Significance of transient luminous events to neutral chemistry: Experimental measurements

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[1] For many years it has been suggested that transient luminous events (TLE) occurring over thunderstorms may produce significant modifications to neutral atmospheric chemistry. Some have speculated that large ionisation increases from red sprites, one type of TLE, could result in enhancements of odd nitrogen. In this study we make use of nighttime NO_2 observations by the GOMOS instrument to test whether TLE are producing significant NO_x enhancements in the middle atmosphere on a regional scale. Comparing regional variations of NO_2 with 2–3 order of magnitude variations in lightning activity, we show that there is no significant impact of red sprites, giant jets or blue jets upon NO_x levels in the stratosphere and mesosphere (20–70 km), within the detection levels of the instrument. While individual TLE may cause a local variation in NO_x, these do not appear to be significant on regional scales (or beyond). **Citation:** Rodger, C. J., A. Seppälä, and M. A. Clilverd (2008), Significance of transient luminous events to neutral chemistry: Experimental measurements, *Geophys. Res. Lett.*, *35*, L07803, doi:10.1029/2008GL033221.

1. Introduction

[2] Red sprites were first recognized by the scientific community about 20 years ago. The first image focused attention on the atmospheric region above thunderstorms, and lead to the identification of a whole host of new phenomena (see the review by *Rodger* [1999]), now collectively termed "Transient Luminous Events" (TLE). It quickly became obvious that red sprites were not a cold phenomena, and involved orders of magnitude increases in middle atmosphere ionisation. Scattering of subionospheric transmissions were first related to sprite occurrence [*Inan et al.*, 1995] and subsequently shown to be due to a "hot electron" discharge process [e.g., *Dowden and Rodger*, 1997], leaving an ionisation increase which persisted for as long as tens to hundreds of seconds [*Nunn and Rodger*, 1999], depending on altitude. A review on the use of subionospheric long-range probing to investigate the impact of lightning upon the lower ionosphere has been presented by *Rodger* [2003].

[3] It has long been known that lightning discharges are a significant producer of odd nitrogen in the troposphere (see the discussion by *Rakov and Uman* [2003]). Some authors have speculated that red sprites might have a significant impact upon the chemistry of the stratosphere and mesosphere, by virtue of their location. For example, it was estimated that red sprites would lead to a two order of magnitude increase in NO above thunderstorms [*Lyons and Armstrong*, 1997]. *Mishin* [1997] reported that a single blue jet event could induce a 10% local perturbation in NO content at 30 km, although this modelling was limited by the significant unknowns associated with the blue jets, which persist to this day. It has been common for researchers in this field to speculate that TLE may significantly impact the middle atmosphere, playing an important role on the regional scale, or indeed on larger scales. One of the specific enquiries of the European Coupling of Atmospheric Layers training network [*Neubert et al.*, 2005] focused on chemical changes to the stratosphere and mesosphere, as part of the wider question "are these high-altitude discharges only pretty and beautiful like rainbows, or do they significantly impact the atmosphere"?

[4] The link between ionisation increases and significant neutral atmospheric variations is a natural one. Ionisation increases results in enhancement of odd nitrogen (NO_x = $NO + NO_2$) and odd hydrogen (HO_x), which play a key role in the ozone balance of the middle atmosphere [e.g., *Brasseur and Solomon*, 2005, pp. 401–404]. Changes in NO_x and O_3 consistent with solar proton-particle precipitation have been observed [*Verronen et al.*, 2005], with long term decreases in upper stratospheric ozone by as much as ~30% [*Seppälä et al.*, 2007].

[5] There are, however, important differences between the particle precipitation processes which have been shown to change middle atmospheric chemistry and TLE. In particular, solar proton events are long-lived compared to TLEs, lasting hours to a few days, while each TLE is roughly tens of milliseconds in duration, with fine-structure on substantially shorter timescales. Short-lived pulses of particle precipitation due to lightning interacting with radiation belt electrons have been shown to be insignificant due to the short timescale of the bursts (0.2 s), despite over a thousand such bursts having occurred over ~8 hours [*Rodger et al.*, 2007]. Most recently, modelling of the impact of a single red sprite upon the middle atmosphere has concluded that each sprite might produce a small modification to the middle atmosphere [*Enell et al.*, 2008], with NO_x enhancements of at most one order of magnitude in sprite streamers. That study considered the possibility that an intense sprite-producing thunderstorm (or mesoscale convection system, MCS) producing multiple TLE in a short time scale might create a NO_x build-up, leading to a significant modification on a regional scale. While the average global red sprite rate is thought to be ~3 per minute [*Ignaccolo et al.*, 2006], hundreds of sprites have been observed over intense storms within a few hours

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[*Lyons*, 2006]. *Enell et al.* [2008] suggest that such intense sprite-producing MCS could lead to local NO_x enhancements at 70 km altitude of 50–500%, filling the volume above the system. Some possible evidence for this comes from NO_2 observations in 500 × 30 km regions from the MIPAS instrument, correlated with lightning observations from the experimental World Wide Lightning Location Network (WWLLN) [*Arnone et al.*, 2008], which reported a lightning-related NO_2 enhancement of ~10% at 52 km, albeit at levels fairly close to the instrument's measurement uncertainties.

[6] In this brief report we examine nighttime NO_2 observations from an Earth observing spacecraft to test the often posed question as to whether TLE may play a significant role in modifying the neutral atmosphere on large scales (regional and global). By considering different altitude ranges we examine whether different TLE (e.g., red sprites, giant jets and blue jets) may play different roles.

2. Variation in Lightning Activity

[7] The average diurnal variation of lightning over land peaks at about ~16 LT [*Williams et al.*, 2000], in contrast to the near-constant levels of oceanic lightning, albeit at much smaller rates on a global scale. However, high peak-current lightning, which is likely be a better proxy for TLE occurrence rates, have been experimentally observed to peak later in the day. Observations from the experimental World Wide Lightning Location Network (WWLLN), which is primarily sensitive to lightning discharges with high peak currents, have shown that ~50% of the WWLLN-lightning over land occurred in the nighttime sector, ranging from ~40% in Asia and Europe, ~55% in Africa and ~60% in the America's [*Lay et al.*, 2007]. Thus a reasonable fraction of the total lightning activity over land will occur during the nighttime, producing a larger possibility of significant atmospheric impact, which could be measured by the GOMOS instrument (see Section 3).

[8] In order to search for a possible TLE-produced modification to NO_x in the middle atmosphere, we exploit the very strong regional and seasonal variations in lightning levels. Satellite observations now allow some confidence in the average geographical distribution of total lightning activity and global flash rate. Five years of Optical Transient Detector (OTD) observations have been combined to produce lightning density distributions averaged over the year [*Christian et al.*, 2003], which show strong seasonal variations driven by the different amounts of land in the northern and southern hemispheres. Globally, ~85–90% of lightning activity occurs above land [*Christian et al.*, 2003, Figure 7], an effect which is amplified when contrasting regions above land with those above oceans at the same longitude. Figure 1 shows global geographical maps of total lightning activity (in units of flashes km^{-2} yr^{-1}) taken from the OTD Low Resolution Annual Climatology dataset. Overlaid on this plot are white boxes showing the selected land and ocean regions which will be examined for possible atmospheric influence of TLE influence. The boxes are marked "L" or "S" depending on whether they are land and ocean regions. Each of the boxes is 15° wide in longitude and 20° wide in latitude. In contrast, mesoscale convective systems are circular weather systems with horizontal scales of 250–2500 km [*Goodman and MacGorman*, 1986], i.e. roughly 2.2–22° of longitude at the equator. The land regions were selected to include high-lightning density areas in the major "chimney" regions of the Earth. We select regions where TLE have been observed (Mid-western USA), positive polarity high charge moment lightning rates occur (Central Africa and SE Asia [*Hobara et al.*, 2006]). Finally we include a "control" region where nighttime lightning rates are high but appear to be dominated by large negative polarity lightning [*Lyons et al.*, 1998] and thus low positive polarity-associated TLE rates (e.g., red sprites) are therefore expected (Florida). Using 2006 WWLLN observations, the local-time peak of large current lightning activity for the selected land regions are: Mid-West 19 LT, Florida 19 LT, Africa 18 LT and SE Asia 18 LT.

[9] The ODT lightning density data shows the very strong differences between the intense thunderstorm regions over land and oceanic regions at the same longitudes. Figure 2 (left) indicates how the seasonally varying flash rate changes in the 8 regions selected in Figure 1. The solid curves in this panel show the selected land regions, while the dotted curves are those for the oceanic regions. As we have selected high-lightning zones, the peak flash rates in each zone of ~6 (Africa), ~4 (Florida), ~3.4 (Mid-west) and ~2 (SE Asia) flashes per second are significant when contrasted with the mean total global flash rate of 45 ± 5 flashes s^{-1}.

[10] Figure 2 (right) shows the ratio between the land and oceanic regions, for the selected regions. In all cases there is two-three orders of magnitude peak difference between the selected land and oceanic regions. The large differences between the land and oceanic regions selected, along with the strong seasonal variations seen for each chimney region provides a large "signal" to search for in NO_x measurements.

3. GOMOS Measurements of NO_2

[11] We use GOMOS [*Hauchecorne et al.*, 2005] NO_2 measurements at altitudes from 20–70 km, to examine the significance of TLE-produced nighttime NO_x. In the stratosphere the NO_x gases are in photochemical balance during the daytime. After sunset NO is quickly converted into NO_2 in reaction with O_3, and thus the nighttime NO_2 measurements provided by GOMOS are a good representation of stratospheric NO_x. In this study we have used GOMOS observations (GOPR version 6.0c, T_{star} ≥6000 K) from the selected geographic regions during July 2002–October 2006. We require the solar zenith angle at the tangent point to be >107°, and >90° at the satellite point, to avoid stray light conditions. The quality of GOMOS measurement selection is discussed in more detail by *Seppälä et al.* [2007].

4. Testing TLE Impacts

[12] Figure 3 shows the seasonal variation of GOMOS measured nighttime NO_2 above our selected land and oceanic regions. The upper section shows the NO_2 partial column vertically integrated over altitudes 30–50 km, while the lower section shows this for 50–70 km. The monthly QBO phase at 30 mb is shown by crosses (westerly) or circles (easterly) in the SE Asian panel in the lower altitude

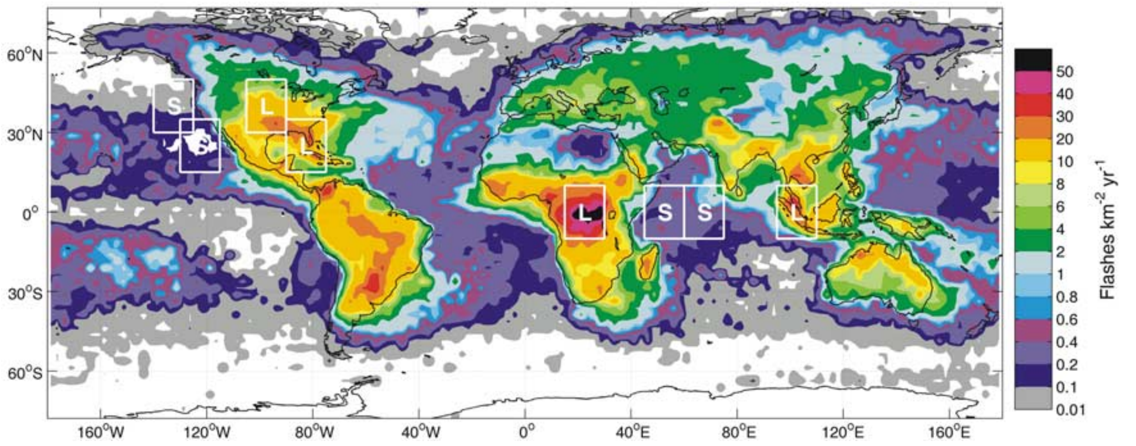

Figure 1. The annualized geographical distribution of total lightning activity determined from 5 years of OTD data in units of flashes km^{-2} yr^{-1}, after *Christian et al.* [2003]. Superimposed are white boxes indicating the land and oceanic regions, marked with a white "L" or "S", respectively, selected for examination in this study.

range, taken from the NOAA QBO U30 Index. The lower of these two altitude ranges includes that affected by blue jets (~20–45 km), while the upper range covers part of the region in which red sprites have been observed to occur (~50–90 km). Giant jets span (~20–75 km) across both these altitude ranges. Figure 3 includes bars showing the measurement uncertainties at the 1 standard deviation level. In the lower altitude range the land and oceanic NO_2 column values are generally very similar, with observations taken at the same time in the different regions falling within the uncertainty levels. Both the land and oceanic 30–50 km NO_2 column values for the American regions display the annual variation of mid-latitude NO_2 column values previously reported for 20–50 km columns [*Hauchecorne et al.*, 2005], with maximum values during the northern hemisphere summer. While we expect high TLE activity in the Mid-West, and comparatively low activity in Florida, these panels show very similar NO_2 variation. In equatorial regions the 20–50 km NO_2 column values were reported to have little annual variation, which may explain why no clear yearly cycles are seen in Figure 3 in the African and SE Asian panels. The higher altitude 50–70 km NO_2 column values in the right panel rarely show any significant variation between the land and oceanic regions once the measurement uncertainties are taken into account.

5. Discussion and Summary

[13] The NO_2 column values shown in Figure 3 do not show evidence for large NO_2 increases above the high-lightning land regions, or seasonal variations which match that seen in the regional lightning activity. The possible effect of the quasi-biennial oscillation (QBO) on the NO_2 measurements below 50 km [*Randel and Wu*, 1996] is taken into account by comparing land and sea regions covering the same latitude range. We therefore conclude that there is no significant perturbation in the nighttime NO_x levels across the altitude range from 30–70 km, inside which blue jets, giant jets and red sprites occur. TLE occurring across this altitude range do not appear to be significant to the NO_x chemistry of the upper stratosphere and mesosphere on a regional scale. *Mishin* [1997] indicates that blue jet-induced NO perturbations are larger at lower altitudes, and hence we have also examined the GOMOS-observed NO_2 partial column from 20–30 km altitude. Again, no significant difference is found between land or ocean regions, or

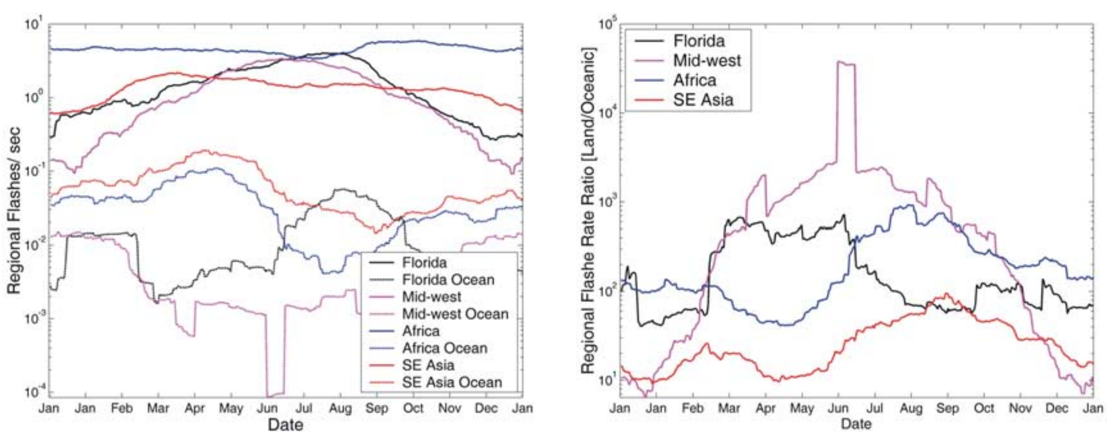

Figure 2. Seasonal variation in the OTD measured flash rate in the selected regions. (left) The flash rate in the land (solid) and oceanic (dotted) regions and (right) the ratio of the land to oceanic flash rates.

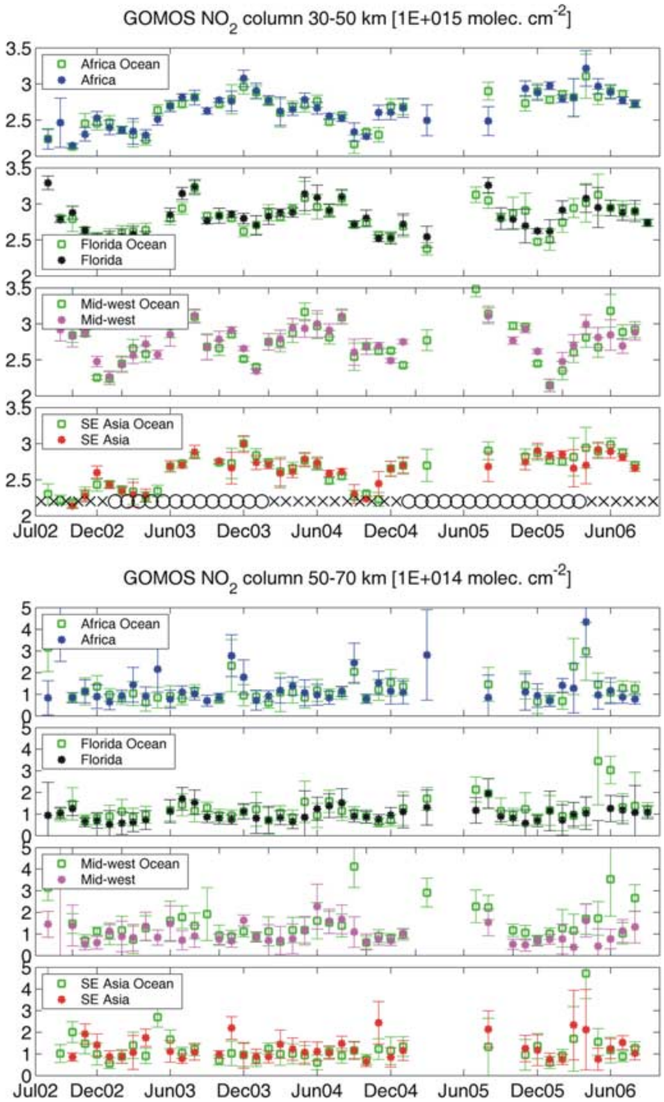

Figure 3. Seasonal variation of GOMOS measurements of nighttime NO_2 for the land and oceanic (squares) selected regions. (top) The NO_2 30–50 km column density, with the monthly QBO phase at 30 mb is shown by crosses (westerly) or circles (easterly), and (bottom) this for 50–70 km. Measurement uncertainties are shown at the 1 standard deviation level.

correlating with the average seasonal lightning variations (not shown).

[14] The photochemical lifetime of NO_x in the upper stratosphere varies from a few days to about a month [*Brasseur and Solomon*, 2005]. It is possible that zonal winds could rapidly distribute this NO_x, such that the differences in the NO_x-levels above the land and oceanic lightning regions are considerably less well defined than the lightning densities themselves. Given that zonal winds can be a few tens of meters per second at the considered regions, strong TLE-produced NO_x enhancements occurring over high lightning regions could be rapidly zonally averaged, such that one might expect little significant difference between NO_x observations made above our selected land and oceanic regions. However, the upper stratospheric lifetime of NO_x is still small when compared with the seasonal variations in the lightning sources, which would modulate the NO_x-levels even if the production which occurs above land at a given latitudes was rapidly "smeared" across all longitudes. The GOMOS measurements do not show evidence for a coherent lightning-driven seasonal pattern in NO_x.

[15] For some time it has been argued that TLEs might have a significant impact upon on the neutral chemistry of the atmosphere. One likely route would be through the production of NO_x due to TLE-ionisation. In this study we have made use of the large spatial and seasonal variations in lightning flash rates to search for a regional-scale TLE impact, assuming that seasonal variations in lighting occurrence will be an effective proxy for TLE occurrence. Even when there is a factor of ~1000 between average lightning flash rates, the GOMOS-measured NO_2 column values show no suggestion of significant perturbations in the nighttime measurements in the altitude range spanning 20–70 km, within the detection levels of the instrument. While TLE may produce local changes in NO_x around the event itself, there is little evidence that TLE are significant to the neutral chemistry of the middle atmosphere, unless

there is weak coupling between lighting occurrence, positive polarity high charge moment flashes, and overall TLE occurrence.

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