

**Uptake of HNO₃ on
UT aerosols**

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Growth of upper tropospheric aerosols due to uptake of HNO₃

S. Romakkaniemi¹, H. Kokkola¹, A. Petzold², and A. Laaksonen¹

¹University of Kuopio, Department of Applied Physics, Kuopio, Finland

²Deutsches Zentrum für Luft und Raumfahrt Oberpfaffenhofen, Institut für Physik der Atmosphäre, Wessling, Germany

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Correspondence to: S. Romakkaniemi (sami.romakkaniemi@uku.fi)

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Abstract

The effect of nitric acid on the equilibrium size distributions of upper tropospheric aerosols is calculated as a function of relative humidity. It is shown that HNO_3 concentrations above a few tenths of a ppb can cause substantial increases in haze mode particle concentrations at relative humidities at about 60% and above. The effect can be strongly magnified when letovicite particles are present in addition to sulfuric acid aerosols. This is mainly due to the lowering of the deliquescence RH of letovicite in the presence of gaseous nitric acid at low temperatures. We have also compared equilibrium calculations of the HNO_3 effect with observations of increased haze mode concentrations at relative humidities above 50% (Petzold et al., 2000). Nitric acid mixing ratios on the order of 0.5-2 ppb may explain the observed increase of haze mode particles at least partially.

1. Introduction

Aerosols in the upper troposphere and stratosphere are mainly composed of H_2SO_4 and H_2O but also ammoniated sulfate and other components have been observed (Xu et al., 2001; Sheridan et al., 1994; Murphy et al., 1998). It has been shown that binary $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ aerosols can effectively take up HNO_3 and turn into supercooled ternary solutions before freezing homogeneously (Tabazadeh et al., 1994; Kärcher and Solomon, 1999). Uptake of HNO_3 increases the hygroscopic mass of aerosols, causing further growth of the droplets due to absorption of water. This affects both the optical properties of the aerosols and the stratospheric heterophase chemistry.

Recently, Petzold et al. (2000) made observations about the occurrence of haze-mode aerosols in the upper troposphere. The number of haze mode particles increased abruptly as the saturation ratio exceeded 0.5. It was argued that the particles were likely to be composed of ammoniated sulfates undergoing deliquescence transition forming liquid droplets (Petzold et al., 2000). There are also some other observations

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on large spherical particles in the upper troposphere (Sassen et al., 1998; Del Guasta et al., 1998). The sphericity of the particles indicates strongly that they are in fact liquid droplets.

Recent thermodynamic equilibrium model calculations have shown that HNO₃ can lower the deliquescence relative humidity (DRH) of ammoniated salts at upper tropospheric conditions (Lin and Tabazadeh, 2002). It was also shown that the most likely ammoniated salt to exist at temperatures near 200 K is letovicite (NH₄)₃H(SO₄)₂. The DRH of letovicite is normally 87% at 210 K, but the presence of 2 ppb HNO₃ can decrease it down to 43%. The highest observed volume mixing ratios (VMR's) of HNO₃ in the upper troposphere are as high as 3 ppb (Laaksonen et al., 1997). High nitric acid VMR's are possible in air masses with continental influence or near flight corridors. Also, sedimentation of nitric acid particles from the stratosphere can cause increased HNO₃ concentrations in the UT. In the future, increased air traffic and/or increased NO_x emissions on the ground may cause high upper tropospheric nitric acid concentrations to occur more frequently, which in turn, may influence the number concentrations of haze mode particles.

In this study, equilibrium equations are used to model nitric acid and water partitioning between gas phase and different aerosol size classes in order to study the influence of HNO₃ on upper tropospheric particle growth at different relative humidities. Assuming that sulfuric acid and ammoniated sulfates can be considered completely involatile, the equilibrium requirements are that the saturated vapor pressures of water and nitric acid above the droplet surfaces have to equal the respective partial pressures in the gas. We also calculate the HNO₃-dependent DRH of letovicite. As a case study, equilibrium calculations are compared with the observations of Petzold et al. (2000).

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2. Theory

The equilibrium saturation ratio S_i of vapor i over a multicomponent droplet surface is obtained from

$$S_i = \frac{p_i}{p_{si}} = f_i X_i \exp\left(\frac{2\sigma v_i}{RT r}\right), \quad (1)$$

5 where p_i is the partial pressure, p_{si} is the saturation vapor pressure, X_i the mole fraction, f_i is the activity coefficient, v_i is the partial molar volume of species i , σ is the surface tension of the solution, R is the gas constant, T is the temperature, and r is the droplet radius.

To describe the equilibrium growth of aerosols we need to write separate equilibrium (Köhler) equations for every condensing vapor. In this study we have equations for water and HNO₃. At normal tropospheric conditions, Eq. (1) can be used for water as is, but the amount of HNO₃ in the air is small compared to water and we need to take into account the fact that condensation depletes the vapor phase. Using the ideal gas law ($pV = nRT$) we can write an equilibrium equation that takes account of the mass balance of species i as

$$S_i = \frac{(n_{ti} - n_{di})CRT}{p_{si}} = X_i f_i \exp\left(\frac{2\sigma v_i}{RT r}\right), \quad (2)$$

where n_{ti} is the total amount of moles of substance i in the system per one droplet, n_{di} is the amount of moles of i in a droplet, and C is the concentration of droplets.

The droplet radius r and the number of moles n_{di} of condensable substances i (in our case water and nitric acid) can be calculated from Eqs. (2) for a monodisperse droplet population with an iterative method at a given temperature if the total concentrations of H₂O and HNO₃ are known. For the present calculations, the theory is expanded for a discrete particle size distribution including N size classes to obtain sufficient accuracy (see Kokkola et al., 2003, for more details). Equations (2) are thus used to calculate the equilibrium size and the composition of aerosols in each size class as

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a function of S_i in a closed system. In practice, this can be done with the following method. Equation (2) is modified so that n_{di} and C are replaced with $n_{di,k}$ and C_k where k denotes the number of size class. The number of moles per droplet n_{ti} in size class b under iteration can be expressed as

$$n_{ti,b} = \frac{n_{tot,i} - \sum_{k \neq b} n_{di,k} C_k}{C_b} \quad (3)$$

where $n_{tot,i}$ is the total number of moles of substance i in the system (not per one droplet). By substituting Eq. (3) into Eq. (2) we get a new equation for HNO₃ in size class b

$$\frac{(n_{tot,n} - \sum_k n_{dn,k} C_k) RT}{\rho_{sn}} = X_n f_n \exp\left(\frac{2\sigma v_n}{RT r_b}\right) \quad (4)$$

Now we have two equilibrium equations for every size class, Eq. (1) for water and Eq. (4) for HNO₃. These equations are iterated one size class at time until all size classes are in equilibrium with both condensing vapors. This method is similar to the mass flux iteration method (see e.g. Jacobson, 1999).

During the iteration we also calculate the saturation ratio of liquid phase letovicite. If the saturation ratio is over unity, letovicite is in the solid phase. As soon as the saturation ratio drops below unity, deliquescence occurs and letovicite is moved from the solid phase to the liquid phase.

In the present study, the vapor pressures of H₂O and HNO₃ over the solution droplets and the saturation ratio of liquid phase letovicite are calculated using the thermodynamic electrolyte model of Clegg et al. (1998). Data for surface tensions and densities of multicomponent solutions composed of water, nitric acid, sulfuric acid and ammonia are not available, and some approximations are needed. The system is therefore simplified by excluding the effect of ammonia on surface tension and density of the solution, whereby those quantities can be calculated using the expression given by Martin et al. (2000), valid for H₂SO₄/HNO₃/H₂O-solutions.

3. Growth factors

In the following calculations concerning upper troposphere we assume (unless otherwise stated) that the total pressure $p = 200$ mbar and the initial temperature and water vapor saturation ratio are $T_0 = 206$ K and $S_0 = 0.3$. In conformity with [Petzold et al. \(2000\)](#), we define the haze mode to consist of droplets larger than $0.72 \mu\text{m}$ in diameter. Both H_2SO_4 and NH_3 are assumed to remain in the droplets during the equilibrium growth in accordance with the original distribution. The size distributions are presented in Table 1. Distribution A is similar to that reported in the article of [Petzold et al. \(2000\)](#). The dry radius is obtained by dividing values given at $S = 0.38$ by a factor of 1.4, which is estimated to be the growth factor of $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ -solution droplets at that saturation ratio.

With these initial values, homogeneous freezing would take place at $S \gtrsim 0.78$ ($T \sim 199$ K) depending on droplet size and timescale of the process ([Koop et al., 2000](#)).

3.1. $\text{H}_2\text{SO}_4/\text{HNO}_3/\text{H}_2\text{O}$

In Fig. 1a we present growth curves for distribution A at 3 different HNO_3 VMR's as a function of saturation ratio. The growth due to HNO_3 uptake is clear.

Figure 1b presents growth factors for the same distribution and reveals that the equilibrium growth factors are not size-independent. Because the Kelvin effect tends to increase the saturated vapor pressures, the concentrations of H_2O and HNO_3 have to decrease with decreasing droplet diameter in order to ensure equilibrium for all sizes. Previously, [Carslaw et al. \(1997\)](#) have noted that the equilibrium composition of stratospheric ternary droplets are size dependent (however, they did not calculate the effect on growth factors).

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3.2. $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$

In Fig. 2, growth curves are presented for distribution B (see Table 1) containing an additional mode of letovicite. Figure 2 shows the growth of the letovicite mode as a function of S (black lines). It can be seen that with 1 ppb of HNO_3 the DRH of letovicite is at 47.8%. At that point, the particles instantly take up HNO_3 and H_2O to maintain equilibrium. The GF due to pure deliquescence is approximately 1.15. The rest of the growth is caused by simultaneous uptake of HNO_3 and H_2O . The total GF at the DRH is 1.65.

It can also be seen that the DRH is size dependent. The smallest particles persist in the solid state to higher RH than do the bigger ones. The first particles to become deliquescent are those with the highest HNO_3 content in the aqueous coating. Because of the Kelvin effect, the highest equilibrium concentration occurs on the biggest particles. Smaller particles with a lower HNO_3 concentration in the liquid layer experience deliquescence at a higher RH.

4. Effect of increased HNO_3 and letovicite concentrations on the number of haze particles

From the examination above it is clear that both increased HNO_3 , and increased letovicite concentrations lead to increased numbers of haze mode particles. This effect is further demonstrated in Figs. 3 and 4 presenting the ratio of haze mode particles to all particles as a function of HNO_3 concentration for 11 different aerosol particle distributions (Table 2) containing different amounts of sulfuric acid and letovicite. Figure 3 shows that at 60% relative humidity, nitric acid concentrations below about 0.2 ppb have little effect on the haze mode. However, above 0.2 ppb, enough of HNO_3 is absorbed by the droplets to cause a substantial increase in N_{haze} . At 75% RH, only 0.1 ppb of nitric acid is needed for a similar effect. This is due to the lowered HNO_3 vapour pressure over solution droplets and HNO_3 dependent deliquescence of letovicite. With

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increasing HNO_3 concentrations, the fraction of haze mode particles to all particles can increase to more than 5% depending on the amount and size of letovicite particles present.

Even without letovicite, the haze mode concentration can increase to more than 1 cm^{-3} , but the amount of HNO_3 needed is higher than in the presence of letovicite particles. Altogether, the variation in the haze mode concentrations for the different size distributions shown in Table 2 is close to an order of magnitude at both 60% and 75% relative humidities. Nitric acid causes a substantial enhancement of haze mode particles already when the aerosol distribution is composed of sulfuric acid alone, however, the presence of letovicite magnifies the effect greatly.

5. A case study

Petzold et al. (2000) observed pronounced increases of upper tropospheric haze mode particles at relative humidities above 50%. Here, we compare our calculations to the observation in order to examine whether nitric acid may have contributed to the haze mode concentrations. To be able to carry out the comparison, we define two modes with size limits $0.37 \mu\text{m} < D < 0.72 \mu\text{m}$ (accumulation mode) and $0.72 \mu\text{m} < D < 10 \mu\text{m}$ (haze mode). Due to the measurement setup, the modes defined by Petzold et al. (2000) were $0.37 \mu\text{m} < D < 0.69$ and $0.75 \mu\text{m} < D < 10 \mu\text{m}$, but this difference should not influence our analysis too much. Figure 5 shows the concentration ratios N_{acc}/N_{14} and N_{haze}/N_{14} , where N_{14} is the concentration of all particles with $D > 0.014 \mu\text{m}$, as a function of the saturation ratio of water vapor at different nitric acid VMR's when the aerosol is composed of sulfuric acid and water (distribution A of Table 1). Also the partitioning of nitric acid between the gas and the liquid phases is shown. The data-points in Fig. 5 are the observations of Petzold et al. (2000).

The observed increase in the number of accumulation mode particles is consistent with the modeled increase caused by uptake of nitric acid on liquid $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$. It seems that an average of less than 1 ppb of HNO_3 is needed to explain the behaviour

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of the accumulation mode particle concentration, and VMR's above 2 ppb would lead to an overestimation of the increase in N_{acc}/N_{14} . A small increase in number of the accumulation mode particles can be seen even without HNO₃ but not nearly enough to explain the observations.

5 The increase in the number of haze mode particles is more problematic. It clearly cannot be explained without the influence of HNO₃. However, even nitric acid is not able to explain all of the observations; with haze mode particles, more than 2 ppb of HNO₃ would be needed to explain a large part of the increase in the haze mode. By adjusting the initial size distribution, the model results could be made to match
10 the observations somewhat better, i.e. a larger increase in haze mode would be seen at a given nitric acid VMR. However, this would inevitably cause an increase also in the accumulation mode concentration, spoiling the agreement obtained with the size distribution A of Table 1.

[Petzold et al. \(2000\)](#) proposed, that their observations could be explained with particles undergoing deliquescence. In Fig. 6 we compare our results for distribution B (see
15 Table 1) containing letovicite to their observations. The increase in haze mode particle concentration fits the measurements better than with distribution A. This is because with proper selection of HNO₃ concentration the DRH of letovicite can be adjusted to occur at $S = 0.5$. The amount of HNO₃ needed is close to 1 ppb. After deliquescence
20 more HNO₃ is partitioned in droplets containing letovicite than in H₂SO₄/H₂O droplets. That can be seen by comparing the growth of H₂SO₄ droplets in Figs. 1 and 2.

It can be seen that our equilibrium model predicts too high accumulation mode concentration if we adjust the haze mode concentration behavior within observations. In order to make the letovicite particles to increase the haze mode enough, the letovicite
25 distribution is such that the accumulation mode of distribution B has more particles than that of distribution A already at $S = 0.3$ (see Table 1 and Figs. 2 and 4). Growth of the letovicite particles due to the deliquescence and nitric acid uptake from sizes below the accumulation mode is not strong enough to bring them directly to the haze mode as deliquescence takes place. Growth factors over 2 (up to 3) would be needed for

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particles undergoing deliquescence to explain the observations, whereas for letovicite the growth factor is only 1.65.

However, discussing the agreement between observations and modeling results should consider the random-type nature of the observations as presented by Petzold et al. (2000). The data were obtained by compiling all observations in the upper free troposphere during the POLSTAR 2 experiment. In the case of high ozone concentrations > 220 ppb and simultaneously observed low relative humidities $\leq 20\%$ data were identified as from the lower stratosphere and were excluded. In total, 40% of the data originate from drier air masses with $S \leq 0.45$, 15% originate from air masses close to ice saturation with $S \sim 0.50$, and the remaining 45% originate from humid air masses above ice saturation with $S \geq 0.55$. Hence, the air masses close to ice saturation are not equally represented compared to more dry and more humid air masses, respectively. Additionally, no Lagrangian flights with the focus on particle formation and growth were performed during POLSTAR 2. The gap between the accumulation mode and the haze mode which occurs in the model results compared to the observations, is likely to be explainable by the missing information for air masses around ice saturation. The size distributions A and B used for the modeling studies are both likely to occur compared to the observed average number density of $\sim 1 \text{ cm}^{-3}$ for particles in the size range $0.37 \mu\text{m} < D < 0.72 \mu\text{m}$. Hence, the observations do not allow for an unambiguous preference for one of the selected size distributions.

As reported by Krämer et al. (2003), the VMR of HNO_3 during the POLSTAR 2 experiment was of the order of 1 ppb. The model results for the accumulation mode are in good agreement with the HNO_3 observations. Without overrating the observations, they also support the model results which indicate a considerable growth of an upper tropospheric haze mode in the presence of sufficient HNO_3 .

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6. Conclusions

We have shown that uptake of nitric acid by upper tropospheric aerosols causes notable increases in haze mode particle concentrations at relative humidities above 50%. At a relative humidity of 75%, a few tenths of a ppb of HNO_3 is enough to have an effect when the aerosol is composed of pure sulfuric acid. The presence of letovicite can substantially enhance the nitric acid effect, decreasing the threshold to about 0.1 ppb, and causing a severalfold increase in the haze mode concentrations at a given HNO_3 concentration. Our equilibrium calculations indicate that uptake of nitric acid by sulfuric acid aerosol may at least partially explain the observed increases of upper tropospheric haze- and accumulation mode particles at relative humidities above 50%. A letovicite mode improved the agreement between calculations and observations somewhat. The required HNO_3 levels were on the order of 0.5–2 ppb.

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Table 1. Parameters for log-normal distributions used in the calculations. In dist B the mode 3 is composed of letovicite with sulfuric acid coating.

dist	property	mode 1	mode 2	mode 3
A	N [cm ⁻³]	112.0	0.1	
	D_{pg} [μm]	0.053	0.518	
	σ_g	1.70	1.35	
B	N [cm ⁻³]	112.0	0.1	1.25
	D_{pg} [μm]	0.050	0.518	0.270
	σ_g	1.70	1.35	1.30

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Table 2. Parameters for log-normal distributions used in the calculations. Modes 1 and 2 are composed of sulfuric acid and mode 3 is composed of letovicite.

dist	N [cm ⁻³]			D_{pg} [μm]			σ_g		
	1	2	3	1	2	3	1	2	3
C ₁	99	-	1	0.05	-	0.27	1.7	-	1.3
C ₂	98	-	2	0.05	-	0.27	1.7	-	1.3
C ₃	97	-	3	0.05	-	0.27	1.7	-	1.3
C ₅	95	-	5	0.05	-	0.27	1.7	-	1.3
C ₁₀	90	-	10	0.05	-	0.27	1.7	-	1.3
D ₀	100	0.1	0	0.05	0.50	0.20	1.7	1.35	1.3
D ₁	99	0.1	1	0.05	0.50	0.20	1.7	1.35	1.3
D ₂	98	0.1	2	0.05	0.50	0.20	1.7	1.35	1.3
D ₃	97	0.1	3	0.05	0.50	0.20	1.7	1.35	1.3
D ₅	95	0.1	5	0.05	0.50	0.20	1.7	1.35	1.3
D ₁₀	90	0.1	10	0.05	0.50	0.20	1.7	1.35	1.3

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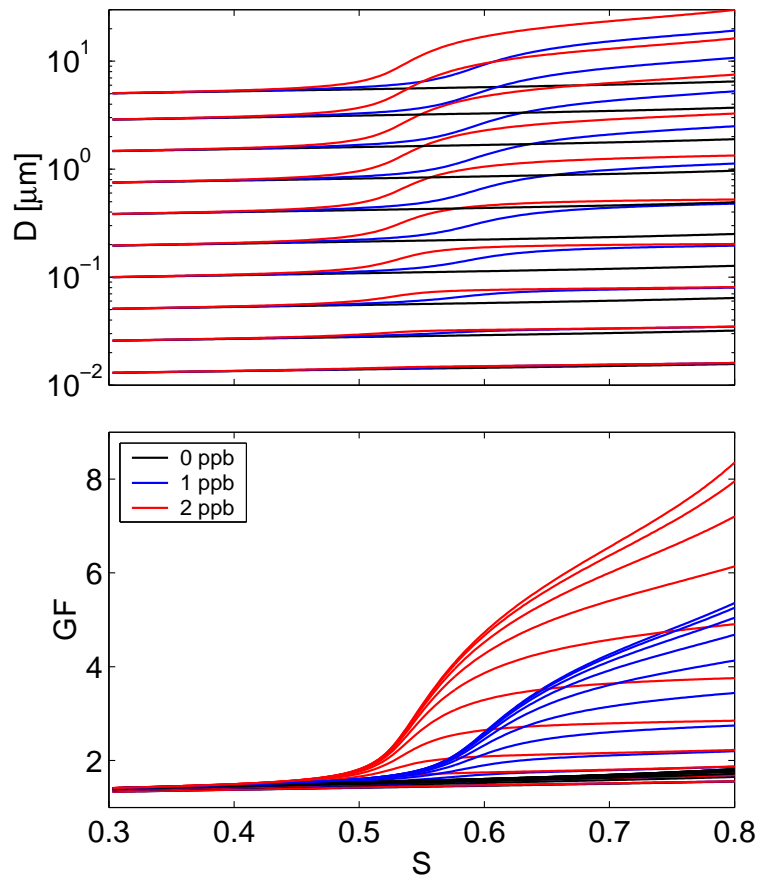


Fig. 1. (a) Growth curves for different size classes according to dist A in Table 1. (b) Growth factors for different size classes for the same distribution.

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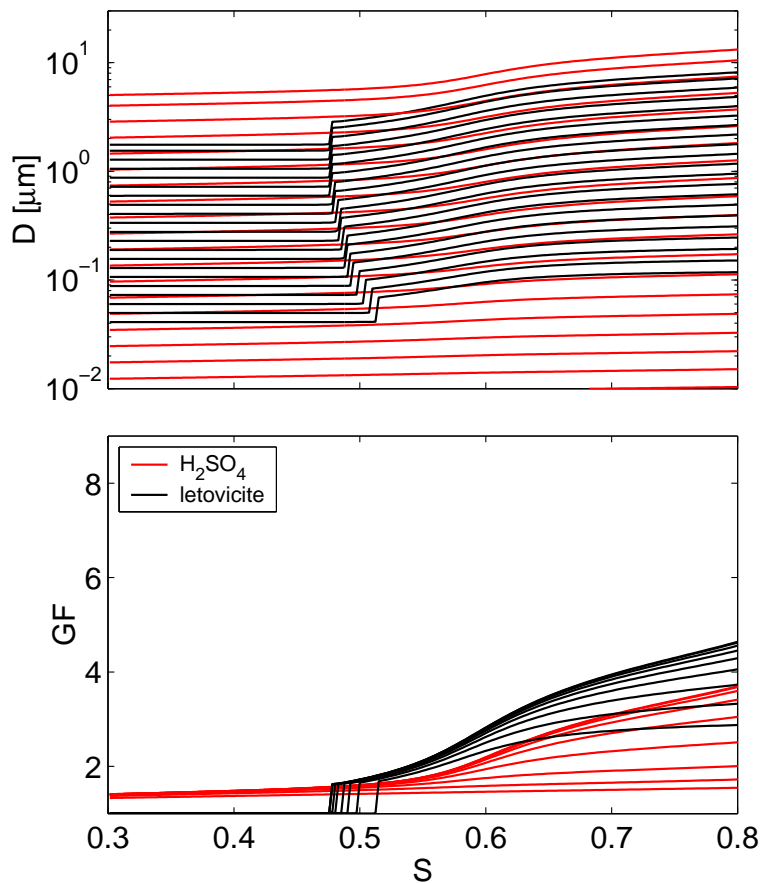


Fig. 2. (a) Growth curves and (b) growth factors for different size classes according to dist B in Table 1. HNO_3 VMR is 1 ppb.

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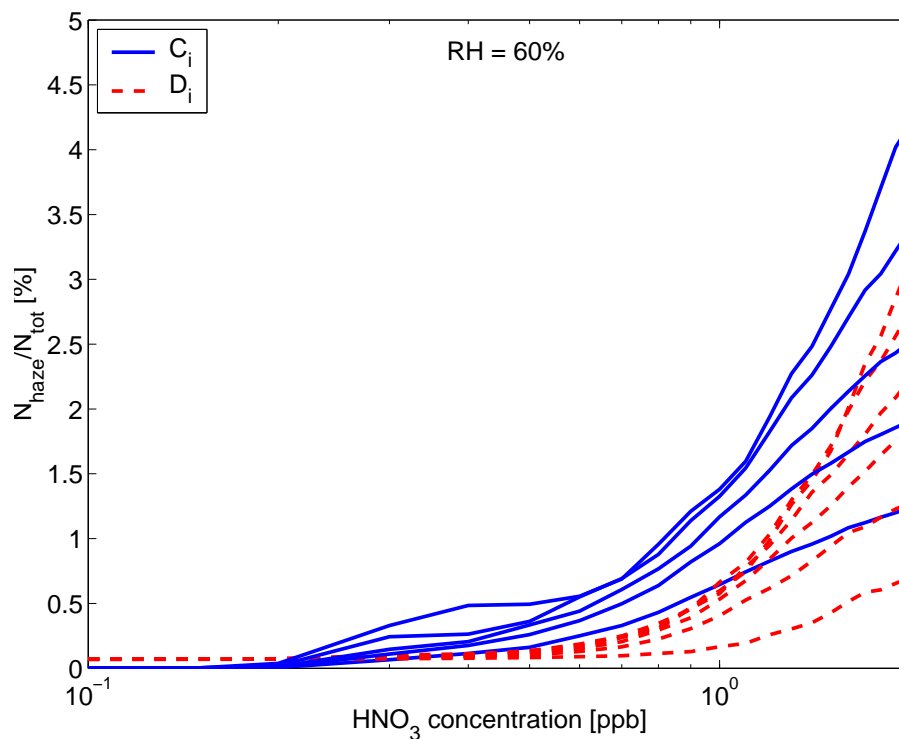


Fig. 3. Number concentration ratio $N_{\text{haze}}/N_{\text{tot}}$ as a function of HNO_3 concentration at 60% RH. The lowermost curves of C_i and D_i correspond to distributions C_1 and D_0 in Table 2 and the uppermost correspond to C_{10} and D_{10} , respectively.

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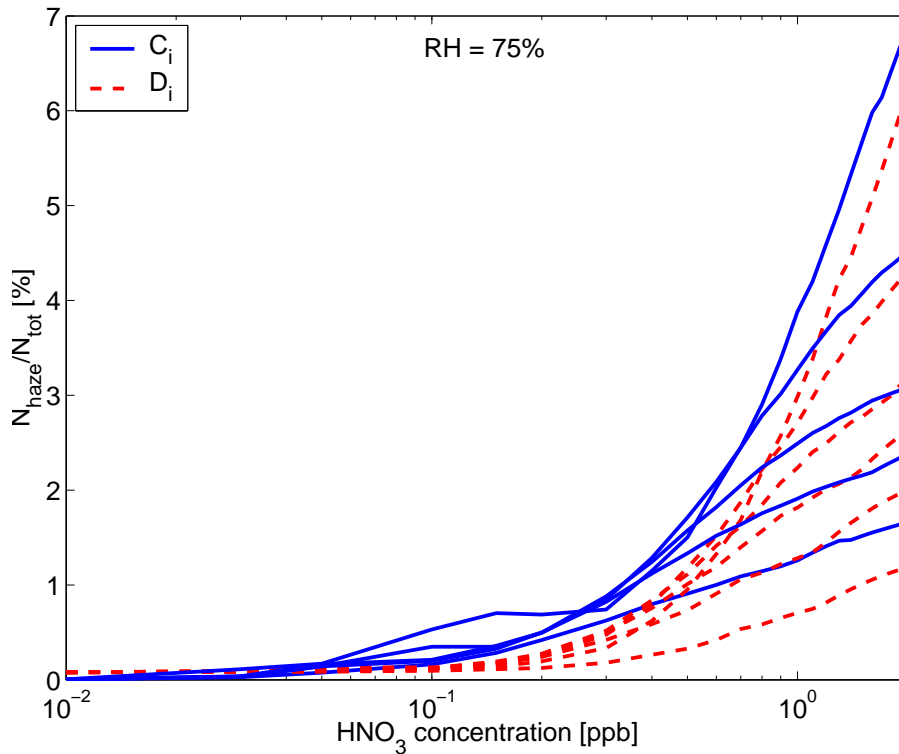


Fig. 4. Like Fig. 3 but RH is now 75%.

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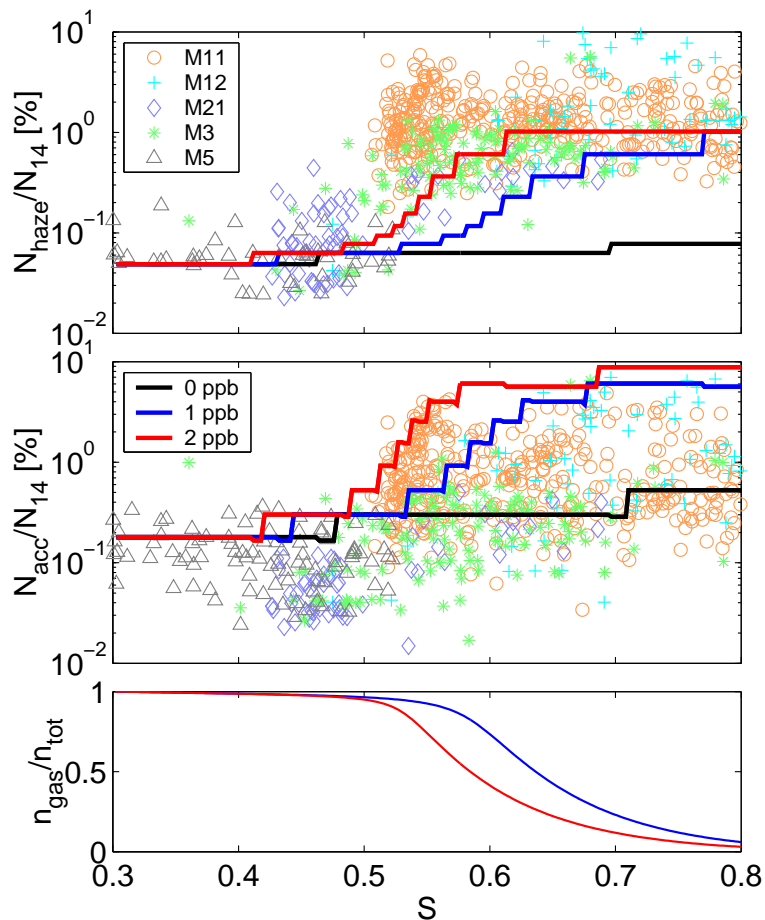


Fig. 5. Comparison to observations of [Petzold et al. \(2000\)](#) for distribution A at three different HNO_3 VMR's. See [Petzold et al. \(2000\)](#) for explanations for different symbols representing the observations.

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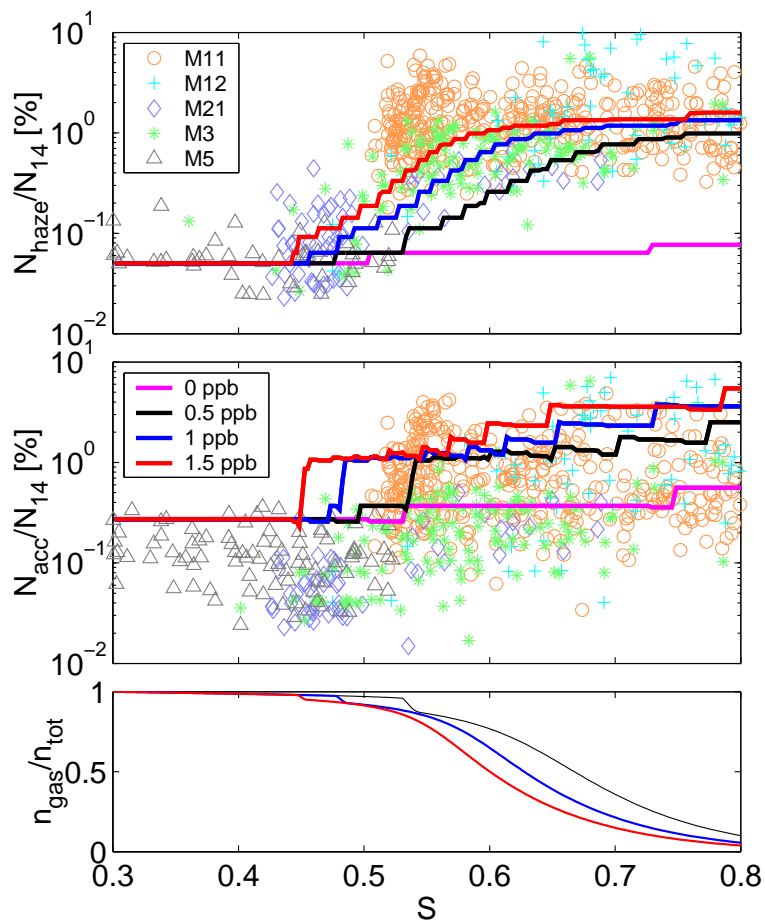


Fig. 6. Comparison to observations of [Petzold et al. \(2000\)](#) for distribution B with four different HNO_3 concentrations.

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