1	Impact of different	energies of	precipitating	particles on NO _x
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2 generation in the middle and upper atmosphere during geomagnetic

3 storms.

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8 ^a Sodankylä Geophysical Observatory, Tähteläntie 62, FI-99600 Sodankylä, Finland. ^b Earth Observation, Finnish Meteorological Institute, P.O. Box 503, FI-00101 9 Helsinki, Finland. 10 ^c Department of Physics, University of Otago, P.O. Box 56, Dunedin, New Zealand. 11 12 ^d Physical Sciences Division, British Antarctic Survey (NERC), High Cross, Madingley Road, Cambridge CB3 0ET, England, U.K. 13 14 15 Corresponding author: Mark Clilverd email: macl@bas.ac.uk tel: +44 1223 221541 fax: +44 1223 221226 16 17 18 Keywords: Solar wind, geomagnetic storms, energetic precipitation, nitric oxide, 19 ozone 20 21 Abstract: Energetic particle precipitation couples the solar wind to the Earth's 22 atmosphere and indirectly to Earth's climate. Ionisation and dissociation increases, 23 due to particle precipitation, create odd nitrogen (NO_x) and odd hydrogen (HO_x) in 24 the upper atmosphere, which can affect ozone chemistry. The long-lived NO_x can be transported downwards into the stratosphere, particularly during the polar 25 26 winter. Thus the impact of NO_x is determined by both the initial ionisation 27 production, which is a function of the particle flux and energy spectrum, as well as 28 transport rates. In this paper we use the Sodankylä Ion and Neutral Chemistry 29 model (SIC) to simulate the production of NO_x from examples of the most realistic 30 particle flux and energy spectra available today of solar proton events, auroral energy electrons, and relativistic electron precipitation. Large SPEs are found to produce 31

higher initial NO_x concentrations than long-lived REP events, which themselves produce higher initial NO_x levels than auroral electron precipitation. Only REP microburst events were found to be insignificant in terms of generating NO_x. We show that the GOMOS observations from the Arctic winter 2003-2004 are consistent with NO_x generation by a combination of SPE, auroral altitude precipitation, and long-lived REP events.

38

39 1. Introduction

40 Over the past few years the importance of High Speed Solar Winds Streams 41 (HSSWS) in the solar wind has become increasingly accepted as a driver of 42 geomagnetic activity within the Earth's magnetosphere (Friedel et al., 2002). While 43 Coronal Mass Ejections (CMEs) are the main source of geomagnetic storms at solar 44 maximum, the declining and minimum phase of solar activity is characterised by an 45 increase in the occurrence rate of high-speed (>500 km/s) solar wind streams 46 emanating from coronal holes (Richardson et al., 2001).

47 Although HSSWS events are not typically associated with large signatures in 48 the D_{st} index (max >-50 nT), they do produce moderate levels of geomagnetic activity 49 which persists for many days. In contrast CME events are more transient, driving high 50 geomagnetic activity for typically only 1-2 days (Richardson et al., 2000). As such, 51 the energy input to the magnetosphere during HSSWS events is comparable to or may 52 exceed the energy input to the magnetosphere during CMEs. There are more 53 significant electron flux enhancements in HSSWS-driven storms compared to CME-54 driven storms, the flux of higher-energy particles peak later in time, and many 55 magnetospheric electromagnetic wave processes are enhanced (Hilmer et al., 2000; 56 Vassiliadias et al., 2007). Energetic particles are ultimately lost to the atmosphere 57 through interactions with magnetospheric waves such as chorus, plasmaspheric hiss,

electromagnetic ion cyclotron waves (EMIC), and Pc5 micropulsations. The altitudes
at which these particles deposit their momentum is dependent on their energy
spectrum, with lower energy particles impacting the atmosphere at higher altitudes
than their more energetic relatives (Rees, 1989; Rodger et al., 2007a).

62 Precipitating particles affect the neutral chemistry of the middle atmosphere. 63 Direct particle impact on N₂ and ion chemical reactions produces atomic nitrogen, 64 part of which reacts to form NO_x (Rusch et al., 1981). Additionally, ion chemistry, 65 involving production and recombination of water cluster ions, leads to production of 66 HO_x (Solomon et al., 1981). Both NO_x (N+NO+NO₂) and HO_x (H+OH+HO₂), 67 although being minor gases, are important catalysts that participate in ozone loss 68 reactions in the upper stratosphere and mesosphere, respectively (Grenfell et al., 69 2006). Ozone, on the other hand, affects the radiative balance, temperature, and 70 dynamics of the atmosphere due to its capability of absorbing solar UV radiation 71 efficiently (Brasseur and Solomon, 2005).

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73 1.1 Solar proton events

74 In the upper stratosphere, larger solar proton events (SPEs) can produce order-75 of-magnitude changes in NO_x concentrations which, due to the long photochemical 76 lifetime of NO_x, can last for months. Related decreases in ozone have been observed 77 to be of the order of several tens of percent (Seppälä et al., 2004; Lopez-Puertas et al., 78 2005). In the presence of a strong underlying polar vortex, NO_x produced above the 79 stratosphere in the MLT region can descend into the stratosphere (Randall et al., 80 2005). In 2003-2004 these conditions were highlighted by Randall et al. (2005) who 81 reported unprecedented levels of spring-time stratospheric NO_x as a result. Rinsland et 82 al. (2005) also observed very high NO_x mixing ratios at 40-50 km in February/March 2004 with the ACE experiment, detecting levels as high as 1365 ppbv. Seppälä et al. (2007b) showed that the NO_x descent period of 2003/04 contained 4 periods of NO_x production, beginning with in-situ stratospheric production by the solar proton events of October/November 2003, and culminating in the descent of thermospherically generated NO_x in January/February 2004. Both Siskind et al. (2000) and Seppälä et al. (2007a) showed that stratospheric NO_x concentrations at altitudes between 23-55 km were well correlated with geomagnetic activity levels (monthly average A_p).

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1.2 Auroral electron precipitation

92 Thermospheric NO concentration is known to enhance significantly during 93 auroral electron precipitation (Barth et al., 2001). The increase of NO in the lower 94 thermosphere at the NO maximum is related to the relative abundances of nitrogen atoms in the ground state $N(^4S)$ and the first excited state $N(^2D)$, which is mainly 95 produced by dissociation of N₂ by precipitating electrons, recombination of NO⁺ and 96 atomic oxygen reacting with N_2^+ ions. The first satellite observations of NO (Rusch 97 98 and Barth, 1975) showed increased amounts of NO in the polar regions. Since then 99 several studies have shown good correlation between satellite observations of 100 thermospheric NO concentration and various indicators of auroral electron 101 precipitation (e.g., Petrinec et al., 2003).

102 A significant fraction of large geomagnetic storms are associated with 103 relativistic electron population increases in the outer radiation belt and the slot region. 104 During storms outer radiation belt fluxes show strong drop-outs in relativistic fluxes, 105 partially due to the D_{st} effect, and partially due to precipitation losses. The increased 106 population decays in part through the loss, i.e., precipitation into the middle and upper

107 atmosphere, over timescales of days to weeks driven by plasmaspheric hiss (Rodger et108 al., 2007b).

109 Even relatively mild geomagnetic disturbances increase the flux into the 110 atmosphere of the relatively low energy electrons that are associated with the aurora 111 (energies 1-10 keV), while large geomagnetic storms can produce relativistic electron 112 precipitation (REP). Energetic electron precipitation from the Van Allen radiation 113 belts occur on different timescales with differing energy ranges, driven by a wide variety of mechanisms. At this point there are still significant gaps in our 114 115 understanding of the processes involved. Essentially all geomagnetic storms 116 substantially alter the electron radiation belt populations (Reeves et al., 2003), in 117 which precipitation losses play a major role (Green et al., 2004). A significant fraction 118 of the energetic particles are lost into the atmosphere (Clilverd et al., 2006a), although 119 storm-time non-adiabatic magnetic field changes also lead to losses through 120 magnetopause shadowing (e.g., Ukhorskiy et al., 2006).

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122 **1.3 Relativistic electron precipitation**

123 Multiple observations exist of REP on very short and relatively long time 124 scales. For example, relativistic electron microbursts are bursty, short duration (<1 s) 125 precipitation events containing electrons of energy >1 MeV (Imhof et al., 1992; Blake 126 et al., 1996). Observations from the SAMPEX satellite show that REP microbursts 127 occur at about L=4-6, and are observed predominantly in the morning sector. 128 Primarily because of this local time dependence microbursts have been associated 129 with very low frequency (VLF) chorus waves, although this is not consistent with the 130 ionospheric signature of rapid REP (Rodger et al., 2007a). Estimates of flux losses 131 due to relativistic microbursts show that they could empty the radiation belt in about a 132 day (Lorentzen et al., 2001), and thus might produce a significant impact upon the 133 neutral atmosphere. In contrast, precipitation events lasting minutes to hours have 134 been observed from balloon-borne sensors (e.g., Millan et al., 2002) and 135 subionospheric VLF measurements (Clilverd et al., 2006a). They occur at about L=4-7, are observed in the late afternoon/dusk sector. The observed loss rates also suggest 136 137 that these minute-hour events could be the primary loss mechanism for outer zone 138 relativistic electrons, although as with the case of REP microbursts significant 139 assumptions are used in the loss rate estimates. It has been suggested that this 140 precipitation may be caused by EMIC waves (Millan et al., 2002), which has been 141 supported by EMIC observations during REP activity (Clilverd et al., 2007a).

142 Evidence of (thermospheric) NO being transported from auroral to lower 143 latitudes after major geomagnetic storms is seen in 1-D modelling and observational 144 studies based on SNOE data (Barth et al., 2003; Barth and Bailey, 2004). However, a 145 consistent approach to latitudinal as well as vertical transport of long-lived NO_x can 146 only be provided by 3D general circulation models. Dobbin et al. (2006) compared 147 runs of their 3D global circulation model CMAT with and without auroral forcing, representative of auroral energy input corresponding to K_p values of 2^+ and 6^- for 148 149 moderate and high activity respectively. CMAT simulations suggest that under 150 moderate geomagnetic conditions, the most equatorward geographic latitudes to be 151 influenced by aurorally produced NO are 30°S and 45°N. Under conditions of high geomagnetic activity, aurorally produced NO is present at latitudes poleward of 15°S 152 153 and 28°N.

In this study we discuss the significance of relatively mid-energy (auroral) electron precipitation, and high-energy relativistic electron precipitation events of different durations, on the neutral atmosphere in the polar regions. This is undertaken 157 in comparison with the effect of solar proton events. The auroral altitude electron 158 precipitation is preferentially related to the occurrence of HSSWS in comparison with 159 CME. Relativistic electron precipitation events are produced by large geomagnetic 160 storms triggered by CME and HSSWS. We model the altitude and effectiveness of NO_x using the Sodankylä Ion and Neutral Chemistry model using the most realistic 161 162 particle flux and energy spectra available today. We also discuss the evidence that the 163 stratospheric polar vortex transports the precipitation-generated NO_x into the 164 stratosphere, and how it can then affect stratospheric winds and temperatures.

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166 **2. Modelling chemistry effects of particle precipitation**

167 Particle precipitation affects the ion-chemistry of the atmosphere. Here we 168 particularly concentrate on the odd nitrogen (NO_x) effects, which will in turn impact 169 ozone concentration, leading to effects on atmospheric dynamics. Figure 1 shows the 170 reaction pathways driven by energetic particle precipitation into the stratosphere, 171 mesosphere, and lower thermosphere. NO_x gases N, NO, and NO₂ are formed primarily in the stratosphere through the reaction $N_2O + O(^1D) \rightarrow 2NO$, and in the 172 thermosphere through both photodissociation and photoionisation of N₂. Precipitating 173 174 charged particles produce NO_x through ionisation or dissociative ionisation of N₂ and O_2 molecules, which results in the formation of N_2^+ , O_2^+ , N^+ , O^+ , and NO^+ . The 175 reactions of these ions lead to formation of both the excited nitrogen atoms $N(^{2}D)$ and 176 the ground state of nitrogen N(⁴S) (Rusch et al., 1981; Solomon et al., 1982). Almost 177 178 all of the excited nitrogen reacts with O₂ to form NO, providing a significant pathway 179 to NO production. The NO produced is converted into NO₂ below ~65 km altitude in 180 various reactions (see e.g. Brasseur and Solomon, 2005, pp. 336-341), but the 181 production of NO₂ is balanced by conversion back to NO either in reaction with O, or by photolysis, the outcome of this balance giving the relative concentrations of NO
and NO₂. During nighttime, when little O is available, and the above reactions are
ineffective, all NO is rapidly converted to NO₂ after sunset.

185 The production of excess amounts of long-lived NO_x in the lower 186 thermosphere and mesosphere can be modelled with a coupled ion-neutral chemistry 187 model. Successful modelling of high energy particle precipitation effects during solar 188 proton events has been recently undertaken using the Sodankylä Ion and Neutral 189 Chemistry model, also known as SIC, which is a 1-D tool for ionosphere-atmosphere 190 interaction studies (Turunen et al., 1996; Verronen et al., 2002). The model is readily 191 applicable in order to accurately model the production-loss balance of NO_x, and its 192 time development, in the cases of both auroral and relativistic electron precipitation.

193 The first version of the model was developed in the late 1980s to facilitate 194 ionospheric data interpretation. A detailed description of the original SIC model, which solved the ion composition only, can be found in Turunen et al. (1996). The 195 196 latest version (v. 6.9.0) solves the concentrations of 65 ions, of which 36 are positive 197 and 29 negative, as well as 15 minor neutral species. A recent, detailed description of 198 SIC is given by Verronen et al. (2005) and Verronen (2006). Below we briefly 199 summarise the main details of the model. The altitude range of SIC is from 20 to 200 150 km, with 1-km resolution. The model includes a chemical scheme of several 201 hundred reactions, and takes into account external forcing due to solar UV and soft X-202 ray radiation, electron and proton precipitation, and galactic cosmic rays. The 203 background neutral atmosphere is generated using the MSISE-90 model (Hedin, 204 1991) and tables given by Shimazaki (1984). The solar flux is estimated by the 205 SOLAR2000 model (Tobiska et al., 2000), version 2.27. The scattered component of 206 the solar Lyman- α flux is included using the empirical approximation given by

Thomas and Bowman (1986). The model includes a vertical transport scheme, as described by Chabrillat et al. (2002), which takes into account both molecular and eddy diffusion. Within the transport code the molecular diffusion coefficients are calculated according to Banks and Kockarts (1973). The Eddy diffusion coefficient profile can be varied using the parameterisation given by Shimazaki (1971). Figure 2 shows a graphical representation of the SIC model structure, showing the inputs, dependencies, and processes with the model.

214 The production of NO_x in the SIC model is dependent on the particle energy 215 spectrum, with lower energy particles ionising the atmosphere at higher altitudes than 216 their more energetic relatives. The left panel of Figure 3 shows the ionisation rates 217 due to a monoenergetic beam of protons with energies spanning 1-1000 MeV, in each case with a flux of 1 proton $\text{cm}^{-2}\text{s}^{-1}\text{sr}^{-1}$. The ionisation rate calculation is based on 218 219 proton energy-range measurements in standard air (Bethe and Ashkin, 1953), as 220 described in detailed by Verronen et al. (2005). These ionisation rate calculations 221 should be contrasted with those given in the right panel, presenting the rates for monoenergetic electron beams with a flux of 100 electrons $cm^{-2}s^{-1}sr^{-1}$. The energy 222 223 ranges span from 4 keV to 10 MeV, representing relatively high energy auroral 224 electrons through to very hard REP. The electron ionisation rates make use of the 225 expressions given by Rees (1989, chapter 3), with effective electron ranges taken 226 from Goldberg and Jackman (1984). In the section below we use the SIC model to 227 simulate the response of the atmosphere to different cases of energetic particle 228 precipitation.

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230 **3. Results from SPE, auroral, and REP case studies**

231 Here we model four separate cases of energetic particle precipitation using the 232 SIC model to simulate the production of NO₂ using the most realistic particle flux and 233 energy spectra available today. For each model run, we also perform a control run. 234 We convert the precipitation energy spectra into ionisation rates as a function of 235 altitude as discussed above, and run these through the SIC model using realistic fluxes 236 at 70°N, 0°E in northern hemisphere winter conditions. We are then able to compare 237 the increase in NO_x and electron number density as a result of the precipitation. The 238 case events studied here are: an example of high levels of proton precipitation flux as 239 seen in the Halloween storm of October 2003; pulsed auroral precipitation from Ulich 240 et al. (2000); relativistic electron precipitation from Gaines et al. (1995) lasting 241 several hours such as those observed by balloon experiments (Millan et al., 2002); and 242 relativistic microbursts from Rodger et al. (2007a).

243

244 **3.1 Solar proton events**

245 The modelled response of the middle atmosphere to a solar proton event is 246 presented in Figure 4 as an example of an event with high levels of proton 247 precipitation. The proton flux levels are taken from the GOES-11 spectra. Electron 248 concentrations shown in the upper panel are enhanced by several orders of magnitude 249 during the periods of extreme forcing. This is directly dependent on the ionisation rate 250 and thus can be used for monitoring the magnitude of the forcing below 80 km. The 251 effects of the SPE events on 28-31 October 2003 and 3-5 November 2003 are easily 252 identified in the upper panel of the figure as increases in electron number density at 253 60-100 km altitudes, particularly on 28 October, and 3 November. Once affected by 254 proton forcing, the concentrations of NO_x (N + NO + NO₂) shown in the lower panel 255 stays at an elevated level. The recovery is slow because of the long chemical lifetime 256 of NO_x especially at high solar zenith angles (SZA, see, e.g., Brasseur and Solomon, 257 2005, pp. 327-358). On 26-27 October, electron number density is enhanced at 60-258 80 km but little change can be seen in the NO_x . However, the largest event on 28–31 259 October leads to enhancements of NO_x and electron number density of several 260 hundred per cent at altitudes above 40 km. In contrast, the effect of the 3–5 November 261 event is small on the already elevated NO_x levels. At altitudes >100 km a diurnal variation in electron number density and NOx can be seen as part of the normal SZA-262 263 driven variability, but the SPE has little influence on it due to the energy of the 264 protons.

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266 **3.2 Auroral electron precipitation**

267 Figure 5 shows the modelled impact of several bursts of auroral electron 268 precipitation on electron number density (upper panel) and NO concentration (lower 269 panel). The electron bursts last 5 min each, starting at 23:05, 23:25, and 23:45 LT. 270 This example is based on studies of bursty aurora of the type observed by Ulich et al. 271 (2000). Assuming that the energy in auroral structures is deposited in monoenergetic 272 sheets embedded within wider regions of electron precipitation with a spread of 273 energies, we can describe the electron spectra as having a combination of Maxwellian 274 and monoenergetic forms, where the monoenergetic component is represented 275 typically by a Gaussian with 10% half-width. This kind of spectrum is called an 276 'inverted-V', and it is a common form in auroral structures with latitudinal widths 277 between 100 m and 100 km (McFadden et al., 1990; Lanchester et al., 1998). The 278 bursts of electrons have a characteristic energy of 5 keV and a total energy flux of 10 mW m⁻², of which 75% is in the monoenergetic component and 25% in the 279 280 Maxwellian component. A large fraction of the total energy is deposited at altitudes

around 110 km by the monoenergetic part, while the Maxwellian part, which has a
wide energy distribution, affects altitudes above 85 km.

283 The response of electron number density to the precipitation of energetic 284 electrons is immediate. The effect is largest at around 110 km altitude, where the 285 density increases by nearly two orders of magnitude. After the first precipitation burst, 286 the electron number density decreases exponentially reaching, after 15 min, a level 287 about one order-of-magnitude larger than the background level prior to the burst. The 288 second and third burst repeats this behaviour in much the same way. After the third 289 burst, electron number densities decrease to values within about 50% of the 290 background after some 2 hours. Between 05:00 and 06:00 LT the Sun begins to rise 291 and solar radiation ionises the atmosphere.

292 Like the electron number density, the NO concentration shown in the lower 293 panel of Figure 5 responds immediately to the precipitation, increasing by a factor of 294 about 1.35 at 115 km, where the relative effect is most pronounced. However, while 295 the electron number density decreases rapidly after the bursts, the NO concentration 296 decreases by only about 10% during the 15 min before the next burst. The second 297 burst causes the NO concentration to increase by a factor of 1.3 from the already 298 elevated level. After an intermediate decrease of 15% or so, the third burst brings 299 about another increase by a factor of about 1.25. Thus there is a cumulative effect of 300 the subsequent electron precipitation bursts on the NO concentration, which is 301 gradually 'pumped up' to about 1.75 times the background level during the third 302 burst. Thereafter it settles after some 2 hours at about 1.65 times the background and 303 remains fairly constant during the rest of the night. After sunrise the NO concentration 304 begins to decrease slowly, and at noon on the following day it is still about 1.25 times higher than that of the control run. The amount of NO produced depends on the total 305

306 energy input, which is a product of the energy flux and the duration of the 307 precipitation. Fifteen minutes of bursts with 10 mW m⁻² flux results in a total energy 308 input per unit area of 9.0 J m⁻², which is a moderate input compared to some other 309 studies (e.g., Barth et al., 2001), who applied a total energy of 64.8 J m⁻².

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311 **3.3 Relativistic electron precipitation**

312 The ionospheric (upper panel) and atmospheric (lower panel) response for 313 long-lasting REP is shown in Figure 6, following the same format as Figure 5. The 314 REP is estimated using the precipitating fluxes measured in the bounce loss cone at 315 L=3.5-4 by the UARS on 18 May 1992 (Gaines et al., 1995). We use this precipitation 316 measurement to provide an indicative example of long-lasting REP, while noting that 317 the significant unknowns at this stage as to the spectrum and fluxes for typical events. 318 The spectrum of this REP event is very hard, containing significant fluxes at energies 319 as high as 5 MeV (Gaines et al., Fig. 3, 1995). Ionisation rates calculated for this REP 320 event using the approach outlined in section 2 lead to increased atmospheric 321 ionisation rates at altitudes as low as ~30-40 km, peaking at ~60 km. The atmospheric 322 impact is then determined by combining these ionisation rates with the SIC model 323 assuming a 3-hour continuous precipitation process. In the upper panel an increase in 324 electron number density can be seen at 50–100 km starting at 12 LT and ending as the 325 REP forcing is turned off at 15 LT. In the lower panel, the NO increases steadily from 326 12-15 LT, peaking at altitude of 60-80 km with a 3 order of magnitude increase over 327 background levels. Unlike the electron number density changes the NO is long-lived 328 following the end of the REP forcing.

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330 **3.4 REP microbursts**

331 In order to estimate the atmospheric impact of REP microbursts we assume a 332 0.1 s burst of 2 MeV monoenergetic relativistic electrons with a REP flux of 100 el.cm⁻²s⁻¹sr⁻¹ taken from the results of Rodger et al. (2007a), who found that such a 333 334 burst produced reasonable agreement with short bursts of relativistic electron precipitation, detected by a subionospheric propagation sensor in Sodankylä, Finland. 335 336 The results from SIC are shown in Figure 7. The REP microburst was applied to the SIC model at 0.5 s and the effect can been clearly seen in the electron number density 337 338 plot in the upper panel. At altitudes of 50 km the enhanced electron number density 339 recovers in ~ 1 s, while at altitudes of ~ 70 km the recovery takes ~ 30 s. No substantial 340 enhancement of NO can be seen in the lower panel. In fact, the NO enhancements are 341 too close to the numerical noise level of the modelling for us to have confidence in the 342 values. Thus multiple forcing events have not been applied to the model study as each 343 and every event produces results too noisy to be trusted. However, the observed 344 repetition rate for microbursts from ground-based measurements is too low for the 345 accumulation to become significant. A similar insignificant NO_x enhancement was found for the considerably softer lightning-generated whistler induced electron 346 precipitation, were >4000 precipitation bursts occurred over ~8 hours (Rodger et al., 347 348 2007c).

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4. Descent of NOx to the stratosphere via the polar vortex

Our case studies using the SIC model have shown that SPEs, auroral altitude electron precipitation, and long-lived REP events produce mesospheric and thermospheric NO_x that is long-lived in polar regions during the winter. In this section we discuss how the NO_x can be transported vertically downwards into the stratosphere and how, once it has arrived, it can influence ozone chemistry and the radiative balance of the lower atmosphere. The balance between the occurrence of descending air during strong polar vortex conditions and the occurrence of significant levels of NO_x generated by energetic precipitation events is fundamental to the delivery of high altitude NO_x into the polar stratosphere. The following section uses the SIC model predictions of NO_x production by SPEs, auroral precipitation, and REP to identify mechanisms operating during the Arctic winter 2003-2004.

The air inside the polar vortex is effectively isolated from lower-latitude air. In the southern hemisphere the polar vortex is generally stable during the entire winter due to the relatively flat topography and the distribution of sea and land around the pole of the Southern hemisphere. The Northern polar vortex is less stable because of wave patterns disturbed by mountain ranges such as the Himalayas with a large interannual variability in its stability (Brasseur and Solomon, 2005, Chapter 6).

368 In the winter polar middle atmosphere transport is largely determined by the 369 polar vortex. In the winter pole, near the polar night terminator, strong temperature 370 gradients lead to formation of the Polar Night Jet (PNJ). As shown in Figure 8a, the 371 PNJ is a strong eastward (westerly) wind in the upper stratosphere-lower mesosphere near 60° N/S latitude, formed due to the thermal wind balance (Solomon, 1999; 372 373 Holton, 2004). The winds in the PNJ, which reach their peak of about 80 m/s near 374 60 km altitude, act as a transport barrier between polar and mid-latitude air, blocking meridional transport and isolating the air in the polar stratosphere and thus forming 375 376 outer edge of the so-called polar vortex. The edge of the winter polar vortex is usually 377 near 60° N/S and it extends from approximately 16 km to the mesosphere.

The isolation is greater, and the polar vortex more stable, in the Antarctic where there is less planetary wave activity affecting the vortex than in the Arctic. In the Arctic, the atmospheric wave activity disturbs the vortex, leading to greater mixing and faster downward motion, compared with those in the Antarctic vortex
(Solomon, 1999). The approximate location of the PNJ is presented in Figure 8a and
the approximate location of the edge of the polar vortex in Figure 8b.

384 The large-scale meridional circulation in the stratosphere is determined by the Brewer-Dobson circulation. The Brewer-Dobson circulation is formed by rising 385 386 motion from the troposphere to the stratosphere in the tropics, poleward transport at 387 stratospheric altitudes and sinking motion at mid- and high latitudes (Solomon, 1999). 388 In the mesosphere, the meridional circulation is formed by a single cell in which 389 rising motion takes place in the summer pole starting from the stratosphere, pole-to-390 pole transport in mesosphere-lower-thermosphere, and downward motion in the 391 winter pole mesosphere, down to the stratosphere. Horizontal transport in the 392 stratosphere and mesosphere is determined by winds in the zonal (longitudinal, u) and 393 meridional (latitudinal, v) directions as presented in Figure 8a. In the polar winter 394 stratosphere the mean zonal winds are in general directed eastwards (westerlies) along 395 with the PNJ. At higher altitudes the zonal winds remain westerly up to about 90 km 396 altitude above which the wind direction is reversed (Brasseur and Solomon, 2005).

397 Inside the polar vortex the vertical descent rate varies from year to year, and 398 also with respect to the distance from the vortex edge, as well as with altitude 399 (Manney et al., 1994; Rosenfield and Schoeberl, 2001). Callaghan and Salby (2002) 400 have shown from model simulations that, in general, the maximum descent rates 401 (2 mm/s, ~5 km/month) in the wintertime Northern Hemisphere (NH) middle and upper stratosphere are found near 60° latitude. At lower altitudes the descent is 402 403 slower, with vertical descent rates of 0.4-0.7 mm/s in the Antarctic middle stratosphere (Kawamoto and Shiotani, 2000). In the mesosphere the downwelling 404 405 rates increase to several mm per second (Callaghan and Salby, 2002) this being due to

due to the cooling rates increasing with altitude (Rosenfield et al., 1994). In the NH,
where the polar vortex is more disturbed than in the SH, there is more year to year
variation in the descent, as changes in the wave activity and frequently occurring
stratospheric warmings affect the vortex conditions (Rosenfield and Schoeberl, 2001;
Brasseur and Solomon, 2005).

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412 **4.2 Arctic winter 2003-2004**

413 Figure 9 presents observed effects of energetic particle precipitation on the 414 NO_x levels and the descent of the NO_x to lower altitudes during the Arctic winter 415 2003-2004 taken from Seppälä et al. (2007b). The top panel of the figure shows the 416 GOES-measured proton flux of protons with energy >10 MeV together with the 417 geomagnetic activity index K_p . The middle panel shows a radio wave ionisation index 418 which indicates enhanced ionisation levels inside the 70-90 km altitude range 419 (Clilverd et al., 2007b), measured through the analysis of radio wave propagation 420 conditions between Iceland and Svalbard. The lower panel presents NO₂ mixing ratios 421 from two satellite instruments complementing each other. The first of the instruments 422 is GOMOS (Global Ozone Monitoring by Occultation of Stars) flying on board the 423 Envisat satellite (Kyrölä et al., 2004). The instrument measures, among others, 424 nighttime ozone and NO₂ vertical profiles in the middle atmosphere (with NO₂ 425 vertical resolution of 4 km in the upper stratosphere and mesosphere). The second 426 instrument is the POAM III (Polar Ozone and Aerosol Measurement) carried on the 427 SPOT-4 satellite, measuring tropospheric and stratospheric daytime ozone, NO₂ (with 428 vertical resolution of < 2 km below 40 km) as well as other minor gases (Lucke et al., 429 1999). In the lower panel of Figure 9 GOMOS nighttime NO2 mixing ratios are 430 shown to illustrate the amount of NO_x in the altitude range 30-70 km in the 65-85°

latitude range (nighttime NO₂ is a good representation of the total NO_x = NO + NO₂ 431 432 in stratosphere-lower mesosphere region, see section 2). To follow the descent of NO_x 433 beyond the polar night (beyond Feb 2004) the daytime NO_2 measurements are shown 434 below the 45 km altitude. Note that the difference in magnitudes between the GOMOS and POAM III observations is due to the diurnal variation of NO₂. The 435 436 radio wave ionisation index shown in the 70-90 km region in Figure 9 indicates that 437 during the winter there were two principal periods showing significant ionisation 438 increases at ~80 km, either created in-situ by particle precipitation or by the ionisation 439 of NO_x by Lyman- α as the NO_x descends from higher altitudes (Seppälä et al., 2007b; 440 Clilverd et al., 2007b).

441 The first period of NO_x production (labelled 1 in Figure 9) occurred during the 442 Halloween storm at the end of October 2003, and can be strongly associated with 443 solar proton precipitation during the storm. Our modelling results presented in this 444 study shows consistency with the GOMOS observations shown in that significant 445 concentrations of NO_x are generated at altitudes of 50-60 km. NO_x descent driven by 446 the polar vortex continues after the end of the SPEs, and transports the NO_x to lower 447 altitudes. The descent of the NO_x enhancement appears to take about 1 month to reach 448 40 km altitude, and the final NO_x levels are about 3 times larger than normal 449 (Jackman et al., 2005).

Following the initial NO_x enhancement a second period of enhanced NO_x is observed from mid-November to mid-December (labelled 2). This appears to descend as well, at approximately the same rate as the first enhancement, but only reaches lower altitudes of 45-50 km before disappearing at all altitudes due to the reconfiguration of the polar vortex at this time (Randall et al., 2005). During this period there were two small solar proton events, and several large geomagnetic

456 storms, which could have generated some NO_x at altitudes >60 km, but the timing and 457 altitude range are consistent with simulation (Semeniuk et al., 2005) where enhanced 458 thermospheric ionisation by low energy electrons was included from the Halloween 459 storm and moderately disturbed periods consistent with HSSWS shortly afterward. 460 Our model simulations of auroral electron precipitation production of NOX at high 461 altitudes (110-120 km in Figure 5) are consistent with this picture.

462 A third period of NO_x enhancement starts on 12/13 January 2004 (labelled 3), 463 and is not associated with any particular storm, but rather with the strengthening of 464 the polar vortex in the upper stratosphere as indicated by cold zonal mean 465 temperatures at 2mB (http://www.cpc.ncep.noaa.gov/products/stratosphere) and 466 consequent strong downward vertical transport from thermospheric altitudes. Between 467 mid-December and early January no significant traces of NO_x are observed at any 468 altitude. The NO_x observed at the start of period 3 shows no sign of been generated at 469 50-60 km as with SPEs, nor does it appear to be created at 60-70 km as with REP. 470 The lack of any geomagnetic storms at the time is consistent with this view. From our 471 model predictions we determine that the most likely source of the NO_x is auroral 472 altitudes (>70 km), although the high concentrations of NO_x suggest a significant 473 period of generation rather than bursts of aurora.

474 The enhancement is first observed at high altitudes and can be followed in each instruments dataset in turn until the enhancement reaches the upper stratosphere 475 476 in April 2004. No single geomagnetic storm or solar proton event can be identified as 477 the source of this NO_x , and the production altitude is reported to be >70 km (Clilverd 478 et al., 2007b), i.e., consistent with an auroral altitude source as modelled in Figure 5. 479 The onset date is consistent with the start of strong downward vertical transport in the 480 polar vortex as the upper stratospheric vortex re-strengthens (see

481 http://www.cpc.ncep.noaa.gov/products/stratosphere) following stratospheric а 482 warming period at the end of December (Clilverd et al., 2006b). The amount of NO_x 483 in this third event is 4 times the background levels once it reaches 40 km, and is 484 significantly longer-lived than the previous two enhancements (4 months compared with 1 month). During the descent period of the third enhancement a secondary 485 486 enhancement of NO_x can be seen in mid-February 2004 at all altitudes above about 487 55 km (labelled 4). The timing of this NO_x increase is coincident with a large 488 geomagnetic storm that occurred on 11 February 2004, which had no associated solar 489 proton event. The NO_x enhancements are significant, and add to the already 490 descending NO_x at 50-55 km, but disappear at higher altitudes after about one week. 491 From our model prediction we find that this secondary enhancement is clearly the 492 result of REP generating NO_x at altitudes of about 55-70 km, i.e., electron energies of 493 200-1000 keV, as shown in our modelling study in Figure 6.

From the analysis of the observations shown in Figure 9 we can determine that all three NO_x generation mechanisms took place during the Arctic winter 2003-2004. Due to the long lifetime of NO_x in the dark polar winter the NO_x accumulated at altitudes of 50-90 km in the early winter because of SPEs and auroral electron precipitation, and also accumulated in the late winter because of auroral electron precipitation and REP.

500

501 **4.3 The ozone balance of the stratosphere**

502 Descending NO_x enhancements from energetic particle precipitation changes 503 the ozone balance of the stratosphere. Since ozone is an important source of heating in 504 the stratosphere, reductions in ozone would be expected to lead to cooling of the 505 stratosphere. By including an experimentally based energetic electron precipitation

506 NO_v production (EEP-NO_v) into a Chemistry-Climate model, Rozanov et al. (2005) 507 were able to identify a 2 K cooling in the polar middle atmosphere at high latitudes 508 (0.5 K cooling at tropical latitudes) compared with a simulation which did not include 509 the EEP-NO_v source. Their model results also showed detectable changes (from -1 K 510 to +2.4 K) in the surface air temperatures in the northern polar region. These were a 511 result of about a 30% decrease in ozone at high polar latitudes within the polar vortex, 512 the ozone decrease outside the vortex being around 3-5%. The model study of 513 Rozanov et al. (2005) was based on a one year simulation, 15 years of observations 514 reported by Sinnhuber et al. (2006) also suggest that there is a relation between 515 energetic electron precipitation (GOES observed electron fluxes) and middle 516 stratospheric ozone observed from ozone soundings in the Arctic. Langematz et al. 517 (2005) have included a REP-NO_x source in a 11-year solar cycle simulation. Their 518 results, like Rozanov et al. (2005), suggest that the NO_x source by particles and its 519 transport from the mesosphere to the stratosphere in the polar vortex are important for 520 stratospheric O_3 . We have shown here that the source of NO_x by particles are a 521 combination of three mechanisms, suggesting more complex variability than in the 522 reported modelling studies above, but all are associated with solar and geomagnetic 523 activity cycles. Detailed estimates, of the influence on stratospheric temperatures, due 524 to each of these three mechanisms will be undertaken in future work.

525

526 **5. Summary**

527 In this study we have discussed the significance of solar proton events, 528 relatively low-energy (auroral altitude) electron precipitation, and high-energy 529 relativistic electron precipitation events of different durations, on the neutral 530 atmosphere in the polar regions. The auroral altitude electron precipitation is 531 preferentially generated by HSSWS in comparison with CME events. In contrast, both 532 CME and HSSWS-triggered geomagnetic storms are likely to produce relativistic 533 electron precipitation events. We have predicted the altitude and effectiveness of NO_x generation at 70°N, 0°E in northern hemisphere winter conditions using the 534 535 Sodankylä Ion and Neutral Chemistry model (SIC), imposing the most realistic 536 particle flux and energy spectra available in the literature at the time of writing. Large SPEs are found to produce higher initial NO_x concentrations than long-lived REP 537 538 events, which themselves produce higher initial NO_x levels than auroral electron precipitation. Only REP microburst events were found to be insignificant in terms of 539 540 generating NO_x.

541 We also analysed the observed effects of energetic particle precipitation on the 542 NO_x levels and the descent of the NO_x to lower altitudes during the Arctic winter 543 2003-2004 as shown by Seppälä et al. (2007b), and discuss them in terms of the case 544 studies modelled using SIC in this paper. We show that the GOMOS observations 545 from the Arctic winter 2003-2004 are consistent with NO_x generation by a 546 combination of SPE, auroral altitude precipitation, and long-lived REP events. Each 547 NO_x generation mechanism occurred independently during the winter, but the longlived NO_x eventually accumulated, and in the presence of a strong polar vortex lead to 548 549 levels of NO_x in the stratosphere that could produce stratospheric wind and 550 temperature changes that are consistent with previously published results.

551

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- 845 **Figure Captions:**
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847 Figure 1. Particle precipitation effects on the ion-chemistry of the atmosphere.

Figure 2. Schematic structure of the Sodankylä Ion and Neutral Chemistry model.
The circles, tilted squares, and squares indicate external input, external models used,
and modules of the SIC model, respectively (taken from Verronen, Fig 4.1, 2006).

Figure 3. Altitude versus ionisation rates for monoenergetic beams of protons 11000 MeV (left) and electrons 4-4000 keV (right).

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Figure 4. The effect of the Halloween solar proton events on electron number density and NO_x as modelled by SIC at 70° N latitude, 0° longitude. The simulated electron number density (log(m⁻³)), from 40-120 km altitude, with time is shown in the upper panel, and the concentration of NO_x (log(m⁻³)) over the same altitude range is shown in the lower panel. Maximum NO_x production occurs at ~50 km altitudes.

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Figure 5. The effect of auroral electron bursts on electron number density and NO concentration. The bursts of electrons have a characteristic energy of 5 keV and a total energy flux of 10 mW m⁻². Upper panel: electron number density during the burst run. Lower panel: the behaviour of NO concentration relative to the background level, i.e., the result of the burst run divided by the result of the control run. The electron bursts last 5 min each, starting at 23:05, 23:25, and 23:45 LT.

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Figure 6. Same as Figure 5, but showing the effect of a 3 hour long burst of REP on electron number density and NO_x levels using the precipitating fluxes measured in the bounce loss cone at *L*=3.5-4 by the UARS on 18 May 1992 (Gaines et al., 1995). The spectrum of the REP event is very hard, containing significant fluxes at energies as high as 5 MeV.

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Figure 7. Same as Figure 5, but showing the effect of a REP microburst on electron number density and NO_x levels. We have assumed a 0.1 s burst of 2 MeV monoenergetic relativistic electrons with a flux taken from those reported by SAMPEX.

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880 Figure 8a: Approximate location of the Northern Hemisphere Polar Night Jet, inside 881 which the polar vortex is formed, and the wind vector directions (zonal *u*, meridional 882 v and vertical w). - 8b: Different phenomena in the winter pole middle atmosphere. 883 On the left side are presented phenomena related to ionisation by precipitating 884 protons, subionospheric radio wave propagation and catalytic reaction cycles of the 885 HO_x and NO_x gases. Also shown are the altitudes regions where the cycles are 886 effective (as indicated by the curly brackets). On the right are shown phenomena 887 related to dynamics. The approximate location of the polar night jet is presented as red 888 circle and the darker colour indicates the area where the peak winds are observed. The 889 red curve depicts the approximate location of the polar vortex. Location of latitudes 890 45° and 60° are indicated with the respective numbers (taken from Seppälä, 2007).

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Figure 9. Combined observations of NO₂ during the Northern Hemisphere winter 2003 – 2004, showing (top) the >10 MeV GOES-11 proton flux cm⁻²s⁻¹sr⁻¹ (heavy line) and K_p index (light line), (middle) high-altitude ionisation levels determined from the subionospheric radio wave index, and (bottom) GOMOS nighttime and POAM III daytime (SS) NO₂ mixing ratios, with the POAM data shown inside heavy boxes. Both data sets have been zonally averaged over 2 days. Note the differing colour scales for the two satellite data sets. These observations show the generation and descent of NO_x into the upper stratosphere. From Seppälä et al. (2007b).





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903 Figure 2. A graphical representation of the SIC model structure. Schematic structure 904 of the Sodankylä Ion and Neutral Chemistry model. The circles, tilted squares, and 905 squares indicate external input, external models used, and modules of the SIC model,

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- **8b**: Different phenomena in the winter pole middle atmosphere. On the left side are presented phenomena related to ionisation by precipitating protons, subionospheric radio wave propagation and catalytic reaction cycles of the HO_x and NO_x gases. Also shown are the altitudes regions where the cycles are effective (as indicated by the curly brackets). On the right are shown phenomena related to dynamics. The approximate location of the polar night jet is presented as red circle and the darker colour indicates the area where the peak winds are observed. The red curve depicts the approximate location of the polar vortex. Location of latitudes 45° and 60° are indicated with the respective numbers (taken from Seppälä, 2007).



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