NO_x descent in the Arctic middle atmosphere in early 2009

C. E. Randall,^{1,2} V. L. Harvey,¹ D. E. Siskind,³ J. France,¹ P. F. Bernath,⁴ C. D. Boone,⁵ and K. A. Walker⁶

Received 22 June 2009; revised 13 August 2009; accepted 18 August 2009; published 25 September 2009.

[1] Measurements by the Atmospheric Chemistry Experiment show that the amount of NO_x (NO + NO₂) produced by energetic particle precipitation (EPP) that descended from the Arctic mesosphere and lower thermosphere into the stratosphere in early 2009 was up to \sim 50 times higher than average in 2005, 2007 and 2008. This is of note because the level of EPP in the preceding months was very low, suggesting that excess production of NO_x was not the cause of the enhancements. Rather, the enhancements are attributed to unusually strong descent in the middle atmosphere. This is the third time on record that extraordinary meteorology contributed to descent of excess NO_x. The results confirm that EPP impacts on the middle atmosphere can be large even in the absence of exceptional EPP, and highlight the need to continually measure NO_x throughout the polar region from the stratosphere to the lower thermosphere. Citation: Randall, C. E., V. L. Harvey, D. E. Siskind, J. France, P. F. Bernath, C. D. Boone, and K. A. Walker (2009), NO_x descent in the Arctic middle atmosphere in early 2009, Geophys. Res. Lett., 36, L18811, doi:10.1029/2009GL039706.

1. Introduction

[2] Energetic Particle Precipitation (EPP) refers to the process by which energetic electrons and protons impinge on the Earth's atmosphere. One consequence of EPP is production of NO_x (NO + NO₂) in the mesosphere and lower thermosphere (MLT), and occasionally in the stratosphere. NO_x has a lifetime of days to weeks in the mesosphere, with longest lifetimes in the polar winter. If dynamical conditions are favorable, the NO_x produced by EPP (hereafter referred to as EPP-NO_x) can thus be transported downward into the stratosphere during the polar winter. This process is referred to as the EPP indirect effect (EPP IE) [*Randall et al.*, 2007]. Once in the stratosphere, EPP-NO_x has a lifetime on the order of months or longer, and catalytically destroys O₃.

[3] Observational evidence for the EPP IE has been given by a number of authors [e.g., *Funke et al.*, 2005; *Jackman et al.*, 2008; *Siskind et al.*, 2000; *Randall et al.*, 1998, 2001,

Copyright 2009 by the American Geophysical Union. 0094-8276/09/2009GL039706

2007]. Randall et al. [2007] showed that interannual variability in the southern hemisphere (SH) EPP IE correlates very well with the level of EPP itself. They suggested that this was due to the fact that interannual variability in SH dynamics is small, so interannual variability in the EPP IE is controlled primarily by changes in EPP-NO_x production, not transport. In the northern hemisphere (NH), however, dynamical variability is high, so the EPP IE does not correlate well with variations in the production of EPP-NO_x as inferred from the Ap index or EPP hemispheric power. The purpose of this paper is to describe observations that show that for the second time in four years, unprecedented meteorological conditions led to very large amounts of descending EPP-NO_x in the NH, even though the level of EPP was well below average. This result is significant in that it confirms that the production of EPP-NO_x is potentially an important element in O₃ depletion regardless of the level of geomagnetic activity, and that EPP influences can often be enhanced by favorable serendipity between space weather and meteorology.

2. Results

[4] Figure 1 compares NO_x descending from the MLT during the Arctic winters from 2004–2009. NO_x mixing ratios are from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) solar occultation instrument [*Bernath et al.*, 2005; see also *Sica et al.*, 2008]. ACE-FTS only samples a single latitude on any given day, which is shown in Figure 1 (top); measurement latitudes nearly repeat from year to year. Figure 1 shows prominent tongues of NO_x descending from the MLT into the Arctic stratosphere in 2004, 2006, and 2009. Because the only significant source of MLT NO_x at the ACE latitudes in winter is EPP, these tongues can unambiguously be identified as EPP- NO_x .

[5] The Arctic EPP IE in 2004 was larger than ever before observed in either the NH or SH; NO_x mixing ratios at 40 km were up to a factor of 4 higher than nominal at some locations [Randall et al., 2005]. Unusual meteorology was a key factor in these enhancements, including a remarkable vortex recovery after a mid-winter sudden stratospheric warming (SSW) and enhanced adiabatic descent in the mesosphere [Hauchecorne et al., 2007; Jin et al., 2005; Manney et al., 2005, 2008a]. Exceptional EPP levels in Oct-Dec 2003 have also been suggested as contributing, although Clilverd et al. [2006] concluded that this was not necessary. Extraordinary meteorology once again prevailed during the Arctic 2006 winter, and was responsible for the tongue of descending NO_x in the MLT that is so prominent in Figure 1 for that year [Manney et al., 2008a, 2008b; Randall et al., 2006; Siskind et al., 2007]. There was only minimal geomagnetic activity in late 2005 and early 2006:

¹Laboratory for Atmospheric and Space Physics, University of Colorado, Boulder, Colorado, USA.

²Also at Department of Atmospheric and Oceanic Sciences, University of Colorado, Boulder, Colorado, USA.

³Space Science Division, Naval Research Laboratory, Washington, D. C., USA.

⁴Department of Chemistry, University of York, Heslington, UK.

⁵Department of Chemistry, University of Waterloo, Waterloo, Ontario, Canada.

⁶Department of Physics, University of Toronto, Toronto, Ontario, Canada.



Figure 1. Zonal average ACE-FTS NO_x (color) in the NH from 1 Jan through 31 Mar of 2004–2009. The white contour denotes CO = 2.0 ppmv; CO increases with increasing altitude. Measurement latitudes (black dots) are shown in the top panel. White regions indicate missing data; vertical black dotted lines denote 1 Feb and 1 Mar. ACE data are unavailable prior to 21 Feb 2004.

Auroral power was well below the average since 1978, the geomagnetic Ap index was lower than it had been since 1988, and there was no evidence of enhanced fluxes of high energy protons or relativistic electrons. Thus, unlike in 2004, the late winter/spring NO_x enhancements of 2006 were more clearly attributed to the dynamical situation.

[6] Measurements of the 2008–2009 Arctic winter show that for the third time in six years, polar winter meteorology was remarkably different from the norm prior to 2004. Manney et al. [2009] describe a major SSW in January 2009 that was the strongest and most prolonged on record; upon recovery, the stratopause reformed in early February at 80°N at an altitude of 80 km, which is arguably more typical of a mesopause altitude than a stratopause altitude. The response of the MLT to such a remarkable warming includes enhanced descent in the polar MLT, and thus transport of EPP-NO_x down toward the stratosphere. This is obvious in Figure 1 (bottom), which shows the prominent tongue of descending NO_x , similar to the tongues in 2004 and 2006. Like in 2006, geomagnetic activity in late 2008 and early 2009 was very low: The average Ap index from Oct 2008 through Feb 2009 was just 4.8, and never exceeded 34 (ftp://ftp.ngdc.noaa.gov/STP/GEOMAGNETIC DATA/ INDICES/KP AP/).

[7] Although not shown, plots of SH ACE-FTS NO_x since 2003 are very similar to the Arctic plots for 2005, 2007, and 2008; further, the Arctic 2006 NO_x mixing ratio enhancements were substantially larger than ever observed in the Antarctic back to 1992 [*Randall et al.*, 2006]. This is of note

because the SH polar vortex is generally larger, more stable, and stronger than the NH polar vortex; it is thus more likely to confine EPP-NO_x to the polar region and not dilute it by mixing with mid-latitude air. We suggest, therefore, that for 2004, 2006, and 2009, the descent rates in the MLT are more directly important for controlling the EPP IE than the vortex structure itself. The white contours in Figure 1 indicate the 2.0 ppmv level of CO, a wintertime tracer of atmospheric motion; that they follow the NO_{x} "tongue" contours until early March confirms that NO_x is indeed descending. In early March photochemistry begins to perturb both CO and NO_x, resulting, e.g., in the sharp decrease in NO_x near 70 km. In the Arctic in 2004, 2006, and 2009, the 2.0 ppmv CO contour reached altitudes as low as 45-50 km in early Mar. In all other winters, in either hemisphere, it never reached lower than ~ 60 km, consistent with less descent in these winters.

[8] Figure 2 quantifies the amount of EPP-NO_x reaching an altitude of 55 km in the Arctic winters of 2004–2009 by correlating CH₄ and NO_x; an anti-correlation is indicative of descending EPP-NO_x [e.g., Randall et al., 2007; Siskind and Russell, 1996]. Much more EPP-NOx reaches 55 km in 2004, 2006, and 2009 than in 2005, 2007, or 2008, with highest NO_x corresponding to lowest CH₄, indicating that it descended from higher altitudes. Although not shown, NH EPP-NO_x mixing ratios at 55 km range from 2 to 20 (10) times higher in 2004, 2006, and 2009 than the highest NO_x mixing ratios observed in other years in the NH (SH) back to 1992. Figure 2 also shows the relationship between CO and CH₄ at 55 km. In the absence of mixing, these tracers should show a tight correlation, with minimum (maximum) values of CH₄ (CO) indicating the highest originating altitude. Maximum CO mixing ratios in 2004, 2006, and 2009 are nearly 3-6 times higher than maximum CO mixing ratios in 2005, 2007, and 2008, indicating much more descent in the mesosphere. This is consistent with the conclusions of Winick et al. [2009], who inferred enhanced descent in the mesosphere in 2004 and 2006 based on analysis of mesospheric OH airglow observations. The tighter correlation in 2004 indicates less mixing, which might partially explain the higher 2004 NO_x values in the top panels.



Figure 2. (top) ACE-FTS CH_4 vs. NO_x in Jan–Mar of the winters shown, at an altitude of 55 km and for latitudes poleward of 50N. For clarity, only every third point is plotted. (bottom) Same as Figure 2 (top), but for CO instead of NO_x .



Figure 3. Ratio of NO_x in 2004, 2006, and 2009 to the average NO_x observed in years 2005, 2007, and 2008, calculated from the data in Figure 1. For guidance, dotted vertical lines indicate 1 Feb and 1 Mar. White diamonds indicate the altitude of the stratopause at the ACE locations.

[9] A more extensive comparison of the relative NO_x enhancements during 2004, 2006, and 2009 is given in Figure 3, which shows the ratio of NO_x observed by ACE in each of these three years to the average NO_x observed at the corresponding latitude, altitude, and time in 2005, 2007, and 2008. The overall morphology in all three winters is similar, but there are significant differences in the details. Largest enhancements in all three years, which exceeded factors of 100, were observed in 2004 near altitudes of 50–55 km. Note that no ACE data were available prior to 21 Feb 2004, so the results are inconclusive regarding 2004 enhancements at this time. Comparing 2006 and 2009, the years with very low EPP, enhancements in 2009 were larger overall, reaching factors of more than 50 (45) at altitudes of 60-65 km (55-60 km) in 2009 (2006). On the other hand, the lowest part of the tongue of $EPP-NO_x$ in 2004 and 2006 extended farther in time and lower in altitude than in 2009. Consistent with this, NO_x/CH₄ correlations show little evidence of the EPP IE at 45 km in 2009, whereas it is quite prominent at 45 km in 2004 and 2006 (not shown).

3. Discussion and Summary

[10] The 2004 and 2006 winters have already been linked to unusual meteorological conditions in the middle atmosphere, specifically with regard to the propagation of planetary and gravity waves [Hauchecorne et al., 2007; Manney et al., 2008a, 2008b; Sathishkumar and Sridharan, 2009; Siskind et al., 2007; Winick et al., 2009]. A complete discussion of the 2009 meteorology is beyond the scope of this paper; however, Manney et al. [2009] have already pointed out certain similarities, such as an unusually strong and persistent SSW that occurred in January and was followed by the reformation of a strong upper stratospheric vortex and displaced (elevated in altitude) stratopause. One difference was that the 2006 SSW was primarily dominated by a planetary wave 1, whereas the 2009 event was dominated by wave 2. Here we highlight how key features in the descending NO_x are linked to key features in the temperature, and by implication, dynamical fields.

[11] Superimposed on the NO_x enhancements in Figure 3 is the zonal average stratopause height, inferred from

temperature maxima measured by ACE between 15 and 90 km. The polar winter stratopause height is typically near 50 km; Figure 3 shows that in these three years, the stratopause height at the ACE measurement latitudes reached well above this altitude, to \sim 75 km in 2009, indicative of enhanced descent in the mesosphere. The onset of NO_x enhancements coincided with formation of the displaced stratopause in both 2006 and 2009. Because of the lack of ACE NO_x measurements prior to 21 Feb 2004, it is impossible to draw conclusions for that year. Nevertheless, the data are strongly suggestive of similar behavior in 2004; temperatures from the Sounding of the Atmosphere using Broadband Radiometry (SABER) indicate stratopause heights as high as 80 km by mid-Jan in 2004 (not shown). Year 2006 shows a strongly displaced stratopause in early Feb that then drops back to ~ 50 km before becoming displaced again; accordingly, NO_x enhancements appear first in early Feb near 70-80 km, before dissipating and then appearing consistently from mid-Feb onward. Similar behavior occurs in 2009, although it is not as dramatic. Thus the evidence is compelling that the NO_x enhancements were caused by increased descent. Finally, it is not surprising that such large EPP-NO_x enhancements have not yet been observed in the SH, since recovery from major SSWs, which are not known to occur in the middle of SH winters, apparently triggered the enhanced descent in the NH MLT. It is ironic, in fact, that the characteristic that makes the SH favorable for the EPP IE – a generally strong and stable vortex that promotes confinement of EPP-NO_x in the polar region - is the same characteristic that prevents the SH from exhibiting exceptionally large EPP-NOx mixing ratio enhancements such as seen in 2004, 2006, and 2009.

[12] To summarize, we have shown that 2009 was the second year on record, 2006 being the first, in which exceptional meteorology in the Arctic stratosphere and MLT led to significant descent of EPP-NO_x in the polar region, even though geomagnetic activity was significantly lower than average. Thus the extraordinary EPP effects on the atmosphere in 2006 were not a one-time occurrence. Similar meteorology was observed in 2004, but was preceded by large solar storms in late 2003. Of these three years, and at the ACE measurement locations, EPP-NO_x enhancements were largest in 2004; enhancements were larger in 2009 than in 2006 above about 60 km, but descended farther in 2006 than in 2009. Significant effects on O_3 are not expected (nor observed) in 2009 because the NO_x mixing ratios did not increase substantially at the altitudes where NOx is most effective at catalytic O3 destruction, from about 22-45 km.

[13] It is important to note the limitations on these conclusions, however, which include the lack of data in the polar night, daily varying and sparse latitude sampling, and the lack of high-latitude data from late March to early May. As an indication of these limitations, Figure 4 shows Arctic 2009 temperatures from the SABER instrument. Note the substantial differences in character between the different latitude bands. As shown in Figure 1, ACE measurement latitudes vary by ~30 degrees from early Jan through Mar, and thus do not always correspond to the most extreme temperatures in the polar region. For example, in early Feb ACE is sampling ~65°N, where there is no indication of a displaced stratopause; at 80–85°N in early Feb, however,



Figure 4. Zonal mean SABER temperatures from 10 Jan to 12 Mar in 2009, in the latitude bands given in each plot. Vertical dotted lines denote 1 Feb and 1 Mar. Black dots denote the stratopause, defined as the maximum temperature from 15-100 km.

the stratopause had already started to reform near 80 km. ACE thus does not sample locations with the most extreme meteorological conditions. In addition, ACE measurements are not made in the polar region in April, when descending NO_x is most likely to reach altitudes where it is the main catalyst of O_3 destruction; for instance, the April 2004 O_3 depletions of up to 60% that were observed by HALOE [*Randall et al.*, 2005] were not seen by ACE. These limitations emphasize the need for continual measurements of NO_x throughout the polar region, from the stratosphere to the lower thermosphere.

[14] The observation of so much descending MLT NO_x in both 2006 and 2009, when EPP was at very low levels, supports the suggestion of *Clilverd et al.* [2006] that the late winter/spring NO_x enhancements that led to the extraordinary O_3 losses in 2004 were in fact not connected to the solar storms of 2003. Instead, they were more likely due primarily to the unusual meteorology, with a contribution from moderate levels of EPP in Jan–Feb 2004. Further research is necessary to determine in detail the differences in EPP-NO_x production and atmospheric meteorology that led to the different magnitudes of the observed EPP IE in the three years, and to understand whether the unusual meteorology can be linked to a specific cause.

[15] Acknowledgments. This work was supported by NASA grants NNX06AC05G and NNX06AE27G. The Atmospheric Chemistry Experiment (ACE), also known as SCISAT, is a Canadian-led mission mainly supported by the Canadian Space Agency and the Natural Sciences and Engineering Research Council of Canada. We thank Gloria Manney for providing us with her submitted manuscript on the 2009 winter and Tom Marshall for input on SABER temperature validation.

References

- Bernath, P. F., et al. (2005), Atmospheric Chemistry Experiment (ACE): Mission overview, *Geophys. Res. Lett.*, 32, L15S01, doi:10.1029/ 2005GL022386.
- Clilverd, M. A., A. Seppälä, C. J. Rodger, P. T. Verronen, and N. R. Thomson (2006), Ionospheric evidence of thermosphere-to-stratosphere descent of polar NO_x, *Geophys. Res. Lett.*, 33, L19811, doi:10.1029/ 2006GL026727.
- Funke, B., M. López-Puertas, S. Gil-López, T. von Clarmann, G. P. Stiller, H. Fischer, and S. Kellmann (2005), Downward transport of upper atmospheric NO_x into the polar stratosphere and lower mesosphere during the

Antarctic 2003 and Arctic 2002/2003 winters, J. Geophys. Res., 110, D24308, doi:10.1029/2005JD006463.

- Hauchecorne, A., J.-L. Bertaux, F. Dalaudier, J. M. Russell III, M. G. Mlynczak, E. Kyrola, and D. Fussen (2007), Large increase of NO₂ in the north polar mesosphere in January–February 2004: Evidence of a dynamical origin from GOMOS/ENVISAT and SABER/TIMED data, *Geophys. Res. Lett.*, 34, L03810, doi:10.1029/2006GL027628.
- Jackman, C. H., et al. (2008), Short- and medium-term atmospheric constituent effects of very large solar proton events, *Atmos. Chem. Phys.*, 8, 765–785.
- Jin, J. J., et al. (2005), Co-located ACE-FTS and Odin-SMR stratosphericmesospheric CO 2004 measurements and comparison with a GCM, *Geophys. Res. Lett.*, 32, L15S03, doi:10.1029/2005GL022433.
- Manney, G. L., K. Krüger, J. Sabutis, S. A. Sena, and S. Pawson (2005), The remarkable 2003–2004 winter and other recent warm winters in the Arctic stratosphere since the late 1990s, *J. Geophys. Res.*, 110, D04107, doi:10.1029/2004JD005367.
- Manney, G. L., et al. (2008a), The high Arctic in extreme winters: Vortex, temperature, and MLS and ACE-FTS trace gas evolution, *Atmos. Chem. Phys.*, 8, 505–522.
- Manney, G. L., et al. (2008b), The evolution of the stratopause during the 2006 major warming: Satellite data and assimilated meteorological analyses, J. Geophys. Res., 113, D11115, doi:10.1029/2007JD009097.
- Manney, G. L., M. J. Schwartz, K. Krüger, M. L. Santee, S. Pawson, J. N. Lee, W. H. Daffer, R. A. Fuller, and N. J. Livesey (2009), Aura Microwave Limb Sounder observations of dynamics and transport during the record-breaking 2009 Arctic stratospheric major warming, *Geophys. Res. Lett.*, 36, L12815, doi:10.1029/2009GL038586.
- Randall, C. E., D. W. Rusch, R. M. Bevilacqua, K. W. Hoppel, and J. D. Lumpe (1998), Polar Ozone and Aerosol Measurement (POAM) II stratospheric NO₂, 1993–1996, J. Geophys. Res., 103, 28,361–28,371, doi:10.1029/98JD02092.
- Randall, C. E., D. E. Siskind, and R. M. Bevilacqua (2001), Stratospheric NO_x enhancements in the Southern Hemisphere vortex in winter/spring of 2000, *Geophys. Res. Lett.*, 28, 2385–2388, doi:10.1029/ 2000GL012746.
- Randall, C. E., et al. (2005), Stratospheric effects of energetic particle precipitation in 2003-2004, *Geophys. Res. Lett.*, 32, L05802, doi:10.1029/2004GL022003.
- Randall, C. E., V. L. Harvey, C. S. Singleton, P. F. Bernath, C. D. Boone, and J. U. Kozyra (2006), Enhanced NO_x in 2006 linked to strong upper stratospheric Arctic vortex, *Geophys. Res. Lett.*, 33, L18811, doi:10.1029/2006GL027160.
- Randall, C. E., et al. (2007), Energetic particle precipitation effects on the Southern Hemisphere stratosphere in 1992–2005, *J. Geophys. Res.*, 112, D08308, doi:10.1029/2006JD007696.
- Sathishkumar, S., and S. Sridharan (2009), Planetary and gravity waves in the mesosphere and lower thermosphere region over Tirunelveli (8.7°N, 77.8°E) during stratospheric warming events, *Geophys. Res. Lett.*, 36, L07806, doi:10.1029/2008GL037081.
- Sica, R. J., et al. (2008), Validation results for the Atmospheric Chemistry Experiment (ACE), Atmos. Chem. Phys., 8, 35–62.
- Siskind, D. E., and J. M. Russell III (1996), Coupling between middle and upper atmospheric NO: Constraints from HALOE observations, *Geophys. Res. Lett.*, 23, 137–140, doi:10.1029/95GL03782.
- Siskind, D. E., G. E. Nedoluha, C. E. Randall, M. Fromm, and J. M. Russell III (2000), An assessment of Southern Hemisphere stratospheric NO_x enhancements due to transport from the upper atmosphere, *Geophys. Res. Lett.*, 27, 329–332, doi:10.1029/1999GL010940.
- Siskind, D. E., S. D. Eckermann, L. Coy, J. P. McCormack, and C. E. Randall (2007), On recent interannual variability of the Arctic winter mesosphere: Implications for tracer descent, *Geophys. Res. Lett.*, 34, L09806, doi:10.1029/2007GL029293.
- Winick, J. R., et al. (2009), OH layer characteristics during unusual boreal winters of 2004 and 2006, *J. Geophys. Res.*, 114, A02303, doi:10.1029/ 2008JA013688.

K. A. Walker, Department of Physics, University of Toronto, 60 St. George Street, Toronto, ON M5S 1A7, Canada.

P. F. Bernath, Department of Chemistry, University of York, Heslington YO10 5DD, UK.

C. D. Boone, Department of Chemistry, University of Waterloo, 200 University Avenue West, Waterloo, ON N2L 3G1, Canada.

J. France, V. L. Harvey, and C. E. Randall, Laboratory for Atmospheric and Space Physics, University of Colorado, Campus Box 392, Boulder, CO 80309-0392, USA. (cora.randall@lasp.colorado.edu)

D. E. Siskind, Space Science Division, Naval Research Laboratory, Code 7640, 4555 Overlook Avenue SW, Washington, DC 20375, USA.