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Could cloud-to-cloud discharges be as effective as cloud-to-ground discharges in producing NO_x?

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ABSTRACT

In global models of tropospheric ozone and oxidized nitrogen, it is usually assumed that cloud-to-cloud discharges are 3 to 10 × less effective than cloud-to-ground discharges in producing nitrogen oxides (NO_x). This assumption is based on a limited number of observations and experiments regarding the acoustic and optical energy spectrum of lightning discharges. We claim that cloud-to-cloud and cloud-to-ground discharges dissipate similar amounts of energy and they may thereby be equally effective per discharge as NO_x producers. Nevertheless, the mechanism of NO_x production by electrical discharges still needs to be clarified, as well as whether there is a vertical differentiation in the production mechanism. We performed sensitivity studies with a global 3-D climatological tracer model (MOGUNTIA). The studies indicate that the simulated tropospheric distributions of oxidized nitrogen and ozone are sensitive to the vertical distribution of the lightning source assumed. Furthermore, it is shown that the strength of the global lightning source used in 3-D global model studies of oxidized nitrogen should be amplified by a factor of 2.6 if cloud-to-cloud discharges are equally effective as cloud-to-ground discharges in producing nitrogen oxides.

1. Introduction

Ozone (O_3) is a key tracer for photochemistry of the background troposphere. Its tropospheric production is modulated by the presence of nitrogen oxide (NO) and nitrogen dioxide (NO_2) (Crutzen, 1973). Hence, an evaluation of tropospheric ozone production requires a correct description of the emission, chemistry and distribution of nitrogen oxides. Lightning has been identified as a potentially very important source of NO_x ($NO_x = NO + NO_2$) in the free troposphere and in remote marine areas, where no other in situ NO_x sources are known to be important (Kasibhatla et al., 1991). Moreover, NO_x emitted by lightning is more readily subject to long-range transport than emissions occurring within the boundary layer. The impact of lightning on the free tropospheric balance of ozone and oxidized nitrogen will therefore depend on the vertical distribution of the lightning source of NO_x. However, our understanding of the mechanisms for the production of nitrogen oxides by electrical discharges and of the actual distribution of this natural source of NO_x is poor.

Two types of lightning discharges are considered according to their vertical allocation: cloud-to-ground and cloud-to-cloud discharges. Following Uman (1987), the most common type of cloud-to-ground discharge is the negative cloud-to-ground discharge. The whole discharge is called a flash, starts in the cloud, and brings to earth several coulombs of negative charge within half a second. The discharge process is initiated by a

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stepped leader, which itself is initiated by a preliminary breakdown within the cloud. The rapid release of return-stroke energy heats the leader channel to a temperature near 30000°K, and generates a high-pressure channel that expands and creates the shock wave that eventually becomes thunder. The leader channel consists of a central core surrounded by a corona sheath. If additional charge is made available at the top of the channel, a continuous dart leader may propagate down the residual first-stroke channel. The dart leader then initiates the second or any subsequent return stroke. Cloud discharges are defined as any lightning flash that does not connect to earth. They can be subdivided into intracloud, intercloud, and cloud-to-air flashes, but there is no experimental data at present to distinguish between the characteristics of these 3 types.

Hitherto, estimates of the global production of NO_x by lightning, applied in global three-dimensional modeling studies of tropospheric ozone and oxidized nitrogen, have assumed that cloud-to-cloud discharges are three to ten times less energetic than cloud-to-ground discharges (Penner et al., 1991; Crutzen and Zimmerman, 1991; Dentener and Crutzen, 1993; Kasibhatla et al., 1993), and thereby 3 to $10\times$ less effective per discharge as NO_x producers. This paper explores the basis for this assumption. We review the calculations by Cooray (1996) for energy dissipation in lightning discharges, and discuss their impact on the strength and distribution of the lightning source of nitrogen oxides. A sensitivity study that addresses the importance of the vertical distribution of the lightning source of NO_x for the distribution of oxidized nitrogen and ozone is presented.

2. Global estimates of nitrogen fixation by lightning

Current estimates of the strength of the lightning source of NO_x are very uncertain. The range estimated by Logan (1983) is 2 to 20 TgN/yr, with a best value of 8 TgN/yr. However, figures as high as 100 TgN/yr have been suggested (Franzblau and Popp, 1989). Using a 3-D global model of the oxidized nitrogen cycle, Gallardo and Rodhe (1995) found that values greater than 20 TgN/yr for this particular source are very difficult to reconcile with observed nitrate wet deposition data. Also, Levy et al. (1995, pers. communication), by comparing a model simulation of tropospheric NO_x with measurements in the remote mid- and upper-troposphere, have bracketed the annual global emission to be in the range between 2 and 6 TgN/yr.

The annual production of NO_x by lightning discharges can be calculated by multiplying the frequency of discharges by the amount of NO_x produced per discharge. The horizontal distribution of lightning, and thereby the total frequency of discharges, can be obtained from satellite observations (Turman and Edgar, 1982; Kotaki and Katoh, 1983; Orville and Henderson, 1986). However, these observations of global lightning activity are given only for isolated years and times of day. Price and Rind (1992) presented a simple lightning activity parameterization that allows a climatological representation of global lightning activity. This parameterization makes use of a semi-empirical relationship between convective cloud top height and lightning flash frequency, with different formulations for continental and marine thunderstorms. The mean global lightning frequency, i.e., the mean number of lightning flashes occurring around the globe per unit time, is estimated to be 100 flashes per second, with a range between 70 to 120 flashes/s (Turman and Edgar, 1982; Borucki and Chameides, 1984; Price and Rind, 1992).

It is, however, even more difficult to assess the amount of NO_x produced per discharge, as the mechanism for this production is not well understood. Theoretical (Hill et al., 1980; Borucki and Chameides, 1984 and others) and experimental (Chameides et al., 1977; Levine et al., 1981; Peyrous and Lapeyre, 1982 and others) studies have focused on the production of nitrogen oxide (NO) in and around the rapidly cooling discharge tube of a lightning stroke, typically a cloud-to-ground discharge. The mechanism usually invoked as responsible for NO production is the Zel'dovitch mechanism (Zel'dovich and Raizer, 1967), by which NO is produced through high-temperature dissociation of molecular nitrogen (N_2) and oxygen (O_2), and subsequent reaction between the atomic species. The emission occurs in the form of NO, and is followed by oxidation of NO to NO_2 by O_3. The equilibrium NO concentration (i.e., the NO concentration at which

NO-producing and NO-destroying reactions are in balance) is a strong function of temperature. The production of NO becomes significant as the temperature rises above 1500 °K (Borucki and Chameides, 1984; Chameides, 1986). The time required to establish thermochemical equilibrium is also a function of temperature, being longer at lower temperatures. So, when rapid cooling takes place, higher equilibrium concentrations of NO than those corresponding to ambient temperature, can be frozen out. On the other hand, Goldembaum and Dickerson (1993) found that NO production is controlled by changes in density, not temperature. They investigated NO production in the expansion phase of a lightning discharge using a hydrodynamic model coupled with the two Zel'dovitch reactions and oxygen dissociation. These authors claim that the rate of production of NO drops off rapidly with decreasing air density and thus altitude. Also, Cooray et al. (1995, pers. communication) have experimentally studied the production of NO_x and ozone by streamers in the corona sheath of lightning discharges. The experiment shows that other discharges than return strokes might be important for the production of these compounds in the troposphere. Thus, further investigation is needed in order to determine the formation mechanism (or mechanisms) of NO_x in electrical discharges (not only in the hot channel associated with return strokes) and to what extent this mechanism is altitude dependent.

Lawrence et al. (1994) presented a review of the estimates of the global fixation of nitrogen by lightning. According to their study, the amount of NO produced by a lightning discharge ranges from 1 to 7×10^{25} molecules of NO. In theoretical and experimental studies, the NO yield per lightning discharge has usually been calculated by multiplying the yield of NO per unit energy by the energy of a typical lightning discharge. The former parameter is estimated by Lawrence et al. (1994) to be in the range of 2 to 13×10^{16} molecules NO per Joule. The latter is believed to be the more uncertain of the two and is estimated to range from 1 to 20×10^{8} Joule per flash. Hence, much of the uncertainty involved in the estimate of the NO yield per discharge is due to our poor knowledge of energy dissipation in lightning flashes.

3. Energy dissipation in lightning flashes

Estimates of energy dissipated in lightning discharges have been made by assuming that a given amount of charge is transferred across a known potential difference (Wilson, 1920; Berger, 1977 and others). However, since the cloud is an extended structure the concept of cloud potential does not have a strict physical meaning. Other estimates have been based on the acoustic and optical energy spectrum of lightning discharges (Conner, 1967; Guo and Krider, 1982). However, this approach requires ad hoc assumptions regarding the relationship between these signatures and the total energy dissipation in the discharge. Other researchers have analyzed the temporal development of the channel properties and the energy dissipation in a lightning channel (Plooster, 1971; Paxton et al., 1986). This approach also needs further assumptions about the shape and amplitude of the return stroke. In general, these estimates neglect the energy dissipation during the leader stage, and only a few of them consider a realistic cloud charge model in the analysis.

Cooray (1996) recently calculated the amount of energy dissipation during different stages of cloud-to-ground discharges, based on electrostatic energy considerations, using realistic cloud charge distributions. According to the classical charge distribution by Malan (1963), there are 3 charge centers in a thundercloud: a positive charge center located at the top of the cloud, a negative charge center below, and a small positive charge pocket at the bottom of the cloud. Cooray's model simulates such a charge distribution and a lightning flash by a leader stage followed by a return stroke. The electrostatic energy of the dipolar system is calculated before and after the occurrence of the discharge. The amount of energy dissipated is then equal to the difference in electrostatic energy between the initial an the final stages. Notice that no assumptions are made regarding the discharge process; the calculation is made in terms of the charge of the centers, the distance between them and the amount of charge neutralized through the discharge. The calculations showed that, for a given amount of charge neutralization, the cloud flash is more energetic than the ground discharge (Table 1). In this analysis, the same cloud charge configuration is used for both cloud-to-ground and cloud flashes. Note that the total energy

Table 1. *Total energy dissipated during a lightning discharge as a function of the amount of charge neutralized during the process (Adapted from Cooray, 1996)*

Charge neutralized (°C)	Energy dissipated ($J \times 10^8$) Ground flash	Cloud flash
1	1.0	2.2
2	2.0	4.3
3	3.0	6.4
4	3.6	8.1
5	4.4	9.8
6	5.2	11.6
7	6.0	13.2
8	6.4	14.5

dissipated in a cloud flash that neutralizes a given amount of charge, is about 2 × the energy released in a stepped leader-return stroke process that neutralizes the same amount of charge. Estimates made from electric field measurements indicate that the amount of charge neutralized by cloud discharges is comparable with the amount of charge neutralized by a typical cloud-to-ground discharge, which is about 7 C (Berger et al, 1975; Brook and Ogawa, 1977). Therefore, we expect cloud discharges to dissipate at least similar amounts of energy as cloud-to-ground discharges. Table 2 summarizes the estimates by Cooray (1996) for a typical cloud-to-ground discharge. In this specific calculation, it is assumed that [5°C] are neutralized in the first stepped leader-return stroke process, and [1°C]in the subsequent processes. Further, it is assumed that most of the energy dissipated in the leader stages, dissipates as corona. Notice that the energy dissipation takes place partly in the leader stage and partly in the return stroke stage.

Table 2. *Energy dissipation in a typical cloud-to-ground discharge according to calculations by Cooray (1996)*

Lightning process		Energy dissipation ($J \times 10^8$)
stepped leader		2.00
first return stroke	Corona Sheath	1.65
	Central Core	1.35
dart leader		0.70
subsequent return stroke	Corona Sheath	0.05
	Central Core	0.25

4. The efficiency of cloud-to-cloud and cloud-to-ground discharges as NO_x producers

To determine the efficiency of cloud and cloud-to-ground discharges as NO_x producers one should have a more detailed knowledge about the discharge process, the amount of energy involved in it, and the mechanism responsible for NO_x formation. Unfortunately, our present understanding of these phenomena is poor, particularly regarding cloud discharges. Cloud-to-ground lightning has been more intensively studied than other types of lightning because of its direct effects on the human environment and because it is more easily observed by optical means (Uman, 1987).

In the absence of better knowledge, researchers have assumed a linear scaling between the amount of energy dissipated and the amount of NO_x formed. However, this assumption might not be adequate. In fact, Chameides (1979) studied the production of NO per energy unit, according to the Zel'dovitch mechanism, as a function of the input energy in a spark discharge and he found a non-linear relationship between both parameters. Also, the model study by Goldembaum and Dickerson (1993) indicates that one cannot make a linear connection between energy dissipation and NO_x production.

As mentioned earlier, when considering the partitioning between cloud discharges and cloud-to-ground discharges as NO_x producers, it is commonly assumed that cloud-to-cloud discharges are three to ten times less energetic than cloud-to-ground discharges. This assumption is based on the observation that optical and acoustic signatures produced by ground discharges are several times larger than the signatures produced by cloud discharges (Holmes et al., 1971; Berger, 1977) and on further assumptions regarding the relationship between these signatures and the total energy dissipation in the discharge. Furthermore, cloud discharges may develop intermittently (Brook and Ogawa, 1977) and therefore contribute less to the cumulative addition of the optical and acoustic signatures as compared to the rapidly developing return strokes. The calculations done by Cooray (1996), discussed in the previous section, show that cloud discharges may actually dissipate similar or even larger amounts of energy compared to cloud-to-ground discharges. Thus, if the production of NO_x per discharge is proportional to the total energy dissipated, then cloud-to-cloud and cloud-to-ground discharges should produce at least similar amounts of nitrogen oxides. However, it must be recalled that this linear scaling might not be adequate. Moreover, as the mechanism for the formation of NO_x in electrical discharges is not satisfactorily known, we cannot specify the fraction of the total energy dissipation involved in the formation of NO_x.

The mechanism usually assumed to be responsible for NO formation in the hot channel of a lightning discharge, the Zel'dovich mechanism, requires high temperatures and a freeze out mechanism for NO production. The freeze out mechanism is needed in order to achieve a rapid cooling of the channel after reaching temperatures higher than 1500°K, allowing higher equilibrium concentrations of NO than those corresponding to ambient temperature. Let's examine whether or not such mechanism could also be active in cloud flashes. Ground flashes are preceded by leader processes (Uman, 1987). Furthermore, all discharges in air that can be produced in the laboratory indicate the presence of leader activity (Les Renardières Group, 1975, 1978). Also, Brook and Ogawa (1977) show evidence, both photographic and electromagnetic field observations, that indicates the presence of leader like activity before neutralization events in cloud discharges. Thus it is reasonable to assume that even in cloud discharges leader stages are an essential part. The available information shows that even during the leader stage of a ground flash the discharge channel is heated very rapidly (i.e., in a few microseconds) to temperatures as high as 30 000°K (Orville, 1968). During the neutralization stage, the temperature may increase even further. Therefore, it is reasonable to assume that the freeze out mechanism for NO is also active in intracloud discharges and that high temperatures, in excess of 1500°K, are reached.

The volume of air processed at high temperatures must also be considered. Theoretical studies on return strokes in ground flashes have shown that the radius of the heated channel depends on the energy dissipation in the channel (Hill et al., 1980), which, in turn, depends on the time integral of the current. On the other hand, Kowalczyk and Bauer (1982) and others have argued that the estimated currents for cloud discharges are much smaller than the currents estimated for ground discharges. However, a small current may dissipate lots of energy if it flows for a long time. Therefore, considerable amounts of air may be processed in connection with discharges characterized by small currents.

Another aspect that must be considered is that, according to Cooray (1996), an important fraction of the total energy dissipation during a lightning discharge takes place during the leader stages. Therefore, NO_x formation might take place during these stages and not only during the return stroke or recoil streamer stages of cloud-to-ground and cloud discharges, respectively.

In summary, our present understanding of the mechanisms for the formation of NO_x in electrical discharges is not sufficient to elucidate, on physical grounds, the ranking between cloud-to-cloud and cloud-to-ground discharges as NO_x producers. Therefore, it is a matter that warrants further investigation. Also, it is well justified to perform sensitivity analyses on the vertical distribution of the lightning source used in large-scale models of tropospheric oxidized nitrogen and ozone chemistry.

5. Importance of the vertical distribution of the lightning source of NO_x

In order to illustrate the importance of the vertical distribution of the lightning source, we use a global 3-D climatological tracer transport model (MOGUNTIA) for oxidized nitrogen and ozone chemistry.

5.1. Model description

MOGUNTIA is an Eulerian tracer model based on advection by monthly averaged winds and parameterized eddy diffusion (Zimmermann, 1984). The horizontal resolution is 10° latitude × 10° longitude, and the vertical is 10 isobaric levels up to 100 hPa. The time-step used is 2 h. A detailed description of the transport representation in MOGUNTIA can be found elsewhere (Zimmermann, 1984; Langner and Rodhe, 1991). The effects of deep cumulus convection are implemented as a stochastic process (Feichter and Crutzen, 1990). The local losses due to dry deposition are written as a first order process (Zimmermann, 1984) described in terms of the deposition velocity over the surface and the depth of the surface layer. Scavenging of the soluble species is also described as a first order process, characterized by scavenging rate, calculated as a function of the precipitation rate at the surface, the fraction of precipitation released at each level, the thickness of the layer where precipitation occurs and the liquid water content of the precipitating cloud (Langner and Rodhe, 1991). We do not distinguish between stratiform and convective precipitation, and sub-cloud scavenging is not included for nitrogen species. The photochemical scheme used includes the main gas phase reactions for CH_4-CO-O_3-HO_x-NO_x-HNO_4-HNO_3 and the reactions that describe the formation and loss of NO_3 and N_2O_5 (Dentener and Crutzen, 1993). The model code also contains the oxidation chemistry of the most abundant non-methane hydrocarbons ethane and propane leading to the formation of organic nitrates (Kanakidou et al., 1991, 1992). The oxidized nitrogen family NO_y is defined in the model as: $NO_y = NO + NO_2 + NO_3 + 2N_2O_5 + HO_2NO_2 + HNO_3 + PAN + PPN + C1$-C3-organic nitrates. The insoluble oxidized nitrogen reservoir is defined as $NOX = NO_y$-HNO_3.

A set of upper boundary conditions based on calculated or observed concentrations of HNO_3 (Gille et al., 1987) and NO_x (Valentin, 1991) are used. The concentrations of NO_x and HNO_3 are fixed at 100 hPa according to these observed or calculated values. The input of stratospheric ozone into the troposphere is represented by the inclusion of a source of 478 TgO_3 per year distributed according to estimates of the global exchange of mass between the stratosphere and the troposphere by Holton (1990) and ozone observations at 100 hPa by Komhyr et al. (1989).

The model considers NO_x-emissions due to industrial sources, soil exhalation, biomass burning and lightning. We use the same emission scenario as Dentener and Crutzen (1993) except for the industrial emission of NO_x due to fossil fuel combustion and the one due to lightning discharges. We adopted the industrial emission distribution based on the Global Emission Inventory Activity (GEIA) data base (Benkovitz et al., 1996), corrected over the territory of the former Soviet Union according to data by Ryaboshapko et al. (1996, Benkovitz et al., 1996).

The emissions of NO_x due to lightning discharges are done as described by Gallardo et al. (1995). The horizontal distribution of the lightning source is based on statistics of cloud top heights from a long-term integration of a General Circulation Model (the ECHAM GCM, Roeckner et al., 1992), applying a parameterization of lightning activity (Price and Rind, 1992).

The vertical allocation of the source is determined by assuming that the lightning emission of NO_x at each grid point in the vertical is proportional to the local air density (i.e., it is distributed as a constant mixing ratio in the air column). The NO_x-emissions due to cloud-to-ground and cloud-to-cloud lightning are distributed between 1000 and 500 hPa, and between 500 hPa and the tropopause level, respectively. Given the ratio between the number of cloud-to-cloud and cloud-to-ground discharges occurring per unit time, one can express the frequency of cloud-to-cloud and cloud-to-ground discharges in terms of the total frequency of discharges. Earlier observations by Prentice and Mackerras (1977) showed a latitudinal variation of this ratio from about 2 at higher latitudes to about 6 at lower latitudes. Recent works by Price and Rind (1993) and Mackerras and Darveninza (1994) have indicated a less pronounced variation of the ratio between the number of cloud flashes and the number of ground flashes. In this work we have adopted the weighted means given by Mackerras and Darveninza (1994).

Further, in order to obtain a global lightning emission field of NO_x some assumptions regarding the amount of NO_x produced per discharge must be made. We assume that the production yield of cloud-to-cloud flashes is α times the production yield of cloud-to-ground discharges. The scalar α is equal to 0.1 (1.0) if cloud-to-cloud discharges are ten times less (equally) effective per discharge than (as) cloud-to-ground discharges in producing NO_x. Finally, the production yield is obtained by imposing a global annual emission of 5 TgN/yr, based on current estimates. Thus, two emission fields of NO_x from lightning were constructed assuming different vertical partitioning between cloud-to-cloud and cloud-to-ground discharges as NO_x producers. A much larger input in the upper part of the model domain results if the same production yield per flash for both types of discharges is adopted. If α is equal to 0.1, about 20% of the lightning emission emanates from cloud-to-cloud discharges, whereas for equal to 1.0, about 70% does.

5.2. Model sensitivity

Lightning emissions of NO_x are very important for the balance of oxidized nitrogen in the middle and upper troposphere (Lawrence et al., 1994). We illustrate this fact in Figs. 1, 2 that show the ratio between the zonally and annually averaged mixing ratio of NO_y calculated with no lightning emissions, and the corresponding NO_y-field calculated with lightning emissions of oxidized nitrogen. Fig. 1 shows the ratio assuming that cloud discharges are ten times less effective than cloud-to-ground discharges per discharge in producing NO_x ($\alpha = 0.1$). Fig. 2 shows the corresponding ratio assuming that cloud discharges are equally effective as cloud-to-ground discharges per discharge in producing NO_x ($\alpha = 1.0$). Notice that more than 40% of the NO_y in extended areas of the free troposphere in the tropics emanates from lightning. Also, up to 30% of the free tropospheric NO_y in a vast region of the Southern Hemisphere originates from lightning. In the Northern Hemisphere, other sources of NO_x than lightning, mainly fossil fuel combustion, explain 90% or more of the NO_y burden. When cloud flashes are assumed to be as effective as ground flashes in producing NO_x per discharge, the region of largest impact of the lightning source is shifted to upper levels in the model's domain.

Let's now explore the effect the changes in the distribution of oxidized nitrogen due to the assumed vertical distribution of the lightning source have on the budget and distribution of ozone. We consider two cases. In the first case we calculate the annual zonally averaged distribution of ozone assuming that cloud discharges are ten times less effective than cloud-to-ground discharges per discharge in producing NO_x. In the second case, we calculate the corresponding ozone distribution assuming that cloud discharges are equally effective as cloud-to-ground discharges per discharge in producing NO_x. Fig. 3 shows the ratio between the two distributions. The largest changes in the ozone distribution occur in the middle troposphere over the tropics and the subtropics of the Southern Hemisphere, following the geographical pattern of the changes in oxidized nitrogen distribution. The global net photochemical production of ozone is enhanced by 5% when cloud flashes are assumed to be equally effective as cloud-to-ground discharges per discharge in producing NO_x. Notice that these are zonally and annually averaged changes, so even larger changes in ozone concentrations occur regionally during

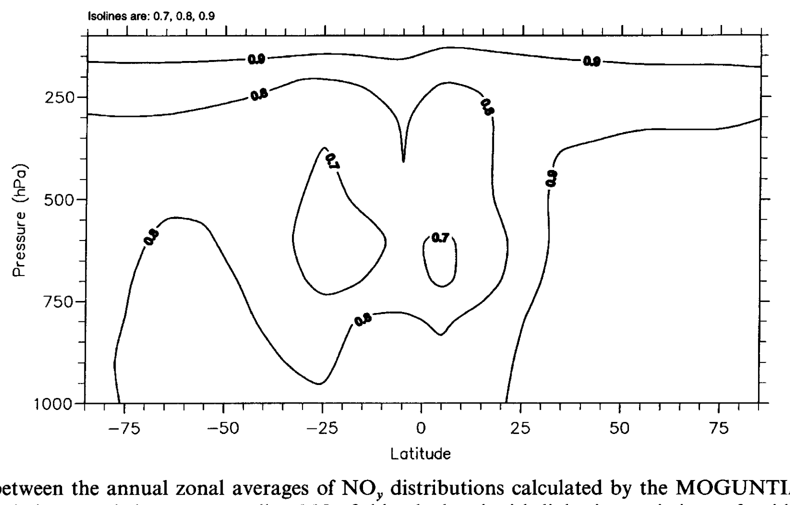

Fig. 1. Ratio between the annual zonal averages of NO_y distributions calculated by the MOGUNTIA model with no lightning emissions, and the corresponding NO_y-field calculated with lightning emissions of oxidized nitrogen: assuming that cloud discharges are ten times less effective than cloud-to-ground discharges per discharge in producing NO_x ($\alpha = 0.1$).

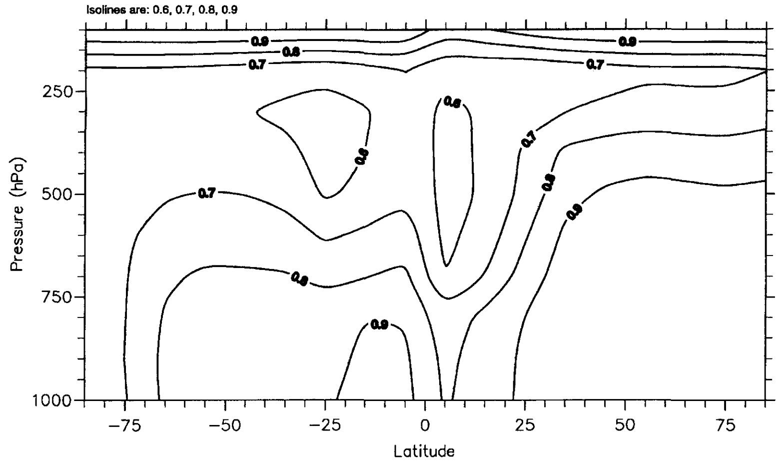

Fig. 2. As Fig. 1, except assuming that cloud discharges are equally effective as cloud-to-ground discharges per discharge in producing NO_x ($\alpha = 1.0$).

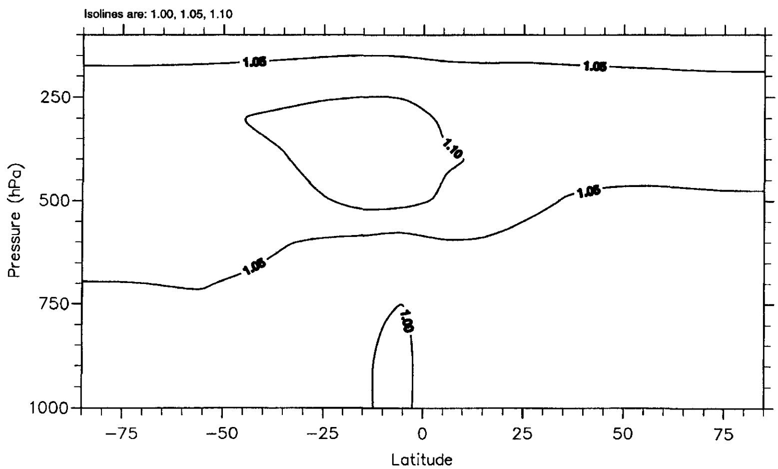

Fig. 3. Ratio between the annual zonal averages of the ozone distributions calculated by the MOGUNTIA model assuming that cloud-to-cloud discharges are equally effective as, or 10 × less effective than cloud-to-ground discharges per discharge in producing NO_x.

the season of highest lightning activity. The troposphere-stratosphere exchange of ozone, followed by the fast exchange in depression systems, is not well resolved in large-scale models and constitutes a major uncertainty in our model simulations of the ozone distribution (Gallardo and Rodhe, 1995; Dentener et al., 1995). A different parameterization of this exchange process might result in a different sensitivity of the model to the vertical distribution of the lightning source. Nevertheless, despite the uncertainties and shortcomings in this model representation, these results illustrate the potential importance of the lightning source of NO_x and its vertical distribution for the budget of oxidized nitrogen and ozone in the troposphere. Moreover, if the emission of NO_x by cloud discharges is increased by assuming $\alpha = 1.0$, the gap between the simulated and the observed NO_x and NO_y mixing ratios in remote sites is diminished (Gallardo and Rodhe, 1995).

Another interesting aspect concerns the strength of the lightning source of NO_x. The model calculations described above were made with a constant total lightning source strength of 5 TgN/yr. Thus, what one sees in Figs. 2, 3, are purely the changes which occur due to changes in the vertical distribution of NO_x emission by lightning. However, if cloud-to-cloud and cloud-to-ground discharges are equally effective per discharge in producing NO_x, the estimate of the strength of the global lightning source should be about two to three times higher than previously believed. This is shown in a simple computation summarized in Table 3. We calculate the annual production of NO_x by cloud-to-cloud and cloud-to-ground discharges in the same way described in Section 5.1. However, for the sake of argument, in the calculation used for Table 3, we normalize the flash frequency distribution obtained to an annual global mean of 100 flashes/s. Further, we assume that the amount of NO_x produced per cloud-to-ground flash is 10^{26} molecules (Noxon, 1976).

Table 3. *Strength of the global lightning source, assuming a global mean lightning frequency of 100 flashes per second, a cloud flash-to-ground flash ratio as described by Mackerras and Darveniza (1994), and that the amount of NO_x produced per cloud-to-ground flash is 10^{26} molecules (Noxon, 1976). The scalar is equal to 0.1 (1.0) if cloud-to-cloud discharges are $10\times$ less (equally) effective per discharge than (as) cloud-to-ground discharges in producing NO_x*

	NO_x produced (TgN/yr)	
Type of discharge	$\alpha=0.1$	$\alpha=1.0$
cloud-to-cloud:		
over land	0.4	3.5
over sea	0.1	1.4
cloud-to-ground:		
over land	1.6	1.6
over sea	0.6	0.6
Total	2.7	7.1

6. Conclusions

Our present understanding of the formation mechanism of NO_x in electrical discharges is not sufficient to determine, on physical grounds, the partitioning between cloud-to-cloud and cloud-to-ground discharges as NO_x producers. Nonetheless, measurements of the optical and acoustic energy spectra of lightning discharges, taken alone, are not sufficient grounds to justify the assumption that cloud-to-cloud discharges are less effective than cloud-to-ground discharges in producing NO_x. A detailed evaluation of the relative importance of both types of discharges as NO_x producers requires a better understanding of the formation mechanism of nitrogen oxides by electrical discharges.

Results from a sensitivity study performed with a global 3-D model of tropospheric oxidized nitrogen, indicate that cloud-to-cloud discharges might be more important than previously assumed. Moreover, if cloud-to-cloud discharges are as effective as cloud-to-ground discharges in producing NO_x per discharge, then the estimate of the strength of the lightning source of NO_x may be 2.6 times larger than previously assumed in 3-D global model studies of oxidized nitrogen.

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