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On the potential impact of large electron concentrations on mesospheric ozone

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Abstract. We use mid- and high-latitude mesospheric O_3 data combined with measurements of Total Electron Content (TEC) to conduct a preliminary study on the short-term coupling between electron concentrations and chemical processes in the upper atmosphere. In particular, we explore the possibility that ion chemistry have an impact on mesospheric ozone concentration under undisturbed atmospheric conditions. Ground-based measurements of mesospheric O_3 were carried out from the Network for the Detection of Stratospheric Change (NDSC) site of Thule Air Base (76.5° N, 68.8° W), Greenland, during January and February of 2002 and 2003, and from Plateau Rosa (or Testa Grigia; 45.9° N, 7.7° E, elev. 3500 m above mean sea level), on the Italian Alps, during several periods of 2004 and from January to March 2005. The Total Electron Content (TEC) values at the geographical coordinates of Thule and Plateau Rosa were obtained from the Global Positioning System (GPS) measurements of differential pseudorange and differential Doppler on L1 and L2 frequency bands. Although results rely on a small number of data points and a more quantitative study is desirable, our preliminary survey suggests that when electron concentrations in the upper atmosphere are sufficiently large, but the atmosphere is not perturbed by strong energy deposition events, a positive correlation between electron content and O_3 concentration may exist.

Key words. mesospheric ozone – total electron content – ion chemistry

1. Introduction

Energy deposition in the upper atmosphere through energetic electron precipitations or solar proton events can be associated with large and rapid variations of electron densities. While the impact of energetic charged particles on mesospheric NO has been observed in the past (e.g., Russell et al. 1984), and extensive modelling has been developed to study potential seasonal changes in stratospheric ozone (Callis, Natarajan & Lambeth 2001), there is still a lack of experimental evidence on the

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short-term impact of protons and electrons on mesospheric ozone concentrations.

Many of the chemical processes affecting mesospheric O_3 concentrations and involving large amounts of charged particles are quite uncertain (Solomon et al. 1981). Generally, energetic charged particles produce odd hydrogen HO_x ($H + OH + HO_2$) through chemistry initiated by ion pair production, and odd nitrogen NO_x ($N + NO + NO_2$) through dissociation of molecular nitrogen (e.g., Crutzen, Isaksen & Reid 1975). Both of these families partecipate in catalytic cycles which are believed to produce the significant depletion of ozone concentrations predicted by atmospheric models and observed by satellite measurements during large energy depositions in the upper atmosphere (e.g., Jackman et al. 2001; Degenstein et al. 2005). However, while large productions of NO_x affect polar ozone on timescales of months and predominantly below 50 km altitude (Callis, Natarajan & Lambeth 2001), HO_x is the most important contributor to the ozone balance above 50 km and is effective only on timescales shorter than a few days (Brasseur & Solomon 1984). In this preliminary study, we look for a measurable correlation between ionospheric electron densities and mesospheric O_3 under undisturbed atmospheric conditions, i.e., in the absence of large energy depositions in the upper atmosphere, in order to evaluate the likelihood of a significant and persistent impact of ion chemistry on polar mesospheric ozone.

2. Measurement description

Ground-based measurements of mesospheric O_3 were carried out from the NDSC site of Thule Air Base (76.5° N, 68.8° W), Greenland, during January and February of 2002 and 2003, and from Plateau Rosa (or Testa Grigia; 45.9° N, 7.7° E, elev. 3500 m above mean sea level), on the Italian Alps, during several one-week periods of 2004 and from January to March 2005 (e.g., de Zafra & Muscari 2004, de Zafra 1995). The rotational emission line of O_3 at 276.923 GHz was observed with a millimeter-wave receiver (GBMS) and two back-end acousto-optical spectrometers. The combination of these two spectrometers allows retrievals of O_3 vertical profiles from 15 to 80 km altitude with an uncertainty of approximately ± 15% (or 0.3 ppmv, whichever is the largest) and a vertical resolution of 7-10 km. The deconvolution of the O_3 emission line is obtained using a constrained matrix inversion algorithm (Twomey 1996). Measured spectra are integrated for ~ 1 hour.

The Total Electron Content (TEC) values at the geographical coordinates of Thule and Plateau Rosa were obtained from Global Positioning System (GPS) measurements of differential pseudorange and differential Doppler on L1 and L2 frequency bands, using a single station solution (Ciraolo & Spalla 1997). TEC values at the geographical coordinates of Thule were calculated from GPS soundings recorded at Thule itself, while TEC values relative to Plateau Rosa were estimated using GPS measurements from stations located in Turin (45.2° N, 7.7° E) and Zimmerwald (46.9° N, 7.5° E). The internal consistency (a measure of precision) of TEC values amounts to 1 to 3 TEC units (10^{16} electrons/m^2) for quiet conditions, while it can reach up to tens of TEC units for disturbed conditions when the electron density gradients can be extremely large. The GPS data referring to these three stations were retrieved at the International GPS Service (IGS) data bank. TEC values used and displayed in this study are averages calculated on time intervals overlapping with the duration of concurrent GBMS ozone measurements.

3. Discussion

Each ion pair produced by energy deposition in the upper atmosphere is thought to form two ozone depleting odd hydrogen constituents up to an altitude of ~ 70 km, with less HO_x being produced above 70 km (Solomon et al. 1981). However, the chains of ion and neutral reactions that are initiated from production of ion pairs and lead to HO_x formation may be interrupted by the recombination of intermediate ions with electrons, therefore reducing the amount of HO_x being formed and potentially leading to an increase of O_3.

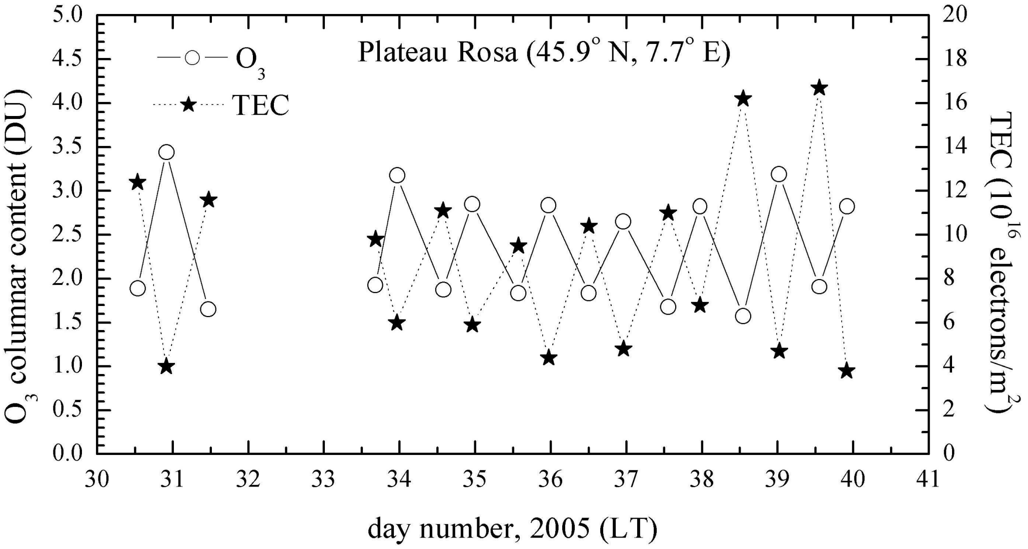

Fig. 1. A sample of O_3 columnar content (O_3 concentration integrated from 57 to 79 km altitude; empty circles connected by solid lines) and TEC (stars connected by dotted lines) time series for Plateau Rosa (Testa Grigia) during 2005. Ozone columnar content values are in Dobson Unit (DU). The diurnal strong anti-correlation due to the 24-hour solar illumination cycle dominates over any potential small direct effect of electron density variations on columnar mesospheric ozone.

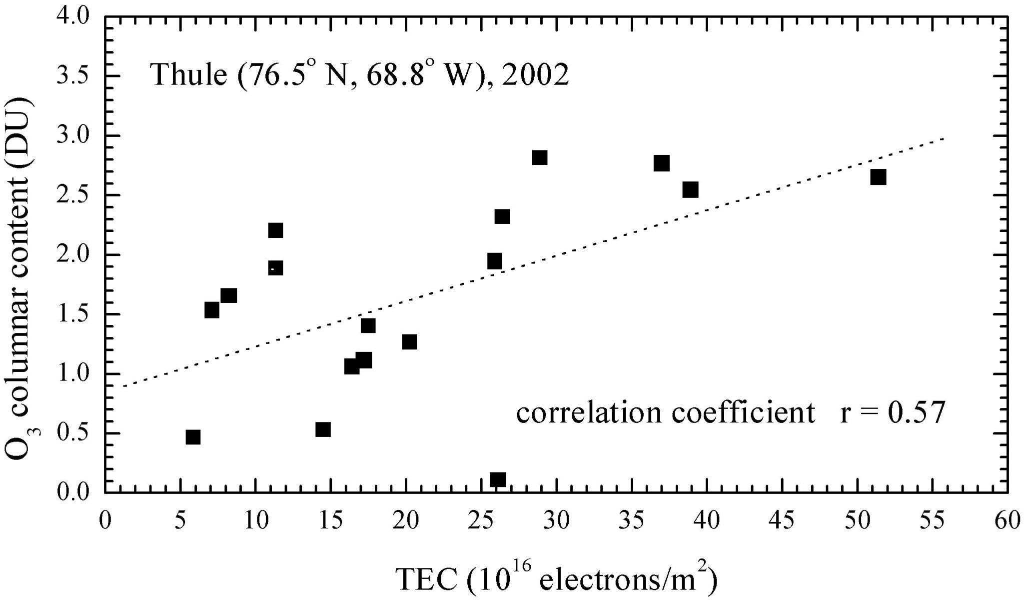

Fig. 2. Correlation scatter plot between TEC values and O_3 columnar contents (O_3 concentration integrated from 57 to 79 km altitude) observed at Thule (76.5° N, 68.8° W) during winter 2002. Ozone columnar content values are in Dobson Unit (DU). The positive correlation is supported by a good correlation coefficient (0.57), although a better statistics is necessary to confirm this qualitative result.

In order to investigate whether short-term variations in mesospheric O_3 content may be correlated with changes in electron concentration, we looked at mesospheric columnar contents of O_3 (O_3 concentration integrated over the 57-79 km vertical range) and paired them with concurrent TEC data. Although electron concentrations peak at altitudes above 200 km, and therefore TEC values are mainly determined by concentrations well above the vertical range of GBMS O_3 measurements, we believe that for this qualitative study TEC values serve the purpose of indicating the relative variations with time of electron densities in the mesosphere. This assumption should hold especially in polar regions, where we focus our attention, because perturbations to the electron distribution at polar latitudes are likely to follow the Earths magnetic field and propagate from the upper to the lower atmosphere. In Fig. 1, TEC values and columnar amounts of mesospheric O_3 measured at Plateau Rosa in 2005 display a strong anti-correlation. Similar results were also obtained with measurements carried out in 2004, Fig. 1 showing only a sample of the typical anti-correlation between mesospheric O_3 and TEC data from Plateau Rosa. However, the diurnal cycles of mesospheric O_3 and TEC (noticeable in Fig. 1) are caused by the diurnal variation of incident solar radiation and are in no way a signature of their mutual interaction: the 24-hour change in solar illumination controls the mesospheric O_3 and electron concentrations and dominates over any potential, minor direct influence of electrons on O_3. Therefore, it is required that the correlation of O_3 columnar content and TEC be investigated using measurements carried out during hours of darkness, in order to avoid the influence of solar radiation, and possibly with larger electron concentrations than those found at mid-latitudes.

Fig. 2 shows TEC data and O_3 columnar contents obtained at Thule during winter 2002, at solar zenith angles $\geq 90°$ (dusk or complete darkness). Although only few points are characterized by TEC values significantly larger than those observed at mid-latitudes, Fig. 2 suggests that a positive correlation between electron content and O_3 concentration may exist when a sufficiently large amount of electrons are present. It is worth stressing that this qualitative result is not necessarily inconsistent with the general finding (e.g., Jackman et al. 2001) of significant depletion of ozone concentrations during large productions of ion pairs. In fact, in unperturbed atmospheric conditions, a local increase of electrons does not necessarily mean an increase in ion pairs production, since transport can play a role in differentiating the distribution of electrons to that of other positive and negative ions.

Two are the main improvements to this study that might clarify whether a positive correlation between electron content and O_3 concentration in the mesosphere exists: 1) obtain electron density measurements in the mesosphere and use those instead of TEC values; 2) as part of planned future GBMS measurement campaigns in Thule, a larger number of O_3 measurements should be carried out daily in order to increase the number of correlation data points available and improve the statistics.

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