

New particle formation in the remote troposphere: A comparison of observations at various sites

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Abstract. Measurements show that new particles are formed by homogenous nucleation over a wide range of conditions in the remote troposphere. In our studies, large nucleation events are found exclusively in regions of enhanced sulfuric acid vapor (H_2SO_{4g}) concentrations, with maximum concentrations never exceeding 5×10^7 molecules cm^{-3} . Although these data suggest that H_2SO_{4g} participated, comparisons between ambient conditions in regions of nucleation to conditions necessary for binary H_2SO_4 water (H_2O) nucleation indicate that the mechanism may vary with elevation. In remote marine regions, at altitudes greater than ~ 4 km above sea level, observations of nucleation in clear air along cloud perimeters are in fair agreement with current classical binary nucleation models. In these regions, the low temperatures associated with high altitudes may produce sufficiently saturated H_2SO_4 for the production of new $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ particles. However, uncertainties with current binary nucleation models limit decisive comparisons. In warmer regions, closer to the earth's surface, measured H_2SO_4 concentrations are clearly insufficient for binary nucleation. Conditions at these sites are similar to those observed in an earlier study where there was circumstantial evidence for a ternary mechanism involving H_2SO_4 , H_2O , and ammonia (NH_3) [Weber *et al.*, 1998], suggesting that this may be a significant route for particle production at lower altitudes where surface-derived species, like NH_3 , are more apt to participate.

I. Introduction

Homogenous nucleation is the formation of new thermodynamically stable particles from condensation of gas-phase species. With current aerosol measurement techniques, studies show that nucleation is sporadic and does not occur uniformly throughout the atmosphere. Nucleation events have been

observed when local emissions or chemical production of precursor species perturbs the atmosphere, or when cloud processing or precipitation scavenging reduces the aerosol surface area. Nucleation does not occur in aged aerosols, which have sufficient surface area to suppress concentrations of precursor gases below levels required to generate new particles. Homogenous and heterogeneous nucleation play a large part in shaping ambient aerosol populations. Homogeneous nucleation increases aerosol number densities, whereas heterogeneous nucleation results in the growth of preexisting particles to larger sizes. Because the influence of tropospheric aerosols on the solar and terrestrial radiation balance is significant, and sensitive to particle size and numbers, these processes are particularly pertinent given concerns with global climate change.

Predicting the occurrence and rate of new particle formation requires an understanding of the mechanism of particle formation. Evidence of homogenous nucleation has been observed in widely varying circumstances, such as in the remote marine boundary layer [Hoppel *et al.*, 1994], in regions of cloud outflow [Hegg *et al.*, 1990; Perry and Hobbs, 1994], and at a remote continental site [Weber *et al.*, 1997]. Both models [Hamill *et al.*, 1982] and indirect measurements of particle composition [Brock *et al.*, 1995] suggest that in the upper troposphere (~ 10 km altitude), particles are formed via nucleating H_2SO_4 and H_2O . In contrast, models simulating nucleation at low elevations must often multiply predicted H_2SO_4 - H_2O nucleation rates by factors of up to 10^7 to obtain new particle production in these regions [e.g., Andronache *et al.*, 1997].

Recent studies [Weber *et al.*, 1995; Weber *et al.*, 1996; Weber *et al.*, 1997], which involved simultaneous measurements of the likely nucleation precursor species, H_2SO_{4g} and H_2O_g , and newly formed "nanoparticles" (3 to 10 nm diameter) have shown that although H_2SO_{4g} appears to play a role, observed particle formation rates are often not explained by current H_2SO_4 - H_2O nucleation models. Here we present a summary of precursor species concentrations and estimates of observed nucleation rates in regions where measurements suggested H_2SO_{4g} participated in new particle formation. These observations are compared to predictions of nucleating H_2SO_4 - H_2O . The comparisons include measurements of nucleation observed in

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widely varying circumstances from several remote regions around the globe.

II. Predicted H₂SO₄-H₂O Nucleation Rates

The classical bimolecular H₂SO₄-H₂O nucleation model that includes the effects of H₂SO₄ hydrates [Jaecker-Voirol and Mirabel, 1989] has been widely used in aerosol physicochemical models to predict H₂SO₄-H₂O nucleation rates in the remote atmosphere (hereafter referred to as the J-M model). A revised classical H₂SO₄-H₂O nucleation model [Wilemski, 1984], which claims to correct a thermodynamic inconsistency inherent with the J-M model, may be in better agreement with laboratory studies of nucleation [Viisanen *et al.*, 1997]. This model was recently parameterized by Kulmala *et al.* [1998] (hereafter the W-K model). The predictions of these two models differ substantially. The W-K model is more restrictive as to conditions where H₂SO₄-H₂O nucleation occurs because it requires higher H₂SO_{4g} concentrations (at a given temperature and relative humidity) to achieve similar nucleation rates. Figure 1 shows the predicted total H₂SO_{4g} relative acidity (RA) and relative humidity (RH) for a nucleation rate of 1 particle cm⁻³ s⁻¹ given by J-M and W-K for temperatures of +25°C and -25°C. Predicted H₂SO₄-H₂O nucleation rates depend only on these three parameters. Both J-M and W-K report nucleation rates as a function of total H₂SO_{4g} concentration. This includes free acid and all monoacid hydrates (i.e., H₂SO₄(H₂O)_n, n ≥ 0). Total RA is the concentration of total H₂SO_{4g} divided by the concentration of free H₂SO_{4g} in equilibrium with pure acid over a flat surface. To convert total acid concentration ([H₂SO_{4g}]_t) to total RA (RA_t), we use the saturation vapor pressure of pure acid given by Ayers *et al.* [1980]. In the following comparisons, it must be kept in mind that current predictions of H₂SO₄-H₂O nucleation rates are uncertain. Both J-M and W-K are based on the liquid drop model and are not a molecular level theory. Recent work [McGraw and Weber, 1998] has shown that the liquid drop model over-predicts the degree of H₂SO₄ hydration. This results in an under-prediction of binary nucleation rates.

III. Observations of Nucleation with Concurrent Measurements of H₂SO₄ and H₂O

Concurrent measurements of H₂SO_{4g}, H₂O_g, and freshly formed particle concentrations permit comparisons between ambient conditions where nucleation is observed to those that would be required for nucleating H₂SO₄-H₂O. We have performed these types of studies in the remote troposphere at the Mauna Loa Observatory, Hawaii [Weber *et al.*, 1995], at a remote continental site in the Colorado Rocky Mountains [Weber *et al.*, 1997], and in the remote Pacific during both the first NASA Pacific Exploratory Mission (PEM-tropics) [Clarke *et al.*, 1998a] and the first Aerosol Characterization Experiment (ACE 1) [Clarke *et al.*, 1998b; Weber *et al.*, 1998].

To compare observations with predictions, regions of new particle formation are identified and observed particle formation rates are estimated. Regions of recent or ongoing nucleation are identified by elevated concentrations of freshly formed particles. Because newly formed particles are about 1.5 nm in diameter, and we can only detect particles larger than ~3 nm, the time for particle growth to a detectable size (order of minutes to an hour) can lead to uncertainty in the conditions which existed at the time of nucleation. To minimize this uncertainty, we identify nucleation by focusing on events where 3-10 nm particle size

distributions indicate that most of the nanoparticles had just reached our instrument's lower detection limit (~3 nm; e.g., see Weber *et al.* [1998]). We also assume nucleation occurred under the most optimal conditions (i.e., highest RA_t) in the vicinity of the identified nucleation region.

Estimates of average nucleation rates are calculated from observed changes in particle concentration and estimates of the time interval over which nucleation occurred. For the ground-based studies at the Mauna Loa Observatory and Idaho Hill, evidence of nucleation was observed almost daily. Here, we only consider one representative day from each study. Both days have been discussed in detail [Weber *et al.*, 1995; Weber *et al.*, 1997]. At both sites, [H₂SO_{4g}]_t typically increased during the morning and reached maximum values of ~10⁷ molecules cm⁻³ near noon. Total particle concentrations (all particles larger than ~3 nm diameter) increased from nighttime levels to ~10³ to 10⁴ cm⁻³ following the peak in [H₂SO_{4g}]_t by about 1 to 2 hours. Some of the lag between maximum [H₂SO_{4g}]_t and total particle concentrations is interpreted as the time required for condensational growth of newly formed particles to detectable sizes. To estimate average particle formation rates, we model the system as a batch reactor under the assumption that the conditions leading to new particle formation were those recorded at the time of maximum [H₂SO_{4g}]_t. Average nucleation rates are estimated from the increase in total particle concentrations (typically of the order of 10³ to 10⁴ cm⁻³) divided by the time for nucleation, which is estimated from the lag between the peak in [H₂SO_{4g}]_t and sharp rise in total particle concentration. Total particle concentrations are used so that growth out of a narrow size range does not need to be considered when estimating the number of particles formed. For Mauna Loa and Idaho Hill, calculations give nucleation rates of about 1 particle cm⁻³ s⁻¹, uncertain to an order of magnitude. Because particles could be formed in significantly shorter intervals than is provided by our estimate, this analysis will tend to under-predict nucleation rates. Thus, it is a conservative approach for predicting conditions required for H₂SO₄-H₂O nucleation because higher saturation's of precursor species would be required to achieve higher nucleation rates.

In an earlier analysis of these data we estimated nucleation rates from measured nanoparticle concentrations and calculated nanoparticle growth rates [Weber *et al.*, 1996]. The approach for estimating nucleation rates in this study yield rates that are consistent with our earlier estimates, if one considers our more recent observation [Weber *et al.*, 1997] that actual nanoparticle growth rates are about an order of magnitude higher than our rates predicted under the assumption that only H₂SO_{4g} and H₂O_g contribute to growth.

In the PEM Tropics study a large nucleation event was observed near the ocean surface, adjacent to a region of precipitation [Clarke *et al.*, 1998a]. The research aircraft flew through the nucleation region, passing a peak in [H₂SO₄]_t, followed downwind by a sharp rise in total particle concentrations. Like the ground based sites, we assume that nucleation occurred when [H₂SO₄]_t was maximal. We estimate the nucleation rate was 1 to 10 cm⁻³ s⁻¹, based on changes in total particle concentrations and the advection time between the locations of the maximum [H₂SO₄]_t and maximum total particle number concentration.

A similar approach for estimating nucleation rates is used for the airborne measurements of ACE 1. In this study large nucleation events were observed downwind of penguin rookeries on Macquarie Island and in air recently vented from clouds. Conditions leading to nucleation downwind of the rookeries

Table 1. Conditions where evidence of new particle formation was observed, and estimated average particle production rates from various studies.

Location	Height above surface, km	$[H_2SO_4]_t$, 10^{16} cm^{-3}	RH, %	T, °C	RA_t , 10^{-4}	J_{obs} , $\text{cm}^{-3} \text{ s}^{-1}$
MLO	ground-based	9.5 (7.8, 9.5)	53 (49, 53)	11 (11, 12)	1.0 (0.8, 1.0)	0.5
Idaho Hill	ground-based	9.8 (8.4, 9.8)	28 (28, 29)	10 (10, 10)	1.2 (1.0, 1.2)	1
ACE 1 MI	0.043 0.55 (0.4, 0.55)	4.4 7.3 (4.4, 7.3)	58 17 (17, 58)	4.4 2.8 (1.5, 4.4)	1.2 2.4 (1.7, 2.4)	1 0.5-10
Pem-Tropics	0.17 (0.16, 0.18)	55 (3.7, 55)	91 (88, 98)	24 (23, 25)	1.3 (0.1, 1.3)	1-10
ACE 1 F17	3.5 (3.2, 4.1)	6.0 (2.0, 6.0)	50 (36, 62)	-13 (-11, -17)	16 (11, 16)	~1
ACE 1 F27	4.2 (4.0, 4.2)	19 (12, 19)	45 (45, 65)	-20 (-19, -20)	157 (83, 157)	~1
ACE 1 F14	6.3 (6.3, 6.3)	2.3 (1.6, 2.3)	67 (55, 67)	-31 (-31, -31)	108 (73, 108)	~1

The locations for the various studies are: MLO, Mauna Loa Observatory, Hawaii (3400 m above sea level); Idaho Hill, Colorado (1070 m above sea level); ACE 1 MI, Macquarie Island (measurements at two different altitudes are shown); ACE 1 F17, F27, and F14 denote flight numbers from the ACE 1 study; Pem-Tropics, first NASA Pacific Exploratory Mission. $[H_2SO_4]_t$ is gas phase concentration of free sulfuric acid and all sulfuric acid mono-hydrates (i.e., total acid) and RA_t is the total relative acidity. J_{obs} is estimated average observed nucleation rate. The table shows measurements recorded at maximum RA_t and ranges () in various regions of nucleation.

have been studied in detailed and are well characterized [Weber *et al.*, 1998]. Nucleation rates are estimated from the increase in total particle concentrations and the time for the air to be advected from the rookery to the measurement location. Again, average nucleation rates are about $1 \text{ cm}^{-3} \text{ s}^{-1}$. Interpretation of the events observed in the vicinity of clouds is less certain. Typically, where evidence for nucleation was detected, total particle concentrations were above background levels by concentrations ranging from about 1×10^3 to $5 \times 10^3 \text{ cm}^{-3}$, similar to levels recorded at other nucleation sites. However, because the aircraft generally skirted the clouds, the precise location of the nucleation region relative to the cloud, and the relative speed of the aerosol detraining from the cloud is unknown. This makes estimates of the time for nucleation difficult. We estimate that these freshly formed particles were less than one hour old. This gives an average nucleation rate of about $1 \text{ particle cm}^{-3} \text{ s}^{-1}$. A similar analysis from the measurements of nucleation near clouds reported by other investigators [Hegg *et al.*, 1990; Perry *et al.*, 1994] give similar average nucleation rates in regions of cloud venting.

Table 1 summarizes measured parameters pertinent to nucleation for the events discussed here. The table gives the measurements recorded at the time of maximum RA_t and the range, in brackets, observed in regions of nucleation. Note that in all cases the $[H_2SO_4]_t$ which led to nucleation was at most $\sim 5 \times 10^7 \text{ molecules cm}^{-3}$ and that average nucleation rates were of the order of $1 \text{ particle cm}^{-3} \text{ s}^{-1}$. The latter permit comparisons between conditions at the various sites where nucleation is observed to those required for a H_2SO_4 - H_2O nucleation rate of $1 \text{ particle cm}^{-3} \text{ s}^{-1}$. This comparison is shown in Figure 1 and is further aided by the high sensitivity of the H_2SO_4 - H_2O nucleation rate to $[H_2SO_4]_t$ (and RA_t). This means that small changes in $[H_2SO_4]_t$ lead to large changes in formation rates making the comparison insensitive to order of magnitude uncertainty in observed nucleation rates. For example, the "error bars" in Figure 1 show the range in RA_t and RH for a nucleation rate ranging from 0.1 to $10 \text{ cm}^{-3} \text{ s}^{-1}$ at -25°C for the W-K model and at $+25^\circ\text{C}$ for the J-M model. At similar temperatures, the ranges in RA_t and RH are similar for both models. This comparison appears more useful than those of observed and predicted

nucleation rates in view of the high sensitivity of nucleation rates to measurement uncertainties in RA and RH .

Figure 1 shows that in the regions of cloud venting for flights 14 and 27 of ACE 1, the conditions are approaching those necessary for nucleating H_2SO_4 - H_2O . The flight 17 measurement is also in a region of cloud venting but does not agree with the binary models. Somewhat better agreement is possible considering that nucleation could have occurred closer to the cloud, in a region of RH higher than shown. Moreover, it has been suggested that mixing processes along cloud perimeters

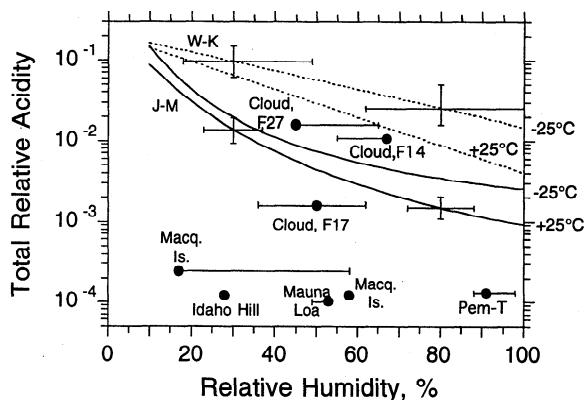


Figure 1. Comparison of conditions that resulted in nucleation events at various sites in the remote troposphere to predictions of onset of H_2SO_4 - H_2O nucleation (i.e., nucleation rate of $1 \text{ cm}^{-3} \text{ s}^{-1}$) using the models of Jaeger-Voirol and Mirabel [1989] (J-M, dotted line) and the parameterized version [Kulmala *et al.*, 1998] of Wilemski's [1984] model (W-K, solid line) at two different temperatures. Nucleation exceeds $1 \text{ cm}^{-3} \text{ s}^{-1}$ for values of (RH , RA_t) above or to the right of the locus of points given by the several curves. Model "error bars" show the conditions for a nucleation rate ranging from 0.1 to $10 \text{ cm}^{-3} \text{ s}^{-1}$. Data are conditions recorded at maximum total relative acidity and "error bars" the relative humidity range observed in the various regions of nucleation. For comparisons between measurements and predictions, refer to Table 1 for the ambient temperature at each measurement.

may also moderately enhance binary nucleation rates over predictions based on average conditions [Nilsson and Kulmala, 1998]. Thus, we conclude that at least for higher elevations in remote marine regions, particles may be formed along cloud perimeters through $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation. However, uncertainties in binary nucleation models limit our certainty of the nucleation mechanism.

The disparity with the binary models is much larger for all other measurements. Given the magnitude of the discrepancy, in these regions, there is little doubt that nucleation was via a different mechanism. Further evidence for this is provided by comparison with the Macquarie Island study [Weber *et al.*, 1998] where nucleation involved biogenic species, possibly ammonia (NH_3) through a ternary mechanism of $\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$. Comparisons of nucleation events at Mauna Loa, Idaho Hill, and Pem-Tropics, to the Macquarie Island measurements, qualitatively suggest that nucleation in these regions may have also been by this mechanism. Note, we cannot rule out the possibility that H_2SO_4 did not participate in these nucleation events. For example, nucleation may have involved low volatile organic species. Furthermore, although observations of high $[\text{H}_2\text{SO}_4]_t$ in regions of nucleation support the hypothesis that H_2SO_4 participated, it does not necessarily prove it; H_2SO_4 could be a surrogate for other photochemical generated nucleation precursor species. However, based on our observations, and measurements of exceedingly low H_2SO_4 vapor pressures over $\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$ mixtures [Marti *et al.*, 1997], we believe a ternary mechanism is a viable explanation.

Our data suggest a trend in nucleation mechanism with elevation. Despite roughly similar $[\text{H}_2\text{SO}_4]_t$ (Table 1), colder temperatures produce higher RA_t at higher altitudes. This results in closer agreement between observations and $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation predictions in these regions. At lower altitudes, the measurements substantially differ from $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ predictions, possibly due to the closer proximity of these sites to surface sources of potential precursor species like NH_3 . This is consistent with observations that ammonia concentrations are generally highest at low elevations and decrease with altitude [Georgii and Müller, 1974]. The result is that in these regions, alternative nucleation mechanisms which involve H_2SO_4 , could permit particle production at much lower $[\text{H}_2\text{SO}_4]_t$ than required for binary nucleation.

Finally, a curious feature of our data is that although locations, ambient conditions, and nucleation mechanisms vary, we consistently observed average nucleation rates of the order of $1 \text{ cm}^{-3}\text{s}^{-1}$. This is due to our observations that large nucleation events tend to increase particle concentrations by 10^3 up to 10^5 particles cm^{-3} . This may result from the self-limiting nature of nucleation, whereby precursor vapors are depleted by forming new particles and the generation of aerosol surface [McGraw and Saunders, 1984]. Thus, the nucleation mechanism may not greatly influence the number of particles formed, however, it will dictate the ambient conditions necessary for its occurrence through the nucleation mechanism's sensitivity to precursor species concentrations (e.g., RH, RA, $[\text{NH}_3]$, etc.). In this way the mechanism determines both the frequency and locations of large atmospheric nucleation events.

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