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On the NO_x Generation in Corona, Streamer and Low Pressure Electrical Discharges

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Abstract: A theory developed by scientists to study nitrogen oxides, NO_x, production by solar proton events shows that the NO_x production rate is approximately equal to the rate of production of ion pairs during the proton impact. Since the bulk of ionization in such events is produced by secondary electron impacts, the same concept is used here to study the NO_x production in low pressure discharges, corona discharges and streamer discharges in which the source of ionization is the electron impacts. Using experimental data pertinent to corona discharges it is established that, as in the case of proton impacts, the rate of NO_x production is approximately equal to the rate of production of ion pairs. The theory in turn is applied to study the NO_x production in streamer and low pressure electrical discharges. The results show that the NO_x production in low pressure discharges depends not only on the energy dissipated but also on the ambient pressure and the electric field. In low pressure discharges the efficiency of NO_x production is given by $k\alpha(p,E)/eE$ NO_x molecules/J, where $\alpha(p,E)$ is the Townsend's first ionization coefficient, p is the atmospheric pressure, e is the electronic charge, E is the electric field and k is the number of NO_x molecules resulting during an ionizing event. This shows that the NO_x production efficiency of a discharge depends not only on the energy dissipation but also on the pressure and the electric field. In the case of streamer discharges, the NO_x molecules produced by the streamer in propagating a unit distance is given by $k/2\omega eE$, where ω is the number of positive ions located in the streamer head.

Keywords: Lightning, return stroke, corona, streamers, nitrogen oxides, NO_x.

INTRODUCTION

An assessment of the global distribution of nitrogen oxides NO_x is required for a satisfactory description of tropospheric chemistry and in the evaluation of the global impact of increasing anthropogenic emissions of nitrogen oxides [1]. In the mathematical models utilized for this purpose it is necessary to have the natural as well as man-made sources of nitrogen oxides in the atmosphere as inputs. Thunderstorms are a main natural source of nitrogen oxides in the atmosphere and it may be the dominant source of nitrogen oxides in the troposphere in equatorial and tropical south pacific [2].

In quantifying the production of NO_x by thunderstorms, scientists have until recently concentrated on the lightning return strokes neglecting all other processes associated with thunderstorms [3]. This view is gradually changing as the theories and experimental data show that not only the return strokes in ground flashes but other discharge events in ground and cloud flashes, such as continuing currents, are also contributing significantly to the NO_x emissions [4]. For example the study conducted by Rahman *et al.* [4] shows that the contribution by continuing currents to the NO_x production in lightning flashes is comparable, if not overwhelm, the contribution by return strokes. However, the physics behind the process which makes continuing currents as efficient as return strokes in producing NO_x is still unknown.

Based on the ionization process that leads to the production of free electrons, electrical discharges taking place in the atmosphere can be divided into two types, namely, 'cold' and 'hot' electrical discharges. In cold electrical discharges free electrons are produced solely by the collisions between energetic electrons and atoms. In these discharges the electron temperature may reach several tens of thousands of degrees whereas the gas and ion temperature remains close to ambient temperature. Corona discharges, streamer discharges and Townsend type electrical discharges taking place at low pressure are several examples of cold discharges taking place in the atmosphere. In hot electrical discharges the gas and ion temperature can also reach several tens of thousands of degrees and the main mechanism in the discharge that generates free electrons is the thermal ionization. In these discharges ions and neutral atoms are heated to such high temperatures through a process called thermalization which facilitates the transfer of energy from free electrons to neutrals *via* ions [5]. This transfer of energy from electrons to neutrals causes the temperature of the neutrals to go up. This increase in the temperature of the neutrals leads to production of copious amount of electrons by the energetic collisions between neutral particles (i.e. thermal ionization). Several examples of hot discharge processes taking place in the atmosphere are return strokes, leaders, M components and continuing currents.

In addition to hot discharges mentioned above, an active thundercloud also produces cold discharges in the form of corona and streamer discharges. However, in quantification of NO_x produced by thunderstorms scientists neglect the con-

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tribution of the cold discharge processes. Moreover, the recent discovery of thunderstorm-created ionization processes in the stratosphere and mesosphere, known as sprites, blue jets and elves, also poses a question as to the effects of these ionization processes on the chemistry of the upper atmosphere. These ionization processes can also be categorized under 'cold' discharges because the pressure at which these discharges take place does not support thermal ionization, making electron impacts the main source of ionization. Indeed, there is a need today to develop procedures to quantify the NO_x production in cold discharges.

The effects of solar proton events in the NO_x production in the upper atmosphere and the effect of NO_x on the chemical balance of the stratosphere was a major concern of the atmospheric scientists since the discovery of the importance of NO_x in ozone production and destruction [6]. In quantifying the NO_x production from these events, scientists utilized the connection between the ionizing events in the atmosphere and the number of resulting NO_x molecules. To facilitate further discussion let us denote the number of NO_x molecules produced per ionizing event in the atmosphere by the parameter k. The theoretical work of Nicolet [7] set the value of k close to unity whereas the investigations of Jackman *et al*. [8] predicted that at altitudes larger than about 80 km k is about 1.5 and for low altitude it is about 1.2-1.3. These results have been used extensively to study the NO_x production by cosmic rays and solar radiation impinging on the Earth's atmosphere. Recently, Rahman *et al*. [9] investigated the validity of this theoretical calculation by studying the NO_x production in air by alpha particles emitted by a radioactive source. The results of this study confirmed these theoretical predictions fixing the value of k to about 1.0. Since the main source of ionization during proton impacts are high energetic secondary electrons, it is reasonable to assume that the number of NO_x molecules produced in discharge processes in which electrons are the main source ionization is approximately equal to the number of ion pairs produced in the discharge. This hypothesis is tested in this paper using the data obtained from corona discharges and then it is utilized to quantify the NO_x production in low pressure discharges and streamer discharges.

TESTING THE HYPOTHESIS USING CORONA DISCHARGES

The working hypothesis of this paper is that in cold electrical discharges, the total number of NO_x molecules generated by the discharge is k times the total number of ion pairs produced in the discharge. Let us test this hypothesis by using the experimental data on corona discharges.

In a recent study, Rehbein and Cooray [10] conducted an experiment to quantify the NO_x production in corona discharges. In the study a corona discharge is maintained in a coaxial geometry and the discharge voltage and the current are measured simultaneously with the concentration of NO_x produced in the discharge chamber. If one neglects the ionization loss processes (i.e. attachment and recombination), the steady state current gives the rate of production of electrons and hence the rate of occurrence of ionizing events in the discharge. Since the total NO_x production in the discharge over a given time interval is known, the data can be used to quantify the number of NO_x molecules produced per ion pair. The results of this calculation gave $k = 0.6$ for negative corona and $k = 1.0$ for positive corona. Even though the NO_x generating efficiency per ion pair is lower in negative corona than in positive corona, this result confirms that the number of NO_x molecules produced in the discharge is approximately equal to the number of ionization events.

NO_X GENERATION IN ELECTRON AVALANCHES AND ITS RELATION-SHIP TO ENERGY DISSIPATION

Consider a single electron accelerating in a background electric field of strength E. The number of ionizations caused by the electron in moving a unit length is equal to the Townsend's primary ionization coefficient $\alpha(p,E)$ which is a function of pressure p and the electric field E. Assuming that one ionization event corresponds to k number of NO_x molecules, the total number of NO_x molecules N_{ul} produced by the ionization processes as the electron moves a unit length is given by

$$N_{ul} = k\alpha(p,E) \tag{1}$$

Now, the energy dissipated as the electron moves a unit length in the background electric field is given by eE, where e is the electronic charge. Thus, the number of NO_x molecules produced per unit energy, N_{ue}, is given by

$$N_{ue} = k\alpha(p,E)/eE \tag{2}$$

Since α/p is a function of the reduced electric field E/p alone, the above equation predicts that N_{ue} is only a function of E/p. Fig. (**1**) shows how the value of N_{ue} varies as a function of E/p. First note that the NO_x production efficiency increases with increasing E/p. This also shows that for a given pressure, the NO_x production per unit energy is not unique but depends also on the applied electric field. Consequently, in any experiment that is designed to obtain the NO_x production efficiency of cold electrical discharges, it is necessary to utilize voltage impulses in which the reduced electric field E/p remains the same.

NO_X PRODUCTION IN STREAMER DISCHARGES

As an electron avalanche propagates towards the anode of a discharge gap, low mobile positive space charge accumulates at the avalanche head. When the avalanche reaches the anode, the electrons will be absorbed into it leaving behind the net positive space charge. Due to the recombination of positive ions and electrons, the avalanche head is a strong source of high energetic photons. These photons will create other avalanches in the vicinity of the positive space charge. If the number of positive ions in the avalanche head is larger than a critical value, the electric field created by the space charge becomes comparable to the background electric field and the secondary avalanches created by the photons will be attracted towards the positive space charge. The electrons in the secondary avalanches will be neutralised by the positive space charge of the primary avalanche, leaving behind a new positive space charge, little bit closer to the cathode. The process repeats itself and the positive space charge head travels towards the cathode as a consequence [11]. This discharge that travels towards the cathode from the anode is called a positive streamer. At atmospheric pressure, the total number of ions in the streamer head ω is about 10^8 and the radius of the streamer head, R_S is about 100 μm [12]. The streamer needs a background field of about 5-10 kV/cm, depending on polarity for continuous propagation. Consider a streamer moving in a uniform electric field of strength E. In order for the streamer to move a unit length in this electric field the total number of ionizing events taking place in the vicinity of the streamer head is about $\omega / 2R_S$ and the total number of NO_x molecules created by the streamer in moving a unit length, N_S is

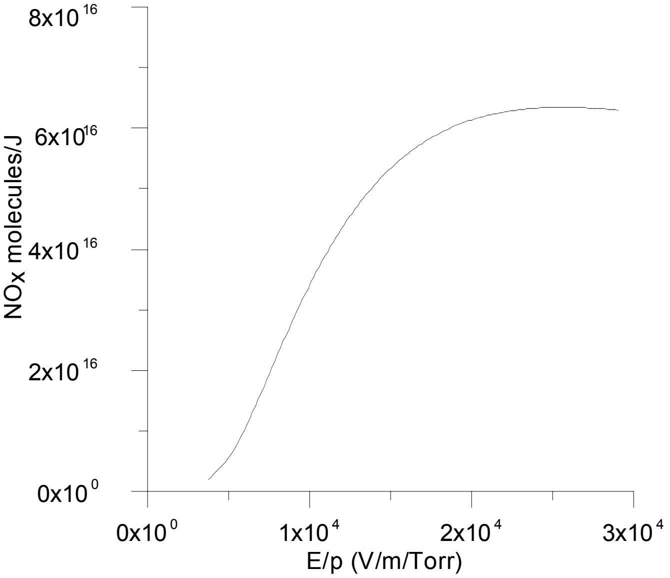

Fig. (1). NO_x production per unit energy N_{ue} as a function of E/p in electron avalanches.

$$N_S = k\omega / 2R_S \qquad (3)$$

On the other hand the amount of energy dissipated by the streamer channel in moving the unit distance is $Ee\omega$. Thus the production efficiency of NO_x in streamer discharges, P_{NOx} in molecules/J is

$$P_{NOx} = k / 2eER_S \qquad (4)$$

Substituting $k = 1$, $R_S = 10^{-4}$ m and $E = 5\text{x}10^5$ V/m (for positive streamers) we find that the streamer will make about 6 x 10^{16} NO_x molecules/J.

In a recent study Cooray and Rahman [13] conducted an experiment in coaxial geometry to measure the NO_x production efficiency of streamer discharges. Let us consider the cylindrical coaxial geometry. Assume that the peak voltage applied to the central conductor of the coaxial system is V. This voltage will create an electric field in the cylinder, which has its highest value at the surface of the conductor and then decreases inversely with increasing radial distance. Consider a streamer discharge initiated close to the inner electrode and moving in this electric field. Assume that the streamer discharge will propagate to a distance where the background electric field reaches E_S, the critical electric field necessary for streamer propagation. If the applied voltage is V then the charge that will be induced on an unit length of the inner conductor of the coaxial arrangement is given by

$$Q = \frac{2\pi\varepsilon_0 V}{\ln(b/a)} \qquad (5)$$

where a and b are the radii of the inner and outer conductors. The electric field at a radial distance r from the inner conductor is then given by

$$E_r = \frac{V}{r\ln(b/a)} \qquad (6)$$

Since the streamers propagate to a distance where the background electric field is E_S, the length of the streamers, l_S, in the coaxial arrangement is given by

$$l_S = \frac{V}{\ln(b/a)E_S} - a \qquad (7)$$

Then the number of ionizing events, N_S, generated during the propagation of the streamer is given by

$$N_S = \frac{\omega l_S}{2R_S} \qquad (8)$$

On the other hand the energy released by the movement of the streamer head in the gap, U_S is given by

$$U_S = e\omega \frac{V}{\ln(b/a)} \ln(l_S/a) \qquad (9)$$

Thus the number of NO_x molecules produced per unit energy, N_{ue}, is given by

$$N_{ue} = \frac{k}{2eE_S R_S \ln(l_S/a)} \qquad (10)$$

In deriving this equation we have assumed that $l_s >> a$. For the experimental conditions reported in the paper, i.e. $a = 0.001$ m, $b = 0.15$ m and $V = 83$ x 10^3 V, the calculated production efficiency is about 1.6 x 10^{16} molecules/J. The measurements produced 1 - 2 x10^{16} molecules/J.

DISCUSSION AND CONCLUSIONS

Studies on the NO_x production in the atmosphere by proton impacts show that the number of NO_x molecules created is almost equal to the number of ion pairs created during the impact. By comparing theory with available experimental data it is shown in this paper that this result is also valid for electrical discharges in which electron impacts are the main source of ionization. Based on this observation the NO_x production in electrical discharges where the electron production depends solely of the impact of electrons with neutral atoms is evaluated. The types of electrical discharges considered in this paper are the corona discharges, electrical discharges at low pressure and streamer discharges.

In a corona discharge associated with current amplitude I, the rate of occurrence of ionizing events (neglecting attachment and re-combination) is I/e, where e is the electronic charge. This is equal to 6.25 x 10^{18} I. Thus, the number of NO_x molecules produced per second in the discharge is given by 6.25 x 10^{18} kI in which the value of k depends on polarity (i.e. $k \approx 1$ for positive polarity and $k \approx 0.6$ for negative). It is of interest to note that this equation can be utilized to estimate the global production of NO_x by ground corona associated with thunderstorms. Various studies estimate that the global corona current associated with ground corona is about 1-2 kA [14]. Substituting this value in the above equation and using $k = 1.0$ for positive corona the number of NO_x molecules produced globally by ground corona per second is estimated to be about (6-12) x 10^{21}. This is equivalent to an annual production of 0.01 *Tg (N)*. During thunderstorms it is not only at ground level that corona discharges are initiated. The thunderstorm itself is a large source of corona discharges. For example, in a thundercloud charges may disperse from regions of high concentration to low concentration through corona discharges and the same could be the vehicle that transports the charges induced in conducting channels during neutralization events in to the bulk of the cloud. The theory as presented in this paper could be applied to obtain the NO_x production from these processes once the magnitude of the currents associated with these processes are known.

Let us consider the results presented in Fig. (**1**). These results are obtained using several assumptions. First it is assumed that the electron generating mechanism in the discharge is the impact ionization due to energetic electrons. Second, it is assumed that the background electric field is uniform in the region in which the discharge is taking place. However, the calculation procedure can be modified rather easily to take into account the effect of non uniform electric fields. Third, it is assumed that the space charge accumulated in space during the discharge will not distort the electric field locally. Due to these assumptions the results as given in Fig. (**1**) are valid for 'Townsend's like' low pressure electrical discharges where the space charge effects can be neglected. On the other hand the data given in Fig. (**1**) can be utilized to study the NO_x production in any cold discharge with space charge distortions provided that the quantity E is replaced by the effective electric field in which the electron avalanches are generated.

An example of a cold discharge in which the space charge effects cannot be neglected is a streamer discharge. In streamers the electron avalanches are initiated in the electric field of the streamer head which is much stronger than the background electric field in which streamers are propagating. In the present paper, in evaluating the NO_x production of streamer discharges in this paper, instead of evaluating the NO_x production in each individual avalanche taking place at the streamer head, a simpler but an equivalent procedure based on the known mechanism of the streamer is utilized. In a streamer discharge electron avalanches are generated in a specially varying electric field. The maximum value of the field which is about 2 x 10^7-4 x 10^7 V/m occurs at the head of the streamer (assuming 10^8 ions located within a radius of about 100 μm-50 μm) and it decreases to about 3x10^6 V/m at a distance of about 200 μm from the head. Recall that our analysis gave about 2 x10^{16} NO_x molecules per Joule for streamer discharges moving in a background electric field of about 5 x 10^5 V/m. Comparison of this NO_x production efficiency with the data given in Fig. (**1**) indicates that in streamer discharges the electron avalanches are generated in an effective electric field of about 5 x 10^6 V/m.

Streamer discharges occurring in the atmosphere are associated mainly with the lightning leaders in ground flashes and cloud flashes. Actually, the propagation of the leader is mediated by streamer bursts emanating from the tip of the leader. They may also originate during the neutralization of these leaders by return strokes. Cooray *et al*. [15] utilized the theory as developed in this paper to study the NO_x production by streamers in lightning leaders.

The experimental observations indicate that the space charge effects cannot be neglected in upper atmospheric discharges known as sprites even though these discharges are taking place at considerably low atmospheric pressures. For example, observations indicate that sprites give rise to streamer like structures. Therefore, the electric field in which the avalanches are growing during the development of sprites may be considerably higher than the background electric field generated by the thunderclouds. However, if the dimension and the ion concentration in these streamer heads are known an analysis similar to that utilized in this paper to study the NO_x production in streamer discharges at atmospheric pressure could be used to evaluate the NO_x production in streamers in sprites. On the other hand, the elves are not associated with streamers and the results presented in Fig. (**1**) could be applied directly if the background electric field that drives them is estimated.

The reason for the development of streamers in sprites, even though they are taking place at considerably low atmospheric pressure, is the large dimensions involved with the region of ionization associated with these electrical discharges. This allows the accumulation of space charge over large volumes affecting the local electric field. On the other hand laboratory discharges generated at the same pressures would not have streamer type discharges because the special dimensions are not large enough in laboratory discharges to support them. The results presented in Fig. (**1**) would be applicable directly in low pressure laboratory discharges. In a study conducted recently by Peterson *et al*. [16], NO_x generated by laboratory discharges at different pressures were measured and based on the results an estimation was made on how the NO_x production efficiency in molecules/J varies as a function of pressure. Based on the results the authors predicted that that the NO_x production efficiency increases with decreasing pressure. Unfortunately, in that study the applied voltage is not given and therefore it is difficult to find out whether the voltage applied at different pressures is such that the *E/p* ratio is the same at different pressures. As shown in the present paper the NO_x production efficiency in these discharges depends on the ratio E/p and any change in this ratio when one moves from one pressure to another will also affects the NO_x production.

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