

1   **Ionospheric evidence of thermosphere-to-stratosphere descent of polar NO<sub>x</sub>**

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5   **Abstract.** During the northern hemisphere winter of 2003-2004 significant levels of  
6   stratospheric odd nitrogen (NO<sub>x</sub>) were observed descending from the mesosphere. Here  
7   we study subionospheric radio wave propagation data from Ny Ålesund, Svalbard,  
8   Norway to determine the origin of the mesospheric NO<sub>x</sub>. A clear change in the radio  
9   wave diurnal variation is observed, starting on January 13, 2004, lasting for 37 days.  
10   The behavior is consistent with the ionization, by Lyman- $\alpha$ , of thermospheric NO<sub>x</sub>  
11   descending into the mesosphere from altitudes above 90 km. Estimates of the  
12   concentration of NO<sub>x</sub> required to produce the observed ionization changes are  
13   consistent with the levels of previously published stratospheric mixing ratios after the  
14   NO<sub>x</sub> has descended into the stratosphere. The radio wave data shows that no significant  
15   proton or electron precipitation events into the mesosphere occurred at this time, and the  
16   mesospheric effects of the large storms in October/November 2003 had abated by late  
17   December 2003.

## 1. Introduction

In this study we analyze ground-based ionospheric data from high latitudes during the northern polar winter of 2003-2004. Subionospheric VLF radio wave propagation is sensitive to changes in ionization at mesospheric altitudes, 50-90 km, including ionization of in situ  $\text{NO}_x$  by Lyman- $\alpha$  [Solomon et al., 1982a], and ionization by particle precipitation. Examples of ionization by particle precipitation are relativistic electron precipitation events during geomagnetic disturbances [Thorne and Larsen, 1976], as well as solar proton events [Westerlund et al., 1969; Clilverd et al., 2005] - including those that occurred in October 2003 [Clilverd et al., 2006]. The effect of increased ionization on propagating radio wave signals is seen as either an increase or decrease in signal amplitude or phase depending on the modal mixture of each signal observed. Using changes in ionospheric propagation conditions during the 2003-2004 winter we examine the changes in mesospheric  $\text{NO}_x$  concentration to contrast two possible source scenarios: descent from the thermosphere and in situ production via high energy particle precipitation, as described above.

Both energetic particle precipitation (EPP, >50 keV electrons, >1 MeV protons) into the mesosphere and low energy particle precipitation into the thermosphere (LEPP, <50 keV electrons, <1 MeV protons) generate enhancements in odd nitrogen. During the polar winter odd nitrogen can survive, and in the presence of strong polar vortex conditions, descend into the stratosphere [Solomon et al., 1982b]. During the northern polar winter of 2003-2004 these conditions existed; Randall et al. [2005] reported unprecedented levels of spring-time stratospheric  $\text{NO}_x$  as a result. Rinsland et al. [2005] also observed very high  $\text{NO}_x$  mixing ratios at 40-50 km in February/March 2004 with

the ACE experiment, detecting levels as high as 1365 ppbv. Although several powerful solar storms occurred at the beginning of the winter period (October and November) there is some uncertainty in the ultimate source of the enhanced  $\text{NO}_x$  because of the breakup of the upper stratospheric vortex in late December 2003. Rinsland et al. [2005] suggested that the EPP-produced  $\text{NO}_x$  could have survived the breakup of the polar vortex in late December, and would have experienced reduced downward transport during this time. Randall et al. [2005] suggested that further periods of EPP occurring after the large storms, and before the end of January, 2004, may have enhanced in situ mesospheric production. Another possibility is that the descent of high altitude auroral-produced  $\text{NO}_x$  (~120 km) caused the enhancement of mesospheric  $\text{NO}_x$  [Natarajan et al., 2004]. In either case the  $\text{NO}_x$  would then descend to the stratosphere in February/March 2004, as observed.

In this paper we show that the descending  $\text{NO}_x$  is first seen at altitudes between 65-90 km on January 13, 2004, about one month prior to the observations made at stratospheric altitudes (section 3). We identify auroral altitudes (~120 km) as the most likely source of the  $\text{NO}_x$  (section 4). We also show that the driver for in situ mesospheric production of  $\text{NO}_x$ , i.e., energetic particle precipitation events (EPP), are not present during this time, and that the effects of the large storms of October/November 2003 have abated in the mesosphere by the end of December 2003. We show that downward transport of thermospheric  $\text{NO}_x$  generated by LEPP is more plausible.

## 2. Experimental setup

Here we use narrow band subionospheric VLF/LF data spanning 20-40 kHz received at Ny Ålesund, Svalbard (79°N, 11°E,  $L=18.3$ ). This site is part of the Antarctic-Arctic Radiation-belt Dynamic Deposition VLF Atmospheric Research Konsortia (AARDDVARK). Figure 1 shows the location of the receiver site (diamond), and the VLF path used during the event period. The transmitter studied is located in Iceland (call-sign NRK) operating at 37.5 kHz. The 2,000 km VLF path is generally located within the winter polar vortex, and ranges in  $L$ -shell from  $L=6$ -18. Ionization effects on VLF/LF wave propagation can be modeled using the Long Wave Propagation Code [LWPC, Ferguson and Snyder, 1990] as long as the induced changes to the ionospheric electron density altitude-profiles are known. To provide this the Sodankylä Ion Chemistry model [SIC, Verronen et al., 2005] was used to determine the effects of increased ionization on the mesospheric electron density profiles. The average path conditions represented by the SIC modeling results were calculated at 70°N, 0°E.

## 3. Iceland to Ny Alesund Data

In Figure 2 we show the amplitude and phase of the Iceland transmitter (NRK, 37.5 kHz) received at Ny Ålesund. Each panel shows three lines. The solid line represents the quiet-day-curve (QDC) estimated from several days averaged throughout the winter period. As reported previously [Clilverd et al., 2006] the QDC exhibits around 7-8 dB lower amplitudes during the daylight hours (09-18 UT) than during the night, plus a phase advance of around 70°. This is as a result of the electron density profile (30-100 km) changing due to daytime solar photo-ionization, increasing the

attenuation of the signal on the propagation path. The two additional lines (dashed and dotted) represent days where the observed diurnal amplitude behavior of the signals is reversed. The daytime amplitudes are higher than at night by around 5 dB (i.e., higher by a factor of 1.8 because  $5\text{dB} = 20\log [1.8]$ ), and the nighttime amplitudes are depressed compared with the QDC by 2-5 dB. The daytime phase advance is larger than for the QDC, extending to about  $100^\circ$ . This is indicative of non-QDC changes in the electron density profiles, caused by processes enhancing ionization levels at some or all mesospheric altitudes. Determining the cause of this change in behavior, and its occurrence frequency during the northern polar winter, allows us to determine whether the increase in mesospheric ionization is a result of either the ionization of descending  $\text{NO}_x$  or ionization directly caused by EPP at any given time.

In Figure 3 we plot the amplitude data for the whole winter period (1 October 2003 – 20 April 2004). The top panel shows the difference between the amplitude of NRK during the day compared with that during the night. The times when solar proton events have been identified (<http://umbra.nascom.nasa.gov/SEP/>) are given by solid vertical lines, and the peak particle flux unit (pfu) for  $>10$  MeV protons  $\text{cm}^{-2}\text{sr}^{-1}\text{s}^{-1}$  is given in brackets. The average QDC value of -7 dB for the winter period is shown by the horizontal dot-dashed line. High daytime amplitude values occur during and after the solar storms of October/November 2003 (day 25-70) – these are indicative of enhanced ionization effects. The figure shows that the mesospheric effects of the October/November 2003 proton events had ended by day 80 (19 December, 2003), which is consistent with GOMOS observations of the recovery of  $\text{NO}_x$  concentrations in the upper stratosphere during December [Hauchecorne et al., 2005]. These

observations indicate that the large solar storms of October/November 2003 are not responsible for the enhanced  $\text{NO}_x$  observed in the mesosphere in January/February 2004 [Randall et al., 2005; Rinsland et al., 2005]. In section 4 we discuss why  $\text{NO}_x$  produced in the thermosphere in October/November 2003 could not have survived long enough to descend into the mesosphere in January 2004.

The lower panel of Figure 3 shows the average nighttime amplitude of NRK. QDC levels are again indicated by a horizontal dot-dashed line (54 dB relative to an arbitrary amplitude level). The October/November 2003 solar storms are associated with lower nighttime NRK amplitudes, typically about 5 dB lower than the nighttime QDC. Both the panels in this figure indicate that enhanced ionization during solar proton events changes the diurnal behavior of the NRK signal. Following the solar proton events, day 105 (January 13, 2004) marks the identification of behavior consistent with enhanced ionization, i.e., high daytime amplitude and low nighttime amplitude. This is shown by the vertical dashed line, although the signature of the event actually takes  $\sim 2$  days to change from QDC levels to those associated with enhanced ionization, indicating a slow onset of the event. However, no associated proton event, or solar storm, either of which might be expected to produce EPP, can be identified at this time. The effect lasts until day 142 (February 19, 2004), i.e., the period lasts 37 days. A second similar period starting on October 22, 2003, is also indicated by a vertical dashed line, but the effects are soon merged with the large solar proton events that occurred a few days later.

#### **4. Descent of High Altitude $\text{NO}_x$**

In order to model the effects of enhanced mesospheric ionization on the Iceland to Ny Ålesund path the SIC model was run with an arbitrary ionization source defined by a proton precipitation energy spectrum based on GOES-11 measurements. We use various additional ionisation rates to drive the SIC model so as to determine the size of the ionisation change required to reproduce the radio wave data. The ionisation rates are characterised in terms of particle fluxes to allow later contrast, even though we only focus on the effects of additional ionisation. To investigate the sensitivity to a range of flux values we scale the  $>50$  MeV flux levels. These are typically  $0.050 \text{ protons cm}^{-2}\text{sr}^{-1}\text{s}^{-1}$  during quiet times, increasing to  $>1000 \text{ protons cm}^{-2}\text{sr}^{-1}\text{s}^{-1}$  during large solar proton events. The ionization of atmospheric constituents changes the electron density altitude profile. The electron density profiles in the altitude range 30-100 km can be applied to the wave propagation code LWPC, and thus the resulting amplitude and phase changes can be calculated for propagation on this path.

Figure 4 shows the result of differing levels of ionization on the Iceland to Ny Ålesund signal. The QDC is given by the solid line, and shows lower amplitudes values during the day by about 5 dB, and a phase advance of about  $100^\circ$  consistent with, but slightly larger than, the observed data. The dotted and dashed lines show the effects of ionization for  $>50$  MeV proton fluxes of 2.5 and  $25 \text{ cm}^{-2}\text{sr}^{-1}\text{s}^{-1}$  respectively. These flux levels represent peak particle flux units (pfu) of  $\sim 10$  and  $100 \text{ protons cm}^{-2} \text{sr}^{-1} \text{s}^{-1}$  for energies  $>10$  MeV, i.e., small solar proton events. The  $2.5 \text{ cm}^{-2}\text{sr}^{-1}\text{s}^{-1}$  fluxes (equivalent to a pfu of 10) lower the nighttime amplitude levels by 2-3 db, and produce increased daytime amplitudes, particularly during sunset. They also increase the phase advance by  $20^\circ$  compared with the QDC. The model response to these flux levels broadly agrees

with the data. In contrast the higher flux levels of  $25 \text{ cm}^{-2} \text{ sr}^{-1} \text{ s}^{-1}$  (equivalent to a pfu of 100) produce a variation similar to the QDC even though there are significant changes to the electron density profiles.

The data presented in Figure 3 confirms that small solar proton events significantly affect the Iceland to Ny Ålesund signal. However, the anomalous period starting January 13, 2004 can not be linked to any satellite-observed enhanced solar proton precipitation or energetic electron precipitation. No clear signature of high geomagnetic activity ( $A_p$  or  $D_{st}$ ), or high speed solar wind can be found that is associated with this period, so the cause is not obviously linked to EPP production of  $\text{NO}_x$  directly in the mesosphere. Additionally the onset time of the event is  $\sim 2$  days, rather than minutes-hours as in EPP events, suggesting a slower more gradual build-up of ionization.

Ionization of  $\text{NO}_x$  by Lyman- $\alpha$  is known to have significant influence on mesospheric electron density profiles. One source of  $\text{NO}_x$  in the mesosphere arrives by vertical transport from auroral altitudes (120 km), where low energy precipitation (LEPP) leads to large increases in energy deposition in that region [Solomon et al., 1982b]. We have shown that the mesospheric  $\text{NO}_x$  produced by the solar storms of October/November 2003 had abated by mid-December 2003 as a result of NO photolysis during the daylight that occurs at 80 km at  $\sim 70^\circ\text{N}$  during the winter months. Similarly the  $\text{NO}_x$  produced in the thermosphere at about 120 km by the storms of October/November 2003 will have disappeared because of NO photolysis during the increased hours of sunlight at these higher altitudes. Semeniuk et al. [2005] showed that the October/November 2003 storms could only produce sufficient  $\text{NO}_x$  to match the FTS observations if the  $\text{NO}_x$  were descended immediately after the storms. The delay of



two months before the actual descent would have significantly reduced the amounts of  $\text{NO}_x$  remaining to levels that would not be significant in terms of the observed mixing ratios in January/February 2004. The smaller storms that occurred after October/November 2003 are likely to have produced significant quantities of  $\text{NO}_x$  at auroral altitudes [Barth et al., 2003]. This would descend during the winter in the presence of the strong polar vortex and downward transport that occurred following the end of the stratospheric warming period at the end of December 2003. Similar effects are regularly observed in the Antarctic polar region even in solar minimum years [Siskind et al., 2000; De Zafra and Smyshlyaev, 2001] strongly suggesting that even moderate levels of solar activity can cause the production of sufficient levels of thermospheric  $\text{NO}_x$  for it to be observed as enhanced  $\text{NO}_x$  descending through the mesosphere to the stratosphere.

Solar Lyman- $\alpha$  radiation (and geocoronal Lyman- $\alpha$  at night) ionizes NO at altitudes of 65-95 km. As a result the descent of  $\text{NO}_x$  from higher altitudes would increase the rate of ionization in the mesosphere, and thus affect the electron density profiles in much the same way as in situ energetic particle precipitation [Solomon et al., 1982a]. The changes observed in the Iceland to Ny Ålesund radio wave data observed from January 13, 2004 are most likely to indicate the start of significant levels of downward  $\text{NO}_x$  transport into the mesosphere from the thermosphere, produced by auroral activity in December and early January, and not the start of a long-lived EPP event producing it in situ (but undetected by satellite borne sensors) or the continuation of the mesospheric effects of the October/November 2003 storms (Figure 3 shows that they had abated by 19 December 2003). The downward transport is consistent with the dynamical

variability shown by the polar vortex at this time. The typical levels of 80 km NO number densities as a result of the descent of thermospheric NO<sub>x</sub> are  $\sim 10^9 \text{ cm}^{-3}$  (c.f.  $\sim 10^7 \text{ cm}^{-3}$  for normal conditions) [Solomon et al., 1982a]. Converting this enhanced level to a mixing ratio as above results in  $\sim 2000$  ppbv consistent with the 1365 ppbv observed by Rinsland et al., [2005].

If the amount of mesospheric NO increased by a factor of 100, then the electron density levels (from Lyman- $\alpha$  ionization) would increase by a factor of  $\sim 10$  because the recombination rate is proportional to both the electron and ion densities. This would cause the ionospheric reflection height for VLF/LF waves to lower by  $\sim 10$  km, which is consistent with the changes produced by the test ionization run in Figure 4 (2.5 flux line) and could therefore explain the anomalous radio wave data in this study.

## **5. In situ Production of Odd Nitrogen**

The Ny Ålesund data is consistent with a picture of quasi-constant excess ionization occurring for 37 days during January and February 2004. We examine the NO<sub>x</sub> change which would be produced by some as yet undetected EPP, with flux sufficient to explain radio wave observations. We can test to see if this level of continuous particle precipitation could generate enough NO<sub>x</sub> in situ in the mesosphere. Our tests used a continuous small proton event with pfu of 5-50, although the effects could have been generated by equivalent electron precipitation. The changes in NO<sub>x</sub> levels were produced by a proton energy spectrum with  $>50$  MeV fluxes of  $2.5 \text{ protons cm}^{-2}\text{sr}^{-1}\text{s}^{-1}$  (equivalent to a pfu of 10) and the maximum effect of the precipitation was at 70 km altitude. Because of the quasi-constant precipitation the level of NO<sub>x</sub> reached an

equilibrium state with a factor of  $\sim 2$  increase. The limiting factor in this increase is because of NO photolysis during the  $\sim 6$  hours of daylight at the SIC modeling location at mesospheric altitudes, where the lifetime of NO against photolysis is only a few days. The SIC model tells us that at 80 km this increase corresponds to NO levels of  $5 \times 10^7 \text{ cm}^{-3}$ . Estimating the neutral atmosphere number density from MSIS as  $5 \times 10^{14} \text{ cm}^{-3}$  results in a mixing ratio of 100 ppbv, with quiet time levels of 50 ppbv. Both of these values are close to the normal background mixing ratio values observed by ACE and not with those associated with the layer of enhanced NO<sub>x</sub> [Rinsland et al., 2005]. Thus the EPP-levels which would explain the radio wave observations only lead to doubling in NO<sub>x</sub> levels, and cannot account for the observed NO<sub>x</sub> increases. Again, transport of thermospheric NO<sub>x</sub> generated by LEPP is more plausible, and more significant, than generation of NO in situ by low-levels of (undetected) EPP.

## 6. Discussion and Summary

During the northern hemisphere winter of 2003-2004 significant levels of stratospheric odd nitrogen (NO<sub>x</sub>) were observed that appeared to be descending from the mesosphere [Randall et al., 2005; Rinsland et al., 2005]. The origin of this mesospheric NO<sub>x</sub> was unclear, either being produced in situ ( $\sim 80$  km) by energetic particle precipitation (EPP), or by descending high altitude NO<sub>x</sub> possibly produced by softer particle precipitation (LEPP) in the auroral zone ( $\sim 120$  km) during or after the large solar storms in October/November 2003.

In this study we investigate the effects of either in-situ particle precipitation in the mesosphere, or ionization of high altitude NO<sub>x</sub> descending into the mesosphere, on

subionospheric radio wave propagation data from Ny Ålesund, Svalbard, Norway. EPP could not be responsible for the in situ formation of the mesospheric NO<sub>x</sub> observed in the radio wave data on January 13, 2004, as no elevated geomagnetic activity or high solar wind speed occurred at the time of the observed anomalous subionospheric signals. Additionally, calculations show that NO<sub>x</sub> produced in situ by EPP which would explain the radio wave data would be only ~2 times the normal levels, not enough to account for the observed NO<sub>x</sub> mixing ratios at 40-50 km [Randall et al., 2005].

The observed radio wave data is more consistent with the descent of high altitude thermospheric NO<sub>x</sub> into the mesosphere starting January 13, 2004, generated by LEPP at ~120 km, brought down by enhanced vertical transport following the end of the stratospheric warming event at the end of December 2003. We find that the NO<sub>x</sub> produced by the solar storms of October/November 2003 had abated by mid-December 2003 due to NO photolysis. Smaller storms that occurred after November 2003 are likely to have produced sufficient quantities of NO<sub>x</sub> at auroral altitudes for it to be observed as enhanced NO<sub>x</sub> descending through the mesosphere to the stratosphere. The extraordinary solar activity in October/November 2003 was not required in order to see the extraordinary NO<sub>x</sub> enhancements in January/February 2004, these enhancements occurred under more typical solar conditions in late December 2003 and early January 2004. The ionization of enhanced NO by Lyman- $\alpha$  radiation would affect the mesospheric electron density profiles in much the same way as in situ ionization from proton precipitation, but only if the NO concentration was ~10-100 times the normal levels. The descent of these high levels of NO during the northern Spring 2004 would reproduce the stratospheric mixing ratios observed at that time [Rinsland et al., 2005].

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(Received N x, 2005; N x 27, 2005;  
accepted N x, 2005.)

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**Figure 1.** The location of the subionospheric propagation path from Iceland to the AARDDVARK receiver site at Ny Ålesund. The Sodankylä Ion Chemistry modeling location is also shown.

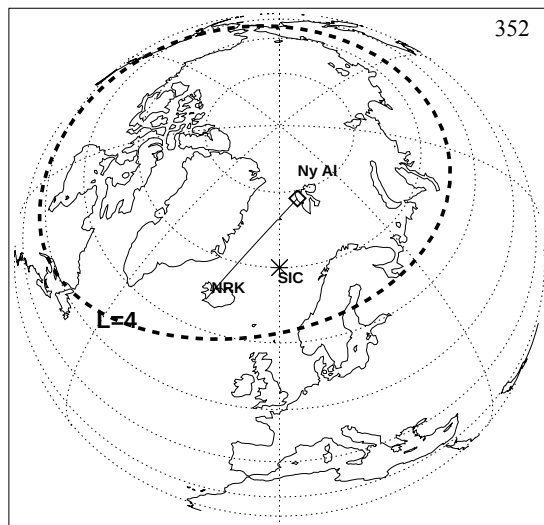
**Figure 2.** The amplitude and phase of the Iceland (NRK, 37.5 kHz) transmitter received at Ny Ålesund, Svalbard, Norway for quiet day conditions (solid line) and two representative anomalous days (dashed and dotted lines).

**Figure 3.** The winter-time differences in day-night amplitude, and average nighttime amplitude for the Iceland transmitter received at Ny Ålesund. Normal values are indicated by the horizontal dashed line. Times of identified solar proton events are given by solid vertical lines, while similar behavior with no identified proton event are shown by the vertical dotted line.

**Figure 4.** The modeled amplitude and phase of the Iceland transmitter received at Ny Ålesund, Svalbard, under the influence of differing levels of proton precipitation flux.

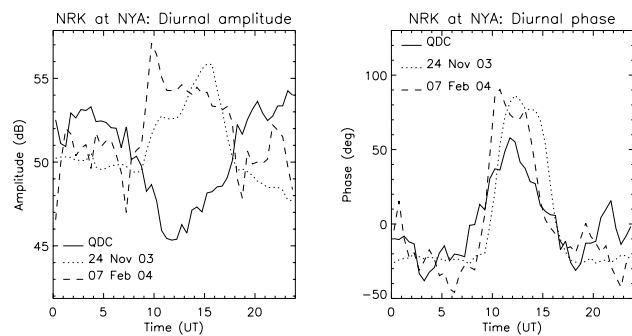


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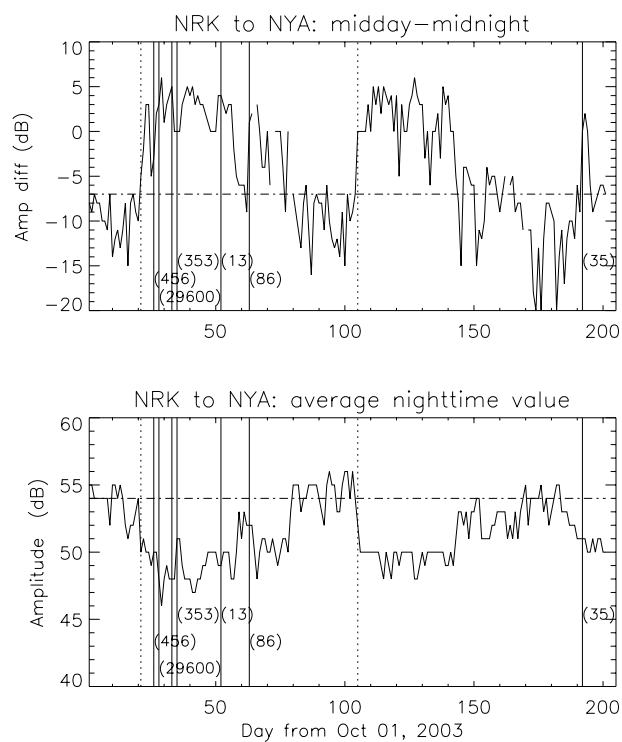


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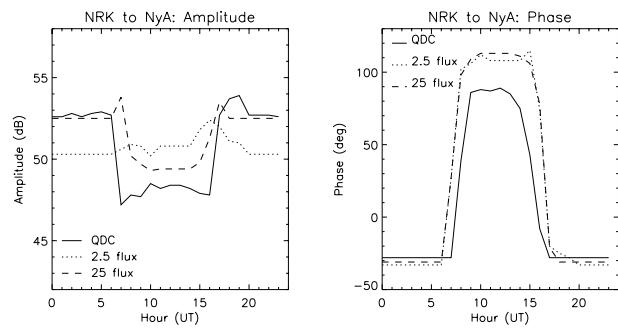
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