Comment on "NO$_x$ production in laboratory discharges simulating blue jets and red sprites" by H. Peterson et al.

S. Nijdam,[1] E. M. van Veldhuizen,[1] and U. Ebert[1,2]

Citation: Nijdam, S., E. M. van Veldhuizen, and U. Ebert (2010), Comment on "NO$_x$ production in laboratory discharges simulating blue jets and red sprites" by H. Peterson et al., *J. Geophys. Res.*, *115*, A12305, doi:10.1029/2010JA015861.

1. Introduction

[1] Knowing the importance of lightning for global atmospheric NO$_x$ production, it is natural to ask how transient luminous events (TLEs) like sprites and jets influence the NO$_x$ content of higher atmospheric layers. This question has been addressed in the past in particular for sprites as they are much more frequent than jets [*Chen et al.*, 2008]. Methods included calculation based estimates on the one hand [*Sentman et al.*, 2008; *Gordillo-Vazquez*, 2008; *Neubert et al.*, 2008; *Enell et al.*, 2008], and observation based estimates on the other hand [*Arnone et al.*, 2008]. *Peterson et al.* [2009] try to address the question through laboratory experiments.

[2] While such a study is certainly a desirable complement to calculations and observations, it will never perfectly model atmospheric conditions and therefore requires care when extrapolating to TLEs. Nevertheless, we argue that *Peterson et al.* [2009] have made several basic conceptual errors, and their study therefore cannot be used for this purpose. The conceptual errors lie (1) in the assumption that it would be sufficient to characterize a discharge only by pressure and current, not distinguishing basically different discharge types, (2) in a wrong application of the similarity laws relating transient cold discharges at different pressures, (3) in a confusion of individual streamer channels within a sprite with carrot sprites as a whole, and (4) in an overestimation of the (local) duration of sprite activity by a few orders of magnitude. Furthermore, we have found a grave calculation error regarding scaling, which has a direct influence on important results reported in this paper.

2. Similarities Between Experiments and Real TLEs

[3] One can use Townsend scaling to compare laboratory and real TLE discharges when the two discharges are of a similar kind and both are dominated by two-body collisions (as is the case for the propagating heads of streamers and sprites). As was shown by *Briels et al.* [2008], it is possible to define properties like the reduced diameter ($n \times d$, with n the air density and d the streamer diameter) that are independent of pressure for a large pressure range.

[4] Further extension of this scaling predicts how current, current density, propagation velocity and more parameters of streamer-like discharges scale with air density. All similarity laws are based on the mean free path length of an electron between collisions with the neutral gas molecules. Therefore, such scaling is only valid when the discharge is dominated by such two-body collisions. This is the case for the active tips of streamers and sprites, but not for sparks, leaders, jets and lightning return strokes.

[5] According to the similarity laws, if the same voltage is applied, all length and time scales scale with the inverse neutral gas density n. Currents are independent of n and therefore current density scales with n^2. Velocities are independent of n. Townsend scaling for comparing streamer experiments with sprites has been discussed, e.g., by *Pasko et al.* [1998], *Rocco et al.* [2002], *Liu and Pasko* [2004], *Ebert et al.* [2006], *Luque et al.* [2007], and *Briels et al.* [2008]. A recent review of the applications and limits of Townsend scaling is given by *Ebert et al.* [2010].

[6] In contrast, *Peterson et al.* [2009] propose that their laboratory experiments are similar to real TLEs because they have the same color, pressure, current density and emission duration. In our view this is incorrect for the following reasons:

[7] 1. It is well known that lightning at (nearly) atmospheric pressure develops in several stages: coronas of cold streamer channels pave the way of hot leader channels, and some leader channels later convert into a very hot return stroke channel. All phases are clearly distinct in their temperature, spectra and chemistry though they all evolve at the same pressure. Therefore, it is clearly insufficient to characterize a discharge only by pressure as *Peterson et al.* [2009] do. Up to now it is frequently assumed that the return stroke channel would be the main source of NO$_x$ production in lightning, but that (implicit) hypothesis can be questioned.

[8] While it is commonly accepted that sprite discharges are a form of streamer or corona discharges, *Peterson et al.* [2009] explicitly mention that they want to avoid the occurrence of coronal discharges in their experiments (their paragraph [19]). They claim that such coronal discharges produce ozone and thus a higher proportion of NO$_2$. But the laboratory equivalent of sprites are just corona (or streamer) discharges.

[9] *Peterson et al.* [2009] stress that their experimental discharges are similar to real TLEs, but this is only similarity in a colloquial sense and does not involve the similarity laws as discussed above. They do not use scaling laws

[1]Department of Applied Physics, Eindhoven University of Technology, Eindhoven, Netherlands.
[2]Centrum Wiskunde & Informatica, Amsterdam, Netherlands.

to compare densities, current densities and dimensions of their discharges with TLEs. The alternative for using scaling laws is to exactly replicate (a part of) a real TLE discharge. In this case the experimental gas density should be equal to that of the TLE discharge. In Peterson et al.'s sprite simulations this is not the case.

[10] From a plasma technological point of view, it should be noted that streamer coronas are used for more than a century to generate ozone for various disinfection purposes [*van Veldhuizen*, 2000] while glow discharges or sparks were clearly discarded for this purpose. The purely empirical finding that streamer coronas produce ozone in a very efficient manner is recently being supported by systematic analysis and subsequent technological improvement. Through voltage pulses lasting only several tens of nanoseconds, *van Heesch et al.* [2008] managed to convert more than 50% of the electrical input energy into O* radicals. The key to success was to power the discharge only during the initial streamer phase while avoiding any secondary streamers or gas heating. The underlying reason for this efficiency is the electron energy distribution in the streamer head with its high transient field; these electrons are very far from any equilibrium as the reduced electric field E/n is locally very high [*Morrow*, 1985; *Dhali and Williams*, 1987]. While (hot and slow) spark and lightning-like discharges can produce about 5×10^{16} molecules joule^{-1} of NO [*Levine et al.*, 1981], (cold and fast) corona or streamer discharges like the ones produced by *Peterson et al.* [2009] are often more efficient and can produce 7×10^{17} molecules joule^{-1} or more.

[11] 2. *Peterson et al.* [2009] estimate the color of the discharges from frames of a video of the discharge. In the images given in Figure 7 of Peterson et al., it is clear that the video is saturated in many cases. In these cases, it is impossible to judge the color from the video. Furthermore, just the fact that the color is similar is no proof that the TLE discharge is similar.

[12] 3. *Peterson et al.* [2009] claim that their current density is similar to sprite current density, but do not proof this claim. There is not much literature on sprite current density, but according to *Cummer et al.* [1998] the combined current of all channels within a carrot sprite is 1.6–3.3 kA. The sprite-like discharges by Peterson et al. have currents of order 10 A and a cross section of order 10^{-3} m^2. This gives a current density of 10 kA/m^2. When we combine this current density with the sprite current reported by Cummer et al., this would give a sprite diameter of less than 1 m. This is lower than estimates of the diameter of one streamer channel in a sprite discharge (which is of order 10–100 m). A real carrot sprite consists of hundreds to ten thousands of these streamers. Therefore, we can conclude that the current density in a real sprite is probably at least 3 orders of magnitude lower than in the laboratory discharges by Peterson et al. Note that even if the current densities would be similar, but the gas density is not, the discharges are not similar in the sense of Townsend scaling as was discussed above.

[13] 4. In sprite discharges, most light is emitted by the moving streamer head. This occurs on short time scales (microseconds). The comparison of the duration of their experiments by *Peterson et al.* [2009] with the 1–2 ms and 0.53 ms duration of sprites from *Pasko* [2007] is misleading.

Pasko mentions that the duration of order 1 ms is a time integration over the motion of the sprite head (page S24 first paragraph). Locally, the sprite channel will only emit light on a time scale of order 1 μs (the length of the streamer head is of order 10 m and its propagation velocity of order 10^7 m/s), as was shown by fast imaging by *McHarg et al.* [2007].

[14] The ~1 ms discharge produced in the lab by *Peterson et al.* [2009] has very different time scales, and therefore very different chemistry. In this long-duration discharge, multistep reactions become much more important, as there is enough time for reaction products to react further. Such a discharge is close to equilibrium, while a sprite (or streamer) is a transient discharge that is very far from equilibrium. In a semiequilibrium discharge, reactions of atoms, ions, radicals and excited species become very important, while a transient discharge is dominated by collisions of fast electrons with neutral gas molecules in the ground state. The reaction products from these collisions only start to play a significant role after the discharge has passed and therefore they are never in equilibrium. Some examples of the different time scales involved in the chemistry of a fast (nanosecond to microsecond) pulse in air can be found in Figure 10 of *Eliasson et al.* [1987] and Figure 5 of *van Veldhuizen et al.* [1996].

[15] Furthermore, *Peterson et al.* [2009] use a damped oscillating voltage and current to drive their discharge (see their Figure 5), while real blue jets and sprites have a pulse-like current of a single polarity. Proper comparison with sprites or blue jets is only possible with a pulse forming network. Examples of such pulse forming networks are C supplies, transmission line transformers and Blumlein pulsers [*Briels et al.*, 2006; *Smith*, 2002].

[16] 5. *Peterson et al.* [2009] mention in their paragraph [22] that sprites consist of a series of streamers, each hundreds of meters long and that this distance is required for the electrons to reach equilibrium with the surrounding electric field. In reality, the streamers in a sprite discharge are tens of kilometers long (and tens to hundreds of meters wide). On the other hand, *Li et al.* [2007] have shown that the electron relaxation length at standard temperature and pressure is about 1.5 μm. By applying Townsend scaling, we can see that at 65 km (0.1 mbar), the electron relaxation length is about 1.5 cm and at 80 km (0.01 mbar) it is about 15 cm. Therefore sprite lengths and electron relaxation lengths are not similar, but differ by 4–5 orders of magnitude.

[17] Summarizing, just the fact that there can be similarities between carefully chosen laboratory experiments and TLEs does not mean that all laboratory discharges represent a real TLE. There are many different types of cold plasmas and they can have vastly different chemistries. This can be determined by pressure, discharge duration, repetition frequency, discharge current density and more, none of which the authors prove to be equal between their discharges and real TLEs. The discharges described by *Peterson et al.* [2009] do not represent real sprites or blue jets. Especially the current duration and waveform in their experiment are very different from those in a real TLE, even though the pressure, color, and gas composition may be similar to TLEs.

3. Comparison Methods for NO$_x$ Production

[18] *Peterson et al.* [2009] use two methods to compare the production of NO$_x$ by their laboratory discharges to real

TLEs. In their first method they compare the energy consumed by their discharges with estimates of the energy of a real TLE. They take a lot of effort to measure the energy dissipated by their laboratory discharge, but then compare this to very rough estimates of TLE energies. These rough estimates have been deduced from optical light emission measurements.

[19] From recent observations by the ISUAL satellite mission, *Kuo et al.* [2008] have calculated the average energy emitted by a sprite discharge to be about 22 MJ. This is close to estimations by *Sentman et al.* [2003] which have a value of 1–10 MJ. Both are much lower than the older rough estimates from *Heavner et al.* [2000] of 250 MJ to 1 GJ that are used by *Peterson et al.* [2009].

[20] As discussed above, *van Heesch et al.* [2008] have shown that O* radical production efficiency of a discharge can vary significantly as function of discharge parameters. *Cooray et al.* [2009] show that also the NO_x production efficiency of electrical discharges not only depends on the energy dissipated in the discharge but also on the shape of the current waveform. This provides an explanation for the different values of NO_x molecules joule^{-1} obtained by different researchers in different experiments. Thus, according to Cooray et al., energy dissipated in a discharge is not suitable as the scaling quantity for extrapolating the laboratory data to lightning flashes. We realize that Cooray et al. appeared later than the paper by *Peterson et al.* [2009] but similar reasoning was already presented by *Cooray et al.* [2008].

[21] In our opinion, the statement by *Cooray et al.* [2008, 2009] that one can never extrapolate laboratory data to lightning flashes is too strong and does not hold if the laboratory discharges are really similar to the geophysical discharges. For discharges that obey Townsend scaling [*Ebert et al.*, 2010], one can use scaling laws to extrapolate laboratory data to geophysical discharges (e.g., comparing streamer discharges with sprites), as long as enough knowledge about both discharges is present.

[22] The second comparison method by *Peterson et al.* [2009] uses the geometric volume of the discharge to compare laboratory discharges to real TLEs. Again, this method has problems with good data from TLEs and depends a lot on a few field measurements.

[23] In the case of sprites, *Peterson et al.* [2009] do not distinguish clearly whether they use the complete volume of one carrot sprite, the volume of one single sprite channel or the volume of all sprite channels together. It seems that they have used the diameter of a single sprite channel as estimated by *Pasko* [2007], but that they did not multiply this cross section by the number of channels in a single sprite. They assume that Pasko's "effective diameter" takes this into account, while it does not, as it is the diameter of a single streamer channel. In conclusion, both comparison methods suffer from the same basic problem: in contradiction to their claims, the laboratory experiments by Peterson et al. are not similar to real TLEs and in their application of scaling they make errors and leave uncertainties.

4. Calculation Error

[24] In their Table 3, *Peterson et al.* [2009] use the geometric method to estimate the NO_x production by a blue jet, by assuming that the production is proportional to volume. However, in their comparison of the two geometries, they make a calculation error of 10^6. The estimations of blue jet NO_x should be 1.7×10^{28} to 6.4×10^{29} instead of 1.7×10^{22} to 6.4×10^{23}. The value of 1.7×10^{22} molecules production of NO_x per blue jet event is one of the most important results given in this paper and it is quoted both in the abstract and in the conclusions. Changing this value to its proper result of 1.7×10^{28} would change some quantitative results in the conclusions and abstract of the paper by 2–3 orders of magnitude.

5. Conclusions

[25] Although we recognize that it is impossible to reproduce exact atmospheric conditions in laboratory experiments, we think laboratory experiments can definitely be used to simulate many aspects of TLEs. However, this requires great care and a good understanding of the relation between the laboratory conditions and the TLE. We argue that the arguments by *Peterson et al.* [2009] regarding the similarities between their experiments and real TLEs and their comparison methods for NO_x production are not supported by good proof or evidence and are in many cases clearly wrong. They use a discharge voltage and current that is a few orders of magnitude longer than sprite and blue jet pulse durations and oscillates instead of having a fixed polarity.

[26] The grave calculation error by a factor 10^6 regarding the geometric estimation of NO_x production by a blue jet disqualifies this paper further. This error affects an important value discussed both in the abstract and in the conclusions of the paper.

[27] For a proper laboratory measurement of NO_x production from sprites, it would be worthwhile to study the efficiency of NO_x production in a similar way as *van Heesch et al.* [2008] have done for the O* production. In order to scale the measured NO_x production to that of TLEs, one would need to combine results of well chosen discharge experiments together with a review of the density dependence of chemical models like used by *Sentman et al.* [2008] and *Gordillo-Vazquez* [2008].

[28] On the other hand, if one only wants to prove that jets and sprites are no significant contributors to global NO_x production, a simple calculation would suffice. The highest estimate of the energy of one sprite or blue jet is about 1 GJ (which is probably an overestimate as we argued above). If we assume that this 1 GJ is used to produce NO with an enthalpy of formation of 90.29 kJ/mole at a 100% conversion efficiency (a clear overestimate), then each such TLE will produce 6.7×10^{27} molecules of NO. If we use NO_2 instead of NO, this number would be about a factor three higher as its enthalpy of formation is 33.1 kJ/mole.

[29] In any case, the resulting maximum (overestimated) NO_x production is similar or slightly higher than the results of *Peterson et al.* [2009] and therefore would lead to the same conclusion that TLEs do not significantly contribute to global NO_x production (when using the same assumptions of TLE occurrence and global NO_x production). Scaling of results from laboratory experiments to TLE energies can only give lower production estimates which will not change this conclusion.

References

Arnone, E., A. Kero, B. M. Dinelli, C.-F. Enell, N. F. Arnold, E. Papandrea, C. J. Rodger, M. Carlotti, M. Ridolfi, and E. Turunen (2008), Seeking sprite-induced signatures in remotely sensed middle atmosphere NO_2, *Geophys. Res. Lett.*, 35, L05807, doi:10.1029/2007GL031791.

Briels, T. M. P., J. Kos, E. M. van Veldhuizen, and U. Ebert (2006), Circuit dependence of the diameter of pulsed positive streamers in air, *J. Phys. D Appl. Phys.*, 39, 5201–5210, doi:10.1088/0022-3727/39/24/016.

Briels, T. M. P., E. M. van Veldhuizen, and U. Ebert (2008), Positive streamers in air and nitrogen of varying density: Experiments on similarity laws, *J. Phys. D Appl. Phys.*, 41, 234008, doi:10.1088/0022-3727/41/23/234008.

Chen, A., et al. (2008), Global distributions and occurrence rates of transient luminous events, *J. Geophys. Res.*, 113, A08306, doi:10.1029/2008JA013101.

Cooray, V., M. Becerra, and M. Rahman (2008), On the NO_x generation in corona, streamer and low pressure electrical discharges, *Open Atmos. Sci. J.*, 2, 176–180, doi:10.2174/1874282300802010176.

Cooray, V., M. Rahman, and V. Rakov (2009), On the NO_x production by laboratory electrical discharges and lightning, *J. Atmos. Sol. Terr. Phys.*, 71, 1877–1889, doi:10.1016/j.jastp.2009.07.009.

Cummer, S. A., U. S. Inan, T. F. Bell, and C. P. Barrington-Leigh (1998), ELF radiation produced by electrical currents in sprites, *Geophys. Res. Lett.*, 25, 1281–1284, doi:10.1029/98GL50937.

Dhali, S. K., and P. F. Williams (1987), Two-dimensional studies of streamers in gases, *J. Appl. Phys.*, 62, 4696–4707, doi:10.1063/1.339020.

Ebert, U., C. Montijn, T. M. P. Briels, W. Hundsdorfer, B. Meulenbroek, A. Rocco, and E. M. van Veldhuizen (2006), The multiscale nature of streamers, *Plasma Sources Sci. Technol.*, 15, S118–S129, doi:10.1088/0963-0252/15/2/S14.

Ebert, U., S. Nijdam, C. Li, A. Luque, T. Briels, and E. van Veldhuizen (2010), Review of recent results on streamer discharges and discussion of their relevance for sprites and lightning, *J. Geophys. Res.*, 115, A00E43, doi:10.1029/2009JA014867.

Eliasson, B., M. Hirth, and U. Kogelschatz (1987), Ozone synthesis from oxygen in dielectric barrier discharges, *J. Phys. D Appl. Phys.*, 20, 1421–1437, doi:10.1088/0022-3727/20/11/010.

Enell, C. F., et al. (2008), Parameterisation of the chemical effect of sprites in the middle atmosphere, *Ann. Geophys.*, 26, 13–27, doi:10.5194/angeo-26-13-2008.

Gordillo-Vazquez, F. J. (2008), Air plasma kinetics under the influence of sprites, *J. Phys. D Appl. Phys.*, 41, 234016, doi:10.1088/0022-3727/41/23/234016.

Heavner, M. J., D. D. Sentman, D. R. Moudry, E. M. Wescott, C. L. Siefring, J. S. Morrill, and E. J. Bucsela (2000), Sprites, blue jets and elves: Optical evidence of energy transport across the stratopause, in *Atmospheric Science Across the Stratopause*, *Geophys. Monogr. Ser.*, vol. 123, pp. 69–82, AGU, Washington, D. C.

Kuo, C. L., et al. (2008), Radiative emission and energy deposition in transient luminous events, *J. Phys. D Appl. Phys.*, 41, 234014, doi:10.1088/0022-3727/41/23/234014.

Levine, J. S., R. S. Rogowski, G. L. Gregory, W. E. Howell, and J. Fishman (1981), Simultaneous measurements of NO_x, NO, and O_x production in a laboratory discharge: Atmospheric implications, *Geophys. Res. Lett.*, 8, 357–360, doi:10.1029/GL008i004p00357.

Li, C., W. J. M. Brok, U. Ebert, and J. J. A. M. van der Mullen (2007), Deviations from the local field approximation in negative streamer heads, *J. Appl. Phys.*, 101, 123305, doi:10.1063/1.2748673.

Liu, N., and V. P. Pasko (2004), Effects of photoionization on propagation and branching of positive and negative streamers in sprites, *J. Geophys. Res.*, 109, A04301, doi:10.1029/2003JA010064.

Luque, A., U. Ebert, C. Montijn, and W. Hundsdorfer (2007), Photoionization in negative streamers: Fast computations and two propagation modes, *Appl. Phys. Lett.*, 90, 081501, doi:10.1063/1.2435934.

McHarg, M. G., H. C. Stenbaek-Nielsen, and T. Kammae (2007), Observations of streamer formation in sprites, *Geophys. Res. Lett.*, 34, L06804, doi:10.1029/2006GL027854.

Morrow, R. (1985), Theory of negative corona in oxygen, *Phys. Rev. A*, 32, 1799–1809, doi:10.1103/PhysRevA.32.1799.

Neubert, T., et al. (2008), Recent results from studies of electric discharges in the mesosphere, *Surv. Geophys.*, 29, 71–137, doi:10.1007/s10712-008-9043-1.

Pasko, V. P. (2007), Red sprite discharges in the atmosphere at high altitude: The molecular physics and the similarity with laboratory discharges, *Plasma Sources Sci. Technol.*, 16, S13–S29, doi:10.1088/0963-0252/16/1/S02.

Pasko, V. P., U. S. Inan, and T. F. Bell (1998), Spatial structure of sprites, *Geophys. Res. Lett.*, 25, 2123–2126, doi:10.1029/98GL01242.

Peterson, H., M. Bailey, J. Hallett, and W. Beasley (2009), NO_x production in laboratory discharges simulating blue jets and red sprites, *J. Geophys. Res.*, 114, A00E07, doi:10.1029/2009JA014489.

Rocco, A., U. Ebert, and W. Hundsdorfer (2002), Branching of negative streamers in free flight, *Physical Review E*, 66, 035102, doi:10.1103/PhysRevE.66.035102.

Sentman, D. D., et al. (2003), Simultaneous observations of mesospheric gravity waves and sprites generated by a midwestern thunderstorm, *J. Atmos. Sol. Terr. Phys.*, 65, 537–550.

Sentman, D. D., H. C. Stenbaek-Nielsen, M. G. McHarg, and J. S. Morrill (2008), Plasma chemistry of sprite streamers, *J. Geophys. Res.*, 113, D11112, doi:10.1029/2007JD008941.

Smith, P. W. (2002), *Transient Electronics: Pulsed Circuit Technology*, Wiley, Chichester, U. K.

van Heesch, E. J. M., G. J. J. Winands, and A. J. M. Pemen (2008), Evaluation of pulsed streamer corona experiments to determine the O* radical yield, *J. Phys. D Appl. Phys.*, 41, 234015, doi:10.1088/0022-3727/41/23/234015.

van Veldhuizen, E. M. (2000), *Electrical Discharges for Environmental Purposes: Fundamentals and Applications*, Nova Sci., New York.

van Veldhuizen, E. M., W. R. Rutgers, and V. A. Bityurin (1996), Energy efficiency of NO removal by pulsed corona discharges, *Plasma Chem. Plasma Process.*, 16, 227–247, doi:10.1007/BF01570180.

U. Ebert, S. Nijdam, and E. M. van Veldhuizen, Department of Applied Physics, Eindhoven University of Technology, Den Dolech 2, Eindhoven NL-5612 AZ, Netherlands. (s.nijdam@tue.nl)