

Interactive comment on “Regime dependence of Aerosol Effects on the Formation and Evolution of Pyro-convective clouds” by D. Chang et al.

MS No.: acp-2014-61

Dear Reviewers,

We would like to thank you for the valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below (line numbers refer to the new version of manuscript).

Reply to Anonymous Referee #2

Authors have addressed my comments adequately, except one erroneous reference.

Wang et al., ACP, 2011 should be:

Wang, Y., Q. Wan, W. Meng, F. Liao, H. Tan, R. Zhang, Long-term impacts of aerosols on precipitation and lightning over the Pearl River Delta megacity area in China, Atmo. Chem. Phys., 11(23), 12421-12436, (2011)

Response: Thanks for the correction. We have corrected this reference. Please see Line 356.

Reply to Anonymous Referee #5

Here is a list of suggestions for minor revision/technical correction (page and line numbers are based on the document acp-2014-61-author_response-version1.pdf)

1. Page7Line153: change “research” to “studies”, or revise the sentence to correct the grammar error.

Response: Accepted. We change “research” to “studies”. Please see Line 153.

2. please resolve the conflict in the descriptions of updraft velocities in pyro-convective clouds at Page7Line176 vs. Page9Line207.

Response: Thanks for the comment. To avoid the conflict, we change the description in Line 207 to be “To cover a wide range of conditions, the updraft velocities range from ca. 0.25 to 20 m s⁻¹ in previous cloud parcel model simulations (Reutter et al., 2009)”. Please see Lines 206-208.

3. Page 8Line196 (and other places applicable): change “changed from ... to...” to “varied between ... and ...”

Response: Accepted. We change “ changed from ... to...” to “varied between ... and ...”. Please see Line 196.

4. Page14Line392: remove “absolute”

Response: Accepted. We delete “absolute”. Please see Line 362.

5. Page18Line 474: change “take” to “taking”

Response: Accepted. We change “take” to “taking”. Please see Line 474.

6. Page18Line478: change “processes” to “contributions”

Response: Accepted. We change “processes” to “contributions”. Please see Line 478.

7. Page18Line486: change “as the evolution of the cloud” to “as the cloud evolves”

Response: Accepted. We change “as the evolution of the cloud” to “as the cloud evolves”. Please see Line 486.

8. Page20Line534: change “3 simulation hour” to” 3 simulation hours” or “3-hour simulations”

Response: Accepted. We change “3 simulation hour” to” 3 simulation hours”. Please see Line 534.

9. Page20Line544-545: please fix the problem of “...scheme...is a ...system”

Response: We change this sentence to be “the cloud microphysics itself is a self-regulatory system”. Please see Line 544-545.

10. Page23Line610: remove ‘several’

Response: Accepted. We delete “several”. Please see Line 610.

11. In the conclusion section, please avoid using acronyms or redefine those important ones that may affect the understanding of your conclusions, as you already did for some of them.

Response: Accepted. In the conclusion section, we redefine those acronyms to make the conclusions easier to comprehend. For example, instead of “As N_{CN} and FF increase”, this sentence is revised to be “As aerosol number concentration (N_{CN}) and fire forcing (FF) increase”. Please see Line 668. “The PA provided further insight into...” is modified to be “The process analysis (PA) provided further

insight into...” Please see Line 697. For the other places, the acronyms are replaced by the full name or redefined. Please see the Conclusion part.

12. Figure 23: it is not specifically discussed in the main text; to deliver a full message, it is better to be adequately described in the figure caption.

Response: Accepted. We add more description in the figure caption, which is “Figure 23. Overview of the research approaches on multi-scale cloud initialization and development. The aerosol-cloud interaction at the microphysical scale, i.e., cloud condensation nuclei (CCN) activation, has been well characterized by the Köhler theory (Köhler, 1936) and by a series of extended equations (Shulman et al., 1996; Kulmala et al., 1997; Laaksonen et al., 1998). When we upscale the activation of a single aerosol particle to aerosol populations at the cloud base, the impact of aerosols on the number of activated CCN still appears simple and can be well described (i.e., the three generic regimes of CCN activation). When considering full microphysics and the larger temporal and spatial scales of a single pyro-convective cloud, the performance of ensemble simulations shows the regime dependence of aerosol effects on the pyro-convective cloud formation and evolution.” Please see Fig. 23.

14 **Abstract**

15 A recent parcel model study (Reutter et al., 2009) showed three deterministic regimes of
16 initial cloud droplet formation, characterized by different ratios of aerosol concentrations
17 (N_{CN}) to updraft velocities. This analysis, however, did not reveal how these regimes
18 evolve during the subsequent cloud development. To address this issue, we employed the
19 Active Tracer High Resolution Atmospheric Model (ATHAM) with full microphysics
20 and extended the model simulation from the cloud base to the entire column of a single
21 pyro-convective mixed-phase cloud. A series of 2-D simulations (over 1000) were per-
22 formed over a wide range of N_{CN} and dynamic conditions. The integrated concentration
23 of hydrometeors over the full spatial and temporal scales was used to evaluate the aerosol
24 and dynamic effects. The results show that: (1) the three regimes for cloud condensation
25 nuclei (CCN) activation in the parcel model (namely aerosol-limited, updraft-limited, and
26 transitional regimes) still exist within our simulations, but net production of raindrops
27 and frozen particles occurs mostly within the updraft-limited regime. (2) Generally, ele-
28 vated aerosols enhance the formation of cloud droplets and frozen particles. The response
29 of raindrops and precipitation to aerosols is more complex and can be either positive or
30 negative as a function of aerosol concentrations. The most negative effect was found for
31 values of N_{CN} of ~ 1000 to 3000 cm^{-3} . (3) The nonlinear properties of aerosol-cloud inter-
32 actions challenge the conclusions drawn from limited case studies in terms of their repre-
33 sentativeness, and ensemble studies over a wide range of aerosol concentrations and other
34 influencing factors are strongly recommended for a more robust assessment of the aerosol
35 effects.

36 **Keywords:** pyro-convective clouds, precipitation, ATHAM, updrafts, aerosol

37

38 **1. Introduction**

39 Clouds have a considerable impact on the radiation budget and water cycle of the Earth
40 (IPCC, 2007). Aerosol effects on clouds and precipitation have been suggested to influ-
41 ence the formation, persistence, and ultimate dissipation of clouds and its climate effects
42 (Stevens and Feingold, 2009; Tao et al., 2012), and hence have been studied intensively
43 through cloud-resolving model simulations, analysis of satellite data, and long-term ob-
44 servational data (Tao et al., 2012).

45 However, aerosol effects are still associated with significant uncertainty in light of
46 the seemingly contradictory results from different studies. For instance, several studies
47 have indicated that increasing aerosol concentrations could reduce cloud fraction and in-
48 hibit cloud formation (Albrecht, 1989; Ackerman et al., 2000; Kaufman et al., 2002;
49 Koren et al., 2004), whereas it is suggested that more aerosols can increase the cloud
50 fraction in other studies (Norris, 2001; Kaufman and Koren, 2006; Grandey et al., 2013).
51 Precipitation from stratiform clouds can be inhibited by elevated aerosol concentration
52 (Zhang et al., 2006), while precipitation from convective clouds can be either suppressed
53 or enhanced (Ackerman et al., 2003; Andreae et al., 2004; Altaratz et al., 2008; Lee et al.,
54 2008; Teller and Levin, 2008; Fan et al., 2013; Camponogara et al., 2014). In addition,
55 changing aerosol concentrations have also been found to exert non-monotonic influences
56 (either positive or negative) on a wide range of cloud properties, such as homogeneous
57 freezing (Kay and Wood, 2008), frozen water particles (Saleeby et al., 2009; Seifert et al.,
58 2012), and convection strength (Fan et al., 2009).

59 One explanation for these seemingly contradictory results is that aerosol effects
60 are regime-dependent, which means that it can vary under different meteorological condi-
61 tions (updraft velocity, relative humidity, surface temperature, and wind shear), cloud
62 types, aerosol properties (size distribution and chemical composition) and observational
63 or analysis scales (Levin and Cotton, 2007; Tao et al., 2007; Khain et al., 2008;
64 Rosenfeld et al., 2008; Fan et al., 2009; Khain, 2009; Reutter et al., 2009; McComiskey
65 and Feingold, 2012; Tao et al., 2012). It is thus important to investigate the regime-
66 dependence of aerosol-cloud interactions and to improve the representation of cloud re-

67 gimes in models (Stevens and Feingold, 2009). If we were able to distinguish under
68 which conditions cloud formation is updraft-limited (aerosol-insensitive) as discussed in
69 Reutter et al. (2009), it would have the advantage that in future work one could for many
70 purposes neglect aerosol effects on clouds in areas that are usually updraft limited.

71 Another challenge in evaluating the aerosol effects lies in the nonlinear properties
72 of aerosol-cloud interactions. Most previous research investigated the response of clouds
73 and precipitation to the perturbation of aerosols based on two or several individual sce-
74 narios, by doubling or tripling the number concentration of aerosol particles. This will be
75 fine for the linear dependence. Since aerosol-cloud interaction is a nonlinear process,
76 such method may not reflect the real aerosol effect. An exemplary case is shown in Fig. 1,
77 in which it is clear that the local derivatives (dY/dX) can be different from $\Delta Y/\Delta X$ de-
78 termined by the difference between A and B cases.

79 Biomass burning generates significant amounts of smoke aerosols, and the fires
80 loft soil particles that contain minerals (Pruppacher and Klett, 1997), both of which could
81 serve as effective cloud condensation nuclei (CCN) and ice nuclei (IN) (Hobbs and
82 Locatelli, 1969; Hobbs and Radke, 1969; Kaufman and Fraser, 1997; Sassen and
83 Khvorostyanov, 2008), thereby affecting the formation of clouds and precipitation. As an
84 extreme consequence of biomass burning, pyro-clouds feed directly from the smoke and
85 heat released from fires (Andreae et al., 2004; Luderer, 2007) and provide a good exam-
86 ple with which to study aerosol-cloud interactions (Reutter et al., 2009).

87 By taking the pyro-convective clouds as an example, here we demonstrate the
88 ability of ensemble simulations to determine the regime dependence and resolve the non-
89 linear properties of aerosol-cloud interactions. Aerosol number concentration, updraft ve-
90 locity (represented by the intensity of fire forcing, which triggers updraft velocities), and
91 key parameters of CCN activation (Reutter et al., 2009) are varied to represent a wide
92 range of aerosol and dynamic conditions. In addition to cloud droplets, the responses of
93 precipitable hydrometeors (raindrops, ice, snow, graupel, and hail) were also investigated.
94 For a better understanding of the mechanisms, we employed the process analysis (PA)
95 method, which documents the rate of change in the mass or number concentration of each

96 hydrometeor type caused by a particular process, thereby enabling the determination of
97 the relative importance of the major microphysical processes under different dynamic
98 forcing and aerosol conditions.

99 **2. Design of numerical experiments**

100 **2.1 ATHAM: model and configuration**

101 The Active Tracer High Resolution Atmospheric Model (ATHAM), a non-hydrostatic
102 model, is used here to study cloud formation and evolution in response to changes in up-
103 drafts and aerosol particle concentration. ATHAM was designed initially to investigate
104 high-energy plumes in the atmosphere and applied to simulate volcanic eruptions and fire
105 plumes (Herzog, 1998; Oberhuber et al., 1998). ATHAM has been used to simulate the
106 evolution of pyro-cumulonimbus clouds (pyroCb) caused by a forest fire and shows re-
107 sults consistent with observations (Luderer, 2007).

108 The model comprises eight modules: dynamics, turbulence, cloud microphysics,
109 ash aggregation, gas scavenging, radiation, chemistry, and soil modules (Herzog et al.,
110 1998; Oberhuber et al., 1998; Graf et al., 1999; Herzog et al., 2003). Cloud microphysical
111 interactions are represented by an extended version of the two-moment scheme devel-
112 oped by Seifert and Beheng (2006), which includes the hail modifications by Blahak
113 (2008), and is able to predict the numbers and mass mixing ratios of six classes of hy-
114 drometeors (cloud water, ice crystals, raindrops, snow, graupel, and hail; detailed in Ta-
115 ble 1) and water vapor. It has been validated successfully against a comprehensive spec-
116 tral bin microphysics cloud model (Seifert et al., 2006). The cloud nucleation (CCN acti-
117 vation) module is based on a lookup table derived from parcel model simulations for py-
118 ro-convective clouds (Reutter et al., 2009). The ATHAM model can execute both 2-D
119 and 3-D simulations. Results of this study are mainly based on 2-D simulations.

120 The meteorological conditions were set up to simulate the Chisholm forest fire
121 (Luderer, 2007; Rosenfeld et al., 2007), which is a well-documented case of pyro-
122 convection. All simulations were initialized horizontally homogeneously with radiosonde
123 data from about 200 km south of the fire on 29 May 2001, which is the same as in Luder-

124 er (2007) (Fig. 2). The vertical profiles of the temperature and dew point temperature re-
125 veal a moderate instability in the atmosphere. Open lateral boundaries were used for the
126 model simulations. The means of wind speed and specific humidity were nudged towards
127 the initial profile at the lateral boundaries. The fire forcing was introduced in the middle
128 grid in the bottom layer of the domain, and its intensity remained constant throughout the
129 simulation of each scenario. Each case was run for 3 simulated hours until the clouds
130 were fully developed and had reached steady state.

131 The 2-D simulations were performed at the cross section of the fire front. The
132 simulation domain was set at 85×26 km with 110×100 grid boxes in the x and z direc-
133 tions. The horizontal grid box size at the center of the x direction was equal to 500 m, and
134 it enlarged towards the lateral boundaries due to the stretched grid (Fig. 3). Such a pro-
135 cess scale with resolution of ca. 1 km has been suggested as the appropriate scale at
136 which to characterize processes related to aerosol-cloud interactions (McComiskey and
137 Feingold, 2012). The vertical grid spacing at the surface and the tropopause was set to 50
138 and 150 m, respectively. The lowest vertical level in our simulation was placed 766 m
139 above sea level, corresponding to the lowest elevation of the radiosonde data, which is
140 close to the elevation of Chisholm at about 600 m (ASRD, 2001). The results of the 2-D
141 simulations are presented and discussed in Sect. 3.

142 **2.2 Aerosol particles and fire forcing**

143 Atmospheric aerosol particles affect cloud formation through two pathways, by acting as
144 CCN and as IN. Following the previous study of Reutter et al. (2009), we limited the
145 scope of aerosol-cloud interactions to CCN activation only. So, in this study, changes in
146 N_{CN} do not directly influence frozen hydrometeors by providing IN, but do so indirectly
147 through their impact on CCN activation and subsequent processes.

148 In the 2-D ensemble simulations, 1302 cases ($31 N_{\text{CN}} \times 42$ fire forcing values)
149 were simulated to evaluate the interplay of aerosol concentration and updrafts on the
150 formation of clouds and precipitation. The N_{CN} varied from 200 to $100,000 \text{ cm}^{-3}$. In each
151 case, N_{CN} was prescribed (distributed uniformly across the modeling domain and kept
152 identical throughout the simulation). A similar prescribed approach has been used in pre-

153 vious studies (Seifert et al., 2012; Reutter et al., 2014). Some previous studies have
154 pointed out that a prescribed aerosol scheme overestimates the magnitude of CCN con-
155 centrations compared to a prognostic aerosol scheme, because it lacks a representation of
156 the efficient removal of particles by nucleation scavenging (Wang et al., 2013).

157 As mentioned above, we used the lookup table of Reutter et al. (2009) for the
158 CCN activation. This table is determined for fresh biomass burning aerosols with a hy-
159 groscopicity parameter κ of 0.2 and a log-normal size distribution (a geometric mean di-
160 ameter of 120 nm and a geometric standard deviation of 1.5, Reutter et al. 2009). For the
161 present study, the aerosol characteristics, such as size distribution, chemical composition,
162 hygroscopicity and mixing state are in fact rather unimportant, compared with the order-
163 of-magnitude changes in the aerosol number concentration (Reutter et al., 2009; Karydis
164 et al., 2012). Therefore, the effects of variations in aerosol characteristics were not con-
165 sidered in our study.

166 In all simulations, clouds were triggered by the fire forcing, which was assumed
167 constant during the simulation. The fire forcing intensity varied from 1×10^3 to 3×10^5
168 W m^{-2} . The correlation between the initial fire forcing and corresponding updraft velocity
169 and temperature at the cloud base was probed and is described in Sect. 3.1.

170 In reality, the composition and quantity of biomass burning emissions depend on
171 the moisture content of fuels, combustion conditions, weather situation, and fire behavior
172 (Bytnerowicz et al., 2009). What's more, the biomass burning plumes can in turn change
173 the relative humidity as well. The aerosol particle number concentrations in biomass
174 burning plumes usually exceed 10^4 cm^{-3} , and can be up to $\sim 10^5 \text{ cm}^{-3}$ (Andreae et al., 2004;
175 Reid et al., 2005). In contrast to regular convection, the updraft velocities in pyro-
176 convective clouds are normally larger than 20~30 m s^{-1} (Khain et al., 2005). On the basis
177 of these facts, within our work more attention is paid to situations with higher aerosol
178 concentration ($>10^4 \text{ cm}^{-3}$) and strong updrafts ($>20 \text{ m s}^{-1}$), which are more representative
179 of pyro-convective clouds.

180 **2.3 Process analysis**

181 Cloud properties are subject to several tens of microphysical processes, e.g., cloud drop-
182 let nucleation, autoconversion, freezing, condensation, evaporation, etc. (Seifert and
183 Beheng, 2006). Elevated concentrations of hydrometeors can be caused either by an in-
184 crease in their sources or by a decrease in their sinks. To improve the understanding of
185 the aerosol-cloud interactions, we employed the process analysis (PA) method to quantify
186 the causation of changes in the concentrations of individual hydrometeor classes.

187 In addition to the standard model output (e.g., time and spatial series of mass and
188 number concentrations of hydrometeors, and meteorological output), our PA method ar-
189 chives additional parameters, i.e., the time rate of change of hydrometeors due to individ-
190 ual microphysical processes under different aerosol and fire forcing conditions. Table A1
191 summarizes all the microphysical processes and their acronyms.

192 **2.4 3-D simulations**

193 In addition, we performed a number of 3-D simulations to investigate its difference to 2-
194 D simulations. As the 3-D simulations are computationally expensive, only 99 cases (11
195 $N_{\text{CN}} \times 9$ fire forcing values) were performed. N_{CN} varied from 200 to 100,000 cm^{-3} , while
196 fire forcing varied between 1×10^3 and $8 \times 10^4 \text{ W m}^{-2}$. The size of the model domain was
197 set at $85 \times 65 \times 26 \text{ km}$ with $110 \times 85 \times 100$ grid boxes in the x, y and z directions. For
198 consistency, the grid resolutions in the x and z directions were the same as for 2-D simu-
199 lations. The minimum grid box size in the y direction was set to 100 m. The results of
200 the 3-D simulations are presented and discussed in supplementary material.

201

202 **3. Results and discussion**

203 **3.1 Relationship between updraft velocity, temperature, and fire forcing**

204 Fire forcing does not affect the cloud activation of aerosols directly, but it can affect acti-
205 vation indirectly by triggering strong updraft velocities. Updrafts are of importance in the
206 formation of clouds and precipitation for redistributing energy and moisture. To cover a

207 wide range of conditions, the updraft velocities range from ca. 0.25 to 20 m s⁻¹ in previ-
208 ous cloud parcel model simulations (Reutter et al., 2009), which represent the range
209 found in trade wind cumulus to thunderstorms (Pruppacher and Klett, 1997).

210 The probability distribution function of vertical velocities (w) at cloud base layer
211 under different fire forcing conditions is shown in Fig. 4a. The velocity on top of the in-
212 put fire forcing is usually the largest, and decreases towards the lateral sides. As the char-
213 acteristic velocities, the maximum velocity at cloud base in Fig. 4a, are plotted against
214 the input fire forcing (range of 1×10^3 to 3×10^5 W m⁻², $N_{CN} = 1 \times 10^3$ cm⁻³) in Fig. 4b.
215 The shaded area indicates the variability of estimation over each simulation period. Ac-
216 cording to the figure, w at cloud base varies monotonically from 1.8 to 27 m s⁻¹ as fire
217 forcing increases from 1×10^3 to 3×10^5 W m⁻². The positive relationship suggests that
218 fire forcing could be a good indicator of vertical velocity. Because it is a variable of cen-
219 tral interest to the cloud research community, the maximum vertical velocity is provided
220 along with the fire forcing values as an additional axis in the following plots.

221 Another variable of key meteorological interest is the maximum temperature at
222 cloud base. To clarify how temperature is affected by fire forcing in our simulations, the
223 relationship between fire forcing and the corresponding maximum temperature at cloud
224 base is shown in Fig. 5. As variations in aerosol number concentrations have very little
225 effect on the temperature profile, we show this relationship for only one aerosol concen-
226 tration ($N_{CN}=5,000$ cm⁻³) as an example. Based on Fig. 5, the cloud base temperature in-
227 creases linearly from 7.6 to 16.4 °C, as fire forcing is enhanced from 1×10^3 to 3×10^5 W
228 m⁻². In order to more clearly convey the effect of the heating imposed in the simulation,
229 we have used this linear relationship to add the maximum cloud base temperature as a
230 secondary axis in the figures.

231 Finally, we note that the horizontal wind shear can also affect the convection
232 strength (Fan et al., 2009), which could be investigated in detail in future studies.

233

234 3.2 Aerosol effects and its regime dependence

235 In this section, the tempo-spatial distribution of each hydrometeor type will be briefly
236 presented, followed by the modeled dependency of various hydrometeors on N_{CN} and fire
237 forcing (FF). Note here only the characteristics of dependency are presented, while the
238 underlying mechanisms will be discussed and interpreted in more detail in Sect. 3.3. For
239 an individual hydrometeor type, the averaged concentrations (over the entire domain and
240 simulation period) were used as metrics in our evaluation, and the condensed water
241 reaching the surface was used as a metric for precipitation.

242

243 3.2.1 Cloud droplets

244 Figure 6 shows the temporal evolution of horizontally-averaged mass concentration of
245 cloud droplets (M_{CD}) under the four pairs of FF and N_{CN} conditions. Under weak fire
246 forcing conditions (LU), the formation of cloud droplets usually occurs from 20 min, and
247 concentrates at an altitude of 4-7 km. The duration of cloud droplets usually last for a
248 short period (40~60 min). Under strong fire forcing conditions (HU), the cloud droplets
249 form earlier (around 5 min), and most cloud droplets are located at a height of 5-9 km.
250 Besides, the cloud droplets reach steady state because of the cycling of cloud formation.

251 To investigate the sensitivity of an individual hydrometeor to changes in N_{CN} and
252 FF , we adopted the definition of relative sensitivity $RS_Y(X)$ (of one variable Y against the
253 variable X) as

$$254 \quad RS_Y(X) = \frac{\partial Y / Y}{\partial X / X} = \frac{\partial \ln Y}{\partial \ln X} \quad (1)$$

255 In this study, X is the factor affecting cloud formation, i.e., N_{CN} and FF , and Y is
256 the mass or number concentration of each hydrometeor type (cloud droplets, raindrops, as
257 well as frozen particles). By using a natural logarithmic calculation of the variables (i.e.,
258 X , Y), the percentage change of an individual parameter relative to its magnitude could be
259 reflected better. This logarithmic sensitivity evaluation has been applied commonly in the

260 assessment of aerosol-cloud interactions (Feingold, 2003; McFiggans et al., 2006; Kay
261 and Wood, 2008; Reutter et al., 2009; Sorooshian et al., 2009; Karydis et al., 2012).

262 Figure 7a shows the dependence of cloud water droplets (N_{CD}) on N_{CN} and FF .
263 The shape of the isolines is generally consistent with the regime designations reported by
264 Reutter et al. (2009). Following Reutter et al. (2009), a value of the $RS(N_{CN})$ to $RS(FF)$
265 ratio of 4 or 1/4 was taken as the threshold value to distinguish different regimes (the
266 same criteria were employed for rainwater and frozen water content). Red dashed lines in
267 Fig. 7a indicate the borders between different regimes. This resulted in an aerosol-limited
268 regime in the upper left sector of the panel (N_{CD} is sensitive mainly to N_{CN} and is insensi-
269 tive to fire forcing), an updraft-limited regime in the lower right sector of the panel (N_{CD}
270 displays a linear dependence on FF and a very weak dependence on N_{CN}), and the transi-
271 tional regime along the ridge of the isopleth (FF and N_{CN} play comparable roles in the
272 change of N_{CD}). The regimes of Reutter et al. (2009) are derived from simulations of the
273 cloud parcel model of CCN activation at the cloud base. Our results demonstrate that the
274 general regimes for CCN activation still prevail, even when considering full microphysics
275 and the larger temporal and spatial scales of a single pyro-convective cloud system. Fig-
276 ures 7c and 7d display the sensitivity of N_{CD} to variations in N_{CN} and FF . Note that the
277 low/high aerosol and fire forcing conditions (LA, HA, LU, and HU) in these figures refer
278 to a group of N_{CN}/FF conditions. LU: low updrafts (1,000–7,000 W m⁻²); HU: high up-
279 drafts (75,000–300,000 W m⁻²); LA: low aerosols (200–1,500 cm⁻³); HA: high aerosols
280 (10,000–100,000 cm⁻³). High sensitivities were found for low conditions of N_{CN} and FF .
281 While there are some deviations (which appear to be random numerical noise), in general,
282 as either N_{CN} or FF increases, the impact on the cloud droplet number concentration of
283 further changes to either the variable becomes weaker (Figs. 7c and 7d). The reduced
284 sensitivity of cloud droplets to aerosols can be explained by the buffering effect of the
285 cloud microphysics, so that the response of the cloud system to aerosols is much smaller
286 than would have been expected.

287 Compared with N_{CD} , the cloud mass concentration (M_{CD}) is less sensitive to N_{CN} ,
288 and an aerosol-limited regime cannot be said to exist for M_{CD} (Fig. 7b and 7e). As a result,
289 there are only two regimes indicated by the red dashed line in the contour plot (Fig. 7b):

290 an updraft-limited regime in the lower right sector of the panel, and a transitional regime
291 in the upper sector (an aerosol- and updraft-sensitive regime). The $RS(N_{CN})$ of N_{CD} is on
292 average 10 times higher than that of M_{CD} , independent of the intensity of the FF . As N_{CN}
293 increases, M_{CD} becomes insensitive to the change of N_{CN} . Averaged $RS(FF)$ values over
294 simulated FF ranges for N_{CD} (0.60) and M_{CD} (0.50) are commensurate (Fig. 7d and 7f, re-
295 spectively), which implies that both the number and mass concentrations of cloud drop-
296 lets are very sensitive to updrafts. These results are derived from simulations with persis-
297 tent fire forcing over the modeling period. We have also examined the case in which the
298 fire forcing was shut down after the first half hour of simulation (not shown). The same
299 regimes were found in these simulations, with boundaries in good agreement with the
300 findings presented in this work.

301

302 3.2.2 Raindrops

303 Figure 8 exhibits the temporal evolution of the horizontally-integrated mass concentration
304 of raindrops under four different conditions. Compared with cloud droplets (Fig. 6), the
305 occurrence of raindrops is much later, especially when N_{CN} and fire forcing are in a high
306 level. Only for LULA case, numerous raindrops can be found in a high altitude (5-7 km),
307 while for other cases, most of raindrops are located below 5 km ($\sim 0^\circ\text{C}$).

308 The response of the raindrop number concentration (N_{RD}) to fire forcing and N_{CN}
309 is more complex (Fig. 9a). The impact of FF on N_{RD} is non-monotonic. In general, en-
310 hanced FF leads to an increase in N_{RD} under weak updraft condition ($< \sim 4,000 \text{ W m}^{-2}$),
311 while further increases in FF result in the reduction in N_{RD} . The aerosol influence varies
312 in the course of N_{CN} change. Under low aerosol condition ($< \sim 1,500 \text{ cm}^{-3}$), increased N_{CN}
313 can enhance the production of N_{RD} . Under high aerosol condition ($> \sim 2,000 \text{ cm}^{-3}$), the in-
314 fluence of N_{CN} on N_{RD} is very small.

315 As FF increases in magnitude, the amount of rain produced (M_{RD}) increases (Fig.
316 9b), but the size of raindrops varies because of the complex behavior of the response of
317 the rain drop number (N_{RD}) to FF (Fig. 9a). The aerosol effect is non-monotonic: M_{RD} in-

318 creases with aerosols in the lower range of N_{CN} values ($< \sim 1000 \text{ cm}^{-3}$), but further increas-
319 es in N_{CN} result in a decrease in M_{RD} . Combining with the relative sensitivities (Figs. 9e,
320 and 9f), the influence of FF is much more significant than that of N_{CN} in most cases. For
321 example, the upper left corner (an aerosol-limited regime for N_{CD}) becomes a transitional
322 regime for M_{RD} with $RS(FF)$ of 0.1 and $RS(N_{CN})$ of -0.06 (Fig. 9). High sensitivities of
323 M_{RD} to N_{CN} are found at low N_{CN} conditions, but the sensitivity decreases as N_{CN} increas-
324 es (Fig. 9e). The N_{CN} plays the most negative role in M_{RD} under intermediate N_{CN} condi-
325 tions (N_{CN} of several 1000 cm^{-3}). In contrast to cloud droplet number concentration, an
326 aerosol-limited regime for M_{RD} hardly exists in our simulations (Fig. 9b). The response of
327 the raindrops to aerosols is much weaker than the response of cloud droplets to aerosols.
328 This finding is consistent with the idea of clouds acting as a buffered system formulated
329 by Stevens and Feingold (2009). Detailed analysis of the microphysical buffering pro-
330 cesses will be presented in Sect. 3.3.2.

331

332 **3.2.3 Frozen water contents**

333 Within our microphysical scheme, frozen water contents are grouped into four main clas-
334 ses: ice crystals, snow, graupel, and hail (Seifert and Beheng, 2006). The time evolution
335 of frozen water content in Fig. 10 suggests that the formation of frozen water content
336 usually occurs in a high level (5-9 km for LU case, and 7-13 km for HU case), and the
337 height of base layer and top layer decreases as time goes by. Under LU condition, the ap-
338 pearance of frozen water content is around 35 min, and lasts for ~ 120 min, with the peak
339 concentration around 50~70 min. Under HU condition, the frozen particles form around
340 10 min, and keep in a steady state.

341 Aerosols exert influence on the frozen water contents via the process of ice nucle-
342 ation (*in*), but the processes that convert between the different hydrometeor classes and
343 water vapor play a greater role in changing the concentrations of frozen particles, espe-
344 cially the processes of drop freezing to form ice (*cfi*) and the vapor condensational
345 growth of ice and snow (*vdi* and *vds* respectively). Figure 11 illustrates the percentage
346 mass contributions of the individual frozen hydrometeor classes to the total frozen mass.

347 The percentages of each hydrometeor are calculated based on average values over the en-
348 tire simulation period. Generally, greater concentrations of aerosols result in more snow
349 and less graupel. This is in agreement with previous studies on convective clouds (Seifert
350 et al., 2012; Lee and Feingold, 2013), and can be explained by the suppression of the
351 warm rain processes under high aerosol condition. High N_{CN} delays the conversion of the
352 cloud water to form raindrops, so that more cloud water content can ascend to altitudes
353 with sub-zero temperatures, hence freeze into small frozen particles (Rosenfeld et al.,
354 2008). Other research has suggested that elevated aerosols could increase the concentra-
355 tion of large frozen particles (graupel/hail) in the convective system (Khain et al., 2009;
356 Wang et al., 2011), which was attributed to the competing effects of aerosols on graupel
357 formation. Since graupel is mainly formed by the accretion of supercooled droplets by ice
358 or snow, the smaller but more abundant supercooled drops under polluted conditions
359 could be either favorable or unfavorable for graupel formation. The percentage of ice
360 crystals does not change much, with ice crystals contributing approximately 20% on av-
361 erage to total frozen particle mass (Fig. 11). It is worth noting that stronger FF leads to
362 increasing concentration of hail. But compared to other hydrometeors, its contribution is
363 not important and the relative percentage is very low.

364 The dependence of total frozen particles on FF and N_{CN} is summarized in Fig. 12.
365 With the enhancement in FF and N_{CN} , both the number and mass concentrations of the
366 frozen water particles (N_{FP} and M_{FP} , respectively) increase. High $RS(N_{CN})$ and $RS(FF)$
367 values were found under low N_{CN} and FF conditions (Fig. 12), respectively. As N_{CN} or
368 FF increases, their impact becomes weaker, as indicated by a decreasing RS . According
369 to the ratio of $RS(FF)/RS(N_{CN})$, both N_{FP} and M_{FP} are within the updraft-limited regime.
370 Again, smaller $RS(N_{CN})$ values for M_{FP} , compared with N_{CD} , illustrate the weaker impact
371 of N_{CN} on the production of frozen particles.

372

373 3.2.4 Precipitation rate

374 Surface precipitation rate is a key factor in climate and hydrological processes. Many
375 field measurements, remote sensing studies, and modeling simulations have attempted to

376 evaluate the magnitude of aerosol-induced effects on the surface rainfall rate (Rosenfeld,
377 1999, 2000; Tao et al., 2007; Li et al., 2008; Sorooshian et al., 2009; Tao et al., 2012).
378 Fig. 13a shows the response of surface precipitation rate (averaged over each 3 h simula-
379 tion) to FF and N_{CN} . The response of surface precipitation to these forcings is similar to
380 that of raindrops (Fig. 9b). The FF play a positive role in the precipitation, and $RS(FF)$
381 shows a decreasing trend as FF increases (Fig. 13c).

382 The effect of N_{CN} is more complex. Both positive and negative $RS(N_{CN})$ were
383 found in our study. There are generally two different regimes: a precipitation-invigorated
384 regime and a precipitation-inhibited regime. In the precipitation-invigorated regime (N_{CN}
385 $< \sim 1000 \text{ cm}^{-3}$), an increase in N_{CN} leads to an increase in the precipitation rate, and a re-
386 duction in $RS(N_{CN})$ (Fig. 13b). In the precipitation-inhibited regime ($N_{CN} > \sim 1000 \text{ cm}^{-3}$),
387 aerosols start to reduce the precipitation, which is reflected in a negative $RS(N_{CN})$. Within
388 the precipitation-inhibited regime, there is also an extreme $RS(N_{CN})$ at a value of N_{CN} of a
389 few thousand particles per cm^3 (Fig. 13b). The threshold to distinguish these two regimes
390 is derived from the current simulated pyro-convective clouds. The cumulus cloud investi-
391 gation in Li et al. (2008) also suggested this non-monotonic trend, with the threshold aer-
392 osol value around 3000 cm^{-3} . The existence of threshold N_{CN} in both studies implies that
393 similar cloud types may have a similar regime dependence, of which the exact shape may
394 differ due to difference in the meteorological conditions, aerosol properties, etc.

395 Based on the ensemble studies, we found that individual case studies result in
396 large uncertainties in evaluating the response of precipitation to perturbations, e.g., N_{CN} .
397 Different selections of the parameter space may result in different or even opposite con-
398 clusions. Therefore, our ensemble study over a wide range of parameter space sheds
399 some lights on these debates.

400 Within our simulations, melting of frozen particles is the biggest contributor to
401 precipitation, and the rain rate is well correlated with the melting rate (Fig. 14). For $N_{CN} >$
402 $1,000 \text{ cm}^{-3}$, increasing N_{CN} results in more small frozen particles (i.e., snow) with low fall
403 velocities. These small frozen particles cannot fall into the warm areas and melt efficient-
404 ly, resulting in a reduced melting rate. For $N_{CN} < 1,000 \text{ cm}^{-3}$, the ratio between large and

405 small frozen particles is not sensitive to N_{CN} anymore and the vertical distribution of fro-
406 zen particles becomes important. Increasing N_{CN} leads to earlier formation of frozen par-
407 ticles at low altitude, which evaporate less and result in more rainfall.

408 In the literature, both positive (Tao et al., 2007) and negative (Altaratz et al., 2008)
409 relationship between aerosols and rain rate have been reported in previous case studies.
410 Our simulations suggest that this apparently contradictory phenomenon might be the ex-
411 pression of the same physical processes under different aerosol and dynamic conditions.

412 Regarding the temporal evolution, low N_{CN} results in earlier rainfall (Fig. 15),
413 which is consistent with current understanding, observations (e.g., Rosenfeld, 1999,
414 2000), and modeling evidence (e.g., the convective cumulus cloud study by Li et al.
415 (2008)). Note that the general relationship between precipitation and aerosols described
416 in this study is based on simulations over a period of 3 hours. Simulations for a longer pe-
417 riod should be carried out in future studies to investigate the influence of aerosols on pre-
418 cipitation over longer time scales as in Fan et al. (2013) and Wang et al. (2014).

419

420 **3.3 Process Analysis**

421 In our simulations, the evolution of hydrometeor concentrations is determined by multi-
422 ple microphysical processes. It is often difficult to tell exactly how aerosol particles affect
423 clouds and precipitation. Here we introduce a process analysis method to help understand
424 the aerosol effects.

425 **3.3.1 Clouds**

426 Figure 16 summarizes the contribution of the microphysical processes that act as the
427 main sources (warm color) and sinks (cold color) for cloud droplets under different aero-
428 sol and fire forcing conditions. For N_{CD} , the dominant source term is the cloud nucleation
429 (CCN activation) process, in which aerosols are activated under supersaturated water va-
430 por and form cloud droplets. As cloud nucleation happens mostly at the cloud base and so
431 is not strongly affected by cloud dynamical feedbacks, the response of N_{CD} shows similar

432 regimes to cloud parcel models (Reutter et al., 2009). To help explain the regime designa-
433 tion, we divide N_{CD} into two factors: an ambient aerosol number concentration (N_{CN}) and
434 an activated fraction (N_{CD}/N_{CN}). Given the aerosol size distributions, the N_{CD}/N_{CN} ratio is
435 determined approximately by the critical activation diameter (D_c) above which the aero-
436 sols can be activated into cloud droplets. The D_c is a function of ambient supersaturation.
437 Stronger updrafts result in higher supersaturation, smaller D_c and hence, larger N_{CD}/N_{CN}
438 ratios. Under high updraft conditions ($>15 \text{ m s}^{-1}$), N_{CD}/N_{CN} is already close to unity
439 (Reutter et al., 2009). A further increase in the updraft velocity will still change the su-
440 persaturation and D_c , but it will not significantly influence the N_{CD}/N_{CN} ratios and N_{CD} . In
441 this case, N_{CD} is approximately proportional to N_{CN} .

442 Under weak updrafts, the N_{CD}/N_{CN} ratio is sensitive to ambient supersaturations.
443 In this case, a larger supersaturation induced by stronger updrafts can effectively change
444 the N_{CD}/N_{CN} ratio and thus N_{CD} is sensitive to the updraft velocity. On the other hand, the
445 stronger dependence of N_{CD}/N_{CN} on the supersaturation also changes the role of aerosols.
446 As more aerosols reduce supersaturation, increasing N_{CN} tends to reduce the activated
447 fraction, N_{CD}/N_{CN} . Taking $N_{CN} = 60,000 \text{ cm}^{-3}$ ($FF = 2,000 \text{ W m}^{-2}$), for example, a 10%
448 increase in N_{CN} causes a 4% decrease in N_{CD}/N_{CN} , whereas a 10% decrease in N_{CN} leads
449 to an 8% increase in N_{CD}/N_{CN} . The impact of changing N_{CN} on the N_{CD}/N_{CN} ratio counter-
450 acts partly or mostly the positive effect of N_{CN} on cloud droplet formation.

451 The changes of M_{CD} are influenced mainly by the following sources: (1) the con-
452 densation of water vapor on the present cloud droplets (vdc) and (2) the cloud nucleation
453 process (cn), and by the following sinks: (3) cloud droplet evaporation (cep) and (4) the
454 accretion of cloud droplets (ac), and (5) the freezing of cloud droplets to form cloud ice
455 (cfi), which includes heterogeneous (Seifert and Beheng, 2006) and homogeneous freez-
456 ing processes (Jeffery and Austin, 1997; Cotton and Field, 2002). Concerning their rela-
457 tive contributions, the net change of condensational growth of droplets (vdc) and cloud
458 droplet evaporation (cep) dominates the change of M_{CD} . As N_{CN} increases, the condensa-
459 tion rate (vdc) does not change much, while the evaporation rate (cep) is raised greatly
460 owing to increased surface-to-volume ratio of smaller cloud droplets. Condensation in-
461 creases M_{CD} and evaporation reduces M_{CD} . In our study, the net effects are negative. A

462 similar result has been reported by Khain et al. (2005) for deep convective clouds. They
463 found that high CCN concentrations led to both greater heating and cooling, and that the
464 net convective heating became smaller as CCN increased. However, the cloud nucleation
465 rate is enhanced and the loss of cloud water due to other sinks (*ac* for weak *FF* condition,
466 and *cfi* for strong *FF* condition) decreases at the same time. This leads to an increasing
467 trend in the total cloud water content with the increase in N_{CN} .

468 Concerning the absolute contribution, increasing *FF* enhances the change rate of
469 the conversion of water vapor to the condensed phase (R_{vdc} and R_{cn}), whose effect is
470 straightforward. The processes of autoconversion (*au*) and accretion (*ac*) are the major
471 sinks at weak updrafts. As *FF* increases, the conversion of cloud droplets to frozen parti-
472 cles, especially to ice (the *cfi* process), becomes increasingly important.

473 The contribution of the microphysical processes in each modeling grid could be
474 observed from the pie charts in Fig. 17 (taking HUHA ($w = 27 \text{ m s}^{-1}$; $N_{\text{CN}}=100,000 \text{ cm}^{-3}$)
475 for example, which is representative of the pyro-convective clouds). Each plot shows the
476 vertical cross sections of the averaged change rate of main processes contributing to
477 cloud water content over 30 simulation minutes. Colors within each pie chart reflect the
478 percentage of contributions in each grid. CCN activation usually starts at cloud base, fol-
479 lowed by *vdc* in the center of the cloud. Towards both sides, cloud droplets convert to
480 water vapor via evaporation. It is worth noting that the pie charts only represent the rela-
481 tive importance of each process at individual simulation grid, not the absolute amount.
482 Though there are fewer *vdc*-dominated grids than *cep*-dominated grids, the total cloud
483 formation rate from *vdc* is still similar to or higher than the *cep* processes. At cloud top
484 with sub-freezing temperature, cloud droplets are frozen to ice crystals via homogeneous
485 and heterogeneous nucleation. At the beginning stage of the cloud (30 min), the cloud
486 droplets concentrate at the center of the modeling domain. As the cloud evolves, it starts
487 to expand, and at the same time the margin area dissipates due to the sink processes (i.e.,
488 *cep*, *cfi*, and *ac*).

489 We are aware that the exact process rates may vary depending on the microphysi-
490 cal schemes used in the simulation (Muhlbauer et al., 2010). Therefore, we stress that the

491 process analysis here is based on the Seifert microphysical scheme (Seifert and Beheng,
492 2006). In the future, further observations from laboratory and field measurements are
493 needed to improve the understanding of aerosol-cloud interactions and to better constrain
494 microphysical parametrizations.

495

496 **3.3.2 Rain**

497 Dynamic conditions strongly influence the pathways of rain formation and dissipation.
498 For weak updraft cases, the warm rain processes, i.e., autoconversion (*au*) and accretion
499 (*ac*) play a big role. Together with melting of snow (*smr*) or graupel (*gmr*), they are the
500 main sources for raindrops (Fig. 18). Under this condition, raindrops may appear at alti-
501 tudes as high as 5–7 km (e.g., Fig. 8a). For high updraft cases, strong updrafts deliver
502 cloud droplets to higher freezing altitudes (Fig. 6). The cloud droplets then turn directly
503 into frozen particles (cloud→ice crystals), without formation of raindrops as an interme-
504 diate stage (cloud→rain→larger frozen particles). Most raindrops are formed from melt-
505 ed frozen droplets and consequently, they appear below ~4 km (Figs. 8c, d). The weaker
506 cloud→rain conversion with higher updrafts also influences the conversion of rain to fro-
507 zen particles, and is the reason why the *rrg* process (riming of raindrops to form graupel)
508 becomes relatively less important as *FF* increases under low aerosol condition.

509 The aerosols also modify the pathways of rain formation. Taking weak updraft
510 cases for example, the accretion process (*ac*) dominates the cloud→rain conversion under
511 low aerosol concentrations, but is replaced by autoconversion (*au*) under high aerosol
512 concentrations (Fig. 18b). The reason for this is that *au* is the process that initializes rain
513 formation. Once rain embryos are produced, accretion of cloud droplets by raindrops is
514 triggered and becomes the dominant process of rainwater production, as observed for
515 shallow clouds (Stevens and Seifert, 2008) and stratiform clouds (Wood, 2005). High
516 aerosol loading delays the occurrence of *au*, inhibiting the initialization of rain and the
517 following accretion processes at the early stage (0–100 min). Melted frozen particles are
518 also a major source of raindrops. Under low N_{CN} conditions, most of them form from
519 melted graupel particles, whereas under high N_{CN} condition, melting of snowflakes be-

520 comes more important. This is consistent with the aerosol impact on the relative abun-
521 dance of frozen particles shown in Fig. 11. A higher aerosol concentration leads to a
522 higher fraction of smaller frozen particles (ice crystals and snowflakes). The main differ-
523 ence between low and high updrafts is that cloud conversion is the main source in the
524 former case, whereas in the latter case, melted graupel/snow particles become the main
525 contributors.

526 Figure 19 illustrates the temporal evolution of the contribution of each process at
527 individual simulation grid (HUHA case). As mentioned before, the warm rain process is
528 quite unimportant under strong *FF* condition (Fig. 18b). However, it is observed that the
529 warm rain process is the leading source of raindrops at the beginning stage (60 min). The
530 raindrops formed from *au* and *ac* are relatively small, which can easily evaporate. The
531 melting of frozen particles to form raindrops becomes more significant after ~90 min,
532 which dominates the production of raindrops. As shown in Fig. 19, although the process-
533 es still continue at 180 simulation minutes, the microphysics has already fully developed
534 during this simulation period. Thus our **3 simulation hours** could cover the characteristics
535 of the formation and evolution of the pyro-convective clouds. What is more, it should be
536 paid attention that long-term simulation may conceal some detailed information, leading
537 to the bias in prediction of hydrometeors.

538 The PA clearly demonstrates that aerosols could significantly alter the microphys-
539 ical pathways and their intensities. Although the variation in individual microphysical
540 process is remarkable, the net result of all processes is not obvious and even unsusceptible
541 to aerosol perturbations. This is especially obvious when we consider the aerosol effect
542 on rain water: it is observed that as aerosols is enhanced by a factor of 500, the intensities
543 of the source processes only decrease by a factor of 10; however, there is only a two-fold
544 change in the net rain water content. This implies that **the cloud microphysics itself is a**
545 **self-regulatory system**, which can produce equilibrium and buffers the effect of aerosol
546 disturbance (negative feedback).

547 The sensitivity of raindrops to aerosols mainly depends on autoconversion param-
548 eterization, and the melting processes, etc. All those parameterizations have very large

549 uncertainties, especially with bulk microphysical parameterizations. For example, most of
550 the autoconversion schemes were developed or evaluated for stratocumulus clouds,
551 which may not be appropriate for convective clouds. Based on the simulations during the
552 convective phase of squall-line development, van Lier-Walqui et al. (2012) presented the
553 uncertainty in the microphysical parameterization by the posterior probability density
554 functions (PDFs) of parameters, observations, and microphysical processes. With the
555 purpose to improve the representation of microphysics, it is of significance to quantify
556 the parameterization uncertainty by using observation data to constrain parameterization.

557

558 **3.3.3 Frozen water content**

559 In this section, we only focus on the interactions between liquid water phase and solid
560 water phase. As the selfcollection and internal conversion between different frozen hy-
561 drometeors could also cause the change in number concentration of total frozen particles,
562 the process analysis for its number concentration is not discussed. As shown in Fig. 20,
563 the effect of FF is straightforward, boosting vapor deposition (vdi) and cloud droplet
564 freezing on ice (cfi). The vdi is always the most important pathway for the formation of
565 frozen particles in our simulations, whereas cfi shows comparable contribution in the
566 HULA case. Over a wide range of N_{CN} and updraft velocities, our results have extended
567 and generalized the results of Yin et al. (2005), in which vdi and cfi were suggested as the
568 dominant processes controlling the formation of ice crystals in individual mixed-phase
569 convective clouds. Although snow is the dominant constituent of frozen particle mass
570 (Fig. 11), the condensation of vapor on ice (vdi) rather than on snow is the major pathway
571 for frozen particles. The increase of snow mass is mostly caused by collecting of ice (ics)
572 and ice self-collection (coagulation of ice particles, $iscs$), which are internal conversions
573 not counted as either a source or a sink of frozen water content. The ice crystals used for
574 conversion to snow derive mostly from the vdi process. Increasing FF enhances the up-
575 ward transport of water vapor and liquid water to higher altitudes where frozen particles
576 can be formed effectively through vdi and cfi . On the other hand, stronger FF reduces the
577 residence time of cloud droplets in the warm environment (to form raindrops), which

578 could explain the attenuation of *rrg* (riming of raindrops to form graupel) as fire forcing
579 increases under low aerosol condition.

580 Positive relationship between aerosols and the frozen water content have been
581 demonstrated in Sect. 3.2.3. As shown in Fig. 20, the increase in frozen water content is
582 achieved through the enhancement of the *vdi* process. The condensational growth rate R_{vdi}
583 is a function of the number concentration (N_{ice}) and size (D_{ice}) of ice, together with the
584 ambient supersaturation over ice (S_{ice}). In our simulations, the averaged S_{ice} and D_{ice} are
585 not sensitive to the aerosol disturbance; it is the N_{ice} that has been increased significantly
586 because of elevated aerosol concentrations. Higher N_{ice} provides a larger surface area for
587 water vapor deposition on the existing ice crystals and increases R_{vdi} . Lee and Penner
588 (2010) have suggested similar mechanisms for cirrus clouds, which was based on the
589 double-moment bulk representation of Saleeby and Cotton (2004).

590 The process of the formation and dissipation of frozen water content in the model-
591 ing area is illustrated in Fig. 21. The ice crystals form firstly at a higher height, followed
592 by the snow production at a lower level. Downdraughts in the margin region are caused
593 mainly by evaporation and melting. Massive melting takes place at the late stage (after 90
594 min), when large frozen particles (i.e., graupel) form. This is in agreement with the fact
595 that the raindrops appear at a late stage and at a lower altitude under strong *FF* condition
596 (Figs. 8c and d).

597 As shown aforementioned, drop freezing parameterizations and ice nucleation pa-
598 rameterizations influence frozen water content dramatically, which involve large uncer-
599 tainties. Ice microphysics is significantly more complicated due to the wide variety of ice
600 particle characteristics. On one hand, the intensities of these processes differ greatly
601 among different microphysical schemes. Eidhammer et al. (2009) have compared three
602 different ice nucleation parameterizations, and found that different assumptions could re-
603 sult in similar qualitative conclusions although with distinct absolute values. The parame-
604 terization with observational constraints agrees well with the measurements. On the other
605 hand, van Lier-Walqui et al. (2012) suggested the processes contributing to frozen parti-

606 cles are dependent on both particle size distribution and density parameters. Parameteri-
607 zation improvement based on observations could help to reduce the uncertainties.

608

609 **3.3.4 Contribution of individual microphysical processes**

610 The ATHAM model consists of **tens of** microphysical processes. However, based on the
611 calculation of their relative contributions, only a few processes play dominant roles in
612 regulating the number and mass concentrations of cloud hydrometeors, suggesting a pos-
613 sibility for the simplification of microphysical schemes.

614 For the number concentration of cloud droplets, the cloud nucleation (*cn*) and *cfi*
615 (freezing of cloud droplets to form ice) processes contribute most to its budget, while
616 other processes together account for less than 10%. For the mass concentration, the net
617 change of *vdc* (condensational growth of cloud droplets by deposition) and *cep* (evapora-
618 tion of cloud droplets) processes determines the variations in the cloud water content. The
619 *cfi* process could contribute ~50% of the sink under LAHU condition. Therefore, when
620 we simulate the mass of cloud droplets, four microphysical processes, i.e., *cn*, *vdc*, *cep*,
621 and *cfi*, account for a large fraction of the budget.

622 The dominant processes that contribute ~90% to the raindrop number concentra-
623 tion under specific conditions are autoconversion (*au*), selfcollection (*rsc*), evaporation
624 (*rep*), melting of ice, snow, and graupel (*imr*, *smr*, and *gmr*). For the raindrop mass con-
625 centration, the contribution of three processes accounts for ~90% under most conditions,
626 which are rain evaporation (*rep*), melting of snow and graupel (*smr*, and *gmr*).

627 For the frozen water content, under weak fire forcing condition, *vdi* (condensa-
628 tional growth of ice crystals by deposition) and *sep* (snow evaporation) contribute ~90%
629 of the source and sink respectively. Under strong fire forcing condition, *vdi* and *cfi* to-
630 gether contribute 90% of the source, while *sep* and *gmr* together are the most important
631 sink (90%).

632 These major processes can capture most of the qualitative and quantitative fea-
 633 tures of pyro-convection processes and this complex model can thus be simplified for
 634 many purposes to improve the computational capacity. Comparison between the compre-
 635 hensive model and simplified framework will be performed and validated in future stud-
 636 ies.

637

638 3.4 Uncertainties due to nonlinearity

639 Aerosol-cloud interactions are regarded as nonlinear processes. In this case, the local aer-
 640 osol effects on a cloud relevant parameter Y , i.e., dY/dN_{CN} can be different from
 641 $\Delta Y/\Delta N_{CN}$, the dependence derived from two case studies. Fig. 1 has shown such an ex-
 642 ample: depending on the case selection, a positive (or negative) dY/dN_{CN} can correspond
 643 to a $\Delta Y/\Delta N_{CN}$ of 0. Then the question arises, how much difference can be expected be-
 644 tween dY/dN_{CN} and $\Delta Y/\Delta N_{CN}$? In the following, we take the responses of the precipita-
 645 tion to aerosols as an example to address this issue.

646 Figure 22 shows the statistics of the relative difference between $\Delta Y/\Delta N_{CN}$ and
 647 dY/dN_{CN} under LU and HU conditions, in which Y represents the precipitation rate. As
 648 precipitation is insensitive to aerosols for $N_{CN} > 10,000 \text{ cm}^{-3}$, only the cases with N_{CN} of
 649 $200 \sim 10,000 \text{ cm}^{-3}$ are chosen in the calculation. The relative difference is defined as:

$$650 \quad \text{Relative difference} = \frac{\frac{\Delta Y}{\Delta N_{CN}} - \frac{dY}{dN_{CN}}}{\frac{dY}{dN_{CN}}} \quad (2)$$

651 and $\frac{\Delta Y}{\Delta N_{CN}}$ is calculated as: $\frac{\Delta Y}{\Delta N_{CN}} = \frac{Y(2N_{CN}) - Y(N_{CN})}{2N_{CN} - N_{CN}}$, in which the aerosol

652 effect is determined by the difference between the reference case and that after doubling

653 N_{CN} . $\frac{dY}{dN_{CN}}$ is the derivative of the precipitation rate at each N_{CN} , representing the local

654 dependence of precipitation on N_{CN} .

655 The histograms in Fig. 22 demonstrate that $\frac{\Delta Y}{\Delta N_{CN}}$ can deviate considerably from
656 $\frac{dY}{dN_{CN}}$, not only for the absolute value but also for the sign. Statistically, most of the rela-
657 tive differences are in the range of -3.7~0.9 (the 25th and 75th percentiles respectively,
658 with the average difference of -3.0) under LU condition, while are between -1.5 and 0.04
659 (the 25th and 75th percentiles respectively, with the mean value of 0.02) under HU condi-
660 tion. The fact that individual case studies may not reveal local aerosol effects demon-
661 strates the importance of ensemble studies in determining the real responses of clouds to
662 aerosol perturbations.

663

664 4. Conclusions

665 In this study, the regime dependence of aerosol effects on the formation and evolution of
666 pyro-convective clouds have been studied in detail (Fig. 23). The main conclusions are
667 summarized as follows:

668 (1) As aerosol number concentration (N_{CN}) and fire forcing (FF) increase, the
669 number concentration of cloud droplets increases. There are three distinct regimes for the
670 cloud number concentration: an updraft-limited regime (high relative sensitivity (RS) ra-
671 tio of $RS(FF)/RS(N_{CN})$), an aerosol-limited regime (low $RS(FF)/RS(N_{CN})$ ratio), and a
672 transitional regime (intermediate $RS(FF)/RS(N_{CN})$ ratio), which agrees well with the re-
673 gimes derived from a parcel model (Reutter et al., 2009). The cloud mass concentration
674 is less sensitive to aerosols, and there are two regimes for mass concentration: an updraft-
675 limited regime, and a transitional regime.

676 (2) The production of rain water content (i.e., M_{RD}) was enhanced with increase in
677 updrafts, and the aerosols could either slightly increase M_{RD} with low aerosol concentra-
678 tion or decrease M_{RD} with large aerosol concentration. The aerosol concentration plays a
679 mostly negative role in M_{CD} under intermediate aerosol conditions (aerosol number con-
680 centration of several 1000 cm^{-3}). M_{RD} was generally within an updraft-limited regime, i.e.,

681 M_{RD} was very sensitive to changes in updrafts, but insensitive to aerosol concentrations
682 ($RS(FF)/RS(N_{CN}) > 4$). The aerosol and **updraft** effects on raindrop number concentrations
683 (N_{RD}) are quite complicated; both of them play the non-monotonic role in the N_{RD} .

684 (3) As updrafts and aerosols increase, the domain-averaged number and mass
685 concentrations of frozen particles (N_{FP} and M_{FP} respectively) were enhanced. N_{FP} and M_{FP}
686 were also within the updraft-limited regime, which is characterized by large
687 $RS(FF)/RS(N_{CN})$ ratio. In this regime, N_{FP} and M_{FP} were directly proportional to fire forc-
688 ing, and independent of aerosols.

689 (4) **Larger fire forcing resulted in more precipitation**, whereas the effect of aero-
690 sols on precipitation was complex and could either enhance or suppress the production of
691 precipitation. The suppression on the precipitation is due to the change in the fraction of
692 small frozen particles and total melting rate of frozen particles. The enhancement on the
693 precipitation resulting from increasing N_{CN} under low aerosol condition is a result of
694 changes in the vertical distribution of frozen particles and its evaporation process.

695 (5) In addition, **when aerosol number concentration and fire forcing became too**
696 **large**, their impact became weaker, as indicated by a decreasing **relative sensitivity (RS)**.

697 **The process analysis (PA)** provided further insight into the mechanisms of aero-
698 sol-cloud interactions. By evaluating the contribution of the relevant microphysical pro-
699 cesses to the formation of an individual hydrometeor, the PA revealed the dominant fac-
700 tors responsible for the changes in hydrometeor number and mass. (1) Cloud nucleation
701 (cn) initializes cloud droplet formation and is the major factor that controls the number
702 concentration of cloud droplets. As expected, the increase in cloud droplet mass can be
703 attributed mostly to the condensational growth (vdc). (2) **Under weak fire forcing condi-**
704 **tion**, autoconversion (au) and accretion (ac) are the main sources of rain droplets. **Under**
705 **strong fire forcing condition**, the major source is the melting of frozen particles. (3) For
706 the frozen content, the condensation of water vapor on existing ice crystals (vdi) is the
707 most important contributor. In addition to CCN activation, the PA also highlights the im-
708 portance of other microphysical processes in regulating cloud evolution, which is worthy
709 of further scrutiny. By identifying the contribution from individual processes, PA may al-

710 so provide an opportunity for the simplification of microphysical schemes. For example,
711 out of 24 microphysical processes that are directly related to the budget of cloud droplets
712 and raindrops, over 90% of the mass and number changes are attributed to only 10 pro-
713 cesses.

714 While the general trend is clear, the inclusion of nonlinear (dynamic and micro-
715 physical) processes leads to a complex and unstable response of clouds to aerosol pertur-
716 bations. This applies to the response of all hydrometeors and precipitation, as indicated
717 by the large standard deviation of **relative sensitivities** in Figs. 7, 9, 12 and 13. This
718 should also hold when variations in other parameters (e.g., meteorological conditions) are
719 introduced. Compared with our results, the **relative sensitivities** derived from cloud parcel
720 modeling are much smoother (Fig. 8 in Reutter et al. (2009)). The difference is probably
721 caused by complex interactions between cloud microphysics and dynamics (Khain et al.,
722 2008; Fan et al., 2009). These highly nonlinear processes result in a more unstable and
723 chaotic response of cloud evolution to aerosol and dynamic perturbations. Because of this
724 non-linearity, sensitivities of clouds based on limited case studies may require caveats,
725 because they may not be as representative as expected, and so cannot safely be extrapo-
726 lated to conditions outside of the range explored. To understand better the role of aerosols
727 in cloud formation, we recommend high-resolution ensemble sensitivity studies over a
728 wide range of dynamic and aerosol conditions.

729 General current understanding and global modelling studies suggest that for cloud
730 droplet number concentration, the updraft-limited regime may be more characteristic of
731 continental clouds, while the aerosol-limited regime may be more characteristic of marine
732 clouds (e.g., Karydis et al., 2012), suggesting that aerosol effects are generally more im-
733 portant for the marine environment. For this case study of pyro-convective clouds, then,
734 we conclude that aerosol effects on cloud droplet number concentrations and cloud drop-
735 let size are likely more important than effects on precipitation, since precipitation is far
736 less sensitive to **aerosol number concentrations** than to updraft velocity. This is in agree-
737 ment with other studies (e.g., Seifert et al., 2012). A recent long-term convective cloud
738 investigation found that microphysical effects driven by aerosol particles dominate the
739 properties and morphology of deep convective clouds, rather than updraft-related dynam-

740 ics (Fan et al., 2013). Therefore, it must still be determined whether this conclusion ap-
741 plies to other cloud types and over longer time scales.

742 In this study, we demonstrate the performance of ensemble simulations in deter-
743 mining the regime dependence of aerosol effects. The use of such regime dependence re-
744 quires caveats because it may differ for different cloud types, aerosol properties, meteorolo-
745 gical conditions and model configurations (e.g., microphysical schemes, dynamic
746 schemes, dimensionality, etc.; the 3-D results are in the supplementary material).

747 In future work, we intend to extend the current studies to: (1) include other types
748 of clouds with other meteorological or atmospheric conditions; (2) investigate the cloud
749 response over longer timescales (Van Den Heever and Cotton, 2007), as different obser-
750 vational scales could introduce biases in the quantification of aerosol effects on clouds
751 (McComiskey and Feingold, 2012); and (3) evaluate the relative contribution of micro-
752 physical and dynamic effects to cloud buffering effects (Stevens and Feingold, 2009;
753 Seifert et al., 2012).

754

755 **Table A1. Symbols and acronyms for individual microphysical process.**

Symbol	Process
<i>cn</i>	Cloud nucleation
<i>cri/s/g/h</i>	Riming of cloud droplets to form ice crystals/snow/graupel/hail
<i>cfi</i> ⁽¹⁾	Freezing of cloud water to form ice crystals
<i>imc/r</i>	Melting of ice crystals to form cloud water/raindrops
<i>au</i>	Autoconversion of cloud water to form rain
<i>ac</i>	Accretion of cloud water by rain
<i>vdc/i/g/s</i>	Condensational growth of cloud droplets/ice crystals/graupel/snow by vapor deposition
<i>in</i>	Ice nucleation
<i>s/g/hmr</i>	Melting of snow/graupel/hail to form raindrops
<i>rsc</i>	Self-collection of raindrops
<i>rfi/s/g/h</i>	Freezing of raindrops to form ice crystals/snow/graupel/hail
<i>rri/s/g/h</i>	Riming of raindrops to form ice crystals/snow/graupel/hail
<i>c/r/i/s/gep</i>	Evaporation of cloud droplets/raindrops/ice/snow/graupel

756 ⁽¹⁾ Here, *cfi* process includes both heterogeneous and homogeneous freezing processes.

757

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764

765

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Table captions

Table 1. Typical characterizations of the frozen hydrometeor classes.

Table 1. Typical characterizations of the frozen hydrometeor classes.

	Diameter (mm)	Density (g cm ⁻³)	Terminal velocity (m s ⁻¹)	
Cloud ice	Columnar crystals	0.01—1 ⁽¹⁾	0.36—0.7 ⁽²⁾	0.013—0.055 ⁽²⁾
	Plate-like	0.01—1 ⁽¹⁾	~0.9 ⁽¹⁾	0.02—0.06 ⁽²⁾
	Dendrites	0.1—3 ⁽¹⁾	0.3—1.4 ⁽¹⁾	0.25—0.7 ⁽³⁾
Snowflakes	2—5 ⁽¹⁾	0.05—0.89 ⁽¹⁾	0.5—3 ⁽¹⁾	
Graupel	0.5—5 ⁽¹⁾	~0.4 ⁽¹⁾	3—14 ⁽¹⁾	
Hail	5—80 ⁽¹⁾	0.8—0.9 ⁽¹⁾	10—40 ⁽¹⁾	

⁽¹⁾ Pruppacher H.R. (1978).

⁽²⁾ Jayaweera and Ryan (1972).

⁽³⁾ Mitchell and Heymsfield (2005).

Figure captions

Figure 1. Conceptual model of the nonlinear relationship between aerosol concentrations and rain rate (Data are from 2-D simulation results of this work).

Figure 2. Atmospheric sounding launched near Edmonton, Alberta on 29 May 2001. The right black line represents the temperature, and the left black line corresponds to the dew-point temperature. This weather information is from the University of Wyoming Department of Atmospheric Science (<http://weather.uwyo.edu/>).

Figure 3. The 110×100 grid points in the computational domain.

Figure 4. Probability distribution function of vertical velocities (w) at cloud base layer under different fire forcing conditions (a); Relationship between input fire forcing (FF) and induced vertical velocity (w) at cloud base (b). The aerosol concentration is $1,000 \text{ cm}^{-3}$. The shaded area represents the variability of estimation ($\pm 1/2\sigma$).

Figure 5. The correlation of fire forcing and the corresponding maximum temperature at cloud base. The shaded area indicates the variability of estimation ($\pm 1/2\sigma$) over each simulation period.

Figure 6. Time evolution of horizontally-averaged cloud water content (g kg^{-1}) as a function of altitude for four extreme cases, which are referred to as (1) LULA: low updrafts ($2,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (2) LUHA: low updrafts ($2,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$); (3) HULA: high updrafts ($300,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (4) HUHA: high updrafts ($300,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$). Maximum values for each episode are also shown.

Figure 7. Number (a) and mass concentration (b) of cloud droplets calculated as a function of aerosol number concentration (N_{CN}) and updraft velocity (represented by FF). Red dashed lines indicate the borders between different regimes defined by $RS(N_{\text{CN}})/RS(FF)=4$ or $1/4$, respectively. Relative sensitivities with respect to N_{CN} (left) and FF (right) for number (panels (c) and (d)) and mass (panels (e) and (f)) concentration of cloud droplets under different conditions. The thick dashed or solid lines represent the mean values under a given condition, and the shaded areas represent the variability of estimation ($\pm 1/2\sigma$). The acronyms indicate LU: low updrafts ($1,000\text{--}7,000 \text{ W m}^{-2}$); HU: high updrafts ($75,000\text{--}300,000 \text{ W m}^{-2}$); LA: low aerosols ($200\text{--}1,500 \text{ cm}^{-3}$); HA: high aerosols ($10,000\text{--}100,000 \text{ cm}^{-3}$).

Figure 8. Same as Figure 6 but for raindrops.

Figure 9. Same as Figure 7 but for raindrops.

Figure 10. Same as Figure 6 but for the frozen particles.

Figure 11. Contributions of individual frozen hydrometeor to total frozen water content under four extreme conditions which are referred to as (1) LULA: low updrafts ($2,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (2) LUHA: low updrafts ($2,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$); (3) HULA: high updrafts ($300,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (4) HUHA: high updrafts ($300,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$).

Figure 12. Same as Figure 7 but for total frozen particles.

Figure 13. Same as Figure 7 but for surface rain rate.

Figure 14. The correlation of rain rate and the melting rate of the frozen particles. The green diamond points are the averaged rain rate under different aerosol concentrations ($FF=10^5 \text{ W m}^{-2}$). The columns represent the integrated melting rate from individual frozen particles.

Figure 15. Time evolution of surface rain rates for the three aerosol episodes ($N_{\text{CN}} = 200; 1,000; \text{ and } 100,000 \text{ cm}^{-3}$ respectively) under LU (low updrafts, $FF=2,000 \text{ W m}^{-2}$) and HU (high updrafts, $FF=50,000 \text{ W m}^{-2}$) conditions.

Figure 16. The pie charts summarize the relative percentage of the microphysical processes involving cloud droplets as a function of N_{CN} and fire forcing (a: number concentration; b: mass concentration). Colors within each pie chart reflect the contribution of processes under the specific condition. Warm colors denote the sources, while cold colors denote the sinks. The acronyms indicate cn: cloud nucleation; vdc: condensational growth of cloud droplets; cep: evaporation of cloud droplets; au: autoconversion; ac: accretion; cfi: freezing of cloud droplets to form ice crystals, including homogeneous and heterogeneous nucleation; crg/h: riming of cloud droplets to form graupel/hail.

Figure 17. The pie charts summarize the vertical cross sections of the change rate of main microphysical processes contributing to cloud water content. Each pie chart shows the averaged contribution over the past 30 min. Colors within each pie chart reflect the percentage of processes in each grid. The black dashed line is the $0.1 \mu\text{g kg}^{-1}$ isoline of the interstitial aerosol, indicating the shape of smoke plume. The meaning of the acronyms is the same as in Figure 16. Warm colors denote the sources, while cold colors denote the sinks.

Figure 18. Same as Figure 16 but for raindrops. The acronyms indicate au: autoconversion; ac: accretion; i/s/g/hmr: melting of ice/snow/graupel/hail to form raindrops; rsc: self-collection of raindrops; ismr: melting of ice and snow to form raindrops; rfi/h: freezing of raindrops to form ice crystals/hail; rep: raindrop evaporation; rrg: riming of raindrops to form graupel; rris: riming of raindrops to form ice and snow.

Figure 19. Same as Figure 17, but for raindrops.

Figure 20. Same as Figure 16 but for the total frozen water content. The acronyms indicate in: ice nucleation; cfi: freezing of cloud droplets to form ice crystals, including homogeneous and heterogeneous nucleation; rfh: freezing of raindrops to form hail; vdi/s/g: condensational growth of ice crystals/snow/graupel by water vapor; rrg: riming of raindrops to form graupel; i/s/gep: evaporation of ice/snow/graupel; s/g/hmr: melting of snow/graupel/hail to form raindrops.

Figure 21. Same as Figure 17 but for frozen particles.

Figure 22. Histograms of the relative difference between $\frac{\Delta Y}{\Delta N_{CN}}$ and $\frac{dY}{dN_{CN}}$ under LU and HU conditions, where Y here denotes precipitation rate. $\frac{\Delta Y}{\Delta N_{CN}} = \frac{Y(2N_{CN}) - Y(N_{CN})}{2N_{CN} - N_{CN}}$, and $\frac{dY}{dN_{CN}}$ is the derivative of the precipitation rate along the variable N_{CN} .

Figure 23. Overview of the research approaches on multi-scale cloud initialization and development. The aerosol-cloud interaction at the microphysical scale, i.e., cloud condensation nuclei (CCN) activation, has been well characterized by the Köhler theory (Köhler, 1936) and by a series of extended equations (Shulman et al., 1996; Kulmala et al., 1997; Laaksonen et al., 1998). When we upscale the activation of a single aerosol particle to aerosol populations at the cloud base, the impact of aerosols on the number of activated CCN still appears simple and can be well described (i.e., the three generic regimes of CCN activation). When considering full microphysics and the larger temporal and spatial scales of a single pyro-convective cloud, the performance of ensemble simulations shows the regime dependence of aerosol effects on the pyro-convective cloud formation and evolution.

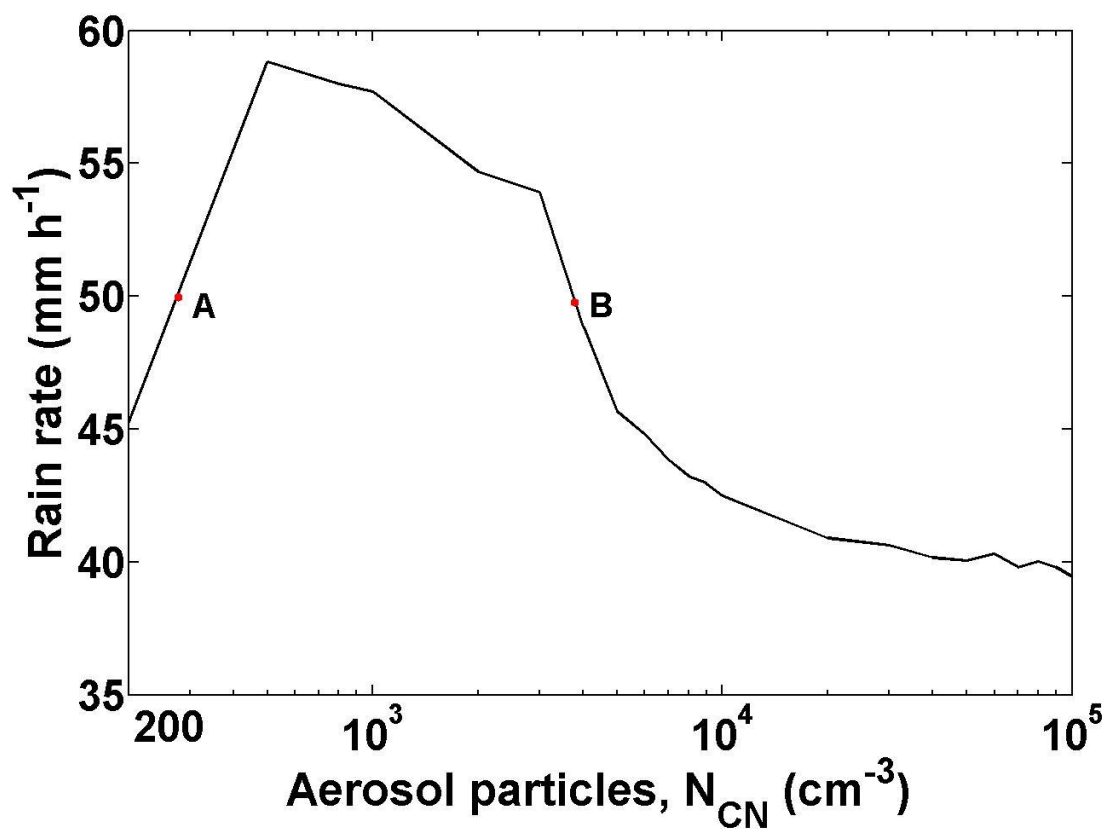


Figure 1. Conceptual model of the nonlinear relationship between aerosol concentrations and rain rate (Data are from 2-D simulation results of this work).

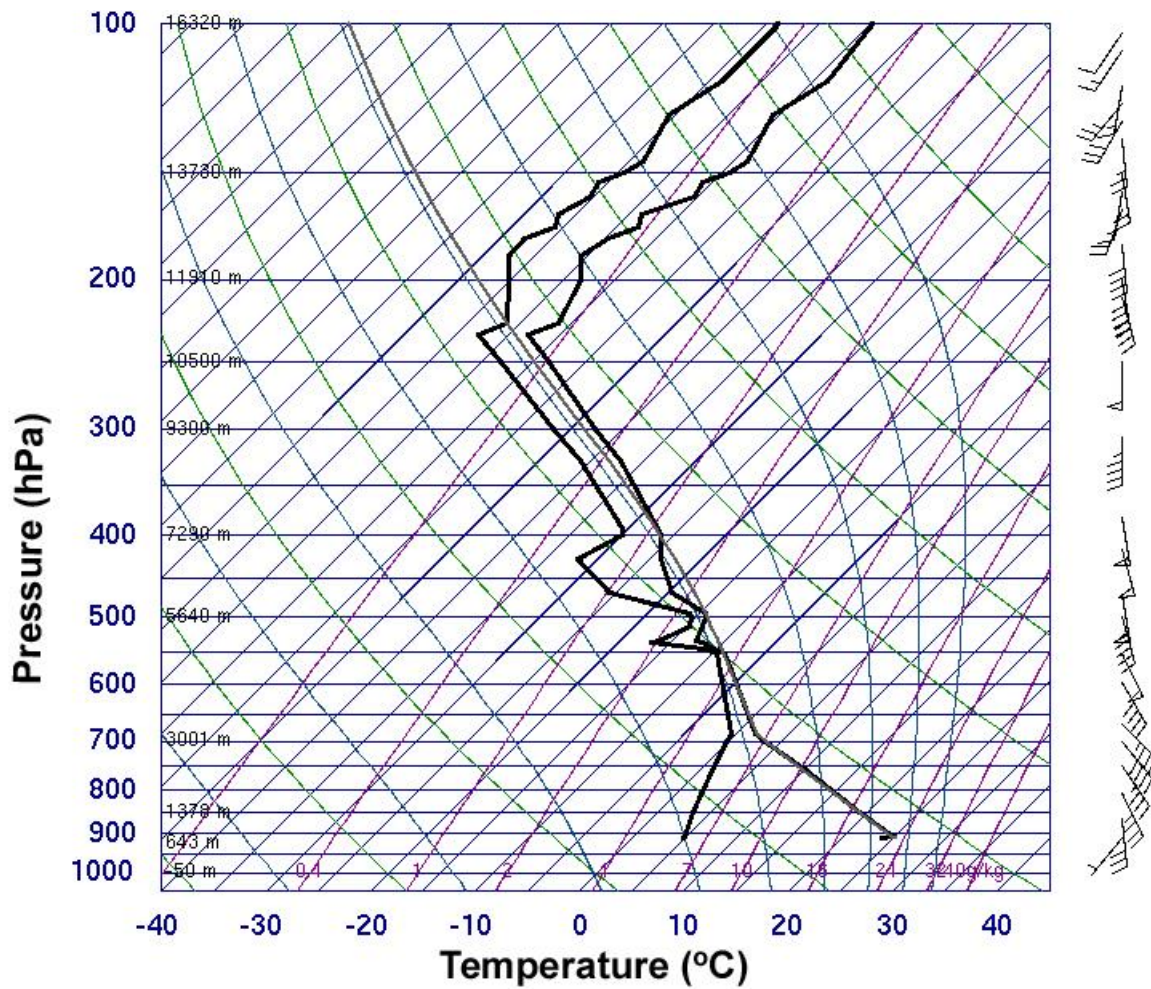
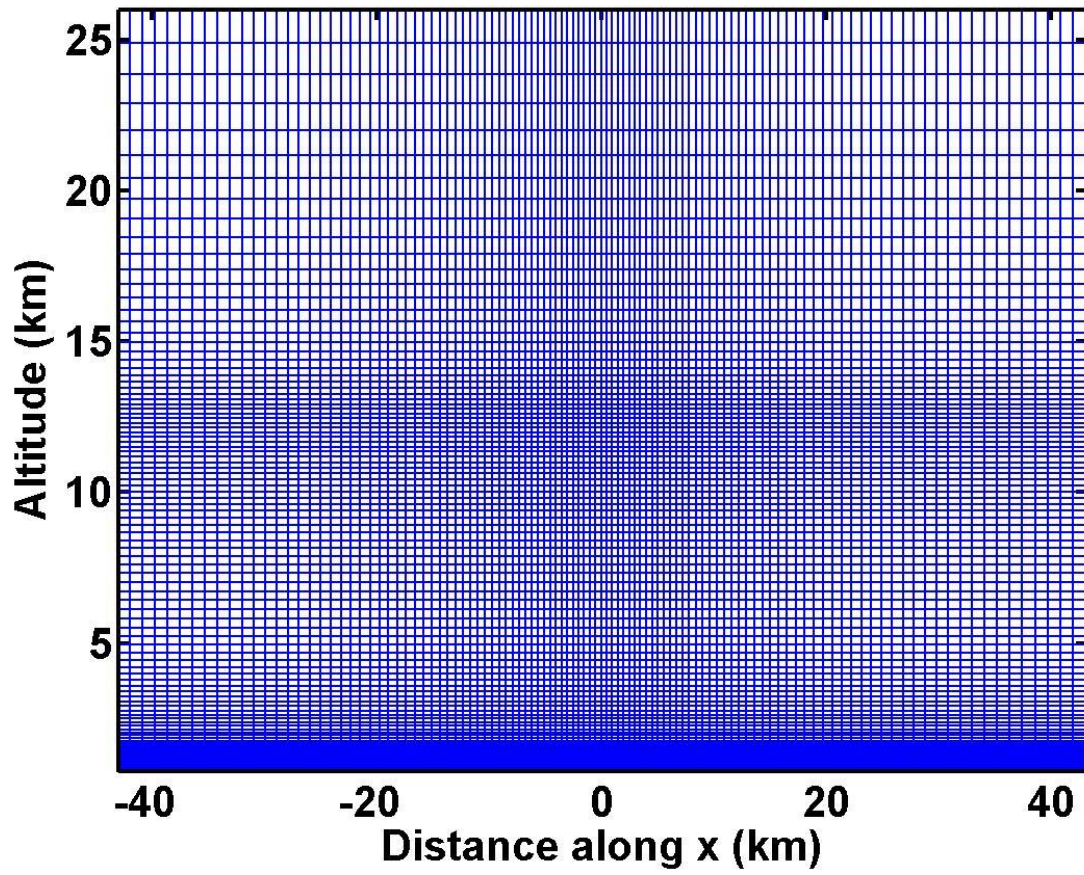


Figure 2. Atmospheric sounding launched near Edmonton, Alberta on 29 May 2001. The right black line represents the temperature, and the left black line corresponds to the dew-point temperature. This weather information is from the University of Wyoming Department of Atmospheric Science (<http://weather.uwyo.edu/>).



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Figure 3. The 110×100 grid points in the computational domain.

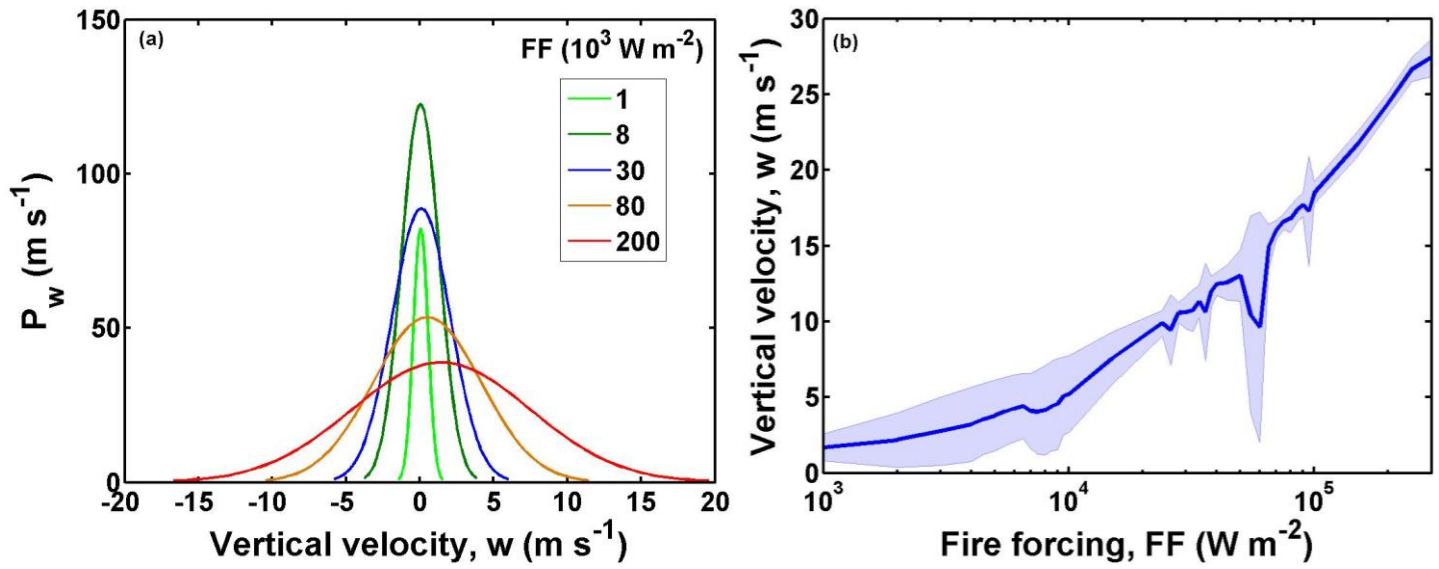


Figure 4. Probability distribution function of vertical velocities (w) at cloud base layer under different fire forcing conditions (a); Relationship between input fire forcing (FF) and induced vertical velocity (w) at cloud base (b). The aerosol concentration is $1,000 \text{ cm}^{-3}$. The shaded area represents the variability of estimation ($\pm 1/2\sigma$).

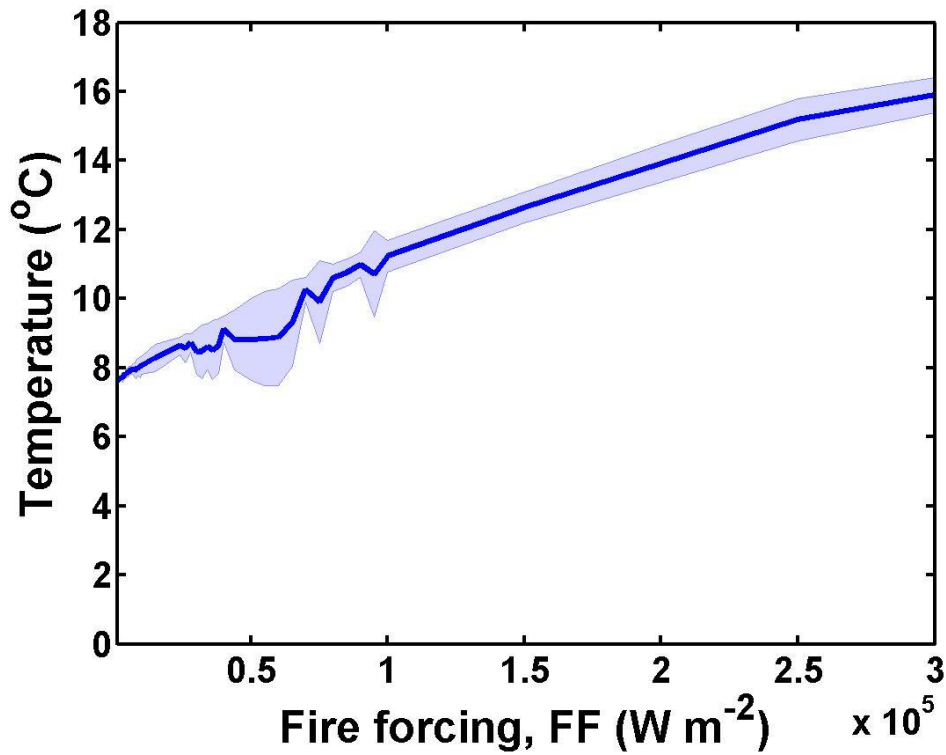
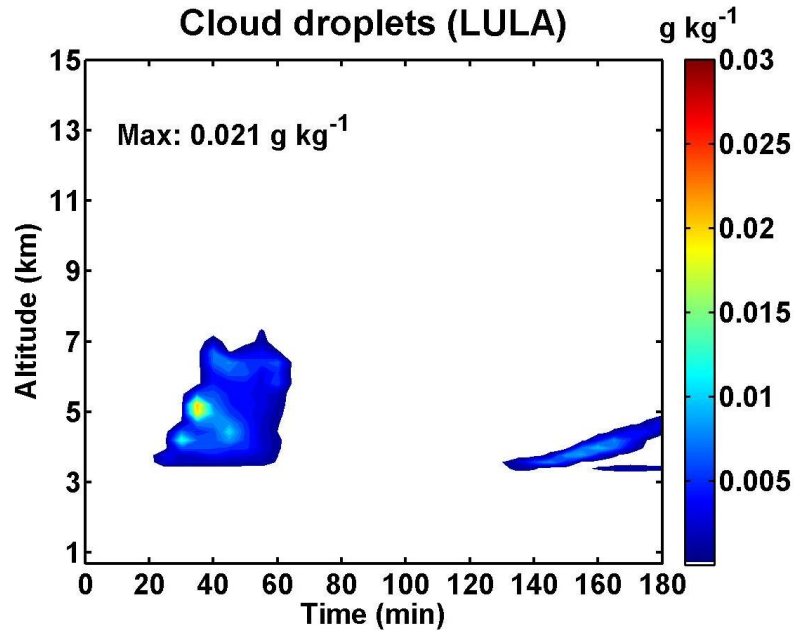
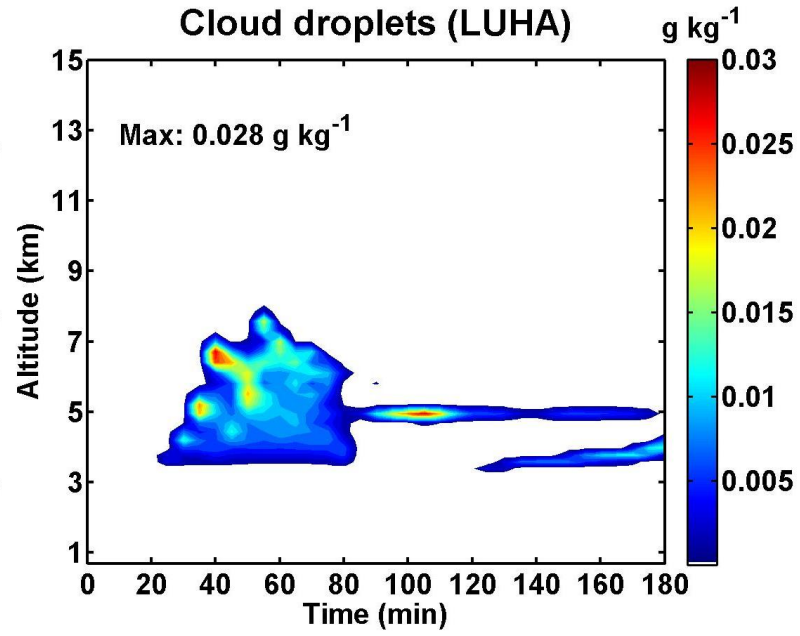


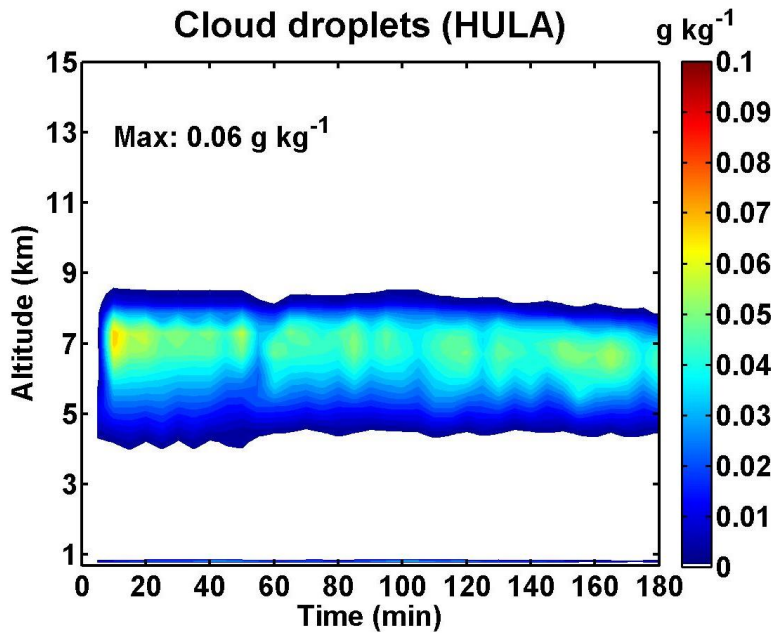
Figure 5. The correlation of fire forcing and the corresponding maximum temperature at cloud base. The shaded area indicates the variability of estimation ($\pm 1/2\sigma$) over each simulation period.



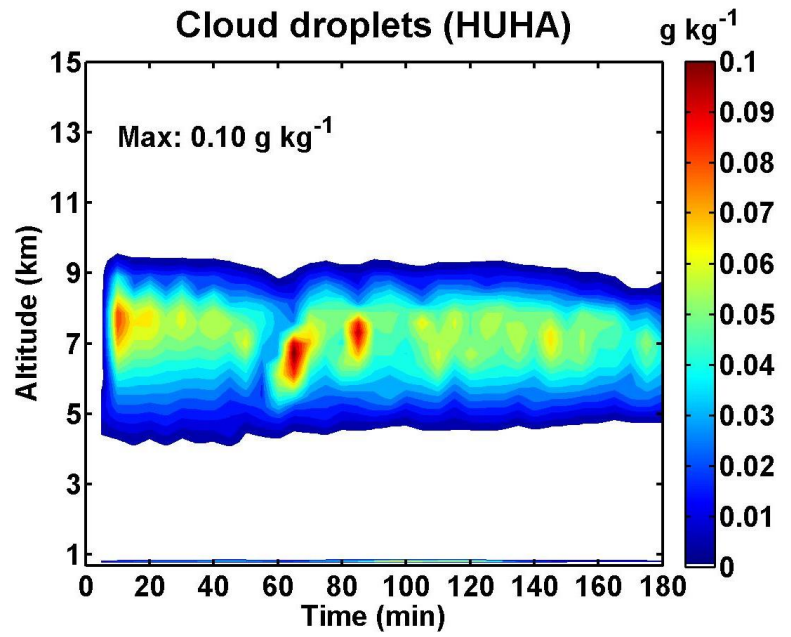
(a)



(b)



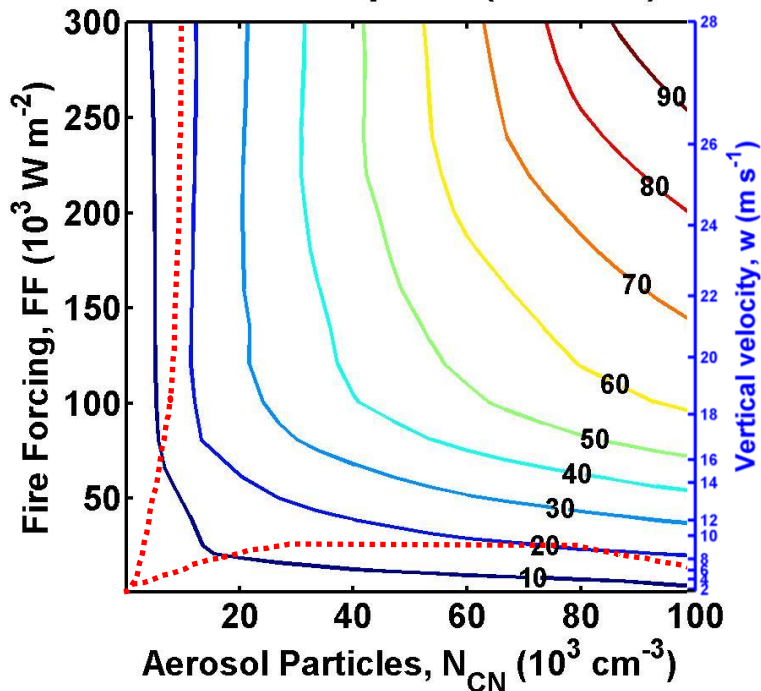
(c)



(d)

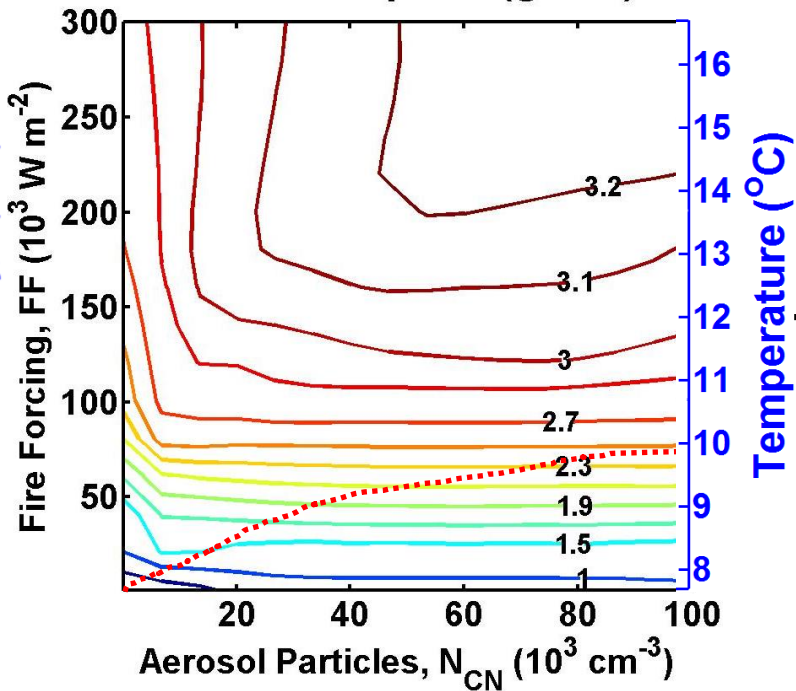
Figure 6. Time evolution of horizontally-averaged cloud water content (g kg^{-1}) as a function of altitude for four extreme cases, which are referred to as (1) LULA: low updrafts ($2,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (2) LUHA: low updrafts ($2,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$); (3) HULA: high updrafts ($300,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (4) HUHA: high updrafts ($300,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$). Maximum values for each episode are also shown.

Cloud droplets (10^6 m^{-3})



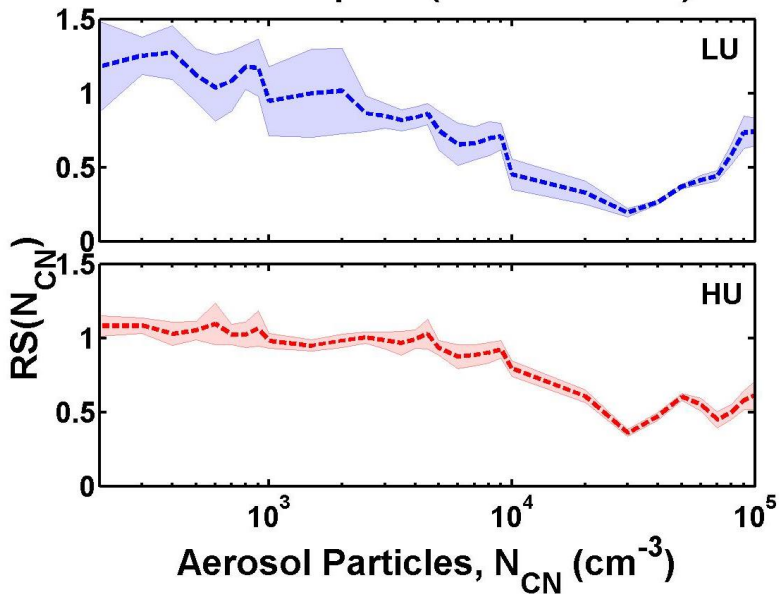
(a)

Cloud droplets (g m^{-3})



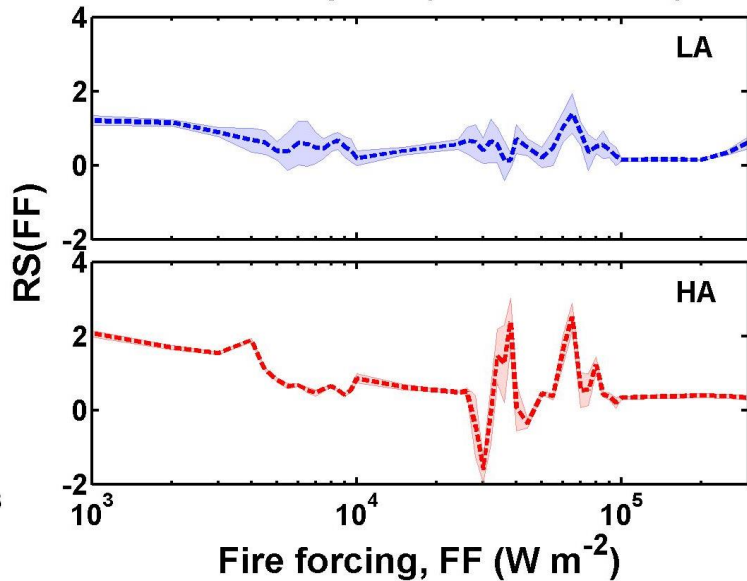
(b)

Cloud droplets (number conc.)



(c)

Cloud droplets (number conc.)



(d)

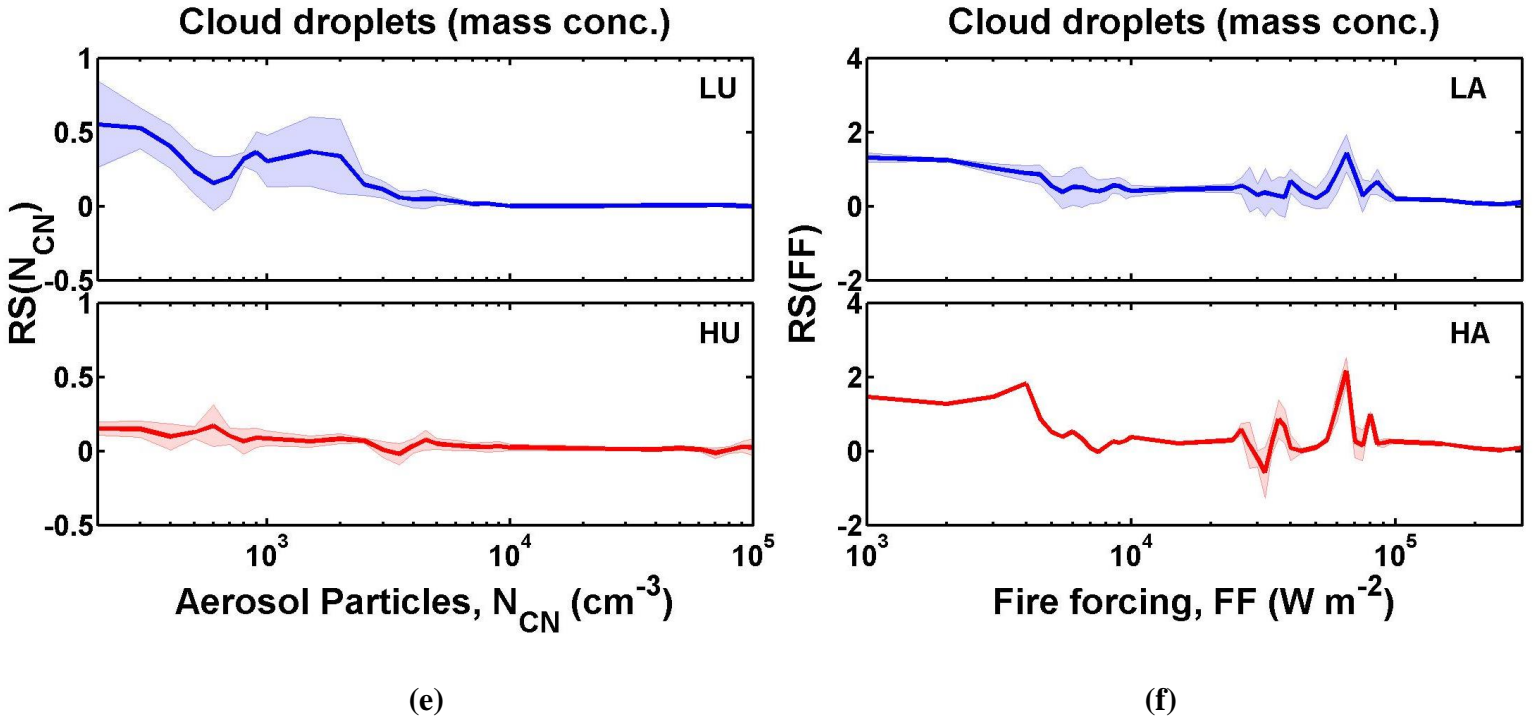
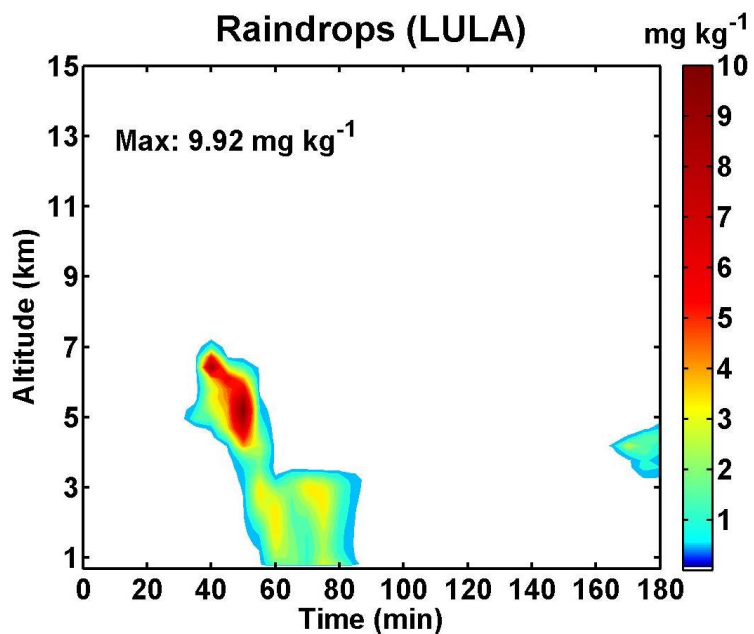
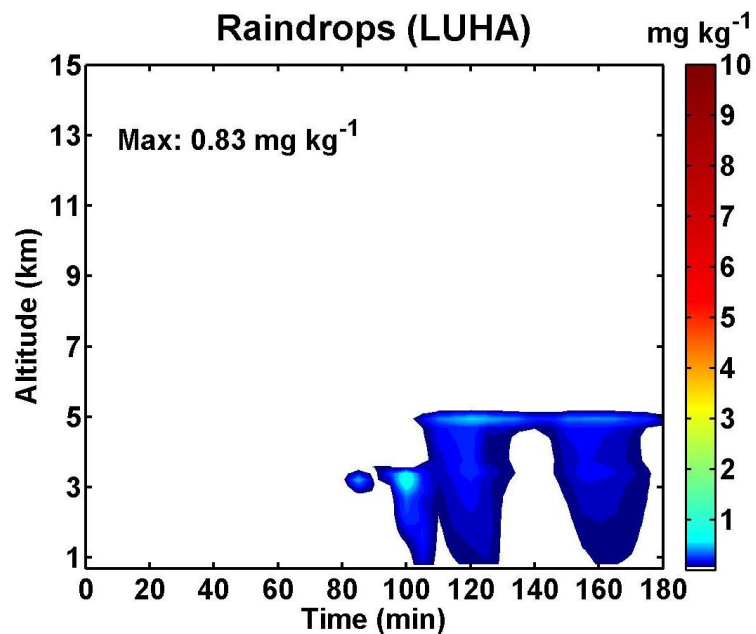


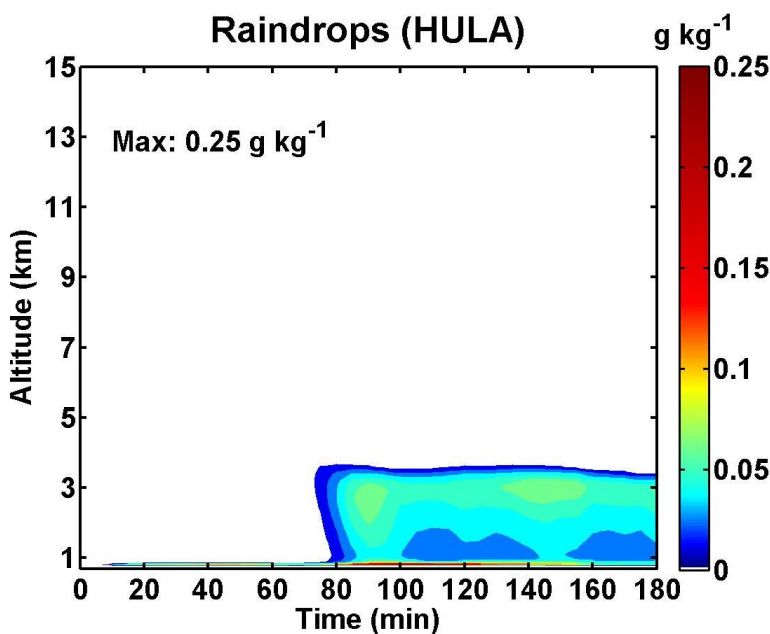
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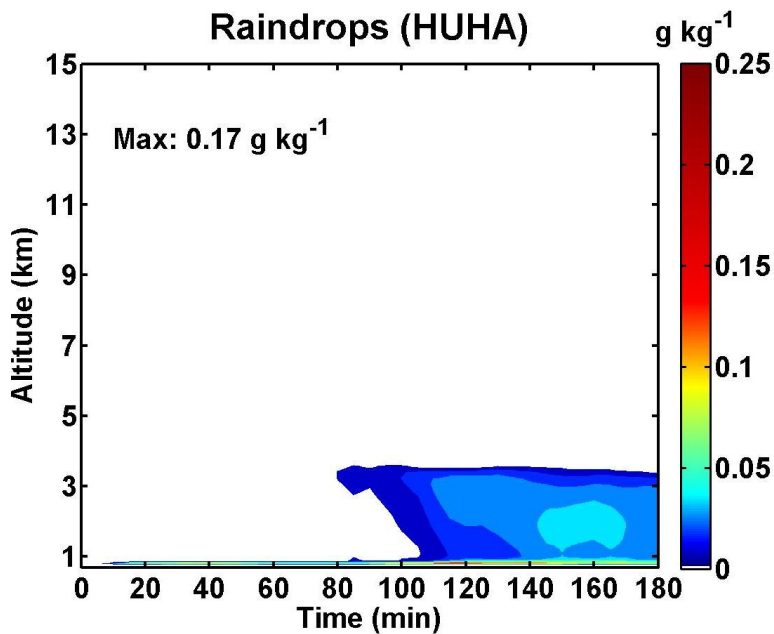
(a)



(b)

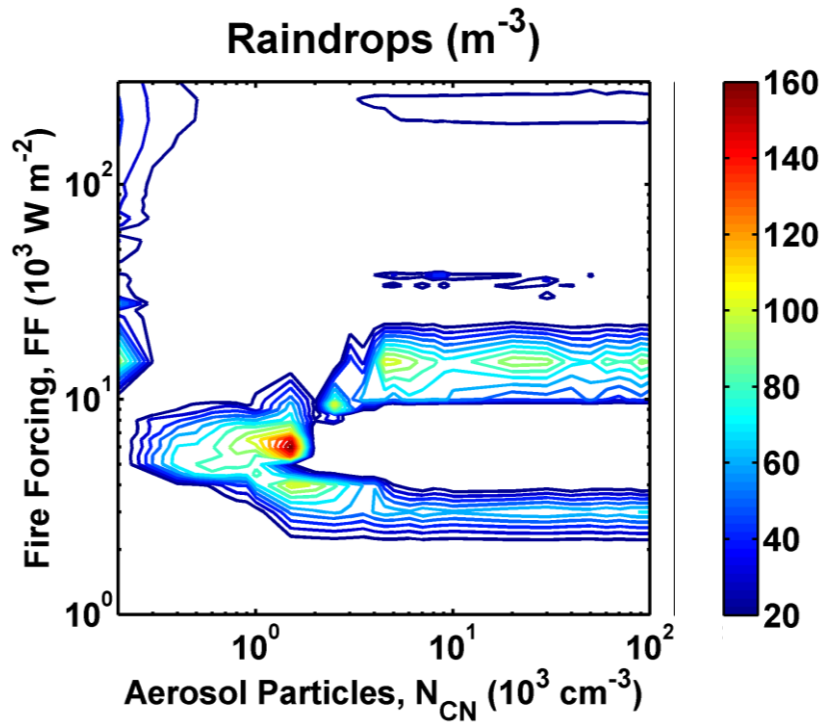


(c)

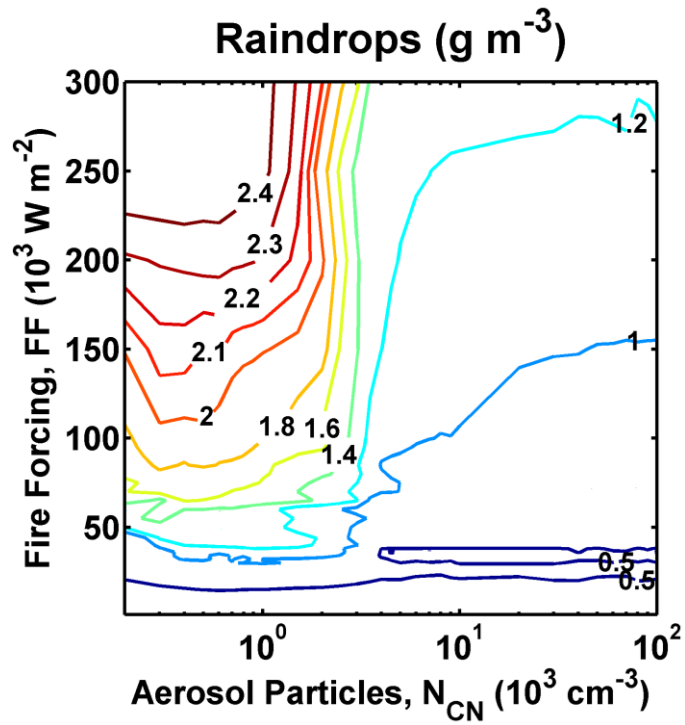


(d)

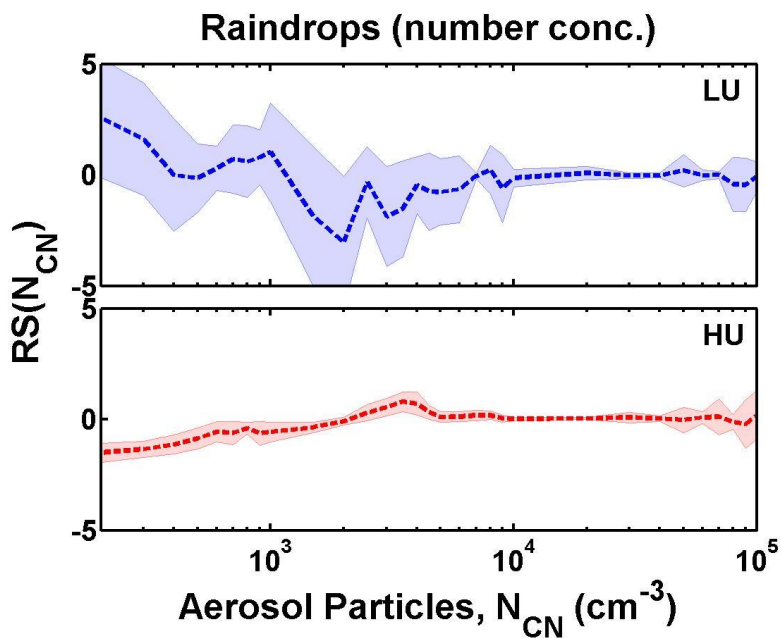
Figure 8. Same as Figure 6 but for raindrops.



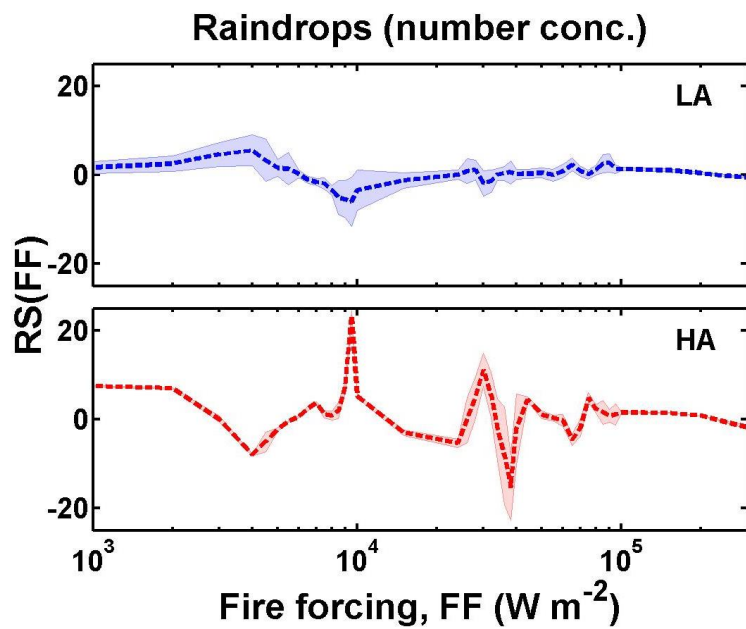
(a)



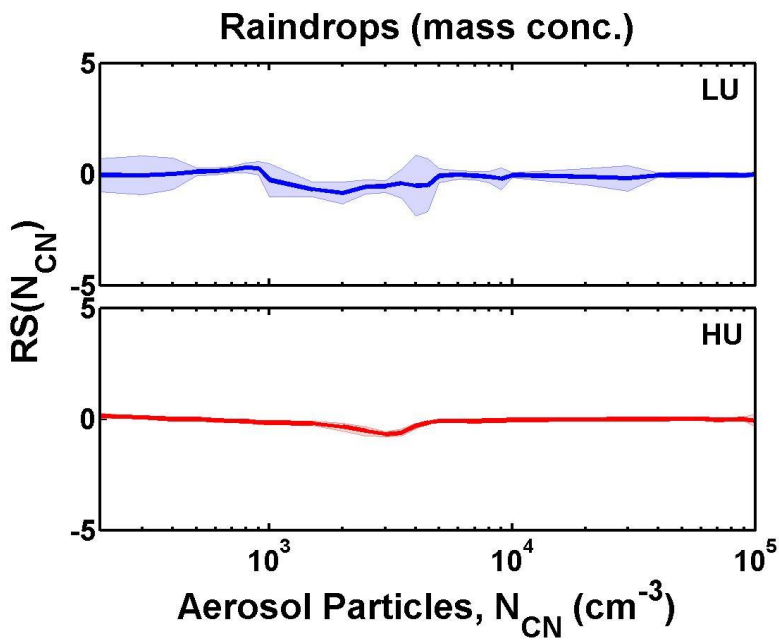
(b)



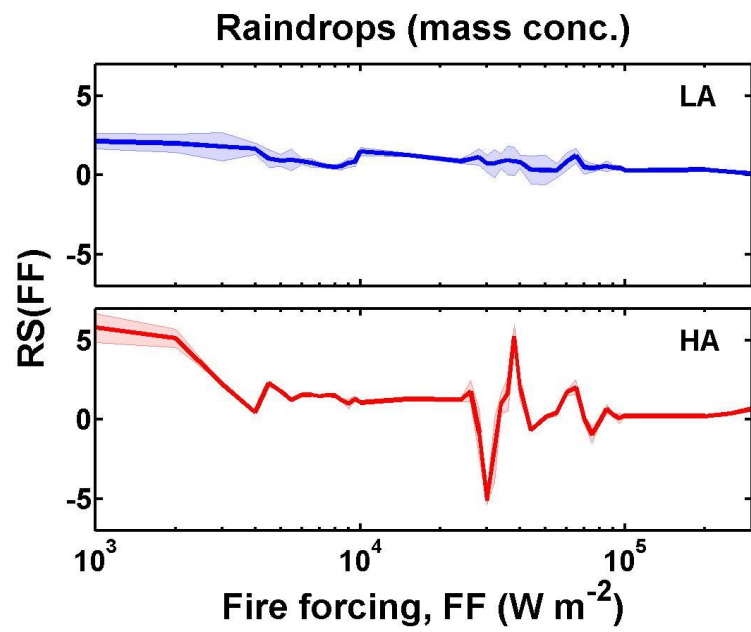
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(d)



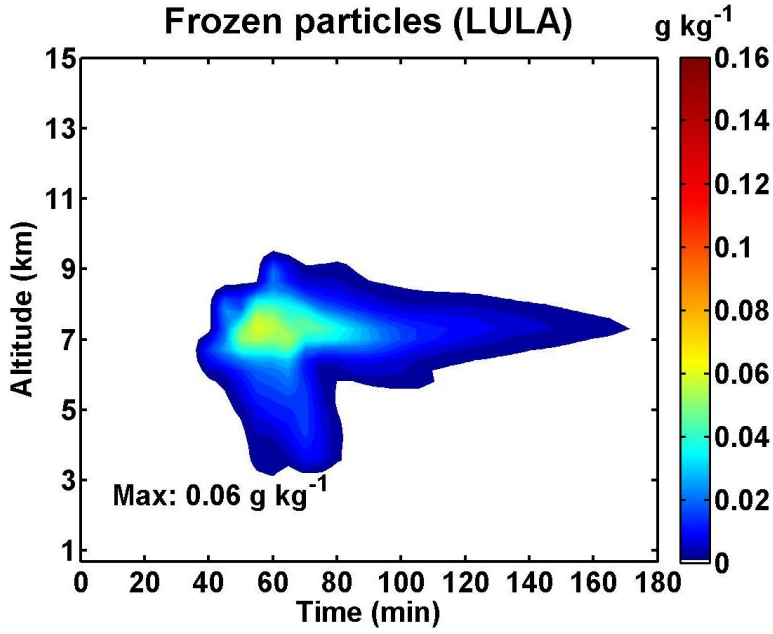
(e)



(f)

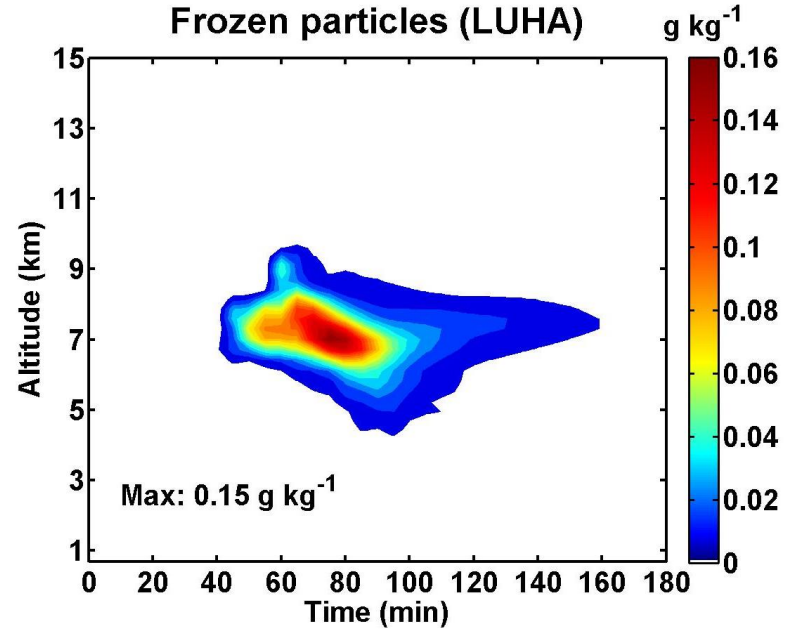
Figure 9. Same as Figure 7 but for raindrops.

Frozen particles (LULA)



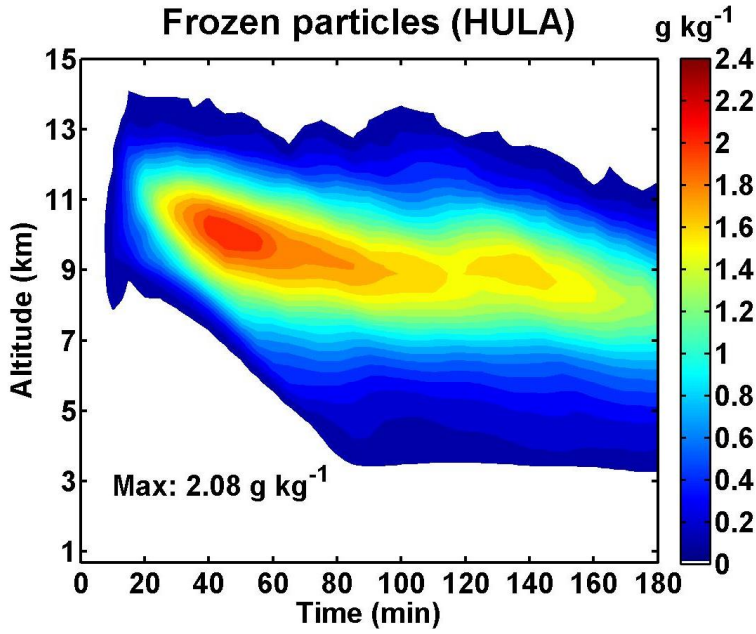
(a)

Frozen particles (LUHA)



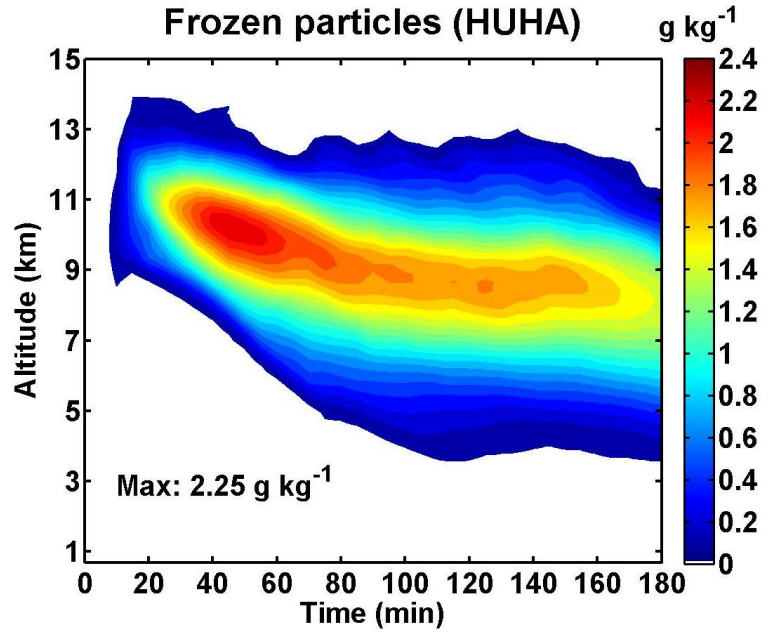
(b)

Frozen particles (HULA)



(c)

Frozen particles (HUHA)



(d)

Figure 10. Same as Figure 6 but for the frozen particles.

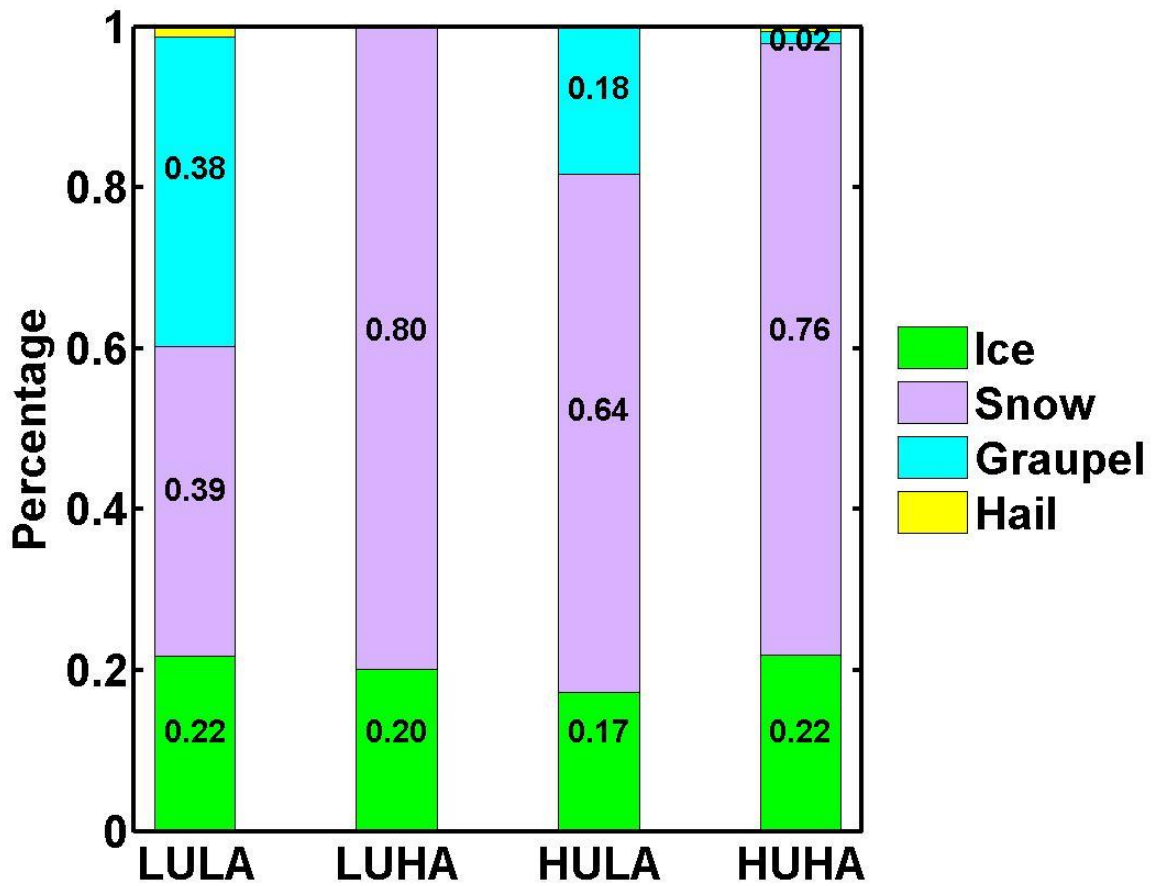
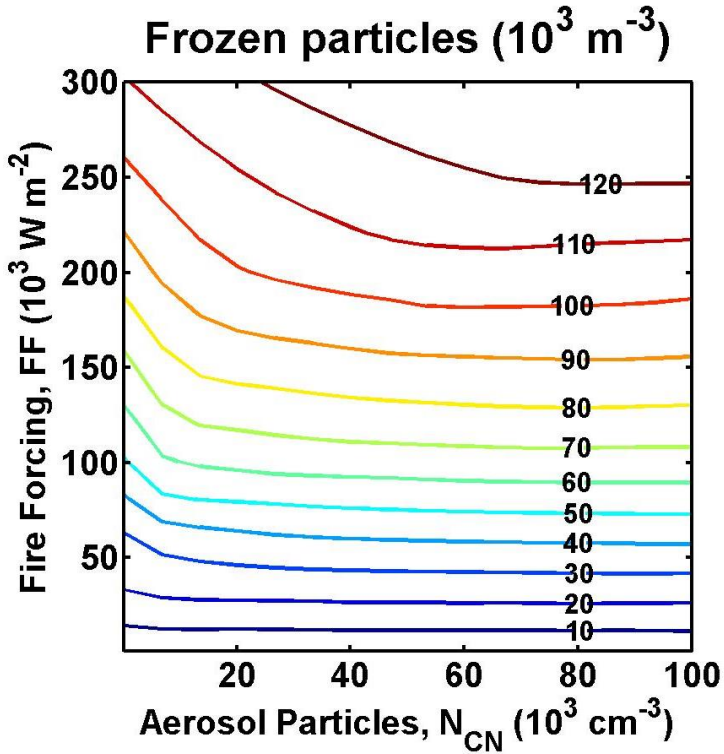
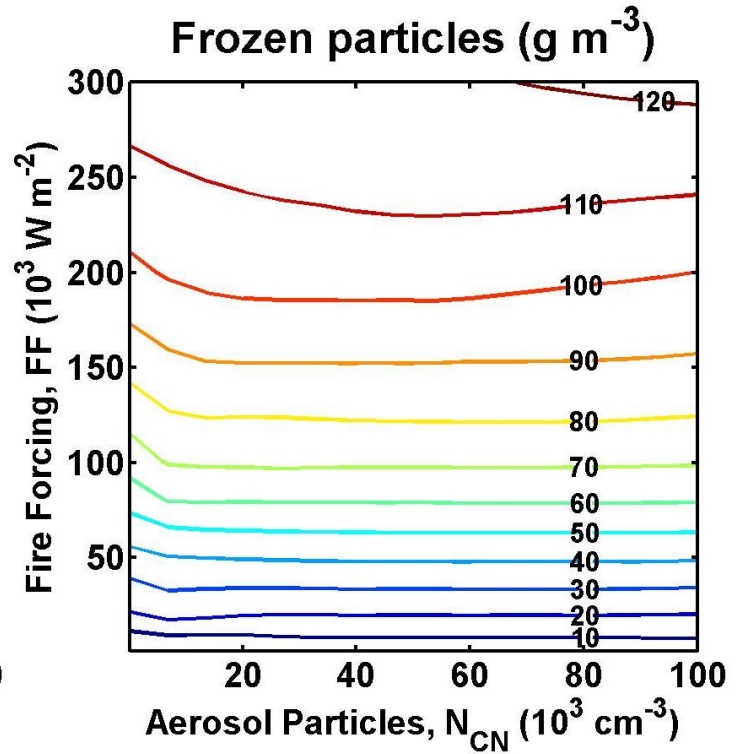


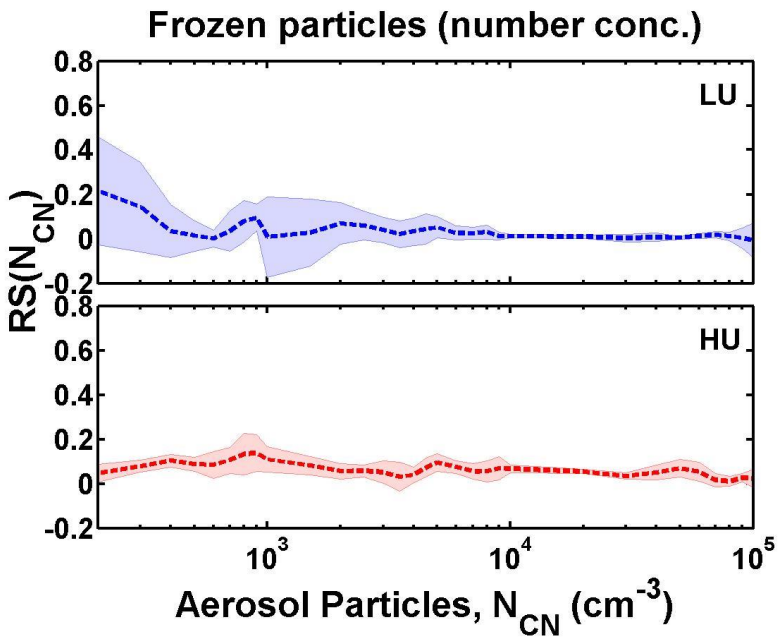
Figure 11. Contributions of individual frozen hydrometeor to total frozen water content under four extreme conditions which are referred to as (1) LULA: low updrafts ($2,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (2) LUHA: low updrafts ($2,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$); (3) HULA: high updrafts ($300,000 \text{ W m}^{-2}$) and low aerosols (200 cm^{-3}); (4) HUHA: high updrafts ($300,000 \text{ W m}^{-2}$) and high aerosols ($100,000 \text{ cm}^{-3}$).



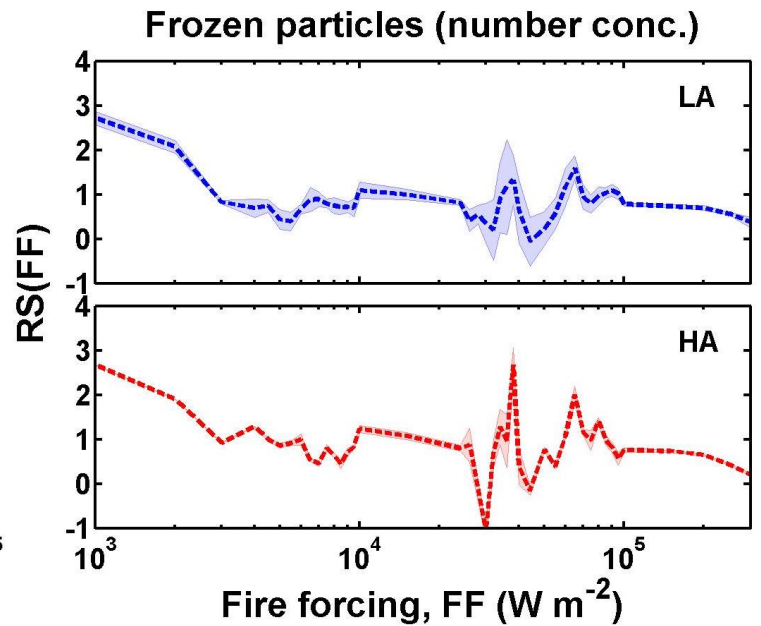
(a)



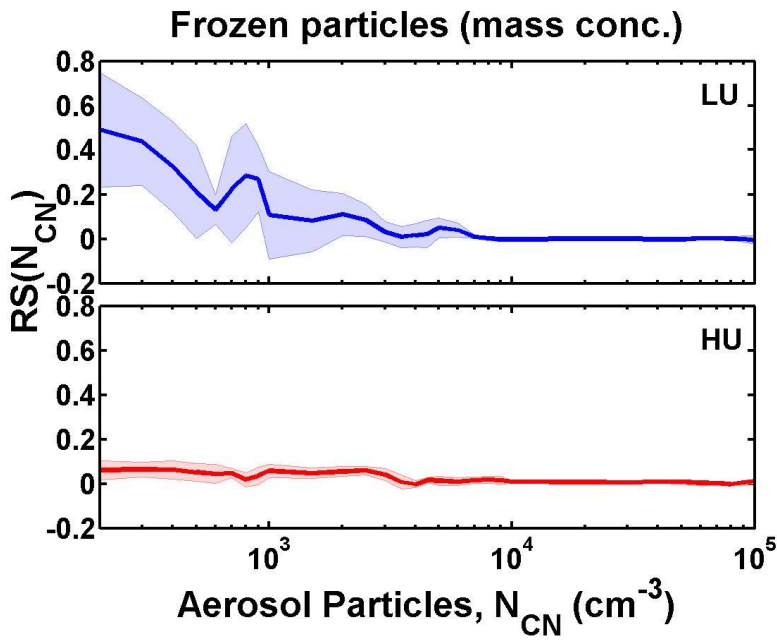
(b)



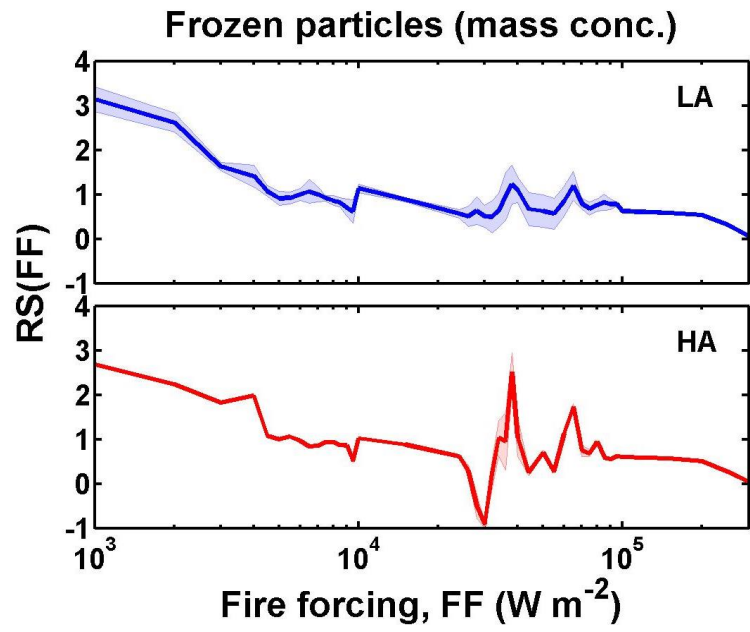
(c)



(d)

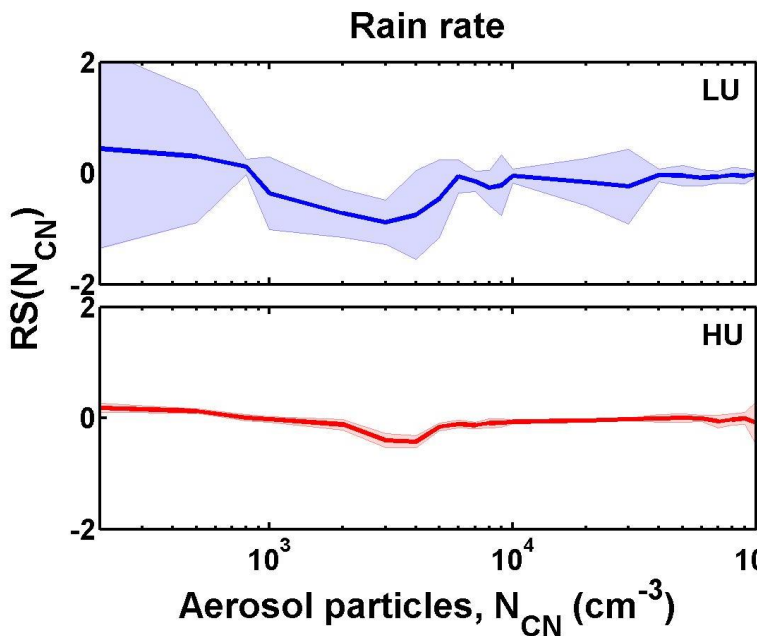
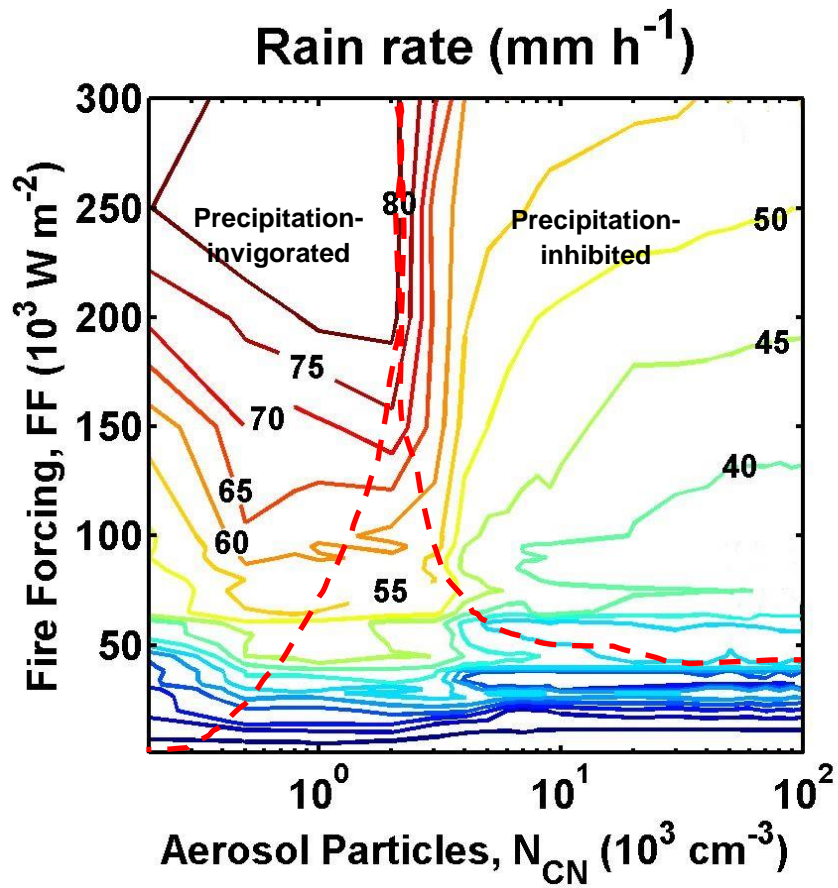


(e)

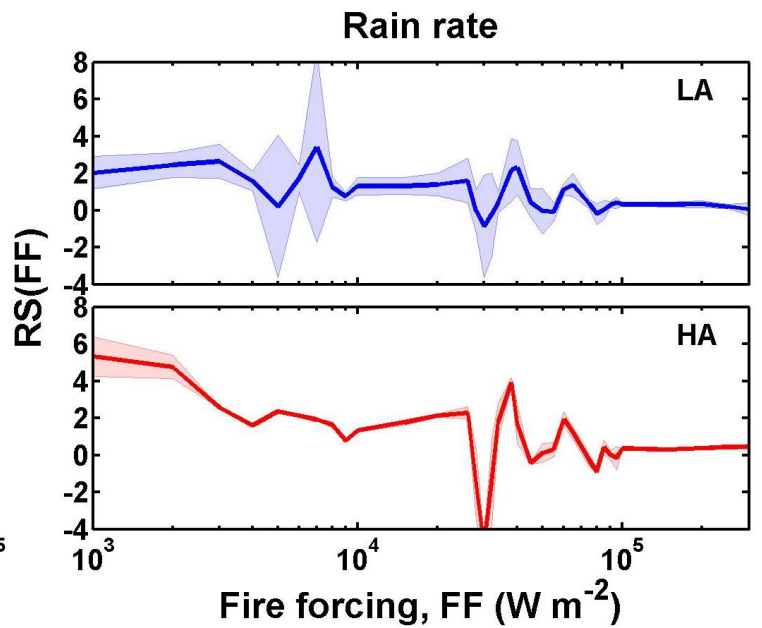


(f)

Figure 12. Same as Figure 7 but for total frozen particles.



(b)



(c)

Figure 13. Same as Figure 7 but for surface rain rate.

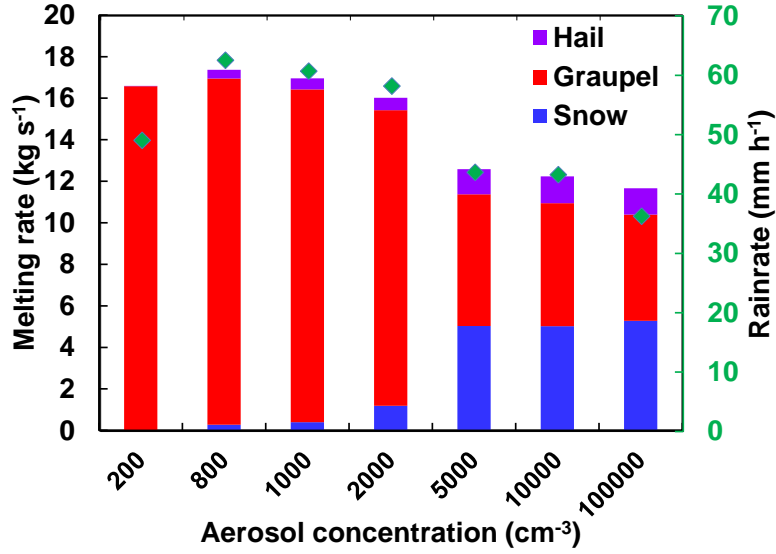


Figure 14. The correlation of rain rate and the melting rate of the frozen particles. The green diamond points are the averaged rain rate under different aerosol concentrations ($FF = 10^5 \text{ W m}^{-2}$). The columns represent the integrated melting rate from individual frozen particles.

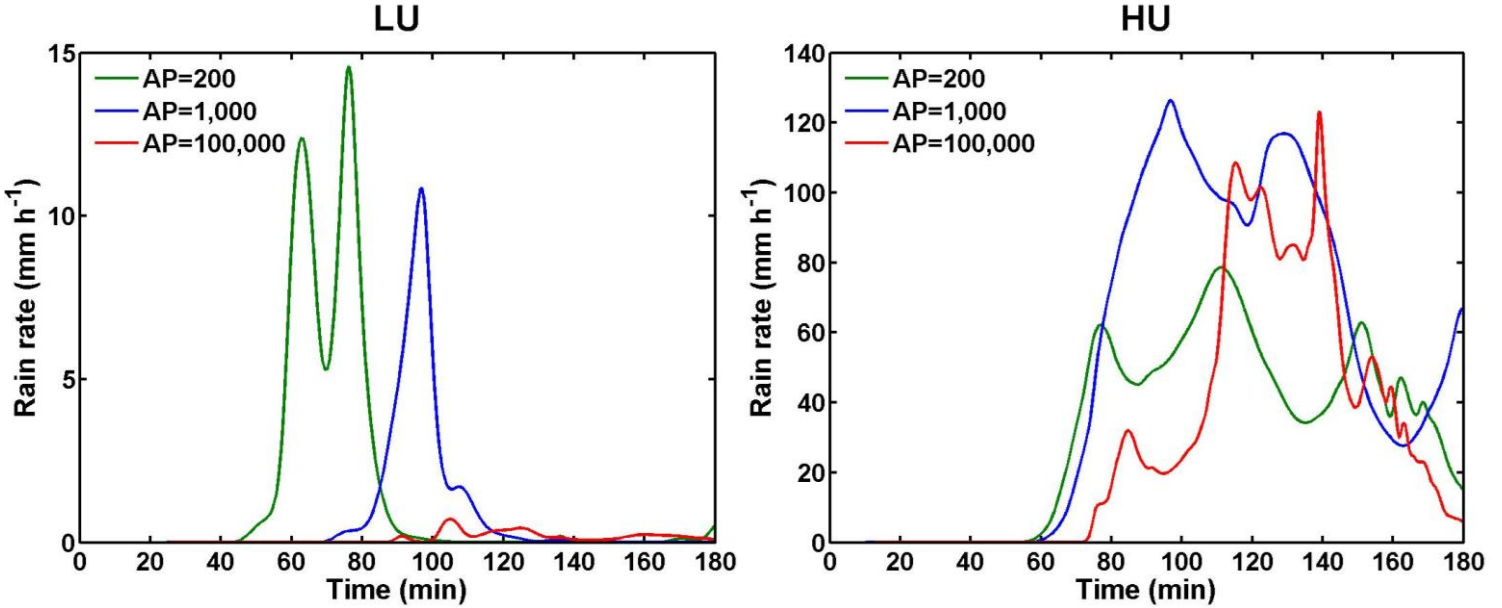


Figure 15. Time evolution of surface rain rates for the three aerosol episodes ($N_{CN} = 200; 1,000; \text{ and } 100,000 \text{ cm}^{-3}$ respectively) under LU (low updrafts, $FF=2,000 \text{ W m}^{-2}$) and HU (high updrafts, $FF=50,000 \text{ W m}^{-2}$) conditions.

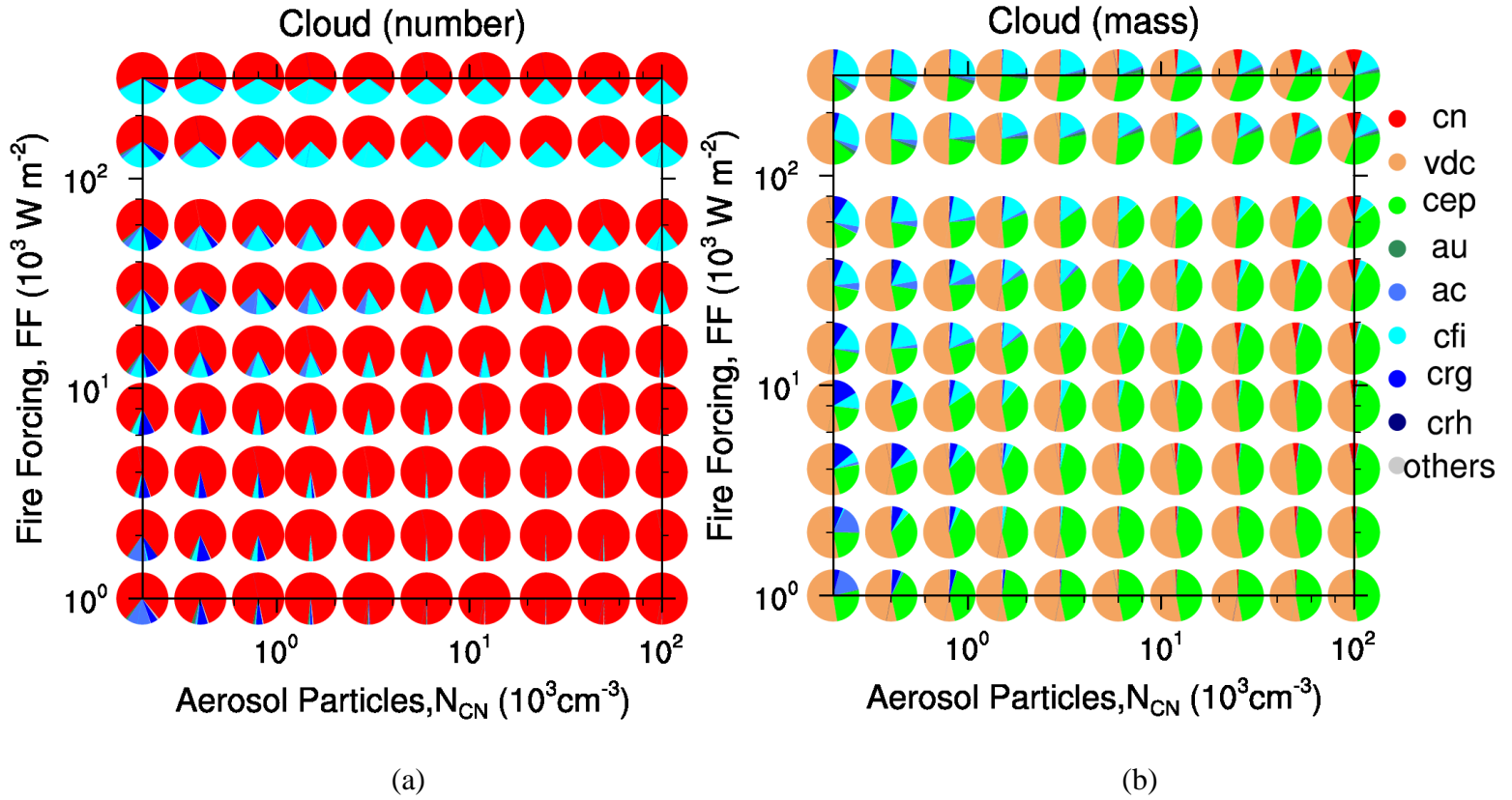
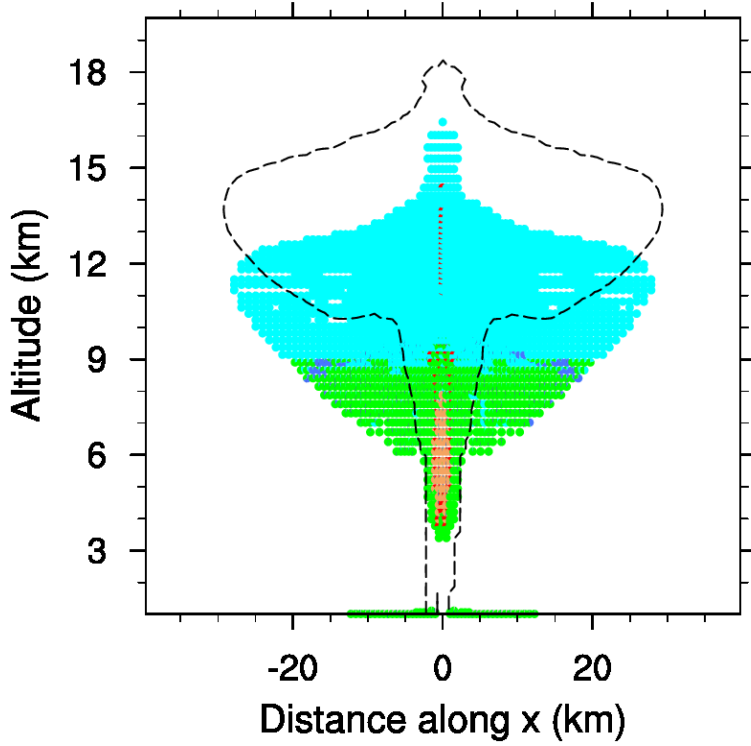
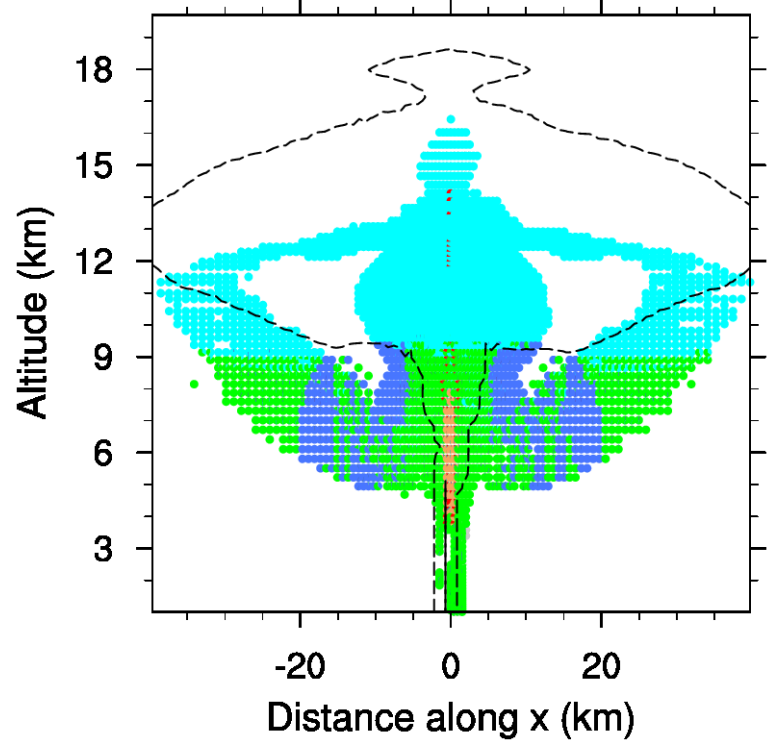


Figure 16. The pie charts summarize the relative percentage of the microphysical processes involving cloud droplets as a function of N_{CN} and fire forcing (a: number concentration; b: mass concentration). Colors within each pie chart reflect the contribution of processes under the specific condition. Warm colors denote the sources, while cold colors denote the sinks. The acronyms indicate cn: cloud nucleation; vdc: condensational growth of cloud droplets; cep: evaporation of cloud droplets; au: autoconversion; ac: accretion; cfi: freezing of cloud droplets to form ice crystals, including homogeneous and heterogeneous nucleation; crg/h: riming of cloud droplets to form graupel/hail.

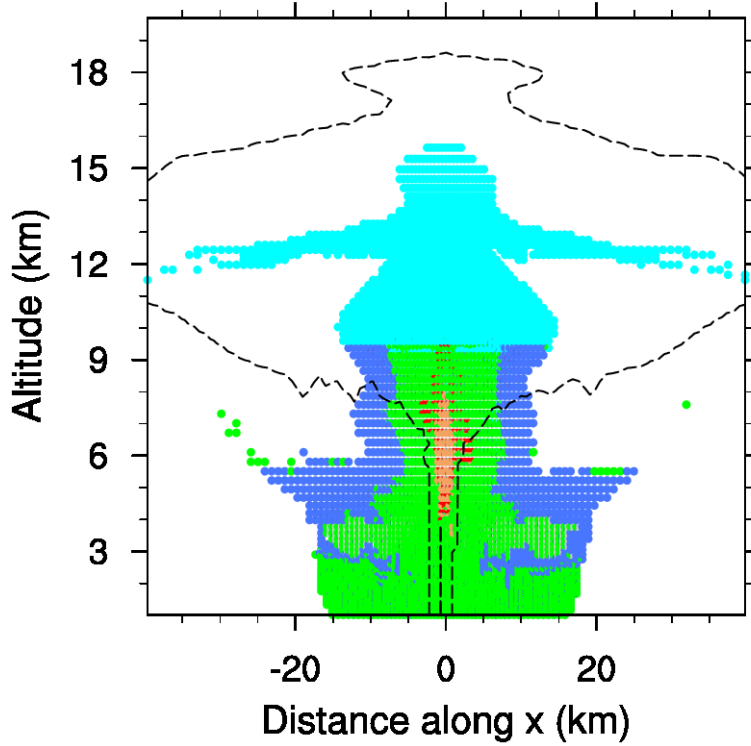
Cloud (30min)



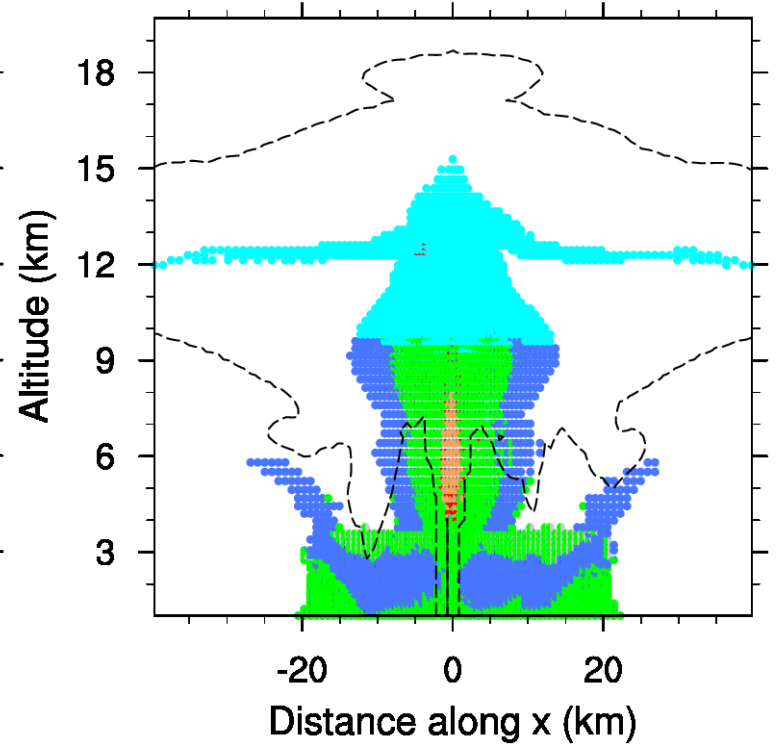
Cloud (60min)



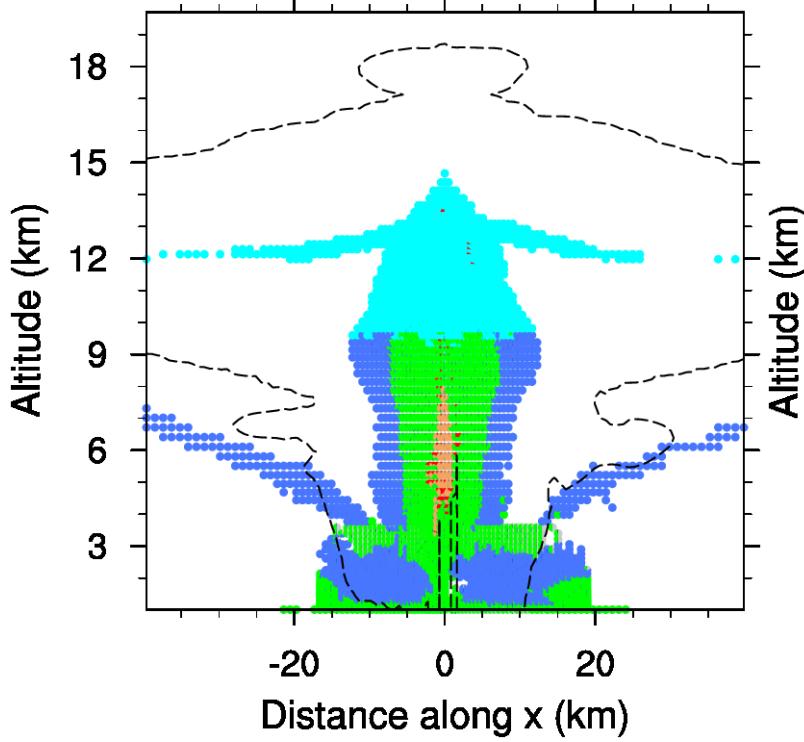
Cloud (90min)



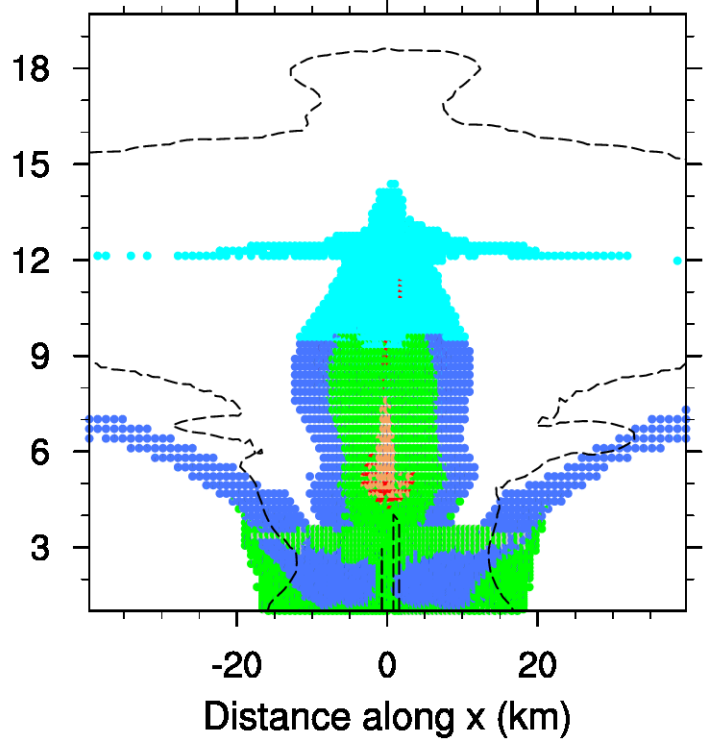
Cloud (120min)



Cloud (150min)



Cloud (180min)



● cn ● vdc ● cep ● au ● ac ● cfi ● crg ● crh ● others

Figure 17. The pie charts summarize the vertical cross sections of the change rate of main microphysical processes contributing to cloud water content. Each pie chart shows the averaged contribution over the past 30 min. Colors within each pie chart reflect the percentage of processes in each grid. The black dashed line is the $0.1 \mu\text{g kg}^{-1}$ isoline of the interstitial aerosol, indicating the shape of smoke plume. The meaning of the acronyms is the same as in Figure 16. Warm colors denote the sources, while cold colors denote the sinks.

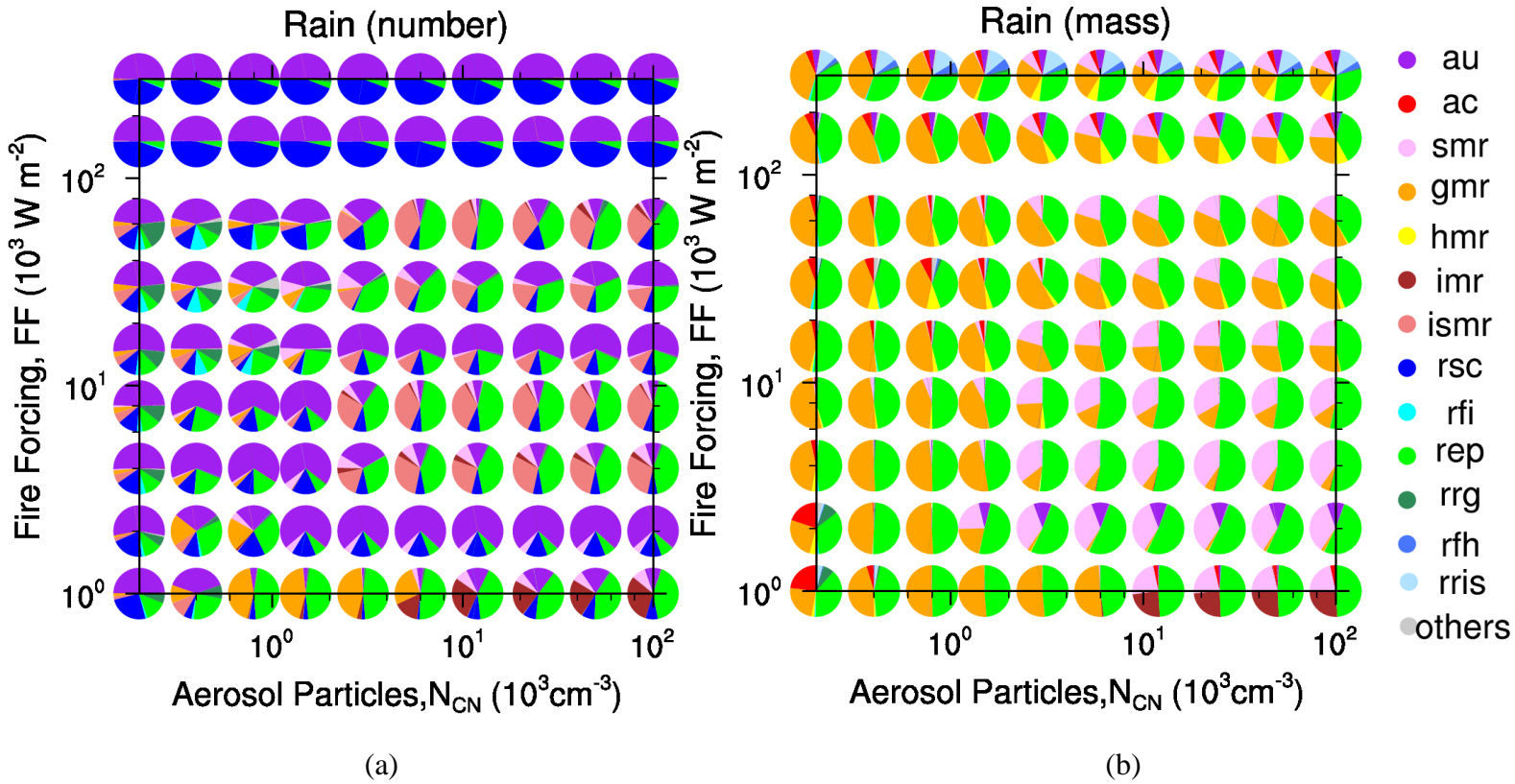
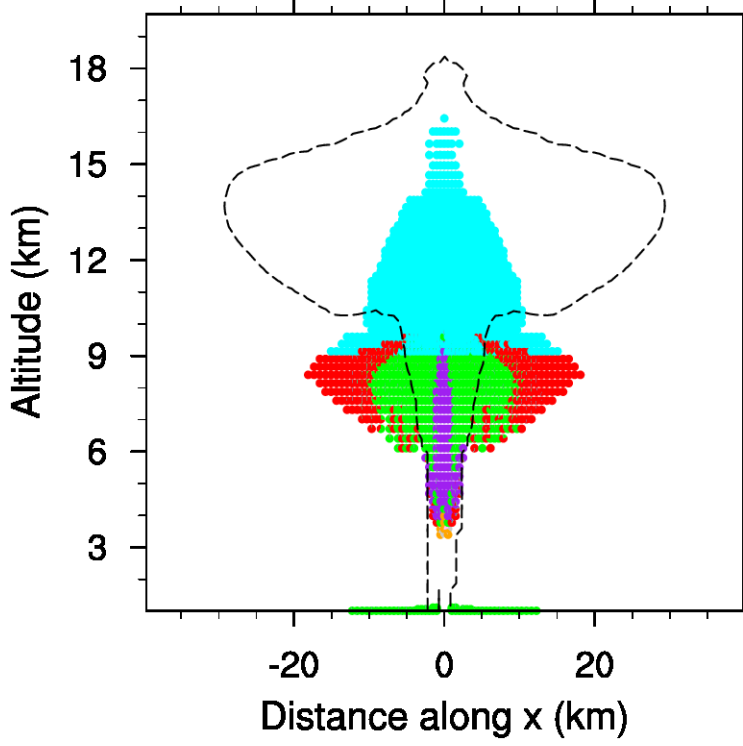
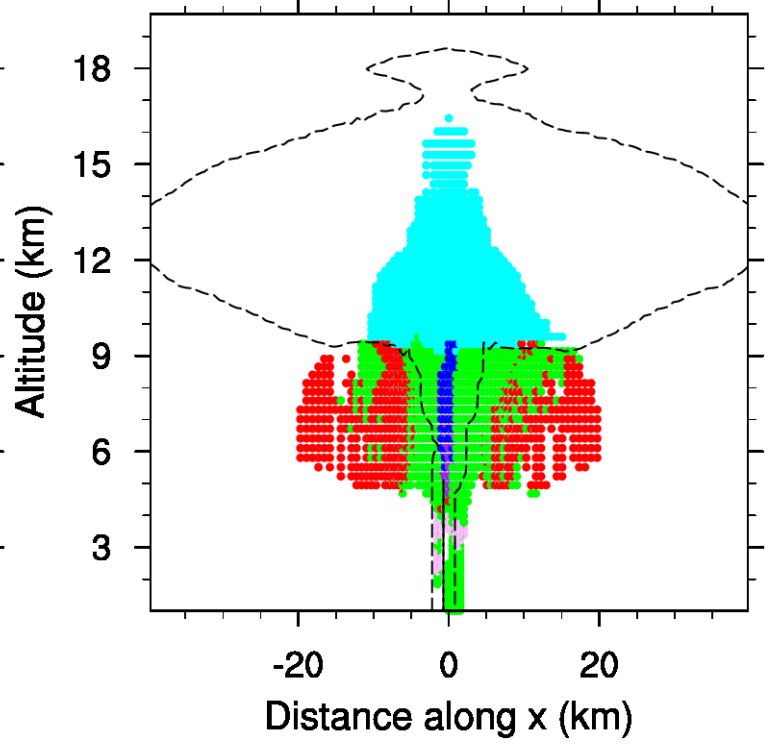


Figure 18. Same as Figure 16 but for raindrops. The acronyms indicate au: autoconversion; ac: accretion; i/s/g/hmr: melting of ice/snow/graupel/hail to form raindrops; rsc: self-collection of raindrops; ismr: melting of ice and snow to form raindrops; rfi/h: freezing of raindrops to form ice crystals/hail; rep: raindrop evaporation; rrg: riming of raindrops to form graupel; rris: riming of raindrops to form ice and snow.

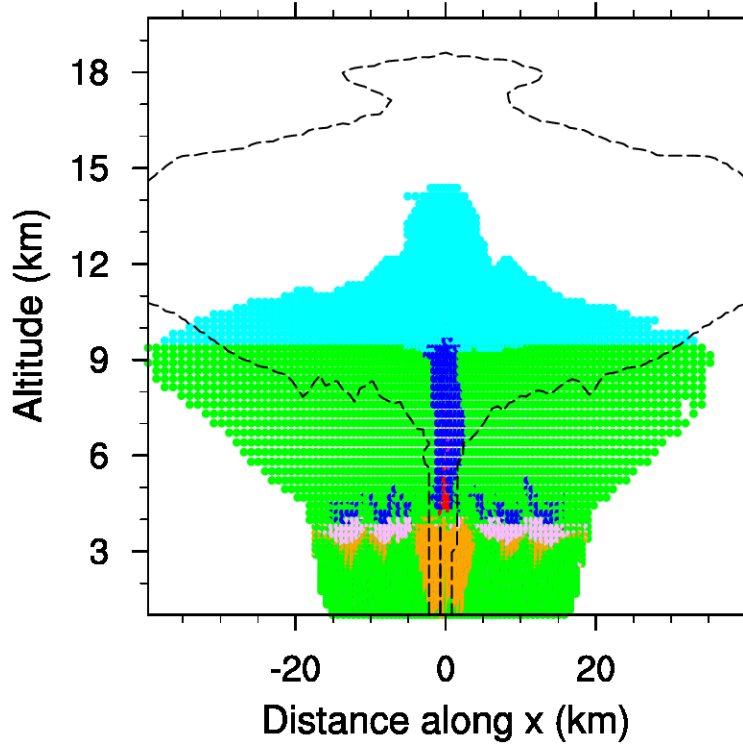
Rain (30min)



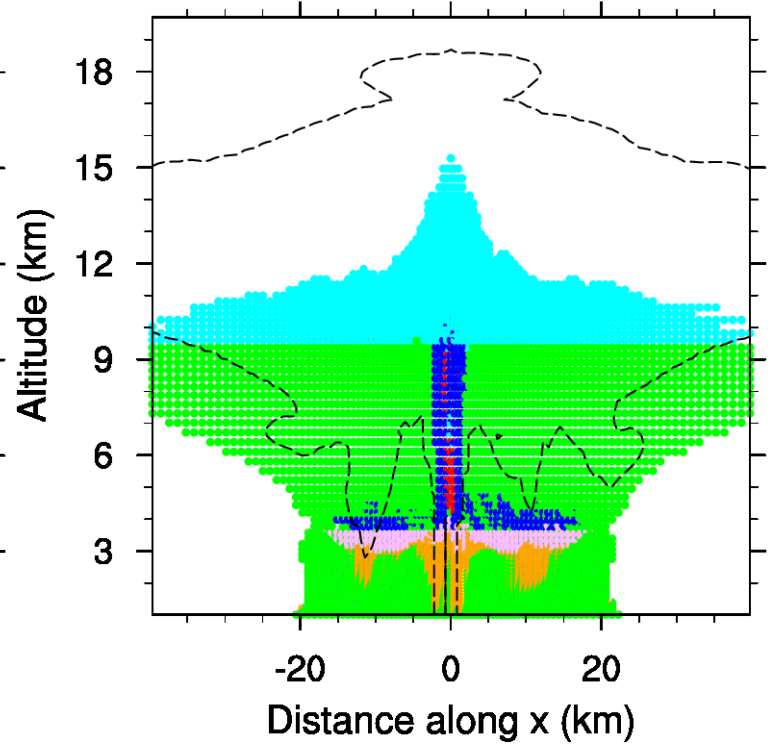
Rain (60min)



Rain (90min)



Rain (120min)



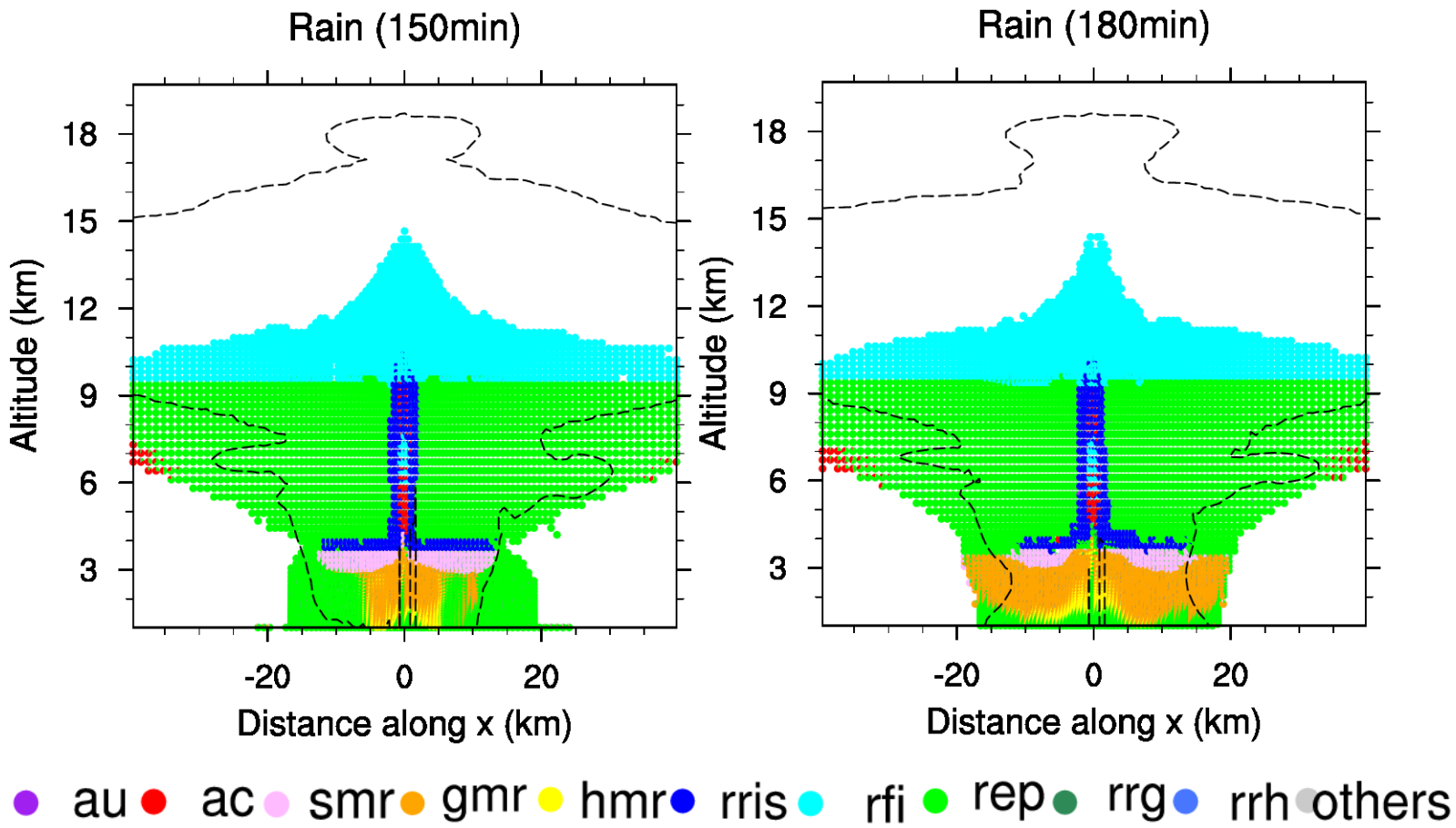


Figure 19. Same as Figure 17, but for raindrops.

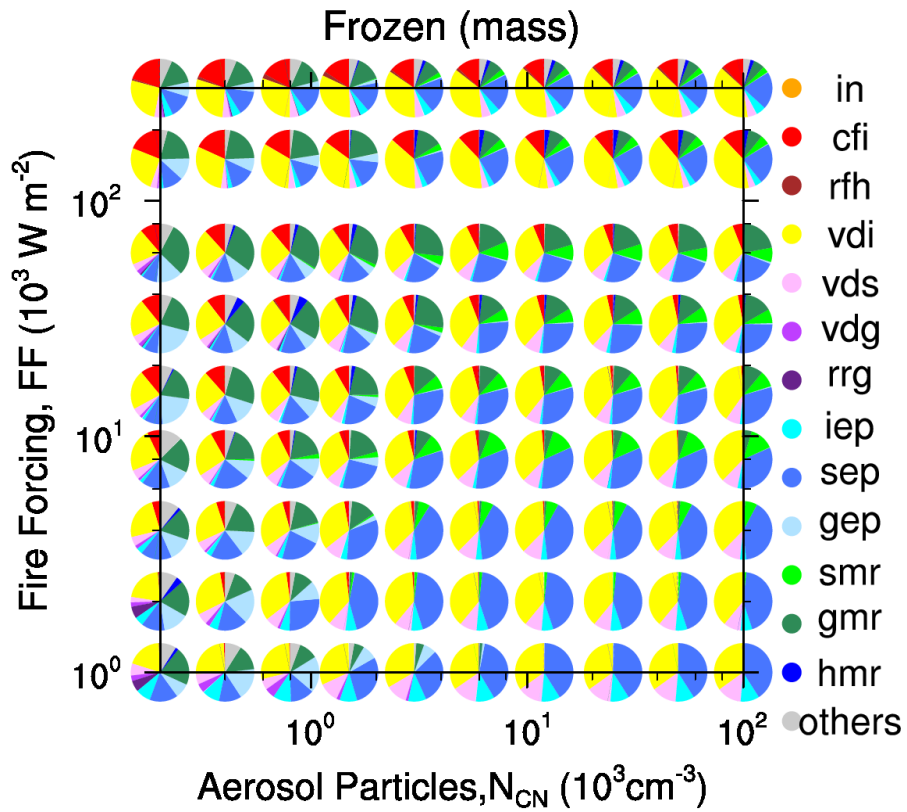
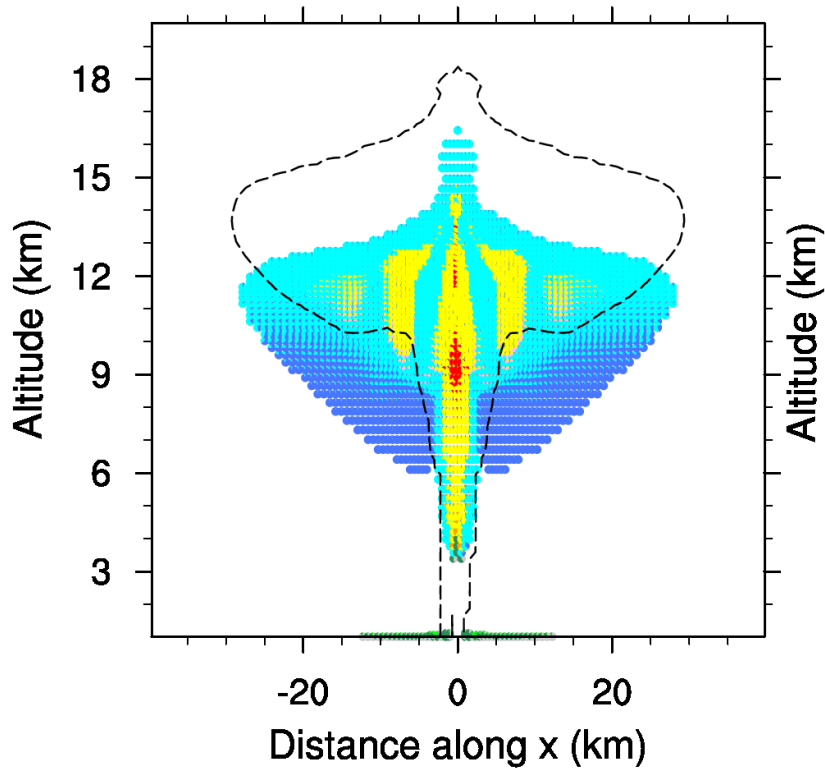
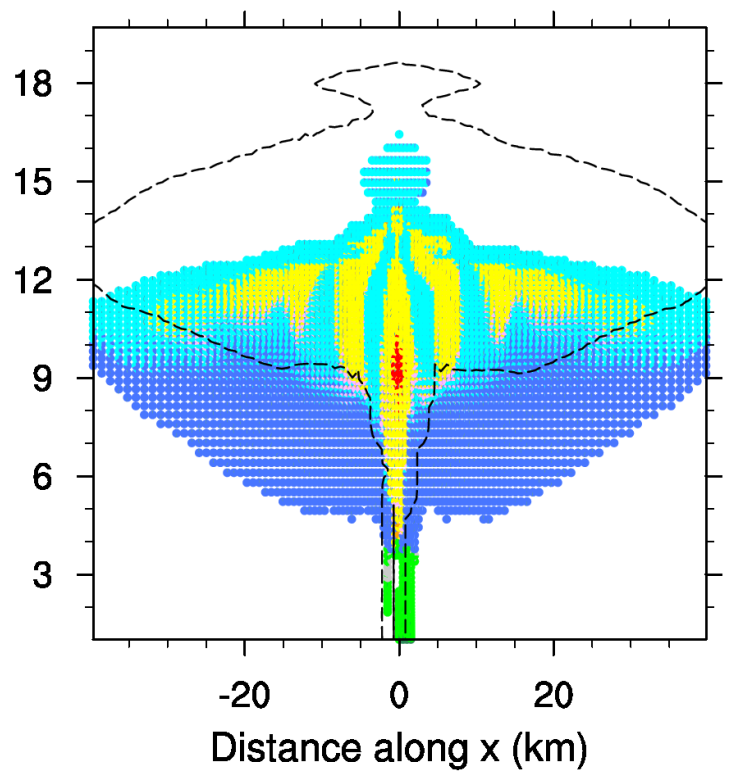


Figure 20. Same as Figure 16 but for the total frozen water content. The acronyms indicate in: ice nucleation; cfi: freezing of cloud droplets to form ice crystals, including homogeneous and heterogeneous nucleation; rfh: freezing of raindrops to form hail; vdi/s/g: condensational growth of ice crystals/snow/graupel by water vapor; rrg: riming of raindrops to form graupel; i/s/gep: evaporation of ice/snow/graupel; s/g/hmr: melting of snow/graupel/hail to form raindrops.

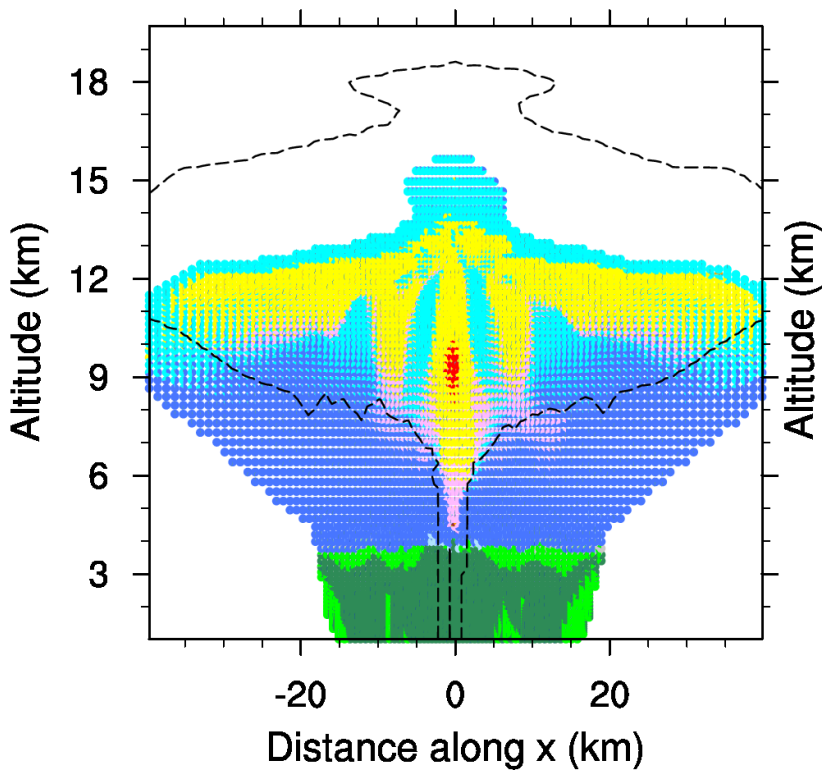
Frozen (30min)



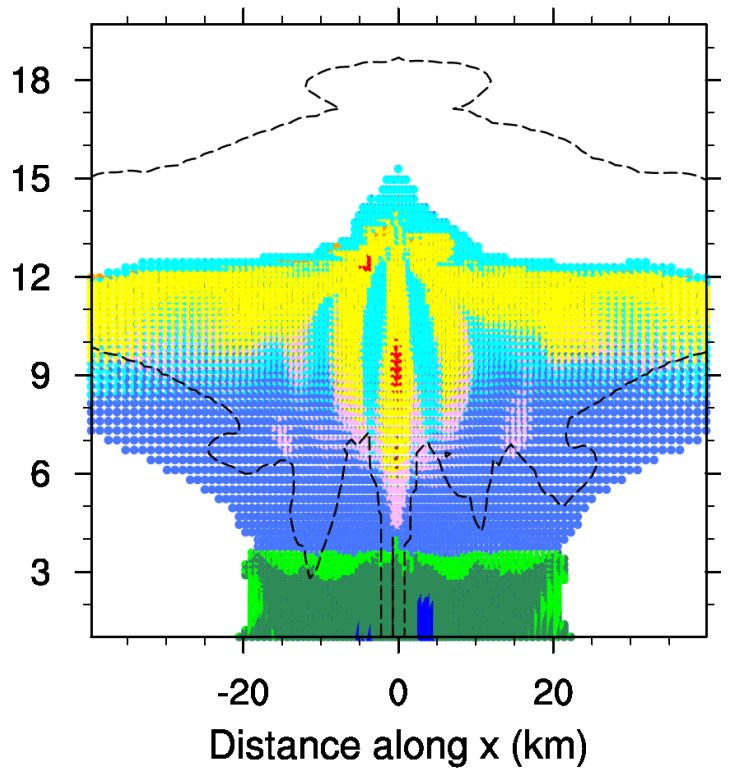
Frozen (60min)



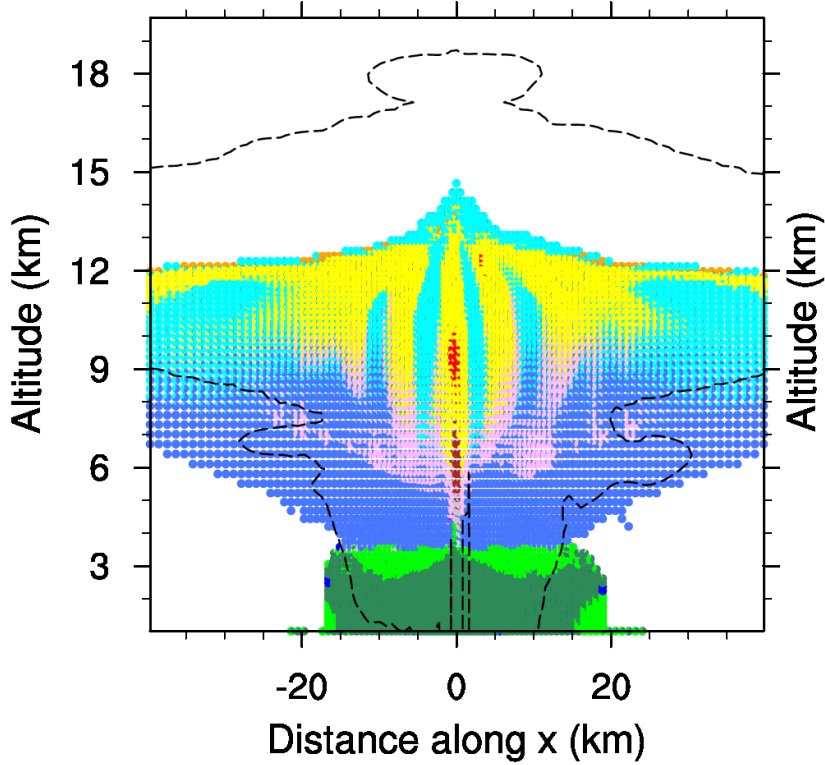
Frozen (90min)



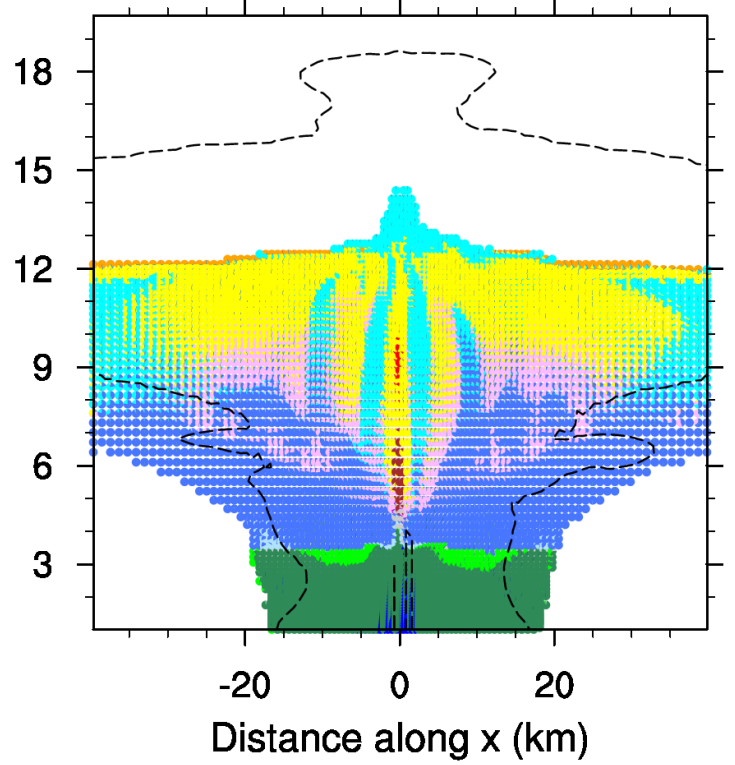
Frozen (120min)



Frozen (150min)



Frozen (180min)



- in ● cfi ● rfh ● vdi ● vds ● vdg ● iep ● sep ● gep ● smr ● gmr ● hmr ● others

Figure 21. Same as Figure 17 but for frozen particles.

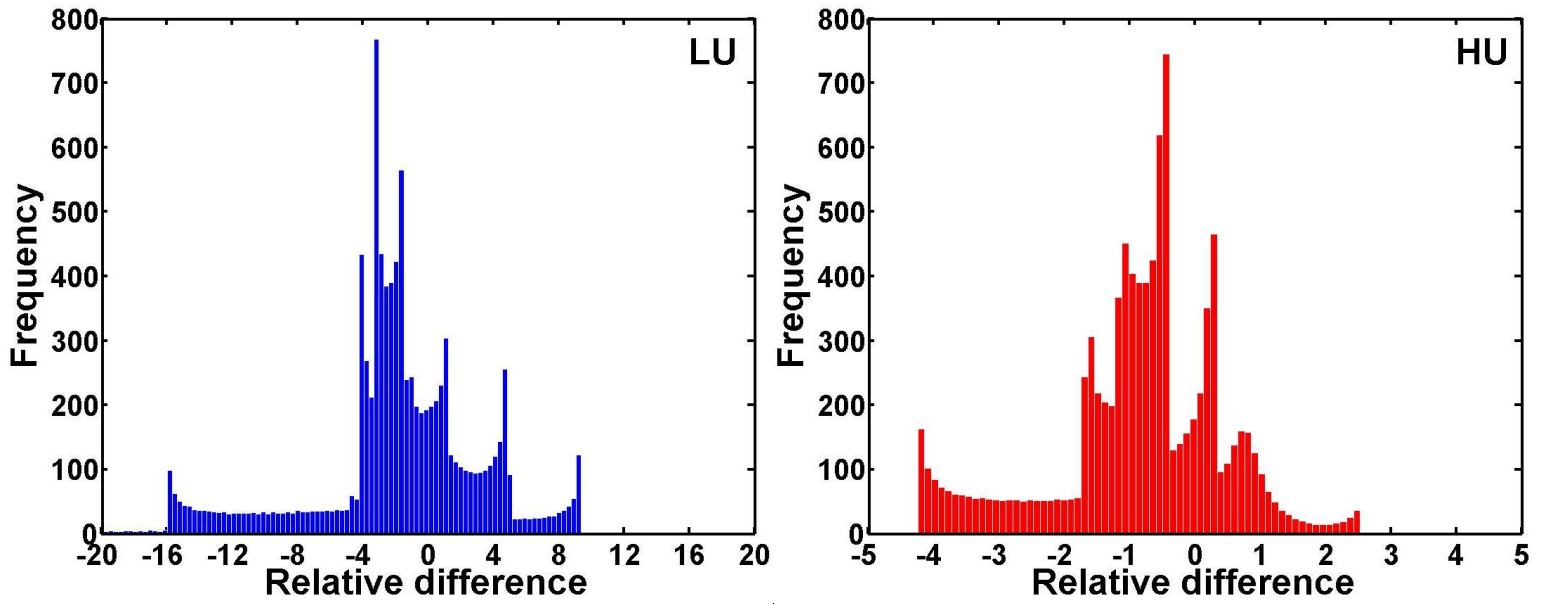


Figure 22. Histograms of the relative difference between $\frac{\Delta Y}{\Delta N_{CN}}$ and $\frac{dY}{dN_{CN}}$ under LU and HU

conditions, where Y here denotes precipitation rate. $\frac{\Delta Y}{\Delta N_{CN}} = \frac{Y(2N_{CN}) - Y(N_{CN})}{2N_{CN} - N_{CN}}$, and $\frac{dY}{dN_{CN}}$ is the derivative of the precipitation rate along the variable N_{CN} .

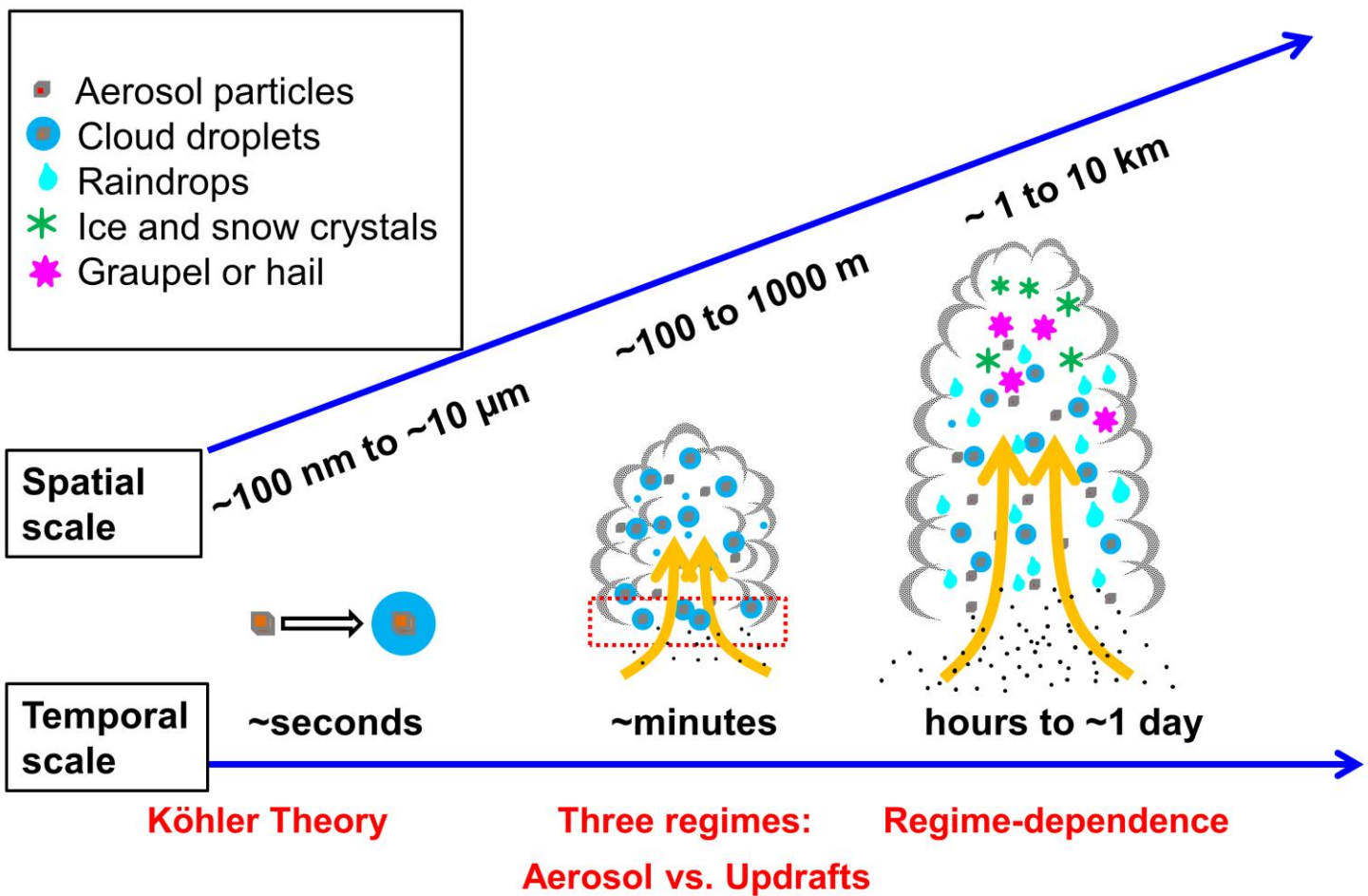


Figure 23. Overview of the research approaches on multi-scale cloud initialization and development. The aerosol-cloud interaction at the microphysical scale, i.e., cloud condensation nuclei (CCN) activation, has been well characterized by the Köhler theory (Kohler, 1936) and by a series of extended equations (Shulman et al., 1996; Kulmala et al., 1997; Laaksonen et al., 1998). When we upscale the activation of a single aerosol particle to aerosol populations at the cloud base, the impact of aerosols on the number of activated CCN still appears simple and can be well described (i.e., the three generic regimes of CCN activation). When considering full microphysics and the larger temporal and spatial scales of a single pyroconvective cloud, the performance of ensemble simulations shows the regime dependence of aerosol effects on the pyroconvective cloud formation and evolution.