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Parameterization of the nitric acid effect on CCN activation

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Abstract. In this paper we present a parameterization of the nitric acid effect on cloud droplet formation. The new parameterization is intended to be used in large scale models in order to obtain regional and global estimates of the effect of nitric acid on cloud drop concentrations and the radiative balance. The parameterization is based on numerical air parcel model simulations and can be applied for unimodal and bimodal lognormal aerosol particle size distributions in a large variety of different conditions. In addition to the aerosol particle distribution and gas-phase HNO_3 concentration, the parameterization requires temperature, total pressure, updraft velocity, and the number concentration of cloud droplets formed at zero nitric acid concentration, as input parameters. The parameterization is also suitable for describing the effect of hydrochloric acid on the cloud drop concentrations, and in practice, the HNO_3 and HCl concentrations can be summed up to yield the total effect. The comparison between the parameterization and the results from numerical air parcel model simulations show good consistency.

1 Introduction

Previous cloud model studies have shown that water-soluble trace gases such as nitric acid (HNO_3) and ammonia (NH_3) can increase the cloud droplet number concentration (CDNC) significantly (Kulmala et al., 1993; Korhonen et al., 1996; Hegg, 2000). The increase in the cloud droplet number concentration will reduce the mean cloud droplet size which in turn increases cloud albedo (Twomey, 1974). Increased CDNC and decreased cloud droplet size can also decrease the precipitation formation efficiency prolonging the cloud lifetime and increasing the cloud coverage (Albrecht, 1989). Thus, the soluble gases can induce similar effects on clouds as the first and the second indirect aerosol effects. Overall, the increase in CDNC will lead to the cooling of the lower troposphere and so the effect is opposite in sign to the warming effect of the greenhouse gases.

The magnitude of the effect of water-soluble trace gases on cloud condensation nucleus (CCN) activation can be calculated using a cloud parcel model, but to estimate the global albedo effect reliably, large scale models such as GCM's need to be used. Because the numerical calculation of the trace gas effect is computationally demanding and there is no analytical solution for deriving the CDNC directly from the properties of aerosol size distribution and environmental variables, the results obtained from parcel model simulations have to be parameterized into a form that can be used in the large scale models. In this study, we are developing a parameterization for the HNO_3 effect on aerosol activation.

In the recent years, many parameterizations for cloud droplet formation have been developed. The most straightforward parameterizations are based on direct measurements relating the number of condensation nuclei to the number of cloud droplets (Gultepe and Isaac, 1999) or relating the non-sea-salt sulfate concentration to the number of cloud droplets (Boucher and Lohmann, 1995; Lowenthal et al., 2004). Such parameterizations do not take into account any variation in the shape of aerosol population or cloud forming dynamics. Twomey (1959) proposed a power law relation (CDNC=cS^k) to determine CDNC as a function of cloud supersaturation S. This relation has later been refined to obtain more accurate results (Cohard et al., 1998, 2000). Some other parameterizations include theoretical calculations combined with detailed numerical parcel model simulations (Nenes and Seinfeld, 2003; Abdul-Razzak and Ghan, 2002). The basic idea in these parameterizations is to estimate the maximum water supersaturation S_{max} in the cloud parcel. S_{max} is compared to the critical supersaturation spectra of the aerosol particle distribution, which can be calculated from Köhler theory as a function of dry particle diameter $S_c(D_p)$ and, in a simplified

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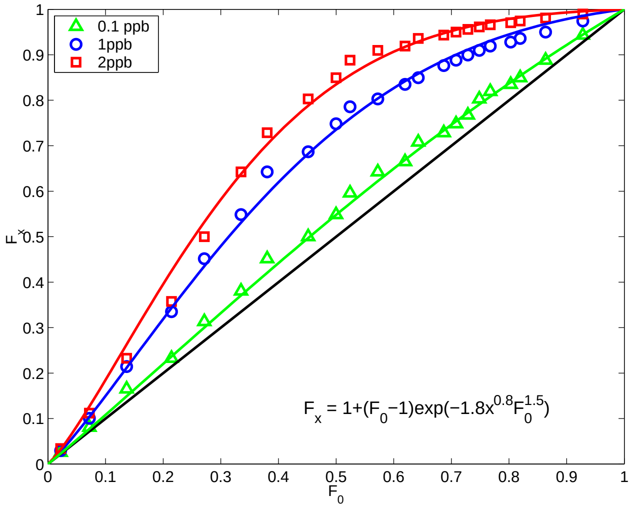

Fig. 1. F_x as a function of F_0 for three different HNO_3 concentrations and lines are calculated with fitted Eq. (2).

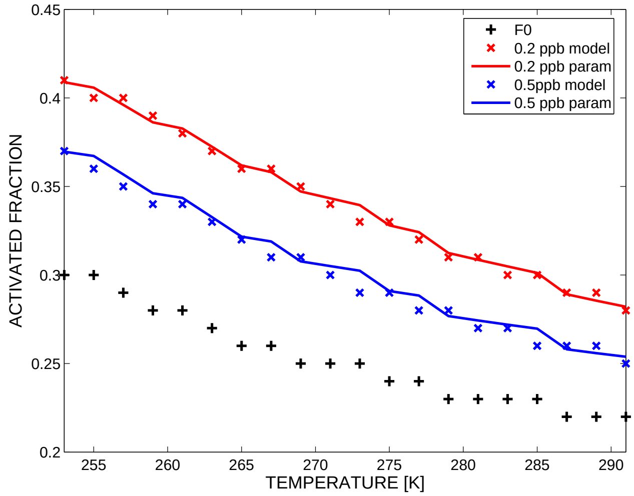

Fig. 2. Activated fraction as a function of temperature for three different HNO_3 concentrations as modeled by parcel model and corresponding predictions by new parameterization.

manner, every aerosol particle with $S_c < S_{max}$ is then supposed to activate to a cloud droplet. These parameterizations take into account the dynamics and the size distribution of the aerosol population, but still the role of condensing gases other than water is undetermined.

2 Parameterization

Condensed hygroscopic compounds lower the particles' S_c, enhancing the ability of an individual aerosol particle to activate to a cloud droplet and thereby increasing CDNC. At the same time, the total mass of the aerosol particle population increases, leading to decrease in S_{max}. This will tend to decrease the CDNC, but in most situations this effect is smaller than the increasing effect of the lower critical supersaturations. In general, the number of activated droplets depends on how the condensed gases are distributed among the particles with different diameters. When the condensation of the trace gases takes place rapidly – i.e. the system is strongly out of equilibrium – small particles collect relatively more of the gases than larger ones, which favors increased activation of cloud drops.

Parameterizing the HNO_3 effect on CCN activation is not a straightforward task to do. If the amount of HNO_3 in the droplets of different sizes could be estimated at the time of activation, the $S_c(D_p)$ could be calculated from Köhler theory and S_{max} could be estimated with methods given by Nenes and Seinfeld (2003) or by Abdul-Razzak and Ghan (2002). The amount of condensed semivolatile matter in droplets with different sizes can be calculated with a size resolved numerical model that solves the differential equations describing the condensation. Condensation depends on the concentration difference between the ambient air and the surface of droplet, and to estimate the condensation rate into a droplet, the surface concentration must be known. However, it turns out that during the activation process, there can be an order of magnitude difference in surface concentrations between small and big droplets and therefore it is difficult to get an analytical solution for partitioning of nitric acid to droplets of different sizes.

Instead of very approximate analytical calculations, we choose to use a more straightforward method. Our new parameterization is based on numerical simulations, which are done with an air parcel model including detailed liquid phase thermodynamics and differential equations for adiabatic expansion and condensation growth of liquid particles (for more information about the model, see Kokkola, 2003). Our strategy is to develop a parameterization which can be used to calculate the activated fraction F_x with x ppb of HNO_3 in the gas phase, when the activated fraction without other condensing gases than water, F_0, is known. The basic form of the parameterization is similar to a parameterization previously presented by Laaksonen et al. (1998)

$$F_x = 1 + (F_0 - 1)\exp\left(f\left(C, T, F_0, V, dist\right)\right), \tag{1}$$

where $f\left(C, T, F_0, V, dist\right)$ is a function of HNO_3 concentration C, temperature T, F_0, updraft velocity V, and properties of the aerosol distribution (denoted with $dist$).

The origin of the mathematical form is in notion that the F_x vs. F_0-curves qualitatively resemble vapor-liquid equilibrium curves in some binary systems. That can be seen in Fig. 1, where F_x is presented as a function of F_0 for some simple distribution at typical conditions. Different F_0 values have been obtained at different updraft velocities and in this case the fitted equation is as simple as

$$F_x = 1 + (F_0 - 1)\exp\left(-1.8x^{0.8}F_0^{1.5}\right). \tag{2}$$

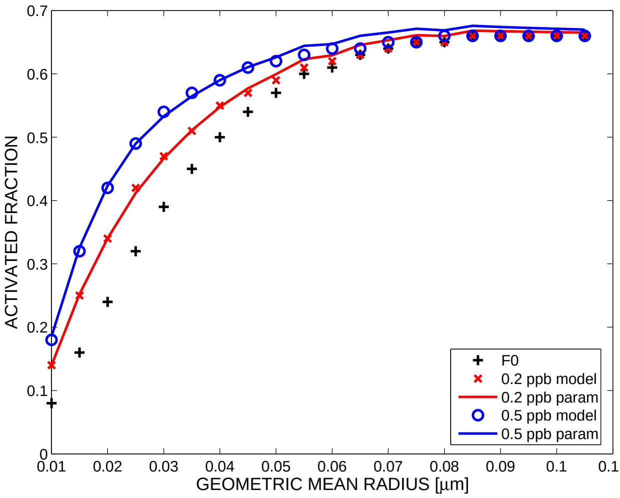

Fig. 3. Activated fraction as a function of geometric mean radius as modeled by parcel model and corresponding predictions by new parameterization when V=0.5 m/s, N=1000 cm^{-3} and σ=1.8.

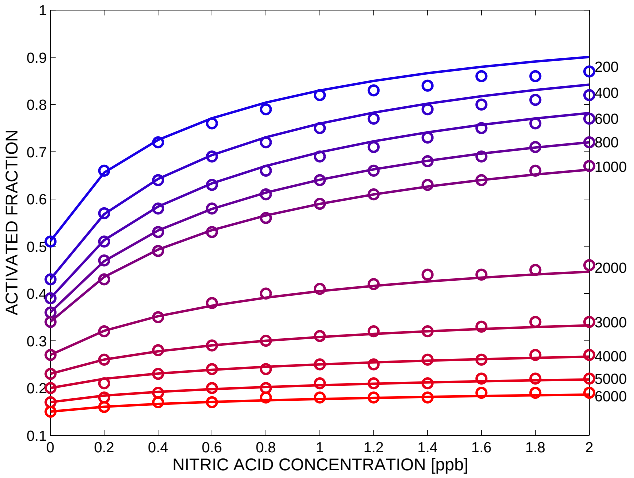

Fig. 4. Activated fraction as a function of aerosol particle and nitric acid concentration. In these model runs r_g is 26 nm, σ is 1.8, T is 273.15 K, and V is 0.5 m/s.

We can see, that the correspondence between Van Laar type Eq. (2) and F_x vs. F_0 data, is good. Beyond that, this form is physically reasonable because it is limited to unity. However, in this simple example only the updraft velocity is varied and to take the other variables also into account, the Eq. (2) must be modified. The final form of the parameterization is presented in Table 1.

With this set of equations, the effect of HNO_3 on aerosol activation to cloud droplets can be calculated for a wide variety of conditions. The parameterization can be used to study liquid phase clouds with the following limitations. The minimum values for T and P are 253 K and 300 mbar, respectively. V has to be positive and higher than 0 m/s. The new parameterization is valid for bimodal lognormal aerosol size distributions. The modes can have different chemical composition, and the single modes can be internally mixed. The particle composition is divided to insoluble and soluble parts based on mass fractions. Inorganic substances are described by ammoniated sulfates with 0:1, 1:1 and 2:1 ammonium-to-sulfate ratios. Also, a sodium chloride mode is possible. Water-insoluble organics can be represented using the insoluble fraction. Water soluble organics are not included in the parameterization. Properties of the different modes are limited in such a way that the geometric standard deviation of both modes need to be higher than 1.2, $r^{\star}_{gi}$ need to be higher than 5 nm and $r^{\star}_{g2} \geq r^{\star}_{g1}$. The functional form of the parameterization (Eqs. 3 to 11) is such, that N_1 can not be zero.

3 Comparison of parameterization with parcel model

We have done simulations with and without nitric acid as a function of one or two basic parameters while the other variables are kept constant. These simulations are presented in Figs. 2–8, where we show comparisons between the new parameterization and parcel model simulations for different aerosol and dynamical properties. In Figs. 2–5 we present comparisons for unimodal and in Figs. 6–7 for bimodal aerosol distributions.

Decreasing temperature increases the HNO_3 solubility in the water solutions and so the condensation takes place at lower RH than at warm conditions. Also, the amount of water in the air is smaller than at warm conditions and so the condensation of water into the droplets is slower, increasing the S_{max}. Overall, there is more time for HNO_3 condensation at low temperatures and so the effect of HNO_3 on CDNC increases with decreasing temperature. This can be seen in Fig. 2, where the activated fraction is presented as a function of temperature for two different HNO_3 concentrations when the updraft velocity is 0.2 m/s. In this case, the aerosol is composed of ammonium sulfate and the size distribution is unimodal with number concentration, geometric mean radius, and geometric standard deviation of 1000 cm^{-3}, 30 nm, and 1.8, respectively. The step-like behavior of the parameterization curve is due to the usage of model results as F_0 values. In the model, the size distribution is discretized to a reasonable number of size classes (bins). In this simulation number of bins was 110 and so the fraction of droplets in one size bin is ∼1%, when the bins are equally spaced in aerosol number. As can be seen in Fig. 2, the difference between model results and predictions of the parameterization are within the uncertainty produced by the discretization.

The strength of the HNO_3 effect depends on the amount of hygroscopic material in the pre-existing aerosol particle distribution. Increase in the mean size or total number concentration of aerosol particles will decrease the HNO_3 effect. As shown in Fig. 3, without nitric acid the CDNC increases with increasing radius and approaches asymptotically some

Table 1. Parameterization for bimodal lognormal aerosol size distribution. Variables: P_{TOT} is total pressure (mbar), C_{HNO_3} is nitric acid concentration (ppb), T is temperature (K), T_0=273.15 K, V is updraft velocity (ms^{-1}), r_{gi} is geometric number mean radius (µm) of mode i, X_{mi} is water soluble mass fraction in aerosol particles in mode i, $r^{\star}_{gi}=r_{gi}X^{0.46}_{mi}$, N is total particle concentration (cm^{-3}), N_i is particle concentration in mode i, σ_i is geometric standard deviation in mode i, F_0 is activated fraction without nitric acid, and F is the activated fraction with C_{HNO_3} ppb of nitric acid in the air. B_c is a variable that takes account the chemical composition and it is 0 for ammonium sulfate and sodium chloride, 1 for ammonium bisulfate and 2 for sulfuric acid. With this set of equations it is supposed that $r^{\star}_{g2}>r^{\star}_{g1}$.

$$r_g = \frac{N_1 r^{\star}_{g1}+N_2 r^{\star}_{g2}}{N} \tag{3}$$

$$B_c = \frac{N_1 r^{\star}_{g1} B_{c1}+N_2 r^{\star}_{g2} B_{c2}}{N_1 r^{\star}_{g1}+N_2 r^{\star}_{g2}} \tag{4}$$

$$A = C_{HNO_3} 1.03^{(308-T)} \tag{5}$$

$$B_1 = 0.138 V^{0.16} \left(r_g\right)^{-2.04} N^{-0.77} A^{0.5} \left(\frac{T}{T_0}\right)^{7.8} 1.57^{B_c} V^{0.2} A^{-0.3} \tag{6}$$

$$B_2 = 2.24 \times 10^{-5} \left(F_0 + 8.54 \frac{A}{N}\right)^{1.4} (V)^{-1.86} A^{0.47} N^{1.94} r_g \left(\frac{T_0}{T}\right)^{2.36} 0.87^{B_c} \tag{7}$$

$$B_3 = \frac{0.22}{0.82^{(B_{c1}-B_{c2})} V^{-0.6} A^{-0.42}} \left(\frac{r^{\star}_{g2}-r^{\star}_{g1}}{r^{\star}_{g1}}\right)^{1.15} \left(\frac{N_2 N_1}{N^2}\right) (F_0+0.24)^{-1.71} \quad , F_0>N_2/N \tag{8}$$

$$B_3 = 0.23 \left(\frac{r^{\star}_{g2}-r^{\star}_{g1}}{r^{\star}_{g1}}\right)^{1.05} \left(\frac{N_2 N_1}{N^2}\right)^{0.13} \left(\frac{F_0 N}{N_2}\right)^{\left(\frac{r^{\star}_{g2}-r^{\star}_{g1}}{r^{\star}_{g1}}\right)\left(\frac{N_2}{N_1}\right)^{1.3}} \quad , F_0<N_2/N \tag{9}$$

$$f_N = \left(\frac{1.75N}{(N_1\sigma_1+N_2\sigma_2)(1+B_3)}\right)^{\left(18.3 r_g^{0.86} F_0^{0.15} N^{0.2} \left(\frac{T_0}{T}\right)^2\right)} \tag{10}$$

$$F = 1 + (F_0 - 1) \exp\left(-2.31 \left(\frac{A P_{TOT}}{900(1+B_1)(1+B_2) N^{0.65} r_g^{1.11}}\right)^{1.17} F_0^{0.54} f_N\right) \tag{11}$$

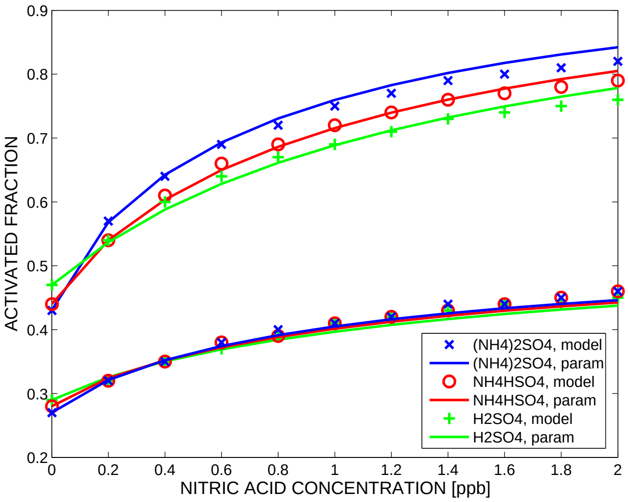

Fig. 5. Activated fraction as a function of aerosol particle number, nitric acid concentration and aerosol particle chemical composition. In the upper set of lines total N=400 cm^{-3} and in the lower set N=2000 cm^{-3}.

limiting value. With HNO_3 in the air this value can be reached with smaller dry particles. In the case of Fig. 3, the

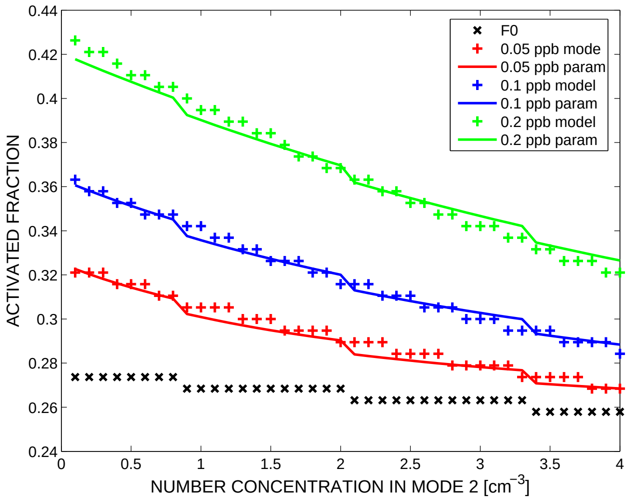

Fig. 6. Activated fraction as a function of number concentration in the coarse mode.

particles are completely water soluble, but in general, some of the aerosol material can of course be insoluble. The effect of insoluble aerosol material is taken into account in the parameterization simply by multiplying the r_g with $(X_m)^{0.46}$.

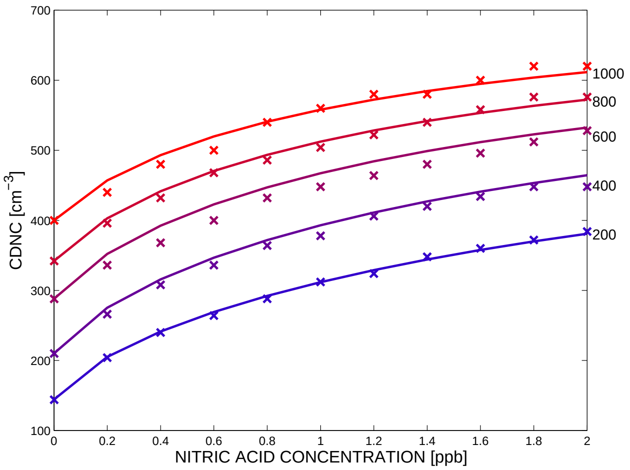

Fig. 7. Number of activated droplets as a function of aerosol particle number concentration in mode 2 and nitric acid concentration.

Table 2. Initial values for cloud model runs for the marine and continental cases examined.

	Marine case		Continental case	
Composition				
mode 1	NH_4HSO_4		$(NH_4)_2SO_4$	
mode 2	NaCl		$(NH_4)_2SO_4$	
	mode 1	mode 2	mode 1	mode 2
N [cm^{-3}]	141	variable	1000	variable
r_g [nm]	26	230	10	30
σ	1.75	2.10	1.70	2.10

In Fig. 4, we show how higher aerosol number concentrations decrease the HNO_3 effect. The increasing particle number concentration decreases the activated fraction, but increases the number of activating particles. Naturally, when the aerosol mass increases, the nitric acid effect decreases. When N=200 cm^{-3}, CDNC increases from 100 cm^{-3} to 160 cm^{-3} as the HNO_3 concentration increases from 0 to 1 ppb and only 0.2 ppb is needed to increase it to 130 cm^{-3}. When N=6000 cm^{-3}, the corresponding CDNC's are 900 cm^{-3}, 1180 cm^{-3}, and 950 cm^{-3}. Based on these numbers it is clear that the HNO_3 effect is more important when the total aerosol number concentration is low. However, with low updraft velocities and narrow size distribution, the HNO_3 effect can be remarkable even when particle number concentrations are thousands per cubic centimeter.

The strength of the HNO_3 effect depends on the acidity of aerosol particles (Hegg, 2000). To take this into account, we have included the variable B_c into parameterization. B_c is related to the number of hydrogen ions released from salt to solution and so it represents the acidity of pre-existing aerosol particles. In the case of acidic particles at low concentration, the uptake of HNO_3 occurs at high relative humidity and so there is not enough time for all HNO_3 to condense before activation occurs. At high aerosol concentration, the total particle mass is higher, allowing faster depletion of HNO_3 and decreasing the importance of particle acidity in regulating the nitric acid effect on the CDNC. Also, the overall effect of nitric acid is lower at high aerosol concentrations, so the effect of aerosol acidity is more difficult to see. In Fig. 5 we show examples of the effect of chemical composition (inducing different acidities) for the same distribution as in Fig. 4, but only for two different values of aerosol particle number concentrations.

The effect of chemical composition is especially important in situations, where the aerosol are externally mixed so that different modes have different compositions. In marine areas, the aerosol are composed of alkaline (sea salt) haze mode and more acidic nucleation and accumulation modes. Due to their large mass, sea salt particles decrease the number concentrations of cloud droplets. HNO_3 tends to condense into aqueous sodium chloride droplets at lower RH than to more acidic aerosols. The NaCl particles are big enough so that all of them will activate even without the presence of HNO_3, thus decreasing the effect of HNO_3 on the CDNC. This is demonstrated in Fig. 6 where the activated fraction is presented as a function of coarse mode particle concentration for three different HNO_3 concentrations. The aerosol size distribution is the same as used by Hegg (2000) in his study and it is presented in Table 2. The updraft velocity and temperature are 0.1 m/s and 273.15 K, respectively.

Continental conditions differ clearly from marine conditions. Both the total number of particles and the shape of the particle size distribution are typically totally different. The number of coarse mode particles in the marine environment is crucial when estimating CDNC but at continental conditions, the fraction of coarse mode particles is almost negligible and it can be excluded from the parameterization. Also, the effect of nucleation mode particles on the CDNC is negligible, thus those particles can also be excluded.

In Fig. 7, we show how the number of particles in mode 2 affects the CDNC and modifies the HNO_3 effect when the aerosol distribution is the continental case from Table 2. In this set of model runs, updraft velocity is 0.5 m/s and temperature is 273.15 K. It turns out that when the number of particles in mode 2 increases, the mode 1 can also be excluded from the parameterization if the updraft velocity is not high enough to activate those particles also. This can be done in situations in which the modes do not overlap. In the case presented in Fig. 7, this can be done only when N_2 is over 1000 cm^{-3}. At lower concentrations, some of the aerosol particles in mode 1 also activate to cloud droplets. In cases, where the modes are clearly distinct and it is known that particles from mode 1 can not activate, it is better to exclude the smaller mode from the parameterization. This is because

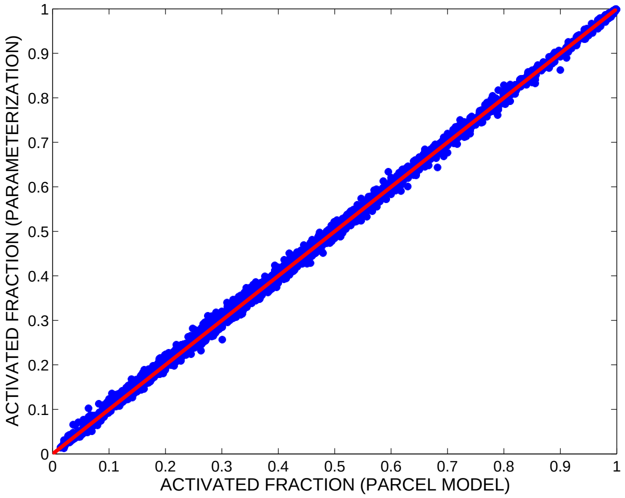

Fig. 8. Activated fraction as predicted by new parameterization and by cloud parcel model.

in the parameterization the activated fraction is defined as the CDNC divided by the total number of aerosol particles represented in the parcel model. Appearance of small particles changes F_0 values and affects therefore the F_x values. It is possible, that the parameterization gives slightly different values for the number of cloud droplets if N is increased by increasing the number of small droplets. However, the deviations due to different F_0 values are of the same order as the deviations shown in the previous figures. Overall, the parameterization is slightly more accurate if a mode with small particles is excluded, but inclusion of small particles does by no means render it useless.

In Figs. 2 to 7, we show examples of the correspondence between the parameterization and cloud parcel model results. The Figures show excellent agreement between the numerical parcel model simulations and the parameterization. In Fig. 8, we compare the prediction of the parameterization with the fraction of aerosol particles that activate with 0.05–3 ppb of HNO_3 in the air. The comparison consists of more than 4000 box model simulations in a wide variety of conditions. In the simulations, the temperature, total pressure and updraft velocity range from 253 K to 293 K, from 500 mbar to 1000 mbar, and from 0.05 m/s to 3 m/s, respectively. The aerosol distribution is lognormal and unimodal or bimodal, total number densities varying between 50 cm^{-3} and 6000 cm^{-3}, geometric mean radius varying between 5 nm and 150 nm (250 nm in the coarse mode), and geometric standard deviation varying between 1.3 and 2.5. In all model runs we have equilibrated both water and nitric acid in the beginning of model run. In doing so we have supposed the system to be closed. The initial RH is chosen to be 95% in all model runs. From Fig. 8 we can see that all data points are very close to the one-to-one line, which corresponds to a perfect match. The highest F to F_0 ratios in the simulations are close to 6 and the median is 1.2.

In order to use our new parameterization, the activated fraction without HNO_3 must be calculated with some parameterization developed for the purpose. We have also tested, that the calculation of F_0 with some other method than the one we have used in deriving the parameterization, will not produce significant errors. It turns out that underestimation of F_0 always increases the relative effect $((F-F_0)/F_0)$ but decreases the absolute effect $(F-F_0)$. However, the error produced by the new parameterization to the relative HNO_3 effect is smaller than the relative error due to the inaccurate F_0 value. The new parameterization can also be used together with simple parameterizations like the one presented by Gultepe and Isaac (1999). To do that, the typical aerosol size distribution and updraft velocity to produce the correlation between CDNC and aerosol number concentration has to be derived.

The new parameterization can also be used to calculate the effect of hydrogen chloride (HCl) on cloud drop number concentrations simply by replacing HNO_3 concentration by HCl concentration in the formulae. According to Kokkola et al. (2003) the effect of HCl on cloud droplet formation is very similar to the effect of HNO_3 and, in practice, the concentrations of HCl and HNO_3 can be summed up to yield the total effect.

4 Conclusions

A new parameterization for calculation of the HNO_3 effect on cloud drop activation has been developed. The parameterization is based on an extensive set of adiabatic cloud parcel model simulations and assumes a unimodal or a bimodal lognormal aerosol size distribution. Predictions of the parameterization have been compared to results obtained by a cloud parcel model for a wide variety of conditions and the comparison shows very good consistency. The parameterization does not include any differential equations or iterations, so it is computationally efficient and easy to use.

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