa/RH relationships per organic species, which was found to be important for biogenic SOA (Rastak et al., 2017).*"

4. It would seem the crux of the conclusions rests on the above process description and how the ARG parameterization takes that information to predict cloud droplet number. ARG would not capture partitioning through the humidity life-cycle, so please elaborate on the link between partitioning within the VBS model at any given RH to feeding parameters into ARG.

In the model, partitioning occurs before activation as a distinct, not synchronous, process. During activation when we feed information into the ARG scheme, there's no partitioning anymore. As we mentioned in the previous answer, water is not considered, so activated particles, which are essentially cloud droplets now, will not be part of any further partitioning. Since this is a box model, the activated particles are calculated as a diagnostic variable in the model, not a prognostic one. Our next paper will be a global model study, where this link can be explored further. Please also see response to comment 1 from Reviewer #1.

5. The title is certainly a question worth asking. However I wonder whether results from a model sensitivity study that, whilst interesting, rests on a framework that does not apparently capture process level phenomena which would influence results can be used to deliver an answer. Starting with responses to the questions above, I would suggest the following statement requires re-phrasing: '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.' There is no indication that process representation within this study has improved on any previous. Conflicting implications on process combinations restrict this evaluation.

This model captures process level phenomena for aerosol microphysics with consideration of nucleation, condensation, and coagulation, all of which affect the activated number concentration calculated by the model at any given time. The new model has in addition organic aerosol partitioning, a process previously missing from the original version of the model, adding an extra process that affects aerosol microphysics. However, we will need to use the global model coupled with cloud microphysics to look at process level phenomena that would affect clouds following aerosol activation. We have also re-phrased the statement above with, "*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 process level phenomena in aerosol microphysics, as applied here for the organics partitioning, would affect cloud microphysics in the global atmosphere and its implications for climate.*" in the manuscript.

For the comment that "There is no indication that process representation within this study has improved on any previous. Conflicting implications on process combinations restrict this evaluation.", we would like to point out that this study isn't an evaluation of the old or the new model. The partitioning process is quite uncertain and the VBS framework is heavily tuned against certain measurements. There are a lot of degrees of freedom in the system and it cannot be evaluated properly in any process-level study without a chamber simulation designed exactly for that purpose, which this paper is not trying to do. In order to test model skill for its climate implications, the evaluation should be performed on a global scale, which we are doing at the moment and plan to publish soon. We will evaluate whether this would improve the model or not, but it is important to note that partitioning affects microphysics in complex ways that are not easy to estimate without a model simulation, and that's what makes this study valuable.

6. To more accurately represent aerosol- cloud interactions through an attempt to account for organic solubility and volatility, more detail is needed before publication. The alternative, of course, is to not present this as an improved representation but deliver it as an existing model sensitivity study which might be better suited to publication in Geoscientific Model Development

We would like to emphasize that we do not study aerosol-cloud interactions here, only the initial phase of cloud formation. Again, we do not claim that the new model has improved in terms of results, but it is different from that of the original model. And as

mentioned in responses to the questions above, it has certainly improved in terms of processes.

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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

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

concentration and dry particle diameter in our model. The parameterization is also a function of geometric standard deviation, which is constant per population in our model as it did in MATRIX (Bauer et al., 2008), as well as a function of aerosol composition and hygroscopicity, as mentioned in the model description, for which we assume a linear increase of solubility with decreasing volatility. The hygroscopicity of the aerosol populations changes with time, as the internal mixing of aerosol populations is altered by aerosol microphysics.

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.

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. We will investigate the
222 effects of condensing organics in a global climate model in the future. The results presented here
223 implicate that in the new model, most areas on Earth would experience less CCN on a typical day,
224 but clean environments with above freezing temperatures, or polluted environments on an
225 extremely dry and hot day, would form more CCN under low updraft velocity conditions, as
226 compared to the old model. We expect that implementing the improved box model in the global
227 scale that includes a two moment cloud microphysical scheme (Morrison and Gettelman, 2008;
228 Gettelman and Morrison, 2015) would more accurately represent aerosol-cloud interactions, which
229 will be our focus on a follow up study. Thus it would offer us valuable insights on how the addition
230 of process level phenomena in aerosol microphysics, as applied here for the organics partitioning,
231 would affect cloud microphysics in the global atmosphere and its implications for climate.

232

241   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	$5 \cdot 10^{-7}$
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, $5 \cdot 10^6$
	Primary organics	$5 \cdot 10^{-6}$, $5 \cdot 10^{-5}$, $5 \cdot 10^{-4}$
	Nonvolatile biogenic organics from terpene source	$1 \cdot 10^{-8}$, $5 \cdot 10^{-6}$, $1 \cdot 10^{-5}$
	Black Carbon	10^{-6}, 10^{-5}, 10^{-4}
Emissions of gases [molecules/cm^3]	VOCs (in sets) Alkenes	$5 \cdot 10^2$, $5 \cdot 10^3$, $5 \cdot 10^4$
	Paraffin	$5 \cdot 10^3$, 10^4, $5 \cdot 10^4$
	Terpenes	10^4, 10^5, 10^6
	Isoprene	10^4, 10^5, 50^6
	NO$_x$	10^5, 10^6, 10^7

437 **Table 3. Minimum and maximum of fractional change in average activated number**
438 **concentration over the last 24 hours between the two models with low, medium and high**
439 **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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442 **Table 4. Minimum and maximum of average activated number concentration over the last**
443 **24 hours of MATRIX and MATRIX-VBS with low, medium and high level emissions at**
444 **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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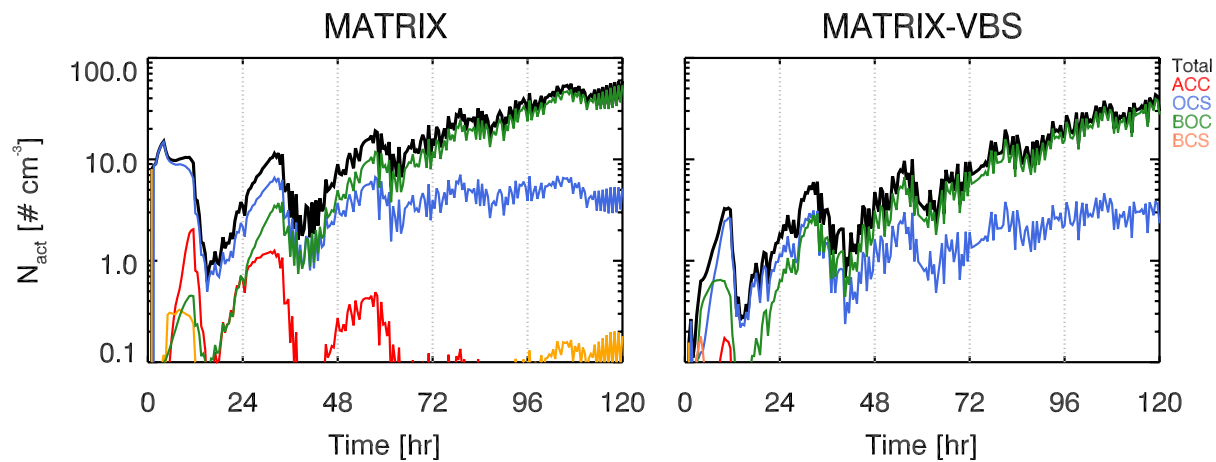
Figure 1. Activated number concentration of aerosol populations (see main text for details) for MATRIX (left) and MATRIX-VBS (right) for 290 K and 40% RH at 30°N latitude with medium emission levels and 0.5 m/s updraft velocity.
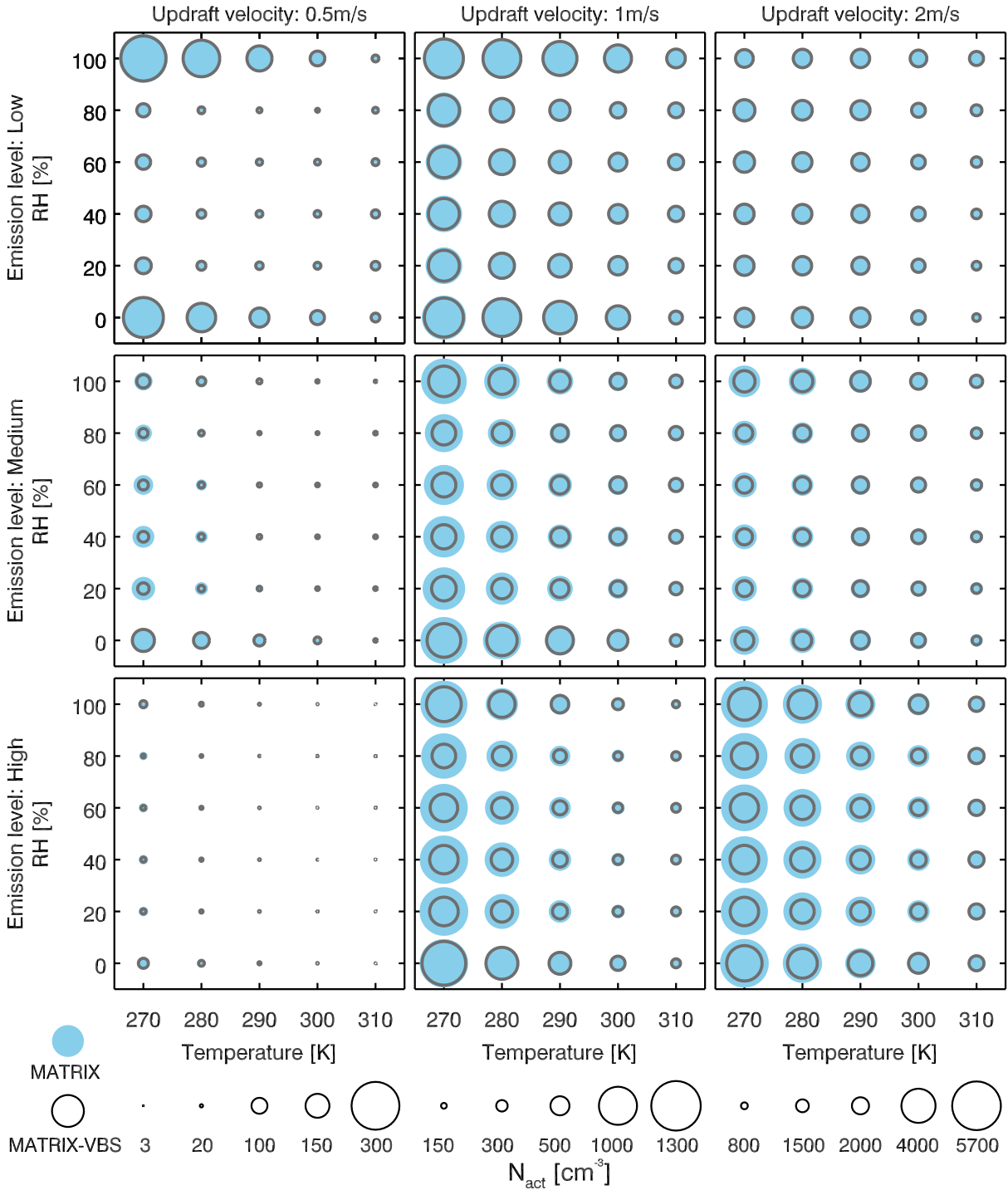
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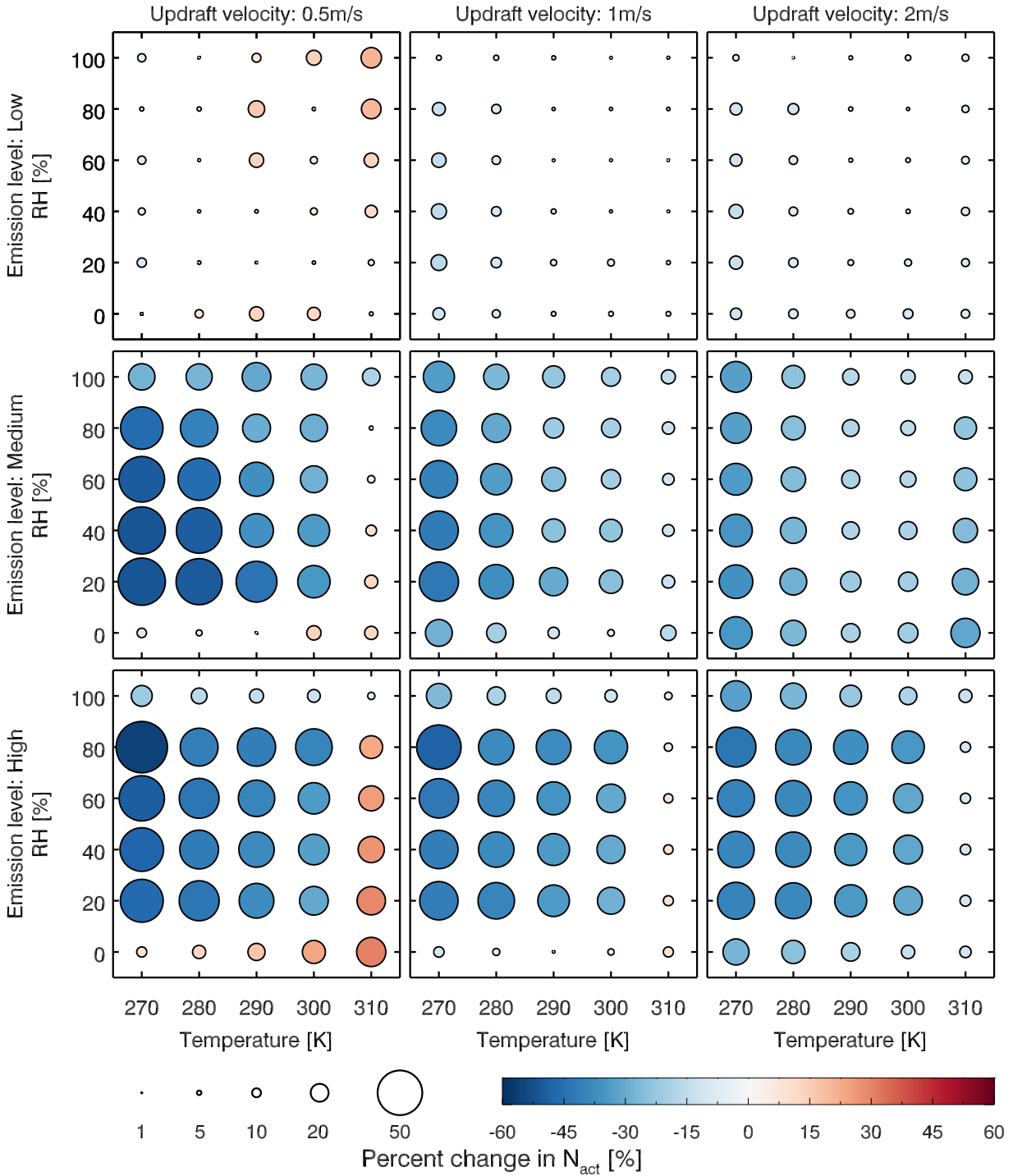
Figure 2. Fractional change of average activated number concentration (size and color of the circles) over the last 24 hours of a 5-day simulation between the two models with low (top row), medium (middle row) and high (bottom row) level emissions at updraft velocities of 0.5 (left column), 1 (middle column) and 2 (right column) m/s.

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456
457 **Figure 3. Average activated number concentration (circle size) during the last 24 hours of a**
458 **5-day simulation in MATRIX and MATRIX-VBS with low (top row), medium (middle row)**
459 **and high (bottom row) emission levels at updraft velocities of 0.5 (left column), 1 (middle**
460 **column) and 2 (right column) m/s. Note difference in scales per column.**

Number concentration by mode

Dry diameter by mode

461
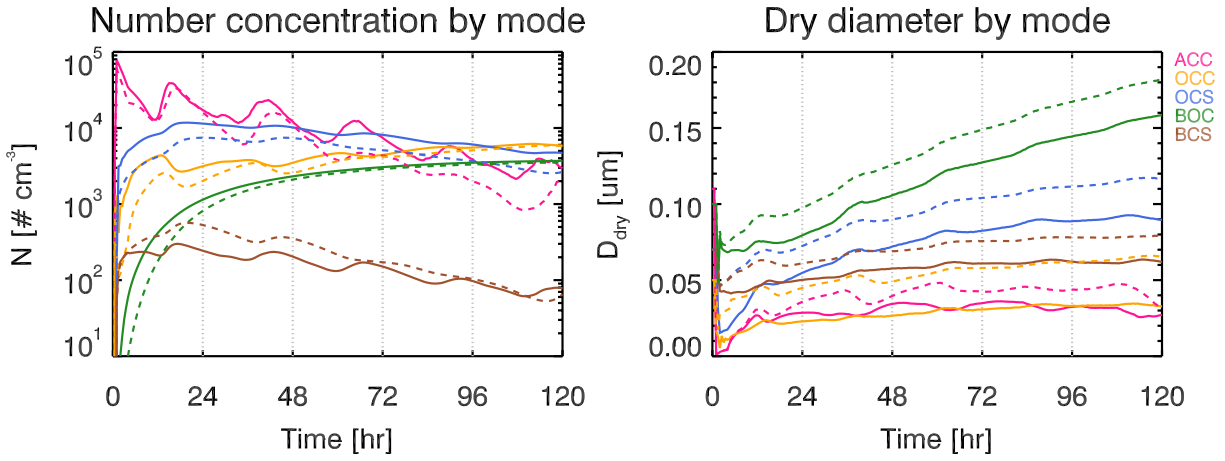
Figure 4. Number concentration (left column) and dry particle diameter (right column) by mode (color lines) for MATRIX (dashed lines) and MATRIX-VBS (solid lines) for the experiments with the same conditions as Figure 1.