Interannual variation of NO$_x$ from the lower thermosphere to the upper stratosphere in the years 1991–2005

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Received 17 June 2010; revised 15 November 2010; accepted 23 November 2010; published 16 February 2011.

[1] The interannual variation of NO$_x$ throughout the year is investigated for the period 1991–2005 at middle to high latitudes using Halogen Occultation Experiment (HALOE) on UARS measurements. We find a clear correlation of NO$_x$ between 80 and 130 km with the auroral electrojet index in both hemispheres, which is fairly independent of season, indicating a relatively frequent NO$_x$ source from precipitating auroral electrons of energies ranging from about 1 keV to several tens of keV. Between 80 and 100 km, NO$_x$ is also highly correlated to fluxes of higher-energy electrons as measured by the Space Environment Monitor (SEM) and its successor, the SEM-2, instruments on POES mostly during autumn and spring, indicating a strong impact of 10–100 keV electrons, which, however, precipitate less frequently than the auroral electrons. Electrons with energies of several MeV were investigated also, and a significant correlation was found with NO$_x$ during some periods. The correlation is smaller and less stable than for the lower-energetic auroral and POES electrons, indicating that the contribution of MeV electrons to the overall NO$_x$ budget is small, at least in the latitude range considered. Also, the altitude range affected, above 60 km, indicates that this impact is probably due to electrons of lower energies (several hundreds of keV instead of several MeV) than the GOES electrons used for the investigation, to which they must be closely related, however. Downward propagation of NO$_x$ is observed in both hemispheres during winter but continues to lower altitudes and lasts longer in the Southern Hemisphere, where the signal can be followed to altitudes around 40 km.


1. Introduction

[2] In the middle atmosphere, NO$_x$ (N, NO, NO$_2$) is highly variable, especially during wintertime at high latitudes and midlatitudes [Siskind and Russell, 1996; Siskind et al., 2000; Siskind, 2002; Randall et al., 1998; Randall et al., 2001; Randall et al., 2006; Randall et al., 2009; Funke et al., 2005; López-Puertas et al., 2005; López-Puertas et al., 2006; Hood and Soukharov, 2006; Lu et al., 2008; Seppälä et al., 2007b; Seppälä et al., 2007a]. During some winters, high values of NO$_x$ can be attributed to the production of NO$_x$ by large solar particle precipitation events [e.g., Jackman et al., 2001, 2005; Randall et al., 2001; López-Puertas et al., 2005; Seppälä et al., 2007a]. However, in most winters, middle atmosphere NO$_x$ values are modulated by the strength of the polar vortex and geomagnetic activity [e.g., López-Puertas et al., 2006; Randall et al., 2009].

[3] High geomagnetic activity triggered by solar Corona Mass Ejections or high-speed solar wind streams leads to the acceleration of electrons in the magnetotail and the outer radiation belts to energies of several keV to MeV. These high-energy electrons are eventually lost to the atmosphere in the auroral zone (60°–75° geomagnetic latitudes) or at geomagnetic latitudes connecting to the radiation belts (~50°–65° geomagnetic latitude). Precipitating electrons ionize the atmosphere, producing N and NO by dissociation and ion chemistry reactions [e.g., Gerard and Barth, 1977; Porter et al., 1976; Rusch et al., 1981]. A positive correlation between geomagnetic activity, precipitating electrons of auroral energies (<10 keV) and NO is well established in the polar lower thermosphere [e.g., Barth et al., 2003; Baker et al., 2001]. A similar correlation between NO$_x$ and energetic electron precipitation represented either by the geomagnetic Ap index or by the electrons’ hemispheric power has been reported mainly during solar winter in the upper stratosphere and mesosphere in both hemispheres [Seppälä et al., 2007b; Lu et al., 2008] and in the middle stratosphere only in the Southern Hemisphere because of the

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0148-0227/11/2010JA015825

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generally less stable polar vortices in the Northern Hemisphere [Randall et al., 2007; Siskind et al., 2000].

[5] An important issue in this context is at which altitudes NOx is produced because of energetic particle precipitation (EPP); this depends on the energy spectrum of the electrons. Energetic electrons during geomagnetic storms are accelerated to energies ranging from keV to MeV; the larger the electrons’ energy, the lower into the atmosphere they can precipitate, with keV electrons reaching the lower thermosphere or upper mesosphere and MeV electrons reaching the upper stratosphere [Berger et al., 1970; Fang et al., 2008]. Thus, in principle, electron precipitation should be possible from the upper stratosphere to the lower thermosphere, though generally fluxes of lower energies are higher, so the higher altitudes should be affected more [e.g., Millan and Thorne, 2007].

[5] So far not much observational evidence is available for an in situ production of NOx due to energetic electron precipitation in the middle atmosphere below ~90 km. One observation of a NOx increase due to geomagnetic disturbances has been reported by Callis et al. [1998] after the 12 May 1992 solar event; in this case, however, the affected area was restricted to altitudes above ~80 km. Another observation seemed to suggest a fairly large in situ production of NOx in the upper stratosphere around 60 km related to an electron precipitation event dominated by electron energies of a few hundred keV on 22 January 2004 [Renard et al., 2006]; however, this was contested by Funke et al. [2007], who pointed out that the large values of NOx observed were also in agreement with transport from the upper mesosphere and that enhanced values at higher altitudes were indeed observed by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument several days before the EPP event. A very significant in situ production of NOx from ~47–70 km was observed by Global Ozone Monitoring by Occultation of Stars (GOMOS) around 15 February in the same winter [Clilverd et al., 2009]. However, measurements from MIPAS covering a similar latitude range are reported for the same period of time and show no evidence for in situ production but do show a clear signal of downwelling of NOx from the upper mesosphere [López-Puertas et al., 2006]. An indirect indication of a potential large contribution of highly relativistic electrons onto the stratosphere comes from the study by Sinnhuber et al. [2006], who found a large interannual variation of stratospheric ozone at very high latitudes during midwinter, which correlates well with the flux of ≥2 MeV electrons in the radiation belts. Thus, observational evidence so far seems to suggest an EPP-NOx source mainly in the upper mesosphere and lower thermosphere, which is transported down into the lower mesosphere and stratosphere during polar winter; whether there is also a direct contribution of EPPs onto the stratosphere and lower mesosphere as a result of precipitating electrons of energies ≥1 MeV is not clear yet, as the observational evidence for this is indirect or ambiguous.

[6] We use the complete time series of NO and NO2 from the Halogen Occultation Experiment (HALOE) on UARS from mid-1991 to late 2005, together with a number of indices of solar and geomagnetic activity as well as electron fluxes to investigate the impact of EPP onto the NOx budget from the midstratosphere to the lower thermosphere in both hemispheres.

2. Data Sets

2.1. HALOE Data

[7] For this investigation, 14 years of NOx (NO + NO2) measurements from the HALOE instrument onboard the UARS satellite [Russell et al., 1993] were analyzed, covering the years 1991–2005. Data of NO and NO2 from HALOE version 19 were used, which are provided as individual measurements gridded onto 141 fixed pressure levels between ~10 and 150 km, giving an approximate vertical spacing of 1 km in the middle atmosphere. As NO2 values are very low during solar illumination in the mesosphere and thermosphere, no NOx measurements are available above ~54 km. So, what is called NOx in the following is NO only in the mesosphere and thermosphere and NO + NO2 in the stratosphere.

[8] Data at latitudes poleward of 40° were averaged into 2 month periods from January/February to December/January of each year. Because HALOE measures in solar occultation, it observes only a very narrow latitude range every day, but the latitudes observed vary quickly from day to day. Data were averaged over 2 months to ensure that measurements in the latitude areas considered are available in nearly all time intervals. Both sunrise and sunset data were used to ensure a reasonable sampling in all time intervals. The HALOE data cover the period October 1991 to November 2005, with only 9 d of measurements in September 2005, however, and only 1 d of measurement in October 2005. Below 40 km, sunrise and sunset NO + NO2 are distinctly different because of nighttime NO3 and N2O5 formation. Below 30 km, NOx is determined by denitrification and denoxification because of polar stratospheric clouds during cold polar winters. As this produces a different type of interannual variation that we do not want to investigate here, the investigation was restricted to altitudes above 35 km, including the caveat that between 35 and 40 km, the data set might be contaminated by nighttime NO3 and N2O5 formation.

[9] In Figure 1 (top), the resulting NOx data set between 35 and 130 km is presented for both hemispheres. A clear downward propagation of NOx from the lower thermosphere to the upper stratosphere is observed in both hemispheres during winter, with a large interannual variation, and generally higher values reaching down into the Southern Hemisphere. Additionally, several large solar proton events (SPEs) can be identified that are most obvious in the summer hemisphere, namely, in July 2000, November 2000 and 2001, and October and November 2003. As the large NOx production due to SPEs dominates the NOx signal in the stratosphere and mesosphere during periods of large SPEs, these periods were excluded from the data analysis. Considered were the following SPEs, chosen because of their modeled global NOx production [Jackman et al., 2008; Jackman et al., 2009]: 14 and 15 July 2000, 9 November 2000, 25 and 26 September 2001, 5 and 6 November 2001, 24 November 2001, 21 and 22 April 2002, 29 October 29 to 3 November 2003, 15–23 January 2005, and 7–17 September 2005. For these SPEs, the 2 month averages containing the
SPE as well as those containing the following month were excluded to account for the longevity of NO\textsubscript{x}, especially during periods of low illumination.

\[10\] In Figure 1 (bottom), the latitudinal coverage of the data set is shown for the whole observation period. As can be seen, a fairly homogeneous data set is sampled during midwinter in both hemispheres, covering the latitude range 40°–50°. At the beginning of the time period, latitudes of around 60° were sampled during summer in both hemispheres, but the summer coverage moved poleward to 60°–70° after 1997. Thus, during summer polar latitudes are sampled and during winter midlatitude air is observed. During spring and autumn, the latitude range changes rapidly, and measurements are averaged over quite large latitude bands as shown by the large standard deviation of the latitudinal means (see Figure 1, bottom), while during midwinter and summer, measurements are averaged over fairly compact latitude areas. In the stratosphere and lower mesosphere, vortex air will be sampled occasionally in both hemispheres during late winter and spring. This may enable us to investigate the impact of downwelling of NO\textsubscript{x} in the polar winter vortex; however, as there is a large year-to-year variation of vortex size and equatorward expansion in the Northern Hemisphere, an artificial interannual variation might be inflicted there because in some years no vortex air is sampled.

2.2. Electron Fluxes, Solar Activity, and Geomagnetic Activity

\[11\] A number of properties were investigated which are related either to the solar cycle or to energetic electron precipitation. All data sets were averaged over 2 month periods from January/February 1991 to November/December 2005. Solar radio flux (F10.7) was used as a proxy for solar activity. Lyman \(\alpha\) radiation was considered as a proxy for solar UV variation also, but as it highly correlates with solar radio flux (correlation coefficient of 0.98 for the 2 month averages used here), only the solar radio flux is shown in the following. The auroral electrojet (AE) index was used to describe geomagnetic activity. The AE index is derived from ground-based measurements of variations of the horizontal magnetic field component at several stations in a similar way to the Ap index, to which it is closely correlated (correlation coefficient of 0.9). However, unlike the Ap index, which is derived from stations all over the world, AE is derived only from stations located in the region of the northern auroral oval, so it directly measures the response of the magnetic field to the auroral electroject AE, i.e., auroral particle precipitation (see also http://wdc.kugi.kyoto-u.ac.jp/aedir/index.html). In this sense, the AE index indicates auroral electron precipitation, i.e., mainly electrons with energies of up to 10 keV [Barth et al., 2003], but it might also be affected by protons or electrons of higher energies.

\[12\] Two different data sets were used to investigate precipitation of relativistic electrons into the atmosphere, electrons with energies from 100 to 300 keV as measured by the particle sensors onboard the Polar Operational Environmental Satellite (POES) spacecrafts, and electrons with energies \(\geq\)2 MeV as measured by particle sensors onboard the Geostationary Operational Environmental Satellite (GOES) spacecrafts. (Usually, the term “relativistic electrons” is used for electrons with energies of more than 1 MeV whose speed is >90% the speed of light. However, electrons with energies of 100 keV also reach relativistic speeds: more than 50% of the speed of light. In the following, electrons with energies from a few tens of keV to <300 keV will be called “moderately relativistic” and electrons with energies of more than 2 MeV will be called “highly relativistic.”) GOES
measurements are taken mostly within the outer radiation belt, measuring particles trapped or in the drift loss cone, but not precipitating into the atmosphere; the POES instruments measure precipitating particles in polar regions. In the following paragraph, the electron data sets used are described in detail.

[13] A number of GOES satellites from GOES-6 to GOES-11 were in orbit during the HALOE period (1991–2005), so that the whole time period is covered by GOES data, though none of the satellites was in orbit for the complete HALOE era. Data from GOES-11 suffer from a spin of the spacecraft, which leads to artificial variations of the measured electron fluxes (http://www.ngdc.noaa.gov/stp/satellite/goes/index.html); electron detectors of all GOES satellites suffer from contamination due to high-energy protons, which may lead to problems during periods of high proton fluxes, i.e., ion storms (http://www.ngdc.noaa.gov/stp/satellite/goes/index.html). A data set merging from different satellites considering these problems was provided by the National Geophysical Data Center (NGDC) (ftp.ngdc.noaa.gov/ftp/solar_data/satellite_environment).

[14] GOES electron fluxes are correlated very weakly with the $AE$ index, with the magnitude of the correlation coefficient lower than 0.3. As the $AE$ index refers to auroral electrons, which have much lower energies than the GOES electrons and do not originate from the radiation belt, it should not correlate well with the GOES electron fluxes.

[15] The POES data originate from the N-12 (1991–1998) and N-15 (1999–2005) satellites, which carry very similar electron detectors, the Space Environment Monitor (SEM) and its successor, the SEM-2. Apart from a small difference in the geometrical factor, which might account for about 5% of the difference between N-12 and N-15, both detectors are identical.

[16] POES electrons are provided in three energy ranges: $>30$ keV, $>100$ keV, and $>300$ keV. The POES electrons were sampled in such a way that two channels were combined, giving electron energies of $30–100$ keV (later called POES low energy) and $100–300$ keV (called POES mid-energy). The highest-energy channel was also used as is, providing electron energies of $300$ keV to $2.5$ MeV (called POES high energy).

[17] POES takes particle measurements both to the top ($0^\circ$ channels) and to the side ($90^\circ$ channels) of the spacecrafts. The $0^\circ$ channels are oriented upward in polar regions and sample precipitating particles there; in midlatitudes, they sample trapped particle populations as well. To construct a data set which represents the maximum particle precipitation for each hemisphere, the particle flux along the satellites’ track has been fitted with a superposition of Gaussian functions, where the apex represents the maximum count rate within the specific precipitation zone. Only the north-bound half of each orbit was used to minimize local time variations, and data were averaged over 1 d, separately for each hemisphere.

[18] The POES electron data are highly correlated with each other, with the highest correlation coefficient being between the low-energy and midenergy channels (0.9 in the Northern Hemisphere and 0.94 in the Southern Hemisphere). In the following, only results from the POES midenergy channel are shown as results from all three channels are very similar. The POES electrons are also correlated with the $AE$ index (correlation coefficients of about 0.6). GOES and POES electron fluxes are positively correlated with moderate correlation coefficients (0.3 to 0.7).

[19] Some properties of the data sets related to energetic particle precipitation (source, energy range, and atmospheric penetration depth) are provided in Table 1. Solar radio flux, POES data, and a merged data set of GOES $\geq 2$ MeV electron fluxes from different satellites were provided by the NGDC. A merged AE data set was provided by the World Geophysical Data Center (WGDC) in Kyoto, Japan. No AE data are available for 1996, so for any correlation with the $AE$ index, this year was omitted.

### 3. Setup and Error Analysis of Correlation Experiments

[20] In the following, we analyze the HALOE NO$_x$ data, together with the solar and geomagnetic data discussed above, to investigate at which altitudes and by which particle populations NO$_x$ is produced in the middle atmosphere. To achieve this, correlations between the interannual variation of NO$_x$ at different altitudes for all 2 month periods with solar radio flux, geomagnetic activity, and electron fluxes are calculated using the Pearson’s product-moment coefficient for both hemispheres in two different ways. In the first setup, NO$_x$ data from 1991 to 2005 were correlated with the solar radio flux, $AE$ index, and electron flux data in an altitude range of 35–130 km throughout the year from January and February to November and December in both hemispheres in such a way that data from the same 2 month period were correlated with each other. In the second setup, correlation coefficients were calculated for a correlation between NO$_x$ varying throughout 12 months starting in autumn and one fixed 2 month period of the $AE$ index corresponding to early winter (May and June in the Southern Hemisphere and November and December in the Northern Hemisphere).
The first setup was chosen to investigate which processes (solar radiation variability, auroral electron precipitation, and midenergy and high-energy electron precipitation from the radiation belts) directly affect atmospheric NO\textsubscript{x} budgets, and at which altitudes. In this sense, areas of high correlation coefficients will be interpreted as regions of direct production of NO\textsubscript{x}. This works well during the summer months. However, during polar winter, vertical transport can be quite efficient, and during a 2 month period NO\textsubscript{x} can be transported quite a long way from its source region; therefore, during polar winter, areas of high correlation coefficient are generated by a mixture of local production and vertical transport. The second correlation experiment was set up to investigate the impact of vertical mixing, i.e., how long a signal of NO\textsubscript{x} production generated during early winter can be traced, and to which altitudes. Results from the first correlation experiment are shown in Figure 2 and discussed in section 4.1, and results from the second correlation experiment are shown in Figure 4 and discussed in section 4.2.

A total of 15 or 14 years of data are available for every 2 month period; however, data during and after large SPEs have been rejected, further reducing the number of data points available. For 14 data points which are not autocorrelated, a correlation coefficient of 0.46 is significant at the 10\% confidence level, and a correlation coefficient of 0.66 is significant at the 1\% confidence level. Contour values in Figures 2 and 4 were sorted accordingly, showing values larger than 0.46 only. Values larger than 0.66, which are considered highly significant, are marked in red. To test the robustness of the results in an independent way, error values were calculated using a bootstrap method in the following way: for every data set (i.e., for all 141 altitudes of the HALOE data set and all 24 time series of monthly means correlated with all indices) a correlation coefficient was calculated for a random distribution of all available data points 100 times, and the average and standard deviation were calculated for this sample. The standard deviation was given as the “bootstrap error” of the respective correlation coefficient. “Bootstrap errors” larger than 0.25 are marked in light gray areas, and those larger than 0.5 are marked in dark gray areas.
as light gray areas in Figures 2 and 4, and values larger than 0.5 are marked as dark gray areas. For most cases, the correlations achieved seem fairly robust insofar as high correlation coefficients, “bootstrap errors” are generally lower than 0.25.

4. Results and Discussion

4.1. Direct Impact of Solar Activity, Auroral Electrons, and Relativistic Electron Precipitation

[22] In Figure 2, the temporal evolution of the correlation coefficient of NO₃ with solar radio flux, the auroral electrojet, and fluxes of relativistic electrons at two different energies is shown over the year as a function of altitude for both hemispheres. Correlation coefficients (CC) significant at least at the 10% confidence level (CC > 0.46) are found for all properties, but at different altitudes and during different time periods. In Figure 3, selected examples are given for periods and altitudes where NO₃ correlates well with solar radio flux (Figure 3a), the AE index (Figure 3b), and POES and GOES electron fluxes (Figures 3c–3e). In the following, the correlations between NO₃ and all four properties considered (solar radio flux, AE index, and POES and GOES electron fluxes) are discussed in detail.

4.1.1. Solar Activity

[23] A significant positive correlation is observed between NO₃ and solar radio flux mostly above 100 km in some months, indicating enhanced NO₃ production due to photoionization for periods of high solar UV radiation. This seems to be in agreement with a positive regression index observed between HALOE NO₂ and the solar Mg II index in the mesosphere at high latitudes by Hood and Soukharev [2006]. High correlation coefficients are restricted mostly to the second half of the year (from July/August to December/January) in both hemispheres. In Figure 3a, one scenario is shown with a high correlation (CC = 0.89) between solar radio flux and NO₃ (October and November in the Southern Hemisphere at 105 km). The correlation appears to be more compact during solar minimum (1991–1998) than during solar maximum (1999–2003). Especially noticeable are the data for 2003, with a very large SPE during October and November and high NO₃ values even in the lower thermosphere. It would seem that the correlation between solar radio flux and NO₃ in the lower thermosphere would be significantly lower if years with large solar proton events were not excluded from the correlation analysis, and this is indeed the case for most of the autumn and winter period, in which large SPEs are especially frequent.

4.1.2. Auroral Electrojet

[24] A significant positive correlation of more than 0.66 is observed between NO₃ and the AE index for most of the observation period in both hemispheres. The largest correlation coefficients of more than 0.86 are restricted mainly to altitudes above 80 km, reaching up to 130 km at some times. During early summer (September and October to November and December in the Southern Hemisphere and March and April to June and July in the Northern Hemisphere), the correlation between the AE index and NO₃ in the mesosphere and lower thermosphere is generally lower and less robust than during autumn and winter, possibly because of the shorter photochemical lifetime of NO₃ in the sunlit mesosphere and lower thermosphere. Significant positive correlations are observed at lower altitudes down to ~50 km, mostly during autumn and winter (Southern Hemisphere, March/April to May/June; Northern Hemisphere, November/December to January/February), probably because of large-scale downwelling at high latitudes during winter. This indicates a particle precipitation source which (because most 2 month periods are affected) appears to be independent of season and fairly frequent; the altitude region affected (80–130 km) corresponds to electrons of energies ranging from 1 keV to several tens of keV [see Fang et al., 2008, Figure 3], with a downwelling of the signal to lower altitudes during autumn and winter. Downwelling of auroral NO₃ during winter is discussed in more detail in section 4.2.

[25] During several 2 month periods, significant correlations between NO₃ and the AE index are observed between 70 and 80 km during summer (April/May, May/June, and July/August in the Northern Hemisphere and February/March in the Southern Hemisphere). As downwelling of air should not play a role during summer, this suggests that occasionally sporadic events of EPP occur where the electron spectra have a significant contribution from higher energies of up to 100 keV [see Fang et al., 2008, Figure 3]. In Figure 3b, one example is shown of a scenario where NO₃ is highly correlated to the AE index (CC = 0.97; March and April in the Southern Hemisphere at 100 km).

4.1.3. POES Electrons

[26] Only results from the POES midenergy channel are shown because, as a result of the high correlation of all three POES channels, results from the other channels are very similar. Significant positive correlations between POES electrons and NO₃ are observed from January/February to June/July and from October/November to December/January at altitudes mostly between 80 and 100 km in both hemispheres. In the Northern Hemisphere, the autumn signal is more pronounced and starts earlier (in September/October) than in the Southern Hemisphere. In the Southern Hemisphere, high values of the correlation coefficient are observed also at much lower altitudes to below 50 km during winter (April/May to June/July), probably due to large-scale downwelling during the winter months. During several months, very high values of the correlation coefficient of more than 0.86 are reached between 80 and 100 km, becoming even more pronounced than the correlation between NO₃ and the AE index in this altitude range. In Figure 3c, one example is shown for a scenario of a very high correlation between NO₃ and the POES electron fluxes (CC = 0.94; October/November in the Northern Hemisphere at an altitude of 80 km). In this case, NO₃ closely follows the POES electron fluxes in all years but 2003, which was not included in the calculation of the correlation coefficient because of the large SPEs in October and November. The vertical and temporal structure of the correlation between NO₃ and the POES electron fluxes as shown in Figure 2 suggests electron precipitation events with particle energies of 10–100 keV [see Fang et al. [2008]], which occur most frequently during spring (February/March and March/April) and autumn (October/November and November/December). It should be noted that this is at the lower end of the energy range observable by the SEM and SEM-2 instruments (see Table 1); no evidence is found for a direct impact of the high-energy part (100 keV to
Figure 3. Variation of NO\textsubscript{x} (red) from year to year compared to solar radio flux (violet in Figure 3a), the AE index (blue in Figure 3b), and the fluxes of relativistic electrons as measured by POES (green) and GOES (violet) (Figures 3c–3e) for selected altitudes and 2 month periods. (a) October and November in the Southern Hemisphere, 105 km; NO\textsubscript{x} compared to solar radio flux. (b) March and April in the Southern Hemisphere, 100 km; NO\textsubscript{x} compared to the AE index. (c) October and November in the Northern Hemisphere, 80 km; NO\textsubscript{x} compared to POES midenergy channel electron flux and AE index. (d) April and May in the Southern Hemisphere, 60 km; NO\textsubscript{x} compared to POES midenergy channel electron flux and GOES electron flux. (e) December and January in the Northern Hemisphere, 60 km; NO\textsubscript{x} compared to POES midenergy channel electron flux and GOES electron flux. Error bars on the NO\textsubscript{x} data denote the statistical error of the mean. Black open circles mark periods likely affected by large solar proton events.
2.5 MeV) of the electron energies observable by the SEM and SEM-2 instruments.

4.1.4. GOES ≥2 MeV Electrons

Significant positive correlations between NO$_x$ and GOES ≥2 MeV electrons are observed during some periods, namely, April/May, May/June, and December/January in both hemispheres: March/April and June/July in the Northern Hemisphere and October/November in the Southern Hemisphere. The altitude range of significant correlation coefficients varies from ~60 to 90 km, higher than expected from the energy range of the GOES electron counters (see Table 1). It should be noted that the correlation coefficients between GOES electron fluxes and NO$_x$ are significantly lower than those between AE index and NO$_x$ or those between POES electron fluxes and NO$_x$ and are mostly accompanied by fairly high values of the “bootstrap error.” Also, the general pattern of positive correlation coefficients is quite similar to that between POES electrons and NO$_x$ as discussed in the preceding section. This suggests that the correlation between GOES electrons and NO$_x$ is not very robust and might at least partially be due to the positive correlation between GOES and POES electron fluxes (CC = 0.3–0.7; see section 2.2). Two examples of scenarios where GOES electron fluxes are significantly correlated with NO$_x$ are shown in Figures 3d and 3e, December/January in the Northern Hemisphere and April/May in the Southern Hemisphere, both at 60 km. For comparison, the POES electron fluxes are shown as well. In April/May (Figure 3d), the correlation between NO$_x$ and POES electrons (CC = 0.80) is higher than that between NO$_x$ and GOES electrons (CC = 0.68), and in most years, the variation between POES and GOES electron fluxes is similar. In 1994, however, NO$_x$ seems to have followed more closely the POES electrons, while in 1995 and 2005, NO$_x$ appears to have followed more closely the GOES electron fluxes. In December/January (Figure 3e), the correlation between NO$_x$ and GOES electrons (CC = 0.73) was larger than that between NO$_x$ and POES electrons (CC = 0.46), and in most years, the NO$_x$ variation appears to follow the GOES electrons. Exceptions are again 1995, when NO$_x$ values were quite high, and 2003, when the GOES electron fluxes were exceptionally high; however, 2003 has been excluded from the calculation of the correlation coefficients because of the large SPE in October and November.

This suggests that there is a contribution of highly relativistic electrons to the overall NO$_x$ budget as argued by Sinnhuber et al. [2006] and Clilverd et al. [2009]; however, their contribution must be significantly smaller than the contributions of auroral electrons (1 to several tens of keV) or moderately relativistic electrons (up to 100 keV) to higher altitudes. It should also be noted that the contribution of highly relativistic electrons is observed mostly at altitudes ≥60 km, which should correspond to lower particle energies than observed by the GOES instruments (see Table 1). Thus, the positive correlation observed between NO$_x$ and GOES electron fluxes around 60 km are more likely due to electrons of energies of several hundred keV, particle populations which are intermediate to the GOES and POES electrons in energy and apparently also in the precipitation behavior.

4.2. Downwelling During Winter

The second setup of the correlation experiment investigates downwelling of EPP-NO$_x$ during winter as observed in the HALOE data set. Results of this setup correlating 2 month averages of NO$_x$ from autumn to late summer of the following year with a fixed 2 month period of the AE index during midwinter (May/June in the Southern Hemisphere and November/December in the Northern Hemisphere) are shown in Figure 4. The AE index is used here because it shows the most significant and temporally stable correlation.

In the Southern Hemisphere, during April/May and May/June, significantly high correlations are observed between NO$_x$ and the AE index above 90 km, indicating direct production due to electrons of less than 30 keV. During the following months, the area of significant correlation coefficients descends downward, reaching altitudes of 40–70 km in June/July and 40–50 km in August/September. After August/September, the signal ceases. Thus, a fairly stable downwelling is observed during the Southern Hemisphere polar winter, starting in late autumn and continuing well into spring, transporting air from the upper mesosphere at least into the midstratosphere. During summer (October/November to February/March), significant negative correlations are observed below 40 km altitude. This signal appears to be quite stable, as it lasts for several months and is accompanied by relatively low bootstrap errors. Since below 40 km sunrise and sunset data can differ because of nighttime N$_2$O$_5$ production (as explained in section 2.1), correlation coefficients were also calculated for sunrise and sunset data separately. Because of the lower sampling rate, correlations are less robust if sunset and sunrise are investigated separately, but it appears that the anticorrelation is
more pronounced for sunrise data than for sunset data, though it is also found in the sunset data in some months. As \( \text{N}_2\text{O}_5 \) formation is highly temperature-dependent and favored by low temperatures, this might indicate a temperature effect.

[31] In the Northern Hemisphere, the picture is less clear; the signal starts in October and November with significant correlation coefficients both above 100 km and around 70 km. The strongest correlation coefficient is observed in December and January, which is also the period when the lowest altitudes of about 55 km are reached. However, a strong signal persists between 60 and 80 km until February and March, when the signal ceases quite abruptly. Much later during spring and early summer, significantly high correlation coefficients are observed at much lower altitudes, around 40 km. This signal continues well into summer (to June/July), but is not considered very robust because of the high values of the “bootstrap error.”

[32] The difference between both hemispheres is due to the differences in vortex stability during the years 1991–2005, with much less stable vortices and major stratospheric warming occurring in many northern winters as noted also by Randall et al. [2007]. The situation might have changed in recent years, when strong downwelling of thermospheric air into the stratosphere has also been observed in the Northern Hemisphere in several winters [Randall et al., 2006; Randall et al., 2009].

5. Conclusion

[33] We have investigated the interannual variation of \( \text{NO}_x \) in 15 years in middle to high latitudes throughout the year in both hemispheres. A correlation analysis of \( \text{NO}_x \) with solar radio flux, the \( \text{AE} \) index as a proxy for auroral electron precipitation, and the fluxes of relativistic electrons measured by both the GOES and POES instruments has shown that \( \text{NO}_x \) in the mesosphere and lower thermosphere is directly affected by electron fluxes with energies ranging from several keV to several hundreds of keV, above ~100 km also by solar radio flux. However, the most temporally stable and probably also the largest contribution to the overall \( \text{NO}_x \) budget appears to come from auroral electrons (1 keV to several tens of keV), which directly affect the altitude region 80–130 km and apparently precipitate quite frequently and independent of season, and from electrons with energies from 10 to 100 keV, which affect the altitude region 80–100 km, and whose impact is more frequently observed during autumn and spring. A small contribution appears to come also from electrons with energies of several hundreds of keV, which affect the altitude region 60–80 km, and are apparently correlated to the fluxes of highly relativistic electrons in the radiation belts as measured by GOES. However, these appear only sporadically, and the correlations observed between \( \text{NO}_x \) and GOES electron fluxes are smaller and less robust than the correlations between \( \text{NO}_x \) and the \( \text{AE} \) index, respectively, with the POES electron fluxes. This suggests that if a contribution of relativistic electrons to upper stratospheric or lower mesospheric \( \text{NO}_x \) exists as argued, e.g., by Sinnhuber et al. [2006] and Clilverd et al. [2009], it is significantly lower than the contribution of electrons of energies of less than 100 keV. However, it should be noted that the studies by Sinnhuber et al. [2006] and Clilverd et al. [2009] both consider polar winter at latitudes significantly higher than those covered by HALOE during winter. Also, highly relativistic electrons of energies of a few MeV might directly affect altitudes below 40 km, which we could not consider here (see section 2.1).

[34] During winter, a clear difference is observed between the Northern and Southern hemispheres in the lower mesosphere and upper stratosphere. In the Southern Hemisphere, \( \text{NO}_x \) from the mesosphere and lower thermosphere progresses farther down and lasts longer than in the Northern Hemisphere because of the different dynamic situations in Northern Hemisphere and Southern Hemisphere winters. As a strong downwelling of air has been observed in several recent winters [Smith et al., 2009, and references therein], the correlation for the Northern Hemisphere might be different if the data set could be extended to the present.

[35] Acknowledgments. The authors acknowledge funding by the Deutsche Forschungsgemeinschaft in the framework of the priority program Climate and Weather of the Sun–Earth System (CAWSES). M.S. gratefully acknowledges funding from the University of Bremen.

[36] Robert Lysak thanks the reviewers for their assistance in evaluating this paper.

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