

# The role of ammoniated aerosols in cirrus cloud nucleation

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**Abstract.** Recent results obtained during the SUCCESS experiment show that aerosols containing ammonium ion are commonly found in the upper troposphere and therefore it is of great importance to investigate the ice freezing properties of such aerosols. For this purpose we present mathematical expressions, derived from recent model calculations and laboratory data, for calculating the deliquescence relative humidity of  $(\text{NH}_4)_2\text{SO}_4$ ,  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{NO}_3$  under upper tropospheric conditions. Using these functions we define the temperature and relative humidity requirements for the deliquescence of dry particles in the upper troposphere. Further using a thermodynamic model of the  $\text{H}_2\text{SO}_4/\text{HNO}_3/\text{NH}_3/\text{H}_2\text{O}$  system, we calculate the variation in the supercooled aerosol composition and saturation ratios of various ionic salts in solution as a function of temperature and relative humidity. Using these values we show salt crystallization in the upper troposphere may occur in two distinct type of ammoniated aerosol solutions. First  $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$  (letovicite) may crystallize in acidic solutions of  $(\text{NH}_4)_2\text{SO}_4$ . Second double salts of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$  may crystallize in nearly neutralized solutions of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ . Finally we discuss how the presence of crystalline salt surfaces in aqueous solutions may alter the mode of cirrus cloud formation from homogeneous to heterogeneous freezing nucleation.

## Introduction

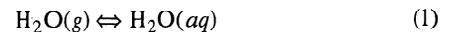
Cirrus cloud formation in the upper troposphere is often modeled by homogeneous nucleation of ice crystals from aqueous  $\text{H}_2\text{SO}_4$  solutions [Sassen and Dodd, 1989; Heymsfield and Miloshevich, 1995; Jensen *et al.*, 1994]. Recently we made significant corrections to the current theories of ice nucleation from aqueous  $\text{H}_2\text{SO}_4$  [Tabazadeh *et al.*, 1997], which can explain the available ice nucleation data obtained in a few rapid lee wave cooling events much better [Sassen and Dodd, 1989; Jensen *et al.*, this issue] than the previous formulations [Jensen *et al.*, 1994; Heymsfield and Miloshevich, 1995]. Studying cirrus formation in cold lee waves can provide useful information about the makeup of the majority of the background upper tropospheric aerosols, which are shown to be mainly composed of aqueous  $\text{H}_2\text{SO}_4$  [Sheridan *et al.*, 1994]. Hence the fact that our cirrus ice nucleation model [Tabazadeh *et al.*, 1997] predicts the measured cirrus cloud nucleation barrier in a few cold lee waves may confirm the measurements of Sheridan *et al.* that upper tropospheric aerosols are sometimes composed of aqueous  $\text{H}_2\text{SO}_4$ . However cooling rates leading to the formation of background cirrus are significantly slower than those experienced in cold lee wave events. Therefore other types of nuclei which do not contribute significantly to the nucleation of lee wave generated cirrus can in fact act as the main nuclei for the formation of background cirrus if they are more efficient in nucleating ice as compared to aqueous  $\text{H}_2\text{SO}_4$ . This is solely related to the rate of cooling since for fast cooling rates the supersaturation that builds up in the air mass can result in the nucleation of the majority of the background aerosols into ice crystals. Whereas, in slower

cooling rates the most efficient nuclei would activate first and their subsequent growth into ice would effectively prevent the nucleation of the majority of the background particles.

Results obtained during the recent SUCCESS field work [Talbot *et al.*, this issue; Coggiola *et al.*, this issue] show that ammoniated and/or nitrated aerosols are present in the upper troposphere. Thus in principle these particles can act as cirrus cloud condensation nuclei if they are more efficient in nucleating ice as compared to pure aqueous  $\text{H}_2\text{SO}_4$  droplets. In this paper we intend to address how the current model of ice formation from sulfuric acid solutions can be influenced by the presence of ammonium and/or nitrate ions in solution.

## Deliquescence Relative Humidity

The vapor-liquid equilibrium between an aqueous salt solution and the water vapor over the solution can be expressed by the following reactions:



where  $g$  refers to the gas phase,  $aq$  refers to the aqueous solution and  $n$  is the moles of salt per mole of water. Using thermodynamic relations associated with the above reactions, Tang and Munkelwitz [1993] have shown that the variation of the deliquescence relative humidity (DRH) of a binary salt as a function of temperature can be calculated from the following relation

$$\ln DRH(T) = \ln DRH(T^*) + \frac{1}{R} \int_{T^*}^T \frac{n(T)\Delta H_s(T)}{T^2} dT \quad (3)$$

where  $\Delta H_s$  is the integral heat of solution,  $DRH(T^*)$  is the measured room temperature value ( $T^* = 298.15$  K) of the deliquescence relative humidity, and  $R$  is the gas constant.

In previous parameterizations of the DRH, the laboratory data is fitted into relation (3) by assuming a constant room temperature value for  $\Delta H_s$  and by using a polynomial temperature fit to the binary salt solubility data for  $n$  [Stelson and Seinfeld, 1982; Wexler and Seinfeld, 1991; Tang and Munkelwitz, 1993]. Also these parameterizations use only laboratory data at higher temperatures. Here we use a different procedure for calculating the temperature variations of the DRH (Table 1). We derive a relation for the temperature dependence of the DRH either directly from recent model calculations by Clegg *et al.* [1997] or by fitting new laboratory data obtained at colder temperatures [Imre *et al.*, 1997]. The advantage of using the results from the model of Clegg *et al.* [1997] is that this model explicitly takes into account the temperature variations of both  $\Delta H_s$  and  $n$ . These temperature dependences are especially important for the  $\text{NH}_4\text{NO}_3$  system since  $\text{NH}_4\text{NO}_3$  exists in several solid phases over the temperature range of interest [Nagatani *et al.*, 1967] and the model of Clegg *et al.* [1997] takes into account the enthalpy changes associated with these solid-solid phase transformations.

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**Table 1.** Deliquescence Relative Humidity Coefficients
$$\ln DRH(\%) = a + \frac{b}{T} + \frac{c}{T^2} + \frac{d}{T^3}$$

Species	a	b	c	d	T-range
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	4.9058e+0	-6.1731e+2	2.0627e+5	-2.0521e+7	254.2<T<320
NH <sub>4</sub> NO <sub>3</sub>	-2.0973e+1	1.8477e+4	-4.5798e+6	3.8801e+8	256.8<T<320
NH <sub>4</sub> HSO <sub>4</sub>	-8.3066e+1	8.1309e+3	-1.8248e+6	1.3686e+8	243.2<T<320
NH <sub>4</sub> HSO <sub>4</sub> *	-4.2179e+1	3.5936e+4	-9.4631e+6	8.2420e+8	243.2<T<300
NH <sub>4</sub> HSO <sub>4</sub> ·8H <sub>2</sub> O*	2.31.82e+2	1.1158e+5	1.3670e+7	---	239.7<T<243.2

Read 4.9058e+0as 4.9058 x 10<sup>0</sup>. The temperature range indicates the limits where the polynomial fit is valid. The lower temperature limit is the eutectic temperature of ice and the corresponding binary salt. Unless indicated the polynomial fits are generated using the model of Clegg *et al.* [1997].

\* These values are fitted to the data of Imre *et al.* [1997].

## H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub>/NH<sub>3</sub>/H<sub>2</sub>O Thermodynamic System

To simulate the thermodynamic properties of the upper tropospheric aerosols would require a complex mixed-electrolyte model which takes into account the various ionic interactions in the aqueous solution. Such a model can be used to predict the equilibrium partitioning of various species between the gas and the aqueous phases. In order to develop a predictive aerosol model for the upper troposphere laboratory data on the hygroscopic properties of the many ammoniated salts that may form in the above system at low temperatures are needed. Unfortunately laboratory data on the ammoniated salts at low temperatures are currently lacking, except for the binary system of NH<sub>4</sub>HSO<sub>4</sub>/H<sub>2</sub>O [Imre *et al.*, 1997a]. For this reason we have used recent work by Clegg *et al.* [1997] to extrapolate the thermodynamic properties of the above system to low temperatures. For the calculations presented here we assumed that the ambient relative humidity is not affected by the water taken up in the aqueous particles.

## Aerosol Processes

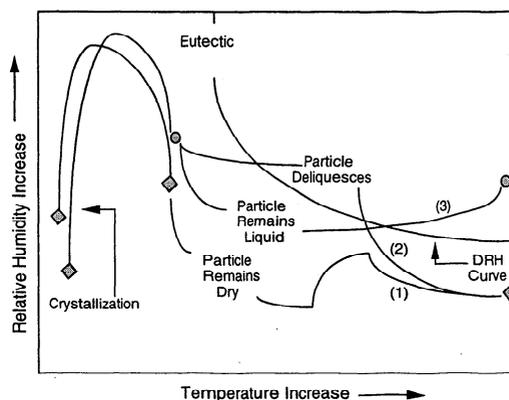
Three general particle transport pathways to the upper troposphere are shown in Figure 1. Basically ammoniated aerosols will start their journey at the surface either as solid or liquid particles. Depending on the transport conditions, a dry particle will remain solid if the particle path remains below the DRH before the eutectic temperature is reached (path 1). After the ambient temperature is cooled below the eutectic point, then further changes in the relative humidity, even if the particle path lies above the DRH curve, will not cause the particle to deliquesce. This occurs since below the eutectic point the solid phase is the thermodynamically stable phase and the only path of obtaining supercooled solution droplets below this point is to start them as liquid droplets at higher temperatures and then cool them below the eutectic point. There are two possible pathways of obtaining supercooled solution droplets in the upper troposphere. First, if the particle is originally dried up at the surface, then it must experience temperature and relative humidity fields which lie above the DRH curve before the eutectic temperature is reached (path 2). Second, a liquid particle will become a supercooled droplet once the particle path crosses the DRH curve (path 3). After transport to the upper troposphere, salt crystallization in solution may completely or partially dry up the liquid particles. This process requires a nucleation barrier, which is usually expressed as the efflorescence relative humidity for the case of a binary solution droplet [Tang and Munkelwitz, 1994; Imre *et al.*, 1997a].

## Deliquescence

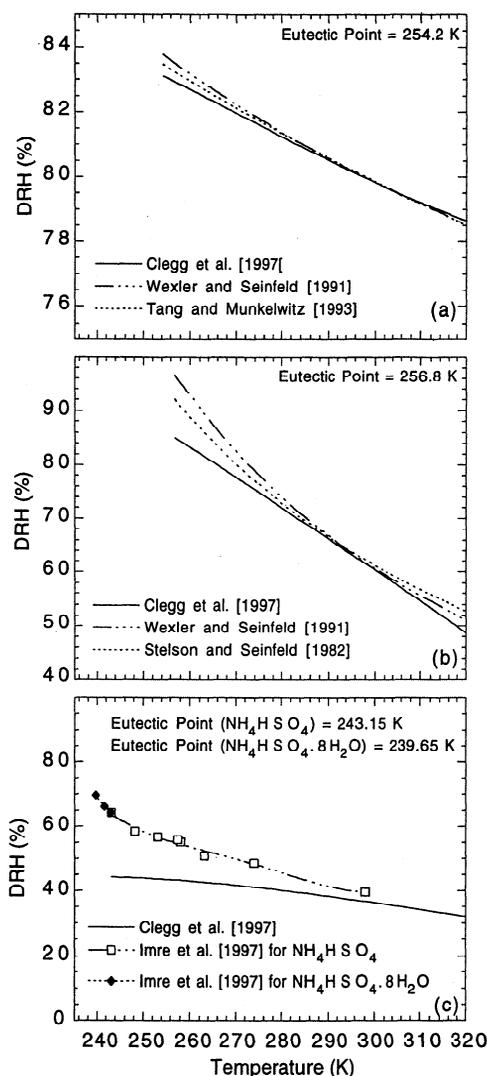
Figure 2 shows the variation of the DRH with temperature for (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, NH<sub>4</sub>NO<sub>3</sub> and NH<sub>4</sub>HSO<sub>4</sub>. In panels (a) and (b) the agreement between the model of Clegg *et al.* [1997] and the previous parameterizations is excellent near and around room temperature values. However, there are noticeable differences at colder (or higher for the case of NH<sub>4</sub>NO<sub>3</sub>) temperatures which are mainly due to the fact that the previous parameterizations used a constant value for the heat of solution. Thus we suggest

using the relations given in Table 1 to compute the DRH for both (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> at colder temperatures. Also from the above discussion if dry (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> particles are cooled in such a way that their path lies below the DRH curve at warmer temperatures, then they will remain dry below their corresponding eutectic temperatures of 254.2 and 256.8 K, respectively. However, it is important to point out that both (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> may become hydrated as the relative humidity increases below the eutectic temperature of the anhydrous salts [Imre *et al.*, 1997a, b]. If this occurs, then the corresponding hydrated forms will have a different DRH curve. Thus the observed deliquescence temperature of these binary systems in the atmosphere may occur at a few degrees below the eutectic temperatures given above for the anhydrous salts if they indeed form hydrated phases (see the discussion below with regard to the NH<sub>4</sub>HSO<sub>4</sub> system).

For the NH<sub>4</sub>HSO<sub>4</sub> system (Figure 2, panel (c)) there is a significant difference between the results obtained from the model of Clegg *et al.* [1997] and the recent laboratory data of Imre *et al.* [1997a]. In Table 1 we present parameterizations for both cases. The cause of this difference is not known at this point and future reinterpretation of either the laboratory data or model parameters may bring the model calculations and the laboratory data into better agreement (for more information on the possible sources of this difference see section 3.7 of Clegg *et al.*). Table 1 also lists parameters for the DRH of NH<sub>4</sub>HSO<sub>4</sub>·8H<sub>2</sub>O based on laboratory data of Imre *et al.* [1997a]. They have observed that near the eutectic temperature of NH<sub>4</sub>HSO<sub>4</sub>, a dry NH<sub>4</sub>HSO<sub>4</sub> particle will absorb exactly 8 water molecules to form a new solid phase of NH<sub>4</sub>HSO<sub>4</sub>·8H<sub>2</sub>O. The eutectic temperature for this phase is a few degrees lower than that of NH<sub>4</sub>HSO<sub>4</sub> (see Table 1). Thus for the NH<sub>4</sub>HSO<sub>4</sub>/H<sub>2</sub>O



**Figure 1.** Particle transport pathways to the upper troposphere. Three general temperature-relative humidity pathways are shown in the plot. Dry and liquid particles are designated as diamonds and circles, respectively. Above the DRH curve the liquid particles are stable and below this curve liquid particles become supercooled. Solid particles can only exist below the DRH curve for temperatures above the eutectic point (see text for more detail).



**Figure 2.** Variation in the DRH as a function of temperature for (a)  $(\text{NH}_4)_2\text{SO}_4$  (b)  $\text{NH}_4\text{NO}_3$ , and (c)  $\text{NH}_4\text{HSO}_4$  calculated from the model of Clegg *et al.* [1997] and the laboratory data of Imre *et al.* [1997]. A comparison between our parameterization and previous works is also shown. In panel (c) the symbols show the laboratory data points of Imre *et al.* [1997].

system a dry particle will remain dry irrespective of changes in the relative humidity as long as the temperature remains below 239.7 K (the eutectic temperature between the  $\text{NH}_4\text{HSO}_4 \cdot 8\text{H}_2\text{O}$  and ice).

It is important to note that for a dry particle, which is composed of a mixture of ammoniated salts, the DRH curve is lower than that of any binary salts that are present in the mixture [Tang and Munkelwitz, 1993; Wexler and Seinfeld, 1991]. For example, if the dry aerosol is composed of a mixture of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ , then the DRH of this mixture is lower than the curves shown for either of the binary components in Figure 2. Thus during transport to the upper troposphere dry particles which contain a mixture of different salts are more likely to deliquesce than are pure salt particles.

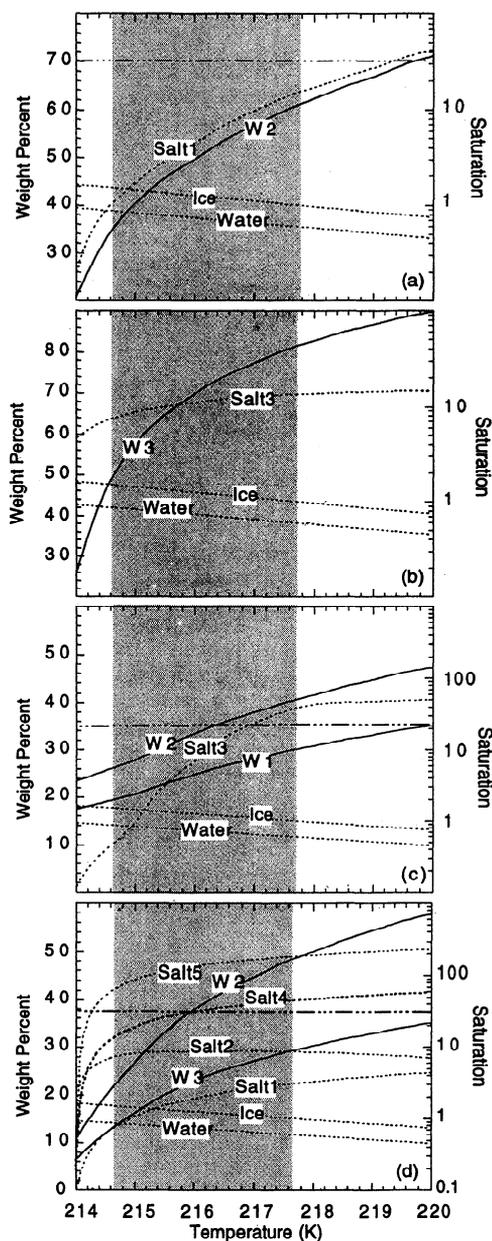
### Crystallization

For the cases where the particle starts as a solution droplet at the surface (path 3 in Figure 1) or deliquesces in its pathway to the upper troposphere (path 2 in Figure 1), we will use a thermodynamic model [Clegg *et al.*, 1997] to investigate the possibility of salt crystallization in the supercooled solutions. Panels (a) and (b) of Figure 3 illustrate the variation in the aqueous aerosol composition as a function of temperature for pure solutions of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ . For

the case of  $(\text{NH}_4)_2\text{SO}_4$ , the horizontal-dashed dotted line shows the salt saturation requirements for nucleating  $(\text{NH}_4)_2\text{SO}_4$  in an aqueous solution droplet at room temperature. This value was obtained by converting the measured efflorescence relative humidity ( $\sim 39\%$ ) of  $(\text{NH}_4)_2\text{SO}_4$  at room temperature [Tang and Munkelwitz, 1994] into a salt saturation ratio using the model of Clegg *et al.* [1997]. The temperature range of interest, for the water vapor mixing ratio chosen for these calculations, is shaded gray in the plot. The higher boundary indicates the point where ice reaches saturation in the atmosphere and the lower boundary marks the temperature at which ice homogeneously nucleates from a  $\text{H}_2\text{SO}_4$  droplet [Tabazadeh *et al.*, 1997]. Panel (a) in Figure 3 shows that  $(\text{NH}_4)_2\text{SO}_4$  is saturated in solution in the gray region, however, the saturation levels stay below the horizontal-dashed dotted line. Thus salt crystallization may or may not occur in solution since the nucleation requirements are probably milder at lower temperatures as compared to room temperature values. Panel (b) of Figure 3 illustrates the same calculations for  $\text{NH}_4\text{NO}_3$ . For this case information is currently lacking on salt saturation requirements even at room temperature. Thus we cannot comment on whether  $\text{NH}_4\text{NO}_3$  saturation in solution, as shown in panel (b), is high enough to allow for crystallization in solution. However laboratory studies usually indicate that  $\text{NH}_4\text{NO}_3$  solution droplets are very hygroscopic and they tend to remain as solution droplets even under very low relative humidity conditions (Dan Imre, private communication). Thus it is very likely that liquid  $\text{NH}_4\text{NO}_3$  particles remain as supercooled droplets in the upper troposphere.

Panel (c) in Figure 3 illustrates the variation in the aerosol composition for a particle which contains an equal mole ratio of  $\text{H}_2\text{SO}_4$  to  $(\text{NH}_4)_2\text{SO}_4$ . The horizontal-dashed dotted line shows the salt saturation requirement for the crystallization of  $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$  (letovicite) in solution at room temperature. We used the same procedure to calculate this value from the room temperature data [Tang and Munkelwitz, 1994], as described above for the  $(\text{NH}_4)_2\text{SO}_4$  system. In the gray area letovicite is highly saturated in solution. Considering that saturation requirements are in general milder at lower temperatures, it is very likely that letovicite will crystallize in this solution. We performed two other sets of calculations by using different mole ratios of  $\text{H}_2\text{SO}_4$  to  $(\text{NH}_4)_2\text{SO}_4$ , mainly (2:1 and 1:2), and obtained similar results to that of 1:1 mole ratio. From these calculations we conclude that letovicite may crystallize in upper tropospheric aerosols, which contain acidic solutions of  $(\text{NH}_4)_2\text{SO}_4$ . Recent studies indicate that the presence of a solid heterogeneous core, such as aircraft generated soot, in an aqueous  $\text{H}_2\text{SO}_4$  particle will result in the lowering of the ice nucleation barrier in solution through a heterogeneous nucleation step [Jensen and Toon, 1997]. Here we suggest that the presence of letovicite cores in solution can similarly change the mode of ice nucleation from homogeneous to heterogeneous freezing and therefore aerosols containing acidic solutions of  $(\text{NH}_4)_2\text{SO}_4$  may be more efficient in nucleating cirrus than pure  $\text{H}_2\text{SO}_4$  solution droplets.

We also performed similar calculations for aerosol solutions, which contain  $\text{H}_2\text{SO}_4$  to  $\text{NH}_4\text{NO}_3$  mole ratios of 1:1, 2:1 and 1:2. For all cases studied, nitrate in solution was driven off from the aqueous particle as  $\text{HNO}_3$ . Basically our calculations show that nitrate will not stay in acidic droplets under relative humidity and temperature conditions found in the upper troposphere. If the initial mole ratio is higher than 1:2, let's assume 1:3, then 2 moles of  $\text{HNO}_3$  will be lost to the gas phase and the remaining aerosol will be a nearly neutral solution of 1:1 mole ratio of  $(\text{NH}_4)_2\text{SO}_4$ : $\text{NH}_4\text{NO}_3$ . Panel (d) in Figure 3 illustrates the variation in the aerosol composition and saturation ratios for a mixture containing a 1:1 mole ratio of  $(\text{NH}_4)_2\text{SO}_4$ : $\text{NH}_4\text{NO}_3$ . In the gray area both  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$  are slightly saturated in the solution and from the above discussions it is unlikely that either of these salts will crystallize in solution. However double salts of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ , mainly  $2\text{NH}_4\text{NO}_3 \cdot (\text{NH}_4)_2\text{SO}_4$  and  $3\text{NH}_4\text{NO}_3 \cdot (\text{NH}_4)_2\text{SO}_4$ , become highly saturated and perhaps may crystallize in the aqueous solution. From these calculations, we suggest that double salts of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$  may crystallize in aerosols, which contain nearly neutralized solutions of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ . As discussed above these salt surfaces



**Figure 3.** The variation in the aerosol composition as a function of temperature. Also shown is the variation in the saturation ratios of ice, water,  $(\text{NH}_4)_2\text{SO}_4$  (Salt1),  $\text{NH}_4\text{NO}_3$  (Salt2),  $(\text{NH}_4)_2\text{H}(\text{SO}_4)_2$  (Salt3),  $2\text{NH}_4\text{NO}_3 \cdot (\text{NH}_4)_2\text{SO}_4$  (Salt4) and  $3\text{NH}_4\text{NO}_3 \cdot (\text{NH}_4)_2\text{SO}_4$  (Salt5). For these simulations the water vapor pressure was fixed at 0.02 mbar.  $W_1$ ,  $W_2$  and  $W_3$  correspond to the weight percentages of the  $\text{H}_2\text{SO}_4$ ,  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$  in solution, respectively. (a) pure  $(\text{NH}_4)_2\text{SO}_4$  (b) pure  $\text{NH}_4\text{NO}_3$  (c) 1:1 mole ratio of  $\text{H}_2\text{SO}_4:(\text{NH}_4)_2\text{SO}_4$  (d) 1:1 mole ratio of  $(\text{NH}_4)_2\text{SO}_4:\text{NH}_4\text{NO}_3$ . The horizontal-dashed dotted lines in panels (a) and (d) show the saturation ratio of  $(\text{NH}_4)_2\text{SO}_4$  at room temperature. The horizontal-dashed dotted line in panel (c) shows the saturation ratio of letovicite in solution at room temperature (see text for more detail).

can change the mode of ice nucleation from homogeneous to heterogeneous freezing and therefore may be better nuclei for cirrus cloud formation as compared to  $\text{H}_2\text{SO}_4$  solution droplets.

## Summary

In this paper we described for the first time how the presence of ammonium and nitrate ions in a sulfate-based aerosol

solution may influence the occurrence of cirrus clouds in the upper troposphere. Based on recent model calculations and laboratory data, we presented functions for calculating the deliquescence relative humidity for a number of different ammoniated particles in the upper troposphere. Basically we have shown particle history during transport to the upper troposphere will govern whether dry ammoniated particles will deliquesce or not. We also showed that if ammoniated droplets are composed of acidic solutions of  $(\text{NH}_4)_2\text{SO}_4$  or nearly neutral solutions of  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ , then salt crystallization may occur in these droplets. Formation of salt surfaces in aqueous particles can alter the mode of cirrus cloud formation from homogeneous to heterogeneous freezing nucleation. This can result in significant changes in the microphysical properties of the background cirrus and in their subsequent radiative impacts in places where ammoniated aerosols are more abundant.

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