

The temporal evolution of the ratio HNO_3/NO_y in the Arctic lower stratosphere from January to March 1997

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Abstract. Aircraft-based measurements of HNO_3 , NO_y , N_2O , and O_3 have been performed in the Arctic lower stratosphere in January (POLSTAR I) and March (STREAM III) of 1997. The two projects employed different aircraft platforms. In addition, NO_y and O_3 were measured using different instruments in the two campaigns. HNO_3 and NO_y were found strongly correlated with correlation coefficients of 0.84 (POLSTAR I) and 0.69 (STREAM III), respectively. The fraction of HNO_3 within NO_y decreased from 96% in January to 59% in March. The decrease is consistent with the lifetime of HNO_3 due to photolysis after polar sunrise. The relationship of NO_y and HNO_3 with N_2O shows that in January NO_y and HNO_3 values were markedly higher than expected, which may indicate nitrification by PSC-II particle sedimentation and evaporation. Contradictory, the ratios NO_y/O_3 observed in January are only slightly elevated. In March, $\text{NO}_y/\text{N}_2\text{O}$ and NO_y/O_3 relations agree well with others reported in the literature. The difference between the NO_y/O_3 and $\text{NO}_y/\text{N}_2\text{O}$ relationships is partly explained by an observed O_3 decrease of about 30% between January and March.

Introduction

Nitric acid (HNO_3), which plays a key role in polar ozone chemistry, is the major component of reactive nitrogen (NO_y) in the lower stratosphere. So far only few simultaneous measurements of NO_y and HNO_3 in the lower stratosphere have been reported [Singh *et al.*, 1997; Fischer *et al.*, 1997], yielding HNO_3/NO_y ratios of about 70–80%. Calculated HNO_3/NO_y ratios [Fahey *et al.*, 1989; Kawa *et al.*, 1992] indicate the rather large range of the fraction HNO_3/NO_y of 40 to 90%. The main source of HNO_3 and NO_y in the stratosphere is thought to be the reaction of nitrous oxide (N_2O) with $\text{O}(^1\text{D})$ yielding two NO molecules which are further oxidized to NO_2 and HNO_3 . In the stratosphere, nitric acid is destroyed mainly by photolysis, leading to the formation of NO_2 . The latter is important for the chlorine deactivation via $\text{ClO} + \text{NO}_2 + \text{M} \rightarrow \text{ClONO}_2 + \text{M}$.

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When reactive nitrogen (mainly HNO_3) is removed from high altitudes (around 20 km) and released at lower altitudes (11 to 15 km) by PSC-II particle sedimentation and evaporation, the efficiency of chlorine deactivation is reduced. These so-called denitrification/nitrification events have been regularly observed in the Antarctic winter stratosphere [cf. Fahey *et al.*, 1990a], but only rarely in the Arctic stratosphere. For example, in February 1995, substantial denitrification was observed at altitudes around 19 km [Sugita *et al.*, 1998], while renitrification was observed between 10 and 13 km [Fischer *et al.*, 1997; Arnold *et al.*, 1998].

In late winter/early spring, sunrise after the polar night, followed by photodissociation of HNO_3 leads to formation of NO_x and, via reaction (1), of ClONO_2 . This is expected to shift the partitioning within NO_y towards NO_x and ClONO_2 and to a decrease of the HNO_3 fraction within NO_y .

This paper reports on measurements of HNO_3 , NO_y , N_2O , and O_3 during two aircraft campaigns in winter 1997 in the Arctic lower stratosphere. We focus on a strong reduction of the ratio HNO_3/NO_y observed from January to March.

Measurements

The POLSTAR 1 (Polar Stratospheric Aerosol Experiment 1) included 7 measurement flights between January 24 and February 06, 1997. The STREAM III (Stratosphere-Troposphere Experiment by Aircraft Measurements III) included 5 flights between March 9 and 25, 1997. The operational base for both campaigns was Kiruna, Sweden (69°N, 20°E). During the POLSTAR 1 mission the German research aircraft *Falcon*, owned and operated by DLR (Deutsches Zentrum für Luft- und Raumfahrt) was used, while the STREAM mission used the Dutch Cessna *Citation* research aircraft, operated by Delft University of Technology. Both research aircraft are twin-jets with a maximum flight altitude of approximately 14 km.

NO_y and O_3 in the POLSTAR I project were measured by DLR, Oberpfaffenhofen [Feigl, 1998]; NO_y in the STREAM III campaign was measured by the Max Planck Institute for Chemistry (MPI-CH), Mainz [Wienhold *et al.*, 1998]; O_3 during STREAM III was measured by the Institute for Marine and Atmospheric Research, Utrecht University (IMAU); in both campaigns N_2O was measured by MPI-CH; during the STREAM II project, additionally, N_2O was measured on two flights of the STREAM III campaign by the Institute for Stratospheric Chemistry, Forschungszentrum Jülich (FZJ) [Bujok *et al.*, 1998]; in both campaigns HNO_3 was measured by the Max Planck Institute for Nuclear Physics (MPIK), Heidelberg [Schneider *et al.*, 1998a]. For a detailed description of the IMAU and MPI-CH measurements see Fischer *et al.* [1997] and references therein.

It has to be taken into account that the NO_y measurements have been performed by two different groups (DLR and MPI-CH) and have not yet been directly compared. On the other hand, the gold converter technique is a well established measuring sys-

tem for stratospheric NO_y measurements. N_2O was measured during two flights of the STREAM III campaign by two different techniques (gas chromatograph, FZJ, and tunable diode laser, MPI-CH). The mean deviation (2σ) between both instruments was 1% (Flight #1) and 1.8% (Flight #2). Also, the calibration standards of both measurement systems have been compared, yielding agreement of $\leq 1\%$. Note also that the accuracy of the NO_y measurements is $\pm 25\%$ [Fischer *et al.*, 1997; Feigl, 1998], while that of HNO_3 is only $\pm 40\%$, due to the accuracy of the rate coefficient of the calibration reaction [Möhler and Arnold, 1991; Schneider *et al.*, 1998b]. IMRMS measurements of gaseous HNO_3 in the Arctic lower stratosphere can be affected by sampling and evaporation of cloud and aerosol particles containing HNO_3 . During the residence time of the probe gas inside the flow tube (approximately 50 ms) small particles are able to evaporate. Since the sampling is not isokinetic, an enhancement factor of particle sampling of about 4 is to be expected. During the POLSTAR I campaign where occasionally high cirrus clouds were observed in the tropopause region, an overestimation of HNO_3 can not be excluded. However, this does not affect the present data discussion since it focuses only on stratospheric data, selected by the criterion $\text{N}_2\text{O} < 300$ ppbv. In the STREAM III campaign the HNO_3 measurements suffered from an additional uncertainty due to collision induced ion dissociation. A correction has been applied relying on the measured ion ratio O^+/CO_3^+ , yielding a correction factor of about 1.1. This increased the overall uncertainty to $\pm 50\%$.

Results and discussion

Figure 1 presents the nitric acid volume mixing ratios measured during the POLSTAR I (a) and STREAM III (b) campaigns. The entire data sets are shown as a function of potential temperature Θ . The error bars indicate the accuracy (POLSTAR I: 40%, STREAM III: 50%) of the measurements. During the POLSTAR I mission, slightly higher potential temperatures of up to 365 K were reached. Highest nitric acid mixing ratios of up to 4 ppbv were observed at a Θ level of about 357 K. During the STREAM III campaign maximum HNO_3 mixing ratios reached only 2.5 ppbv at a Θ level of 352 K.

For further analysis, the data were averaged over 5 minutes, and a subset was selected by the criterion $\text{N}_2\text{O} < 300$ ppbv to exclude tropospheric air masses. This rather conservative criterion

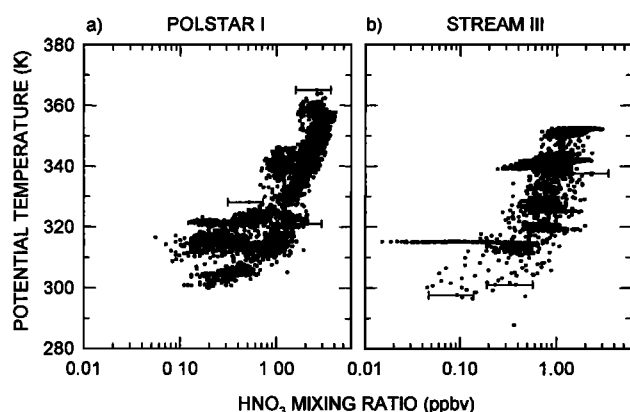


Figure 1. Nitric acid mixing ratios as a function of potential temperature (Θ) from (a) POLSTAR I (24.01.97 - 06.02.97) and (b) STREAM III (09.03.97 - 25.03.97). The error bars indicate the accuracy (POLSTAR I: 40%, STREAM III: 50%).

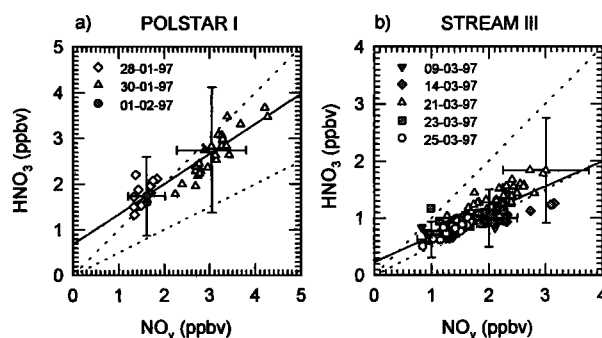


Figure 2. HNO_3 as a function of NO_y during POLSTAR I (a) and STREAM III (b). Solid lines: linear regression. Dotted lines: 50% and 100% level, respectively. The error bars indicate the systematic uncertainties. Mean ratios HNO_3/NO_y are: POLSTAR I: 0.96 ± 0.18 ; STREAM III: 0.59 ± 0.05 .

led to the rejection of 3 flights so that only 4 flights from the POLSTAR I mission (28-01-97, 30-01-97, 01-02-97, 04-02-97) are included in the data discussion. We note that NO_y was not measured on the flight of 04-02-97.

NO_y partitioning

Figure 2 presents the overall relationships between HNO_3 and NO_y during both campaigns, i.e. for all flight tracks characterized by $\text{N}_2\text{O} < 300$. The accuracy of the measurements is indicated by the error bars (HNO_3 : 40% and 50%, respectively; NO_y : 25%). The dotted lines indicate HNO_3 fractions within NO_y of 50% and 100%. The solid lines represent linear regressions through the data points. Both species are well correlated with a correlation coefficient r^2 of 0.84 during POLSTAR I and 0.69 during STREAM III. The most remarkable feature evident from Figure 2 is the different HNO_3 fraction comparing January and March 1997. While the mean ratio HNO_3/NO_y (for $\text{N}_2\text{O} < 300$ ppbv) of the three POLSTAR I flights is 0.96 ± 0.18 , it is only 0.59 ± 0.05 (mean and standard deviation) during the five STREAM III flights.

Figure 3 shows the temporal evolution of the HNO_3/NO_y ratio between January 28 and March 25, 1997. The thick bars indicate mean values and standard deviations for the individual flights, the thin error bars indicate the absolute accuracy of the measure-

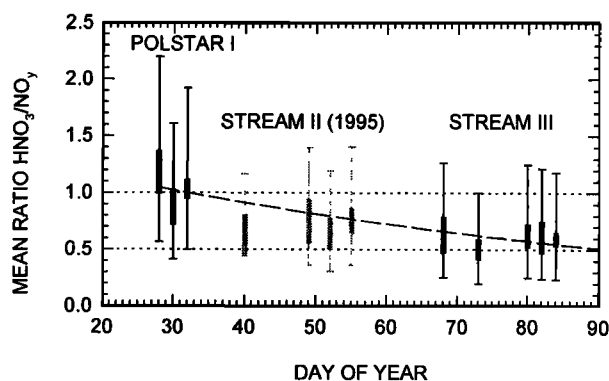


Figure 3. Temporal evolution of the ratio HNO_3/NO_y between January and March 1997. Thick bars: Standard deviation; Thin error bars: Systematic uncertainty. The dashed line represents the functional fit $y = a \times \exp(-t/\tau)$ to the 1997 data yielding a timescale of HNO_3 decay of 86 days. Also indicated are the HNO_3 to NO_y ratios measured in February 1995 (STREAM II, Fischer *et al.* [1997]).

ments: 25% for NO_y, 40% for HNO₃ during POLSTAR I, 50% for HNO₃ during STREAM III. These errors were added linearly, resulting in a "worst case" scenario (i.e. the upper error bar limit, for example, is reached when HNO₃ was overestimated by 40% and NO_y was underestimated by 25%). The accuracy of the HNO₃ measurements of 40–50% is mainly determined by the error in the rate coefficient of the calibration reaction [Möhler and Arnold, 1991; Schneider et al., 1998b]. The HNO₃ fraction measured in January (96%) is very high, and the mean value of flight #1 (97-01-28) is even larger than 1. Thus, an overestimation of HNO₃ is rather likely, while an underestimation of NO_y during POLSTAR I is unlikely, since the values are already much higher than the calculated NO_y^{*} (see below). Any systematic error in HNO₃ or NO_y, however, does not change the observed decrease of the ratio HNO₃/NO_y.

Nitric acid is destroyed by photochemical conversion, either by photolysis (HNO₃ + hν → OH + NO₂) or by reaction with OH (OH + HNO₃ → H₂O + NO₃). A decrease of the HNO₃ fraction inside the NO_y at the end of the polar night is not completely unexpected, although, to our knowledge, neither measurements or model calculation showing this feature were reported up to now. From January to March, solar exposure is markedly increasing at the polar regions. The diurnally averaged photodissociation rate of HNO₃ for 70°N and 12 km altitude is increasing from about 0.2 × 10⁻⁷ s⁻¹ on January 20 to 2.5 × 10⁻⁷ s⁻¹ at the end of March. The dashed line in Figure 3 represents the functional fit $y = a \times \exp(-t/\tau)$ through the data, yielding a time constant τ of 86 days. This decrease agrees with the time constant for HNO₃ photolysis. Although the measurements discussed here have not encountered the same air masses, air masses sampled in March have seen longer periods of sunlight than those encountered at the end of January.

Also shown in Figure 3 is the ratio HNO₃/NO_y measured during the STREAM II project in February 1995 [Fischer et al., 1997] which yielded an average value of 69%. Although this pertains to measurements two years before the presently discussed data sets, in an environment presumably influenced by nitrification, the values fit rather well to the 1997 picture.

Correlations with N₂O and O₃

Figure 4 shows the volume mixing ratios of HNO₃ and NO_y as a function of N₂O for both measurement campaigns. Linear regressions through the HNO₃ data are represented by the solid

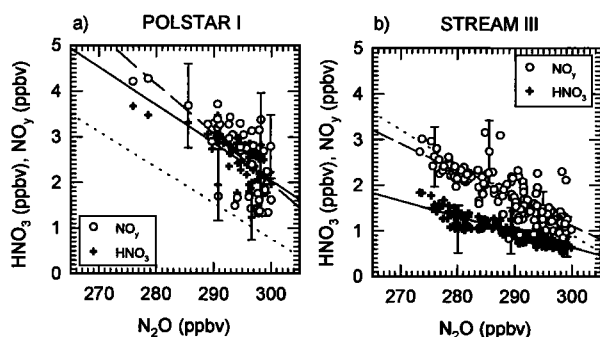


Figure 4. HNO₃ and NO_y as a function of N₂O. a): POLSTAR I; b): STREAM III. Only stratospheric data (N₂O < 300 ppbv) are shown. The data points represent 5 minute averages. The error bars indicate the systematic uncertainty. Dotted line: NO_y-N₂O relationship measured during AASE II [Weinheimer et al., 1993]; dashed line: linear regression of NO_y; solid line: linear regression of HNO₃.

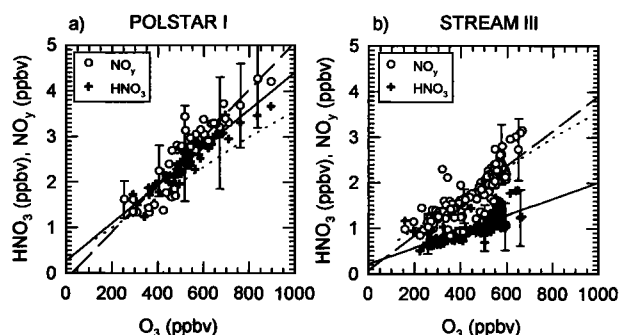


Figure 5. HNO₃ and NO_y as a function of O₃. a): POLSTAR I; b): STREAM III. Only stratospheric data (N₂O < 300 ppbv) are shown. The data points represent 5 minute averages. The error bars indicate the systematic uncertainty. Dotted line: NO_y-O₃ relationship measured during AASE I [Murphy et al., 1993]; dashed line: linear regression of NO_y; solid line: linear regression of HNO₃.

lines, the dashed lines refer to the NO_y data. The STREAM III data fit well to a linear regression with correlation coefficients of 0.66 (NO_y) and 0.76 (HNO₃), while the POLSTAR I data only yield a correlation coefficient of 0.44 (both HNO₃ and NO_y). The functional relationships given by the linear regressions are:

$$\text{POLSTAR I: } \text{NO}_y = -0.1081 \times (\text{N}_2\text{O} - 318.4) \quad r^2 = 0.44$$

$$\text{HNO}_3 = -0.0827 \times (\text{N}_2\text{O} - 324.7) \quad r^2 = 0.44$$

$$\text{STREAM III: } \text{NO}_y = -0.0598 \times (\text{N}_2\text{O} - 318.7) \quad r^2 = 0.66$$

$$\text{HNO}_3 = -0.0339 \times (\text{N}_2\text{O} - 319.2) \quad r^2 = 0.76$$

All values are ppbv. Also indicated, by the dotted line, is NO_y^{*} which is a measure of the expected NO_y [Fahey et al., 1990b]. NO_y^{*} was calculated using the expression $\text{NO}_y^* = \text{slope} \times (\text{N}_2\text{O}_{\text{meas}} - \text{N}_2\text{O}_{\text{trop}})$. We adopted the slopes measured in the AASE-II project [Weinheimer et al., 1993] for January (-0.0778) and March (-0.0765) and the mean tropospheric N₂O values measured during the POLSTAR I (310 ± 4 ppbv) and STREAM III (312 ± 3 ppbv) campaigns. The NO_y data from the STREAM data set are in very good agreement with calculated NO_y^{*}, indicating that no denitrification or nitrification occurred. The POLSTAR I NO_y and HNO₃ data are higher than the calculated NO_y^{*} by up to 2 ppbv. This finding might indicate nitrification by PSC-II particle sedimentation in January 1997, although 2 ppbv of renitrification seem to be very much, since that would require an almost complete removal of NO_y from the 20 km region. A calibration error of N₂O, however, can be excluded, since tropospheric values of both campaigns were equal inside the error limits of 4 ppbv (see above). A systematic overestimation of HNO₃ and NO_y might explain part of the difference (see error bars in Figure 4a), but then NO_y and HNO₃ would have been measured too low during the STREAM III campaign.

Figure 5 gives HNO₃ and NO_y as a function of O₃ for both campaigns. The dashed lines give the linear regressions to the NO_y-O₃ relationships, while the solid lines refer to HNO₃. The functional relationships are:

$$\text{POLSTAR I: } \text{NO}_y = 5.217 \times 10^{-3} \times \text{O}_3 - 0.141, \quad r^2 = 0.82$$

$$\text{HNO}_3 = 4.157 \times 10^{-3} \times \text{O}_3 + 0.264, \quad r^2 = 0.87$$

$$\text{STREAM III: } \text{NO}_y = 3.833 \times 10^{-3} \times \text{O}_3 + 0.077, \quad r^2 = 0.77$$

$$\text{HNO}_3 = 1.793 \times 10^{-3} \times \text{O}_3 + 0.215, \quad r^2 = 0.60$$

All values are ppbv. Also shown, by the dotted line, is the relationship NO_y/O₃ measured in AASE I [Murphy et al., 1993] in January/February 1989 at 59°N (Stavanger, Norway). The POLSTAR I NO_y data are slightly higher than the AASE I data, but the NO_y enhancement is smaller than that deduced from the N₂O reference. If substantial renitrification had taken place, the

ratio NO_x/O₃ should be enhanced by the same value (about 2 ppbv at the highest O₃ levels) as the NO_x-N₂O relationship. This finding is not consistent with the explanation in terms of renitricification.

The STREAM III data agree well with the AASE I data. However, there are also higher HNO₃/O₃ ratios reported in the literature: Weinheimer *et al.* [1993] report a ratio of HNO₃ to O₃ of about 5×10^{-3} ppbv/ppbv, measured in AASE II in January and March 1992. This value is nearly identical to the POLSTAR I data and higher than the STREAM III data. We also observe a decrease in O₃ from January to March: The loss in ozone between the POLSTAR I and the STREAM III measurements is about 30% at N₂O < 275 ppbv, corresponding to about 1% per day. This value is comparable to measurements from 1993 [Bregman *et al.*, 1997] and data deduced from ozonesonde measurements for 1997 [Knudsen *et al.*, 1998]. This O₃ decrease does partly compensate the response of the observed NO_x/O₃ ratio to the NO_x decrease between January and March, in a way that the ratio NO_x/O₃ does not change substantially.

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