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Enhanced NO_x in 2006 linked to strong upper stratospheric Arctic vortex

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[1] Measurements from the Atmospheric Chemistry Experiment show pronounced downward transport of NO_x ($NO+NO_2$) to the Arctic stratosphere in Feb–Mar 2006. NO_x mixing ratios in the upper stratosphere were 3–6 times larger than observed previously in either the Arctic or Antarctic, aside from the extraordinary winter of 2003–2004. There was only minimal geomagnetic activity in late 2005 and early 2006, however, suggesting that NO_x produced via energetic particle precipitation was not significantly elevated. On the other hand, the Arctic polar vortex at stratopause altitudes in Feb 2006 was exceptionally strong, implying greater confinement of air in the polar night. Carbon monoxide data also indicate enhanced confined descent of air from the mesosphere. These results confirm that impacts of EPP on the atmosphere are modulated by meteorological conditions; this has implications for understanding interannual variability and trends in stratospheric NO_x and ozone.

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1. Introduction

[2] One of the main loss mechanisms of stratospheric O_3 is the catalytic NO_x cycle. The main source of global stratospheric NO_x ($NO+NO_2$) is oxidation of N_2O that is transported up from the troposphere. A secondary source is energetic particle precipitation (EPP). The higher the energy of the precipitating particle, the lower in altitude the energy is deposited [*Thorne*, 1980], and the more rare its occurrence. The highest energy electrons ($E > 300$ keV) and protons ($E > 30$ MeV) can produce NO directly in the stratosphere, but precipitate only during periods of strong geomagnetic disturbances such as solar proton events (SPEs). Electrons (protons) with energies lower than $\sim$30 keV (1 MeV), such as those that form the aurora, precipitate routinely and produce NO in the thermosphere.

[3] It is now well documented that NO produced in the mesosphere or thermosphere can descend to the stratosphere where it participates in catalytic O_3 destruction [e.g., *Callis et al.*, 2001; *Randall et al.*, 1998, 2001, 2005; *Siskind et al.*, 2000]. This occurs primarily during the polar winter because NO_x has a photochemical lifetime of days or less in the sunlit upper mesosphere and above. We refer to NO_x produced via EPP as EPP-NO_x, and to the process of EPP-NO_x production followed by descent to the stratosphere as the EPP indirect effect (IE; as opposed to direct production in the stratosphere itself). Implications of the EPP IE for global climate are just beginning to be investigated. Using a coupled chemistry climate model, *Rozanov et al.* [2005] suggest that increased O_3 loss from EPP-NO_x can lead to cooling of up to 2 K in the high latitude middle stratosphere, and possibly to detectable changes in the surface air temperature.

[4] The long term impact of the EPP IE on stratospheric O_3 is poorly quantified because there are few measurements of NO_x throughout the middle and upper atmosphere during the polar night. The only long-term satellite measurements of polar NO_x come from solar occultation (SO) instruments, which require sunlight and thus do not view the region of maximum EPP-NO_x. Nighttime NO_x can be retrieved from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) [*Funke et al.*, 2005], but this data set only began in 2002. Using SO data from 1992 through 2005, C. E. Randall et al. (Energetic particle precipitation effects on the southern hemisphere stratosphere in 1992–2005, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Randall et al., submitted manuscript, 2006) estimate that the EPP IE contributes up to 20% of the southern hemisphere (SH) polar stratospheric NO_x budget; interannual variability is strongly correlated with low and medium energy EPP.

[5] The EPP IE in the northern hemisphere (NH) was first inferred from measurements in 1979 [*Russell et al.*, 1984]. Until the winter of 2003–2004, however, the SO data sets used to investigate long term EPP impacts showed much less evidence for the EPP IE in the NH than in the SH. One possible explanation for this is the more transient nature of the NH polar vortex. The EPP IE is facilitated by confinement of descending air within the polar vortex, hindering horizontal transport to lower latitudes where the EPP-NO_x would be more efficiently dissociated. Therefore, a stronger, more stable polar vortex, as is generally observed in the SH, is expected to lead to more efficient transport of EPP-NO_x to the stratosphere.

[6] The NH winter of 2003–2004 was unusual in two ways. First, large solar storms occurred during late Oct and Nov of 2003, leading to substantial EPP [e.g., *Gopalswamy et al.*, 2005, and references therein]. Second, the upper stratospheric polar vortex in Feb–Mar 2004 was the strongest on record at that time [*Manney et al.*, 2005]. One or both of these characteristics led to observations of unprecedented

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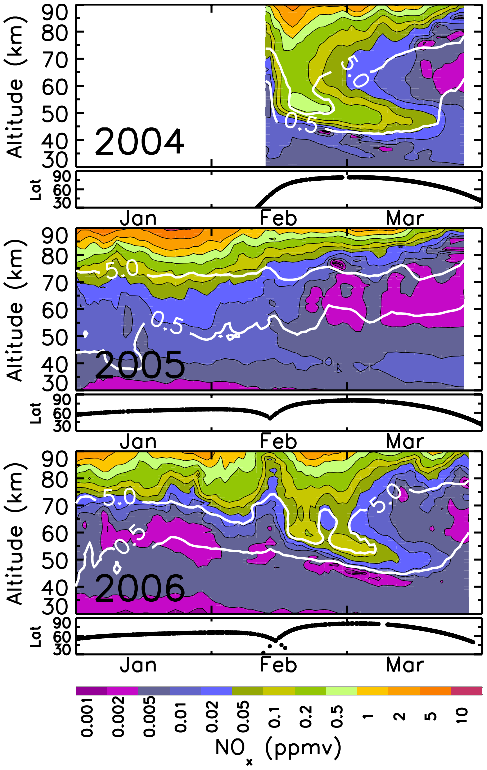

Figure 1. ACE NO_x mixing ratios poleward of 50°N in (top) 2004, (center) 2005, and (bottom) 2006. The bottom plot shows the ACE occultation latitudes in the respective years. White contours show ACE CO (ppmv).

stratospheric NO_x enhancements and O_3 reductions due to the EPP IE, but distinguishing the relative importance of the high particle activity and strong vortex was not possible given the available measurements [e.g., *López-Puertas et al.*, 2005; *Natarajan et al.*, 2004; *Randall et al.*, 2005].

[7] As described below, energetic particle activity in the 2005–2006 Arctic winter was low, but the upper stratospheric vortex was once again extraordinarily strong. Thus even though energetic particle activity was not enhanced, observations show enhanced transport of EPP-NO_x to the stratosphere in Feb–Mar 2006, illuminating the important role that meteorology plays in determining the EPP IE.

2. ACE-FTS Data

[8] Measurements from the SO Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS, hereafter referred to as ACE) [*Bernath et al.*, 2005] are used to document EPP effects on the atmosphere during the Arctic winters of 2003–2004 to 2005–2006. ACE was launched 12 Aug 2003 into a 74° inclination orbit; each day up to 15 measurements are made around a circle of approximately constant latitude in each hemisphere. Latitudes vary in time, with a maximum range of about 85°S to 85°N that is sampled near equinox over a period of about one month. Latitudes poleward of 50°N are sampled continuously from Jan throughout most of Mar (Figure 1).

[9] The ACE instrument operates with 0.02 cm^{-1} spectral resolution in the infrared from 2.2 to 13.3 μm, enabling retrieval of many constituents from 5–100 km with a vertical resolution of ~4 km [*Bernath et al.*, 2005]. The data used here correspond to ACE version 2.2, described in detail by *Boone et al.* [2005]; we use near real time data. Preliminary validation of version 1.0 ACE constituent retrievals has been presented in a special section of *Geophysical Research Letters*, *32*(15), 2005. *McHugh et al.* [2005] show that throughout the range of altitudes over which they are retrieved, ACE NO and NO_2 profiles are qualitatively similar in shape to those retrieved from the Halogen Occultation Experiment (HALOE); ACE NO is lower than HALOE by 10–20% in the stratosphere. A comprehensive analysis of ACE CO measurements, which we use below to identify vertical transport, is presented by *Clerbaux et al.* [2005]; *Jin et al.* [2005] found excellent agreement between ACE CO and correlative data. *McHugh et al.* [2005] found excellent agreement between ACE and HALOE CH_4, another molecule used as a transport diagnostic.

3. Results

[10] Figure 1 shows ACE NO_x mixing ratios poleward of 50°N from Jan–Mar of 2004–2006 (ACE data became available on a regular basis beginning in Feb 2004). CO isopleths are used to estimate vertical motion [*Clerbaux et al.*, 2005]. They show significant descent from the mesosphere to the stratosphere in 2004 and 2006, but not 2005. The tongue of enhanced NO_x descending from the mesosphere into the stratosphere in Feb–Mar 2004, which closely follows the CO contours, is a clear signature of the unprecedented EPP IE that occurred that year [*Randall et al.*, 2005; *Rinsland et al.*, 2005]. A very similar signature is seen in Feb–Mar 2006, although NO_x is not as large as in 2004. Because the only significant polar winter source of NO_x above the stratopause is EPP, this tongue of enhanced NO_x in 2006 can unambiguously be ascribed to the EPP IE. NO_x is larger in Jan 2005 than in Jan 2006; but in Feb–Mar the CO contours suggest weaker descent, explaining the lack of enhanced NO_x at this time. In Mar increasing sunlight dissociates NO_x, leading to low NO_x in the lower mesosphere in all three years.

[11] Historically, the EPP IE has been inferred from the correlation between CH_4 and NO_x, as in Figure 2. Here all individual HALOE (which operated from 1991–2005) and ACE NO_x measurements from Jan–Apr in the NH, and from Jul–Oct in the SH, are plotted vs. CH_4 at 48 km. High NO_x correlating with low CH_4 is indicative of the EPP IE, since this is a signature of NO_x-rich mesospheric air descending to the stratosphere [*Siskind et al.*, 2000]. This signature is obvious in the NH ACE data for 2006, with NO_x values greater than 100 ppbv at the lowest values of CH_4. Remarkably, maximum mixing ratios in 2006 are more than 6 times higher than had been observed previously by HALOE, except in 2004, or than observed by ACE in 2005. Data from the NH 2003–2004 winter has been omitted from Figure 2 for clarity; as described previously, the EPP IE led to unprecedented NO_x enhancements in the upper stratosphere and lower mesosphere in Mar–Apr 2004 [*Randall et al.*, 2005; *Rinsland et al.*, 2005], with maximum mixing ratios greater than 600 ppbv at 48 km.

[12] In contrast to the NH, for which HALOE data show only minimal indication of the EPP IE prior to 2004, the EPP IE is routinely observed in the SH (Randall et al., submitted manuscript, 2006). This is reflected in the elevated NO_x

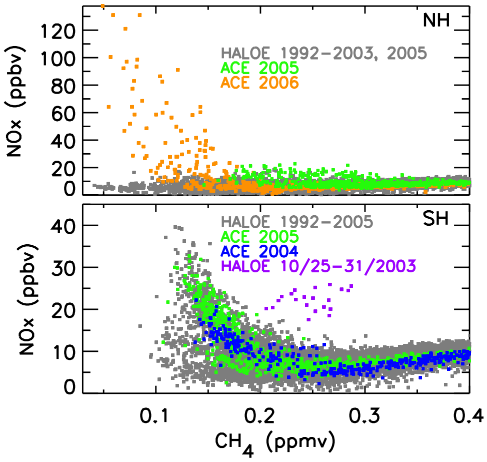

Figure 2. All NO_x measurements at 48 km (top) in the NH from Jan–Apr and (bottom) in the SH from Jul–Oct from HALOE (gray) and ACE for their respective winters of operation. Data from Jan–Apr 2004 is omitted in the NH for clarity. Colors refer to ACE data in 2004 (blue, SH), in 2005 (green, NH and SH), and in 2006 (orange, NH), and SH HALOE data from 25–31 Oct 2003 (purple). Note the different vertical axis scales.

corresponding to CH_4< 0.2 ppmv in the SH plot of Figure 2. The elevated SH NO_x values in October 2003 show direct production of NO in the upper stratosphere from SPEs, and are not indicative of the EPP IE. Even though the EPP IE is obvious in many SH winters, maximum SH mixing ratios observed by HALOE since 1992, and by ACE in 2004–2005, are only about 30% as high as in the NH ACE data in 2006.

[13] Unlike ACE, HALOE only skirted the polar region during winter, so it did not sample the largest EPP-NO_x enhancements. For instance, in 2004 HALOE did not sample poleward of 50°N from Feb 18 through Apr 2, missing the most prominent enhancements seen by ACE (Figure 1). Because of the different sampling, it is not possible to state definitively that the EPP IE in 2006 was larger than in any year other than 2004. It is remarkable, however, that HALOE data from 1992–2005 show maximum NH EPP-NO_x (~15 ppbv) that was comparable only to average SH EPP-NO_x, whereas ACE data from 2004–2006 show maximum NH EPP-NO_x in 2006 that was significantly larger than maximum SH EPP-NO_x.

[14] The large NH EPP IE in 2006 cannot be explained by enhanced EPP. The Ap index, a measure of planetary-scale geomagnetic activity that correlates well with auroral EPP, is shown in Figure 3 for the months of Oct–Feb for the winters of 2003–2004 to 2005–2006. As mentioned above, large solar storms led to substantial geomagnetic activity in Oct–Dec 2003, as reflected in the high Ap index values. The Ap index spikes in Nov 2004 and Jan 2005 occur in association with SPEs. Activity for Oct 2005 through Feb 2006 was significantly lower on average than in either of the previous two years. As also shown in Figure 3, the average NH auroral power over the Oct–Feb period in 2005–2006 has not been this low since 1988, and is well below the average since 1978. There is also no evidence for substantially elevated high energy protons or relativistic electrons during this time period. High speed solar streams have been associated with increases in EPP [*Kozyra et al.*, 2006], and a strong recurrent stream appeared during Oct 2005–Feb 2006. Because of its orientation, however, it produced only weak magnetic activity. Geomagnetic activity was elevated in Sep 2005, but because of photolysis, the descending tongue of NO_x-rich air in Feb 2006 cannot be attributed to NO_x created in the upper mesosphere or thermosphere as early as Sep.

[15] Figures 1–3 thus indicate that even though EPP activity was low in 2005–2006, EPP-NO_x descent was significant. Mesospheric meteorological data are unavailable, so we rely here on stratospheric data, under the assumption that upper stratospheric vortex strength is a proxy for confined descent in the mesosphere. Prior to 2006, the upper stratospheric vortex in Feb–Mar 2004 was the strongest ever recorded, as indicated by low N. pole temperatures and high zonal mean wind speed from 60–80°N [*Manney et al.*, 2005]. Figure 4 shows these diagnostics using United Kingdom Meteorological Office (MetO) analyses since 1992 for the 2000 K (~50 km) potential temperature level. Feb temperatures at the N. pole in 2006 were lower even than in 2004, and wind speed increased rapidly to the record levels of 2004 by late Feb. (Note that the record negative speeds in Jan pertain to periods when the vortex was displaced from the pole, so that the cyclonic winds are reflected as easterly in a zonal mean calculation). Further, potential vorticity gradients (not shown) were large (G. L. Manney, private communication, 2006). These diagnostics indicate that in Feb and early Mar the upper stratospheric vortex was exceptionally strong, and suggest that the large EPP IE in 2006 was a direct result of unusual meteorological conditions that led to confined descent of EPP-NO_x.

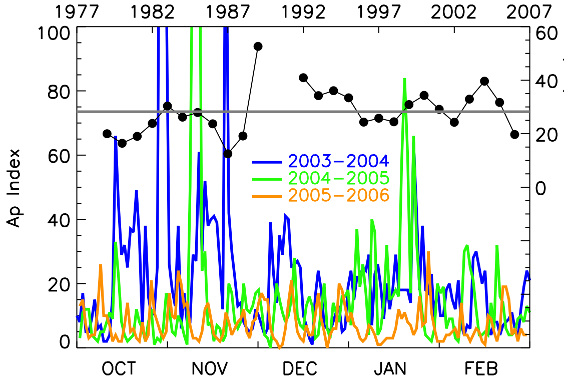

Figure 3. Daily average Ap index for Oct–Feb of 2003–2004 (blue), 2004–2005 (green) and 2005–2006 (orange) as well as average NH auroral power over the months of Oct–Feb from 1978–1979 through 2005–2006 (black dots; top, right axes). Auroral data are not available for the NH winters of 1989–1990 and 1990–1991. Spikes in the Ap Index at 205 and 150 in Oct–Nov 2003 and at 160 in Nov 2004 have been allowed to run off-scale for clarity. The horizontal line denotes the average auroral power for these months/years. Data source is www.sec.noaa.gov.

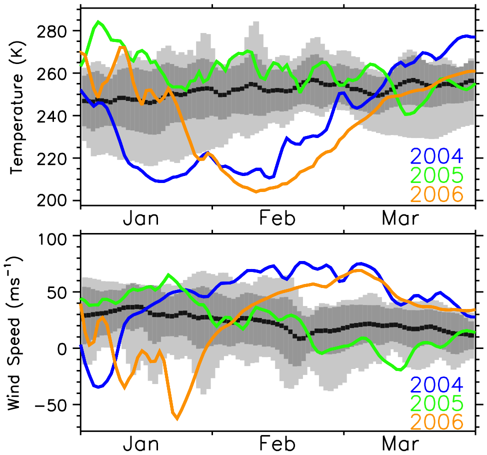

Figure 4. (top) Daily MetO temperature (T) at 90°N (calculated by averaging temperatures at the 87.5°N latitude grid point) and (bottom) zonal mean wind speeds from 60–80°N at 2000 K in 2004 (blue), 2005 (green) and 2006 (orange). The thick black line shows the average T from 1992–2003. The dark gray region represents the average ±1-σ standard deviation, and the light gray region the full range, of daily T values from 1992–2003.

4. Discussion and Summary

[16] Using recent ACE data in comparison to historical measurements from HALOE, we have shown that the amount of EPP-NO_x descending into the Arctic stratosphere was extraordinarily large in Feb–Mar 2006, even though EPP activity was not enhanced in the preceding months. EPP-NO_x observed by ACE at 48 km in Feb–Mar 2006 was more than 6 (3) times higher than observed previously by HALOE in the NH (SH). These comparisons are influenced by the fact that HALOE did not sample latitudes as high as ACE, but still indicate that the 2006 EPP IE was remarkable. We link the large EPP IE in 2006 to unusual meteorological conditions that led to enhanced confined descent from the mesosphere to the stratosphere.

[17] The results confirm the important role of meteorology in determining the magnitude of EPP-NO_x reaching the stratosphere. Two of the last three years have exhibited an unusually strong NH upper stratospheric polar vortex, and in both years the EPP IE was substantial; but in one of those years, EPP activity (production) was low. We therefore conclude that unlike the Antarctic, transport variability is potentially more important in modulating the magnitude of the Arctic EPP IE than variability in EPP itself [e.g., *Callis et al.*, 2001].

[18] The EPP IE has been correlated with stratospheric O_3 loss [*Randall et al.*, 1998, 2001, 2005], but mechanistic interpretation of the observations requires model calculations to separate dynamical and chemical effects. The EPP-NO_x that reached the Arctic stratosphere in 2006 would have mixed with background NO_x as the vortex warmed, so its effects on middle stratosphere O_3 were not obvious in the ACE data. Although predictions vary [*Hood and Soukharev*, 2005, and references therein], it has been speculated that a changing climate will affect future vortex strength. The results here suggest that a stronger (weaker) vortex leads to increases (decreases) in the EPP IE, thereby increasing (decreasing) NO_x in the polar stratosphere. Quantifying such future effects on stratospheric O_3 and possibly climate requires more observations of NO_x throughout the polar night, meteorological data pertaining to the polar stratosphere and mesosphere, and a concerted modeling effort.

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