

# Evaluation of a Quasi-steady state approximation of the cloud Droplet Growth Equation (QDGE) scheme for aerosol activation in global models using multiple aircraft data over both continental and marine environments

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**Abstract.** This research introduces a numerically efficient aerosol activation scheme and evaluates it by using stratus and stratocumulus cloud data sampled during multiple aircraft campaigns in Canada, Chile, Brazil, and China. The scheme  
15 employs a Quasi-steady state approximation of the cloud Droplet Growth Equation (QDGE) to efficiently simulate aerosol activation, the vertical profile of supersaturation, and the activated cloud droplet number concentration (*CDNC*) near the cloud base. The calculated maximum supersaturation values using of the QDGE scheme were compared with multiple parcel model simulations under various aerosol and environmental conditions. The differences are all below 0.18 %, indicating good performance and accuracy of the QDGE scheme. We evaluated the QDGE scheme by specifying observed  
20 environmental thermodynamic variables and aerosol information from 31 cloud cases as input and comparing the simulated *CDNC* with cloud observations. The average of mean relative error (*MRE*) of the simulated *CDNC* for cloud cases in each campaign ranges from 17.30 % in Brazil to 25.90 % in China, indicating that the QDGE scheme successfully reproduces observed variations in *CDNC* over a wide range of different meteorological conditions and aerosol regimes. Additionally, we carried out an error analysis by calculating the Maximum Information Coefficient (MIC) between the mean relative error  
25 (*MRE*) and input variables for the individual campaigns and all cloud cases. MIC values ~~are~~ were then sorted by aerosol properties, pollution level, environmental humidity, and dynamic condition according to their relative importance to *MRE*. Based on the error analysis, we found that the magnitude of *MRE* is more relevant to the specification of input aerosol pollution level in marine regions and aerosol hygroscopicity in continental regions than to other variables in the simulation.

## 1 Introduction

30 Aerosols play an important role in determining ~~affecting~~ the radiation balance of the earth-atmosphere system by scattering and absorbing shortwave radiation and altering the cloud reflectivity and lifetime (Twomey, 1974, 1977; Ghan, 2013; Forster et al., 2016; Ramaswamy et al., 2019; Wang et al., 2020). ~~Currently, A~~aerosol-cloud interactions ~~are remain as~~ one of the largest sources of climate modeling uncertainty ~~(IPCC AR6, Forster et al., 2021)Intergovernmental Panel on Climate Change, 2013).~~

35 Aerosol-cloud interactions are largely driven by the activation of aerosols to form cloud droplets. The addition of activated aerosol to existing clouds can directly change the concentration and size of cloud droplets and thereby affect the microphysical properties and radiative forcing of the clouds. Aerosol activation is controlled by rapid and nonlinear aerosol and cloud microphysical processes (Meskhidze et al., 2005), which have not been explicitly resolved in climate models yet (Fountoukis et al., 2007; Kang et al., 2015). Nenes et al. (2001) pointed out that the cloud droplet activation process is  
40 subject to kinetic limitations, including inertial, evaporation, and deactivation mechanisms, which further adds to the complexity of the aerosol activation.

Early parameterizations of aerosol activation in climate models were based on observations and derived through parameter fitting, using the aerosol number or mass concentration or other Cloud Condensation Nuclei (CCN) proxies (e.g., sulfate mass) to empirically determine the activated *CDNC* (Jones et al., 1994; Boucher and Lohmann, 1995; Jones and Slingo, 1996;  
45 Lohmann, 1997; Kiehl et al., 2000; Menon et al., 2002). Although these parameterizations have the advantages of convenience and low computational burden (Fountoukis et al., 2007), substantial uncertainties are resulting from limited spatiotemporal representativeness and unresolved variations in aerosol properties (Meskhidze et al., 2005). In the recent two decades, physically-based parameterization schemes of aerosol activation have emerged (Abdul-Razzak and Ghan, 2000; Cohard et al., 2000; Fountoukis and Nenes, 2005; Ming et al., 2006; Kivekäs et al., 2008; Khvorostyanov and Curry, 2009;  
50 Shipway and Abel, 2010; Zhang et al., 2015). These schemes are based on the Köhler theory and are used in climate models to parameterize aerosol activation near the cloud base. As Köhler theory fundamentally describes the process by which water vapor condenses and forms liquid cloud droplets, it can be applied to a wide range of atmospheric conditions and aerosol pollution levels. However, considerable approximations of the Köhler theory are employed for application in climate models, which leads to potential biases in comparison with results from more rigorous and accurate simulations of cloud droplet  
55 growth with adiabatic parcel models (e.g., ~~(Ghan et al., (2011)–Ghan et al. (2014))~~). The ongoing increase in computing power (Herrington and Reed, 2020) reduces the need for cost-saving approximations in climate models. In the following, we will introduce a Quasi-steady state approximation of the cloud Droplet Growth Equation (QDGE) that provides an efficient alternative to parameterizations of activated *CDNC* in climate models.

Parameterization schemes of aerosol activation ~~have~~were often been evaluated ~~using~~with adiabatic parcel model simulations.  
60 These models explicitly solve aerosol activation and droplet growth processes by mimicking vertical uplifting of an air parcel containing a specified number of aerosol particles, predicting changes in temperature, humidity/supersaturation,

activation of aerosols, and droplet growth from the cloud base upward. When utilizing identically specified aerosols, the results of a parcel model can be used as a benchmark to evaluate parameterizations. This approach has been [used](#) extensively [used](#) to evaluate activation schemes (Table 1). ~~Alternatively~~However, a less commonly used approach is to evaluate parameterizations by conducting a “closure experiment”, that is, to carry out a parameterized calculation by specifying observed aerosol concentrations and environmental thermodynamic conditions, and then compare the calculated and observed *CDNC* (e.g. Snider and Brenguier, 2000; Guibert et al., 2003; Fountoukis and Nenes, 2005; Kivekäs et al., 2008). Though some parameterizations have been evaluated based on comparisons of simulated and observed *CDNC* from aircraft campaigns, mostly regional data sets [have been](#)were used for very specific meteorological conditions and pollution levels. It is essential to select a wide range of cloud data for different atmospheric conditions and pollution levels to arrive at meaningful conclusions for global climate model simulations.

In this study, we introduce the QDGE scheme and evaluate it by using cloud data from multiple aircraft campaigns in four different regions over the world, covering marine and continental conditions. This paper is organized as follows. The next section describes the QDGE scheme and Sect. 3 summarizes the data and method used for the closure experiment~~and the~~ evaluation. Section 4 illustrates the results of the closure experiment and analyzes the sources of simulation errors, followed by conclusions and discussion in Sect. 5.

**Table 1. A summary of activation parameterizations and the evaluation methods in previous studies.**

| Parameterization               | Evaluation methods                             |
|--------------------------------|------------------------------------------------|
| Abdul-Razzak et al. (1998)     | Parcel model                                   |
| Cohard et al. (2000)           | Parcel model                                   |
| Snider et al. (2003)           | Aircraft measurements                          |
| Fountoukis and Nenes (2005)    | Parcel model; Aircraft measurements            |
| Ming et al. (2006)             | Parcel model                                   |
| Kivekäs et al. (2008)          | Other parameterizations; Aircraft measurements |
| Khvorostyanov and Curry (2009) | Twomey power law (Pruppacher et al., 1998)     |
| Shipway and Abel (2010)        | Parcel model                                   |

## 2 QDGE scheme

### 2.1 Scheme description

Aerosol particles ~~that are~~suspended in [an air](#) parcel ~~of air activate and~~ grow into cloud droplets by condensation of water vapor if supersaturation with respect to water exceeds a critical value. In stratus and convective clouds, aerosol activation is particularly efficient in the vicinity of the cloud base, where supersaturation typically reaches its local maximum. Although observations provide evidence that aerosol activation is not limited to the region near the cloud base, this is omitted in the aerosol activation scheme described here, similar to most~~parcel~~ models and parameterizations.

85 In order to determine the portion of the aerosols that activates and forms cloud droplets, a numerically efficient solution of the condensational droplet growth equation (e.g. Seinfeld and Pandis, 2016) is employed to simulate the growth of an ensemble of aerosol particles near the cloud base. The ~~water vapor saturation ratio and number of activated~~ cloud droplets above the cloud base ~~are is~~ simulated by solving a series of equations that describe assuming a vertically ascending air parcel containing aerosols from below the cloud base, which ascends vertically to produce supersaturated conditions above the cloud base. The vertical velocity of the air parcel ~~of air~~,  $w_c$  (in  $\text{m s}^{-1}$ ), is either specified or parameterized, as described in Sect. 3.2.3.

The change in wet aerosol particle radius,  $R_{pw}$  ~~(in m)~~, by condensation of water vapor as a function of the environmental supersaturation ~~water vapor saturation ratio~~ ( $S$ , e.g. Emanuel, 1994) in the scheme is given by

$$R_{pw} \frac{dR_{pw}}{dt} = \frac{S - S_p}{c}, \quad (1)$$

95 where  $S_p$  is the equilibrium supersaturation ~~water vapor saturation ratio directly~~ over the surface of the particle, which is obtained from  $\kappa$ -Köhler theory (Petters and Kreidenweis, 2007):

$$S_p - 1 = \frac{A}{R_{pw}} - \frac{B}{R_{pw}^3} \quad (2)$$

100 where the parameters  $A$ ,  $B$ , and  $C$  account for thermodynamic conditions in the cloud and physiochemical properties of the aerosol particles and droplets (Appendix A).

As described below, the QDGE scheme solves Eqs. (1) and (2) in combination with energy and moisture budgets to calculate changes in  $S$ .  ~~$S$ -driven by thermodynamic processes.~~ For instance, the thermodynamic equations underlying the QDGE scheme can be used to obtain the temporal evolution of  $S$  in the air during adiabatic ascend near cloud base (Ghan et al., 2011):

$$105 \quad \frac{dS}{dt} = Dw_c - E \frac{dq_w}{dt} \quad (3)$$

where ~~t~~The parameters  $D$  and  $E$  are weak functions of temperature and pressure, and  $q_w$  is the liquid water mixing ratio, which is related to the activated particle-size distribution (Appendix A).

Theoretically, each growing aerosol particle will compete with others for the water vapor in the environment, and the particle size increases according to Eq. (1) and affects the environmental supersaturation through Eq. (3). Eqs. (1-3) are ~~complexly coupled in a complex manner thus hardly have an analytical solution.~~

$$A = , \quad (3)$$

$$B = \kappa , \quad (4)$$

and

$$C = + , \quad (5)$$

115 where  $\kappa$  is the aerosol hygroscopicity,  $\sigma$  the surface tension of the solution/air interface (which is approximated by the surface tension of water here),  $\rho_w$  the density of water,  $M_w$  the molecular weight of water,  $R$  the universal gas constant,  $T$  the

temperature, the dry aerosol particle radius, the saturation vapor pressure, the latent heat of vaporization, the modified thermal conductivity of air accounting for non-continuum effects, the modified diffusivity of water vapor in air accounting for non-continuum effects (Seinfeld and Pandis, 2016). Petters and Kreidenweis (2007) and Kreidenweis et al. (2008) provided tabulated values of the hygroscopicity parameter  $\kappa$  for a variety of chemical compounds, based on laboratory data and modeling. They found that parameterized water contents are often within experimental uncertainty. However, the accuracy of this approach tends to decrease with decreasing aerosol water content. In particular, simulations of highly concentrated, non-ideal aqueous solutions with strong electrostatic interactions between ions with the Aerosol Inorganic Model (AIM; Wexler and Clegg (2002); <http://www.aim.env.uea.ac.uk/aim/aim.html>) give evidence for systematically different results at low aerosol water contents for some compounds (Kreidenweis et al., 2008). In order to improve biases at low relative humidity, the original method was extended to account for variations in  $\kappa$  with relative humidity in the QDGE scheme. Specifically, piecewise linear relationships between  $\kappa$  and aerosol water activity for different chemical components were determined based on results from AIM.

~~Direct~~ However, the numerical solution ~~iss~~ of Eq. (1) are applicable but computationally expensive. For example, Eq. (3) indicates that the balance between the enhancement of  $S$  ~~S~~ due to the air parcel uplifting, and given that the reduction of  $S$  ~~S~~ due to the condensation growth of activated particles, rate of water vapor depends on the aerosol size distribution and chemical composition, which leads to ~~at~~ the highly non-linear behavior variation of  $SS$  with time in the ascending parcel of cloud air/~~height~~ the supersaturation water vapor saturation ratio vertical profile. The condensation growth is ~~also~~ non-linearly related to the environmental conditions and aerosol properties (Eqs. 1 ~~and~~ 2). Therefore typically, a time steps much shorter than 1 second ~~is typically~~ are required to numerically solve these equations, which implies computational expenses that would prohibit applications in climate models (Khain et al., 2015). For instance, adiabatic ascending parcel models (e.g. Chen et al., 2016; Peng et al., 2005) to numerically solve Eqs. (1-3) require a very high time resolution, typically with a time step of  $10^{-3}$  to  $10^{-4}$  seconds. The parcel model results are regarded as the most accurate numerical solution and can be used as the benchmark to verify the parameterization of activation and condensation processes.

In large-scale stratus clouds, the maximum supersaturation (usually less than 0.2 %) occurs about 100 m above the cloud base, that is, the rate of  $S$  change is  $0.002\% \text{ m}^{-1}$  or so. A similar conclusion can be derived from the change of supersaturation and temperature (combined with a lapse rate of atmospheric temperature; (Pandis et al. (1990))). Therefore, it is reasonable to assume a scale of several seconds (or meters) at which the supersaturation is approximately constant in the air parcel. Consequently ~~However~~ In this study, we introduce a parameterization for climate model to achieve numerical efficiency ~~can be achieved by using~~ use a Quasi-steady state ~~approximated~~ approximation to solve the Droplet Growth Equation (QDGE), which ~~assumes~~ can be derived by using ~~that~~ the local ~~approximation~~  $S \approx \text{const}$  is approximately a constant. ~~in~~ Eq. (1), which can ~~then~~ be conveniently expressed as follows,

$$\frac{dx}{du} = \delta - a \left( \frac{b}{x^{1/2}} - \frac{1}{x^{3/2}} \right), \quad (46)$$

150 for the time period from  $t$  to  $t + \Delta t_s$  ( $\Delta t_s$  is a sub-timestep, roughly several tens of seconds in a climate model), with variable substitutions for particle size,  $x = R_{pw}^2/2$ , and for time,  $u = t|S-1|/C$ , and parameters that are given by:

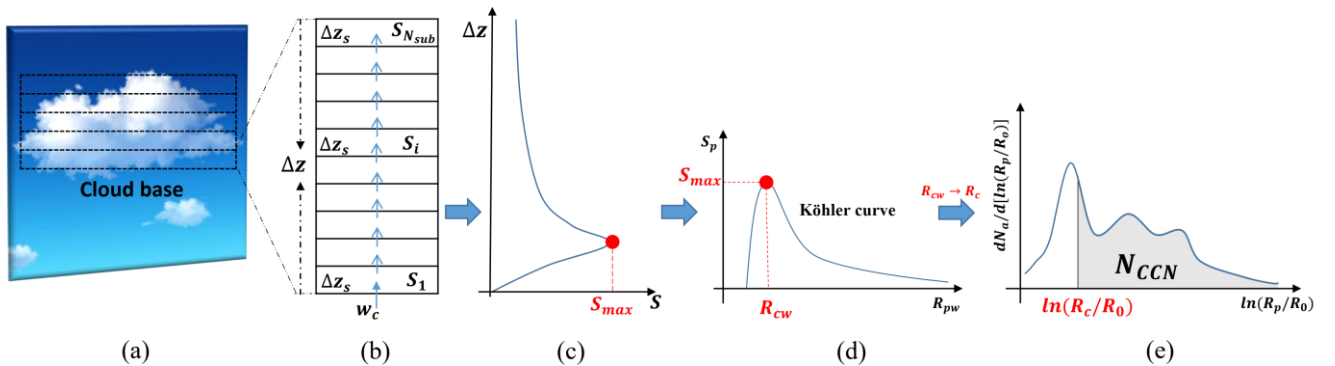
$$\delta = \begin{cases} -1, & \text{if } S < 0.1, \\ 1, & \text{if } S \geq 0.1, \end{cases} \quad (57)$$

$$a = \frac{B}{2^{3/2}|S-1|}, \quad (68)$$

$$b = \frac{2A}{B}. \quad (79)$$

In the QDGE aerosol activation scheme, numerical efficiency is achieved by using pre-calculated solutions  $x(u)$  of Eq. (46), are used, which are provided as the wet particle size  $R_{pw}$  dependent on different values of  $a$ ,  $b$  and  $\delta$  in the form of look-up tables (LUTs) for different values of  $a$ ,  $b$ , and  $\delta$  in the model to calculate  $R_{pw}$  for different values of  $a$  and  $b$ . The  $S$ -dependent parameters  $a$  and  $\delta$ , and  $u$ , are determined through an iterative procedure, for each time step and vertical level near cloud base, as described in the following.

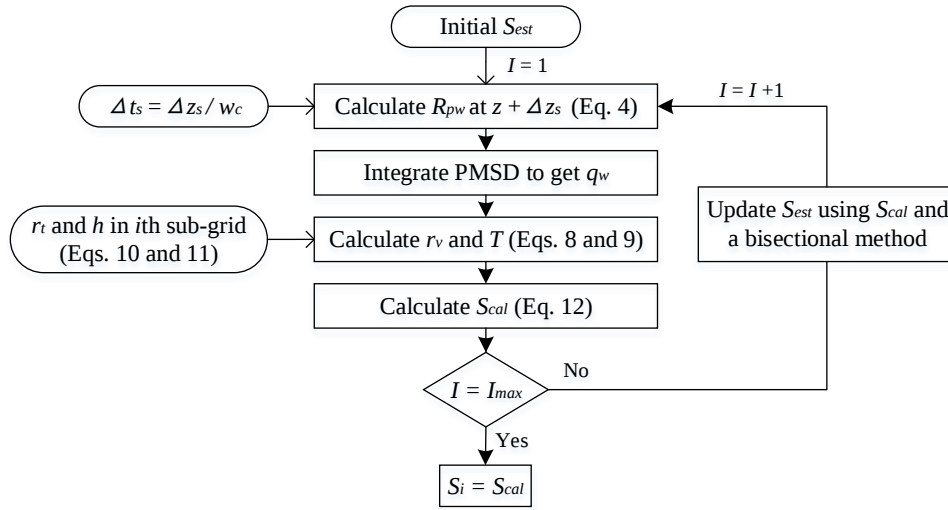
The process major steps of the QDGE scheme to calculate the aerosol activation are shown in Fig. 1. A vertical grid with  $N_{sub}$  sub-levels and (grid spacing  $\Delta z_s = \Delta z/N_{sub}$ ) is employed in the QDGE scheme, where  $\Delta z$  is the grid spacing in the atmospheric host model, near cloud base (Fig. 1a-b). Calculations are only performed for the first host model grid layer above the cloud base, with typical values  $\Delta z_s \approx 1 - 10$  m. The local approximation with constant  $S$  applies in each sub-level  $\Delta z_s$ , and a vertical profile of  $S$  is eventually obtained within the host model grid  $\Delta z$  (Fig. 1c). The iterative calculation to obtain  $S$  at each sub-level is described below, to ensure that the supersaturation maximum ( $S_{max}$ ) is captured and sufficiently well resolved in model applications of the aerosol activation scheme.



170 **Figure 1. A schematic diagram to show the major steps of the QDGE scheme.**

The growth calculations are performed for a sub-ensemble of aerosol particles which are selected from the full dry aerosol size distribution at regular size intervals,  $\Delta\chi = 1/p\Delta\phi$ , where  $p$  is on the order of 5–20 and  $\Delta\phi$  is the simulated particle size range of Aitken and accumulation mode aerosols, expressed in terms of a dimensionless particle size parameter  $\phi = \ln(R_p/R_0)$ , with  $R_0 = 10^{-6}$  m. In this study, we set  $p$  to 6 for the closure experiment, meaning that 6 discrete aerosol particle sizes are used. Sizes of other particles in the continuous aerosol size distribution are obtained from linear interpolation between the sizes of the particles in the discrete 6 member sub-ensemble.

In each sub-level  $\Delta z_s$ , the supersaturation  $S$  in each sub-grid (i.e.  $S_i$  in Fig. 1b, where  $i = 1, \dots, N_{sub}$ ) and the  $S$ -dependent parameters in Eq. (46) are obtained through an iterative calculation, which explicitly requires the conservation of mass and energy. The flow chart of the iterative calculation is as shown in Fig. 2.



**Figure 2. The schematic flow chart of the iterative calculation for the sub-grid supersaturation  $S_i$ , where  $I$  is the number of iterations. PMSD is the particle mass-size distribution. Total water mass mixing ratio,  $r_t$ , and liquid water static energy,  $h$ , are conserved.**

Specifically, at the beginning of an iteration, an initial value of supersaturation ( $S_{est}$ ) is first specified (“best-guess estimate”) and Eq. (46) is integrated over the sub-time-step  $\Delta t_s = \Delta z_s / w_c$  to obtain a first estimate of the particle wet sizes  $R_{pw}$  at the sub-level  $z + \Delta z_s$ . Next, an integration over the particle mass-size distribution (PMSD) yields a first estimate of the liquid water mixing ratio,  $LWC q_w$ , at  $z + \Delta z_s$  (Fig. 2), subject to the initially specified  $S_{est}$  value of the water vapor saturation ratio. Secondly,

The subsequent calculations are based on the total water mass mixing ratio,  $r_t$ , and liquid water static energy,  $h$ , in the ascending parcel of air are calculated, as defined as by:

$$r_t = r_v + q_w LWC, \quad (49)$$

$$h = gz + c_p T - L_v LWC q_w ,$$

(914)

where  $r_v$  is the water vapor mass mixing ratio,  $T$  is the temperature,  $g$  is the gravitational constant, and  $c_p$  is the heat capacity at a constant pressure of dry air. ~~Currently, only adiabatic processes are considered, and therefore total water mass mixing ratio,  $r_t$ , and liquid water static energy,  $h$ , in Eqs. (11) and (12) are conserved in each sub-level as the parcel ascends from  $z$  to  $z + \Delta_z$ . The total water mass mixing ratio and liquid water static energy at the lower and upper boundaries of the~~ current host model grid (with the superscripts  $L$  and  $U$  respectively) are first calculated using Eqs. (8) and (9). Then, the total water mass mixing ratio ( $r_t^i$ ) and liquid water static energy ( $h^i$ ) in the  $i$ th sub-level are obtained by linear interpolation, given by:

$$r_t^i = \frac{N_{sub}-i+1}{N_{sub}} r_t^L + \frac{i-1}{N_{sub}} r_t^U , \quad (10)$$

$$h^i = \frac{N_{sub}-i+1}{N_{sub}} h^L + \frac{i-1}{N_{sub}} h^U . \quad (11)$$

Knowing  $r_t^i$  and  $h^i$ ,  $r_v$  and  $T$  in the  $i$ th sub-level can be determined derived from Eqs. (8) and (11) using the estimated  $q_w$ , using the first estimate of  $q_w$  LWC, as described above. Subsequently, these results are used to update the supersaturation  $S$  is calculated water vapor saturation ratio, based on the standard definition of the water vapor saturation ratio,

$$S + 1 = \frac{r_v \left( \frac{1 + \frac{r_v}{0.622} r_* / \varepsilon}{1 + r_v / \varepsilon 0.622} \right)}{r_*} ,$$

(122)

where  $\varepsilon \equiv 0.622$ , and  $r_*$  is the saturation water vapor mass mixing ratio in the air parcel, which and of air, which depends on  $T$ . Subsequently

And then, the updated value calculated supersaturation ( $S_{cal}$ ) after each iteration is compared to and the initial estimate  $S_{est}$  of  $S$  are compared and are used to determine an An improved estimate of  $S$  is determined using a bisectional method that

minimizes the difference between different available estimates of  $S$  through iteration, as shown in Fig. 2. The method enables quickly converges to a good-enough estimated desired value of  $S$  with the maximum number of iterations ( $I_{max}$ ) as 4,

which solves Eq. (64) and satisfies all necessary constraints according to Eqs. (840), (449), and (4212). Here, the maximum number of iterations ( $I_{max}$ ) was set to 4 for the model applications discussed below. The iterations are repeated in

each complete and the sub-grid level to calculate  $S_i$  results are is available at  $z + \Delta_z$ . The calculations are repeated in order to

obtain  $S$  at the next higher level above until the vertical profile of supersaturation is results are available at all  $N_{sub}$  levels (Fig. 1b).

The maximum value of the simulated vertical supersaturation profile of  $S$ ,  $S_{max}$ , is used to diagnose the critical particle size,  $R_{cw}$ , based on Eq. (2) (Fig. 1c-d). Once  $S_{max}$  and the critical wet radius ( $R_{ew}$ ) are calculated in the grid, All particles with

225 ~~a wet/dry~~ radius larger than  $R_{cw}/R_e$  are taken as the activated particles to become ~~cloud condensation nuclei, cloud droplets.~~  
 Consequently, the cloud condensation nuclei number concentration ( $N_{CCN}$ ) is obtained by integrating the activated aerosol  
 size distribution accordingly (Fig. 1e). Above cloud base, a uniform number of ~~vertical profile of the the activated particles~~  
~~cloud droplet equal to the value at cloud base number mixing ratio is assumed,~~ equals to the value calculated at cloud base, in  
 good agreement with observations and detailed simulations of using cloud-resolving models (Gerber et al., 2008; Slawinska  
 230 et al., 2012; Jarecka et al., 2013).

In each grid of the host model, the dry aerosol number-size distribution is represented as particle numbers at regular size  
 intervals,  $\Delta\chi = 1/p\Delta\varphi$ , where  $p$  is the number of size bins.  $\Delta\varphi$  is the particle size range covering both Aitken and  
 accumulation modes, expressed in terms of a dimensionless particle size parameter  $\varphi = \ln(R_p/R_0)$ , with  $R_0 = 10^{-6}$  m. In  
 this study, we set  $p$  to 6, meaning that 6 discrete aerosol particle size bins are used. The continuous aerosol size distribution  
 235 (such as Fig. 1e) can be obtained from linear interpolation using the particle numbers in 6 discrete size bins.

Currently, only adiabatic processes are considered in each sub-level. Therefore, total water mass mixing ratio ( $r_t$ ) and liquid  
 water static energy ( $h$ ) are conserved as the parcel ascends from  $z$  to  $z + \Delta z$ . ~~Although the above calculations assume the~~  
~~adiabatic ascending of air parcel. However, entrainment could have potential impacts on the~~energy and moisture profiles in  
 clouds may be affected by entrainment processes. ~~host model grid. Besides, you can~~Therefore, ~~We and we~~ additionally  
 240 consider the impact of entrainment ~~modifying to consider the effect of entrainment on the  $S$  profile if necessary. The~~  
~~entrainment is considered to have a direct impact on~~  $r_t$  and  $h$  above the cloud base by using

$$r_t^{Ue} = r_t^L + (r_t^U - r_t^L)\exp(-e\Delta z), \quad (13)$$

$$h^{Ue} = h^L + (h^U - h^L)\exp(-e\Delta z), \quad (14)$$

where  $r_t^{Ue}$  and  $h^{Ue}$  are the ~~values of~~ total water mass mixing ratio and liquid water static energy considering the entrainment  
 245 of air, with a specified entrainment rate given by  $e$ , respectively. ~~effects, which~~ These can be used to replace  $r_t^U$  and  $h^U$  in  
 Eqs. (10) and (11) when entrainment needs to be considered ~~occurs~~.

Note that Eq. (3) can only be used for an adiabatic processes and does not work if there is entrainment or radiative cooling of  
 the air, e.g. the formation of cloud droplets in radiation fog. In contrast, the QDGE scheme is much more general, as outlined  
 above. The QDGE scheme can be easily modified for simulations of entrainment and radiation fog if required.

250 Finally, ~~the maximum value of the simulated vertical supersaturation~~water vapor saturation ratio profile,  $S_{max}$  (Fig. 1c), is  
 selected and used to diagnose the critical particle size,  $(R_{cw})$ , which separates activated from non-activated particles, i.e. by  
 requiring that  $S_{pmax} = S_{max}$  (Fig. 1d). Particles with sizes that are equal to or greater than the critical size ( $R_e$ , the dry size  
 corresponding to  $R_{cw}$ ) are assumed to be activated. ~~Consequently, the cloud condensation nuclei number droplet number~~  
~~concentration ( $N_{CCN}$ ) is obtained by integrating the activated particle size distribution accordingly (Fig. 1e).~~

255 Above cloud base, a uniform vertical profile of the cloud droplet number mixing ratio is assumed, in good agreement with  
 observations and detailed simulations of clouds (Gerber et al., 2008; Slawinska et al., 2012; Jarecka et al., 2013). Also, you  
 can set the entrainment rate to consider the effect of entrainment on the vertical profile if necessary. ~~exp~~The QDGE scheme

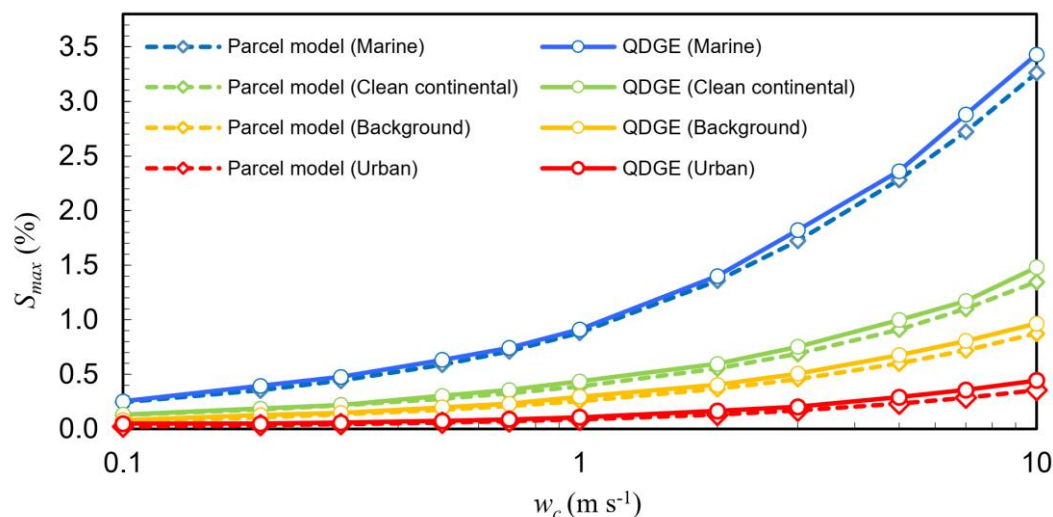
calculates the activated particle number near the cloud base. Above cloud base, a uniform number of vertical profile of the the activated particles cloud droplet equal to the value at cloud base number mixing ratio is assumed, in good agreement with observations and detailed simulations of using cloud resolving models (Gerber et al., 2008; Slawinska et al., 2012; Jarecka et al., 2013)

## 2.2 Comparison with a parcel model

In this subsection, we examine the performance of the QDGE scheme by comparing it with parcel model results by conducting a series of experiments as described in Ghan et al. (2011).

The parcel model can numerically solve the droplet growth equations in a most accurate way, by representing aerosol size distributions with finely discretizing bins and utilizing a very short time step to trace the supersaturation variation with time/height (Ghan et al., 2011).

For the comparisons, we assume a tri-modal lognormal size distribution (Whitby, 1978) of ammonium sulfate aerosol, consistent with the experimental setup in Ghan et al. (2011) (Table B1). The environmental conditions in the simulations cover with a wide range of  $w_c$  values ( $0.1 - 10 \text{ m s}^{-1}$ ) and four different aerosol regimes (Marine, Clean continental, Background, and Urban). When conducting QDGE simulations, we set the number of sub-levels ( $N_{sub}$ ), the maximal number of iterations ( $I_{max}$ ), and the number of size bins ( $p$ ) as 60, 4, and 6, respectively, which are the same as those in the following closure experiment (Sect. 4.1). Comparison between the results from the simulations are shown in Fig. 3, in which the parcel model results are identical to those in Ghan et al. (2011). In general, the QDGE scheme performs well with lower  $w_c$  but overestimates the  $S_{max}$  when  $w_c$  is larger than  $2 \text{ m s}^{-1}$ . The differences in  $S_{max}$  between parcel model and the QDGE scheme in all experiments are within 0.18 % (with an average of 0.05 %), much lower than the differences between parcel model and four state-of-the-art activation schemes (within approximately  $\pm 1.5 \%$ ) in Ghan et al. (2011). This indicates that the QDGE scheme achieves a high accuracy in simulating the processes of activation and condensation growth of cloud droplets under the specified conditions.



**Figure 3. Comparison between the calculated maximum supersaturation ( $S_{max}$ ) from the QDGE scheme (solid line) and the parcel model (dashed line, Ghan et al., 2011-Ghan et al., 2014).**

In contrast to the QDGE scheme, the four activation- schemes considered by Ghan et al. (2011) are based on parameterized and simplifying assumptions about the physical processes involved in the formation of clouds droplets, using the vertical grid of the host model. Therefore, the QDGE scheme can be used for a broader range of environmental and aerosol conditions than these schemes, in general. Although the QDGE scheme mimicks the parcel model well, it is also numerically more efficient. Typically a parcel model simulation will take several minutes, while the QDGE scheme only consumes 0.1 seconds for the same case using single core on Intel Xeon E5-2660 v2.

One more advantage of the QDGE scheme is the potential scale adaptivity for different vertical grids. The accuracy of the simulated supersaturation profile increases with the specified number of sub-levels ( $N_{sub}$ ) and number of iterations ( $I_{max}$ ). Therefore, as the super-computer capabilities for climate model simulations are improved, the QDGE scheme will provide a more accurate solution for the activation process and easily adapts to the accuracy requirement for high-resolution GCMs in the future.

An earlier version of the QDGE scheme has been successfully used for simulations with the 5<sup>th</sup> generation of the Canadian atmospheric global climate model (CanAM5). It is currently being tested in additional models.

The QDGE aerosol activation scheme has been previously used to assess Arctic indirect radiative forcing (Arora et al., 2015) and to determine the sensitivity of Arctic clouds to changes in future surface seawater dimethyl sulfide concentrations (Mahmood et al., 2019).

3.1 Campaign description

The worldwide cloud data used for the evaluation were sampled from four aircraft campaigns. The locations and instrument information of the four campaigns are shown in Fig. 44 and Table 2. The Canada (CAN) campaign provided marine stratus cloud data observed during the Radiation, Aerosol and Cloud Experiment (RACE) in 1995 off the coast of Nova Scotia, Canada (Peng et al., 2002). The Chile (CL) campaign provided marine stratocumulus clouds data observed during the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx), for near-climatological atmospheric conditions off northern Chile and southern Peru (Wood et al., 2011). The Brazil (AMA) campaign provided continental stratus clouds data observed in Manaus, Brazil during the Green Ocean Amazon (GoAmazon2014/5) Experiment (Martin et al., 2016). The China (CN) campaign provided polluted continental stratus clouds data sampled in Beijing, China by the Beijing Weather Modification Office (Liu et al., 2020). These worldwide datasets comprise continental (CN and AMA), coastal (CAN), and marine (CL) meteorological conditions. Additionally, they cover different levels of human influence on clouds, with an observed range of the mean aerosol number concentration ( $N_a$ ) within 100 m below the cloud base from 282  $\text{cm}^{-3}$  to 1350  $\text{cm}^{-3}$ .

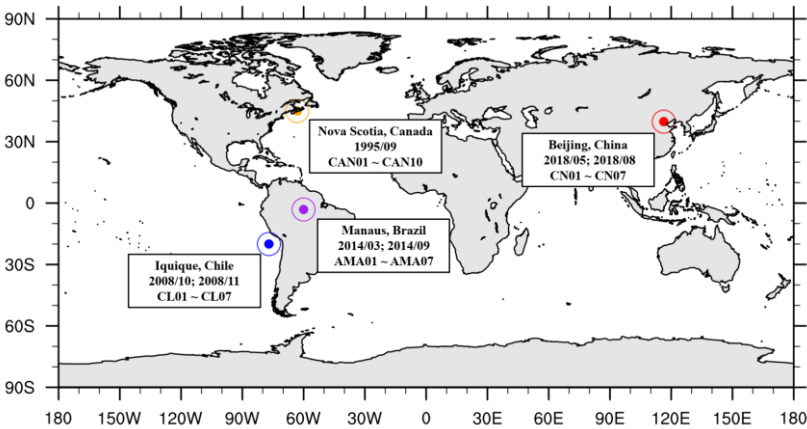


Figure 44. The geographical distribution of 31 selected cloud cases in the four aircraft campaigns. The text boxes provide the locations, the periods, and the names of the cloud cases for each campaign.

Table 2. An overview of the four aircraft campaigns in this study.

| Name            | CAN                                     | CL                                     | AMA                                      | CN                                       |
|-----------------|-----------------------------------------|----------------------------------------|------------------------------------------|------------------------------------------|
| Date            | 1995/09                                 | 2008/10; 2008/11                       | 2014/03; 2014/09                         | 2018/05; 2018/08                         |
| Location        | Nova Scotia, Canada                     | Iquique, Chile                         | Manaus, Brazil                           | Beijing, China                           |
| Cloud type      | stratus                                 | stratocumulus                          | stratus                                  | stratus                                  |
| Campaign name   | RACE                                    | VOCALS-REx                             | GoAmazon2014/5                           | /                                        |
| CDNC instrument | FSSP (15 bins, 2.0~47.0 $\mu\text{m}$ ) | CAS (20 bins, 0.6~56.3 $\mu\text{m}$ ) | FCDP (20 bins, 1.5~150.0 $\mu\text{m}$ ) | FCDP (20 bins, 1.5~150.0 $\mu\text{m}$ ) |

| Aerosol instrument                    | PCASP (15 bins,<br>0.13~3.00 $\mu\text{m}$ )<br>ASAP (13 bins,<br>0.183~2.37 $\mu\text{m}$ ) | PCASP (30 bins,<br>0.09~3.00 $\mu\text{m}$ ) | PCASP (30 bins,<br>0.09~3.45 $\mu\text{m}$ )      | PCASP (30 bins,<br>0.10~3.00 $\mu\text{m}$ ) |
|---------------------------------------|----------------------------------------------------------------------------------------------|----------------------------------------------|---------------------------------------------------|----------------------------------------------|
| Chemistry instrument                  | AMS                                                                                          | AMS                                          | AMS                                               | /                                            |
| <i>LWC</i> instrument                 | King hot-wire probe                                                                          | King hot-wire probe                          | King hot-wire probe<br>and Johnson-Williams probe | King hot-wire probe                          |
| Atmospheric condition instrument      | AIMMS                                                                                        | AIMMS                                        | AIMMS                                             | AIMMS                                        |
| Number of selected cloud cases        | 10                                                                                           | 7                                            | 7                                                 | 7                                            |
| Number of cases for $w_c$ calculation | 2                                                                                            | 3                                            | 5                                                 | 4                                            |
| $N_a$                                 | 476                                                                                          | $282 \pm 116$                                | $846 \pm 819$                                     | $1350 \pm 916$                               |

Note: is the integrated number of particles detected by aerosol instruments and averaged within 100 m below the cloud base. The definition of cloud base and selection of cloud cases refer to Sect. 3.2.1. Calculation of  $w_c$  refers to Sect. 3.2.3.

320 Aerosol and cloud measuring instruments utilized in the four campaigns are briefly presented in Table 2. The observed variables mainly include [the \*CDNC\*](#), [the cloud liquid water content \(\*LWC\*\)](#), the aerosol number-size distribution, the chemical compositions of aerosol, and atmospheric condition parameters. For the measurement of [the \*CDNC\*](#), the forward scattering spectrometer probe (FSSP) was used in the CAN campaign. The cloud, aerosol, and precipitation spectrometer (CAS) was used in the CL campaign. The fast cloud droplet probe (FCDP) was used in the AMA and CN campaigns. Although FCDP, 325 FSSP, or CAS can observe cloud droplets with a particle size up to 150  $\mu\text{m}$ , we only integrated the number for droplets with a particle size of 2 to 30  $\mu\text{m}$  to derive the *CDNC*. Because cloud droplets larger than 30  $\mu\text{m}$  are subject to collision-coalescence, and droplets smaller than 2  $\mu\text{m}$  may be deactivated by evaporation (Fountoukis and Nenes, 2005). For the measurements of the *LWC*, the King hot-wire probe was used in all campaigns, and the Johnson-Williams probe was also equipped as an alternative option in GoAmazon2014/5. In terms of the aerosol observation, all the four campaigns utilized an 330 onboard passive cavity aerosol spectrometer probe (PCASP), and some flights during the CAN campaign used the atmospheric solids analysis probe (ASAP), providing aerosol number concentration in multiple size bins roughly from 0.1 to 3  $\mu\text{m}$ . We integrated the number for particles within the detected size range to determine  $N_a$ . In the CAN, AMA, and CL campaigns, the mass concentrations of aerosol chemical species, including  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ , and organics (*org*), were measured using the aerodyne aerosol mass spectrometer (AMS). The CN campaign lacked data for aerosol chemical 335 composition (see Sect. 3.2.2). For the CL campaign, five aircraft (i.e. Lockheed C-130, BAe-146, Gulfstream-1, Dornier-228, and Twin Otter) carried out observations (Wood et al., 2011). In order to ensure data integrity and consistency for aerosol number-size distribution and chemical composition measurements in the subsequent analysis, we only selected data from the Gulfstream-1 flights. The atmospheric condition parameters ( $T$ , pressure ( $P$ ), relative humidity ( $RH$ ), vertical velocity ( $w$ )) were mainly observed by the airborne integrated meteorological measurement system (AIMMS), in all campaigns. For the

CL campaign, vertical velocity data were not available from the Gulfstream-1 flights, thus we used the observed  $w$  data from the Twin Otter flights that occurred simultaneous with Gulfstream-1 flights. Some meteorological variables that are required by the QDGE scheme, particularly including  $r_v$ ,  $r_t$ , and  $h$ , were not available from the aircraft observations. Therefore, we calculated these based on other variables (Sect. 3.2.4). Detailed descriptions of the aforementioned observational instruments and data quality control procedures can be obtained from the relevant publications for the different aircraft campaigns (Li et al., 1998; Peng et al., 2002; Wood et al., 2011; Kleinman et al., 2012; Martin et al., 2016, 2017; Wang et al., 2020).

## 3.2 Data processing for closure experiment

### 3.2.1 Data extraction

The flow chart of data extraction and processing is shown in Fig. 52. In the first step, we conducted a screening of observational data to obtain suitable cloud cases fulfilling the following conditions (Step 1 in Fig. 25). First, we selected cloud cases with continuous  $LWC$  profile with  $T > 0^\circ\text{C}$  and  $LWC \geq 0.05\text{ g cm}^{-3}$  in each layer, identifying the height of the cloud base as  $H_{low}$  (see Fig. BA11). Second, we checked whether the  $LWC$  near the cloud base approximately satisfies the wet adiabatic assumption, that is, nearly free from entrainment. As shown in Fig. BA11, we plotted the observed  $LWC$  and the adiabatic  $LWC$  ( $LWC_{ad}$ ) profiles, the later ones were calculated by assuming that  $LWC$  increases linearly with the height above cloud base ( $H_c$ ), i.e.  $LWC_{ad} = C_w H_c$ .  $C_w$  is the adiabatic liquid water lapse rate, which is a function of temperature (Brennguier, 1991). For liquid clouds, the value of  $C_w$  varies from  $0.5 \times 10^{-3}$  to  $3.0 \times 10^{-3}\text{ g m}^{-4}$  (Peng et al., 2002). For the cases shown in Fig. AB14,  $C_w$  ranges from  $0.6 \times 10^{-3}$  to  $2.8 \times 10^{-3}\text{ g m}^{-4}$ . The mean of  $C_w$  in each cloud case is shown in Table BA12. Considering that the entrainment rate  $e$  was set to  $1.0 \times 10^{-3}\text{ m}^{-1}$  (weak entrainment, Barahona and Nenes, (2007)) when running the QDGE scheme in order to be close to the real atmosphere, we identify the nearly adiabatic part in the cloud case (i.e. data sampled between  $H_{low}$  and  $H_{high}$  in Fig. A4B1) for obtaining the observed cloud properties for evaluating the simulation. Third, we excluded the impact of collision-coalescence in the selected cloud cases, by ensuring that the water contents of cloud droplets with size greater than  $30\text{ }\mu\text{m}$  were less than  $0.05\text{ g cm}^{-3}$ . Finally, we checked to make sure each cloud case has  $N_a$  larger than  $CDNC$ . Ultimately, we obtained 31 eligible cloud cases were selected, as shown in Fig. A4B1. Table B2A4 listed the observed data in the selected cloud cases,  $CDNC_o$  and  $LWC$  were averaged over the adiabatic part of each cloud case,  $N_a$  and  $RH$  were averaged within 100 m below the cloud base. As shown in Step 2 of Fig. 52, we classified data samples of each cloud case into cloudy and clear conditions by utilizing the following criteria. Data sampled inside the cloud (cloudy condition) requires that  $LWC \geq 0.05\text{ g cm}^{-3}$ ,  $CDNC > 10\text{ cm}^{-3}$ , and  $RH \geq 99.5\%$ , and data samples outside the cloud (clear condition) requires that  $LWC < 0.05\text{ g cm}^{-3}$ ,  $N_a > 10\text{ cm}^{-3}$ , and  $RH < 99.5\%$ .

During each flight, the sampling along the horizontal flight track was continuous, which allowed us to better characterize the cloudy conditions or atmospheric conditions inside or outside the cloud. In all the 31 selected cloud cases, we were able to

extract data samples at  $nl$  levels ( $l_{di}, di = 1, 2, \dots, nl$  from the cloud base; where  $nl$  is usually 4, at least 2.) along horizontal flight tracks in each cloud case, and calculated the mean value of the observed variable  $v$  ( $V_{v, l_{di}}$ ) along the horizontal track in each level  $l_{di}$ .  $V_{v, l_{di}}$  is then extended to the vertical model levels ( $L_{fj}, fj = 1, 2, \dots, NL$ ; where  $L_{fj}$  refers to the interfaces of the vertical layers in the model, i.e.  $\Delta z = L_{fj+1} - L_{fj}$ ) for running the QDGE scheme, which is Step 3 as shown in Fig. 52.

The extension proceeded with the following rules: The meteorological variables profile in clear condition, such as  $T$ ,  $P$ , and  $r_t$ , were extended downwards to the surface by using hydrostatic equation and ideal gas law, then extended to the top by linear extrapolation, and interpolated between  $l_1$  and  $l_{nl}$ . The aerosol mass and number profiles were extended to surface and top by linear extrapolation and interpolated between  $l_1$  and  $l_{nl}$ .  $RH$  was filled between  $l_1$  and  $l_{nl}$  by linear interpolation.

For each cloud case, the data samples in the clear air were used to obtain aerosol-related input information for the model simulations (number and mass concentrations of aerosol components in different particle size sections) and the profiles of meteorological parameters. The data samples in cloudy conditions were used to obtain the vertical velocity and  $LWC$  as input for the model, and to provide measured  $CDNC$  for comparisons with model results and closure verification. ~~Here The the simulated ., and  $LWC$  values in the boundaries of the host model grid werewere converted into  $q_w$  tofor calculateinge the initial  $r_t$  and  $h$  for applying in the QDGE scheme (Fig. 2 and Eqs. 8-12).~~ These are Steps 4, 5, and 6, as shown in Fig. 52 and

described in the next three subsections.

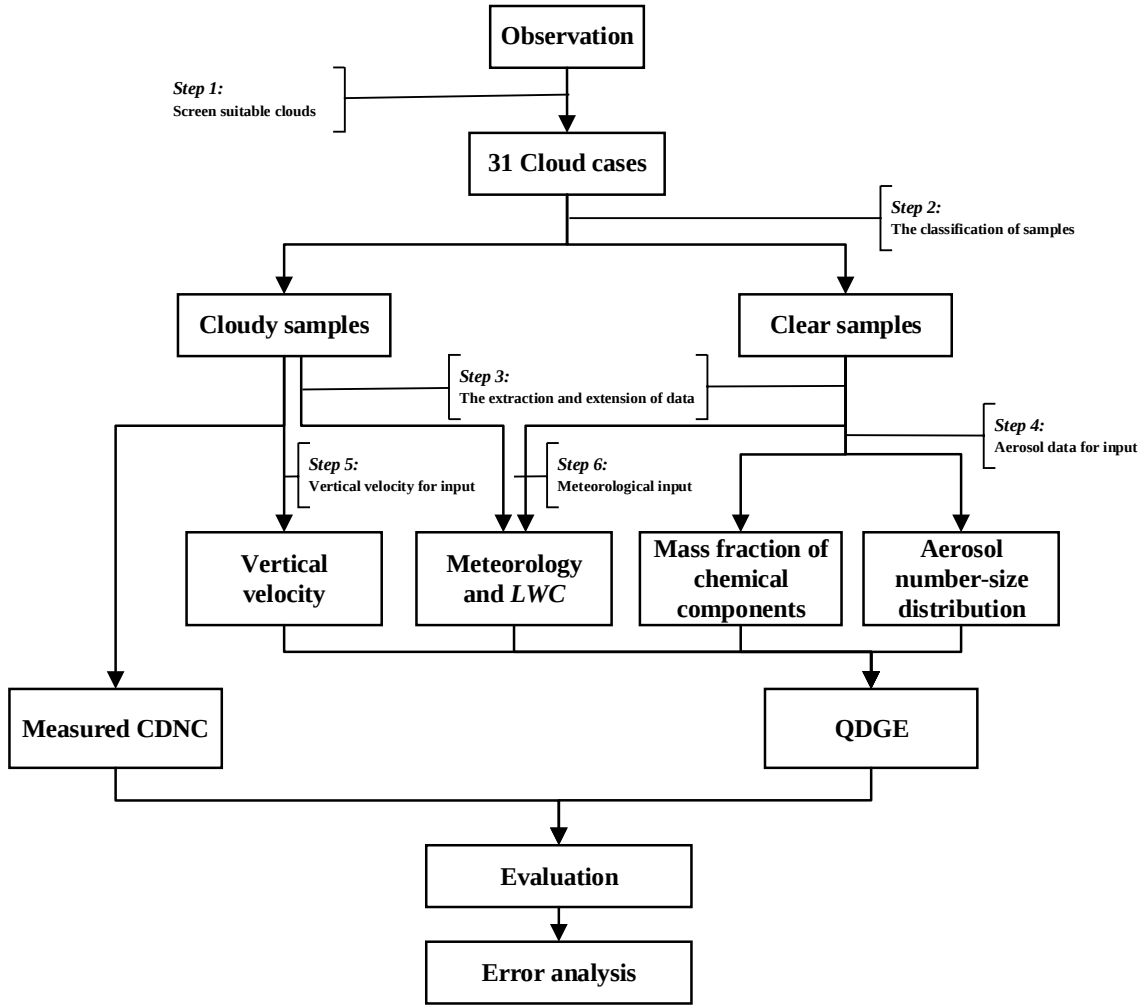
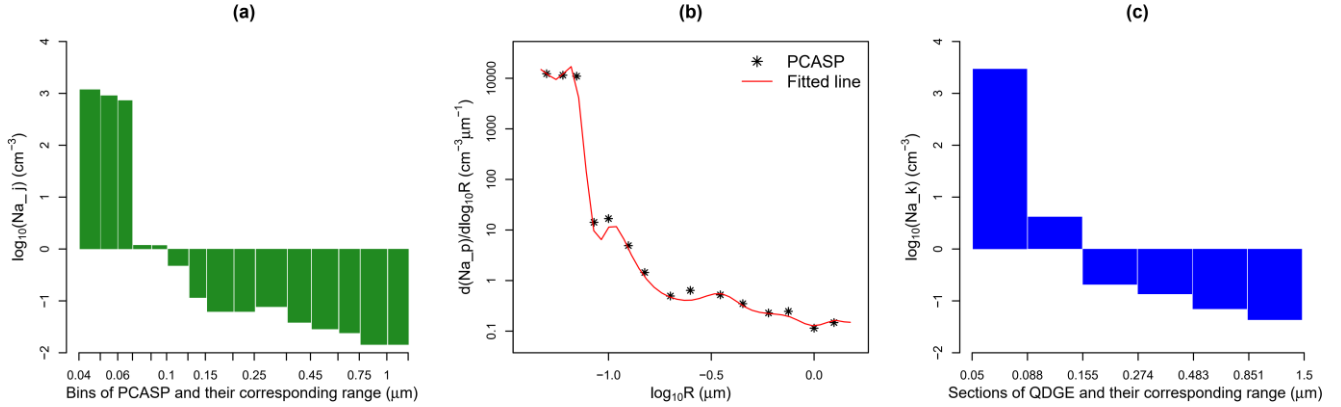


Figure 52. A flow chart to schematically show the data extraction and processing for this work.

### 3.2.2 Aerosol data for input

In each of the cloud cases from the different aircraft campaigns, aerosol number concentrations  $N_{a\_pj}$  ( $pj = 1, \dots, njp$ ; where  $njp$  is the number of size bins detected in observation, see Table 2) sampled by ASAP or PCASP were categorized in 13, 15, or 30 bins. The size-resolved aerosol number concentrations were subsequently interpolated to a common particle size distribution (PSD) with 6 prescribed size sections for model input based on the following method (as depicted in Fig. 63). First, we used the aerosol number concentration in each size bin of the PCASP (or ASAP) data to fit a continuous PSD using cubic spline interpolation (Fig. 63b). Second, we integrated the fitted PSD to obtain the aerosol number concentration  $N_{a\_k}$  ( $k=1, \dots, 6$ ) in the aerosol size sections employed by the QDGE scheme (the dry aerosol particle radius boundaries are at 0.050, 0.088, 0.155, 0.274, 0.483, 0.851, 1.500  $\mu\text{m}$ , as shown in Fig. 63c). By utilizing this method, the total  $N_a$  obtained

by integration over the 6 QDGE sections was slightly different from the observed total aerosol number due to the fitting of PSD, thus we further weighed the total fitted aerosol number concentration by the observed aerosol number to ensure the conservation of total number concentration (i.e., the total  $N_a$  integrated over the QDGE sections in Fig. 63c is the same as the aerosol number integrated over the observed PSD in Fig. 63a). Finally, the PSD of the aerosol number concentration in 6 sections (Fig. 63c) was used as input to the QDGE scheme.



**Figure 63.** The processing of the observed aerosol number-size distribution for the input to the QDGE scheme. (a) shows the observed aerosol number concentration in each size bin sampled by PCASP, (b) the particle size distribution curve (red line) fitted to the observations (the asterisks refer to the observations that were derived from (a)), and (c) aerosol number concentration in 6 size sections, as prescribed in model simulations with the QDGE scheme.

For each of the CAN, AMA, and CL campaigns, the AMS provided measurements of chemical components over the entire campaign, providing concentrations of  $NH_4^+$ ,  $NO_3^-$ ,  $SO_4^{2-}$ ,  $Cl^-$ , and *org*. The various chemical components in the aerosol were assumed to be internally mixed, thus all aerosol particles with the same size have the same composition~~different components share the same aerosol number concentration in each size section~~. To obtain the PSD of mass concentration of each chemical component, we made use of the AMS measurements. For continental campaigns such as CN and AMA, we assumed that aerosols are composed of  $NH_4NO_3$ ,  $(NH_4)_2SO_4$ ,  $NH_4Cl$ , and organics (*Org*) (Shilling et al., 2018; Zhou et al., 2019; Li et al., 2020). For coastal or oceanic campaigns such as CAN and CL, we took sea salt ( $NaCl$ ) into account, too. For the CAN, AMA, and CL campaigns, we converted the AMS data of ion mass ( $AMS_{ci}$ ,  $ci$  is  $NO_3^-$ ,  $SO_4^{2-}$ ,  $Cl^-$ , or *org*) to the mass of each chemical component ( $m_c$ ,  $c$  is  $NH_4NO_3$ ,  $(NH_4)_2SO_4$ ,  $NH_4Cl$ , organics (*Org*), or  $NaCl$ ).

$$m_{NH_4NO_3} = \frac{AMS_{NO_3^-}}{M_{NO_3^-}} M_{NH_4NO_3}, \quad (153)$$

$$m_{(NH_4)_2SO_4} = \frac{AMS_{SO_4^{2-}}}{M_{SO_4^{2-}}} M_{(NH_4)_2SO_4}, \quad (146)$$

$$m_{NH_4Cl} = \frac{(1-\alpha)AMS_{Cl^-}}{M_{Cl^-}} M_{NH_4Cl},$$

(175)

$$m_{NaCl} = \frac{\alpha AMS_{Cl^-}}{M_{Cl^-}} M_{NaCl},$$

(186)

425  $m_{Org} = AMS_{Org},$

(179)

where  $M_{ci}$  and  $M_c$  are the molecular weight of ion  $ci$  and chemical component  $c$ , respectively. Here we assume that concentrations of  $NH_4^+$  are sufficiently high to balance all anions. The mass of sea salt in different campaigns is controlled by a given factor  $\alpha$  to partition the amount of  $Cl^-$  in sea salt and continental chemical components. We set the values of  $\alpha$  as 0, 90%, and 95% for AMA, CAN, and CL campaigns. That is, 90% and 95% of  $Cl^-$  are attributed to sea salt in the coastal campaign CAN and the oceanic campaign CL, respectively. Based on the calculated mass concentration of each chemical component, the average density of aerosol can be obtained:

$$\rho_a = \frac{\sum_{c=1}^5 m_c}{\sum_{c=1}^5 m_c / \rho_c},$$

(1820)

435 where  $\rho_c$  is the density of each component  $c$ , and they are 1725, 1769, 1527, 1900, and 1400 kg m<sup>-3</sup> for  $NH_4NO_3$ ,  $(NH_4)_2SO_4$ ,  $NH_4Cl$ ,  $NaCl$ , and  $Org$ , respectively (Ferek et al., 1998; Nakao et al., 2013). Consequently, we can obtain the mass concentration (unit kg cm<sup>-3</sup>) of each component  $c$  in section  $k$  following this equation:

$$Mass_{c,k} = \frac{m_c}{\sum_{c=1}^5 m_c} \cdot N_{a,k} \frac{4\pi}{3} R_k^3 \rho_a,$$

(1219)

440 where  $R_k$  is the median radius of section  $k$ .

Since no AMS data are available for the CN campaign, we assumed the mass fraction of different chemical components according to contemporaneous measurements in Beijing, China (Zhou et al., 2019; Li et al., 2020), as shown in Table A2B3.

Under the assumption of  $\rho_a = 1600$  kg m<sup>-3</sup> (Levy Zamora et al., 2019),  $Mass_{c,k}$  in the CN campaign can be obtained from Eq. (1921).

445 Finally, we obtained the number concentration of total aerosol and the mass concentration of each chemical component from PCASP/ASAP and AMS measurements in each cloud case and calculated aerosol number and mass concentrations in 6 prescribed size sections following the above procedures (Step 4 in Fig. 52). We then used the aerosol information as input to drive the QDGE scheme.

### 3.2.3 Vertical velocity for input

450 The averaged updraft velocity ( $w_+$ ) and sub-grid vertical velocity ( $w_{sub}$ ) obtained from the observed vertical velocity ( $w$ ) samples in clouds were used to calculate  $w_c$  ( $w_c = w_+ + w_{sub}$ ) as input for running the QDGE scheme (Step 5 in Fig. 25). The updraft velocity is a key variable for parameterizing aerosol activation. Peng et al. (2005) pointed out that using a characteristic value of the vertical velocity distribution (0.8 times the standard deviation of the distribution) is a good approximation for simulating the nucleated cloud droplet number of marine stratus when running the parcel model.

455 Meskhidze et al. (2005) also gave a method to calculate  $w_+$ , which had the optimal closure for cumulus and stratocumulus clouds. Here, we derived a universal method for calculating  $w_+$  in stratus and stratocumulus based on the above two studies. According to Meskhidze et al. (2005), the averaged updraft velocity ( $w_+$ ) can be calculated by probability density function (PDF) of  $w$ ,  $p(w)$ :

$$w_+ = \frac{\int_0^\infty wp(w)dw}{\int_0^\infty p(w)dw} \quad .$$

460 (2022)

For the normal PDF with the mean velocity  $w_0$  and standard deviation  $\sigma$ ,  $p(w)$  can be represented as

$$p(w) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{(w-w_0)^2}{2\sigma^2}\right) = \beta\phi(\omega) \quad ,$$

(231)

where  $\omega = \beta w + \gamma$ ,  $\beta = 1/\sigma$ ,  $\gamma = -w_0/\sigma$ , and  $\phi(\omega)$  is the standard normal PDF.

465 Use Eq. (23) into Eq. (22) we obtain

Take Eq. (21) into Eq. (20) and obtain

$$w_+ = \frac{\phi(\gamma)}{(1-\Phi(\gamma))\beta} - \frac{\gamma}{\beta} = \frac{\phi(\gamma)}{(1-\Phi(\gamma))} \sigma + w_0 \quad ,$$

(242)

where  $\Phi(\gamma)$  is the cumulative distribution function of the standard normal PDF that can be represented by error function

470 (erf):

$$\Phi(\gamma) = \int_{-\infty}^{\gamma} \phi(t)dt = \frac{1}{2}(1 + \operatorname{erf}(\frac{\gamma}{\sqrt{2}})).$$

(253)

Especially, when  $w_0 = 0$ ,

$$w_+ = \frac{\phi(0)}{(1-\Phi(0))} \sigma = \sqrt{\frac{2}{\pi}} \sigma \cong 0.8\sigma \quad ,$$

475 (264)

which is consistent with the characteristic velocity pointed by Peng et al. (2005) used for assessing cloud droplet closure for stratocumulus clouds sampled in the CAN campaign.

A sub-grid vertical velocity ( $w_{sub}$ ) is needed for the QDGE scheme, and it can be derived from the square root of the Turbulent Kinetic Energy ( $TKE$ ) following Morrison and Pinto (2005):

$$w_{sub} = \sqrt{\frac{2}{3} TKE} \quad (275)$$

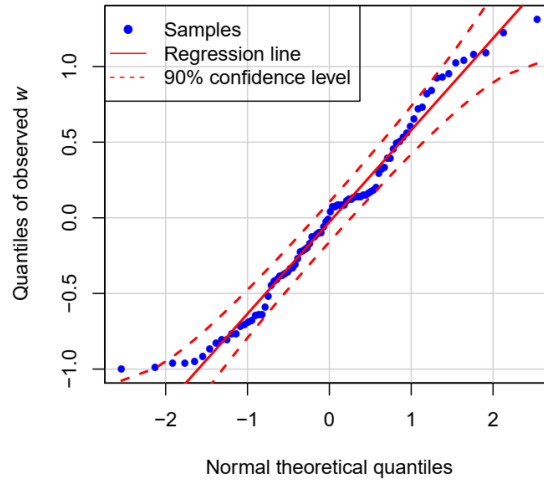
where the  $TKE$  is given by can be calculated according to its definition, which is half the sum of the variances (square of standard deviations) of the velocity components:

$$TKE = \frac{1}{2} (\overline{(u')^2} + \overline{(v')^2} + \overline{(w')^2}) \quad (286)$$

In this study, we assume that no horizontal movement occurs in cloud during the horizontal flight tracks, that is,  $\overline{(u')^2} = \overline{(v')^2} = 0$  and  $\overline{(w')^2} = \sigma^2$ . Therefore, the sub-grid vertical velocity can be represented by  $\sigma$ :

$$w_{sub} = \frac{\sigma}{\sqrt{3}} \quad (297)$$

If the observed  $w$  in each selected cloud case obeyed the normal distribution, we could calculate  $w_c$  ( $w_c = w_+ + w_{sub}$ ) following Eqs. (224) and (279) as input for running the QDGE scheme easily. We checked the normality of  $w$  distribution by drawing a quantile-quantile (Q-Q) plot using the observed  $w$  values along the horizontal flight track of the cloud case, taking CN01 as an example in Fig. 74. The linearity between the Q-Q plot of observed  $w$  samples and a standard normal distribution indicates that  $w$  data does indeed follow the normal distribution.



**Figure 74.** A normal quantile-quantile plot for comparing the observed  $w$  sampled by aircraft in cloud case CN01 with a standard normal distribution. The linearity of the data points (blue) suggests that the observed  $w$  are normally distributed.

In the four campaigns of this study, 4 cloud cases in CN, 2 cases in CAN, 5 cases in AMA, and 3 cases in CL have enough data samples to obtain the PDF of  $w$  (Table 2), as plotted for checking the normality of  $w$  distribution in Fig. A2B2.

500 However, the  $w$  PDF in two of the CAN cloud cases does not conform to the normal distribution very well (panel (5) and (6) of Fig. A2B2). So, we used the mean and standard deviation of  $w$  distribution in Peng et al. (2005) to obtain  $w_c$  in the CAN campaign. For the CN, AMA, and CL campaigns, we directly calculated the  $w_c$  from available data samples for the cloud cases plotted in Fig. A2-B2 and used their mean values for cloud cases lacking enough  $w$  values in each campaign (Table A4B2).

### 505 3.2.4 Meteorological input

Some meteorological variables ( $T$ ,  $P$ ,  $RH$ , and  $LWC$ ) can be obtained from AIMMS measurements directly, though, others ( $r_v$ ,  $r_t$ , and  $h$ ) need to be calculated according to available variables (Step 6 in Fig. 25). We obtained  $r_v$  by the following equation:

$$r_v = \frac{\varepsilon 0.622 e_* RH}{P - e_* RH}, \quad (3028)$$

where  $e_*$  can be estimated by referring to Murray (1967):

$$e_* = 6.1078 e^{\left(\frac{17.2694(T-273.16)}{T-35.86}\right)}. \quad (3129)$$

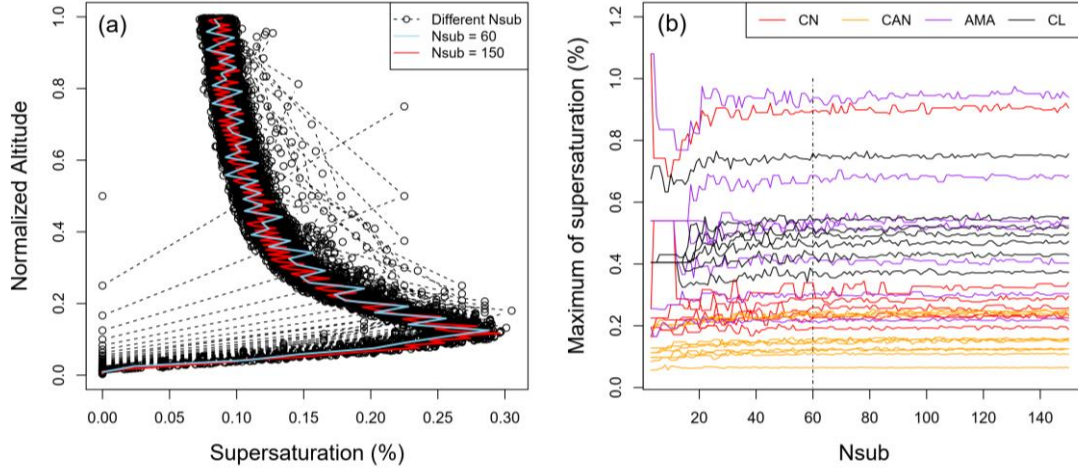
515 Then,  $r_t$  and  $h$  can be obtained by Eqs. (849) and (449) from  $r_v$  and other available variables. All meteorological variables were extracted and interpolated to model levels, as described in Sect. 3.2.1. The profiles of measured meteorological variables served as the initial state to drive the QDGE scheme.

### 3.2.5 Determination of $N_{sub}$

520 As mentioned in Sect. 2.1, the QDGE scheme simulates vertical profiles of supersaturation to determine  $S_{max}$ , for a vertical grid with the size  $\Delta z_s = \Delta z / N_{sub}$ , where  $\Delta z$  is the grid size of the atmospheric host model. The accuracy of the simulated supersaturation profile generally increases with  $N_{sub}$ , though, large values of  $N_{sub}$  imply higher computational burdens. For applications of the QDGE scheme in atmospheric models, it is therefore important to determine an optimal value of  $N_{sub}$  that yields sufficiently accurate supersaturation profiles at acceptable costs.

525 Figure 85a plots the vertical profiles of  $S$  simulated by the QDGE scheme with different  $N_{sub}$  values for the cloud case CN01. The results show that each profile with  $N_{sub} \geq 3$  produces a well-defined maximum of  $S$  ( $S_{max}$ ), which approaches to a stable value as  $N_{sub}$  is further increased. All cases seem to converge to a similar value as  $S_{max}$  with  $N_{sub} = 150$ , as plotted in Fig. 58a. Figure 58b shows the variation of  $S_{max}$  with the increasing  $N_{sub}$  for all cloud cases in the four campaigns. Overall,  $S_{max}$  fluctuates dramatically with  $N_{sub} < 10$ , but plateaus when  $N_{sub}$  is greater than 60 (10 for CAN). Results obtained for  $N_{sub} = 150$  and  $N_{sub} = 60$  are similar. The mean relative error and correlation coefficient between  $S_{max}$  with

$N_{sub} = 150$  and that with  $N_{sub} = 60$  are 1.97% and 0.9997, respectively. Therefore, we used  $N_{sub} = 60$  in this study  
 530 ( $N_{sub} = 10$  for CAN). Further discussion regarding the selection of  $N_{sub}$  are provided in Sect. 5.



**Figure 58.** (a) Vertical profiles of the simulated supersaturation for different  $N_{sub}$  (1-150) in the QDGE scheme for the cloud case CN01. (b) Changes of the maximum supersaturation with different  $N_{sub}$  for all cloud cases in the four campaigns.

### 3.3 Statistical parameters for evaluation and error analysis

535 The QDGE scheme simulates the  $CDNC$  ( $CDNC_M$ ) in each cloud case, based on  $S_{max}$ . Noting that  $CDNC_M$  is not exactly the same as  $N_{CCN}$  here, as we take wet particles with a size between 2 to 30  $\mu m$  to compare with the observed one. Considering that aerosol activation is particularly efficient in the vicinity of the cloud base in stratus and convective clouds, the QDGE scheme only calculates the  $CDNC$  at the cloud base (Sect. 2.1). Here, we considered the effect of weak entrainment on the vertical profile of the cloud droplet number mixing ratio ~~in order~~ to be close to the real cloud base in the atmosphere (Sect. 3.2.1). Therefore, we evaluated the simulation-effect performance of the QDGE scheme by comparing  $CDNC_M$  with the vertically average value of the observed  $CDNC$  ( $CDNC_O$ ) in the nearly adiabatic part of the cloud (between  $H_{low}$  and  $H_{high}$  in Fig. A4B1) (Sect. 3.2.1), given by:

$$CDNC_O = \frac{1}{N_O} \sum_{H=H_{low}}^{H_{high}} CDNC_{O,H}, \quad (32)$$

where  $N_O$  is the number of samples between  $H_{low}$  and  $H_{high}$ , and  $CDNC_{O,H}$  is the observed  $CDNC$  in height  $H$ .

545 Correspondingly, the ~~mean bias (MB) and~~ mean relative error ( $MRE$ ) of each cloud case can be calculated, as follows:

$$MRE = \left| \frac{CDNC_M - CDNC_O}{CDNC_O} \cdot 100\% \right|, \quad (33)$$

where  $MRE$  of each cloud case will also be used for subsequent error analysis.

To evaluate the overall accuracy of the QDGE scheme, we also calculated the mean values of  $CDNC_O$ ,  $CDNC_M$ ,  ~~$MB$~~ ,  $MRE$  for cloud cases in each campaign, namely  $\overline{CDNC_O}$ ,  $\overline{CDNC_M}$ ,  ~~$\overline{MB}$~~ , and  $\overline{MRE}$ . Besides, the  ~~$R$ -square ( $R^2$ )~~ ( $R$  is the Pearson correlation coefficient) between the  $CDNC_O$  and  $CDNC_M$  in each campaign was also calculated.

To quantify the contributions of different physical variables to errors in the simulated  $CDNC$  with the QDGE scheme, we calculated the Maximum Information Coefficient (MIC) (Reshef et al., 2011), which provides a measure for the strength of the relationship between each input variable and  $MRE$ . MIC can be a good measure to capture the association between the attributive variable and  ~~$MRE$~~  for different types of relationships, such as linear, exponential and many complex functional relationships (Reshef et al., 2011). There is no need to standardize the data before the MIC calculation and the calculations have low computational complexity and high robustness. However, it should be noted that the association here does not refer to a specific correlation, such as temporal or spatial correlation, or positive or negative correlation, but refers to the strength of a certain relationship between the variable and  ~~$MRE$~~ . The MIC value is always between 0 and 1. The higher the MIC value, the stronger the association between the input variable and  $MRE$ , that is, the input variable contributes more significantly to the  $MRE$ . Here, we calculated the MIC base on the minepy package in Python (Albanese et al., 2018), and set the parameters required in MIC as the default settings suggested by the code developers. Different parameters had an insignificant effect on the relative importance of variables and  ~~$MRE$~~ .

We calculated the MIC between  $MRE$  and each one of the following input variables: the relative humidity ( $RH$ ), the mean vertical velocity ( $w_+$ ) and the sub-grid vertical velocity ( $w_{sub}$ ) to represent environmental and dynamic conditions; the total aerosol number ( $N_a$ ) as a proxy of pollution level; the hygroscopicity of aerosol ( $K_m$ ) weighted by composition volume fraction, and the effective radius of aerosol PSD ( $R_{e,a}$ ) to represent the chemical and size properties of the aerosol. Here,  $K_m$ , and  $R_{e,a}$  are defined as:

$$K_m = \frac{\sum_{c=1}^5 \frac{m_c \kappa_c}{\rho_c}}{\sum_{c=1}^5 \frac{m_c}{\rho_c}}, \quad (34)$$

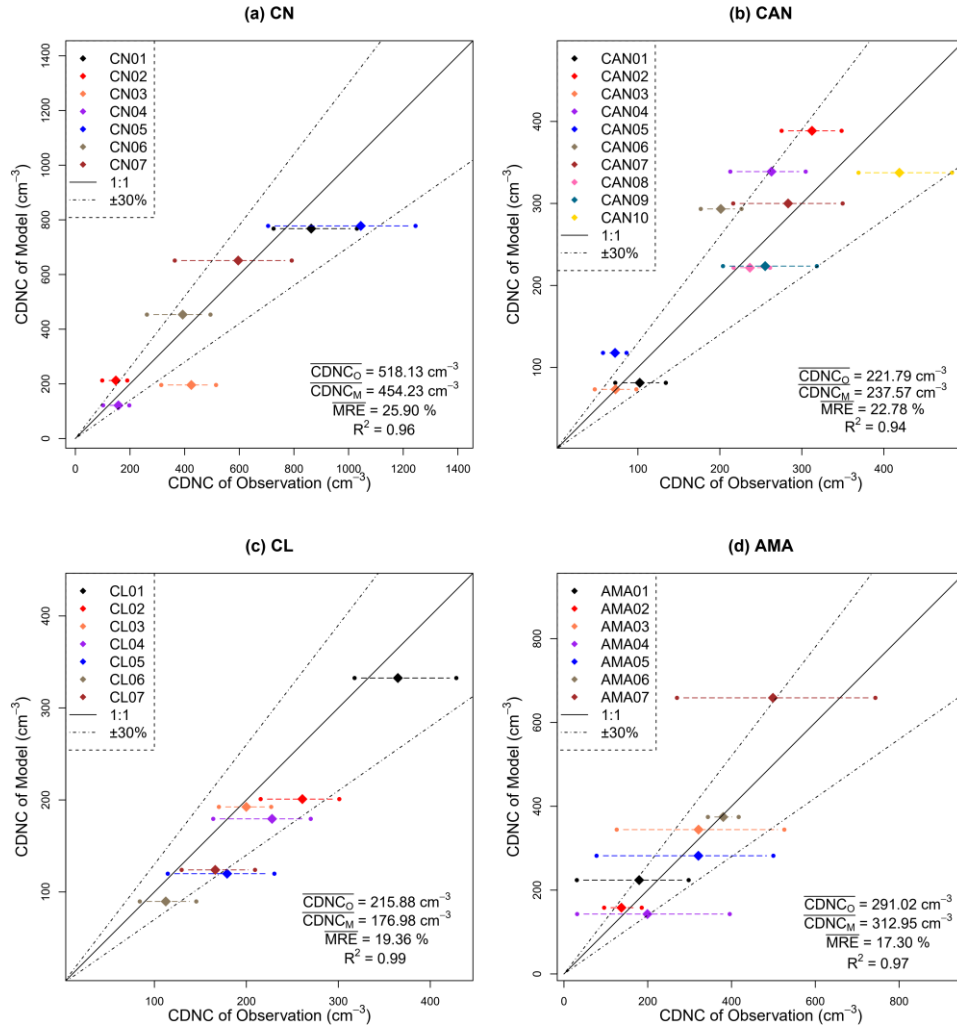
$$R_{e,a} = \frac{\sum_{j=1}^{n_j} R_j^3 N_{a,j}}{\sum_{j=1}^{n_j} R_j^2 N_{a,j}}, \quad (35)$$

where  $\kappa_c$ , the hygroscopicity of component  $c$ , is accounted for variations with relative humidity in the QDGE scheme ([Appendix A Sect. 2](#)).  $R_j$  represents the middle radius in the  $j^{th}$  particle size bin observed by PCASP or ASAP (see Sect. 3.2.2 and Table 2). For MIC calculation, the values of input variables derived from observations are listed in Table ~~A1~~ [B2](#) for each cloud case.

## 4 Results

### 4.1 Closure experiment

580 The results of the closure experiment are shown in Fig. 96. Almost all  $CDNC_M$  values fall within 30 % of the mean observations in the clouds.  $R^2$  is above 0.94 for all campaigns, which indicates a good agreement between simulation and observation. For the four campaigns covering marine to continental conditions, the  $\overline{MRE}$  values are all below 26 % ~~and the values are within  $\pm 20$  %~~. The AMA campaign produces the best agreement between model results and observations, with a  $\overline{MRE}$  value of 17.30 %. On the other hand, the CN campaign produces a poor agreement, with a  $\overline{MRE}$  value of 25.90 %.  
585 ~~However, cloud droplet number concentrations are underestimated for all cloud cases for the CL campaign (Fig. 9c==~~  
~~—19.36 %), which may be related to the high activation ratio ( $AR$ , the ratio of  $N_a$  to  $CDNC_o$ , see Table A4B2) in this region.~~  
 $AR$  in all CL cases are higher than 60 %, suggesting that the marine environment is favorable for more aerosol particles to be activated. If particles with a smaller size than the detection limit of PCASP (about 10 nm) are activated, it could lead to an underestimation of the simulated  $CDNC$  in the CL campaign.



590 **Figure 96.** A closure experiment between  $CDNC_O$  and  $CDNC_M$  for each cloud case in the (a) CN, (b) CAN, (c) CL, and (d) AMA  
 595 campaigns. The horizontal dashed lines represent the range of the observed  $CDNC$  within the 25% and 75% quantiles.

In order to provide further context, we compare the  $\overline{MRE}$  values of this study to previous studies with different aerosol activation parameterizations and aircraft measurements, as shown in Table 3. The  $\overline{MRE}$  values are relatively high for those early parameterizations, basically around 50 %. In the recent two decades, the performance of physically-based parameterization has been significantly improved, as is evident from a reduction of the  $\overline{MRE}$  to about 30 %. For instance, one of the schemes (Fountoukis and Nenes, 2005) achieved remarkable closure (with  $\overline{MRE}$  of 13.5 %) for continental cumuliform/stratus. In this study, the QDGE scheme performs decently (the  $\overline{MRE}$  values are all below 26 %) in four different regions, indicating that the scheme is suitable for simulations of cloud droplet number concentrations over a wide range of different meteorological conditions and different levels of aerosol pollution.

600 **Table 3. Comparison of results from simulations with activation schemes and the QDGE method (Mainly referring to Fountoukis et al. (2007))**

| Parameterization or Model                              | $\overline{MRE}$ (%) | Observed cloud type             | Location                   | Reference                                         |
|--------------------------------------------------------|----------------------|---------------------------------|----------------------------|---------------------------------------------------|
| Flossmann et al. (1985)                                | ~50.00               | Continental stratocumulus       | North of England           | (Hallberg et al., 1997)                           |
| UWyo parcel model*                                     | <50.00               | Marine stratocumulus            | Tenerife, Spain            | (Snider et al., 2003; Snider and Brenguier, 2000) |
| Fountoukis and Nenes (2005); Nenes and Seinfeld (2003) | ~30.00               | Coastal stratus                 | Monterey, California, USA  | (Meskhidze et al., 2005)                          |
| Fountoukis and Nenes (2005)                            | 13.50                | Continental cumuliform /stratus | Cleveland and Detroit, USA | (Fountoukis et al., 2007)                         |
| Kivekäs et al. (2008)                                  | ~35.00               | Continental stratus             | North of Finland           | (Kivekäs et al., 2008)                            |
|                                                        | 17.30                | Continental stratus             | Manaus, Brazil             |                                                   |
|                                                        | 19.36                | Marine stratocumulus            | Iquique, Chile             |                                                   |
| QDGE scheme                                            | 22.78                | Costal stratus                  | Nova Scotia, Canada        | This work                                         |
|                                                        | 25.90                | Continental stratus             | Beijing, China             |                                                   |

a. UWyo parcel model, available at <http://www.das.uwyo.edu/ccp/web>

#### 4.2 Error analysis

Although the performance of the QDGE scheme is good in different aircraft campaigns, it is useful to analyze sources of biases in the simulations. Following the procedures described in Sect. 3.3, we calculated the Maximum Information Coefficient (MIC) between ~~MRE~~ $\overline{MRE}$  and the input variables of the QDGE scheme, including aerosol properties ( $K_m$ , and  $R_{e,a}$ ), thermodynamic state ( $RH$ ), pollution level ( $N_a$ ), and atmosphere dynamic conditions ( $w_+$  and  $w_{sub}$ ), as shown in Table ~~A4B2~~[A4B2](#). The MIC values for all cloud cases and each campaign have been shown in Table 4.

For almost all campaigns, the aerosol number concentration and the hygroscopicity, have the most significant impacts on  $\overline{MRE}$ . This is consistent with the ~~change of environmental supersaturation (Eq. (3))droplet growth equation~~[change of environmental supersaturation \(Eq. \(3\)\)](#), according to which the variation of supersaturation  $S$  with height is essentially determined by the competition between the production of  $S$  by adiabatic cooling and the reduction in  $S$  from condensational growth of the particles, the latter mainly depends on the number and solubility of the aerosol particles. In detail,  $N_a$  has a greater impact on  $\overline{MRE}$  in marine regions (CAN and CL), but  $K_m$  is more significant in continental regions (CN and AMA). In marine regions, where  $N_a$  is relatively low (Table 2), a small fluctuation in  $N_a$  can cause noticeable changes in the simulated  $S_{max}$  and CDNC, which makes  $\overline{MRE}$  more sensitive to  $N_a$ . However, in continental areas,  $N_a$  is relatively high, and the change in hygroscopicity becomes more important to  $\overline{MRE}$ . The atmospheric humidity and the dry size of the aerosol particle also have non-negligible impacts on  $\overline{MRE}$ . Both affect the hygroscopic growth of aerosol particles and the reduction in  $S$ . Overall, the atmosphere dynamic conditions have the most

insignificant impact on  $MRE$ , which may be attributed to the weak variation of them in stratus and stratocumulus clouds (Table B2A4).

The MIC values also help to explain the relatively poor simulation performance of some campaigns. The chemical properties of the aerosol, which affect  $K_m$ , are very important for the simulation in the continental region, but the CN campaign lacks AMS data and we applied the same chemical composition for all cloud cases, based on earlier measurements in this region (Sect. 3.2.2). Given the importance of the chemical properties, simultaneous measurements of chemical components probably would have helped to enhance the accuracy of simulated  $CDNC$  for the CN campaign. Another possible cause of biases in simulated  $CDNC$  for the CN campaign is a much larger standard deviation of observed  $N_a$  (see Table 2) than that of other campaigns, which could be responsible for the error in the simulated  $CDNC$ . However, it should be noted that although the CAN campaign is characterized by the presence of coastal clouds and smaller variations in  $N_a$ , its  $MRE$  is higher than the AMA campaign, which may be related to the application of uniform updraft velocity in simulations for the CAN campaign (Sect. 3.2.3 and Table A4B2).

Overall speaking, the errors in the simulated  $CDNC$  is largely relevant to the missing data in observation (such as CN and CAM campaign), the analysis of MIC and error sources here could provide a good reason to develop and improve measurement strategies in the future aircraft campaigns.

**Table 4. The calculated MIC values between  $MRE$  and different input variables for all cloud cases and each campaign.**

|           | CN    |           | CAN   |           | CL    |           | AMA   |           | ALL   |
|-----------|-------|-----------|-------|-----------|-------|-----------|-------|-----------|-------|
| $K_m$     | 0.522 | $N_a$     | 0.610 | $RH$      | 0.522 | $K_m$     | 0.522 | $N_a$     | 0.343 |
| $RH$      | 0.522 | $K_m$     | 0.396 | $N_a$     | 0.470 | $N_a$     | 0.522 | $K_m$     | 0.315 |
| $N_a$     | 0.470 | $R_{e,a}$ | 0.396 | $K_m$     | 0.292 | $w_+$     | 0.470 | $RH$      | 0.242 |
| $w_+$     | 0.470 | $RH$      | 0.396 | $R_{e,a}$ | 0.198 | $w_{sub}$ | 0.470 | $R_{e,a}$ | 0.202 |
| $w_{sub}$ | 0.470 | $w_+$     | 0.000 | $w_+$     | 0.198 | $RH$      | 0.292 | $w_+$     | 0.170 |
| $R_{e,a}$ | 0.292 | $w_{sub}$ | 0.000 | $w_{sub}$ | 0.198 | $R_{e,a}$ | 0.198 | $w_{sub}$ | 0.170 |

## 5 Conclusions and discussion

In this paper, we introduce a numerically efficient aerosol activation scheme, which calculates the maximum cloud supersaturation and cloud droplet number concentration ( $CDNC$ ) by employing a Quasi-steady state approximation of the cloud Droplet Growth Equation (QDGE) scheme. The QDGE scheme utilizes the look-up tables and ~~an~~ iterative ~~method~~ calculation for solving the sub-level variation of supersaturation and deriving the maximum supersaturation and the activated particle number-size distribution-mass-and-energy budgets for efficient applications of the scheme in the large-scale grid of climate models. The comparison between the results of the QDGE scheme and a parcel model shows that biases in the maximum supersaturation under different environmental and aerosol conditions are within 0.18 % (with an average of 0.05 %), consistent with the ~~highindicating the decent accuracyand reasonable performance of of the QDGE scheme.~~ Whereafter,We we evaluated the simulated  $CDNC$  with worldwide cloud data sampled during four aircraft campaigns,

645 covering a wide range of different meteorological conditions and different levels of aerosol pollution. The aerosol information, updraft velocity, and meteorological conditions were carefully extracted from aircraft measurements and applied to drive the QDGE scheme. The simulated CDNC is compared with the observed correspondence in the nearly adiabatic part of the cloud, for evaluating the performance of the scheme. The average values of the mean relative error ~~and the mean bias~~ in the four campaigns are all within 26% ~~and  $\pm 20\%$ , respectively~~, indicating that the QDGE scheme can reasonably simulate the activated *CDNC* on a regional or global scale. We also investigated the potential sources of error in the simulated *CDNC* and found that the magnitude of the mean relative error is mostly relevant to the aerosol number concentration in marine regions and to aerosol hygroscopicity in continental regions than to other variables in the simulation. Several points are worthy of mentioning for future work. The QDGE scheme can be further optimized in several aspects. First,  $N_{sub} = 60$  generates reasonably good results in four different regions in this study, but this number is a little high and the computation will be too demanding to apply in general circulation models. Second, the iterative calculation to derive supersaturation in each sub-grid level can be computationally expensive. Therefore, both adjustments on  $N_{sub}$  number and optimization on the iteration would be necessary before the QDGE scheme is applied in the climate model. ~~Last, we also want to evaluate the QDGE scheme by comparing it with parcel model simulations, to further identify the sources of error related to the approximations in the scheme.~~ These works would be considered in future studies.

## 660 Appendix A: Parameters

The parameters  $A$ ,  $B$ ,  $C$ ,  $D$ , and  $E$  in Eqs. (1-3) are given by

$$A = \frac{2M_w\sigma}{RT\rho_w}, \quad (A1)$$

$$B = \kappa R_p^3, \quad (A2)$$

$$C = \frac{\rho_w RT}{e_* D'_v M_w} + \frac{L_v \rho_w}{K'_a T} \left( \frac{L_v M_w}{RT} - 1 \right), \quad (A3)$$

$$665 \quad D = \frac{g M_w L_v}{c_p R T^2} - \frac{g M_a}{RT}, \quad (A4)$$

$$E = \frac{P M_a}{e_* M_w} + \frac{M_w L_v^2}{c_p R T^2}, \quad (A5)$$

670 where  $\kappa$  is the aerosol hygroscopicity,  $\sigma$  is the surface tension of the solution/air interface (which is approximated by the surface tension of water here),  $\rho_w$  is the density of water,  $M_w$  is the molecular weight of water,  $R$  is the universal gas constant,  $T$  is the temperature,  $R_p$  is the dry aerosol particle radius,  $e_*$  is the saturation vapor pressure,  $L_v$  is the latent heat of vaporization,  $K'_a$  is the modified thermal conductivity of air accounting for non-continuum effects,  $D'_v$  is the modified diffusivity of water vapor in air accounting for non-continuum effects (Seinfeld and Pandis, 2016),  $g$  is the gravitational constant,  $M_a$  is the molecular weight of dry air,  $P$  is the atmospheric pressure, and  $c_p$  is the heat capacity at a constant pressure of dry air.

Petters and Kreidenweis (2007) and Kreidenweis et al. (2008) proposed provided tabulated values of the hygroscopicity-a  
 parameter  $\kappa$  for representing the hygroscopicity of aerosol with a variety of chemical compounds, and provided tabulated  
 values of  $\kappa$  based on laboratory data and modeling. They found that the aerosol water content (the ratio of wet aerosol  
 volume to the dry aerosol volume) parameterized on  $\kappa$  was generally within the experimental uncertainty, but biased at low  
 relative humidity (Kreidenweis et al., 2008; Petters and Kreidenweis, 2007). parameterized aerosol water contents are often  
 within experimental uncertainty. However, the accuracy of this approach tends to decrease with decreasing aerosol water  
 content. In particular, (Kreidenweis et al., (2008) Kreidenweis et al., (2008) also evaluated the calculated aerosol water  
 content based on  $\kappa$  simulations of highly concentrated, non ideal aqueous solutions with strong electrostatic interactions  
 between ions with the Aerosol Inorganic Model (AIM; Wexler and Clegg (2002)), which gives evidence for systematically  
 different results from a rigorous thermodynamic model at low aerosol water contents for some compounds. In order to  
 improve biases at low relative humidity, the original method was extended to account for variations in  $\kappa$  with relative  
 humidity in the QDGE scheme. Specifically, piecewise-linear relationships between  $\kappa$  and aerosol water activity for different  
 chemical components were determined based on results from AIM.

**Appendix B: Tables and FiguresA**

Table B1. Aerosol distribution and property parameters, referring to Whitby (1978) and (Ghan et al., (2011b).

| Aerosol type      | Mode number<br>concentration (cm <sup>-3</sup> ) | Mode radius (um)   | Mode geometric<br>standard deviation | $\kappa$ | $e$ | Condensation<br>coefficient |
|-------------------|--------------------------------------------------|--------------------|--------------------------------------|----------|-----|-----------------------------|
| Marine            | 340, 60, 3.1                                     | 0.005, 0.035, 0.31 | 1.6, 2.0, 2.7                        |          |     |                             |
| Clean continental | 1000,800,0.72                                    | 0.008, 0.034, 0.46 | 1.6, 2.1, 2.2                        | 0.7      | 0   | 1                           |
| Background        | 6400,2300,3.2                                    | 0.008, 0.038, 0.51 | 1.7, 2.0, 2.16                       |          |     |                             |
| Urban             | 106000,32000,5.4                                 | 0.007, 0.027, 0.43 | 1.8, 2.16, 2.21                      |          |     |                             |

Table BA21. A summary of observed ( $CDNC_o$ ,  $N_a$ ,  $RH$ , and  $LWC$ ), derived ( $AR$ ,  $Sol$ ,  $C_w$ ,  $K_m$ ,  $R_{e,a}$ ,  $w_+$ , and  $w_{sub}$ ), simulated and  
 evaluative ( $CDNC_M$ , ~~MB~~, ~~a~~ and  $MRE$ ) variables of each cloud case in four campaigns.

| Case | Observed variables              |                              |             |                                | Derived variables |              |                                                                   |       |                          |                               |                                   |                                 | Simulated and<br>evaluative variables       |                                            |
|------|---------------------------------|------------------------------|-------------|--------------------------------|-------------------|--------------|-------------------------------------------------------------------|-------|--------------------------|-------------------------------|-----------------------------------|---------------------------------|---------------------------------------------|--------------------------------------------|
|      | $CDNC_o$<br>(cm <sup>-3</sup> ) | $N_a$<br>(cm <sup>-3</sup> ) | $RH$<br>(%) | $LWC$<br>(g cm <sup>-3</sup> ) | $AR$<br>(%)       | $Sol$<br>(%) | <del><math>C_w</math></del><br>$10^{-3}(10^{-3}$<br>$(g cm^{-4})$ | $K_m$ | $R_{e,a}$<br>( $\mu m$ ) | $w_+$<br>(m s <sup>-1</sup> ) | $w_{sub}$<br>(m s <sup>-1</sup> ) | $CDNC_M$<br>(cm <sup>-3</sup> ) | <del><math>MRE</math></del><br>$MRE$<br>(%) | <del><math>MB</math></del><br>$MRE$<br>(%) |
| CN01 | 863.25                          | 3016.27                      | 67.92       | 0.20                           | 28.62             | 65.00        | 0.69                                                              | 0.37  | 0.23                     | 0.469                         | 0.340                             | 767.86                          | -11.05                                      |                                            |

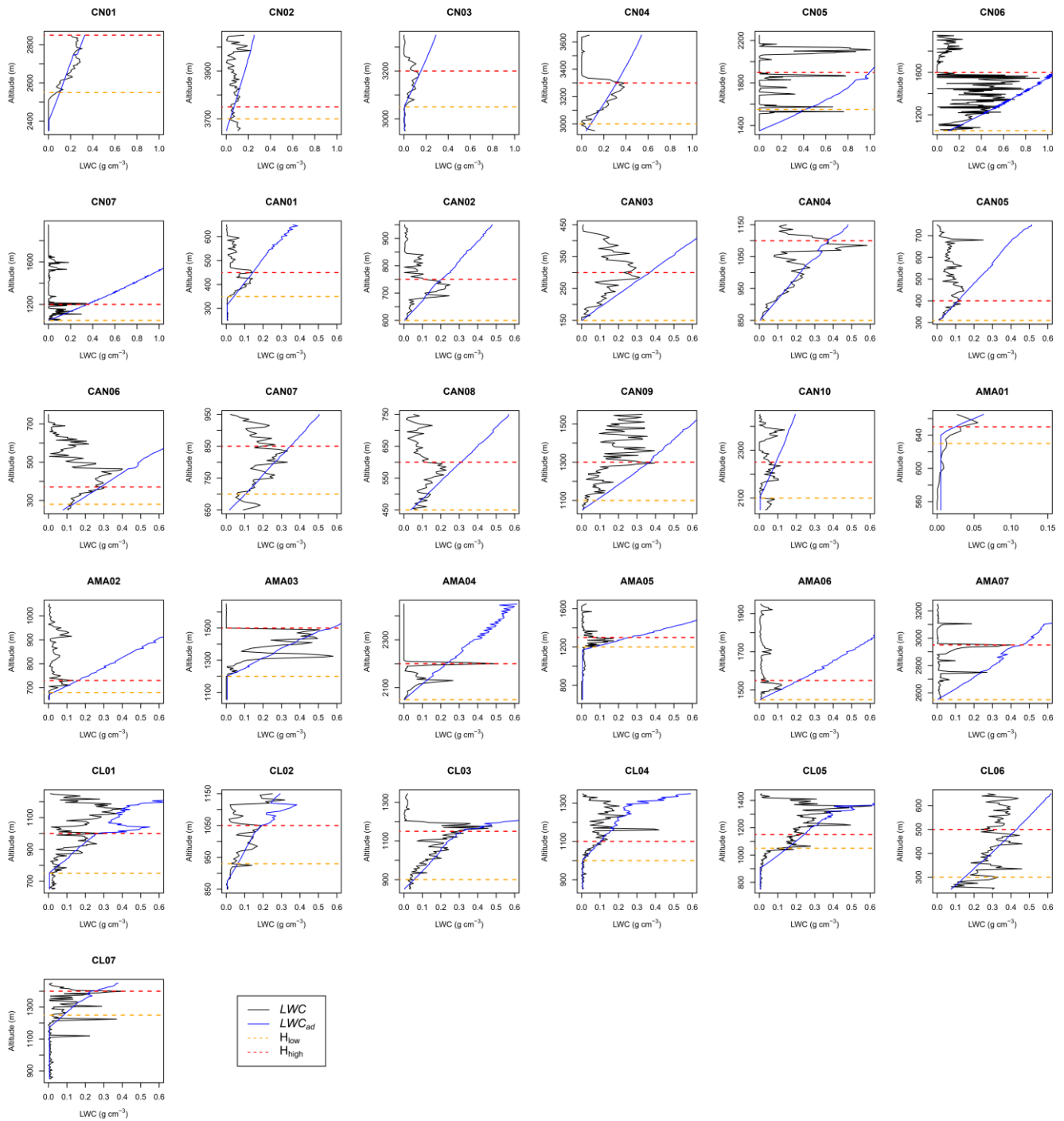
|  |       |         |         |              |      |       |       |      |             |      |       |       |        |                  |
|--|-------|---------|---------|--------------|------|-------|-------|------|-------------|------|-------|-------|--------|------------------|
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 11.05            |
|  | CN02  | 148.17  | 372.77  | 61.89        | 0.06 | 39.75 | 65.00 | 0.71 | 0.39        | 0.41 | 0.609 | 0.441 | 212.3  | <del>43.28</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 43.28            |
|  | CN03  | 424.41  | 432.05  | 61.89        | 0.08 | 98.23 | 65.00 | 1.04 | 0.39        | 0.15 | 0.609 | 0.441 | 195.84 | <del>53.86</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 53.86            |
|  | CN04  | 157.49  | 1738.09 | 57.71        | 0.12 | 9.06  | 65.00 | 0.81 | <u>0.40</u> | 0.98 | 0.609 | 0.441 | 121.33 | <del>22.96</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 22.96            |
|  | CN05  | 1044.72 | 1550.93 | 88.12        | 0.43 | 67.36 | 65.00 | 1.99 | 0.33        | 0.18 | 0.714 | 0.516 | 777.82 | <del>25.55</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 25.55            |
|  | CN06  | 392.89  | 850.10  | 72.42        | 0.22 | 46.22 | 65.00 | 1.93 | 0.35        | 0.56 | 0.444 | 0.314 | 453.34 | <del>15.39</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 15.39            |
|  | CN07  | 596.01  | 1486.6  | 66.79        | 0.11 | 40.09 | 65.00 | 2.36 | 0.37        | 0.22 | 0.609 | 0.441 | 651.10 | <del>9.24</del>  |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 9.24             |
|  | CAN01 | 102.28  | 108.26  | 95.27        | 0.12 | 94.48 | 62.50 | 1.03 | 0.54        | 0.84 | 0.299 | 0.215 | 81.26  | <del>20.55</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 20.55            |
|  | CAN02 | 312.43  | 461.86  | 82.95        | 0.23 | 67.65 | 73.95 | 1.37 | 0.76        | 0.17 | 0.299 | 0.215 | 388.57 | <del>24.37</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 24.37            |
|  | CAN03 | 72.69   | 110.60  | 97.07        | 0.28 | 65.72 | 79.40 | 2.40 | 0.68        | 0.3  | 0.299 | 0.215 | 73.31  | <del>0.85</del>  |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 0.85             |
|  | CAN04 | 263.02  | 547.91  | <u>86.30</u> | 0.22 | 48.00 | 73.95 | 1.50 | 0.71        | 0.67 | 0.299 | 0.215 | 338.82 | <del>28.82</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 28.82            |
|  | CAN05 | 72.12   | 176.43  | <u>84.60</u> | 0.11 | 40.88 | 62.50 | 1.15 | 0.65        | 0.28 | 0.299 | 0.215 | 117.77 | <del>63.30</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 63.30            |
|  | CAN06 | 201.15  | 441.24  | 90.82        | 0.19 | 45.59 | 73.95 | 1.67 | 0.66        | 0.85 | 0.299 | 0.215 | 293.30 | <del>45.81</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 45.81            |
|  | CAN07 | 283.26  | 673.60  | 84.23        | 0.18 | 42.05 | 73.95 | 1.67 | 0.74        | 0.18 | 0.299 | 0.215 | 299.97 | <del>5.90</del>  |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 5.90             |
|  | CAN08 | 236.61  | 561.35  | 79.83        | 0.25 | 42.15 | 73.95 | 1.82 | 0.79        | 0.22 | 0.299 | 0.215 | 221.63 | <del>6.33</del>  |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 6.33             |
|  | CAN09 | 255.29  | 1064.55 | 79.83        | 0.26 | 23.98 | 73.95 | 1.51 | 0.79        | 0.31 | 0.299 | 0.215 | 223.57 | <del>12.43</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 12.43            |
|  | CAN10 | 419.06  | 609.57  | 81.25        | 0.21 | 68.75 | 73.95 | 0.62 | 0.78        | 0.12 | 0.299 | 0.215 | 337.48 | <del>19.47</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 19.47            |
|  | CL01  | 364.78  | 493.78  | 54.36        | 0.15 | 73.88 | 72.25 | 2.54 | <u>0.60</u> | 0.13 | 0.618 | 0.447 | 332.53 | <del>8.84</del>  |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 8.84             |
|  | CL02  | 260.91  | 339.76  | 64.86        | 0.13 | 76.79 | 84.79 | 2.70 | 0.59        | 0.13 | 0.537 | 0.389 | 200.93 | <del>22.99</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 22.99            |
|  | CL03  | 199.93  | 309.33  | 41.98        | 0.18 | 64.63 | 80.27 | 1.86 | 0.74        | 0.14 | 0.618 | 0.447 | 192.45 | <del>3.74</del>  |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 3.74             |
|  | CL04  | 227.94  | 272.76  | 40.43        | 0.09 | 83.57 | 70.36 | 1.53 | 0.96        | 0.13 | 0.618 | 0.447 | 179.44 | <del>21.28</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 21.28            |
|  | CL05  | 179.08  | 187.54  | 57.02        | 0.19 | 95.49 | 79.45 | 2.06 | 0.63        | 0.12 | 0.618 | 0.447 | 119.84 | <del>33.08</del> |
|  |       |         |         |              |      |       |       |      |             |      |       |       |        | 33.08            |

|       |        |         |       |      |       |       |      |      |      |       |       |        |                            |
|-------|--------|---------|-------|------|-------|-------|------|------|------|-------|-------|--------|----------------------------|
| CL06  | 112.37 | 141.17  | 67.65 | 0.31 | 79.60 | 83.83 | 2.19 | 0.58 | 0.33 | 0.429 | 0.310 | 89.67  | <del>-20.20</del><br>20.20 |
| CL07  | 166.17 | 226.35  | 58.74 | 0.22 | 73.41 | 91.20 | 1.21 | 0.72 | 0.20 | 1.189 | 0.694 | 123.98 | <del>-25.39</del><br>25.39 |
| AMA01 | 179.50 | 307.47  | 90.50 | 0.09 | 58.38 | 17.94 | 1.07 | 0.07 | 0.86 | 0.761 | 0.55  | 223.88 | <del>24.72</del><br>24.72  |
| AMA02 | 137.19 | 296.02  | 84.32 | 0.10 | 46.34 | 27.56 | 1.01 | 0.12 | 0.68 | 1.074 | 0.777 | 158.08 | <del>15.23</del><br>15.23  |
| AMA03 | 321.21 | 548.11  | 78.67 | 0.30 | 58.60 | 26.58 | 1.03 | 0.12 | 0.77 | 1.203 | 0.870 | 344.32 | <del>7.19</del><br>7.19    |
| AMA04 | 199.21 | 368.46  | 78.25 | 0.32 | 54.07 | 26.58 | 1.06 | 0.11 | 0.76 | 1.628 | 1.178 | 142.86 | <del>28.29</del><br>28.29  |
| AMA05 | 320.88 | 445.44  | 77.21 | 0.30 | 72.04 | 18.91 | 0.99 | 0.07 | 0.72 | 0.959 | 0.595 | 281.98 | <del>12.12</del><br>12.12  |
| AMA06 | 380.27 | 1535.06 | 59.22 | 0.13 | 24.77 | 16.86 | 1.46 | 0.12 | 0.20 | 1.074 | 0.777 | 374.47 | <del>1.53</del><br>1.53    |
| AMA07 | 498.91 | 2419.76 | 68.04 | 0.32 | 20.62 | 29.36 | 1.03 | 0.11 | 0.35 | 1.245 | 0.901 | 658.73 | <del>32.03</del><br>32.03  |

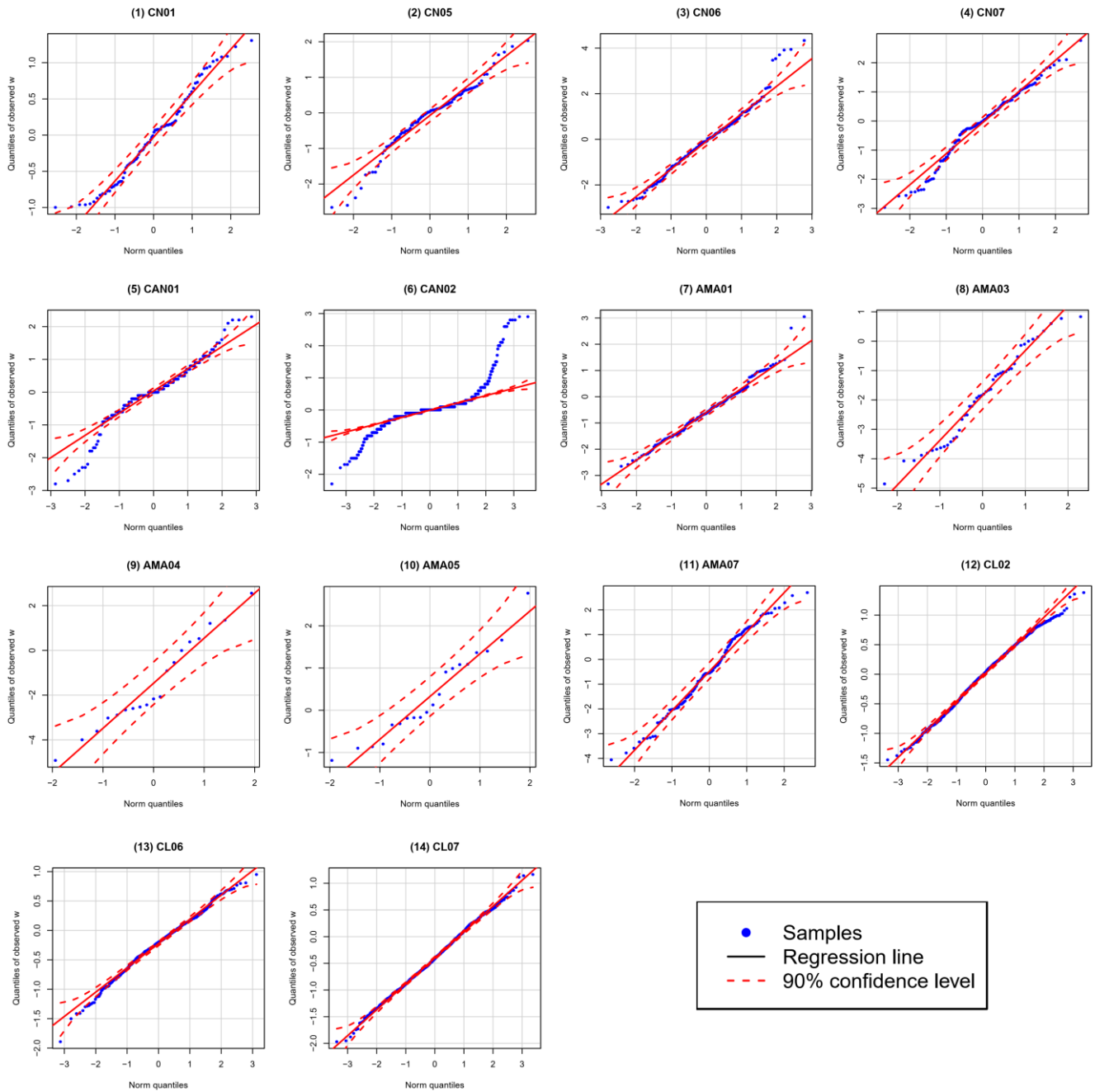
**Table BA23.** The observed mass fractions of different aerosol compositions in Beijing, China in two previous studies, as well as the assumed fractions used in this work.

| Date              | Particle size range | Sampler           | <i>org</i> fraction | $SO_4^{2-}$ fraction | $NO_3^-$ fraction | $NH_4^+$ fraction | $Cl^-$ fraction | Reference          |
|-------------------|---------------------|-------------------|---------------------|----------------------|-------------------|-------------------|-----------------|--------------------|
| Summer, 2017/2018 | PM1                 | ACSM <sup>a</sup> | 37%                 | 26%                  | 22%               | 14%               | 1%              | Zhou, et al., 2019 |
| Summer, 2018      | PM2.5               | ACSM              | 34%                 | 31%                  | 22%               | 13%               | ~1%             | Li, et al., 2020   |
| Summer, 2018      | 0.01~3um            | PCASP             | 35%                 | 29%                  | 22%               | 13%               | 1%              | This work          |

a. ACSM: Aerosol Chemical Speciation Monitor.



**Figure AB41.** The profiles of observed  $LWC$  (black) and adiabatic  $LWC$  ( $LWC_{ad}$ , blue) for 31 liquid water cases.



700 Figure BA22. The normal quantile-quantile plot for comparing the observed  $w$  sampled by aircraft with a standard normal distribution, for each cloud case with sufficient data. The linearity of the data points (blue dots) suggests that the observed  $w$  are normally distributed under a 90 % confidence level.

Code and data availability. The version of the QDGE scheme used to produce the results used in this paper, as well as the  
705 input data and scripts to run the model and the data to produce the key plot for the simulations, are archived on Zenodo and  
can be accessed at <https://doi.org/10.5281/zenodo.4841035> (Wang et al., 2021).

Author contribution. HW processed all data, conducted all simulations and analyses, and wrote the manuscript. YP led the  
work, designed the experiment, and refined the manuscript. KS developed the initial model version of the QDGE scheme  
and provided a summary of the approach, and ~~contributed to the writing of~~ refined the manuscript. YY, WZ, and DZ helped  
710 with the data usage in the China campaign and refined the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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