HO$_2$ Generation Above Sprite-Producing Thunderstorms Derived from Low-Noise SMILES Observation Spectra

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Abstract  No direct observational evidence of sprite-produced active radicals has been presented owing to the difficulty of observing a small event area in the nighttime mesosphere, whereas sprite chemical models have indicated that sprite discharge locally affects the atmospheric composition. We present the first observational evidence of a HO$_2$ production above sprite-producing thunderstorms from the coincidence of temporal-spatial observations of HO$_2$ spectra, sprite events, and thunderstorms by two space instruments, a submillimeter-wave limb spectrometer and ultraviolet/visible Imager and a ground-based very low frequency radiation lightning detection network. A total of three areas was identified with enhanced HO$_2$ levels of approximately 10$^{25}$ molecules. A chemical sprite model indicates an increase in HO$_2$ in the considered altitude region; however, the predicted production due to a single sprite event is smaller than the observed enhancement. Our observational results suggest that sprites potentially contribute about 1% of nighttime background HO$_2$ generation at altitudes of 75–80 km globally.

Plain Language Summary  HO$_2$ radical, a key atmospheric oxidant in the mechanisms that influence the atmospheric composition of our planet, has been predicted to be generated by lightning-induced upper atmospheric discharge, called sprites, from water vapor. However, there has been no observational evidence of sprite-producing active radicals due to the difficulty in specific observations that identify these radical productions by sprite, caused by factors such as a very small molecular abundance and narrow event area in the upper atmosphere. For the first time, we present observational evidence of a rise in HO$_2$ amount above sprite-producing thunderstorms by using a unique method that combined observations between HO$_2$ detection by a super-high-sensitive space spectrometer, sprite emission detection using a satellite imager and ground-based lightning detection using expand radio frequency receivers. The amount of HO$_2$ generated in the sprite event area can be approximated to be 40–190 times greater than the background values. Sprite chemical model simulations reproduced HO$_2$ enhancement by sprite events. These pieces of evidence suggest that sprites potentially contribute about 1% increase of background level of global HO$_2$ generation during nighttime at altitudes of 75–80 km, which can contribute to a change in the Earth’s atmospheric composition.

1. Introduction

Current technical advancements in areas such as measurement sensitivity and multiple-satellite observations have allowed us to examine single sudden events, although the statistical approach has been universally used to understand atmospheric phenomena generally. The understanding of sprite phenomena has been improved by the global coverage provided by satellite observations (e.g., Blanc et al., 2007, Chern et al., 2003, Garipov et al., 2013, Jehl et al., 2013, Sato et al., 2015, and others). The electric fields and electron energies of sprite streamers were inferred from high-quality morphological and spectral data of ultraviolet (UV)/visible imagers (e.g., Adachi et al., 2006; Kuo et al., 2005) and global distributions and occurrence frequency are estimated by long-term global observations (e.g., Chen et al., 2008). These observations were also
used to constrain the parameters of sprite model studies (Liu & Pasko, 2005; Liu et al., 2006) to reveal the mechanisms of the optical emission processes of sprites. Measuring atmospheric composition produced by single sudden sprite events has proven very difficult so far, although advanced heterodyne radio technologies have enabled the detection of minor species, such as active radicals in the upper atmosphere, by space submillimeter wave sounders (e.g., Kikuchi et al., 2010; Siegel, 2007; Waters et al., 2006; Yamada et al., 2018, and others).

Sprites are one of the most familiar types of the various upper atmospheric lightning phenomena, which are characterized as transient luminous events (Neubert et al., 2008; Pasko et al., 2012). Since the discovery of the sprite in 1990 (Franz et al., 1990), it has been suggested that transient luminous events, particularly sprites, generate active radicals and ions by ion-neutral chemistry models (e.g., Arnone et al., 2014, Evtushenko et al., 2013, Gordillo-Vázquez, 2008, Hiraki et al., 2008, Parra-Rojas et al., 2015, Winkler & Notholt, 2014, and others), although no conclusive observational evidence of chemical impact has been reported so far. Sprite discharges are induced by conventional air breakdown, caused by lightning-driven electric fields above thunderstorms (Hu et al., 2007; Pasko et al., 1995). They appear in an altitude range of 40–90 km, with a horizontal extent of several tens of kilometers. The duration of sprite luminescence ranges from a few to several tens of milliseconds (Barrington-Leigh et al., 2001). Estimates of the global rate of sprite occurrence range from one to three events per minute (Chen et al., 2008; Ignaccolo et al., 2006; Sato & Fukunishi, 2003); that is, there is approximately one sprite event for every thousand lightning flashes, as there are approximately 40–50 lightning flashes per second (Christian et al., 2003). It is recognized that once a sprite is produced, they appear repeatedly in the same region (Jing et al., 2008), although not every thunderstorm produces sprites.

Previously, two independent satellite instruments were used in an attempt to investigate sprite chemistry by observing lower mesospheric NO2 trends with tropospheric lightning activity (Arnone et al., 2008, 2009; Rodger et al., 2008); however, no conclusive observational evidence has been reported thus far. Arnone et al. (2008) showed a statistical difference between the amount of NO2 observed by the Michelson Interferometer for Passive Atmospheric Sounding mid-infrared emission spectrometer and thunderstorms activity within a field of view (FOV) of 300 × 30-km regions recorded by the Worldwide Lightning Location Network (WWLLN). Their statistical analysis and follow-up work (Arnone et al., 2009) indicates that NO2 enhancements of about 10% at 52-km height and tens of percent at 60-km height immediately after thunderstorm activity. Rodger et al. (2008) compared regional variations of column amounts of NO2 observed by the Global Ozone Monitoring by Occultation of Stars UV-visible spectrometer with respect to the lightning activity obtained from Optical Transient Detector observations (Christian et al., 2003) in the tropical and northern midlatitude regions (Rodger et al., 2008). The area between land and ocean regions was compared, because ~85–90% of lightning activity occurs above land (Christian et al., 2003). They reported no evidence of any correlation within the detection levels of the Global Ozone Monitoring by Occultation of Stars measurements.

In this study, we present the first investigation of HO2 generated by sprite events above sprite-producing thunderstorms from the coincidence of temporal-spatial observations of HO2 spectra, sprite events, and thunderstorms by two space instruments, submillimeter-wave limb spectrometer and UV/visible Imager and a ground-based very low frequency (VLF) radiation network used for lightning detection. Single limb scan HO2 spectra were observed using the Submillimeter-Wave Limb-Emission Sounder (SMILES) with full local time coverage. The SMILES instrument is suitable for single-scan HO2 detection because the HO2 single-scan detection limit is on the order of 1 ppbv in the nighttime mesosphere, which is an order of magnitude better than previous microwave/submillimeter limb instruments (Baron et al., 2011; Kasai et al., 2013; Kikuchi et al., 2010; Kreiling et al., 2013). A total of 127 emissions including sprites, halos, and gigantic jets was extracted from the Imager of Sprites and Upper Atmospheric Lightning (ISUAL) (Chern et al., 2003) observation data, and VLF radiation from lightning strokes was recorded by receiver sites of WWLLN (Lay et al., 2004) during the SMILES observation period from 12 October 2009 to 21 April 2010.

This paper is organized as follows. Section 2 describes observation data and analysis of the HO2 spectra, sprite image, and lightning distribution at their spatiotemporal coincidence. In section 3, we estimate the amount of HO2 enhancement in a sprite event area for the cases presented in section 2 and discuss a qualitative comparison with a sprite chemical model and a possible global impact of HO2 generation by sprite events. Finally, in section 4, we conclude our main results.
We excluded the following cases to find spatiotemporal coincidence between SMILES and ISUAL observations: (1) longitudinal and latitudinal differences between the SMILES observation area and the ISUAL sprite detection location are more than 3° and 1.5° because of the uncertainty in sprite location, SMILES...
Figure 1. (a–c) Observation geometry of the SMILES FOV (yellow and blue lines), the estimated locations of sprites (red dots) and their uncertainty (red boxes), and lightning occurrence distribution between SMILES and ISUAL observations obtained from WWLLN data (grayscale color map) for Events A, B, and C, respectively. Altitude ranges of the SMILES-FOV are 75–90, 77–90, and 80–90 km for Events A, B, and C, respectively. SMILES and ISUAL observed from left to right in these figures. (d–f) Images of sprites and their parent lightning emissions detected by the ISUAL for each event. (g–i) Distributions of the mean intensity in the range of $649701.5 \pm 0.8$ MHz for the background atmosphere (white columns). Blue columns represent the mean line intensity near sprite events. Yellow columns represent the mean line intensity three scans before and after.

A total of three cases is found in the coincidence conditions between ISUAL and SMILES observations, using the available data from 14 and 18 November 2009 and 9 March 2010. We refer to these as Events A, B, and C. The time and locations of each event are summarized in Table 1. The differences in latitude/longitude between the tangent points of the SMILES LOS and estimated locations of sprite were $0.8^\circ/2.7^\circ$, $-1.4^\circ/2.7^\circ$, and $0.1^\circ/1.7^\circ$, respectively, in each event. The shortest distances between the observation line of sight of the SMILES and the estimated areas of the sprite locations are <10, 110, and <10 km, respectively. The
Figure 2. (a–c) The SMILES observation spectra at anterior, coincidence, and posterior positions in each event. The first and third columns for each event show the observation spectra at anterior and posterior positions from event area, respectively, and the second column shows the SMILES observation spectra at the event area (red lines). These positions are able to be referred from the A1–A3, B1–B3, and C1–C3 scans as shown in Figure 1. The first, second, third rows for each event show the SMILES observation spectra for the HO2 transition at 649.701 GHz, HO2 transitions at 625.660 and 625.664 GHz, and O3 transition at 625.371 GHz, respectively. The blue vertical lines represent the frequency of the molecular transitions. The horizontal axes show frequency offset from the transition frequency. The tangent heights of SMILES observation spectra are 75, 77, and 80 km for each event.

The elapsed time of the SMILES observations since ISUAL detection are 2.4, 1.5, and 4.4 hr for each event. The LOS geometry of the SMILES limb observations, estimated locations of sprites, and lightning distribution between ISUAL and SMILES observations for each event are shown in Figures 1a–1c. ISUAL detected a carrot sprite, sprite halo with faint fine-scale structure, and a part of a sprite in each respective event, as shown in Figures 1d–1f. The widest sprite emissions are estimated to be 17, 30, and 8 km at altitude ranges of 55–75, 75–80, and 50–70 km, respectively, shown in Figures S1 in the supporting information. In Event A, a part of the line of sight of the SMILES single-shot scan grazed the air mass of the sprite occurred, whereas there is a possibility that additional sprite events not detected by ISUAL occurred around the area in which WWLLN detected lightning. WWLLN recorded 382 lightning strokes at 159.5–160.0°W and 21–21.5°N located beneath the SMILES FOV during the time between ISUAL and SMILES observations in Event A. In Event B, it is unlikely that the SMILES LOS grazed the estimated area of the sprite halo, although the sprite halo has a wide width and large position uncertainty, and there are active thunderstorms likely producing sprites around the SMILES line of sight. In Event C, a part of the SMILES line of sight covered the sprite air mass.

As shown in Figure 2, local spectral enhancements of HO2 are detected at the tangent altitudes of 75, 77, and 80 km in Events A, B, and C, respectively. The line intensities and root-mean-square noise of HO2 spectra at each event are 1.3 ± 0.2, 2.1 ± 0.2, and 2.0 ± 0.2 K for each event, while adjacent to the the air mass, anterior/posterior positions of SMILES sprite observation, were 0.37/0.21, 0.87/0.54, and 0.76/0.73, respectively. The line intensity is calculated as the mean intensity in the range of 649701.5 ± 0.8 MHz which covers the Doppler width from the center line of HO2 at 300 K. We extract background atmospheric observations for each coincident observation satisfying the following conditions: within 45 days and 5° of the solar zenith angle and latitude from each respective event, the mean line intensities of HO2 spectra in these background profiles for each event are 0.55, 0.73, and 0.53 K with one standard deviation of 0.34, 0.46, and 0.41 K, with a total of 104, 157, and 322 background observations, respectively. Figures 1g–1i show the distributions of the line intensities for the background atmospheric observations with line intensities for the sprite events and three scans before and after the SMILES observation at the sprite events. Local spectral enhancements of other transition frequency of HO2 (625.66 GHz) are also detected by another side band frequency of SMILES at the same place and time. Slight decreases in O3 spectral intensities at each event are observed by SMILES, although it is difficult to separate these perturbations from spectral noises and latitude and altitude variations of O3 abundance. Further observations carefully dealing with the background latitudinal and altitudinal variations are needed because of detecting the perturbation in small event from the large amount of background O3.
These local enhancements were most likely caused by sprite events, rather than from other events or variations. Although solar proton and energetic electron precipitation events are known to enhance the nighttime abundance of HOx (H, OH, and HO2) in the high-latitude region owing to the reaction between electrons and hydrated cluster ions (Jackman et al., 2011, 2014; Solomon et al., 1981), their activity was quite low at the low-latitude region and during the SMILES observations that took place from 12 October 2009 to 21 April 2010 (Andersson et al., 2014). The diurnal variation in mesospheric HOx is mostly controlled by the photolysis of H2O by sunlight (Kreyling et al., 2013), which only depends on the local time and lifetime of HOx species, making it incapable of producing the local enhancement. Horizontal transportation by zonal winds may impact the estimated HOx productions as demonstrated in Arnone et al. (2014) for nitrogen oxides. The zonal winds are between 0 and 40 m/s at November (Events A and B) and between −20 and 0 m/s on March (Event C) near 75- to 80-km altitudes (Baron et al., 2013). We estimated that the distances the air masses are moved away from the LOS are smaller than 300, 200, 300 km for Events A, B, and C, respectively. These ranges are of the same order of magnitude than the area of the thunderstorms. Hence, the transportation only induces a small underestimation of HO2 abundances estimated in this analysis. Future studies will be performed with global chemical model to account for dynamical effects.

3. Estimation of HO2 Amount Produced by Sprite Events

HO2 number densities in the sprite event area are estimated with a least squares fitting by using the Atmospheric Radiative Transfer Simulator (Buehler et al., 2018; Eriksson et al., 2011). The simulation assumes that there is an HO2 enhanced area where all the additional HO2 is contained. This box is called the enhanced HO2 box. The altitudes of the enhanced HO2 boxes for estimating the number density of HO2 are 75, 77, and 80 km, respectively. Line parameters include line position, intensity, pressure broadening, energy levels, and partition functions of HO2 were taken from Perrin et al. (2005). The atmospheric profiles include the volume mixing ratio of HO2, temperature, and pressure. Vertical atmospheric profiles are obtained from the averaged values of the SMILES Level-2 research (L2r) product data Version 3.0.0, an updated version of Version 2.1.5 (Baron et al., 2011; Kasai et al., 2013; Kreyling et al., 2013), at adjacent observation scans. The calculated spectra are convoluted with frequency response functions of acousto-optic spectrometers (Mizobuchi et al., 2012).

The enhanced air column abundances from an initial value, ΔobsHO2, are also calculated as below:

$$\Delta_{\text{obs}} \text{HO}_2 = V_{\text{box}} (l) \left[ n_{\text{fit}} (l) - n_{\text{back}} \right],$$

where $l$, $V_{\text{box}}$, $n_{\text{fit}}$, and $n_{\text{back}}$ are the length of the enhanced HO2 box, the column volume of the enhanced HO2 box, the best fit number density of HO2 in the enhanced HO2 box, and the averaged values of the SMILES L2r product data at adjacent observation scans, respectively. We assume that the SMILES FOV has an elliptical shape with a diameter of 3 km vertically and 6 km horizontally.

The best fit densities and one standard deviation errors of the fitting densities of HO2 in the enhanced HO2 boxes are $38 \pm 10 \times 10^6$, $38 \pm 5 \times 10^6$, and $160 \pm 20 \times 10^6$ molecules/cm$^3$ for averaged densities of the adjacent air mass of $0.56 \times 10^6$, $1.0 \times 10^6$, and $0.85 \times 10^6$ molecules/cm$^3$, respectively, in the case of the size of the enhanced HO2 box, $l$, is same as the horizontal width of sprite emission ISUAL detected. The number densities of HO2 in the enhanced HO2 boxes are $68 \pm 18$, $38 \pm 5$, and $190 \pm 24$ times larger than the background values. The root-mean-square of the residual between the simulated and observed spectra is 0.23, 0.20, and 0.21 K, while the root-mean-square noise of the observed spectrum is 0.21, 0.21, and 0.22 K, respectively, as shown in Figures S2a–S2c in the supporting information.

The $\Delta_{\text{obs}} \text{HO}_2$ for each event is nearly constant relative to the length of the sprite box as shown in Figure S3. They were $8.9 \pm 2.5 \times 10^2$, $16 \pm 2.0 \times$, and $17 \pm 2.0 \times 10^{24}$ molecules for each event. Table 1 summarizes the $\Delta_{\text{obs}} \text{HO}_2$ with elapsed times from the occurrence of the ISUAL sprite to the SMILES HO2 observation, locations of sprite, local time of HO2 observations, tangent heights of SMILES LOS, and horizontal widths of sprite emissions at each tangent height of SMILES LOS for each event. The estimated amounts of HO2 enhancement in the sprite event area are much larger than those of the latitudinal variation as shown in Figures S2d–S2f in the supporting information.

We also investigated the sprite-induced chemical disturbances in the mesosphere on the timescale of a few hours after a sprite event by using a one-dimensional atmospheric chemistry and transport model (see...
supporting information Text S1 for the model description and simulation results). The model simulation showed that once HO$_2$ is formed, the enhanced concentrations remain basically constant for a few hours, although the enhancements are much smaller than the enhancements observed by SMILES as shown in Figure S4 in the supporting information. Multiple sprite events are likely necessary to explain the observed HO$_2$ enhancement. The long-lasting HO$_2$ increase in the model may allow an accumulation of HO$_2$ enhancements. There is a short-term decrease in ozone at 75 km with recovery after 1,500 s. The model indicates the HO$_2$ enhancements are caused by a conversion of water molecules into HO$_2$ through reactions and recombination of proton hydrates, which is similar to the situation in solar proton events. There are also possibilities of different electric field parameters than used in our model and missing chemical processes in the model. The preliminary model simulation only accounts for streamer tip electric fields. The effect of afterglow fields (Gordillo-Vázquez & Luque, 2010) has not been considered yet. They will be addressed in future simulations. Further model studies of a time range of several hours after sprite initiation should focus on factors related to the above reactions and take into account horizontal transport processes.

Furthermore, we estimated a global production of HO$_2$ molecules by sprite events during nighttime at altitudes of 75–80 km from the observational results. The total amount of HO$_2$ produced by the nighttime sprite events can be approximately $10^{28}$ molecules if the occurrence frequency were assumed to be one to three events per minute, while the total amount of background HO$_2$ can be $10^{26}$ molecules if background concentration of HO$_2$ were assumed to be $0.9 \times 10^6$ molecules/cm$^3$ at the altitude range. Thus, the production of $10^{26}$ molecules of HO$_2$ by a sprite event area is potentially accounts for 1% of the global amount of nighttime HO$_2$ at 75–80 km. The background concentration of HO$_2$ is derived from the average of SMILES L2r product data in nighttime at 75–80 km during the SMILES observation period. This estimation simplified the sprite and atmospheric conditions such as the strength of electric field in sprite discharges and the background atmospheric composition. To be clear, the retrieved production is not necessarily showing the production of HO$_2$ from single sprites but rather the accumulation of production by the entire sprite-producing thunderstorm. As Arnone et al. (2014) demonstrated, it is important to investigate the climate chemistry sensitivity to sprite HO$_2$ production without the above simplification. The impact of sprite chemistry on the Earth’s atmosphere may increase in the future because the source of HO$_2$ production and trigger of sprite occurrence are mesospheric water vapor and tropospheric lightning, which are increased by greenhouse gas emissions and global warming, respectively. Global warming is predicted to increase tropospheric lightning activity (5–10% for every 1 °C of warming) (Krause et al., 2014; Michalon et al., 1999; Price & Rind, 1994; Romps et al., 2014; Trapp et al., 2007) although the prediction has a large uncertainty in the lightning parameterization (Finney et al., 2018). Long-term trends of increase in mesospheric carbon dioxide (CO$_2$) and water vapor (H$_2$O) have been reported owing to recent improvement in the accuracy of global measurements (Chandra et al., 1997; Nath et al., 2018; Nedoluha et al., 2017; Qian et al., 2017). These trends, approximately +5.5% of CO$_2$ and <10% of H$_2$O per decade, are related to anthropogenic CO$_2$ and methane (CH$_4$) emissions, respectively, and affect radiative forcing and mesospheric composition. Further observations and model studies regarding sprite and atmospheric discharge chemistry are needed to assess their potential impact on the atmosphere of the Earth.

4. Conclusions

In this study, we detected for the first time the local enhancements of HO$_2$ above sprite-producing thunderstorms at an altitude range of 75–80 km by using SMILES, ISUAL, and WWLLN data. All three areas showed that the enhancement of HO$_2$ amounts to be approximately $10^{25}$ molecules. The chemical sprite model indicates an increase in HO$_2$ in the considered altitude region for several hours after a sprite discharge; however, the predicted production of HO$_2$ due to a single sprite event is smaller than the observed enhancement. The production of $10^{25}$ molecules of HO$_2$ at a sprite event area potentially accounts for approximately 1% of the global amount of nighttime HO$_2$ at altitudes of 75–80 km according to the simplified estimation. The investigation of the impact of sprite discharge events on global atmospheric composition becomes more important in the scenario of increase in both lightning activity and H$_2$O abundance in the upper atmosphere. Further observations and model studies regarding sprite and atmospheric discharge chemistry are needed to assess their potential impact on the Earth’s atmosphere.
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