

## Stratospheric effects of energetic particle precipitation in 2003–2004

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[1] Upper stratospheric enhancements in NO<sub>x</sub> (NO and NO<sub>2</sub>) were observed at high northern latitudes from March through at least July of 2004. Multi-satellite data analysis is used to examine the temporal evolution of the enhancements, to place them in historical context, and to investigate their origin. The enhancements were a factor of 4 higher than nominal at some locations, and are unprecedented in the northern hemisphere since at least 1985. They were accompanied by reductions in O<sub>3</sub> of more than 60% in some cases. The analysis suggests that energetic particle precipitation led to substantial NO<sub>x</sub> production in the upper atmosphere beginning with the remarkable solar storms in late October 2003 and possibly persisting through January. Downward transport of the excess NO<sub>x</sub>, facilitated by unique meteorological conditions in 2004 that led to an unusually strong upper stratospheric vortex from late January through March, caused the enhancements. **Citation:** Randall, C. E., et al. (2005), Stratospheric effects of energetic particle precipitation in 2003–2004, *Geophys. Res. Lett.*, 32, L05802, doi:10.1029/2004GL022003.

### 1. Introduction

[2] Through dissociation and ionization processes, energetic particle precipitation (EPP) leads to routine production of odd nitrogen in the mesosphere and thermosphere, and to sporadic production in the stratosphere when very highly energetic particles are involved. EPP also indirectly affects the stratosphere via descent of EPP-produced mesospheric NO<sub>x</sub> (NO + NO<sub>2</sub>) to the stratosphere during polar winter and spring, where it plays a major role in ozone chemistry. EPP coupling of the upper and lower atmosphere was first examined more than two decades ago [Solomon et al.,

1982], and observational evidence for its occurrence is prevalent [e.g., Russell et al., 1984; Jackman et al., 1995; Rinsland et al., 1996; Randall et al., 1998, 2001a; Siskind et al., 2000; Callis et al., 2001; Natarajan et al., 2004 (hereinafter referred to as N04)]. EPP occurs throughout the polar night, but the stratospheric response depends strongly on the energy and number of precipitating particles (i.e., the altitude and intensity of the source), and on the efficiency of vertical transport across the stratopause.

[3] In Nov and Dec 2003, enhancements in northern hemisphere (NH) upper stratospheric HNO<sub>3</sub> and NO<sub>2</sub> were measured by the MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) instrument [Orsolini et al., 2005] (hereinafter referred to as O05), and enhancements in NO<sub>2</sub> were measured by the GOMOS (Global Ozone Monitoring by Occultation of Stars) instrument [Seppälä et al., 2004]. These authors attributed the enhancements to NO<sub>x</sub> produced by EPP during the unusually powerful solar storms and associated solar proton events (SPEs) of Oct–Nov 2003 (see special section on Violent Sun–Earth Connection Events of October–November 2003, *Geophysical Research Letters*, 32, 3, 2005). In Apr 2004, unprecedented enhancements in NH upper stratospheric NO<sub>x</sub>, accompanied by significant reductions in ozone, were observed by the Halogen Occultation Experiment (HALOE). The enhancements were tentatively ascribed to indirect effects (mesospheric production followed by descent) of the Oct–Nov solar storms (N04). In this paper we describe measurements from numerous satellite instruments to provide a more comprehensive picture of EPP effects in 2003–2004 on the stratosphere. Included are data from the POAM (Polar Ozone and Aerosol Measurement) II & III, SAGE (Stratospheric Aerosol and Gas Experiment) II & III, HALOE, MIPAS, and OSIRIS (Optical Spectrograph and InfraRed Imager System) instruments. We examine the time dependence of NO, NO<sub>2</sub>, and O<sub>3</sub> from Nov 2003 through Jul 2004, place the observations in historical context, and investigate the origin of observed NO<sub>x</sub> enhancements.

### 2. Stratospheric NO<sub>x</sub> Enhancements and O<sub>3</sub> Reductions in 2003–2004

[4] Figure 1 compares 2004 measurements of NO<sub>2</sub> (or NO<sub>x</sub> in the case of HALOE) and O<sub>3</sub> at 40 km to data in previous years from four solar occultation instruments: HALOE (1991–present), SAGE II (1984–present) and POAM II & III (1994–1996 and 1998–present). Because of their mid-inclination orbits, the HALOE and SAGE II measurement latitudes vary rapidly in time, generally reaching polar latitudes for several days each month, but at different times each year (see Randall et al. [2001b] for representative latitude sampling of HALOE and SAGE II).

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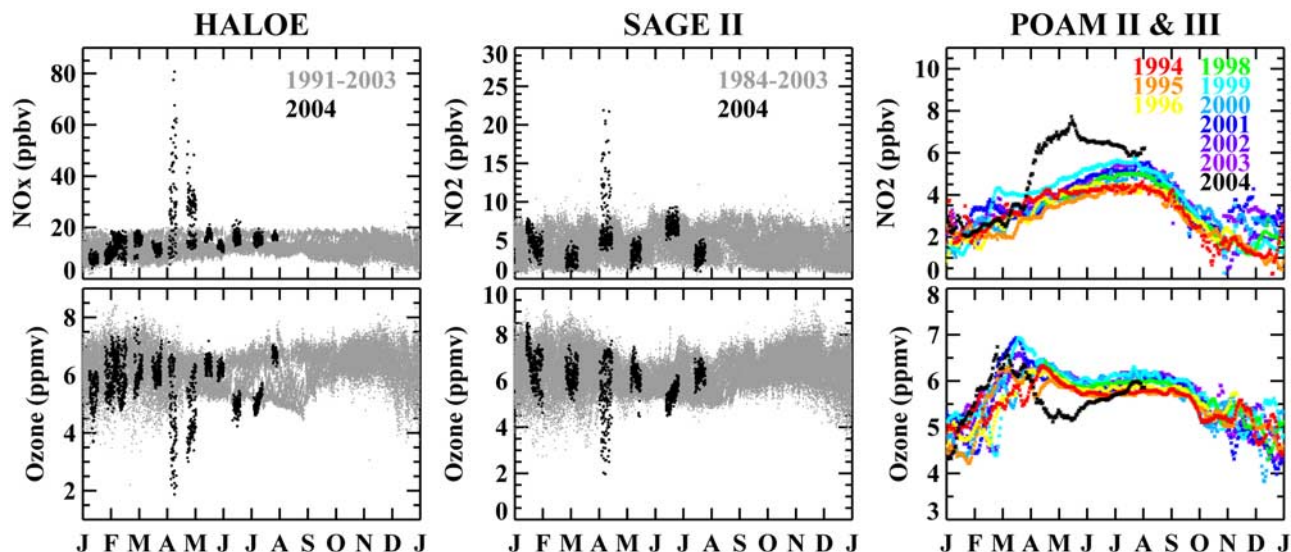
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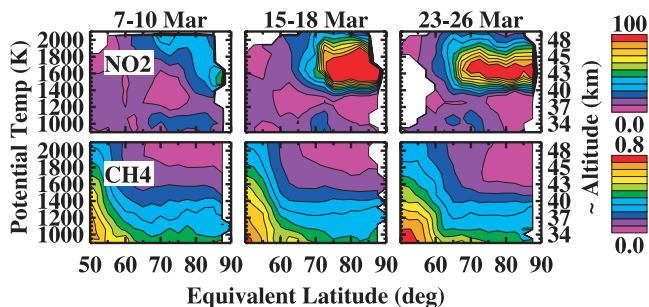
**Figure 1.** Mixing ratios at 40 km for (top left) HALOE  $\text{NO}_x$ , (top middle) SAGE II  $\text{NO}_2$ , (top right) POAM  $\text{NO}_2$  and (bottom)  $\text{O}_3$  for the respective instruments. POAM III  $\text{NO}_2$  has been scaled as described by *Randall et al.* [2001a]. Tick marks on the horizontal axis denote the first day of each month.

Thus every individual NH measurement with error less than 30% is depicted, including local sunrise (SR) and sunset (SS) data. POAM measurement latitudes repeat annually, varying only slowly in time between  $54^\circ$  and  $71^\circ$  in the NH, so 7-day running averages are shown; only local SS measurements are acquired by POAM in the NH.  $\text{NO}_x$  (or  $\text{NO}_2$ ) enhancements and  $\text{O}_3$  reductions in Apr 2004 are unmistakable in these data sets:  $\text{NO}_x$  mixing ratios are up to a factor of 4 higher than ever observed previously, and  $\text{O}_3$  is reduced by more than 60% in some cases. Further analysis (not shown) reveals that the anomalies are confined to latitudes poleward of  $\sim 40^\circ\text{N}$ , and that enhancements of 10–50% persist through July (the last month of the analysis) in all three data sets above 40 km. N04 show the photochemical consistency of the  $\text{NO}_x$  enhancements and  $\text{O}_3$  reductions in Apr HALOE data. Figure 1 gives a more complete picture of the time dependence and places the observations into the context of measurements dating back to 1984.

[5] The polar-orbiting MIPAS instrument senses infrared emission, and is thus capable of measuring atmospheric constituents globally during night and day. Figure 2 shows MIPAS  $\text{NO}_2$  and  $\text{CH}_4$  data plotted as a function of equivalent latitude (EqLat, the latitude that would enclose the same area between it and the pole as a given potential vorticity (PV) contour) and potential temperature, for several 4-day periods in Mar 2004 (MIPAS has not been operating since late Mar 2004).  $\text{NO}_2$  mixing ratios typically decrease with increasing altitude and EqLat in the upper stratosphere, due to less production from  $\text{N}_2\text{O}$  oxidation, destruction by atomic nitrogen, and descending  $\text{NO}_x$ -poor air. In 2004, however,  $\text{NO}_2$  enhancements above 40 km appear in early Mar at high EqLat, reaching values of over 150 ppbv by mid-Mar before declining slightly in late Mar. Corresponding  $\text{CH}_4$  plots indicate that the  $\text{NO}_2$  enhancements occur in air that descends slowly throughout Mar;  $\text{CH}_4$  mixing ratios of 0.1–0.2 ppmv signify air of mesospheric origin, consistent with the Apr enhancements shown by N04.

[6] The geographic distribution of nighttime MIPAS  $\text{NO}_2$  for 24–26 Mar at 1640 K ( $\sim 43$  km) is shown in Figure 3, and reveals enhanced  $\text{NO}_2$  nearly filling the vortex. The decline in  $\text{NO}_2$  near the pole is still under investigation. The polar-orbiting OSIRIS instrument senses scattered light with near-global, daytime coverage, when diurnally-varying  $\text{NO}_2$  mixing ratios are at their smallest. OSIRIS is not sensitive above 1600 K, where the peak of the late Mar enhancements was observed by MIPAS. By mid-Apr, however, the peak enhancement had descended below 1600 K. The OSIRIS  $\text{NO}_2$  map shown in Figure 3 indicates that the enhancements at this time and altitude were still confined largely to the vortex, but with substantial inhomogeneity perhaps due to increased mixing as the vortex began to break down.

[7] The POAM III and SAGE III solar occultation measurements have continual high-latitude coverage throughout the year, and can thus be used to follow the temporal progression of the enhancements in altitude.

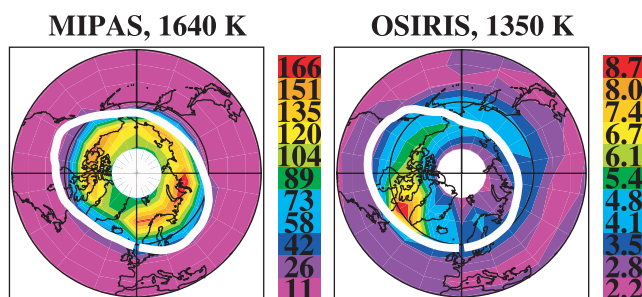


**Figure 2.** Potential temperature/Equivalent Latitude plots of MIPAS  $\text{NO}_2$  (top, solar zenith angle (SZA)  $> 95^\circ$ ) and  $\text{CH}_4$  (bottom) in 2004 for the dates shown in each column. These are similar to zonal mean contour plots but use the vortex-centered EqLat coordinate system. Contour increments are 10 ppbv for  $\text{NO}_2$  and 0.08 ppmv for  $\text{CH}_4$ ; white indicates missing data. The vortex edge is at  $\sim 60^\circ$  EqLat.

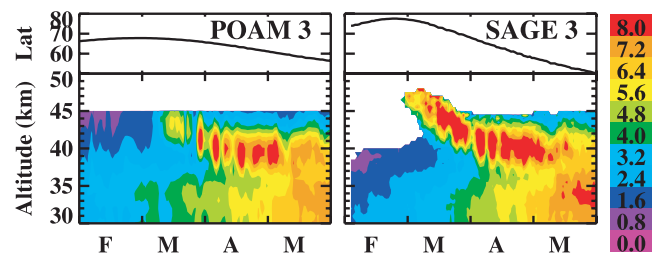
Contour plots of three-day running average  $\text{NO}_2$  profiles from POAM III and SAGE III are shown in Figure 4. By 1 Mar, enhanced  $\text{NO}_2$  is evident in the SAGE III data above 45 km, and both data sets show significant enhancements at 45 km by 9 Mar. The enhanced layer descends in time during Mar and Apr, with a maximum near 38–40 km by early May. The data shown in Figures 2–4, as well as the fact that particle activity in Mar–Apr 2004 (not shown) was insufficient to produce significant  $\text{NO}_x$  directly in the stratosphere, confirm that the extraordinary spring/summer enhancements in Figure 1 arise from descent of  $\text{NO}_x$ -rich air from the mesosphere prior to the beginning of Mar.

### 3. Origin of the Spring/Summer $\text{NO}_x$ Enhancements

[8] Two requirements must be satisfied to explain the 2004  $\text{NO}_x$  enhancements: a significant source of mesospheric  $\text{NO}_x$ , and efficient descent to the upper stratosphere. The anomalous nature of the 2004 enhancements suggests that one or both of these requirements was met in an extraordinary way. The Oct–Nov 2003 solar storms were certainly exceptional, but were they the ultimate source? That significant  $\text{NO}_x$  was produced from the stratosphere to the thermosphere during the Oct–Nov 2003 solar storms is evident from numerous satellite observations [Jackman *et al.*, 2005; Seppälä *et al.*, 2004; O05]. Ideally, the descending layer of enhanced  $\text{NO}_2$  shown in Figure 4 would be traced back in time and upward in altitude to its origin, but no reliable POAM III or SAGE III  $\text{NO}_2$  data exist above the highest altitudes shown here. However, the measurement of  $\text{NO}_x$  ( $\text{NO} + \text{NO}_2$ ) enables the examination of enhancements up into the mesosphere. Polar ( $>60^\circ\text{N}$ )  $\text{NO}_x$  data from the Atmospheric Chemistry Experiment (ACE) are available from mid-Feb to late-Mar 2004, and show an enhanced layer descending inside the vortex during this time from  $\sim 55$  to 45 km (C. P. Rinsland *et al.*, ACE stratospheric measurements of  $\text{NO}_x$  and long-lived tracers during February and March 2004, submitted to *Geophysical Research Letters*, 2005). Using HALOE data, N04 also show  $\text{NO}$  enhancements inside the vortex in mid-Feb above 50 km. Further



**Figure 3.** (left) Orthographic map of MIPAS  $\text{NO}_2$  (ppbv,  $\text{SZA} > 95^\circ$ ) at 1640 K ( $\sim 43$  km) for 24–26 Mar 2004. Latitudes range from  $30^\circ\text{N}$  to the pole, and  $0^\circ$  longitude is at the bottom. The white contour denotes  $60^\circ$  equivalent latitude, approximately the vortex edge. The white circle near the pole indicates missing data. (right) Same as left but for OSIRIS  $\text{NO}_2$  at 1350 K ( $\sim 40$  km) for 12–13 Apr 2004; only ascending node data (local solar time 12–18 hrs) are included with  $\text{SZA}$  ranging from  $72$ – $85^\circ$ .



**Figure 4.** Three-day running averages of (left) POAM III and (right) SAGE III SS  $\text{NO}_2$  mixing ratios (ppbv) from 1 Feb to 31 May 2004. For Feb–Apr, only data inside the vortex is included. Measurement latitudes are shown at the top of each plot.

analysis of HALOE data (not shown) reveals late January 2004  $\text{NO}$  enhancements greater than 200% compared to mean values in 1992–2003; the enhancements occur between 60 and 70 km, and the HALOE measurement latitude at this time was  $\sim 47^\circ\text{N}$ . Because of the sparse latitude sampling of solar occultation instruments, these data are not definitive. At a minimum, they suggest that the anomalous springtime enhancements originated in the polar region above 60 km before the end of January. Nevertheless, there are no satellite measurements of  $\text{NO}_x$  in the polar mesosphere/thermosphere from Nov through Jan that unambiguously connect the late Jan enhancements to the storms.

[9] This leads to the question of whether it is possible that the source of the springtime enhancements was EPP production of  $\text{NO}_x$  after, not during, the Oct–Nov storms. A major warming resulted in a breakup of the upper stratospheric vortex in late Dec 2003; the vortex re-gained strength throughout Jan 2004, and by Feb–Mar was the strongest on record since at least 1979 [Manney *et al.*, 2004]. Therefore, assuming meteorological conditions in the mesosphere mirrored those in the upper stratosphere,  $\text{NO}_x$  produced in the Oct–Nov storms that had descended to the mesosphere by late Dec probably experienced substantial mixing to lower latitudes. Here it would have been photolyzed, reducing the probability for observing enhanced  $\text{NO}_x$  in the stratosphere by Mar. On the other hand,  $\text{NO}_x$ -rich air in the polar lower mesosphere in late Jan most likely experienced less transport to midlatitudes, resulting in an unusually high probability of being transported to the stratosphere. Thus, nominal EPP that occurred well after the Oct–Nov storms, together with the unique dynamical situation in 2004, could have resulted in  $\text{NO}_x$  enhancements in the springtime upper stratosphere qualitatively similar to those seen in Figure 4. Determining whether  $\text{NO}_x$  production after the major warming, in combination with the unprecedented vortex strength, was sufficient to explain the magnitude of the anomalous springtime enhancements is the subject of future work.

### 4. Summary

[10]  $\text{NO}_x$  and  $\text{O}_3$  data from numerous satellite instruments have been analyzed to present a broad overview of EPP effects on the NH stratosphere from Nov 2003 through Jul 2004. EPP effects are ubiquitous in 2003–2004 satellite data, providing a unique opportunity to test our understand-



ing of atmospheric coupling via EPP.  $\text{NO}_x$  production from the upper stratosphere through the mesosphere occurred in Nov as a direct result of strong solar storms in Oct–Nov 2003. Substantially elevated mesospheric  $\text{NO}_x$  was observed in Jan as well; whether this was a result of  $\text{NO}_x$ -rich air descending after the initial storms, or new production from smaller SPEs and energetic electrons, is still undetermined. By late Jan, the satellite data indicate a systematic descent of  $\text{NO}_x$ -rich air in the vortex that finally led to unprecedented (in the NH data sets shown here, which span 1984–present)  $\text{NO}_x$  enhancements and  $\text{O}_3$  reductions in the upper stratospheric vortex from Mar–May 2004. The anomalies declined as the vortex broke up, but were still evident even in Jul. The extraordinary springtime  $\text{NO}_x$  enhancements and  $\text{O}_3$  reductions are attributed to a combination of EPP and an exceptionally strong vortex from late Jan into Apr that confined  $\text{NO}_x$ -rich air to the polar region, and allowed relatively unmixed descent in the polar lower mesosphere and upper stratosphere. Detailed 3-D modeling including explicit calculation of  $\text{NO}_x$  production in the upper atmosphere by precipitating electrons and protons over a wide range of energies is necessary to definitively attribute, or not, the observed  $\text{NO}_x$  enhancements to the powerful solar storms of Oct–Nov 2003.

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