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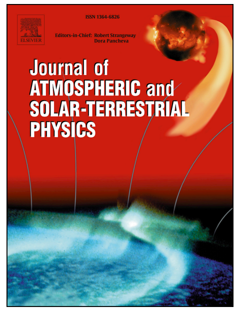

Solar proton events and stratospheric ozone depletion over northern Finland

M.H. Denton, R. Kivi, T. Ulich, C.J. Rodger, M.A. Clilverd, R.B. Horne, A.J. Kavanagh

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Solar proton events and stratospheric ozone depletion over northern Finland

M. H. Denton[1,2], R. Kivi[3], T. Ulich[4], C. J. Rodger[5], M. A. Clilverd[6], R. B. Horne[6], A. J. Kavanagh[6].

1. Space Science Institute, Boulder, CO 80301, USA.
2. New Mexico Consortium, Los Alamos, NM 87544, USA.
3. Arctic Research Centre, Finnish Meteorological Institute, Sodankylä, Finland.
4. Sodankylä, Geophysical Observatory, Sodankylä, Finland.
5. Department of Physics, University of Otago, New Zealand.
6. British Antarctic Survey (NERC), Cambridge, UK.

ABSTRACT:

We examine the variation of stratospheric ozone over northern Finland using ozonesonde observations from 1845 stratospheric balloon flights launched between 1989 and 2015 from near Sodankylä. The annual variation of the ozone partial pressure is examined and seasonal variations are explored and quantified. Direct links between the measured ozone partial pressure and common solar-wind parameters are also examined. A superposed-epoch analysis of the observations based on 191 solar proton events (SPEs) reveals a clear drop in the ozone partial pressure that commences following SPE-arrival at Earth. This analysis shows a reduction in stratospheric ozone in the winter/early-spring months (when the polar vortex is active over northern Finland), in contrast to summer/early-autumn months where no decrease is detected. By subtracting the natural seasonal variations in ozone partial pressure the SPE-driven reduction in ozone between 16 km and 24 km altitude is quantified. Analysis indicates that the ozone partial pressure during winter/early-spring is reduced, with a minimum reached ~8 days following the SPE arrival. On average, the ozone partial pressure is reduced by ~10% between 16-24 km altitude and takes ~40 days to return to its previous level. To the best of our knowledge, this is the first comprehensive statistical study, on a regional basis, that provides direct, and long-term in-situ evidence for ozone depletion by SPEs in the northern hemisphere.

CORRESPONDING AUTHOR: M. H. Denton (mdenton@spacescience.org)

1. Introduction

Solar proton events (SPEs) arise in association with energetic events on the Sun and energisation processes in interplanetary space (e.g. *Reames* [1999]; *Kurt et al.* [2004], *Tylka et al.* [2006], *Oh et al.*, [2010], and references therein). Upon arrival at Earth the solar protons may enter the upper atmosphere and collide with neutral particles at altitudes that are dependent on the incident energy of the protons [*Seppälä et al.*, 2008]. At this point they induce chemical changes to the local neutral population. Such energetic particle precipitation (EPP) is regularly implicated in the production of species such as odd nitrogen (NO_x) (e.g. *Crutzen et al.* [1975]; *Shumilov et al.* [2003]; *Clilverd et al.* [2005]), which are themselves implicated in the subsequent depletion of stratospheric ozone. Odd-nitrogen species are long-lived during darkness and can descend to stratospheric altitudes via the high-latitude polar vortex, during the polar winter. Once at stratospheric altitudes NO_x may cause chemical destruction of the in-situ ozone (e.g. *Jackman et al.* [1995]; *Jackman et al.* [2009]). Other chemical pathways for ozone destruction are also available (e.g. *Jackman et al.* [2009]; *Damiani et al.* [2008, 2009, 2012]). Observational case studies of large individual SPEs reveal that such ozone depletions (in the mesosphere and stratosphere) do indeed occur (e.g. *Weeks et al.* [1972]; *Heath et al.* [1977]; *Thomas et al.* [1983]; *Lopéz-Puertas et al.* [2005]; *Seppälä et al.* [2004; 2006; 2008]). Modelling studies of such events have also helped ascertain the long and short term implications for atmospheric ozone balance (e.g. *Jackman and McPeters* [1985]; *Jackman et al.* [1996]; *Rodger et al.* [2008]; *Jackman et al.* [2009]). More general recent studies of the effects of EPP have outlined some of the chemical changes that occur during SPEs (e.g. *Verronen and Lehman* [2013]) and also recently drawn links between geomagnetic activity, particle precipitation, and changes in polar surface air temperature [*Seppälä et al.*, 2009]. However, a complete accounting of the chemical changes in the atmosphere resulting from EPP, and their relative importance, remains a major unsolved issue in magnetospheric and atmospheric physics *[Denton et al.*, 2016].

In the current study we aim to expand upon current knowledge by using a large database of ozone measurements constructed from 1845 *ozonesonde* balloon flights, launched from Sodankylä in northern Finland, between 1989 and 2015 [*Kivi et al.*, 2007]. Initially, these data are analysed to examine the seasonal/annual changes that occur in the ozone partial pressure in this region. Following this, links between the stratospheric ozone partial pressure and various common solar-wind parameters are explored. Subsequently, we carry out a statistical analysis of SPE-induced changes in stratospheric ozone during 191 SPE events that took place over the same time period as the ozonesonde observations. Finally, we quantify the observed reductions measured in the ozone partial pressure following SPEs and discuss the implications of our findings, in comparison with other reported observations.

2. Ozonesonde Data Set

The ozone profiles used in this study were obtained by the electrochemical concentration cell (ECC) type of ozonesonde [*Deshler et al.*, 2008; 2017, *Kivi et al.*, 2007, *Smit and ASOPOS Panel*, 2014]. These instruments are capable of providing profile measurements of ozone concentration from the surface to the lower stratosphere (~30-34 km) with precision better than 2-3 % [*Deshler et al.*, 2008]. Vertical resolution of the soundings is about 10 meters, as data is typically analyzed in 2 second intervals, while the effective vertical resolution is of the order of 100-150 meters, given that the sensor response time is 20-30 seconds. The sondes used in this study have been launched on a regular basis from Sodankylä, Finland (67.4 N, 26.6 E) since 1989 during all seasons. Sondes are normally launched once per week around local noon. These data are supplemented by the frequent ozonesonde campaigns that have taken place in winter/spring season, significantly increasing the number of launches and the available dataset. This measurement program has resulted in 1845 soundings over the 27-year time period studied here. Balloon ozonesonde data have the advantage over satellite data that they provide high spatial resolution altitude profiles of the ozone distribution throughout the lower stratosphere. However, while these data are certainly suitable for study of changes in the ozone distribution over northern Finland, it should be noted that results will only apply on a local/regional basis. The details of the ozonesonde sounding system and principles of the data set homogenization are explained in detail in *Kivi et al*. [2007].

3. Analysis and Results

3.1 Seasonal and Annual Variations of Stratospheric Ozone

As previously shown by *Kivi et al.* [2007], there is a substantial annual variation in the stratospheric ozone partial pressure over Sodankylä during the year with a peak ozone level occurring around April and a minimum occurring around September (see *Kivi et al.* [2007], Figure 4). The annual cycle of ozone in the Arctic lower stratosphere is caused by the global annual cycle in ozone transport from lower latitudes towards the poles (e.g. *Butchart*, [2014], and references therein). This transport is strongest in winter and early spring and the ozone variability is highest during this period. The stratospheric ozone concentrations in late spring and summer decrease continuously at high latitudes since the combination of ozone production and transport are too slow to offset the destruction of ozone via reactions involving NO_x. Significant destruction of ozone also occurs during via catalytic reactions involving chlorine monoxide (ClO) and bromine monoxide (BrO), but only during periods of sunlight (i.e. not during the polar winter) (e.g. *Jackman et al.* [2009]; *Damiani et al.* [2008, 2009, 2012]). To take account of the annual and seasonal variability of ozone in our analysis, we initially extend the analysis of *Kivi et al.* [2007], who calculated the average monthly variation of stratospheric ozone between 1989 and 2003. Here, this calculation is extended, using a further twelve years of data to cover the years 1989 to 2015 inclusive (cf. Section 3.3).

The average ozone partial pressure (in mPa) is calculated as a function of geopotential altitude (in km) and month of the year, for the entire ozonesonde dataset between 1989 and 2015. The results of this analysis are shown in Figure 1. (Note: As is common with sonde data we use geopotential altitude (height) rather than pressure units on the y-axis - the geopotential altitude approximates the altitude above sea-level of a particular pressure level). Clearly there is significant variability in the measured ozone on an inter-annual basis, which is largely due to inter-annual variability in chemical and dynamical factors influencing ozone. This inter-annual variability is strongly pronounced in the northern hemisphere, where dynamical variability is relatively large from year to year in contrast to the Antarctic stratosphere. However, significant vortex ozone depletions have also been observed in the northern hemisphere [*Manney et al.*, 2011].

To explore the annual variations in the ozone data we examine the entire dataset of balloon-borne ozonesonde measurements made above northern Finland between 1989 and 2015, plotted in Figure 2 (top panel). It is clear that the regular seasonal variations (cf. Figure 1) are a prominent feature in this dataset, but there are also large changes on a year-to-year basis. For example, the measurements around 2001, close to solar maximum, are noticeably higher than most other periods.

To quantify these changes we also plot these data on a seasonally-adjusted *difference-from-mean* basis (Figure 2, bottom panel). The mean ozone partial pressure for the appropriate month and altitude (i.e. from the data shown in Figure 1) is subtracted from each individual measured ozone point in the data set. Doing this should remove seasonal trends from the data and allow variations due to other causes to be detected. The *seasonally-adjusted* difference-from-mean parameter plotted in Figure 2 provides quantification of the strength of the variations on an annual basis. For example, for the data around 2001, the measured ozone is at times ~8 mPa higher than the mean value (at around 20 km altitude). In contrast, the measured ozone partial pressure is at times ~8 mPa lower than its mean value in 1993 (at around 15 km altitude). The years around solar maximum (when the solar EUV flux and F10.7 index are higher) show evidence of generally higher ozone levels compared to the years around solar minimum(when the solar EUV flux and F10.7 index are lower). However, this certainly isn't always true in every year with elevated F10.7 and there is much variation. The causes of annual variations are also known to include: (i) "internal" terrestrial effects such as the quasi-biennial oscillation (QBO) cycle and volcanic eruptions that perturb aerosol concentrations, etc., (ii) longer-term "external" effects such as the 11-year solar cycle, and (iii) longer-term internal trends in stratospheric ozone as a result of anthropogenic causes [*Kivi et al.*, 2007; *Manney et al.*, 2011]. A more comprehensive discussion of the annual cycle, inter-annual variability and also day-to-day variability of stratospheric ozone over Sodankylä may be found in *Kivi et al.* [2007].

3.2 Variations of Stratospheric Ozone with Solar and Geophysical Parameters

The literature contains various modelling and case-study results that show links between the state of the incoming solar wind, the subsequent state of the magnetosphere, and the terrestrial ozone population, in particular ozone in the mesosphere and ozone in the stratosphere. Considering the stratosphere, two physical mechanisms have been suggested that are thought to cause changes in the ozone population based on solar-wind driving; (i) instantaneous destruction of O_3 due to EPP by extremely energetic incident protons (with energies greater than ~100 MeV), whereby these penetrate the atmosphere directly to near-stratospheric altitudes and cause immediate ozone dissociation, and (ii) a longer time-scale process whereby odd-nitrogen species, for example, are created at mesospheric altitudes (by lower-energy particle precipitation due to both electrons or protons) and these then descend to stratospheric altitudes during the polar winter. At stratospheric altitudes these species cause chemical destruction of ozone. The latter process is often termed in the indirect effect, and the former the direct effect. Initially, we aim to test the efficacy of the direct effect by exploring links between solar-wind/geophysical parameters with the stratospheric ozone partial pressure. To investigate near-instantaneous changes in the ozone population we aim to correlate between the measured ozone partial pressure and the level of various geophysical parameters measured 24 hours previously.

Figure 3 contains plots of the ozonesonde observations as a function of altitude, and four geophysical and solar-wind parameters, namely: (i) The Auroral Electrojet (AE) index, a commonly used parameter that is a good proxy for magnetospheric substorm activity [*Davis and Sugiura*, 1966; *Gjerloev et al.*, 2004], (ii) the solar-wind electric field parameter ($-v_{sw}B_z$) calculated as the negative product of the solar-wind velocity (v_{sw}) and the z-component of the solar-wind magnetic field (B_z), and measured in units of mV m^{-1}, (iii) the adjusted F10.7 index, a measure of the incident radio flux at 10.7 cm, and a good proxy for ionising solar EUV radiation [*Tapping*, 2013, and references therein], and (iv) the Disturbance Storm Time (Dst) index [*Sugiura*, 1964; *Sckopke*, 1966], a widely used measure of magnetospheric storm strength and a proxy for the strength of the Earth's ring current, measured in units of nT. All parameters used are taken from the hourly OMNI2 database [*King and Papitashvili*, 2005]. The aim of this analysis is to search for trends showing near-instantaneous changes in the ozone population as a result of changes driven by external solar wind parameters. If present, such changes could then be linked to direct destruction of stratospheric ozone by energetic particle precipitation.

It is clear from the plots shown in Figure 3 that there is little evidence for such changes on a systematic basis. None of the plots show clear trends in the ozone partial pressure as a result of increases or decreases in the parameters examined. Perhaps the most surprising result here is that there is no clear correlation with the F10.7 index since stratospheric ozone is known to be correlated with solar irradiance [*Kivi et al.*, 2008]. In Figure 1 the highest ozone levels (above the mean) come in years around solar maximum although this isn't by any means a hard and fast rule. For example, in 2006 near solar minimum, the average ozone levels were above average even though the F10.7 index during the year rarely exceeded 100. It is known that other physical variables also strongly affect the annual transport and destruction of stratospheric ozone during each year and that these can certainly suppress variations based on the UV variation alone (see for example *Kivi et al.* [2007] and *Manney et al.* 2011]).

Whilst the literature does contain example case-studies of some of the largest geomagnetic storms resulting in rapid decreases in stratospheric ozone, it is also known that many of these 'extreme' events contain multiple drivers of geomagnetic activity such that no single index currently accounts for all causes of driving. The results from Figure 3 provide evidence that using fluctuations in a single geophysical index alone are not sufficient to use as a predictor of subsequent changes in the near-instantaneous ozone partial pressure in the stratosphere. If such changes are occurring, a more sophisticated analysis to reveal them is required.

3.3 The Effects of Solar-Proton Events (SPEs) on Stratospheric Ozone

In order to explore changes in stratospheric ozone, beyond the systematic seasonal variations shown in Figure 1, and the annual variations shown in Figure 2, we examine external driving of the atmosphere by solar-proton events. Many studies of non-terrestrial drivers of changes in the ozone population focus on such events, since the particle spectrum is sufficiently hard that the most energetic particles can penetrate deep into the mesosphere, into the stratosphere, and even to ground level (e.g. *Solomon et al.* [1981], *Thomas et al.* [1983], *Jackman and McPeters.* [1985], *Clilverd et al.* [2005], *Verronen et al.* [2011] *Damiani et al.* [2016], *Hocke* [2017]). For the most energetic solar-terrestrial event yet known in the modern era, the 1859 Carrington Event, it has even proved possible to model the effects of extremely high fluxes of solar protons on the ozone balance within the atmosphere [*Rodger et al.*, 2008]. However, in contrast to event studies of very large events, or theoretical studies of long-term trends, it has proven difficult to make estimates of the long-term effects of more frequent, but less extreme, events particularly on a regional level. A recent study by *Damiani et al.* [2016] has addressed this issue, in part, for the southern hemisphere by showing a 10-15% decrease in stratospheric ozone over Antarctica using limb-sounding methods from the Aura satellite. That study found that descent of mesospheric NO_x down to stratospheric heights was the primary cause of such stratospheric ozone depletion in the southern hemisphere. The authors also highlighted the limited observational evidence available to quantify particle precipitation effects upon the ozone budget in general [*Damiani et al.*, 2016]. Here, we attempt to address this issue for the northern hemisphere.

As a basis for the analysis we utilize a list of 191 Solar Proton Events taken from the published list by the US National Oceanic and Atmospheric Administration (NOAA) Space Weather Prediction Center (SWPC) and freely available via file transfer protocol (ftp://ftp.swpc.noaa.gov/pub/indices/SPE.txt). At the time of the analysis, only one SPE event was listed beyond the end of 2015. The epoch times taken from the NOAA list are the start-times of each individual SPE, at a time resolution of one-hour.

Analysis is then carried out by performing a superposed epoch study of the measured ozone during these events. All available ozonesonde data are binned as a function of epoch time (time from SPE-onset) and geopotential altitude. The final grid for this binning is one day in time and 1 km in geopotential altitude, and spans a total of

90 days: 30 days prior to the SPE through to 60 days after the SPE. In addition, a set of 2500 randomly selected epochs between 1989 and 2015 are generated using the methodology of *Park and Miller* [1988]. The ozone data are then sorted based on these random epochs with this analysis providing a control data set against which the SPE analysis can be compared. Note that no account is made of short-term diurnal effects.

Figure 4 contains a selection of plots resulting from this analysis and shows the ozone partial pressure as a function of geopotential altitude and epoch time for (a) 2500 random epochs selected between 1989 and 2015 (left column), and (b) 191 solar proton events that occurred during the same interval (right column). The plots show the averaged (mean) ozone partial pressure up to 40 km altitude (top row). No adjustment is made for seasonal variations in the data at this point. Also shown are line plots of the integral of the ozone partial pressure for altitudes between 16 and 24 km (middle row) for these events. This altitude region contains the bulk of stratospheric ozone (cf. Figure 1). The red points represent the individual data (i.e. the mean ozone partial pressure) and the blue line is a 15-day running box-car average of these data. (Note: the reduction in the running mean commences slightly prior to zero-epoch due to the 15-day averaging, although the individual (red) data points indicate ozone remains high until SPE arrival). The grey shading indicates the standard deviation of the superposition, whilst the thin orange, black, and purple lines represent the upper quartile, the median, and the lower quartile of the superposition.

The plots in the top two rows of Figure 4, although suggestive of a decrease in total stratospheric ozone following SPE occurrence, take no account of the large seasonal variations in the ozone partial pressure known to exist over Sodankylä. In order to remove the seasonal variations in the data contributing to the superposed epoch analysis the mean ozone partial pressure for the month in question (taken from the values in Figure 1) is subtracted from each data point. The resulting quantity is thus seasonally independent. The bottom row in Figure 4 shows the summation of these 'difference-from-mean' values. Here, a negative value indicates a seasonally independent (i.e. a true) reduction in ozone partial pressure. As can be seen from Figure 4, there are only small fluctuations in the ozone partial pressure in all three plots for the random epochs. However, for the SPEs there is clear evidence for a decrease in the ozone partial pressure that commences close to the zero epoch and reaches a minimum value after around 8 days. This is evident especially in the line plots showing the summed ozone partial pressure Here, the decrease in the ozone partial pressure (summed between 16-24 km) is of the order of 5%. The seasonally adjusted ozone partial pressure immediately prior to the zero epoch is slightly above the mean value but then falls rapidly around zero epoch, reaching a minimum after around 8 days, and then remains below the seasonally adjusted mean value for around 40 days.

While it is unlikely that protons with energies lower than ~10-100 MeV will directly reach the atmosphere above Sodankylä due to geomagnetic field rigidity effects (see *Rodger et al.* [2008], Figure 8), these protons will produce NO_x at higher latitudes. This is then likely to be rapidly mixed throughout the polar vortex, and thus may contribute to indirect, time-delayed ozone depletion, particularly when the polar vortex extends to the Sodankylä region. This occurs around 50% of the time between December and April and never between July and October [*Kivi et al.*, 2007 - Table 3]. In order to test for such seasonal trends in the ozone observations we further sub-divide the data shown in Figure 4 based upon day of year.

Figure 5 shows the same series of plots as shown in Figure 4, but this time the data are only shown for the months July, August, September, and October (JASO) - periods when the polar vortex is absent over northern Finland [*Kivi et al.*, 2007]. In comparison with Figure 4, it is clear that changes in ozone following SPEs during these months are practically absent. There are fluctuations around the mean level throughout the epoch period, but there is little evidence for any systematic decrease in ozone partial pressure during these SPEs.

Figure 6 shows the same series of plots as shown in Figure 4 and Figure 5, but here the data are only shown for the months January, February, March, and April (JFMA); periods when the polar vortex is frequently present over northern Finland (~50% of the time) and can cause more rapid descent of long-lived NO_x during hours of darkness. In contrast with the plots in Figure 4 and Figure 5, it is clear that a large reduction in the stratospheric ozone partial pressure occurs immediately following SPE arrival at Earth. This reduction persists for many days. Again, the reduction in the running mean commences slightly prior to zero-epoch in the 15-day running average, although the individual (red) data points do not show evidence of a decrease until zero epoch. We also note that prior to zero-epoch the ozone for the SPE events is already greater than the mean value - this is due to the fact that the SPEs are more common during the years around solar maximum, when ozone levels tend to be slightly elevated. In general, years around solar maximum have higher ozone concentrations on average, although this trend clearly isn't true in all years. It is strongly depends on other factors that affect the production and the loss of ozone (e.g. catalytic loss of ozone in the springtime caused by ClO and BrO reactions) [cf. *Manney et al.*, 2011].

Clearly, the reduction in ozone commencing at zero epoch is significant, being of the order of 10% during JFMA when the polar vortex is active over northern Finland. A minimum decrease is reached ~8 days on average after the SPE and ozone remains at a level below the mean value for ~40 days, and continues to fluctuate beyond this time. This result supports the interpretation that destruction of stratospheric ozone is occurring, due to the external driver of solar-wind proton precipitation. While the reduction in measured stratospheric ozone following SPE-arrival is clearly evident in Figure 6, the descent of NO_x in the polar vortex at these lower altitudes is quite slow: around 8 km/month at ~50 km altitude [*Manney et al.*, 1994; *Rinsland et al.*, 2005]. Hence, a more-gradual response in the observed ozone decrease might be expected, rather than a decrease immediately following SPE arrival at Earth. A delay in the destruction of ozone, due to NO_x production via electron precipitation, is also possible (e.g. *Andersson et al.* [2014], *Randall et al.* [2015]).. Further studies are required to pin down the ultimate causes of the ozone depletion revealed here, and such studies are underway. Based on the literature, it is anticipated that these causes likely will include an admixture of direct and indirect physical mechanisms.

3.4 Stratospheric Ozone Changes During High-speed Solar-wind Streams (HSSs)

Stimulus for the work carried out in this manuscript originally arose from the knowledge that high-speed solar-wind streams (HSSs) are associated with enhanced activity in the Earth's magnetosphere and subsequently with large increases in energetic particle precipitation originating from the plasma sheet and/or radiation belts, noted by a number of authors (e.g. *Sandanger et al.*, [2007], *Longden et al.* [2008], *Rodger et al.* [2008], *Denton et al.* [2009], *Borovsky and Denton* [2009], *Morley et al.* [2012], *Kavanagh et al.* [2012], *Hendry et al.* [2013], *Clilverd et al.* [2013], *Blum et al.* [2015]). Although the energy spectrum during HSS-events is much softer than during SPEs there is still ongoing energetic particle precipitation, primarily due to relativistic (>1 MeV) electrons. Given the long-lived nature of HSSs such precipitation persists for much longer time intervals whether as continuous low-energy precipitation [*Whittaker et al*., 2014;] or in the form of electron microbursts [*Nakamura et al.*, 2000; *Lorentzen et al.*, 2001; *Blum et al.*, 2015]. Indeed, ozone decreases in the middle atmosphere have been clearly observed during medium energy precipitation [*Andersson et al.*, 2014]. Using the data set described above, a search for substantial changes in stratospheric ozone partial pressure in the days following HSS-arrival, including times when the polar vortex was active, was carried out. The results from that study proved inconclusive (figure not shown).

4. Discussion and Conclusions

The analysis and results shown above highlight the difficulty in extracting changes in stratospheric ozone due to a single variable. However, by removing the monthly mean variations from the measured ozone partial pressure, and by concentrating on intervals when the polar vortex is active, and hence can assist in the descent of NO_x species, it has been clearly demonstrated that solar protons are causing substantial stratospheric ozone depletion of the order of 10%, following SPEs.

Determination of ozone variability on the Earth's climate is of great topical interest although reviews of the behaviour of ozone throughout the atmosphere have tended to concentrate on internal terrestrial variables [*Staehelin et al.*, 2001], or long-term solar changes [*Haigh*, 2003] rather than directly on short-term solar-induced forcing via SPEs. We argue that external driving of the Earth's stratospheric ozone budget was somewhat under-appreciated originally with studies initially concentrating on effects in the upper stratosphere and mesosphere [*Rusch et al.*, 1981; *Solomon et al.*, 1981; 1983]. Changes in the lower stratosphere due to external driving are now receiving more attention in the literature. Where solar-proton effects on climate are considered within global climate models, the effects have been reported as not statistically significant with regard to the annually averaged temperature and the total ozone variation (e.g. *Jackman et al.* [2009]). Although incorporation of the effects of short-term solar-proton fluctuations in global coupled climate models is currently quite rare, the effects of solar protons have been included in other modelling studies previously (e.g. *Jackman and McPeters* [1985]; *Jackman et al.* [1996]; *Rodger et al.* [2008]). SPEs have also been shown to produce local changes to stratospheric ozone on a case-by-case basis (e.g. *Weeks et al.* [1972]; *Heath et al.* [1977]; *Thomas et al.* [1983]; *Lopéz-Puertas et al.* [2005]; *Seppälä et al.* [2006]; *Seppälä et al.* [2008]). However, since most energy is deposited above 40 km, the direct effects of SPEs in general have not been considered important for quantification of total global ozone losses (e.g. *Sinnhuber et al.* [2003]), despite the long-lived nature of NO_x species generated at higher altitudes allowing their descent to occur over a period of weeks or months (e.g. *Randall et al.* [2001]). The full effects of SPEs may not be appreciated by study of single events. In this study, superposed epoch analysis of 191 SPEs has enabled a longer-duration statistical investigation of the average changes in ozone partial pressures to be carried out. Our work emphasises the need to incorporate energetic particle precipitation into climate models and we acknowledge the recent efforts in this direction (e.g. *Matthes et al.* [2017]).

5. Summary

In summary, we have analyzed data from 1845 balloon ozonesondes launched from Sodankylä, northern Finland, between 1989 and 2015. The mean monthly variation of stratospheric ozone has been calculated. The data have been analysed with respect to a number of geophysical variables (the AE index, the F10.7 index, the solar wind electric field parameter, and the Dst index), although no direct correlations to between stratospheric ozone and these indices (with a 24 hour time lag) was found.

To the best of our knowledge, this is the first comprehensive statistical study using balloon ozonesonde data that has provided direct, and long-term, in-situ evidence for stratospheric ozone depletion by SPEs in the northern hemisphere.

The results from this study are:

1. The average stratospheric ozone measured over northern Finland is highly variable during the year. The mean of the ozone partial pressure is greatest in winter and early spring and minimised in summer and early autumn.

2. There is no clear evidence for direct correlations between the stratospheric ozone partial pressure and the AE index, the F10.7 index, the solar-wind electric field, or the Dst index measured on the previous day.

3. A superposed epoch analysis of 191 SPEs examined between 1989 and 2015 indicates a fall in the ozone partial pressure of ~5% with a minimum reached ~8 days (on average) after the SPE arrival. The ozone partial pressure remains reduced below its mean value for ~40 days.

4. When only time intervals are considered when the polar vortex is present over northern Finland (i.e. late winter) the fall in ozone partial pressure is ~10% with the minimum again reached after ~8 days (on average)

after SPE arrival. The ozone remains reduced below its mean value for ~40 days. In contrast, no decrease in ozone is found following SPEs that occur during the northern hemisphere summer months of July-October when the polar vortex is not present.

It is intended that this study will highlight the fact that the average stratospheric ozone reduction is significant (~10%) following SPEs during the northern polar winter. We also intend that this knowledge be utilised to separate anthropogenic causes of ozone loss from natural causes. As noted in Section 2, balloon ozonesonde data have the advantage over satellite data that they provide high spatial resolution altitude profiles of the ozone distribution throughout the lower stratosphere. We plan on future analysis of other measured ozone data sources to investigate if the above findings are truly global or local (i.e. polar latitudes only) in nature. A follow-up study, using the same methodology but utilizing global satellite data coupled with multi-station balloon ozonesonde data from inside and outside the polar vortex, would certainly address this issue. In an era when separating anthropogenic ozone depletion from natural ozone depletion is receiving widespread attention, the results from this current study, summarised above, are particularly timely in that they provide quantification of the expected average ozone reduction for a number of events rather than quantifying such losses during single-event case-studies. Such averaged results should prove simpler to include in global climate models.

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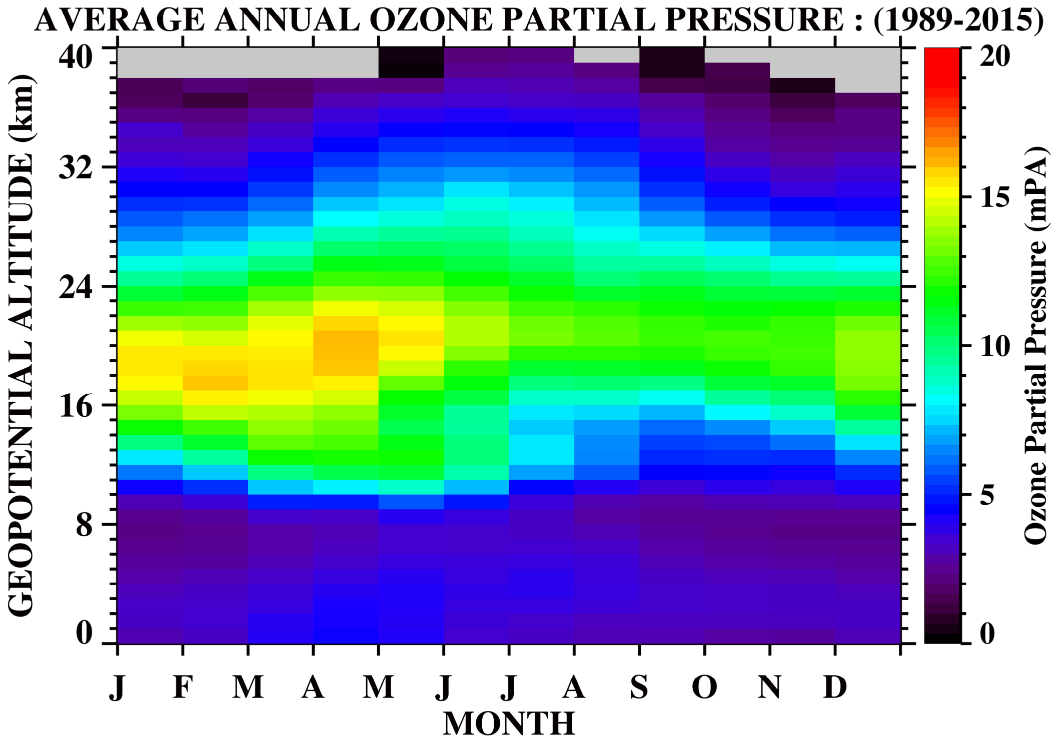


FIGURE 1: The averaged ozone partial pressure (in mPa) as a function of geopotential altitude and month-of-the-year for 1845 balloon ozonesondes launched between 1989 and 2015 from Sodankylä, northern Finland. Ozone is higher in winter/early-spring months and lower in summer/autumn months. (Note: This analysis updates the previous analysis of *Kivi et al.* [2007], Figure 4).

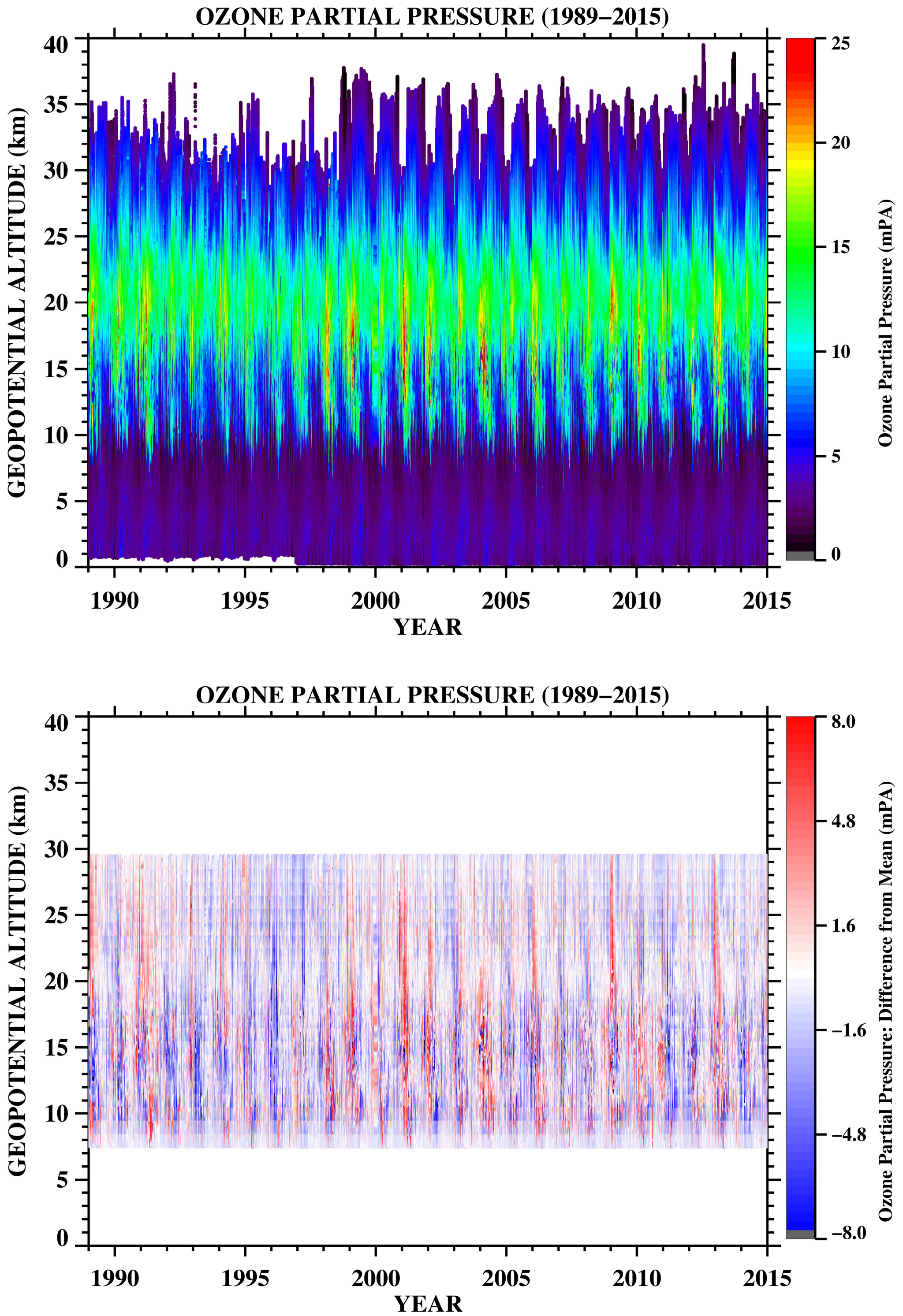


FIGURE 2: Showing a time-series of all ozonesonde data from Sodankylä between 1989 and 2015. The top plot shows the ozone partial pressure as a function of year and geopotential altitude. The bottom figure shows the same data (8-30 km altitude only), but this time adjusted for the seasonal mean variations using the data from Figure 1. Positive values (red) indicate measurements that are higher than the long-term mean and negative (blue) values show measurements that are lower than the long-term mean.

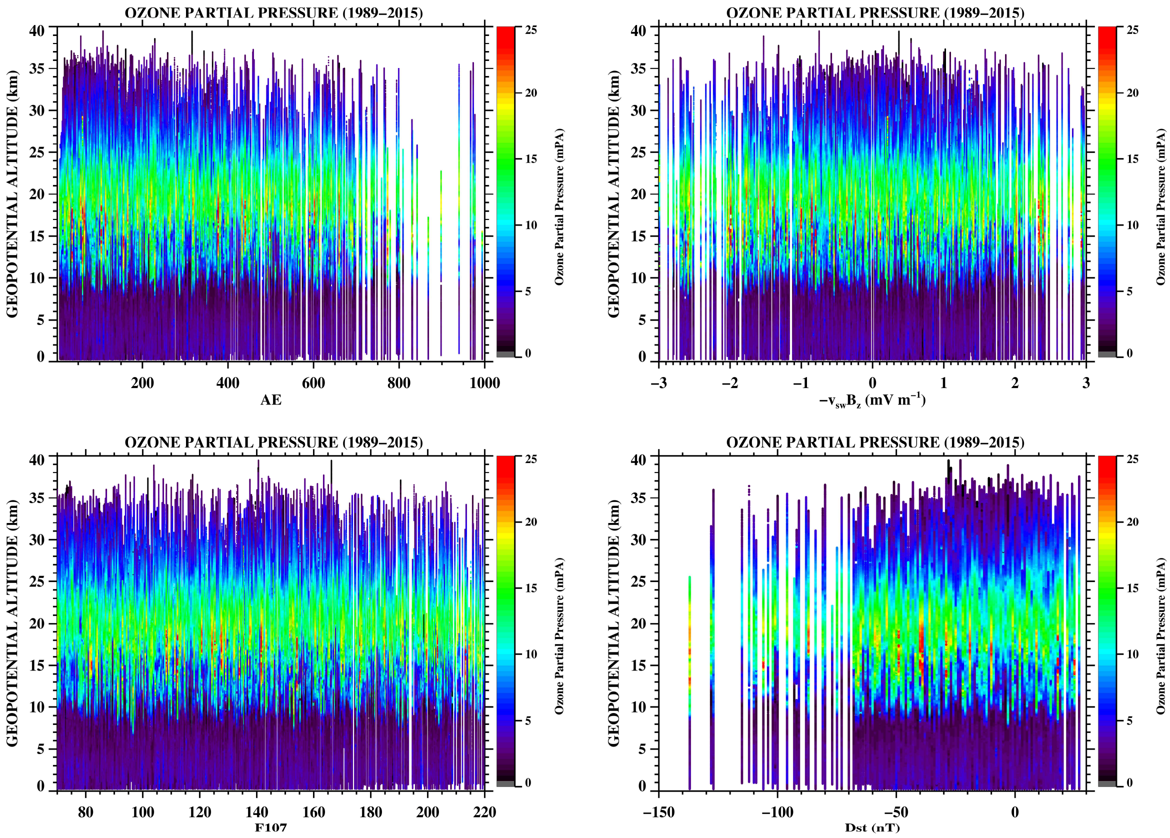
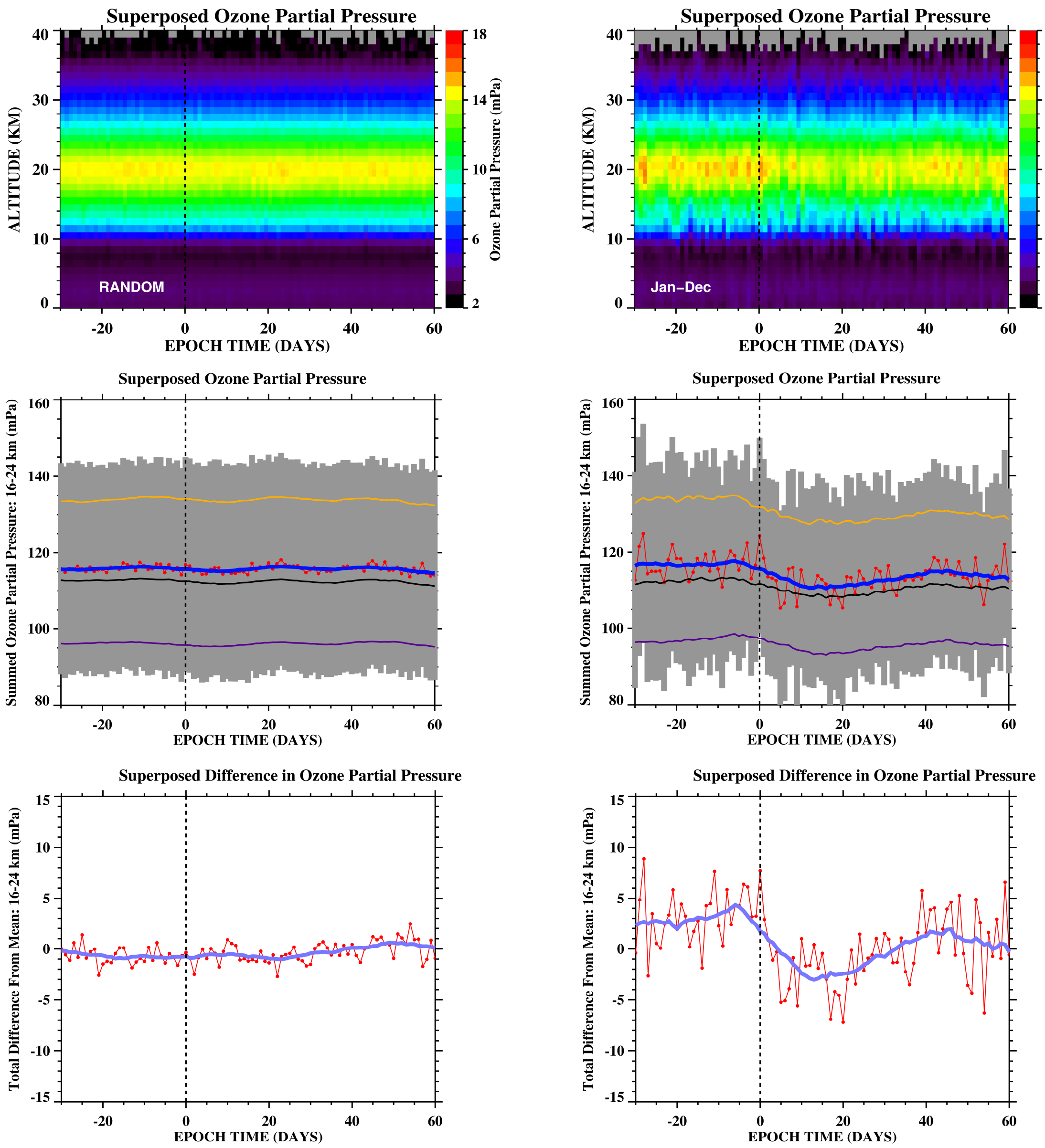


FIGURE 3: Plots showing the variation of the ozone partial pressure as a function of geopotential altitude in kilometres with (i) the AE index, (ii) the solar-wind electric field parameter $-v_{sw}B_z$, (iii) the F10.7 index, and (iv) the Dst index. There is little evidence for systematic changes in the ozone partial pressure with any of these four parameters.



<u>**FIGURE 4**</u>: Showing the ozone partial pressure as a function of geopotential altitude and epoch time for (a) 2500 random epochs selected between 1989 and 2015 (left column), and (b) 191 solar proton events that occurred during the same interval (right column). The plots show the averaged ozone partial pressure up to 40 km altitude (top row), the integral of the ozone partial pressure between 16 and 24 km altitude (middle row), and the integrated difference-from-mean quantity, also between 16 and 24 km altitude (<u>note</u>: date become sparse in for altitudes >~35km). The red points represent the individual data and the blue line is a 15-day running box-car average for these data. The grey shading indicates the standard deviation of the superposition, whilst the thin orange, black, and purple lines represent the upper quartile, the median, and the lower quartile of the superpositions. There is a clear decrease in the ozone partial pressure following the arrival of solar proton events.

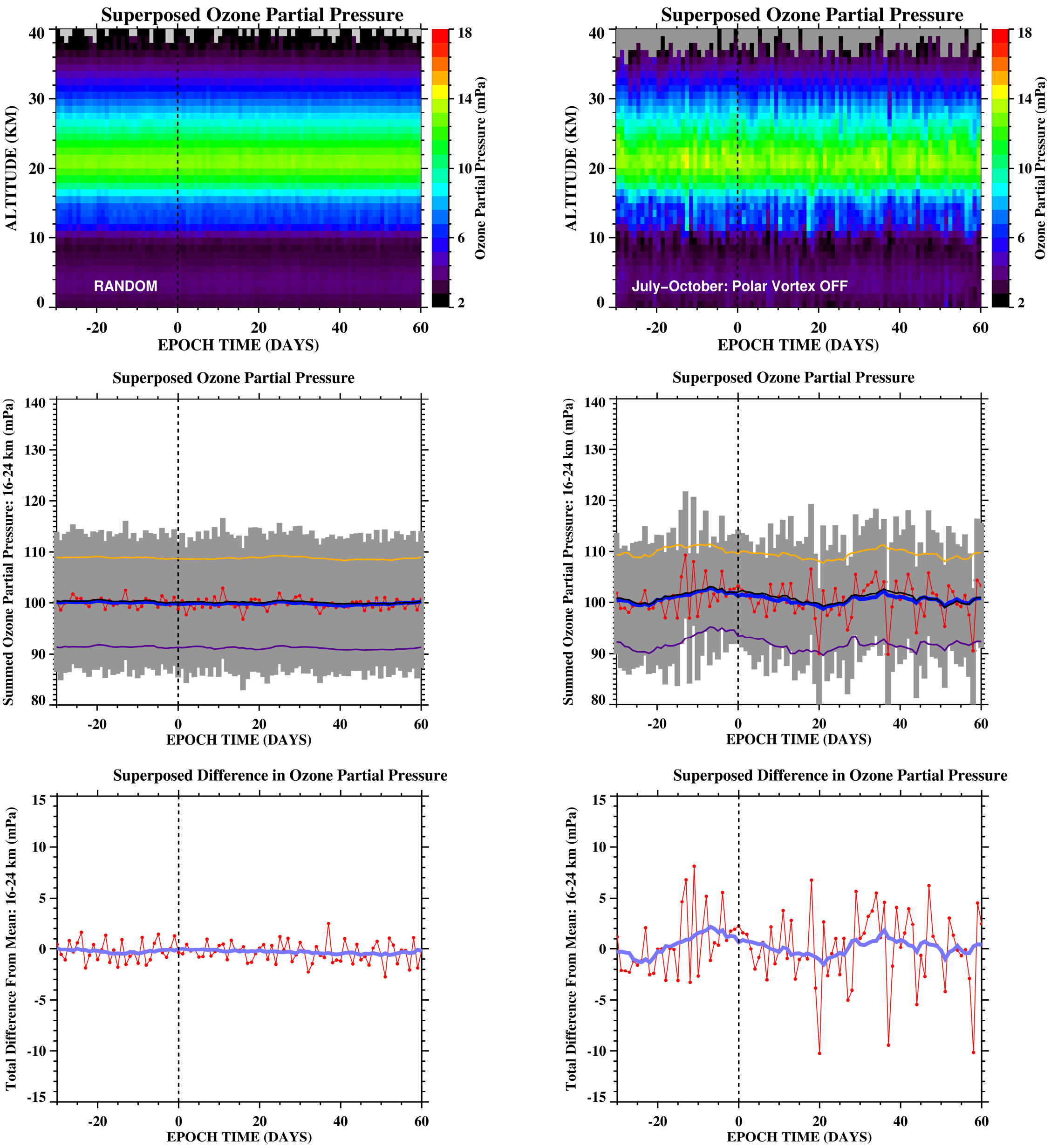


FIGURE 5: The same format as shown in Figure 4, but here data are only plotted for July, August, September, and October when the polar vortex is INACTIVE over Northern Finland. There is no clear trend for changes in the stratospheric ozone population following the SPEs.

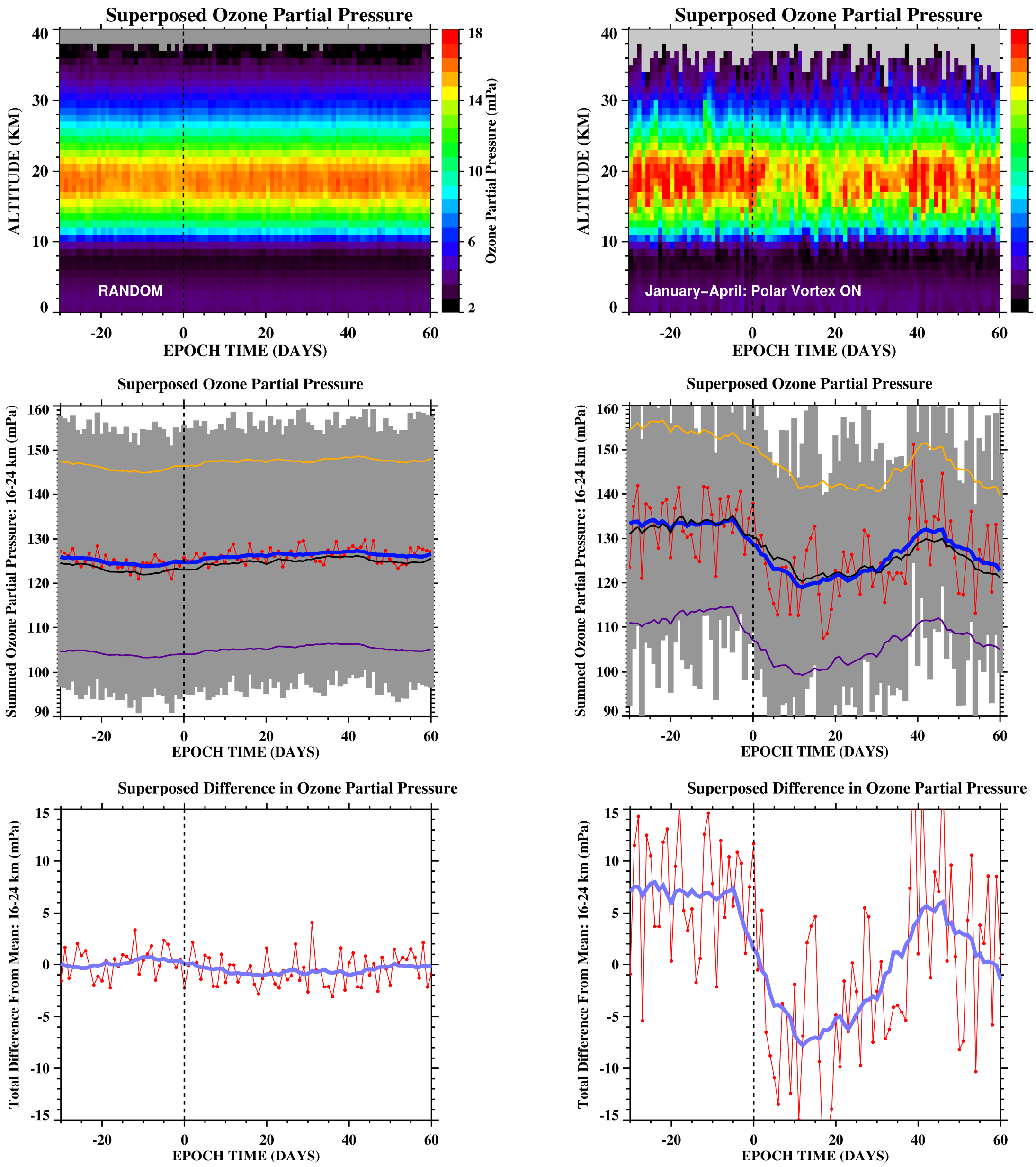


FIGURE 6: The same format as shown in Figure 4 and Figure 5, but here data are only plotted for January, February, March and April, when the polar vortex is ACTIVE over Northern Finland. There is a clear trend for a decrease in the stratospheric ozone population following the SPEs.

<u>HIGHLIGHTS</u>:

- No link between stratospheric ozone and geophsyical indices (24 h lag)
- Stratospheric ozone falls ~10% after solar proton events - below mean for ~40 days.
- Ozone decrease only occurs during polar winter months, no effect in summer.