

Uptake of NO_y on wave-cloud ice particles

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Abstract. In a flight through a wave cloud during SUCCESS on 2 May 1996, simultaneous forward- and aft-facing NO_y inlets were used to infer the amount of condensed-phase NO_y present on ice particles that were up to a few minutes old. Condensed-phase amounts were 25-75 pptv, or 10-20% of gas-phase NO_y. Given the rapid HNO₃ uptake on ice observed in the laboratory, a model calculation implies that virtually all of the gas-phase HNO₃ will be depleted in the first 1-2 minutes after the appearance of ice. Thus the NO_y observations are consistent with the laboratory results only if the ambient HNO₃/NO_y ratio is 10-20%.

1. Introduction

Interactions between gas molecules and particles have proven to be important processes in the chemistry of the lower stratosphere. With the frequent occurrence of cirrus clouds in the upper troposphere there is potential for such processes to be important there as well. Indeed it has been suggested that cirrus ice particles may lead to the chemical destruction of ozone [Borrmann *et al.*, 1997; Kley *et al.*, 1996; Reichardt *et al.*, 1996]. In addition, ice particles are likely to play a role in the removal, or at least the temporary sequestration and downward transport, of NO_y species such as HNO₃ and HO₂NO₂ [Abbatt, 1997; Zondlo *et al.*, 1997]. Ice particles could also play a role in the interconversions among NO_y species. The NO_y content of wave cloud ice particles is the subject of this paper.

The partitioning of NO_y between the gas and condensed phases is an important aspect of the NO_y budget. Species will be subject to different chemical and transport processes depending on whether they are in the gas or condensed phase. Photolytic and chemical reactions will be different, and the vertical transport will be different, as particles will sediment. The presence of NO_y species in ice particles could result from several processes: (1) They could be in the ice nuclei responsible for ice formation. (2) Their presence could result from chemical conversions that occur in/on the particles and that leave the product species in the condensed phase. Or, (3) they could be adsorbed onto the ice surface during or after growth.

During SUCCESS we employed forward- and aft-facing inlets to measure NO_y. Since cloud ice particles are collected with enhanced efficiency by the forward-facing inlet and discriminated against by the aft, the difference between the two simultaneous NO_y measurements enables a measurement of the amount of NO_y in the particles. In this paper we present measurements of condensed-phase NO_y made during a flight of the NASA DC-8 through a wave cloud over the Front Range of the Colorado Rockies on 2 May 1996. The wave cloud provides a unique

sampling opportunity since the particles are only minutes old and likely nucleated on sulfuric acid aerosols [Jensen *et al.*, this issue].

2. Measurements

The "aft-inlet" NO_y was sampled through a horizontal inlet tube oriented perpendicular to the aircraft window. The tube's end face was a 45° cut, oriented 45° away from directly aft. This inlet provided a measurement that was dominated by gas-phase NO_y but that may also include NO_y present on aerosols smaller than 1 μm or so which are likely to be ingested (approximate calculations according to Vincent *et al.* [1986] show that particles with diameters smaller than 1.4 μm are sampled with efficiencies greater than 50%). Gas-phase NO and O₃ were sampled through a separate aft-facing inlet. This instrument has previously flown on the DC-8 [Weinheimer *et al.*, 1993] and on the NCAR Sabreliner [Ridley *et al.*, 1994]. Data were collected at 1 sec in SUCCESS. The overall error in a 10-sec average NO_y value of 500 pptv is 34 pptv (1-σ). The precision is 16 pptv (1-σ).

NO_y was also sampled through a forward-facing inlet using a separate detection channel. This inlet was subsokinetic and collected particles preferentially relative to the gas phase. Condensed-phase NO_y is inferred by taking the difference between the forward- and aft-inlet signals and dividing by the effective enhancement factor that is obtained by integration of the size-dependent enhancement factor over the size spectrum. This enhancement is treated as in Fahey *et al.* [1989], though for the ice particles of interest here, 10-20 μm in diameter, the enhancement factors are very close to the high-mass limit and so not as size-dependent as for the smaller particles in PSCs. Since the high-mass limit is well-defined, and simply equal to the ratio of the true air speed and the known flow velocity inside the inlet tube, enhancement factor errors due to flow around the inlet are minimal. The high-mass enhancement factor is altitude-dependent since a constant mass flow is drawn into the inlet. It has a value of 43 for the case described in this paper at 41 kft. Integrations of the size-dependent enhancement factor over the particle size spectrum give values which are generally within a few percent of this upper limit when in the wave cloud and not at the very edge. On the other hand, the flow around the fuselage could also impact particle concentrations. Using the semi-empirical approach of King [1984], we have estimated a shadow zone depth for 10-μm diameter particles of 4 cm at the fuselage station (530 in.) of the inlet. The inlet stuck out 37 cm into the flow, and at this point the King approach yields a particle concentration increase of 6% due to the flow past the fuselage. This 6% is too uncertain to use for data correction; it is small enough, however, to indicate that this is not a serious problem. The overall error in condensed-phase NO_y is not known. The two principal sources of error are likely (1) uncertainties about particle sampling due to flow around the fuselage and (2) temporary losses of HNO₃ to the walls that diminish the peaks and also lead to an inlet memory effect (illustrated later).

Heat was applied to both NO_y inlet lines and out to the inlet tips. The tips were maintained at 50°C. The inlet lines external to

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the aircraft were kept at 75°C , while those in the cabin were 50°C . For the forward-facing inlet, it is expected that ice particles will impact in a 90° bend; this will facilitate evaporation, as will dynamic heating.

Ice particles were detected by the Multi-Angle Spectrometer Probe (MASP) [Baumgardner *et al.*, this issue] and the Counterflow Virtual Impactor (CVI) [Twohy and Gandrud, this issue]. The MASP measured spectra of particle concentration, surface area, and volume over the diameter range of $0.3\text{--}40\ \mu\text{m}$. The CVI measured the ice water content for diameters larger than $6\ \mu\text{m}$.

3. 960502 Wave-Cloud Observations

Figure 1 shows a portion of the flight track of the DC-8 as it sampled a wave cloud in the vicinity of Boulder, Colorado. The dynamics and microphysics of this case are the subjects of other papers in this volume [Dean-Day *et al.*, Jensen *et al.*]. The aircraft flew legs perpendicular to the wind, along the upwind edge of the cloud, as well as legs along the wind, primarily upwind of the cloud but also into and out of the leading edge. For 20:18:20–20:19:00 (all times are UT, either hour:minute:second or hour:minute), the DC-8 flew approximately downwind, and for 20:27:50–20:29:05 approximately upwind inside the cloud. These periods are valuable for looking at air parcel evolution. For 20:20:00 to 20:28:00 the DC-8 flew in a straight line along the ragged upwind edge of the cloud, going in and out of cloud.

Figure 2a shows the NO_y measurements from the forward- and aft-facing inlets along with the particle total volume measured by MASP. The increases in forward-inlet NO_y are clearly associated with the presence of particles detected by MASP. There are several increments in forward-inlet NO_y of 1000–3000 pptv (Figure 2a, raw signal, including enhanced particle contribution), which when divided by the enhancement factor of about 40 give actual mixing ratios of about 25–75 pptv. These amounts are 10–20% of the NO_y mixing ratio from the aft-facing inlet. Note the common spike just after 20:22 that occurred in the absence of particles. This is DC-8 exhaust at the time when the plane crossed its own track (Figure 1), and NO showed a similar spike (not shown). Note, also, the failure of the forward-inlet NO_y to return to the aft-inlet baseline after going through particles. This is a forward-inlet “memory” effect that decayed over a time of minutes following a cloud encounter.

Figure 2b has an expanded scale, more clearly showing that there are deficits in aft-inlet NO_y coincident with particles. These

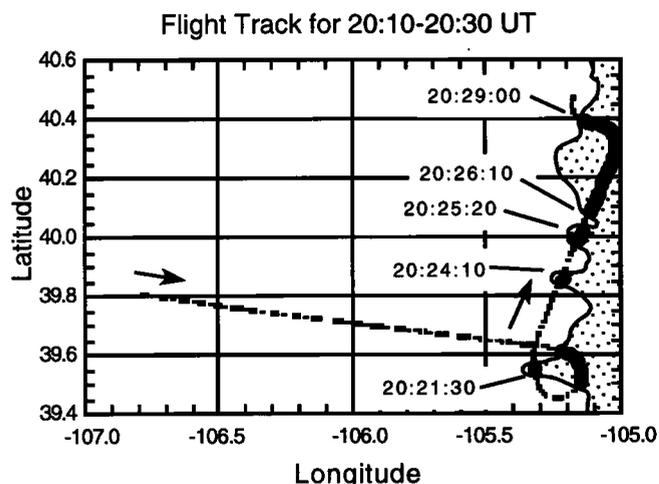


Figure 1. Flight track of the DC-8 prior to and during the wave cloud penetrations (large points, those with MASP surface area greater than $1000\ \mu\text{m}^2\ \text{cm}^{-3}$). The eastward leg is in the direction of the wind.

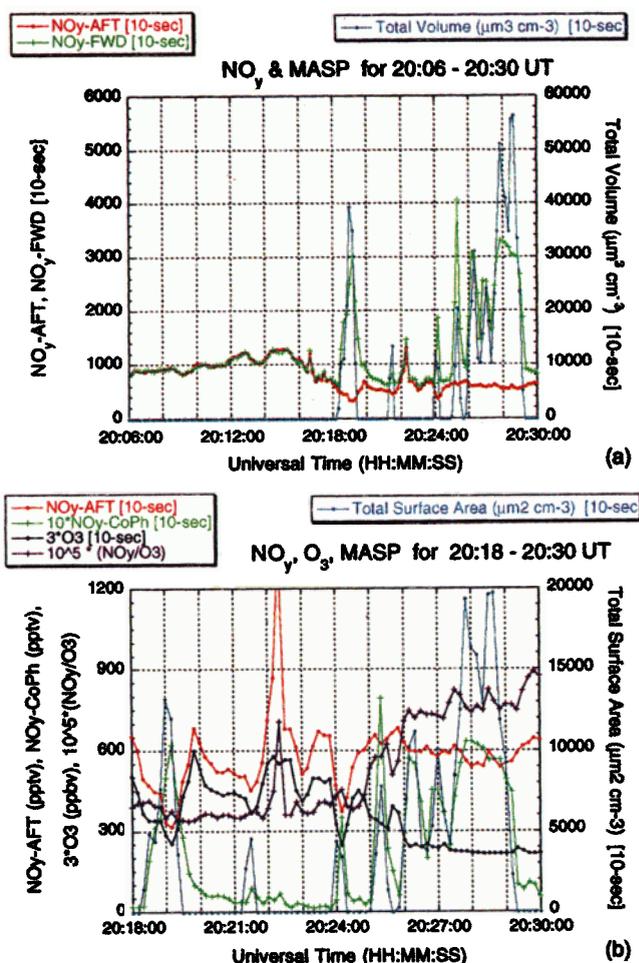


Figure 2. (a) Forward- and aft-inlet NO_y and MASP total volume upwind of and then along the upwind edge of the wave cloud. These “raw” forward-inlet NO_y measurements include the enhanced signal from particles. (b) Aft-inlet NO_y , condensed-phase NO_y , O_3 , NO_y/O_3 , and MASP surface area during the penetrations of the upwind edge of the wave cloud. Note that aft-inlet NO_y and O_3 , on the one hand, are anti-correlated with condensed-phase NO_y and MASP surface area, on the other.

deficits, however, are not primarily due to uptake on particles, as the O_3 mixing ratio shows the same correlations with the ice (Figure 2b). At 20:19:10 the NO_y decrement is about 50%, and there is a comparable decrement in O_3 . Since NO_y and O_3 are often positively correlated—note the relative constancy in Figure 2b of the NO_y/O_3 ratio during this period of large variation in the individual species—the primary cause of this 50% variation in NO_y is a change in air composition not caused by the ice. Even though the DC-8 flew at a constant pressure altitude during this period, the vertical motions associated with the cloud caused the in-cloud and out-of-cloud air to differ, and this effect is especially pronounced as the DC-8 was at and within a few hundred meters either side of the tropopause during this period (as indicated by microwave temperature profiler data [Dean-Day *et al.*, this issue]). As a result, we cannot expect to discern a smaller uptake-caused deficit in gas-phase NO_y in the presence of these larger changes which occurred synchronously. These correlations are characteristic of the contrast between stratospheric and tropospheric air. When NO_y and O_3 are high, the H_2O amount is low and there is not enough H_2O to form cloud particles; the air is more stratospheric. When NO_y and O_3 are low, the H_2O amount is high and cloud particles form; the air is more tropospheric.

Until 20:26, the anti-correlations between aft-inlet NO_y and MASP volume and surface area are dominated in this manner by air composition changes. However, for 20:26–20:30, at the north end of the track, the air is more uniformly tropospheric, as the O₃ mixing ratio drops to a uniform 70 ppbv and the NO_y/O₃ ratio changes to a higher, more tropospheric value of 8×10^{-3} . Thus for the period of 20:27:40 to 20:29:20 the aft-inlet NO_y deficit of 10–12% (relative to aft-inlet NO_y just outside this period) is possibly due to uptake on particles.

The amount of condensed NO_y can be expressed in two ways relative to the ice amount: (1) as a condensed-phase mixing ratio, NO_y to H₂O, as if it were distributed throughout the total volume of the ice particles, and (2) in units of monolayer coverage of the total ice surface area, as if it were distributed over the surface of the ice. CVI-measured ice water contents (IWCs) have peaks in the range of 3–6 mg m⁻³ (not shown). For IWCs greater than 3 mg m⁻³ the condensed-phase mixing ratios (NO_y/H₂O) have a range of 1.8–4.5 ppm by number of molecules. When the MASP total volumes are converted to IWCs using ice density, the inferred condensed-phase mixing ratios are in the range of 0.1–0.9 ppm. These ratios are significantly smaller than those derived from the CVI, as the MASP IWCs are significantly larger. The source of this MASP-CVI discrepancy is not known. The model of *Jensen et al.* [this issue] agrees more closely with the CVI measurements. Using the CVI-supported, model-calculated surface areas of about 2000 μm² cm⁻³, 25–75 pptv of NO_y would cover, as HNO₃, about 1.5–4.4% of a monolayer over the total particle surface area. Measured MASP surface areas lead to fractional surface coverages of 0.3–1.2%; these smaller fractions are consistent with the greater ice amounts determined by MASP.

4. Assessment of NO_y Uptake

Three mechanisms were mentioned earlier to account for the presence of NO_y in the particles. Our measurements do not allow us to distinguish among the three, as we are not in a position to assess (1) the prior presence of NO_y in the ice particle nuclei, nor (2) chemical conversion in/on the particles, but we can assess (3) the uptake of NO_y as HNO₃ on the basis of laboratory studies of the incorporation of HNO₃ in ice. The field measurements do not indicate whether the condensed NO_y resides throughout the volume or on the surface of the ice particles, but these possibilities can be assessed, first for the volume and then for the surface.

Davy and Somorjai [1971] found that the solubility of HNO₃ in ice is “on the order of parts per million or less.” Since CVI condensed-phase mixing ratios of 1.8–4.5 ppm by number were found (0.1–0.9 ppm from MASP), this suggests that the ice may have contained a significant fraction of the condensed NO_y throughout its volume, provided that the capacity is near the upper limit amount derived by *Davy and Somorjai* for growth of ice from the liquid. Alternatively, it may be the case that the adsorbed HNO₃ is excluded from the crystal lattice as it grows, causing the HNO₃ to accumulate on the surface of the growing crystal.

Recent work of *Abbatt* [1997] and *Zondlo et al.* [1997] shows that there is rapid uptake of HNO₃, up to approximately monolayer coverage, on the surface of pristine ice that is nearly in equilibrium with H₂O vapor. Since under these conditions there is little or no net growth of the ice, these are not the same conditions as occur during particle growth in the atmosphere where there may be significant co-condensation of the HNO₃ with the H₂O. However, these results allow us to assess the capacity of the ice particle surface to take up the available HNO₃ and also to determine whether there is adequate time during the minutes of particle growth in the wave cloud for uptake to occur. *Zondlo et al.* found a lower limit of 0.006 for the uptake coefficient during the initial rapid adsorption period, decreasing to 2×10^{-4} at the time of monolayer coverage; their apparatus did not allow them to obtain a value for the fast initial uptake. *Abbatt* found a lower

limit of 0.2 for the uptake coefficient of HNO₃ during the initial period of rapid uptake. Since the amount of condensed-phase NO_y is enough to cover only 1.5–4.4% of a monolayer (0.3 to 1.2% from MASP), it is clear that the ice particle surface has more than enough capacity to take up all of the condensable NO_y in the rapid uptake regime (uptake coefficient of tenths), even if none were distributed throughout the volume of the ice. It should be noted that the laboratory measurements assume that the ice surface area at the molecular scale is not significantly larger than the area at the larger scale. The same assumption is also made in the interpretation of the in-cloud measurements.

Figure 3 addresses the rate of uptake by applying the model of *Jensen et al.* [this issue] to the along-wind entry into the cloud just after 20:18. Here the wind speed and aircraft true airspeed have been used to convert the measured DC-8 time series into a time series as seen by an air parcel (as in *Jensen et al.*). Figure 3 shows the modeled HNO₃ uptake given these assumptions: an uptake coefficient of 0.2, an initial HNO₃/NO_y ratio of 15%, and an initial NO_y of 500 pptv. The calculations show that only 1–2 minutes is required to deplete nearly all of the 15% of NO_y that is assumed to be HNO₃, once appreciable ice has condensed. This is evident in the leveling off of the condensed-NO_y curve at 75 pptv. (The complementary decrease in gas-phase NO_y is difficult to discern.) For an uptake coefficient of 0.2 and a surface area of 2000 μm² cm⁻³ (the ultimate model value), the exponential time constant for HNO₃ depletion via uptake is 38 sec.

Note that the exit from the cloud at 20:29 (Figure 2b) shows the same qualitative behavior as measured and modeled for 20:18. Since the aircraft is traveling against the wind, the parcel time-in-cloud increases when going toward the left from 20:29 in Figure 2b (see also Figure 1). As for just after 20:18, there is a leveling-off of the condensed-phase NO_y after the initial rise upon entry of parcels into the cloud.

Thus the wave cloud observations are consistent with the rapid uptake observed in the laboratory, provided that the ambient HNO₃/NO_y ratio is only about 15%. This is generally below what can be expected on the basis of models. For example, the HNO₃/NO_y ratio in the NCAR IMAGES model [*Müller and Brasseur*, 1995] is 40–50% for these conditions [J.-F. Lamarque, personal communication]. Also, the HNO₃/NO_y ratio in the Harvard/GISS 3-D model above 8 km ranges between 30% and 40% on average for the SUCCESS sampling region and season, while the Harvard diel steady state model predicts a ratio of between 40% and 50% (expected to be higher than actual because it imposes steady state) [*Jaeglé et al.*, this issue; L. Jaeglé, personal communication]. The HNO₃/NO_y ratio of 10–20% estimated here from the condensed-phase NO_y measurements is

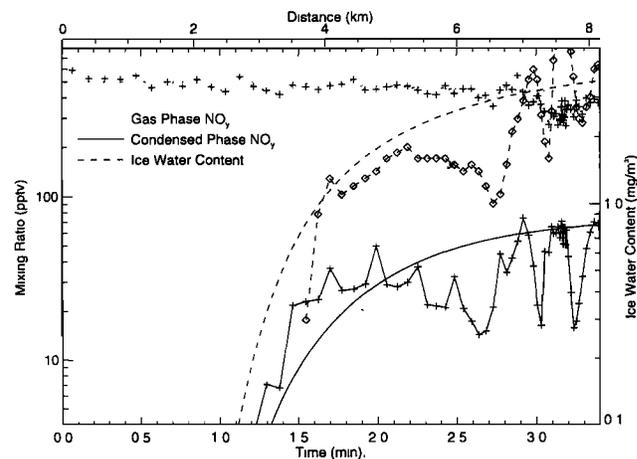


Figure 3. Model calculation of HNO₃ uptake for the downwind penetration just after 20:18 UT. Three model-observation pairs are plotted. The ice water content is from the CVI.

clearly lower than given by these models. On the other hand, it is consistent with recent HNO₃ measurements, albeit in different locales. Singh *et al.* [1996a] show an HNO₃/NO_y ratio of 10-15% (their Figure 10a) for 7-12 km altitudes in the midlatitude/subtropical Pacific (25-42°N) of PEM-West A. Singh *et al.* [1996b] show a ratio of about 10% (their Figure 12c) for 7-12 km altitudes in the South Atlantic of TRACE A.

Nonetheless, two alternative possibilities should be considered: (1) HNO₃ could be a larger fraction of NO_y than 15% but this is all that is taken up due to interference from species present in the atmosphere and not present for the laboratory measurements. The ice surface encountered by a HNO₃ molecule in the atmosphere is not the pristine surface of the laboratory experiments, but rather one which may already have many other non-H₂O species present, so it is possible that the laboratory uptake coefficients do not apply. (2) The value of 10-20%, for the fraction of ambient NO_y that is condensed, could be in error. This is not likely since it is derived from two sources with some independence. First, the forward-inlet NO_y measurement gives these ratios using the enhancement factor integrated over the MASP size spectrum. As noted above, enhancement factor errors are less severe when in or approaching the large mass limit, as is the case here. Second, the aft-inlet NO_y deficits observed at a time when NO_y/O₃ is relatively constant, 20:27:40-20:29:20 (in comparison to aft-inlet NO_y just outside this period), are 10-20% as well. Since this inference does not require knowledge of enhancement factors, it confirms that the HNO₃/NO_y ratio is only 10-20%.

5. Summary

Simultaneous measurements of aft-inlet and forward-inlet NO_y in the wave-cloud of 2 May 1996 are consistent with laboratory measurements of rapid uptake of HNO₃ provided that only 10-20% of the ambient NO_y is HNO₃. For this case, the laboratory-derived uptake coefficients imply that most of the HNO₃ is taken up in 1-2 minutes after the formation of ice particles, as they grow to diameters of 10-20 μm with ice water contents of 3-6 mg m⁻³. If there are other condensable NO_y species in significant abundance, such as HO₂NO₂, that are also taken up rapidly, then HNO₃ would need to be a smaller fraction of NO_y. If such measurements are repeated it would be useful to have measurements of species such as gas-phase HNO₃, HO₂NO₂, and CH₃C(O)OONO₂ in order to address this uncertainty. Also, laboratory simulations of the growth of ice in the presence of HNO₃ and HO₂NO₂ and other atmospheric species (e.g., H₂O₂) would be useful for determining whether the condensed species are distributed throughout the volume or are present primarily on the surface. If primarily on or near the surface, then they are more readily available for reaction with other gas-phase species. Such studies would also show whether other species may interfere with the uptake of HNO₃.

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