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1 **Ice Multiplication by Fragmentation of Spherical Freezing of Drops:**

2 **A Theoretical Investigation**

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ABSTRACT

10 The ice multiplication by fragmentation associated with collision-freezing
11 of supercooled drops is investigated. A zero-dimensional dynamical system
12 describing the time evolution of the number density of the supercooled drops
13 and the ice crystals in a mixed-phase cloud is developed. The analytical so-
14 lutions of this system are derived for various asymptotic limits. Especially,
15 when the ice fragmentation by freezing supercooled drops is considered in
16 its isolation, a complete analytical solution is available, which shows that
17 the originally-existed supercooled drops are simply all converted into the ice
18 crystals multiplied by a fragmentation number per freezing. When constant
19 generation of both supercooled drops and ice crystals is considered, the ice-
20 crystal number tends to linearly increase with time with the rate given by the
21 sum of the primary-ice and the the supercooled-drop sources with the latter
22 multiplied by the fragmentation number per freezing. The study concludes
23 that the ice multiplication by fragmentation of freezing drops can be a cru-
24 cial process in certain situations. Yet its multiplication tendency is limited by
25 availability of supercooled rain or drizzle and by whether the freezing tem-
26 perature allows more than one fragment to be emitted per frozen drop so as
27 to allow a positive feedback with collisional raindrop-freezing. When there
28 is no more rain or drizzle, the multiplication stops. When there is copious
29 supercooled rain initially and temperatures are optimal, then this fragmenta-
30 tion can be more prolific than the other multiplication processes such as the
31 Hallett-Mossop process and the ice-ice collision process.

1. Introduction

Coincident measurements of concentrations ice particles in clouds and of active ice nucleus (IN) aerosols in the environment have shown a discrepancy by orders of magnitude, in clouds too warm for homogeneous freezing (Hobbs 1969, Auer *et al.* 1969). This has been explained in terms of fragmentation of ice. Several pathways of fragmentation have been observed in the laboratory, but their relative importance has been unclear.

One possible type of fragmentation is shattering during collision–freezing of supercooled drops. Generally the rate of freezing of a drop is limited by the rate of dissipation of latent heat. Initially, the latent heat from freezing is converted to thermal energy of the drop, raising its temperature close to 0°C almost instantaneously. In this first stage, needles of ice permeate the drop. In the subsequent stage, the latent heat flows down the gradient of temperature to the surroundings, once a steady state has been reached. The rate of dissipation to the air governs the freezing rate. The fastest rate of freezing occurs for freezing as near as possible to the drop surface. Hence, the freezing occurs from the outside inwards, with a shell of ice encasing the liquid core.

It is experimentally observed that as the ice shell thickens, the expansion on freezing can sometimes cause the shell to shatter (Wildeman *et al.* 2017). Splinters of ice can then be emitted into the air. Many earlier laboratory experiments (Latham and Mason 1961, Brownscombe and Thorndike 1968, Dye and Hobbs 1968, Hobbs and Alkezweeny 1968, Takahashi and Yalashita 1969) measured this fragmentation for drops in free-fall: See Mossop (1970) as a review. Only a few splinters are typically emitted per frozen drops (usually less than about 1-10 depending on temperature and size). On the other hand, there is a positive feedback that can lead to all supercooled drops being frozen quite quickly: as ice splinters are emitted they can grow and after a minute or so are large enough to collide efficiently with supercooled drops, freezing them.

56 Phillips *et al.* (2001) quantified the effect of this multiplication mechanism by raindrop-
57 splintering for a New Mexican convective cloud. It was found that it was less prolific than the
58 Hallett–Mossop (1974) process of rime-splintering between -3 and -8°C when cloud-droplets
59 freeze on impact with graupel. However, Lawson *et al.* (2015) simulated another observed case
60 with a spectral bin microphysics parcel model and found the positive feedback to account for the
61 observed ice.

62 Phillips *et al.* (2018) provided a comprehensive formulation to treat rain– or drizzle–drop freez-
63 ing fragmentation by pooling published data from past laboratory studies in the literature. Two
64 modes were represented:

- 65 • Mode 1: spherical drop-freezing when incident ice particle is less massive than the drop or due
66 to immersed IN activating, with an outer ice shell growing inwards and breaking;
- 67 • Mode 2: non-spherical drop-freezing when incident ice particle is more massive than the drop
68 and ice particles may be present in the splash.

69 Only mode 1 has been observed in the lab comprehensively. For mode 2 a theoretical approach
70 for the physics of the collision was invoked. After creating an empirical formulation for mode 1
71 alone, Phillips *et al.* (2018) applied it to a simulation of a composite case of deep convection from
72 the tropical Atlantic in ICE-T (Ice in Clouds Experiment–Tropical). They found observed ice
73 concentrations were predicted in a bin microphysical parcel model with the new schemes for an
74 assumed probability of 50% (-10°C) for the chance of any drop in the splash containing ice due to
75 partial freezing on impact.

76 The empirical scheme for numbers of secondary fragments per drop, N , from spherical drop-
77 freezing of mode 1 was expressed by Phillips *et al.* (2018) with a 2D phase-space of drop diam-
78 eter and freezing temperature. A peak of N near -15°C was observed. Construction of a zero–
79 dimensional (0D) model of monodisperse populations of crystals, graupel (from drop-freezing)

80 and supercooled drops without fallout showed that the positive feedback with exponential growth
81 of ice concentrations only occurs for $N > 1$, which is found at diameters of 0.1 mm between about
82 -13 and -17°C , 0.2 mm (or 0.4 mm) between about -10°C and -20°C and 1.6 mm when colder
83 than -7°C . When mode 2 was included in the 0D model, the realm of instability was extended to a
84 wider range of temperature.

85 The purpose of the present paper is to extend this investigation of the efficiency of the ice frag-
86 mentation by spherical drop-freezing from such a theoretical point of view. A similar 0D dynam-
87 ical system is applied for this purpose, generalized to include a source of raindrops from coales-
88 cence, as presented in the next section. By taking the similar methodologies, Yano and Phillips
89 (2011) and Yano *et al.* (2016) have investigated the efficiency of the Hallett–Mossop and the ice–
90 ice collision ice–multiplication processes. Yano *et al.* (2016) further list the various methodologies
91 applicable for interpreting the behavior of a dynamical system, and in that manner, the basic nature
92 of a given microphysical process described by the given dynamical system can be elucidated.

93 The most basic strategy is to develop an asymptotic expansion by assuming a certain physical
94 parameter to be small. We emphasize that a given physical parameter may not be necessarily small
95 arithmetically, but it can still be introduced as a small parameter in order to obtain insights to the
96 basic behavior of the system. This most basic strategy is systematically applied in the present
97 study. After introducing a formulation of the problem in the next section, theoretical analysis
98 considers first the ice fragmentation process of mode 1 in its isolation in Sec. 3 and the feedbacks
99 are quantified. General analyses are presented in Sec. 4. Obtained results are further discussed in
100 Sec. 5.

101 **2. 0D Model**

102 *a. Basic Assumptions*

103 We consider a cloud at subzero temperatures consisting of supercooled drops, ice crystals, and
 104 graupel. Their number densities are designated by n_r , n_i , and n_g , respectively. Collisions between
 105 ice crystals and supercooled drops cause freezing to the latter leading to formation of graupel,
 106 associated with N (up to 10–100) secondary fragments of ice emitted by each frozen drop. The
 107 rate that supercooled drops freezes by collision of ice particles may be given by $Kn_r n_i$, where K is
 108 a coefficient describing the product of the collision efficiency and rate of geometric sweep-out of
 109 air volume per second (collection kernel, $\text{m}^3 \text{sec}^{-1}$).

110 We further assume constant generations, c_0 and c_i , of supercooled rain and ice crystals, respec-
 111 tively. We also assume that the supercooled rain is lost by fall with a characteristic time scale of
 112 τ_f . Some of those additional processes may be turned off in the following as required.

113 Here, being consistent with our earlier studies (Yano and Phillips, 2011, Yano *et al.* 2016,
 114 Phillips *et al.* 2018), no spatial dimension is considered.

115 *b. Mathematical Formulation*

116 From the physical assumptions just introduced above, the mathematical description of this mi-
 117 crophysical system is presented by

$$118 \quad \dot{n}_r = c_0 - Kn_r n_i - n_r / \tau_f \quad (2.1a)$$

$$119 \quad \dot{n}_i = c_i + KN' n_r n_i \quad (2.1b)$$

120
 121 Note that the drop–freezing is initiated by capture (loss) of an ice particle, and leads to a gain of N
 122 ice particles by fragmentation so that $N' = N - 1$ more ice particles are found after drop–freezing
 123 in total per drop frozen. This means that for some conditions of drop size and freezing temperature
 124 (as reviewed by Pruppacher and Klett 1997), there can be $N' < 0$, such as at diameters less than
 125 about 0.1 mm near the optimum of -15°C and 0.5–1 mm at temperatures several degrees warmer
 126 or colder than this. For a monodisperse population of drops, when $N' < 0$, then there is capture

127 of splinters causing a decrease with time of the crystal concentration with incomplete freezing of
 128 the population of drops. This corresponds to a situation in reality with a continuum of drop sizes
 129 in any cloudy volume but with insufficiently numerous large drops to cause any explosion of ice
 130 concentration (Sec. 1).

131 Additionally, the time evolution of the graupel number, n_g , is presented by

$$132 \quad \dot{n}_g = Kn_r n_i - n_g / \tau_g$$

133 also assuming that the graupel is lost from the cloud-microphysical system by fall-out with a
 134 characteristic time-scale, τ_g . However, the graupel number, n_g , does not affect the subsequent
 135 evolution of the remaining part of the system. Thus, in the following, the evolution of n_g will not
 136 be considered.

137 *c. Nondimensionalization*

138 Nondimensionalization is crucial to ensure a systematic investigation of a given system, be-
 139 cause it reduces the parameters characterizing the system to a smaller number of nondimensional
 140 parameters, thus a parameter space for the investigation is much reduced.

141 In the following, we add the subscript $*$ to the nondimensional variables in order to distinguish
 142 them from the dimensional variables. Here, we introduce an unspecified time scale, τ , for nondi-
 143 mensionalizing the time. Although the fall time-scale, τ_f , of rain is a natural choice for the scale,
 144 we retain a certain freedom for the model analysis in this manner. Thus,

$$145 \quad \frac{d}{dt} = \frac{1}{\tau} \frac{d}{dt_*}. \quad (2.2)$$

146 The particle number densities may be nondimensionalized by

$$147 \quad n_i = n_{i*} / K \tau, \quad (2.3a)$$

$$148 \quad n_r = n_{r*} / KN' \tau. \quad (2.3b)$$

150 By substituting the expressions (2.2), (2.3a, b) into Eqs. (2.1a, b), we obtain a nondimensional-
 151 ized system:

$$152 \quad \dot{n}_{r*} = \hat{c}_0 - \hat{K}n_{i*}n_{r*} - \varepsilon_s n_{r*}, \quad (2.4a)$$

$$153 \quad \dot{n}_{i*} = \hat{c}_i + \hat{K}n_{r*}n_{i*}. \quad (2.4b)$$

154
 155 Here, we have introduced the following nondimensional parameters:

$$156 \quad \hat{c}_0 = KN'\tau^2 c_0, \quad (2.5a)$$

$$157 \quad \hat{c}_i = K\tau^2 c_i, \quad (2.5b)$$

$$158 \quad \varepsilon_s = \tau/\tau_f. \quad (2.5c)$$

160 Note that \hat{c}_0 and \hat{c}_i are the nondimensional generation rates of liquid and ice; ε_s measures the
 161 magnitude of the rain sedimentation rate. Furthermore, the dimensionless tag, \hat{K} , is introduced,
 162 multiplying the freezing–fragmentation terms so as to indicate a contribution of this process to the
 163 solutions. Formally $\hat{K} = 1$ by nondimensionalization when this process is present and may be set
 164 zero otherwise. Note that the time scale, τ , is defined by Eq. (3.5) below in retrospect.

165 **3. Basic Analysis: Freezing–Drop Fragmentation Process in Isolation**

166 As the simplest case, the shattering of ice particles by freezing drops is considered in isolation
 167 away from the other processes. We obtain this situation by setting, $\hat{c}_0 = \hat{c}_i = \varepsilon_s = 0$ in the above
 168 general system (2.4):

$$169 \quad \dot{n}_{r*} = -\hat{K}n_{r*}n_{i*}, \quad (3.1a)$$

$$170 \quad \dot{n}_{i*} = \hat{K}n_{r*}n_{i*}. \quad (3.1a)$$

172 Here, the tag, \hat{K} , for the ice fragmentation process is still retained. Note that there is no nondi-
 173 mensional parameter controlling this system. Thus, even in numerical terms, the interest of the

174 investigation of this system only depends on the initial condition. As seen below, the initial con-
 175 dition can further be normalized, because the characteristic time-scale, τ , of the system remains
 176 arbitrary due to the absence of rain sedimentation.

177 *a. Full Solution*

178 We identify two equilibrium solutions in this system: either $n_{r*} = 0$ or $n_{i*} = 0$ and the other
 179 number density remains an arbitrary finite value. As suggested by Yano and Phillips (2011), as
 180 well as Yano *et al.* (2016: See especially their Sec. 6.2), the standard procedure would be to
 181 perform the linear perturbation analysis against these equilibrium states for inferring the tendency
 182 of the system away from these equilibrium states.

183 However, in the present case, a full analytical solution is available for Eqs. (3.1a, b). This is
 184 realized by noting that the sum of the two particle number densities is conserved, as seen by
 185 taking the sum of Eqs. (3.1a, b):

$$186 \quad \frac{d}{dt_*}(n_{r*} + n_{i*}) = 0. \quad (3.2)$$

187 In dimensional terms, the above conservation law is

$$188 \quad \frac{d}{dt}(N' n_r + n_i) = 0.$$

189 Thus, more precisely, the sum of the number densities, weighted by the ice-fragmentation number,
 190 N' , on the rain water number density, n_r , is conserved with time.

191 From Eq. (3.2), n_{i*} can be written in terms of n_{r*} by

$$192 \quad n_{i*} = n_0 - n_{r*}, \quad (3.3)$$

193 where $n_0 = n_{r*}(0) + n_{i*}(0)$ is the initial total particle number. By substituting the above expression
 194 into Eq. (3.1a), we obtain a differential equation solely in terms of n_{r*} . It can be readily solved to
 195 obtain:

$$196 \quad n_{r*} = (n_{r*}(0) + n_{i*}(0)) \left(1 + \frac{n_{i*}(0)}{n_{r*}(0)} e^{\hat{K} n_0 t_*}\right)^{-1}. \quad (3.4a)$$

197 Further substitution of the above solution into Eq. (3.3) leads to a solution for n_{i^*} :

$$198 \quad n_{i^*} = (n_{r^*}(0) + n_{i^*}(0)) \left(1 + \frac{n_{r^*}(0)}{n_{i^*}(0)} e^{-\hat{K}n_0 t^*}\right)^{-1}. \quad (3.4b)$$

199 Note that the time-scale, τ , introduced above for non-dimensionalising the system remains ar-
 200 bitrary. Thus without loss of generality, we may set the initial condition to $n_0 = 1$, which implies
 201 that

$$202 \quad \tau = K^{-1}(N'n_r(0) + n_i(0))^{-1} \quad (3.5)$$

203 by referring to Eq. (2.3a, b). Note that the dimensionless characteristic time-scale for the expo-
 204 nentials of both n_{r^*} and n_{i^*} is just $1/n_0$ in Eq (3.4a, b). This characteristic time-scale becomes τ
 205 when dimensionalised. In other words, the initial particle number dictates the characteristic time-
 206 scale of the system, as shown by Phillips et al. (2018). Furthermore, the behaviour of the system
 207 can be investigated systematically by solely changing the initial condition for n_{r^*} as a result. The
 208 examples of time evolution for those normalizations are shown in Fig. 1.

209 It may also be worthwhile to note that the solutions (3.4a, b) are given in dimensional terms by:

$$210 \quad n_r = (n_r(0) + n_i(0)/N') \left[1 + \frac{n_i(0)}{N'n_r(0)} e^{K(N'n_r(0) + n_i(0))t}\right]^{-1} \quad (3.6a)$$

$$211 \quad n_i = (N'n_r(0) + n_i(0)) \left[1 + \frac{N'n_r(0)}{n_i(0)} e^{-K(N'n_r(0) + n_i(0))t}\right]^{-1} \quad (3.6b)$$

212
 213 The result shows that regardless of the initial condition, the system exponentially approaches to a
 214 state only with the ice crystals, $n_i = N'n_r(0) + n_i(0)$ by converting all the supercooled drops into
 215 the ice. As a result, we also see that the state with $n_r \neq 0$ and $n_i = 0$ is unstable, and that with
 216 $n_r = 0$ and $n_i \neq 0$ is stable against small perturbations.

217 Finally, the IE factor, $f(t) = n_i(t)/n_i(0)$, evolves as

$$218 \quad f(t) = \frac{1 + N'n_r(0)/n_i(0)}{1 + N'n_r(0)/n_i(0) e^{-K(N'n_r(0) + n_i(0))t}}$$

219 and the eventual ice enhancement reaches the maximum $f = 1 + N'n_r(0)/n_i(0)$ as $t \rightarrow \infty$.

220 *b. Perturbation Analysis*

221 A particular interest here is how the ice–number evolution is modified by slightly modifying its
 222 value by n'_{i^*} . As a result, the rain–water number is also perturbed by, say, n'_{r^*} . In this isolated
 223 setting, the total number density is conserved, thus we may set

$$224 \quad n'_{r^*} = -n'_{i^*}. \quad (3.7)$$

225 The perturbation equation for n'_{i^*} is given by

$$226 \quad \dot{n}'_{i^*} = \hat{K}(n_{i^*}n'_{r^*} + n_{r^*}n'_{i^*})$$

227 or by substituting the number–concentration constraint (3.7),

$$228 \quad \dot{n}'_{i^*} = \lambda n'_{i^*},$$

229 where

$$230 \quad \lambda = \hat{K}(n_{r^*} - n_{i^*}) \quad (3.8)$$

231 may be considered a feedback parameter characterizing this perturbation evolution. We see that
 232 at the initial stage when the rain–water is relatively abundant, $n_{r^*} - n_{i^*} > 0$, and the feedback is
 233 positive (splintering feedback), whereas when the ice number has substantially multiplied, $n_{r^*} -$
 234 $n_{i^*} < 0$, and the feedback becomes negative (drop–depletion feedback)

235 For any system in general, a feedback parameter may be defined as the rate of change of a
 236 forcing, Q , of system evolution with respect to its response (a change in its state variable, x) with
 237 $\lambda = dQ/dx$. Here, $\lambda > 0$ for a net positive feedback and $\lambda < 0$ for a net negative feedback. The
 238 forcing may be a function of variables quantifying several processes ($Q = Q(x, X_1, X_2, \dots, X_n)$). By
 239 performing this derivative as a partial derivative with respect to only one process variable, then the
 240 derivative quantifies the feedback from that process ($\lambda_i = (\partial Q / \partial X_i)(dX_i/dx)$). The total feedback
 241 parameter is the sum of partial derivatives corresponding to all the processes by the chain rule:

242 $\lambda = \sum_i \lambda_i$. Thus the net feedback parameter has contributions from all the positive and negative
 243 feedbacks of the system.

244 One can regard the above evolution equation ($\dot{n}_{i^*} = \hat{K}n_{r^*}n_{i^*}$) for ice crystal number as arising
 245 from two separate processes: splintering of freezing drops, controlled by n_{r^*} , and depletion of
 246 drops by collision with crystals, controlled by n_{i^*} . The evolution equation of the system (Eq. 3.8)
 247 may be viewed as a special case of a more general equation, $\dot{n}_{i^*} = Q$ where $Q = Q(n_{i^*}, n_{r^*})$,
 248 with rain and ice concentrations being hypothetically ‘independent’ contributions to the forcing Q
 249 causing a response in ice concentration. The feedback strength has contributions from both pro-
 250 cesses: $\lambda = dQ/dn_{i^*} = \partial Q/\partial n_{i^*} + \partial Q/\partial n_{r^*} dn_{r^*}/dn_{i^*} = \lambda_1 + \lambda_2$ and with dn_{r^*}/dn_{i^*} expressing
 251 the linkage between rain and ice concentrations by the evolution equation for the drop deple-
 252 tion process. Considering $Q = \hat{K}n_{r^*}n_{i^*}$, the feedback parameter thus has the two contributions,
 253 $\lambda_1 = \partial Q/\partial n_{i^*} = \hat{K}n_{r^*} > 0$ measuring the feedback from splintering (a positive feedback) and
 254 $\lambda_2 = \partial Q/\partial n_{r^*} dn_{r^*}/dn_{i^*} = \hat{K}n_{i^*} \times (-1) < 0$ measuring the feedback from drop depletion (a neg-
 255 ative feedback). Thus, we arrive again at the same equation as above, $\lambda = \lambda_1 + \lambda_2 = \hat{K}(n_{r^*} - n_{i^*})$.

256 The identified characteristics of the system is summarized in the phase-space of (n_{r^*}, n_{i^*}) in
 257 Fig. 2. Positive and negative feedbacks ($\lambda > 0$ and $\lambda < 0$) are realized over the regions of insta-
 258 bility and stability (separated by the thick line, $n_{i^*} = n_{r^*}$), where splintering and drop-depletion
 259 feedbacks prevail respectively. The system travels along the trajectory (line with arrow, in which
 260 $n_{r^*} + n_{i^*} = n_0$) from one equilibrium point that is unstable ($n_{r^*} = n_0$) to the other that is stable
 261 ($n_{i^*} = n_0$). While travelling along the trajectory, the system goes from the realm of instability
 262 ($n_{r^*} > n_{i^*}$) to that of stability ($n_{r^*} < n_{i^*}$).

263 4. General Analysis

264 When the general case with $\hat{c}_0 \neq 0$, $\hat{c}_i \neq 0$, $\varepsilon_s \neq 0$ is considered, a full analytical solution is no
 265 longer available, thus various asymptotic expansion methods are called for.

266 *a. Feedback Analysis*

267 Probably the most useful starting point is to note that both the supercooled–drop and ice–particle
 268 number densities, n_{r^*} and n_{i^*} , can evolve independently in absence of the drop freezing and the
 269 subsequent ice fragmentation. We may consider this as a leading–order behaviour of the system
 270 (Eq 2.4) by setting $\hat{K} = 0$:

$$271 \quad n_{r^*}^{(0)} = (n_{r^*}(0) - \frac{\hat{c}_0}{\epsilon_s})e^{-\epsilon_s t^*} + \frac{\hat{c}_0}{\epsilon_s}, \quad (4.1a)$$

$$272 \quad n_{i^*}^{(0)} = n_{i^*}(0) + \hat{c}_i t^*. \quad (4.1b)$$

274 The ice–fragmentation process is introduced as a process at $O(\hat{K})$. Here, taking \hat{K} as a per-
 275 turbation parameter for the asymptotic expansion is merely a device for analytically inferring the
 276 behavior of the system. Recall that in fact, $\hat{K} = 1$ by definition. However, in this manner, we
 277 can investigate how the evolution of the system is modified in the presence of ice fragmentation.
 278 Physically, this amounts for investigating the feedback to the system due to a presence of ice
 279 fragmentation.

280 Thus, a full solution may be approximately obtained by adding an $O(\hat{K})$ –contribution to the
 281 above leading–order solution:

$$282 \quad n_{r^*} = n_{r^*}^{(0)} + \hat{K} n_{r^*}^{(1)}, \quad (4.2a)$$

$$283 \quad n_{i^*} = n_{i^*}^{(0)} + \hat{K} n_{i^*}^{(1)}. \quad (4.2b)$$

285 The $O(\hat{K})$ –equations are given by

$$286 \quad \dot{n}_{r^*}^{(1)} = -n_{r^*}^{(0)} n_{i^*}^{(0)} - \epsilon_s n_{r^*}^{(1)}, \quad (4.3a)$$

$$287 \quad \dot{n}_{i^*}^{(1)} = n_{r^*}^{(0)} n_{i^*}^{(0)}. \quad (4.3b)$$

289 Here, note that the first term, $n_{r^*}^{(0)} n_{i^*}^{(0)}$, in the left–hand side of both equations represents the feed-
 290 back to the system due to the ice–fragmentation process. It may also be important to note that this

291 term is nonlinear, but since this term is given, the above equations can readily be transformed into
 292 an integral form

$$293 \quad n_{r*}^{(1)} = -e^{-\varepsilon_s t_*} \int_0^{t_*} n_{r*}^{(0)} n_{i*}^{(0)} e^{\varepsilon_s t} dt_*, \quad (4.4a)$$

$$294 \quad n_{i*}^{(1)} = \int_0^{t_*} n_{r*}^{(0)} n_{i*}^{(0)} dt. \quad (4.4b)$$

296 By substituting the leading-order solutions (4.1a, b) into the above, and performing the integrals,
 297 we obtain

$$298 \quad n_{r*}^{(1)} = \left[\frac{\hat{c}_0}{\varepsilon_s^2} (n_{i*}(0) - \frac{\hat{c}_i}{\varepsilon_s}) - (n_{r*}(0) - \frac{\hat{c}_0}{\varepsilon_s}) (n_{i*}(0) t_* + \frac{\hat{c}_i}{2} t_*^2) \right] e^{-\varepsilon_s t_*} \\
 299 \quad - \frac{\hat{c}_0}{\varepsilon_s^2} \left[(n_{i*}(0) - \frac{\hat{c}_i}{\varepsilon_s}) + \hat{c}_i t_* \right], \quad (4.5a)$$

$$300 \quad n_{i*}^{(1)} = (n_{r*}(0) - \frac{\hat{c}_0}{\varepsilon_s}) \frac{n_{i*}(0)}{\varepsilon_s} (1 - e^{-\varepsilon_s t_*}) \\
 301 \quad + \frac{\hat{c}_i}{\varepsilon_s} (n_{r*}(0) - \frac{\hat{c}_0}{\varepsilon_s}) \left[\frac{1}{\varepsilon_s} - \left(\frac{1}{\varepsilon_s} + t_* \right) e^{-\varepsilon_s t_*} \right] + \frac{\hat{c}_0}{\varepsilon_s} (n_{i*}(0) t_* + \frac{\hat{c}_i}{2} t_*^2). \quad (4.5b)$$

303 We see that apart from the exponentially decaying tendency associated with the sedimentation, the
 304 feedback is merely algebraic with no obvious destabilization tendency to the system.

305 The solutions may be further simplified by setting the initial condition to $n_{r*}(0) = \hat{c}_0/\varepsilon_s$ and
 306 $n_{i*}(0) = 0$:

$$307 \quad n_{r*}^{(1)} = -\frac{\hat{c}_0 \hat{c}_i}{\varepsilon_s^2} \left[\frac{1}{\varepsilon_s} (e^{-\varepsilon_s t_*} - 1) + t_* \right], \quad (4.6a)$$

$$308 \quad n_{i*}^{(1)} = \frac{1}{2} \frac{\hat{c}_i \hat{c}_0}{\varepsilon_s} t_*^2. \quad (4.6b)$$

310 The solution by this feedback analysis would be indicative of the evolution of the system at onset
 311 of ice fragmentation. Note that the solution for n_{r*} may further be approximated to

$$312 \quad n_{r*}^{(1)} \simeq -\frac{1}{2} \frac{\hat{c}_i \hat{c}_0}{\varepsilon_s} t_*^2 \quad (4.7)$$

313 to the limit of $t \rightarrow 0$. We see that the dimensionless concentrations of rain water and ice crystals
 314 decrease and increase at the same rate, initially. This is consistent with the splintering positive

315 feedback of ice multiplication (noted above for the simplest system) being boosted by the rates
 316 of primary generation of crystals and drops, as well as by the fall-out time of drops being long
 317 (compared to the characteristic time for glaciation). Fall-out of drops has a damping effect on the
 318 splintering positive feedback.

319 *b. Taylor–Expansion Solution*

320 The feedback analysis of the last subsection suggests that the initial tendency of the system due
 321 to the ice fragmentation is merely algebraic. It further suggests that an exact initial tendency of the
 322 system may be obtained by directly applying the Taylor expansion to the solution. Thus, we set

$$323 \quad n_{r*} = \sum_{j=0}^{\infty} n_{r*,j} t_*^j, \quad (4.8a)$$

$$324 \quad n_{i*} = \sum_{j=0}^{\infty} n_{i*,j} t_*^j. \quad (4.8b)$$

326 In substitution of these Taylor expansions into the original full equations (2.4a, b) leads to a solu-
 327 tion. In deriving the solution, we note particularly

$$328 \quad n_{r*} n_{i*} = \sum_{j=0}^{\infty} \left[\sum_{l=0}^j n_{r*,l} n_{i*,j-l} \right] t_*^j.$$

329 With $j = 0$, we obtain

$$330 \quad n_{r*,1} = \hat{c}_0 - \hat{K} n_{r*,0} n_{i*,0} - \varepsilon_S n_{r*,0}, \quad (4.9a)$$

$$331 \quad n_{i*,1} = \hat{c}_i + \hat{K} n_{r*,0} n_{i*,0}. \quad (4.9b)$$

333 Here, $n_{r*,0}$ and $n_{i*,0}$ are defined by the initial conditions. With $j = 1$, we further obtain

$$334 \quad n_{r*,2} = -\hat{K}(n_{r*,0} n_{i*,1} + n_{r*,1} n_{i*,0}) - \varepsilon_S n_{r*,1}, \quad (4.10a)$$

$$335 \quad n_{i*,2} = \hat{K}(n_{r*,0} n_{i*,1} + n_{r*,1} n_{i*,0}). \quad (4.10b)$$

337 In order to simplify the expressions, again, we set $n_{r^*,0} = \hat{c}_0/\varepsilon_s$ and $n_{i^*,0} = 0$. Then we obtain

$$338 \quad n_{r^*,1} = 0,$$

$$339 \quad n_{i^*,1} = \hat{c}_i,$$

$$340 \quad n_{r^*,2} = -\hat{K} \frac{\hat{c}_i \hat{c}_0}{\varepsilon_s},$$

$$341 \quad n_{i^*,2} = \hat{K} \frac{\hat{c}_i \hat{c}_0}{\varepsilon_s}.$$

343 Putting them together, the initial tendency of the solution is given by

$$344 \quad n_{r^*} = \frac{\hat{c}_0}{\varepsilon_s} (1 - \hat{K} \hat{c}_i t_*^2) + \dots, \quad (4.11a)$$

$$345 \quad n_{i^*} = \hat{c}_i (t_* + \frac{\hat{K} \hat{c}_0}{\varepsilon_s} t_*^2) + \dots. \quad (4.11b)$$

347 This solution (4.11a, b) may be compared with (4.7) and (4.6b) obtained in the last subsection. We
 348 see that the feedback analysis underestimates the effect of the ice fragmentation by half due to its
 349 perturbation treatment.

350 *c. Feedback Analysis with Distorted Physics*

351 A variation to the feedback analysis in Sec. 4.a is to differentiate the strength of the ice freezing–
 352 fragmentation to the hydrometeor types. Thus, we re–write Eqs. (2.4a, b) as

$$353 \quad \dot{n}_{r^*} = \hat{c}_0 - \hat{K}_1 n_{i^*} n_{r^*} - \varepsilon_s n_{r^*}, \quad (4.12a)$$

$$354 \quad \dot{n}_{i^*} = \hat{c}_i + \hat{K}_2 n_{r^*} n_{i^*}. \quad (4.12b)$$

356 Here, we may artificially assume that the freezing fragmentation is negligible for the supercooled
 357 drops, thus $\hat{K}_1 \ll 1$, and the effect of freezing–fragmentation is only felt by the ice particles to the
 358 leading order, assuming $\hat{K}_2 = O(1)$. Thus, we solve the problem with the following asymptotic

359 expansion:

$$360 \quad n_{r_*} = n_{r_*}^{(0)} + \hat{K}_1 n_{r_*}^{(1)} + \dots$$

$$361 \quad n_{i_*} = n_{i_*}^{(0)} + \hat{K}_1 n_{i_*}^{(1)} + \dots$$

363 Though such an assumption is hardly justified from a physical basis, this procedure provides a
 364 useful insight into the evolution of the system during ice fragmentation. For this reason, we shall
 365 call this method the *feedback analysis with distorted physics*.

366 The leading-order solution for n_{r_*} is, thus, given by Eq. (4.1a), whereas we solve the full prob-
 367 lem for n_{i_*} to the leading order:

$$368 \quad \left(\frac{d}{dt_*} - \hat{K}_2 n_{r_*}^{(0)} \right) n_{i_*}^{(0)} = \hat{c}_{i_*}$$

369 This equation is linear in respect to $n_{i_*}^{(0)}$, and its solution can easily be written in an integral form:

$$370 \quad n_{i_*}^{(0)} = [n_{i_*}(0) + \int_0^{t_*} \hat{c}_i \exp(-\int_0^{t'_*} \hat{K}_2 n_{r_*}^{(0)} dt''_*) dt'_*] \exp(\hat{K}_2 \int_0^{t_*} n_{r_*}^{(0)} dt_*)$$

371 The second integral is readily performed and we obtain

$$372 \quad \int_0^{t_*} n_{r_*}^{(0)} dt_* = \frac{1}{\varepsilon_s} (n_{r_*}(0) - \frac{\hat{c}_0}{\varepsilon_s}) (1 - e^{-\varepsilon_s t_*}) + \frac{\hat{c}_0}{\varepsilon_s} t_*$$

373 However, the first integral

$$374 \quad \int_0^{t_*} \exp(-\hat{K}_2 \int_0^{t'_*} n_{r_*}^{(0)} dt''_*) dt_* = \int_0^{t_*} \exp[-\frac{\hat{K}_2}{\varepsilon_s} (n_{r_*}^{(0)}(0) - \frac{\hat{c}_0}{\varepsilon_s}) (1 - e^{-\varepsilon_s t_*}) - \frac{\hat{K}_2 \hat{c}_0}{\varepsilon_s} t_*] dt_*$$

375 is not readily integrable, thus we focus on the two limits, $t_* \sim 0$ and $t_* \rightarrow \infty$.

376 (i) When $t_* \sim 0$, noting $1 - e^{-\varepsilon_s t_*} \simeq \varepsilon_s t_*$, the integral becomes

$$377 \quad \int_0^{t_*} \exp(-\hat{K}_2 \int_0^{t'_*} n_{r_*}^{(0)} dt''_*) dt_* \simeq \frac{1}{\hat{K}_2 n_{r_*}^{(0)}(0)} (1 - e^{-\hat{K}_2 n_{r_*}^{(0)}(0) t_*}),$$

378 and the solution is

$$379 \quad n_{i_*}^{(0)} \simeq (n_{i_*}^{(0)}(0) + \frac{\hat{c}_i}{\hat{k}_2 n_{r_*}^{(0)}(0)}) e^{\hat{K}_2 n_{r_*}^{(0)}(0) t} - \frac{\hat{c}_i}{\hat{k}_2 n_{r_*}^{(0)}(0)}. \quad (4.13a)$$

380 (ii) When $t_* \rightarrow \infty$, the integral asymptotically approaches to:

$$381 \int_0^{t_*} \exp(-\hat{K}_2 \int_0^{t'_*} n_{r_*}^{(0)} dt''_*) dt_* \rightarrow -\frac{\varepsilon_s}{\hat{K}_2 \hat{c}_0} \exp[-\frac{\hat{K}_2}{\varepsilon_s} (n_{r_*}^{(0)}(0) - \frac{\hat{c}_0}{\varepsilon_s}) - \frac{\hat{K}_2 \hat{c}_0}{\varepsilon_s} t_*] + \frac{1}{\hat{K}_2 n_{r_*}^{(0)}(0)},$$

382 thus

$$383 n_{i_*}^{(0)} \rightarrow (n_{i_*}^{(0)}(0) + \frac{\hat{c}_i}{\hat{K}_2 n_{r_*}^{(0)}(0)}) \exp[\frac{\hat{K}_2}{\varepsilon_s} (n_{r_*}^{(0)}(0) - \frac{\hat{c}_0}{\varepsilon_s}) + \frac{\hat{K}_2 \hat{c}_0}{\varepsilon_s} t_*] - \frac{\varepsilon_s \hat{c}_i}{\hat{K}_2 \hat{c}_0}. \quad (4.13b)$$

384 It is seen that in both limits, the ice–crystal number increases exponentially with time when the
385 decrease of supercooled drops by freezing is neglected.

386 We now turn to $O(\hat{K}_1)$ in order to see the modifications of the both particles numbers by the
387 exponential ice–fragmentational multiplication:

$$388 (\frac{d}{dt_*} + \varepsilon_s) n_{r_*}^{(1)} = -n_{r_*}^{(0)} n_{i_*}^{(0)}, \quad (4.14a)$$

$$389 (\frac{d}{dt_*} - \frac{\hat{K}_2}{\hat{K}_1} n_{r_*}^{(0)}) n_{i_*}^{(1)} = \frac{\hat{K}_2}{\hat{K}_1} n_{r_*}^{(1)} n_{i_*}^{(0)}. \quad (4.14b)$$

391 Though it is possible to explicitly write down the full asymptotic solutions for the same two
392 limits as considered for the leading order, the expressions are lengthy, and not particularly illu-
393 minating. For this reason, we focus on the case with $n_{r_*}(0) = \hat{c}_0/\varepsilon_s$ and $n_{i_*}(0) = 0$. Note that
394 coincidentally, in this case, the two leading–order solutions (4.13a, b) agree. We also focus only
395 on the most dominant exponential term in the solution of Eqs. (4.14a, b), neglecting the various
396 exponentially–decaying terms that follow.

397 After these simplifications, the first–order modification to the supercooled rain density, n_{r_*} , due
398 to the freezing is given by:

$$399 n_{r_*}^{(1)} = -\frac{\varepsilon_s \hat{c}_i}{\hat{K}_2 (\hat{K}_2 \hat{c}_0 + \varepsilon_s^2)} e^{(\hat{K}_2 \hat{c}_0 / \varepsilon_s) t_*} + \dots \quad (4.15a)$$

400 The result shows that as the first–order effect, collision freezing exponentially depletes the su-
401 percooled water due to the exponential multiplication of ice. If this result is taken literally, the
402 supercooled water would be completely depleted over a finite time. In reality, as the supercooled

403 water begins to deplete, the ice fragmentation also slows down, thus it is more likely that super-
 404 cooled water will be depleted in slower rate.

405 In turn, the first-order modification to the ice-particle number is:

$$406 \quad n_{i*}^{(1)} = -\frac{\hat{c}_0 \hat{c}_i}{\hat{K}_2 \hat{c}_0 + \varepsilon_s^2} \left[\frac{\hat{K}_2^2 \hat{c}_0^2 + \varepsilon_s^2 \hat{K}_2 \hat{c}_0 + \varepsilon_s^4}{\varepsilon_s \hat{K}_2 \hat{c}_0 (\hat{K}_2 \hat{c}_0 + \varepsilon_s^2)} + \frac{\hat{c}_0 \hat{c}_i}{\hat{K}_2 \hat{c}_0 + \varepsilon_s^2} t_* \right] e^{\hat{K}_2 n_{r*}^{(0)}(0) t_*} + \dots \quad (4.15b)$$

407 We see the exponential ice multiplication tendency is first, suppressed by the constant factor (the
 408 first term), and further decreases linearly with time (the second term). Thus, the dominant expo-
 409 nential term becomes negative over a finite term, and the only remaining positive contributions are
 410 neglected exponentially-decaying terms. We interpret that the result suggests that though the ice
 411 multiplies with time, the growth is weaker than exponential.

412 *d. Asymptotic Tendency towards $t_* \rightarrow \infty$*

413 The analysis of the last subsection suggests (though it does not show) that the supercooled water
 414 would deplete with a rate slower than exponential, and also the ice multiplies by fragmentation
 415 with a rate slower than exponential. The most likely case is that the supercooled water decreases,
 416 and the ice particles increase both algebraically with time. Thus, to the asymptotic limit towards
 417 $t_* \rightarrow \infty$, we may set

$$418 \quad n_{r*} \sim n_{r*0} t_*^{-\alpha}, \quad (4.16a)$$

$$419 \quad n_{i*} \sim n_{i*0} t_*^\beta. \quad (4.16b)$$

421 Here, both α and β are expected to be positive constants, and the symbol \sim suggests that we are
 422 only concerned with the asymptotic tendency toward $t_* \rightarrow \infty$.

423 Substitution of the asymptotic expressions (4.16a, b) into Eqs. (2.4a, b) leads to

$$424 \quad -\alpha n_{r*0} t_*^{-(\alpha+1)} \sim \hat{c}_0 - \hat{K} n_{i*0} n_{r*0} t_*^{-\alpha+\beta} - \varepsilon_s n_{r*0} t_*^{-\alpha}, \quad (4.17a)$$

$$425 \quad \beta n_{i*0} t_*^{\beta-1} \sim \hat{c}_{i*} + \hat{K} n_{r*0} n_{i*0} t_*^{-\alpha+\beta}. \quad (4.17b)$$

426

427 In order this asymptotic expression to be valid, the most dominant terms in terms of the power in
 428 t_* must balance.

429 In Eq. (4.17a), we find $t_*^{-(\alpha+1)} \ll t_*^{-\alpha} \ll t_*^{-\alpha+\beta}$, thus the dominant expected asymptotic balance
 430 is:

$$431 \quad \hat{c}_0 - \hat{K} n_{i*0} n_{r*0} t_*^{-\alpha+\beta} \sim 0. \quad (4.18)$$

432 This balance is achieved when $-\alpha + \beta = 0$, or $\alpha = \beta$. It may be important to note that in asymp-
 433 totic limit of $t_* \rightarrow \infty$, the equation for the supercooled rain number becomes quasi-stationary (*i.e.*,
 434 the temporal tendency in the left hand side does not contribute to the leading order), and also the
 435 sedimentation no longer plays a leading role (*i.e.*, the 3rd term is the right hand side does not
 436 contribute to the leading order).

437 By substituting $\alpha = \beta$ into Eq. (4.17b), we find

$$438 \quad \beta n_{i*0} t_*^{\beta-1} \sim \hat{c}_{i*} + \hat{K} n_{r*0} n_{i*0}.$$

439 Since the right hand side is positive definite, it must be balanced by a constant growing tendency
 440 of the ice number, thus

$$441 \quad \beta = 1,$$

442 and also $\alpha = 1$ as a result.

443 Constants, n_{r*0} and n_{i*0} , may be determined in a more straightforward manner by considering
 444 the conservation of the total particle number, which is obtained by taking the sum of Eqs. (2.4a, b):

$$445 \quad \frac{d}{dt_*} (n_{r*} + n_{i*}) = \hat{c}_0 + \hat{c}_i - \varepsilon n_{r*}.$$

446 As $t_* \rightarrow \infty$, $n_{r*} \rightarrow 0$, thus the above conservation law asymptotically approaches to

$$447 \quad \dot{n}_{i*} \sim \hat{c}_0 + \hat{c}_i. \quad (4.19)$$

448 Substituting Eq. (4.16a) into Eq. (4.19) with $\beta = 1$, we immediately obtain $n_{i*0} = \hat{c}_0 + \hat{c}_i$. By
 449 further substituting this result into Eq. (4.18), we find $n_{r*0} = \hat{c}_0[\hat{K}(\hat{c}_0 + \hat{c}_i)]^{-1}$. Thus, the asymptotic
 450 solution towards $t \rightarrow \infty$ becomes

$$451 \quad n_{r*} \sim \frac{\hat{c}_0}{\hat{K}(\hat{c}_0 + \hat{c}_i)} t_*^{-1}, \quad (4.20a)$$

$$452 \quad n_{i*} \sim (\hat{c}_0 + \hat{c}_i) t_*. \quad (4.20b)$$

454 In the limit of the long time scale, the ice crystals multiply by fragmentation linearly with time
 455 at a rate defined by the sum of the supercooled-drop and primary-ice sources.

456 Finally, the asymptotic solution becomes when dimensions are included:

$$457 \quad n_r \sim \frac{c_0}{\hat{K}K(N'c_0 + c_i)} t^{-1}, \quad (4.20a)$$

$$458 \quad n_i \sim (N'c_0 + c_i)t. \quad (4.20b)$$

460 The definition of IE ratio, $f \equiv n_i^*(t^*)/n_i^{*(0)}(t^*)$, is the ratio of the total ice concentration to
 461 the primary ice concentration. Thus it is a measure of the extent of secondary ice production.

462 So in the general case, if \hat{c}_i is non-zero and $t \rightarrow \infty$:

$$463 \quad f = (N'c_0/c_i + 1)/(n_i(t=0)/(tc_i) + 1) \rightarrow 1 + N'c_0/c_i \quad (1)$$

464 The maximum IE ratio is approached on a timescale of $n_i(t=0)/c_i$.

465 Otherwise without continuous primary ice nucleation, $\hat{c}_i = 0$ implies:

$$466 \quad f(t) \rightarrow N'c_0t/(n_i(t=0))$$

467 taking the dimensional expressions for the source terms. Now there is no upper limit on the IE
 468 ratio, and it increases with time at a constant rate of $N'c_0/n_i(t=0)$.

469 The supercooled–drop number decreases only weakly with time at a rate $\sim t_*^{-1}$ maintaining a
 470 quasi–stationarity with the generation rate balancing with the collision freezing rate. The rain
 471 sedimentation does not play a leading role.

472 *e. Perturbation Analysis*

473 Finally, the perturbation analysis considered in Sec. 3.b may also be performed for the full
 474 problem. However, the procedure is more involved with the fact that the source and the sink terms
 475 are involved in the full system, thus the total number density is no longer conserved, and it is no
 476 longer possible to invoke a simple constraint as Eq. (3.7). Instead, we have to consider explicitly
 477 the two perturbation equations for both n'_{r^*} and n'_{i^*} :

$$478 \quad \dot{n}'_{r^*} = -(\hat{K}n_{i^*} + \varepsilon_s)n'_{r^*} - \hat{K}n_{r^*}n'_{i^*}, \quad (4.21a)$$

$$479 \quad \dot{n}'_{i^*} = \hat{K}(n_{i^*}n'_{r^*} + n_{r^*}n'_{i^*}). \quad (4.21b)$$

481 The analysis of this system is further involved due to the fact that the linear operator in the right–
 482 hand side also evolves with time. However, the problem can be simplified, as implicitly assumed
 483 in Sec. 3.b, when the evolution of the reference state, (n_{r^*}, n_{i^*}) , is considered much slower than
 484 that of the perturbations. In this case, the time derivative in the left–hand side may be replaced by
 485 an eigenvalue, λ , and the above problem reduces to that of finding the eigenvalues for the linear
 486 operator in the right hand side, which is determined from

$$487 \quad \lambda^2 - (\hat{K}n_{r^*} - \hat{K}n_{i^*} - \varepsilon_s)\lambda + \hat{K}^2n_{i^*}n_{r^*} - (\hat{K}n_{i^*} + \varepsilon_s)\hat{K}n_{r^*} = 0.$$

488 Solving it for λ , we obtain the two eigenvalues (adding the subscripts 1 and 2 for distinction):

$$489 \quad \lambda_1 = \frac{(\hat{K}n_{r^*} - \hat{K}n_{i^*} - \varepsilon_s)}{2} \left[1 + \left\{ 1 + \frac{4\varepsilon_s\hat{K}n_{r^*}}{(\hat{K}n_{r^*} - \hat{K}n_{i^*} - \varepsilon_s)^2} \right\}^{1/2} \right],$$

$$490 \quad \lambda_2 = \frac{(\hat{K}n_{r^*} - \hat{K}n_{i^*} - \varepsilon_s)}{2} \left[1 - \left\{ 1 + \frac{4\varepsilon_s\hat{K}n_{r^*}}{(\hat{K}n_{r^*} - \hat{K}n_{i^*} - \varepsilon_s)^2} \right\}^{1/2} \right].$$

491

492 The first eigenvalue reduces to Eq. (3.8) in the limit of $\varepsilon_s \rightarrow 0$, whereas the second reduces to
 493 $\lambda_2 = 0$. The latter is simply interpreted as a manifestation of the conservation of the total particle
 494 number number (3.7).

495 When a finite sedimentation rate ($\varepsilon_s \neq 0$) is considered, the threshold from the positive to neg-
 496 ative feedback for λ_1 reduces by ε_s (*i.e.*, bias towards a negative “feedback”). Thus, the neutral
 497 line shifts from $n_{i*} = n_{r*}$ to $n_{i*} = n_{r*} - \varepsilon_s$: the critical ice number density becomes smaller in the
 498 presence of sedimentation. Some examples of the neutral lines are plotted in Fig. 3a for selective
 499 sedimentation rates, ε_s . In the presence of sedimentation, however, the neutral line no longer sep-
 500 arates between the stable and the unstable regimes, but the stable regime upper left of the neutral
 501 line identified in Fig. 2 also destabilizes due to the second eigenvalue, λ_2 . Most importantly, in
 502 these general situations, the perturbation evolution of the system is no longer interpreted in terms
 503 of a single “perturbation” parameter, but the actual perturbation evolution is determined by a linear
 504 combination of two exponential tendencies characterized by two eigenvalues, sensitively depend-
 505 ing on the initial perturbation. Trajectories in the phase-space are shown in Fig. 3a for the special
 506 case of $\varepsilon_s = 0$ and $\hat{c}_i = 0$ by numerical integration of Eq (2.4), with primary ice only implicitly
 507 represented by an initial nonzero value of ice concentration, $n_{i*}(t^* = 0) = 0.001$. The simulations
 508 are integrated until $t^* = 10^3$.

509 The evolution over time of the IE ratio, f , for the same trajectories is shown in Fig. 3b. Two
 510 stages of the glaciation are evident. First, there is the usual exponential growth of the ice concen-
 511 tration by an IE ratio of up to 10^3 in a dimensionless time of $t^* < 1-10$. During this stage, the
 512 rain concentration is depleted almost to zero. If the integration of the corresponding dimensional
 513 equations were performed, this initial stage would correspond to the time-scale of explosive frag-
 514 mentation noted above of $\tau = 1/(K(n_i(0) + n_r(0)N')$). Second, there is the subsequent exponential
 515 relaxation to the asymptotic solution when $t^* > 1-10$, coinciding with a balance between supply

516 of fresh drops ($\hat{c}_0 > 0$) and their depletion by fragmentation and collisions with ice splinters. In
517 this second stage, the rain concentration decreases steadily, inversely proportional with time.

518 **5. Discussions**

519 *a. Efficiency of the Ice Multiplication Processes*

520 Our theoretical investigation reveals that the ice multiplication by fragmentation of the freezing
521 drops is not always as potentially effective as the two other ice multiplication processes examined
522 by Yano and Phillips (2011), and Yano *et al.* (2016): Hallett–Mossop and ice–ice collision pro-
523 cesses. As shown in the Appendix C of Yano and Phillips (2011), the ice multiplication rate by the
524 Hallett–Mossop process is linearly proportional to the existing graupel number density. This leads
525 to an exponential multiplication of ice with time, assuming that the system is in quasi–equilibrium
526 between the ice particles and graupel, and also the supercooled cloud–droplets are re–adjusted to
527 an equilibrium number as graupel sweeps them through as the latter falls. As carefully analyzed
528 in Yano *et al.* (2011: see the Appendix especially), the ice multiplication by ice–ice collision is
529 fundamentally nonlinear, being proportional to the square of the ice–number density. In the ideal-
530 ized state with infinite water–vapor supply, this leads to an explosive increase of the ice number to
531 infinity within a finite time.

532 Compared to those rather strong multiplication tendencies found with these two processes, the
533 ice multiplication by fragmentation of freezing drops often tends to be rather modest. This stems
534 from the fact that this process relies on a number of supercooled drops available in the system,
535 and it essentially limits the possible number of ice fragments that can be generated. When the
536 ice fragmentation by freezing drops is considered in its isolation, as in Sec. 3, this conclusion
537 is especially clear: the final total number of ice fragments found is simply $N'n_r(0) + n_i(0)$ with
538 $N = N' + 1$ the number of secondary ice fragments generated per freezing collision, and $n_r(0)$,
539 $n_i(0)$ are the initial numbers of supercooled drops and ice crystals, respectively.

540 When both supercooled–water rain and primary–ice sources are added to the system, as in Sec. 4,
541 the behavior of the system becomes more involved. However, in the limit of the long time–scale,
542 the ice fragmentation number simply increases linearly with time by the rate that both supercooled
543 water and primary ice are supplied. We see that such a multiplication tendency is, generally, much
544 weaker than any of those found in Hallett–Mossop or ice–ice collision process.

545 However, in some observational situations, fragmentation of freezing drops *does* become a pri-
546 mary process of ice multiplication, especially when there is copious supercooled rain initially and
547 temperatures are optimal. A case during the ICET is such an example.

548 *b. Comparison with the Observation*

549 A quantitative observational comparison with the present theoretical results is provided by Law-
550 son *et al.* (2015), who present the measurements during ICET over U.S. Virgin Islands in July
551 2011. Their flights measuring cloud properties were performed in ascending order so that a
552 Lagrangian interpretation of data following the convective updrafts can be developed. The La-
553 grangian interpretation would also be further facilitated by considering the ice–fragmentation pro-
554 cess in its isolation as in Sec. 3.

555 In this framework, the key conserved quantity is the total particle number with the supercooled
556 drops multiplied by the fragmentation number per collision freezing. It also follows that the total
557 water content, as a sum of liquid and ice, is also conserved. Hence, the scatter plots for liquid and
558 ice values, both in terms of the number density and the water content would form a linear line with
559 a negative slope. The scatter plots using the measurements for varying heights (temperature levels)
560 averaged over various convective cores for both quantities in Fig. 4 clearly present a negative
561 correlation both for the number density (a) and the water content (b), being consistent with the
562 theory presented in Sec. 3, though the distributions are hardly aligned to any line.

563 The strong scatter may be understood by two reasons: 1) the plots are hardly form a single
 564 convective event, but a collection of many convective cores, though all of them are in similar
 565 regime; 2) the source and the loss terms for both liquid and ice, neglected in the theory, would not
 566 be negligible in practice. Considering those two caveats, these two scatter plots would rather be
 567 considered a good support for the idealized theory presented in Sec. 3.

568 A rather surprising result from their data analysis is a fast glaciation process: they found that
 569 3–5 g m⁻³ supercooled liquid are almost all glaciated over 3 min time at an elevation of 1–1.5 km
 570 higher within an updraft core. Here, this rapid glaciation rate is easily explained by the theoretical
 571 analysis in Sec. 3. Note that the collision efficiency is defined by

$$572 \quad K = \pi r_d^2 v_T$$

573 in terms of the radius, r_d , of the supercooled drops, and the differential vertical velocity, v_T ,
 574 between the supercooled drops and ice crystals. Assuming, $r_d \sim 10^2 \mu\text{m} \sim 10^{-4} \text{ m}$, $v_T \sim 1 \text{ m/s}$,
 575 we obtain

$$576 \quad K \sim r_d^2 v_T \sim 10^{-8} \text{ m}^3/\text{s}$$

577 Also setting $n_r \sim 1 \text{ cm}^{-3} \sim 10^6 \text{ m}^{-3}$, $N' \sim 1$, the characteristic glaciation rate is estimated by

$$578 \quad N' K n_r \sim 10^{-1} \text{ 1/s},$$

579 and the characteristic time scale is $\tau \sim 10 \text{ sec}$ by following the definition (3.5). Noting from
 580 Fig. 1 that it typically takes about ten characteristic time scales for completing the glaciation, a
 581 time required for completing the glaciation is also only 10^2 sec , being fairly comparable with the
 582 observational estimate of 3 min.

583 *c. Asymptotic Expansion Approaches*

584 The present study has employed various asymptotic–expansion methods in order to elucidate
 585 the basic behavior of the system for ice fragmentation by collision freezing of supercooled drops.

586 Here, we emphasize that not all the asymptotic solutions presented herein are equally useful. Some
587 of them are not precise even in asymptotic sense, but merely for providing qualitative insights.
588 Nevertheless, they help to identify more rigorous methods for deriving more precise solutions.
589 For example, the feedback analysis in Sec. 4.a suggests the initial tendency of the system is alge-
590 braic in time, that further motivates us to determine the exact initial tendency by the Taylor expan-
591 sion method (Sec. 4.b). Though the exact Taylor–expansion result demonstrates that the feedback
592 analysis halves the initial parabolic tendency, at the same time, without support of the feedback
593 analysis, the Taylor–expansion solution is justified only in a strong limit of $t \rightarrow 0$, because in
594 general, the presence of an exponentially growing tendency invalidates the Taylor–expansion so-
595 lution very rapidly in time. Another example is an approach of distorted physics considered in
596 Sec. 4.c. Again, though this analysis is not quantitatively accurate, the result suggests to consider
597 an algebraic tendency of the solution in the long time–scale limit, as considered in Sec. 4.d.

598 Asymptotic analyses presented herein are hardly exhaustive either, because some asymptotic
599 analyses are not particularly illuminating. For example, it is possible to consider the modification
600 of the solution presented in Sec. 3 by adding the sources, \hat{c}_0 and \hat{c}_i , as perturbation terms. However,
601 such an analysis hardly elucidates a rather drastic change of the system behavior by adding finite
602 sources, as considered in Sec. 4.

603 Some of the analyses turn out to be rather subtle. For example, an alternative feedback analysis
604 by another assumption of distorted physics, $\hat{K}_2 \ll \hat{K}_1 \sim 1$, may also be considered. In this case, the
605 system exponentially approaches new equilibrium states up to first order. Yet it is rather difficult
606 to draw any physically meaningful conclusions from this solution.

607 *d. Stochasticity of the Ice Fragmentation Process*

608 Finally, we should keep in mind that the fragmentation number, N' , of ice per collision freezing is
609 not a fixed number even when a collision between a drop and an ice particle with fixed diameters

610 is considered. One reason is that the number of fragments per spherically frozen drop (mode
611 1) varies by orders of magnitude depending on temperature. Vertical motions due to sloped in-
612 cloud drafts combined with sedimentation create variability of the ambient temperature. Equally,
613 even for identical conditions of freezing temperature and drop size, the number of drop fragments
614 varies dramatically from drop to drop. Macroscopically, it may be assumed to be probabilistic.
615 Kolomeychuk *et al.* 1975 observed that about 10% of all drops in their experiment (1.6 mm)
616 emitted about 90% of all the detected splinters. Considering only drops freezing between -15°C
617 and -18°C , 90% of the splinters were from a third of the drops.

618 Details of the collision process are complex for the most purposes of cloud microphysical mod-
619 eling, and the outcome of the fragmentation number by collision freezing may simply be consid-
620 ered a random number (*cf.*, Kolomeychuk *et al.* 1975). As a result, the ice fragmentation may be
621 considered a stochastic process. This is an aspect, where further investigation may be warranted.
622 Here, we present short general theoretical observations for facilitating further studies.

623 As a whole, the contribution of stochasticity would also be best understood by adding this com-
624 ponent as a perturbation, say, $\delta N'$. In the general case considered in Sec. 4, under the asymptotic
625 limit to the long time scale, the ice–particle number, n_i , asymptotically evolves solely depend-
626 ing on the strength of the two hydrometeor sources. This asymptotic tendency is not affected,
627 even when the fragmentation number fluctuates randomly. The latter effect only affects the time
628 evolution of the supercooled–drop number, n_r . It can be shown that the resulting fluctuation, n'_r ,
629 is dictated by a Brownian motion induced by white noise, $\delta N'$, under a linear drag with a drag
630 coefficient linearly increasing with time.

631 On the other hand, when the ice fragmentation process is considered in isolation, as in Sec. 3, we
632 find that stochastic fluctuations, $\delta N'$, of fragmentation induce multiplicative noise. As found the

633 case for an analogous problem with ice–ice collision multiplication by Yano and Phillips (2016),
634 this multiplicative noise process may contribute to the whole system in a nontrivial manner.

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639 mechanisms for ice production in clouds and their relation of aerosol conditions.

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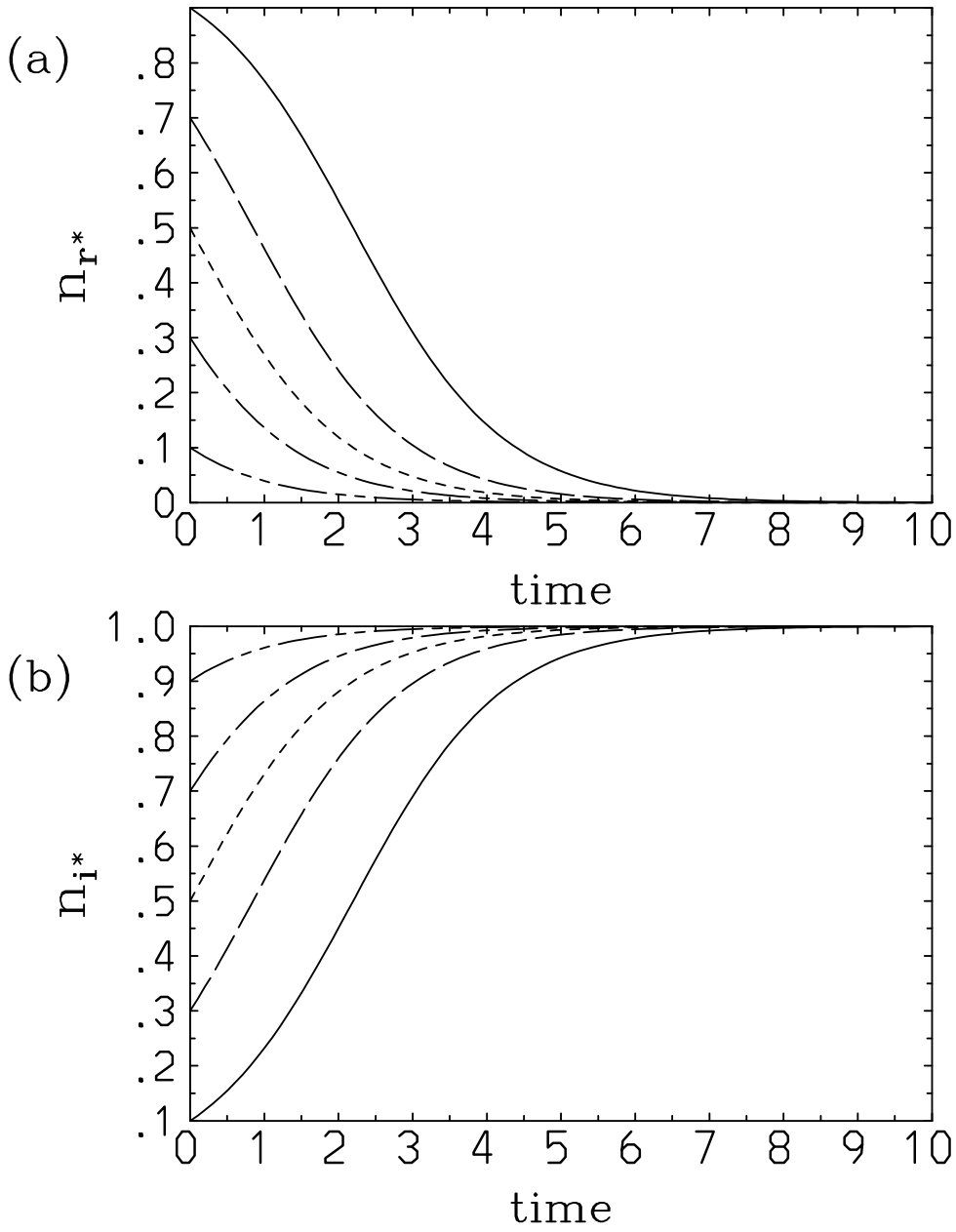
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679	*LIST OF FIGURES	Time evolution of the particle number densities with various	
680		initial conditions when the ice–fragmentation process is considered in its isolation: for (a)	
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695 FIG. 1. Time evolution of the particle number densities with various initial conditions when the ice-
 696 fragmentation process is considered in its isolation: for (a) supercooled drops, n_{r^*} , and (b) ice crystals, n_{i^*} .
 697 All the variables are nondimensionalized as described in the text.

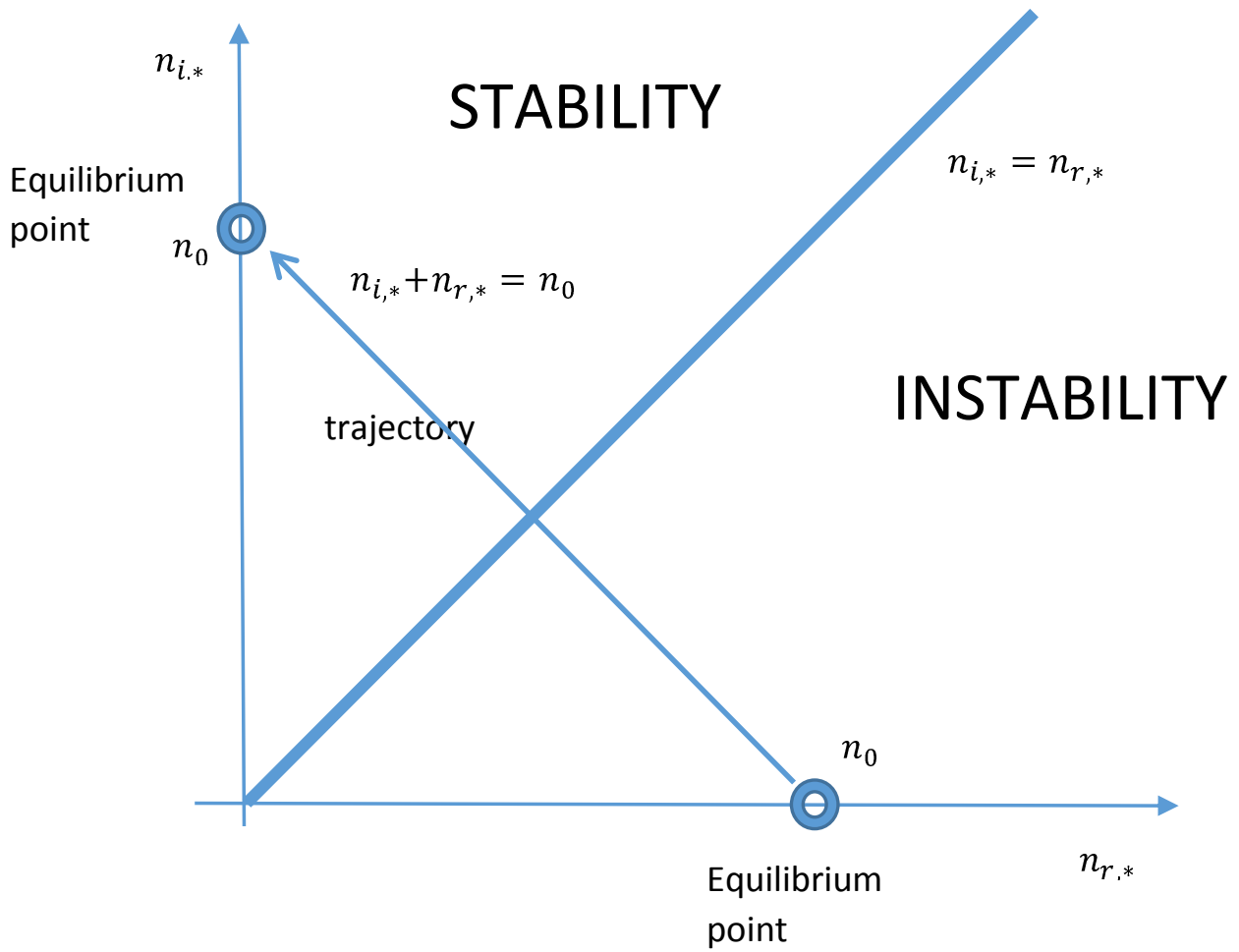
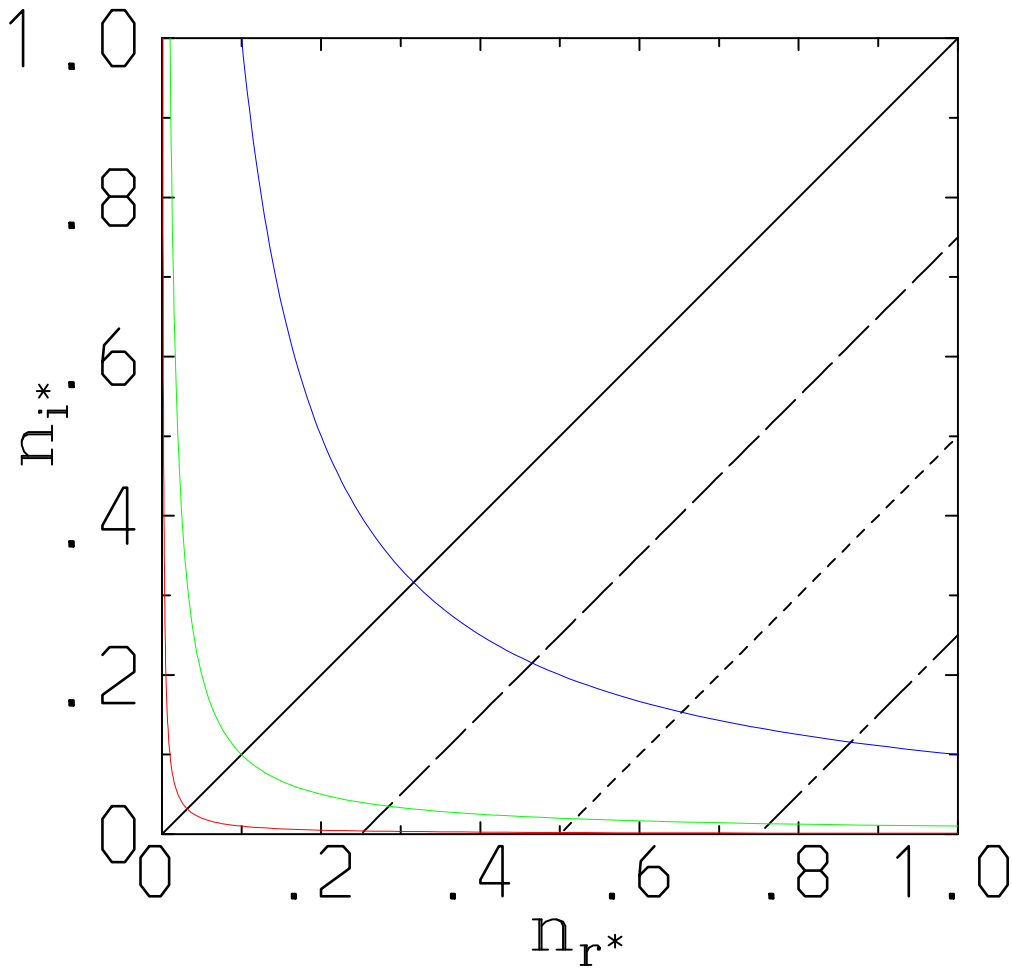
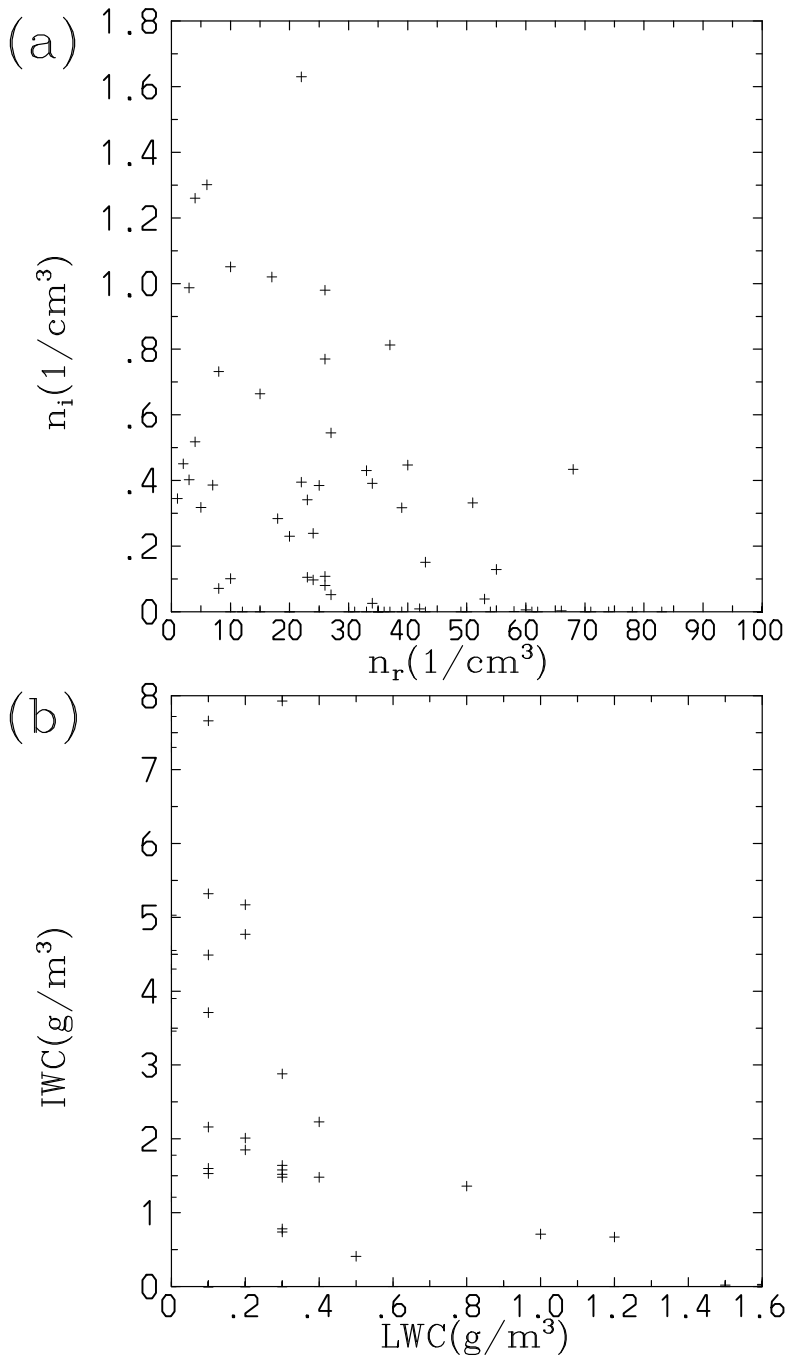


FIG. 2. The stability characteristics of the system when no source and sink is considered as in Sec. 3.



698 FIG. 3. The stability characteristics of the system in the general case: shown are the neutral lines with selective
699 choice of the sedimentation rates: $\varepsilon_s = 0$ (solid), 0.25 (long dash), 0.5 (short dash), and 0.75 (chain dash). Note
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702 shown based on the asymptotic solution (4.20) with $\hat{c}_0 = 10^{-1}$ (blue), 10^{-2} (green), and 10^{-3} (red): the system
703 evolves in the direction of decreasing n_{r^*} and increasing n_{i^*} .



704 FIG. 4. Scatter plots between the liquid and the ice values observed during the ICE-T campaign. The values
 705 marked by + symbol are measurements for updraft cores: (a) particle number density ($1/\text{cm}^3$), (b) water content
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