

The hydroxyl radical as an indicator of SEP fluxes in the high-latitude terrestrial atmosphere

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Abstract

The low background values at nighttime of the mesospheric hydroxyl (OH) radical make it easier to single out the atmospheric response to the external solar forcing in Polar Regions. Because of the short lifetime of HO_x, it is possible to follow the trails of Solar Energetic Particle (SEP) events in the terrestrial atmosphere, as shown by Storini and Damiani (2008). The sensitivity of this indicator makes discernible not only extreme particle events with a flux peak of several thousand pfu [1 pfu = 1 particle/(cm² s sr)] at energies >10 MeV, but also those with lower flux up to about 300 pfu. Using data from the Microwave Limb Sounder (MLS) on board the EOS AURA satellite, we examined the correlation of OH abundance vs. solar proton flux for almost all the identified SEP events spanning from November 2004 to December 2006 (later on no more SEP events occurred during Solar Cycle no. 23). The channels at energies greater than 5 MeV and 10 MeV showed the best correlation values ($r \sim 0.90$ – 0.95) at altitudes around 65–75 km whereas, as expected, the most energetic channels were most highly correlated at lower altitudes. Therefore, it is reasonably possible to estimate the solar proton flux from values of mesospheric OH (and viceversa) and it could be useful in studying periods with gaps in the records of solar particles.

In addition, the SEP events of September 2005, characterized by an evident hemispheric asymmetry of the SEP-induced OH formation, have been examined separately. The impact of the SEPs on the hydroxyl radical was appreciably stronger in the Northern Hemisphere than in the Southern at altitudes above roughly 60 km and also the nighttime OH layer was involved. Preliminary analyses performed with other MLS data (i.e., O₃, H₂O, T) suggest that atmospheric in situ conditions could play a relevant role in explaining the hemispheric asymmetry of OH.

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1. Introduction

Among solar-terrestrial coupling phenomena, solar energetic particle events play a significant role. They consist of transient particle (mainly proton) emissions from the Sun, associated with solar flares and coronal mass ejections (CMEs) generally driven by shock waves. They travel in the interplanetary medium and can enter the terrestrial environment if the solar source region is magnetically well con-

nected to the Earth (see, for instance, Storini et al., 2005 for the flare sources associated with most of the relativistic SEP events recorded during Solar Cycle no. 23). In this case, geostationary satellites [like those of the GOES (Geostationary Operations Environmental Satellite) series; for an overview of the satellite platform and instruments, see the GOES Data Books available at: <http://www.ngdc.noaa.gov/stp/GOES/goes.html>] are able to record them. The energy spectra of SEP events can be very different from one event to the other. They can reach GeV energies and can be recorded at ground level by the neutron monitor network as Ground Level Enhancements (GLEs), especially in the polar

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cap regions (geomagnetic latitudes greater than 60°). Typically the SEP flux is inversely proportional to energy, and the counting-rate increase observed at neutron monitor energies, which is linked with the particle flux around the Pfofzer Maximum level (see Pfofzer and Regener, 1935), could be not proportional to the SEP flux at lower energies (these particles interacting mostly at the mesospheric level).

The occurrence of large SEP events deserves special attention because of its implications for the chemistry of the middle atmosphere (we recall that the NOAA/Boulder definition of SEP event states that the event starts when the proton flux $J [E > 10 \text{ MeV}]$ is greater than/equal to 10 pfu for three consecutive 5-min intervals and the event ends when J falls below 10 pfu). SEPs produce enhanced ionization roughly between 90 km and 30 km, in addition to the other more constant sources (such as X-rays, UV, extreme UV, Lyman α and β , and galactic cosmic rays; see Brasseur and Solomon, 2005). The result of these phenomena is the triggering of reaction paths connected to both ion and neutral chemistry which affect reactive components like NO_x ($\text{N} + \text{NO} + \text{NO}_2$) and HO_x ($\text{H} + \text{OH} + \text{HO}_2$). They are able to decrease the ozone by efficient catalytic cycles, in the stratosphere and mesosphere, respectively (see Jackman and McPeters (2004) for a review). Recent studies, using data from new satellite sensors, have been focusing on the SEP-induced effects on other reactive components like ClO_x ($\text{Cl} + \text{ClO} + \text{ClO}_2$) (von Clarmann et al., 2005) and on reservoir species, e.g., N_2O_5 , ClONO_2 , HNO_3 , N_2O , HOCl , HCl (see, among others, López-Puertas et al., 2005b; Orsolini et al., 2005; Verronen et al., 2008; Funke et al., 2008; von Clarmann et al., 2005; Damiani et al., 2009). Further, the Whole Atmosphere Community Climate Model has been used in two recent companion papers (Jackman et al., 2008; 2009) to study short-, medium- and long-term effects in the polar atmosphere induced by the main SEP events of the satellite age. In these papers, attention is drawn to some discrepancies between the above model and satellite data (e.g., N_2O_5 , ClONO_2 , HNO_3) and to the stratospheric ozone decreases of $>10\%$ for up to 5 months after the largest events. Finally, the passage of solar energetic particles influences the atmospheric temperatures (because of the correlated ozone decrease, ozone being a strong absorber of solar radiation), winds (Jackman et al., 2007; Krivolutsky et al., 2006), noctilucent clouds (von Savigny et al., 2007) as well as the polar mesosphere summer echoes recorded by ground based radars (Morris et al., 2009).

Several models showing the rise of NO_x and HO_x after the occurrence of large SEP events have been constructed since the late seventies (e.g., Crutzen et al., 1975; Solomon et al., 1981) but, only after August 2004 did the continuous availability of hydroxyl and peroxy (HO_2) radical data coming from the Microwave Limb Sounder (MLS) on board the near polar orbit EOS – AURA satellite (Schoeberl et al., 2006; Pickett, 2006) make it possible to check models against experimental data (i.e., Verronen et al., 2006, 2007).

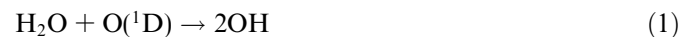
Nowadays the release of the improved MLS OH data version 2.2 (Pickett et al., 2008; Livesey et al., 2007; MLS data available at: <http://mirador.gsfc.nasa.gov/>) makes it possible to investigate OH variability up to 0.003 hPa ($\sim 86 \text{ km}$) with good vertical resolution ($\sim 2.5 \text{ km}$). It allows analysis of the OH response to the nighttime SEP flux in the upper mesosphere also beyond the range of elevated abundance of OH radicals centered on about 82 km, as shown by Pickett et al. (2006b). In a previous study (Damiani et al., 2008), we could not take into account this factor because the MLS OH data Version 1.5 had an increasing error with altitude and a low vertical resolution in the upper mesosphere.

Because of the short lifetime of HO_x species (Solomon et al., 1981), the hydrogen radicals follow the trail of the incoming SEP in the terrestrial atmosphere (Storini and Damiani, 2008). This is because their variability is mainly linked to the atmospheric chemistry, contrary to what happens with the NO_x variability, in which both chemical and transport phenomena are present. In this way, the mesospheric OH abundance can be utilized as an indicator for SEP presence in the terrestrial environment, as discussed in the following sections.

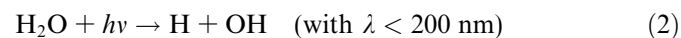
2. The hydroxyl abundance in the polar atmosphere

The main source of mesospheric OH radicals is the water vapor which reacts with atomic oxygen or which is photolysed by UV radiation (Brasseur and Solomon, 2005).

Below 65–70 km the main path is:



and above 65–70 km:



In Polar Regions the abundance of the OH radicals depends on the sunlight presence; hence, the summer abundance is very different from the winter. The low availability of excited atomic oxygen, the very modest photolysis process and the lack of transport from mid latitudes to the inner polar vortex (the majority of the HO_x is produced at mid latitudes) lead to a low hydroxyl radical concentration during the winter season compared with summer. Moreover, the dissimilar abundance of mesospheric water vapor in winter ($\sim 1 \text{ ppmv}$) and summer ($\sim 8 \text{ ppmv}$) is likely to contribute to the different behavior of the OH (Hervig et al., 2003). Therefore, in Polar Regions elevated OH values are observed in summer (some ppbv) and lower ones in winter ($\ll 1 \text{ ppbv}$), roughly between 40 and 70 km. Furthermore, in the upper mesosphere (above 70 km) there are two peaks at slightly different altitudes. They appear at about 75 km and 82 km during day and night hours, respectively (Pickett et al., 2008).

The odd hydrogen is mainly produced at mid latitudes, transported to the polar winter thermosphere and subsequently descending towards the upper mesosphere. In the

middle/lower mesosphere, the concentration of atomic hydrogen is only a small fraction of the total HO_x molecules. This is because the high atmospheric density results in the existence of the so called third body and the atomic hydrogen can react with molecular oxygen to form HO_2 . When this reaction becomes less important, the H concentration increases and the ozone destruction by the atomic hydrogen, with subsequent OH production, becomes the main path. These processes are involved in the formation of the upper mesospheric OH layer (at ~ 82 km) at nighttime (see Pickett et al., 2006b).

The lifetime of the HO_x components is very short in the middle mesosphere (less than 1 h at 80 km; e.g., Brasseur and Solomon, 2005) and this prevents transport from influencing the OH distribution there, but at slightly higher altitudes it grows quickly (at 85 km it is ~ 30 days; see Pickett et al., 2006b) and dynamics can assume an important role.

Since the main factor influencing the OH abundance is the solar zenith angle (SZA), a very limited variability of its mixing ratio under constant nighttime condition is expected. Therefore, during the winter months in Polar Regions the almost exclusive source of OH variability arises from the SEP event occurrence. However, recent experimental results showed variation of abundance and altitude of the mesospheric OH layer under unusual meteorological conditions (Damiani et al., 2010). The upper panel of Fig. 1 shows the MLS OH abundance (nighttime OH profiles, $\text{SZA} > 95^\circ$) in the region within the geo-

graphic latitudes $75\text{--}82^\circ\text{N}$ during January–March 2006, when no SEP events occurred and under low geomagnetic activity (see the lower panels of Fig. 1). The sudden descent of the OH layer and its settling at an altitude ~ 5 km lower than usual, with larger values and width until early March, are clearly visible.

The variability of the layer of OH^* (i.e., vibrationally excited hydroxyl radical at the nominal altitude of ~ 87 km) induced by the meteorology is an aspect recently discussed by Winick et al. (2009). Under extreme events of strong air descent (i.e., northern winter of 2004 and 2006) this layer is shown to undergo a downward displacement of $\sim 5\text{--}8$ km due to the descending of O_x ($\text{O} + \text{O}_3$). Since it is deactivated by collisional quenching at lower altitudes, the same cause similarly also provoked the variability reported in Fig. 1. The OH layer in the ground vibrational state experienced a similar behavior also in February/March 2009 and in both cases the downward displacement of this layer induced an OH mixing ratio (at the altitude of 0.015 hPa) of the same order of magnitude of that caused by the SEP events of January 2005 (Damiani et al., 2010).

Although these events, lasting only a few weeks and occurring on very rare occasions (although they are not so rare in recent times, i.e., extra-ordinary descent occurred after stratospheric warming events in 3 out of 6 Northern Hemisphere winters during 2004–2009; Randall et al., 2009), are more or less isolated, we want to remark that they could contaminate the OH radicals utilized as a SEP

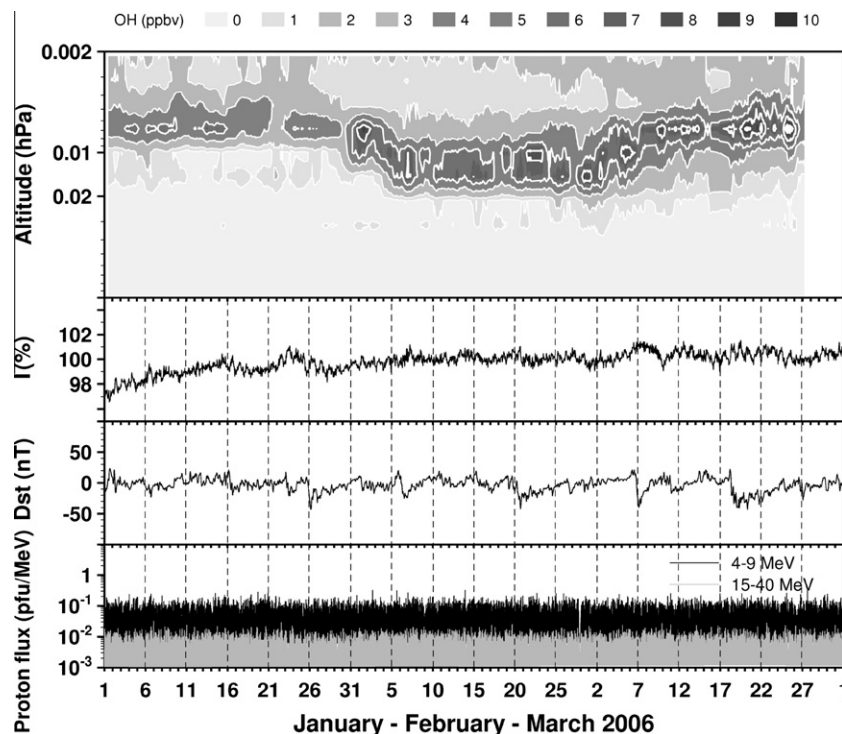


Fig. 1. From top to bottom panels: time series of MLS OH abundance in the region within the geographic latitudes $75\text{--}82^\circ\text{N}$, cosmic ray intensity of the Rome neutron monitor (data available at: http://webusers.fis.uniroma3.it/~svirco/pag_2.html), Dst geomagnetic index, GOES 11 Proton flux at 4–9 MeV and 15–40 MeV channels.

indicator. For this reason, it is more useful to find traces of SEP impact in the lower-middle mesosphere at about 65–75 km where they are much more evident because there the low OH background is never influenced by the variability of the upper OH layer.

In order to evaluate the effects of the SEPs with moderate flux (here, moderate flux means roughly $100 \text{ pfu} < J < 5000 \text{ pfu}$ at the GOES channel $>10 \text{ MeV}$) on hydroxyl radicals, it is necessary to consider time intervals with a low background of OH. Damiani et al. (2009), considering the SEP flux in the GOES channel $>10 \text{ MeV}$, elaborated a rank list for the 10 largest SEP events occurring during Solar Cycle no. 23. The effects of the SEP events such as those of January 2005 (Seppälä et al., 2006; Storini and Damiani 2008), May 2005, September 2005 and December 2006, if compared with the more famous and investigated ones of July 2000 or October 2003 (Jackman et al., 2001, 2005; Seppälä et al., 2004; von Clarmann et al., 2005; López-Puertas et al., 2005a,b; Rohen et al., 2005), are much less striking. In fact, studying the response of MLS OH after the SEP events of January 2005, Damiani et al. (2008) outlined that a clear OH signature was present only in the winter hemisphere whereas in the summer hemisphere the effects were hidden by the elevated background values. Conversely, after the largest SEP events, the larger production of HO_x makes it possible to record important variations even in the mesospheric chemistry of the summer hemisphere in spite of the high background of HO_x .

It is important to remark that the HO_x diurnal cycle also allows us to separate some variability related to moderate SEP events. The OH levels increase right after sunrise and follow the SZA daily cycle. Then the OH decreases soon after sunset. In this way, the OH background changes quickly under twilight conditions with high and low values during the day and night, respectively (Solomon et al., 1981; Pickett et al., 2006a). Therefore, also during a few

night hours, it is possible to observe a variability induced by the SEPs.

3. The hydroxyl radical as an indicator of SEP events

The possibility of utilizing the MLS OH as an indicator of SEPs in the terrestrial environment is connected to the presence of a constant abundance of OH inside the Polar Regions during the winter months. The geographic latitudinal range $75\text{--}82^\circ$ is a suitable region to perform such a study which can be conducted by zonal means on a daily basis. We choose this region for three reasons. It is completely inside the polar caps where the SEP's impact is quite uniform; it is an area where the condition of winter night is roughly maintained for many months; it is the core of the polar vortex thus it is less disturbed by planetary waves. Taking into account the availability of MLS OH, we studied the relationships between the OH abundance and the SEP events from November 2004 to December 2006 (after which no more SEPs occurred) during the descending phase of Solar Cycle no. 23. The main SEP events occurred in November 2004, January–May–September 2005 and December 2006 (see Table 1 for the complete list of the SEP events). Since our analysis requires that MLS OH is observing during nighttime, the SEP-induced effects have been investigated in the Northern Hemisphere (NH) from October 2004 to March 2005 (plus December 2006) and in the Southern Hemisphere (SH) from April 2005 to September 2005. Our condition of nighttime is met when the $\text{SZA} > 95^\circ$. The MLS data has been checked following the recommendations of Pickett et al. (2008) and Livesey et al. (2007). The numbers of OH profiles utilized for the daily zonal means are $\sim 250\text{--}200$ in mid-winter, decreasing to ~ 50 (100) at the beginning/end of the winter in NH (SH) owing to the incoming solar illumination. In this way, the relative error of the means is $\sim 5\%$ for most of the period.

Table 1
Features of the analyzed SEP events. First four columns: SEP event start, time of its maximum flux, proton flux and number of samples per SEP utilized in the correlations of Fig. 2 (see the text). Last three columns: OH increase (%) at 55 km, 65 km and 75 km (referred to the pre-SEP day).

SEP events (pfu unit)				OH increase (%)		
Start (YY, MM DD/UT)	Maximum (Day/UT)	Proton flux at $>10 \text{ MeV}$	Number of samples per SEP	$\sim 55 \text{ (km)}$	$\sim 65 \text{ (km)}$	$\sim 75 \text{ (km)}$
04, November 01/0655	01/0805	63	0	–	–	–
04, November 07/1910	08/0115	495	9	–	99	397
05, January 16/0210	16/1840	365	2	76	234	279
05, January 17/1240	17/1750	5040	3	1221	2131	894
05, January 20/0650	20/0810	1860	4	575	1030	448
05, May 14/0525	15/0240	3140	4	160	379	328
05, June 16/2200	17/0500	44	0	–	–	–
05, July 14/0245	15/0345	134	6	–	–	–
05, July 27/2300	29/1715	41	8	–	–	–
05, August 22/2040	23/1045	330	4	–	93	110
05, September 08/0215 ^a	11/0425	1880	0	171	198	104
05, September 14/0040 ^a	15/0905	235	0	–	–	–
06, December 06/1555	07/1930	1980	7	110	954	940
06, December 13/0310	13/0925	698	4	–	315	543

–: No evaluation (see the text)

^a Excluded from the correlation analyses.

As expected the values of mesospheric OH are quite similar for the two hemispheres because they depend mainly on the solar illumination, and we selected the data to keep this constant. As a reference we can state that in the middle of winter the typical OH abundance is ~ 0.01 ppbv, 0.1 ppbv and 1 ppbv at ~ 55 km, 65 km and 75 km, respectively, and that, usually, the relative standard deviation of the quiet periods is ~ 20 – 30% . Since the September 2005 SEP event occurred under a slightly different condition of the solar illumination, such events have been examined separately (see next section). Indeed, although the OH profiles recorded in SH also in September keep our nighttime condition for ~ 100 samples per day, the OH abundance of the zonal means is roughly twice the mid-winter values at the above-mentioned altitudes (not beyond ~ 75 km). This is discernible in the SH data whereas, for the NH, during the corresponding season, such high abundance is not seen. Therefore, the reason for such behavior is not very clear. It could depend on the different hemispheric abundance of the gas source (we checked MLS O_3 and H_2O but a definitive conclusion was not reached), on the employed SZA which could not assure a complete absence of sunlight (although it seems quite improbable at least at ~ 50 – 60 km) and finally on the different local solar time (LST) of the satellite passage (in the selected regions, MLS data in NH are recorded before sunrise whereas in SH the measurement occurs after sunset, thus different values of $[O]$ could cause this hemispheric discrepancy in OH, see Eq. (2) in Section 2).

Fig. 2 shows the correlation values (r) of daily proton flux vs. MLS OH for various altitudes and energies. On the left side of Fig. 2 we used the OH increment induced by SEPs (i.e., the difference between the daily OH mixing ratio of SEP influenced days and the daily OH mixing ratio of the pre-event day) and on the right side the actual daily OH value of the SEP influenced days. For these analyses, 51 samples (i.e., 51 days for which the proton flux was

above the background level observed by GOES) have been employed. For each event, the number of days was selected by using the proton channel where the flux enhancement lasts longer as compared to typical values (usually at energies greater than 5 MeV). Then, in order to correlate the proton flux against the OH abundance at different energies, for each event we used the same number of days for all channels. As mentioned above, the SEP events of September 2005 have been not included in this kind of analysis. The quite similar results of the correlations on the right and left side of Fig. 2 indicate that, normally, the SEP-induced variations are deeply dominant over the day to day variability, at least during the selected days.

The channels at energies greater than 5–10 MeV reach the best correlation values (up to 0.95) at altitudes roughly between 65 and 75 km, whereas at lower altitudes the r -value decreases. As expected, the most energetic channels have an opposite trend, hence low correlation coefficients at high altitudes and high values at low altitudes. Since only a few SEP events reached a noticeable flux at elevated energies (e.g., >50 MeV), at these levels the correlation values are lower than at others. Note also that at ~ 80 km the best correlation occurs for energies greater than 5–10 MeV. This occurs because the flux at energies greater than 1 MeV, which energy should impact similar altitudes, is also influenced by the protons of the Earth's radiation belt. Therefore, when utilizing it as indicator of SEP events, we have a lower accuracy.

The three columns on the right side of Table 1 report the OH radical increase (%), induced by the investigated SEP events, at three different altitudes. We did not report the variations for the smaller events because of their negligible or absent impact on the daily OH (comparable to the day to day variability). The lowest flux able to provide a reasonable variation in the daily OH is ~ 300 pfu. Note the largest enhancements induced by the SEP events of January 2005 (maximum increase of more than 2000% at

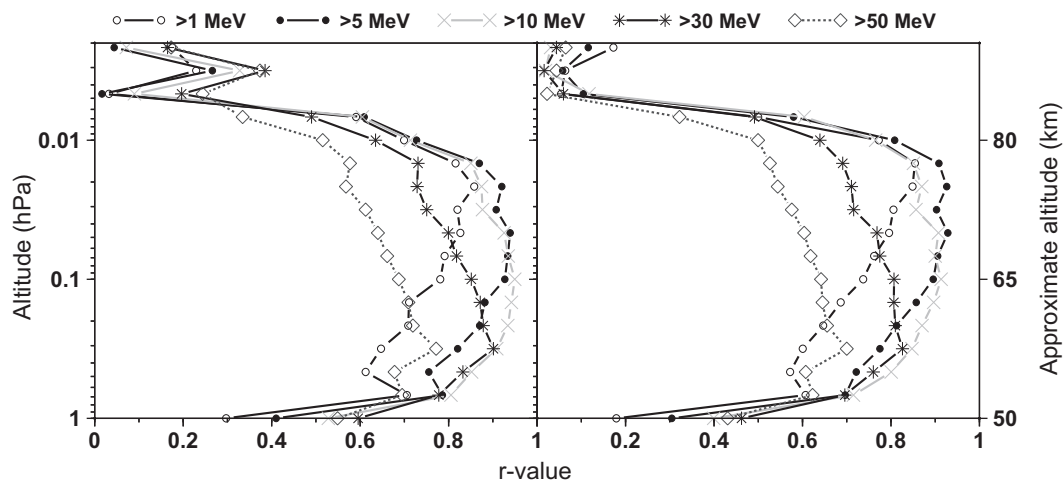


Fig. 2. Correlation coefficients (r -values) of proton flux vs. OH mixing ratio for various altitudes and energies. Left side: proton flux vs. OH increment (i.e., the difference between the daily OH mixing ratio of SEP influenced days and the daily OH mixing ratio of the pre-event day) induced by SEP events. Right side: proton flux vs. actual daily OH of SEP influenced days.

~65 km) and the limited effects of the SEPs of May and September 2005 in spite of their considerable flux. The short-lived flux enhancement of the May event and the high background of atmospheric OH in September (see above) prevented a larger impact on the mesospheric chemistry. In the next sections, the features of the SEP events of September 2005 and their impact on the hydroxyl radical and other minor components will be examined in detail.

4. The SEP events of September 2005

Since we are interested in comparing the effects of the September 2005 SEP events in both hemispheres, OH abundances recorded within belts with the same geomagnetic latitudes and comparable solar illumination are examined. Owing to the LST of the satellite's passage in the investigated regions, we chose to analyze the MLS OH profiles between 60° and 70° geomagnetic latitudes. We made the transformation from the geographic coordinates to the Corrected GeoMagnetic (CGM) coordinates by the service available at http://omniweb.gsfc.nasa.gov/vitmo/cgm_vitmo.html. The algorithm is solely based on the Definite/International Geomagnetic Reference Field (DGRF/

IGRF) models for Epochs 1900–2010. In this way, we employed OH profiles inside two geomagnetic latitude belts between 60° and 70°N and S, corresponding to ~48°N to ~77°N and ~45°S to ~82°S geographic latitudes.

As a consequence of this choice, 5946 profiles of MLS OH v2.2 in the NH and 6164 in the Southern have been used to study the impact of solar particles during September, 2005. The number of samples utilized per day [usually, more than 100 (150) at night and 100 (70) during the day, in the Northern (Southern) hemisphere] assures a good relative precision of the zonal averages also in the upper mesosphere (see Pickett et al. (2008) for a detailed discussion on the precision and accuracy of the MLS OH and HO₂ data).

As a first step, in order to single out the SEP-induced effects in both hemispheres, we filtered the data by utilizing only nighttime profiles. In addition, for a comparison, we chose to show also the OH data recorded during the day. Due to the strong daily variability of the HO_x following the SZA, it is important to recall that in September the SH has a nighttime longer than the NH (to help the reader we can state that, e.g., on September 1 at 70°N there are ~7 night hours whereas at 70°S ~13 hours).

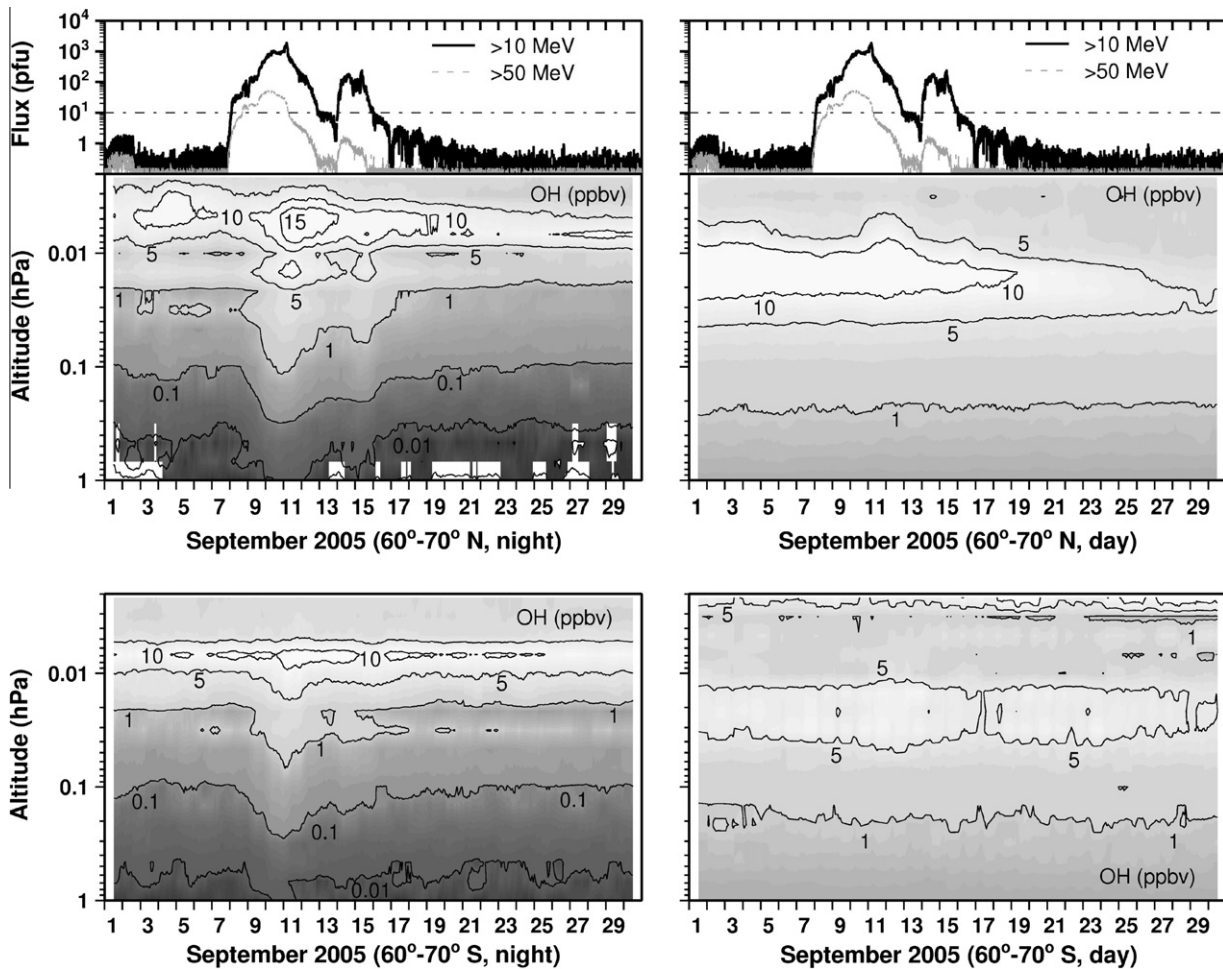


Fig. 3. Main panels: time series of MLS OH abundance between 60° and 70° geomagnetic latitude in Northern (upper panel) and Southern (lower panel) hemispheres (left side: night hours; right side: day hours). Small top panels: time series of the GOES 11 proton flux at >10 MeV and >50 MeV.

Fig. 3 (left side) shows the time series of the OH radicals during September 2005 in both hemispheres for night hours (SZA > 95°; North hemisphere: ~2.3–3.9 hour LST and South hemisphere: ~17.3–1.0 hour LST) from 1 hPa (~50 km) to 0.002 hPa (~88 km – we extended by ~2 km the upper limit of the data to improve the clarity of the figure). Note the sudden rise of about an order of magnitude in the upper mesosphere. The OH enhancement begins roughly at the same time as the SEP (on September 8) and it reaches a peak at a slightly delayed moment in the lower and upper mesosphere according to the different energies involved (this is because the more energetic the particles are the earlier their impact on the terrestrial atmosphere occurs). Moreover, in order to better evaluate the potentiality of OH as a SEP indicator, it is important to observe the further modulation on the OH radicals induced by the moderate SEP events that occurred on September 14 (see Fig. 3). The GOES 11 proton flux on these days was only ~200 pfu at >10 MeV (see the flux profile in the top panels), nevertheless it was able to leave its mark on the OH. Also the SH shows SEP effects, but they are less pronounced than in the NH in both the middle and upper mesosphere.

The right side of Fig. 3 shows the OH abundance in September 2005 in the same regions but during day (SZA < 80°; NH 11.4–13 hour LST and SH 14.2–15.9 hour LST). As already stated before, the background of the hydroxyl radical is now completely different: the OH values present in the lower mesosphere are greatly enhanced compared with at the same altitudes at night. Moreover, there is an OH peak just below 0.01 hPa in both hemispheres. Under these conditions the SEP signature in the OH seems to be completely absent. We can just observe the modulation present above 0.01 hPa in the NH after the most intense SEP event, but there are no indications to assert that it depends on the solar proton flux.

To better point out the enhancements due to SEP events, the following Fig. 4 shows the time series of OH (recorded inside the same belts as the previous Fig. 3) at 0.1 hPa (~65 km), 0.02 hPa (~75 km), 0.01 hPa (~80 km) and 0.006 hPa (~82 km) in September 2005 during night hours. A more intense impact on the NH is evident at all altitudes either as an absolute value or percentage increases. The pre-SEP OH abundance is similar in both hemispheres only at the lower layer (0.1 hPa), whereas the upper layers show higher OH values in the SH. At 0.1 hPa the Northern OH peak is more than twice the Southern, whereas at higher altitudes this difference is reduced because of the higher OH background of SH. Furthermore, we can single out the small peak due to the SEP event of 14 September in both hemispheres, and also in this case the effects are smaller in the Southern region.

It is interesting to remark that also the layer of elevated OH above 0.01 hPa is involved (see the OH trend at 0.006 hPa in Fig. 4). Increased OH values are evident especially in the NH from about 10 to 18 September (see the contour line of 10 ppbv in Fig. 3) even if it is not simple

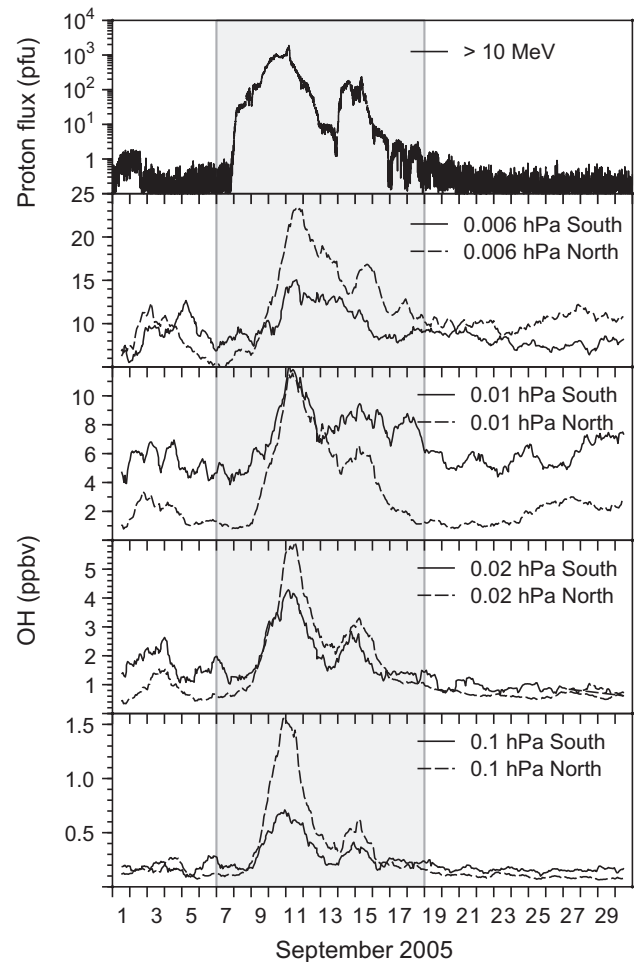


Fig. 4. From top to bottom panels: time series of the GOES 11 proton flux at >10 MeV and MLS OH abundance at 0.006 hPa (~82 km), 0.01 hPa (~80 km), 0.02 hPa (~75 km) and 0.1 hPa (~65 km) for September 2005 between 60° and 70° geomagnetic latitudes of both hemispheres.

to discern the contribution of the SEPs over the pre-existing seasonal trend of hydroxyl radicals. Anyhow, during the maximum solar particle flux, the OH layer in the NH seems to rise to at least twice the level observed in pre-SEP days (despite its elevated background) and the OH increase is present also at altitudes higher than SH ones. These features are in contrast to past models (e.g., Solomon et al., 1981) and other experimental results which showed a feeble impact of solar protons on the HO_x species above 80 km. For instance, with respect to the nighttime OH layer, the maximum mixing ratio recorded by MLS is ~12 ppbv for the SEP event of January 17, 2005 whereas it is ~22 ppbv for September 11, 2005 (values averaged over the geographic latitudinal range 60–70°N, see also the power panel of Fig. 5). Obviously the different season partly contributes to increase the September background over which the additional SEP-induced OH appears (see below).

During the SEP event of September 2005 a somewhat larger contribution of less energetic protons than in other

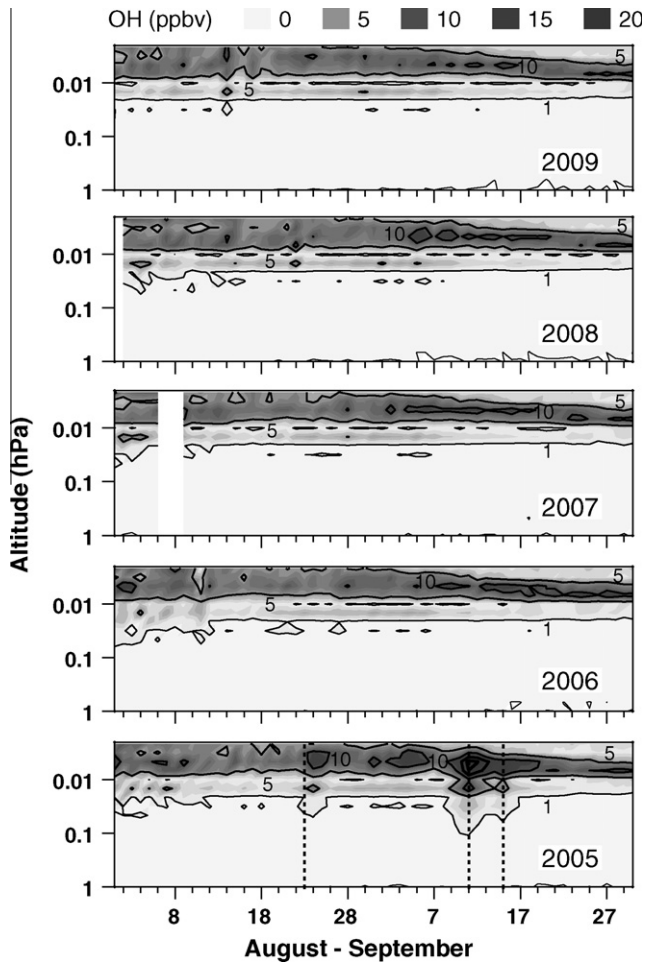


Fig. 5. Contours of OH mixing ratio in the geographic latitudinal range 60–70°N in August–September from the years 2005 (bottom) to 2009 (top).

events examined was noted (e.g., at the channel >1 MeV the daily proton flux is 7169 pfu vs. 13,222 pfu for January 17 and September 11 2005, respectively). Since they impact at elevated altitudes, this could explain the large OH mixing ratio present in the upper mesosphere of the NH during the event. Furthermore, the intense increase of the nighttime OH layer could not depend only on the proton flux but, for example, also on energetic electron precipitation (as already suggested by Verronen et al. (2007) regarding the SEP events of January 2005), although it is not simple to check this (see Rodger et al., 2007).

Since the seasonal evolution could play a role in explaining the larger SEP-induced OH abundance of the NH compared with the SH, Fig. 5 shows the daily MLS OH in August–September for the years 2005–2009, facilitating the separation of the different contributions between the seasonal evolution and external solar forcing. The OH values are averaged over the geographic latitudinal band 60–70°N to highlight the seasonal trend representative of the polar vortex region. The descent and the increased mixing ratio of the OH layer are roughly present every year in September. Note also the similar shape of the SEP-induced

OH increase of September 2005 shown in Figs. 3 and 5 which suggests that such changes are not confined to the geomagnetic polar cap.

Further analyses with MLS data (see Fig. 6 for September 2005) revealed that the descending trend of the OH layer in September follows the seasonal downward displacement of the ozone and water vapor in the mesopause. In particular, the upper limit of the OH layer moves down together with the lower boundary of the secondary maximum of the O_3 . Therefore, the OH variations are consistent with changes in H_2O and O_3 , being both the main sources of OH at these altitudes.

The highest OH mixing ratios (from about 10 to 18 September), caused by the SEP events, are further enhanced because they occurred over a pre-existing seasonal evolution. On the contrary, the increased OH in the early days of September 2005 (before the SEP) seems related only to the seasonal evolution and not to external forcing. Several elements lead us to conclude this. Firstly, the enhancement is not related to any solar particles increase (the slight modulation occurred on September 1 and 2 has a too low a flux to be taken into account); then there is no evidence of OH variability in the SH during the same days; finally, Fig. 5 shows that this variation is quite similar to the natural variability of the other years.

5. Hemispheric OH asymmetry induced by the SEP events of September 2005

The asymmetric effects of the SEPs on the hemispheric OH are a fascinating aspect but explaining its origin it is not a simple task. To our knowledge this is the first time that the evaluation of the hemispheric impact of a SEP event on the hydroxyl radical is performed by data recorded under roughly similar conditions. Past studies have been focused on the asymmetric response of the ozone but they were conducted in different seasons and under different SZAs. For instance, Rohen et al. (2005) showed the interhemispheric O_3 difference for the SEP events of October/November 2003. In such conditions, O_3 depletion in the winter hemisphere stronger than in the summer was emphasized and the cause was attributed to the summer photolysis of the ambient H_2O and therefore higher ambient, non-SEP-produced HO_x concentrations. There, OH data recorded in both hemispheres under roughly similar nighttime conditions are compared. Due to the extreme reactivity of the hydroxyl radical, a similar SEP-induced OH abundance is expected (excluding the seasonal variability of OH), but this is not confirmed by Figs. 3 and 4 (where the same bands of geomagnetic latitude have been examined in both hemispheres). Because of the improbability of a relevant hemispheric asymmetry of the solar proton impact on the polar atmosphere, the reason for the different OH response to SEPs is unlikely to be connected to the external forcing but rather to different in situ atmospheric conditions.

In Fig. 6, the hemispheric abundance of the OH and its gas sources and temperature are examined inside the geo-

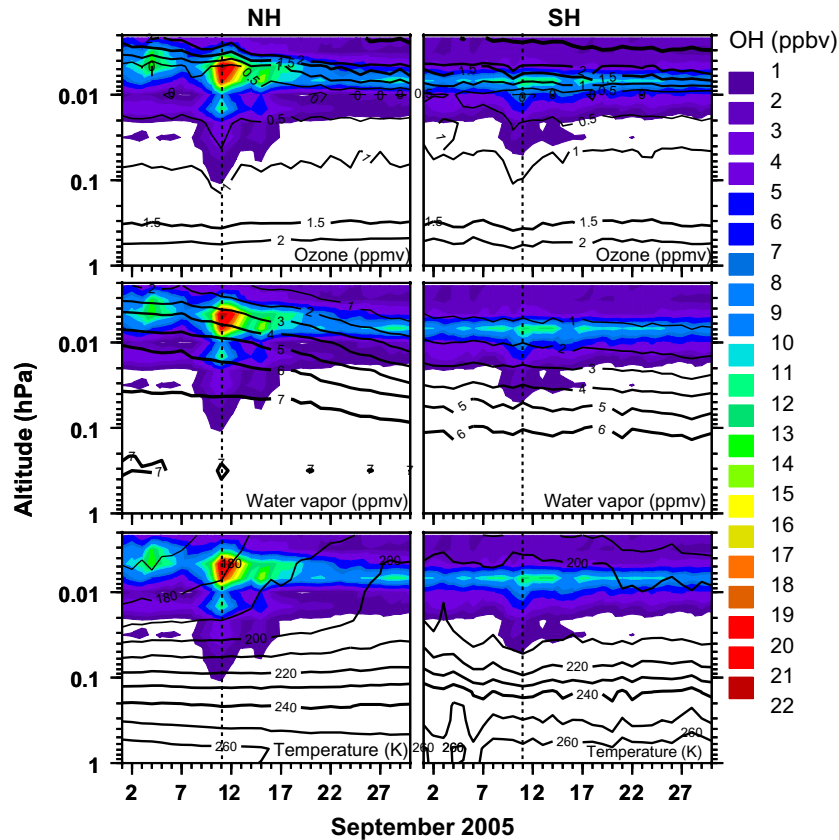


Fig. 6. From top to bottom panels: contours of O_3 and H_2O mixing ratio and Temperature in the geographic latitudinal range $60\text{--}70^\circ\text{N}$ (left side) and S (right side) during September 2005. Filled contours in all panels: OH mixing ratio in the range 1–22 ppbv (OH < 1 ppbv not shown).

graphic latitudinal band between 60° and 70° . The asymmetry of the SEP-induced OH formation reported for the upper mesosphere (filled contours in all panels) is similar to Fig. 3. In order to quantitatively assess these features, specific modeling studies will be needed and this is beyond the scope of this paper, nevertheless we want to resolve some points.

The production of SEP-induced HO_x occurs via water cluster ions. As is known, in the mesosphere their presence is possible up to about 80 km (but this limit is variable with season) and depends mainly on the water vapor and atomic oxygen concentration and on the temperature. Obviously the first element increases the hydration order of the ions whereas the elevated oxygen atom concentration prevents the formation of hydrated ions (see, for instance, Osepian et al., 2008). During September 2005 there is a different hemispheric distribution of ozone at the mesopause (at least until September 20, see Fig. 6, upper panels) and this suggests that similar hemispheric patterns exist also for the atomic oxygen [which is roughly confirmed by the MSIS)-Mass Spectrometer Incoherent Scatter) model; Hedin, 1991], allowing SEP-induced OH formation in the NH above the theoretical limit of 80 km. In addition, the greater abundance of H_2O in the NH compared with the SH (more than twice in upper mesosphere, see the middle panels of Fig. 6) should contribute to enhance the SEP-induced OH production in the NH. Finally, the tempera-

ture could also play an important role since low temperatures foster the hydration ion formation. Therefore, the large difference of hemispheric temperature (Fig. 6, lower panels) is a further element to take into account in explaining the asymmetric OH formation.

However, the OH asymmetry is also present at lower altitudes (~ 0.1 hPa) where the hemispheric gas mixing ratios and temperatures are quite similar. Therefore, other parameters not considered here (the electron/ion concentration; the different SZA of MLS OH, e.g., on September 11 the average SZA of the profiles is $\sim 104^\circ$ for the NH and $\sim 119^\circ$ for the SH) could also play a significant part in explaining the OH asymmetry.

6. Concluding remarks

It is known that the signature of SEP events such as those which occurred in 2005 (with flux roughly lower than 5000 pfu at energy >10 MeV) is easily detectable in winter inside the Polar Regions because of the low OH background (e.g., Damiani et al., 2008). This work illustrates the possibility of utilizing the mesospheric OH abundance recorded in Polar Regions under nighttime conditions as a SEP indicator. Almost all SEP events occurring in the descending phase of Solar Cycle no. 23 (since late 2004 when the MLS started to be operational) have been examined and the correlation of the OH abundance vs. the SEP

flux between 1 and 0.002 hPa is shown for the GOES proton channels at >1 , >5 , >10 , >30 , >50 MeV. The best correlation ($r \sim 0.90$ – 0.95) for the energies >5 MeV and >10 MeV is at ~ 65 – 70 km whereas, as expected, the most energetic channels have their best correlation at lower altitudes. The channel at >50 MeV showed minor correlation values because of the insufficient number of events with consistent flux at similar energies (note that the analyses are made on a daily basis). The OH variation produced by SEP events with a daily flux greater than ~ 300 pfu (for the channel >10 MeV) is discernible from the day to day variability. During nighttime, the maximum OH increase (i.e., more than 2000%) was recorded at ~ 65 km after the SEP event of January 17, 2005 corresponding to the highest flux among the examined SEP events. Therefore, the research performed shows that during SEP events it is reasonably possible to estimate the solar proton flux from values of OH (and vice versa) at altitudes of ~ 50 – 86 km and this could be useful in studying periods with gaps in the records of solar particles (e.g., failures on the satellite instruments).

These analyses have been possible because of the comparable OH mixing ratio present in both polar hemispheres during the winter. In September the nighttime OH background pre-SEP is slightly higher than the values for the winter months, therefore, the SEP events of September 2005 have been studied separately analyzing their features inside different geographic/geomagnetic regions. The SEP-induced impact on the hydroxyl radicals of both hemispheres are examined and, thanks to the season (September) and the LST of the MLS OH profiles, it was possible to compare the effects under similar atmospheric conditions. The investigated regions are between 60° and 70° geomagnetic latitude because this is a good compromise between the LST of the satellite passage and its position inside the geomagnetic polar cap where the effects are known to be more intense. Because of the same geomagnetic latitudes utilized, the hemispheric asymmetry of the SEP-induced OH formation (see Figs. 3 and 4) should not depend on the external solar forcing but on different in situ atmospheric conditions. So further analysis in regions between 60° and 70° geographic latitudes has been performed.

The main results of the analysis of the SEP events recorded during September 2005 are:

1. Well-discernible OH enhancement due to the September 2005 SEP events in both hemispheres during nighttime hours.
2. No discernible SEP impact on the OH profiles recorded at the same location during daytime hours.
3. On the OH nighttime profiles the SEP signature is relevant from about 1 hPa (~ 50 km) up to the enhanced OH layer centered at about 82 km (Pickett et al., 2006b). The middle mesospheric OH rise, recorded just after the SEPs, is higher by more than one order of magnitude compared with the values for quiet days.

4. Also, the upper mesospheric OH layer was subjected to significant increase due to SEP events, particularly in the NH, where this layer was at higher altitude. This seems to be in contrast with what is expected from past models because they predicted no effects above 80 km. The possible influence of energetic electrons (impacting similar altitudes) was not investigated. In September 2005 the high flux of low proton energy, able to impact the upper mesosphere, compared with other investigated events should be noted.
5. The OH increases occurring in NH in the early days of September (before the SEPs) above 0.01 hPa were attributed to the seasonal evolution of the hydroxyl radical. Further analyses considering the time evolution of water vapor and ozone showed that the seasonal trend of OH follows the evolution of O_3 and H_2O .
6. The hemispheric asymmetry of the SEP-induced OH formation is an element partly explained. Despite the greater illumination present in the NH compared with the SH and consequently higher ambient HO_x due to H_2O photolysis (which should attenuate the SEP-induced OH production), the impact of the energetic solar particles is more evident in the hydroxyl radical recorded in the NH. The different hemispheric O_x distribution, the higher H_2O mixing ratio and the colder temperatures present in the upper mesosphere of NH compared with the SH are elements potentially able to influence the SEP-induced OH because its formation occurs via water cluster ions. Nevertheless, hemispheric differences in the OH are present also in the middle mesosphere where the above-mentioned parameters are roughly similar in both hemispheres. Hence, specific modeling studies are necessary to explain this phenomenon in detail.

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