

Denitrification in the Arctic winter 2004/2005: Observations from ACE-FTS

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[1] Arctic denitrification in the winter 2004/2005 is analyzed using measurements from the satellite instrument ACE-FTS. Two methods based on changes in the correlations of odd nitrogen (NO_y) versus methane and versus an artificial long-lived tracer are employed. The observations show that significant denitrification occurred in the region 15 km–20 km and that it was spatially inhomogeneous within the polar vortex. Before the vortex breakup in mid-March 2005 a maximum denitrification of 8 ± 0.8 ppbv, accounting for a relative reduction of over 50%, was observed at ~ 450 K (~ 17 km) using both methods. When averaged over the polar vortex for the first half of March, the denitrification at this level was about 4.4 ppbv and 5.0 ppbv from the artificial tracer method and the correlation method, respectively. **Citation:** Jin, J. J., K. Semeniuk, G. L. Manney, A. I. Jonsson, S. R. Beagley, J. C. McConnell, C. P. Rinsland, C. D. Boone, K. A. Walker, and P. F. Bernath (2006), Denitrification in the Arctic winter 2004/2005: Observations from ACE-FTS, *Geophys. Res. Lett.*, 33, L19814, doi:10.1029/2006GL027687.

1. Introduction

[2] Denitrification refers to the permanent removal of odd nitrogen (NO_y) from the polar lower stratosphere, and is caused by the sedimentation of polar stratospheric clouds (PSCs) that contain nitric acid (HNO_3). Polar ozone loss in spring is caused by catalytic reactions between odd oxygen ($\text{O} + \text{O}_3$) and active chlorine (Cl, ClO and HOCl) and active bromine (Br, BrO, and HOBr), which are activated through heterogeneous reactions on the surface of PSCs and are deactivated through reactions with nitrogen dioxide (NO_2) and methane (CH_4). Therefore denitrification can delay chlorine deactivation and consequently prolong polar ozone reduction [e.g., Santee *et al.*, 1996; Rex *et al.*, 1997; Tabazadeh *et al.*, 2000; Waibel *et al.*, 1999]. This phenomenon occurs frequently in the Antarctic [e.g., Fahey *et al.*, 1990; Santee *et al.*, 2004] because of the low winter temperatures. It occurs less frequently and denitrification is less extensive in the Arctic as a result of the often disturbed Arctic polar vortex and consequent higher temperatures. However,

significant denitrification has been observed in the Arctic for several exceptionally cold winters [Sugita *et al.*, 1998; Kondo *et al.*, 2000; Fahey *et al.*, 1990, 2001; Popp *et al.*, 2001].

[3] The Arctic winter 2004/2005 was particularly cold [Kleinböhl *et al.*, 2005; Manney *et al.*, 2006], and there was significant polar stratospheric cloud formation (A. Y. Zsetsky *et al.*, manuscript in preparation, 2006). Consequent severe ozone loss has been reported [Jin *et al.*, 2006; Manney *et al.*, 2006; von Hobe *et al.*, 2006; Singleton *et al.*, 2006], and denitrification has been seen in airborne [Kleinböhl *et al.*, 2005] and EOS MLS [Schoeberl *et al.*, 2006] observations. In addition, chlorine activation and deactivation during this winter was analyzed by Dufour *et al.* [2006]. In this study we use the version 2.2 data from the satellite instrument ACE-FTS [Bernath *et al.*, 2005] to analyze the denitrification for this winter. ACE-FTS is a Fourier Transform Spectrometer with a vertical resolution of ~ 3 –4 km; details of the retrieval approach are described by Boone *et al.* [2005].

[4] A useful approach to study polar denitrification is the correlation method [e.g., Fahey *et al.*, 1990; Popp *et al.*, 2001]. In this method, the deviation from the standard correlation of NO_y versus a long-lived tracer is interpreted as denitrification. However, it has been shown that this method can overestimate the extent of denitrification [Michelsen *et al.*, 1998] since tracer correlations can also be altered by mixing processes. Rex *et al.* [1999] used a mixing-line method to analyze denitrification assuming that mixing occurs in a single mixing event. Later, Plumb *et al.* [2000] showed that the mixing-line method was unable to remove all of the mixing effects. To avoid some of these problems, Esler and Waugh [2002] composed an artificial tracer that has a linear relation with NO_y , so that mixing does not change the correlations, to get a robust estimate of denitrification.

[5] The signature of depleted NO_y must be viewed with caution as HNO_3 can be temporarily sequestered in PSCs, such as supercooled ternary solutions (STS), which are sufficiently small that they sediment too slowly for HNO_3 removal. A potential indirect signature of denitrification via sedimentation is the increase of NO_y mixing ratios at lower altitudes when large PSC particles such as HNO_3 –containing Nitric Acid Trihydrate (NAT) “rocks” evaporate [cf. Waibel *et al.*, 1999; Kleinböhl *et al.*, 2005].

[6] In this study we estimate the denitrification from both the correlation method and the artificial tracer method. We also observe the signature of evaporated NO_y at lower altitudes.

2. Results and Discussion

[7] We have analyzed ACE-FTS measurements between 350 K–1000 K (~ 12 km–35 km) in the Arctic (50°N –

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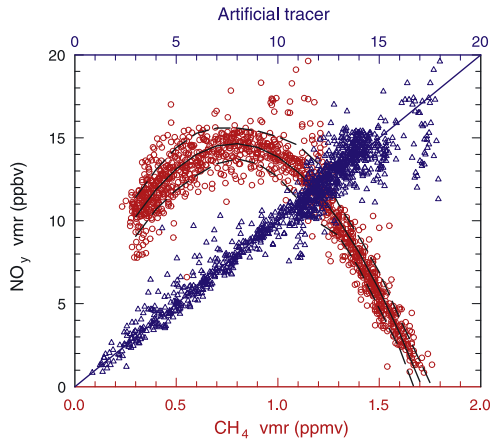


Figure 1. Correlations of NO_y/CH_4 (red circles) and NO_y /artificial tracer (blue triangles) between 350 K–1000 K (~ 12 km–35 km) for 1–15 January 2005. The black solid line is a 3rd order polynomial fit for the NO_y/CH_4 correlation. The black dashed lines represent the $1\text{-}\sigma$ error range of the fit. The blue solid line represents a linear fit that has NO_y equal to the artificial tracer.

80°N) for the period January–March 2005. We consider measurements north of 50°N with sPV (scaled potential vorticity [Manney *et al.*, 1994]) $> 1.8 \times 10^{-4} \text{ s}^{-1}$ above 500 K and sPV $> 1.4 \times 10^{-4} \text{ s}^{-1}$ between 350 K–500 K to be inside the polar vortex. sPV is calculated from NASA’s Global Modeling and Assimilation Office Goddard Earth Observation System version 4.0.3 (GEOS-4) meteorological analyses [Bloom *et al.*, 2005].

[8] Some of the odd nitrogen species (HNO_3 , ClONO_2 , NO_2 , N_2O_5 , and NO) were not retrieved in the lower stratosphere because of low densities, thus NO_y is derived as follows

$$\text{NO}_y = \text{HNO}_3 + \text{ClONO}_2 \quad (\text{potential temperature} < 450 \text{ K}),$$

$$\text{NO}_y = \text{HNO}_3 + \text{ClONO}_2 + \text{NO}_2 + 2 \times \text{N}_2\text{O}_5 \quad (450 \text{ K} \leq \text{potential temperature} < 600 \text{ K}),$$

$$\text{NO}_y = \text{HNO}_3 + \text{ClONO}_2 + \text{NO}_2 + 2 \times \text{N}_2\text{O}_5 + \text{NO} \quad (\text{potential temperature} \geq 600 \text{ K}).$$

We estimate that errors in the NO_y estimates are less than 10%.

[9] Figure 1 shows the correlation of NO_y versus CH_4 for 1–15 January 2005 and a polynomial fit to the data

$$\text{NO}_y = 0.878305 \times \text{CH}_4^3 - 20.1389 \times \text{CH}_4^2 + 30.1194 \times \text{CH}_4 + 2.99409. \quad (1)$$

where NO_y and CH_4 are volume mixing ratios in ppbv and ppmv, respectively. After 15 January 2005 the deviation from this correlation is employed to derive the NO_y change in the polar vortex. However, because of the curvature of the NO_y/CH_4 relationship, the correlation can change with

horizontal or vertical mixing. Therefore, using NO_y and CH_4 observations within the $1\text{-}\sigma$ error range of the above polynomial fit and simultaneous N_2O , CFC-11, and OCS observations between 350 K–1000 K for 1–15 January 2005, we compose an artificial tracer along the lines proposed by *Esler and Waugh* [2002] with the coefficients determined by a linear regression.

$$\begin{aligned} \text{Artificial tracer} = & 1.180 \times 10^{-2} \text{ CH}_4 \text{ (ppbv)} \\ & - 3.522 \times 10^{-2} \text{ N}_2\text{O} \text{ (ppbv)} \\ & - 4.778 \times 10^{-3} \text{ CFC-11 (pptv)} \\ & - 3.376 \times 10^{-2} \text{ OCS (pptv)} + 7.740 \quad (2) \end{aligned}$$

A linear correlation of NO_y with the artificial long-lived tracer for 1–15 January 2005 is obtained and shown in Figure 1. Because of the linear relationship, the correlation is insensitive to mixing processes. Here the linear fit that has NO_y equal to the artificial tracer is taken to be the representative of pre-denitrification conditions. Thus we attribute any deviation from this correlation after 15 January 2005 to denitrification or renitrification.

[10] Figure 2 shows the NO_y reductions, which are the differences between the observed NO_y and the NO_y derived from the correlation methods, for the period 1–15 January 2005. Generally, the results from the two methods are very similar and show no significant bias as expected for this period. The $1\text{-}\sigma$ deviations from zero are almost the same above ~ 380 K (~ 14 km), and are about 0.5–1.2 ppbv between 380 K–700 K.

[11] NO_y reductions inside the polar vortex between 16 January–15 March 2005 are shown in Figure 3. In order to exclude the possibility of temporary sequestration within PSC particles, observations with temperatures above and below the threshold temperature T_{NAT} for NAT formation are shown separately in this figure: we assume that no sequestration occurs above T_{NAT} . T_{NAT} is derived from simultaneous ACE-FTS water vapor and HNO_3 observations using the formula of *Hanson and Mauersberger* [1988]. We note that there are more data points available from the correlation method than from the artificial tracer method. This is because we required all four species to be available for the artificial tracer while for some occultations

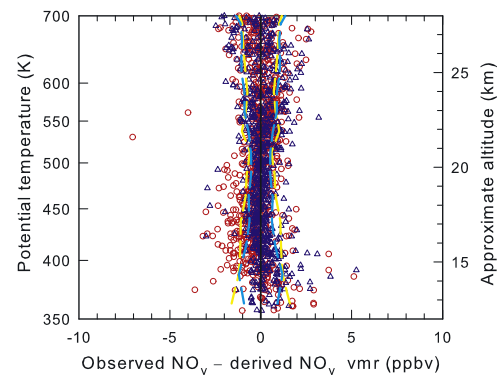


Figure 2. NO_y reductions for 1–15 January 2005 from the correlation method (red circles) and the artificial tracer method (blue triangles). The yellow and cyan dashed lines represent the $1\text{-}\sigma$ deviations from zero for the two methods, respectively.

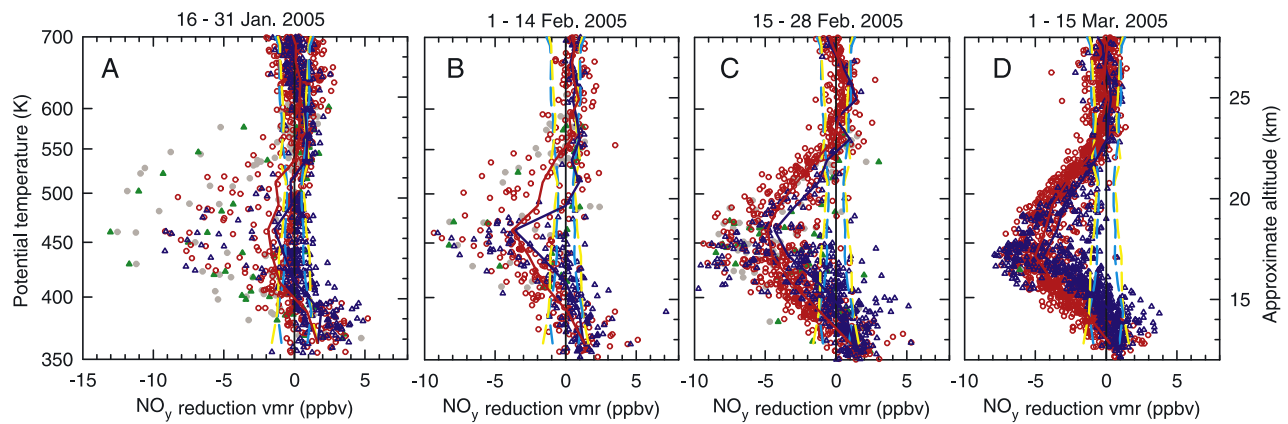


Figure 3. NO_y reductions inside the polar vortex for the periods 16 January 2005–15 March 2005. The open red and closed grey circles are calculations from the correlation method, and the open blue and closed green triangles are calculations from the artificial tracer method. The open and closed symbols are observations for temperatures above and below T_{NAT} , respectively. The red and blue solid lines are averages of the results with temperatures above T_{NAT} from the two methods. The yellow and cyan dashed lines are the same as in Figure 2.

not all of them were retrieved on account of low densities. Individual comparisons (not shown) indicate that differences between the results from the two methods can be seen up to 500 K, with the largest differences, over 5 ppbv for some observations, occurring between 380 K–425 K after the middle of February. This suggests that the denitrification is overestimated by the correlation method because of mixing [Michelsen *et al.*, 1998; Rex *et al.*, 1999; Plumb *et al.*, 2000]. Despite the significant differences for some of the individual denitrification estimates, the difference between their averages is less than 1 ppbv above ~ 425 K (~ 16 km) and less than 2 ppbv below this level.

[12] The NO_y departure from the gas phase was observed as early as in the second half of January 2005 (Figure 3a). This likely reflects the presence of a pool of extremely cold air over northern Canada and Scandinavia at the end of the month [e.g., Dufour *et al.*, 2006]. NO_y reductions occurred between 400 K–550 K (~ 15 km–23 km). Sequestration plus denitrification ($T < T_{\text{NAT}}$) resulted in NO_y reductions as large as 12 ppbv. The largest denitrification ($T > T_{\text{NAT}}$) present during this period occurred at ~ 450 K (~ 17 km), and was about 9 ± 0.8 ppbv from the correlation method and from the artificial tracer method, corresponding to a relative reduction of about 60%. The error is the $1-\sigma$ deviation from zero of the NO_y reduction for 1–15 January 2005. However, the average of the vortex denitrification for this period was limited to < 2 ppbv (Figure 3a), suggesting that the denitrification was not widespread.

[13] We also note an NO_y increase of ~ 5 ppbv at ~ 400 K (~ 15 km) and below. Since neither mixing nor horizontal transport can change the correlations to produce the NO_y enhancement, we attribute the increase to the sedimentation and evaporation of HNO_3 -containing particles [cf. Waibel *et al.*, 1999]. Several typical HNO_3 profiles indicating the increase at ~ 400 K in January are shown in Figure 4. The increase of HNO_3 at lower levels appears to be a signature of removal from upper levels, not withstanding the effect of horizontal wind shear along altitudes. Such renitrification has also been observed at potential temperatures above 340 K during the Polar Aura Validation Experiment (PAVE) flights at the end of January 2005 [Dibb *et al.*, 2006] and by

EOS MLS at ~ 15 km from the end of December 2004 [see also Schoeberl *et al.*, 2006].

[14] The denitrification intensified by the end of February (Figures 3b and 3c), which is consistent with the meteorological context. The potential PSC area in which the air temperature is below T_{NAT} at 475 K remained large (over 10×10^6 km²) until the middle of February, and did not totally disappear until the middle of March [Kleinböhl *et al.*, 2005]. We note that there were more ACE-FTS inner vortex observations available in March than in January (see equivalent latitude change in work by Dufour *et al.* [2006]) because of the poleward shifting of the orbit. It is difficult, therefore, to determine quantitatively how the denitrification intensified during this period.

[15] By the middle of March 2005, when the polar vortex broke up [Manney *et al.*, 2006], the vortex air was significantly denitrified between 400 K–500 K (~ 15 km–20 km) (Figure 3d). Both methods yield maximum denitrification estimates of about 8 ± 0.8 ppbv, accounting for over 50% relative reductions locally, at ~ 450 K (~ 17 km). The maximum was slightly smaller than the value at the end of February. We note that the difference in denitrification

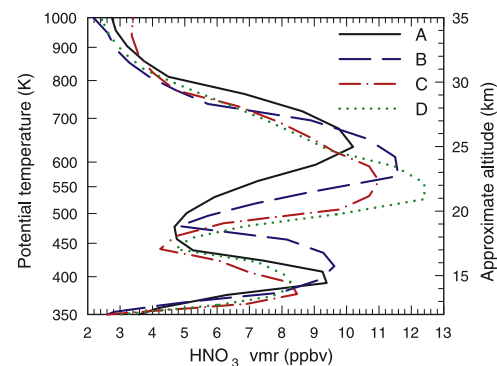


Figure 4. Several typical HNO_3 profiles. Line A, at (63°N , 96°W) on 13 January 2005; line B, at (63°N , 66°W) on 16 January 2005; line C, at (65°N , 80°W) on 22 January 2005; and line D, at (65°N , 63°W) on 23 January 2005.

was likely because different air parcels were sampled so their denitrification histories were different as opposed to this being a signature of release of NO_y from PSC particles. The denitrification average reached a maximum by the end of February: about 5.0 ppbv and 4.4 ppbv at ~ 17 km from the correlation method and the artificial tracer method, respectively. The altitude of the peak descended slightly but the maximum averages remained unchanged for 1–15 March 2005.

[16] The denitrification within the vortex was horizontally inhomogeneous in extent. Figure 5 shows the denitrification estimates plotted versus equivalent latitude (EqL, the latitude that would enclose the same area between it and the pole as a given potential vorticity contour) between 430 K–460 K where the largest NO_y reductions occurred. The broad range of denitrification estimates at high EqL suggests that the denitrification did not occur uniformly. This reflects the non-uniform distribution of temperatures below T_{NAT} inside the vortex. Meteorological analyses show that the region of the lowest temperatures during January and February was localized over Scandinavia and the north Atlantic so that vortex interior air was not processed homogeneously.

[17] Our denitrification estimates are in reasonable agreement with results from other measurements and modeling. For example, the maximum average of about 3.7 ppbv at around 450 K in early February 2005 (Figure 3b) is close to the value of 3.1 ± 0.8 ppbv obtained from airborne measurements for the same period [Kleinböhl *et al.*, 2005]. However, the maximum average from the ACE-FTS data occurred ~ 2 – 3 km lower than for the aircraft data, and the largest individual NO_y reduction was about 3 ppbv larger than the largest reduction of HNO_3 in the aircraft study, noting that HNO_3 constitutes $> 85\%$ of NO_y in lower polar vortex. The differences can be attributed to the coarse vertical resolution (6–10 km) of the aircraft data and the different sampling in the inhomogeneously denitrified polar vortex. The estimated maximum relative NO_y reduction in early March is slightly smaller than the 70% predicted by modeling [von Hobe *et al.*, 2006] for early March. On the other hand, the maxima of individual observations and relative reductions are comparable to analyses using balloon-borne and airborne in-situ measurements in previous cold Arctic winters such as the winter 1994/1995 [Sugita *et al.*, 1998; Waibel *et al.*, 1999] and the winter 1999/2000 [Popp *et al.*, 2001].

[18] EOS MLS data also indicate significant Arctic denitrification for this winter [Schoeberl *et al.*, 2006]. The maximum denitrification reached 18 ppbv in January and in February, and reached 14 ppbv in early March at ~ 450 K. These values are ~ 6 – 8 ppbv larger than the ACE-FTS maxima. The difference can, in part, be attributed to the fact that EOS MLS HNO_3 mixing ratios are up to 30% larger than those from ACE-FTS [Froidevaux *et al.*, 2006]. However, we note that comparisons of MIPAS/ENVISAT HNO_3 measurements with Odin/SMR, ACE-FTS, and balloon measurements suggest good agreement with differences generally $< 15\%$ below 30 km [Mencaraglia *et al.*, 2006; Wang *et al.*, 2006].

[19] Although the temperature in the Arctic polar vortex was particularly low in the winter 2004/2005, the air with temperature below the threshold temperature for ice formation only constituted a very small fraction of the vortex

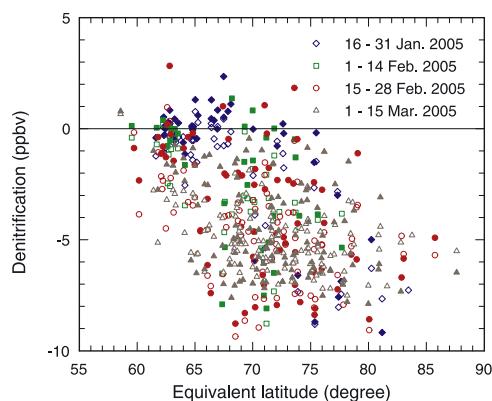


Figure 5. NO_y reductions between 430 K–460 K versus equivalent latitudes. The open and closed symbols are results from the correlation method and the artificial tracer method, respectively.

volume and was not continuously present [Kleinböhl *et al.*, 2005; Jiménez *et al.*, 2006]. As a result, no noticeable dehydration was observed by ACE-FTS, and EOS MLS only observed one single dehydration event of up to 0.5 ppmv [Jiménez *et al.*, 2006]. These observations are similar to those for previous cold Arctic winters [World Meteorological Organization, 2003], and are consistent with the simulations showing that Arctic denitrification is caused by the sedimentation of large HNO_3 -containing particles rather than by the sedimentation of ice particles [Carslaw *et al.*, 2002].

3. Summary

[20] Significant Arctic denitrification was observed by ACE-FTS in the polar vortex between 400 K–550 K (~ 15 km– 23 km) in the winter 2004/2005 although no obvious dehydration was measured. ACE-FTS observed denitrified air from the middle of January to the middle of March. The denitrification, analyzed by a correlation method and an artificial tracer method, occurred inhomogeneously inside the vortex. Before the vortex breakup at the middle of March, both methods give a maximum local denitrification of 8 ± 0.8 ppbv at ~ 450 K (~ 17 km) although the correlation method produces larger denitrification estimates than the artificial tracer method for some individual observations because of episodic mixing [Manney *et al.*, 2006; Jin *et al.*, 2006; Schoeberl *et al.*, 2006]. The denitrification intensified by the end of February and the maximum average reached ~ 4.4 – 5.0 ppbv before the vortex broke up. Sedimentation and evaporation of PSC particles appear to have led to an NO_y increase below ~ 15 km. The magnitude of the denitrification observed by ACE-FTS is smaller than estimates from EOS MLS observations, but is consistent with other observations and modeling results for this winter and comparable to the denitrification in previous cold winters.

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