

SNAP CODE:	111000
SOURCE ACTIVITY TITLE:	OTHER SOURCES AND SINKS <i>Lightning</i>
NOSE CODE:	301.10.01
NFR CODE:	N/A

1 ACTIVITIES INCLUDED

Lightning and corona discharge during thunderstorm events cause atmospheric chemical reactions to take place at high voltages and high temperatures. These reactions cause the production of NO_x in the atmosphere. Such production processes are, strictly speaking, not real emissions as the compounds involved (primarily N₂ and O₂) are not injected into the atmosphere but are present anyway. However as these processes can not adequately be described by conventional atmospheric models on one hand, and their impact is eventually identical to those from (anthropogenic) emissions on the other hand, they are easy to be compared on the emission level and thus are frequently treated as such.

2 CONTRIBUTIONS TO TOTAL EMISSIONS

Global NO_x production by lightning has been estimated in the range of 3-5 TgN/yr [1]. For the U.S., 40% of the yearly lightning-generated NO is estimated to be produced during the summer months [2]. Other estimates using the calculation schemes given below indicate that the lightning NO comprises only 3% of the total NO_x emissions budget, with a maximum contribution of 24% at the maximum hour with respect to anthropogenic emissions at a definite period in summer for the U.S. North-East [3].

These figures apply to emissions within the whole troposphere. Emissions in the boundary-layer (circa lowest 1 km) are obviously considerably less. In reporting emissions under the joint EMEP/CORINAIR system, care must be taken to report only emissions between the ground and 1 km - see section 8.

This activity is not believed to be a significant source of PM_{2.5} (as of December 2006).

3 GENERAL

3.1 Description

The electrical discharge of lightning creates plasma channels in the atmosphere characterized by the high fraction of ionic loads and high temperatures. Major compounds of the atmosphere, notably nitrogen, oxygen, and water, may be ionized and then undergo chemical transformation. While the exact pathway of such transformations is largely unknown, a few assessments have been made [4, 5]. A major species to be produced is nitric oxide (NO), but also other compounds containing nitrogen, oxygen and hydrogen atoms are being formed. Crucial for the formation is the high temperature during the flash (up to 30000 K) and the

subsequent rapid cooling below 1500 K which prevents the freshly formed NO from immediate destruction.

3.2 Definitions

Lightning: Atmospheric discharge during thunderstorm events

CG - discharge: Flash starting in the cloud, bringing several coulomb of negative charge to earth within about 0.5 s (negative discharge)

IC - discharge: Flash that does not connect to earth: intracloud, intercloud and cloud-to-air flashes

3.3 Techniques

While lightning exhibits different characteristics depending on whether it is cloud-to-ground (CG), cloud-to-cloud or within-cloud (inter- and intra-cloud, IC), emission estimation techniques have not been resolved to this level of detail. It has been reported that IC discharges may be up to ten times less efficient in producing NO_x than the CG discharges [4]. However, newer information suggests that these discharges may be nearly equal [1, 5]. The amount and distribution of NO produced is believed to be dependent on the energy and the frequency of lightning strokes, which in turn is dependent on cloud temperatures and cloud heights. IC lightning is known to be more frequent than CG lightning. The ratio has been correlated to the cold cloud thickness (cold cloud = below freezing), representing the size of the electric field involved that may determine the number of IC flashes [6]. Despite of generally large variations of this ratio, a dependence on geographical latitude has been found using cold cloud thickness as a parameter.

3.4 Emissions

Out of the compounds being formed in lightning discharges, numbers are given only for NO and NO₂ as NO_x. These emissions seem to be the most relevant.

Differentiation needs to be made for IC and CG lightning, especially with respect to their injections into the atmosphere as relevant for models. IC lightning occurs at altitudes above about 5 km and may be neglected in some boundary layer models, while CG lightning is expected to reach from the ground to about 7 km high (north of 30 deg. Latitude) or 10 km high (south of 30 deg. Latitude). The NO formed is distributed decreasing with height as a function of air density [7]. For a 7 km flash, about 20% of the emissions would then occur in the lowest 1000m, and 80% between ground level and 5 km.

In [3] the IC component -- here only calculated in the detailed methodology -- is assumed to add an extra 21% NO at 60 deg. N and 61% at the equator. All IC flashes are assumed to occur above a height of 5 km.

3.5 Controls

There are no controls to natural emissions by definition.

4 SIMPLER METHODOLOGY

The number of lightning flashes can be obtained from measurements (see section 5), or from estimations. In the latter case, the flashes are estimated from meteorological data on thunderstorm occurrence and from the geographical latitude of the area considered. Support for these estimations may be given by satellite data [8]. Emissions are then calculated according to [3] :

$$CG_{NO} = E \times M$$

where CG_{NO} is the NO produced by the cloud-to-ground part of the lightning flash

$$E = 4 \times 10^8 \text{ J per cloud-to-ground flash and}$$

$$M = 9 \times 10^{16} \text{ molecules NO / J}$$

Calculated in mass units, this yields 2.75 kg NO_x (calculated as NO_2) per flash of lightning. About 20% of this amount is assumed to be emitted below 1 km altitude, 80 % below 5 km altitude.

5 DETAILED METHODOLOGY

The difference of the detailed methodology is that the number of lightning flashes is actually counted and cloud-to-cloud flashes are included. Data for the U.S. are available from the East Coast lightning detection network, or from the lightning strike data archive from Global Atmospheric, Inc. in Tucson, AZ. In many European countries, especially in Western Europe, national networks are operative and may be accessible through the respective national meteorological service. These networks do not include cloud-to-cloud (IC) flashes however.

Emissions now are calculated as [9]:

$$LNO = (N_{CG} \cdot EF_{CG} / \epsilon_{CG}) + [(N_{CG} / \epsilon_{CG}) \cdot (10/(1 + (\Phi/30)^2) - 1)] \cdot EF_{IC}$$

where:

LNO = NO emissions for lightning flashes in study area, molecules NO

N_{CG} = Number of cloud-to-ground (CG) flashes recorded

ϵ_{CG} = Efficiency of the CG network

EF_{CG} = Emission factor for NO for each CG lightning flash

Φ = Latitude of the study area in degrees

EF_{IC} = Emission factor for NO for each inter- or intra - cloud (IC) lightning flash

The emission factors needed for calculation are given in section 8. For the U.S. East Coast, the efficiency has been reported to be 0.7 [9]. The equation takes care of the fact that IC lightning is, dependent on the latitude, about four times as frequent as CG lightning. While recent investigations [6] indicate a less pronounced latitude dependence than the one given here, the results are virtually identical at 40 deg. Latitude.

Emissions from IC flashes are assumed to take place above 5 km altitude only. In contrast, 80% of the CG-lightning emissions are expected at altitudes below 5 km, and 20% even below 1 km.

6 RELEVANT ACTIVITY STATISTICS

Meteorological data on lightning frequencies need to be obtained. Lightning climatologies are being produced or are available in the meteorological offices of many countries. The data may either derive from reporting thunderstorm events, from observation networks, or from satellite information.

7 POINT SOURCE CRITERIA

No point sources.

8 EMISSION FACTORS, QUALITY CODES AND REFERENCES

A large variety of emission factors is given in the literature from laboratory as well as field investigations. According to [10], the low, median and high end of these estimates may be given at 0.36×10^{26} , 4×10^{26} , and 30×10^{26} molecules NO per flash. As indicated in [10] and also discussed in [3], the highest of these figures (from [11]) is not supported by studies modelling nitrate deposition. [1] estimates global NO_x production from a best fit approximation between a global model and observations from regions where lightning is expected to be a major source. This study yields results close to the lowest of the three factors given, thus we recommend to apply the factor of 0.36×10^{26} molecules NO (2.75 kg NO_x) for each flash of lightning as EF_{CG} . Only part of these emissions should be reported: See Table 8.1.

Literature data [4, 9] suggest an emission factor for IC lightning of an order of magnitude lower than for CG lightning. We tentatively are recommending that EF_{IC} be set equal to 0.36×10^{25} molecules NO (275 g NO_x). Recent theoretical considerations [5] however indicate that such a low emission factor might not be realistic. The total energy dissipated in an IC flash should then be at least as high as in GC flashes. Even considering the decreased NO formation at high altitude the EF_{IC} should be considerably higher then (maybe a factor 5). However quantification is missing, and the emissions only concern altitudes above 5 km anyway (where they are relevant primarily on the global scale). Thus an update will only be given at a later stage, when new evidence emerges.

All recommended emission factors are compiled in the following table for the respective altitudes. Note that reporting will only be necessary for lightning emissions up to one km at this stage. The upper layer emissions may be needed at a later stage only.

Table 8.1: Recommended emission factors per flash of lightning in molecules NO and kg NO_x (calculated as NO₂), respectively.

ALTITUDE	EF _{CG} (molecules)	EF _{CG} (kg)	EF _{IC} (molecules)	EF _{IC} (kg)	COMMENT
below 1 km	0.72x10 ²⁵	0.55	0	0	report
1km to 5 km	2.16x10 ²⁵	1.65	0	0	do not report !
above 5 km	0.72x10 ²⁵	0.55	0.36x10 ²⁵	0.275	do not report !
total	3.6 x10 ²⁵	2.75	0.36x10 ²⁵	0.275	do not report !

Because of the uncertainty in the NO production factors, we assume a quality code of D. Additional information on uncertainties can be obtained from [1] and [12].

9 SPECIES PROFILES

Emissions are given for NO_x. While virtually all of the oxidation product is originally NO, a considerable part is transformed to NO₂ very quickly (depending on ozone availability etc.). This part may be in the order of 25% of the original NO [11].

10 UNCERTAINTY ESTIMATES

The uncertainty with the emission factors has been estimated a factor of three, however the validity of these results have to be checked with respect to those literature estimates giving results different by up to an order of magnitude (see [5] and [10]).

11 WEAKEST ASPECTS/PRIORITY AREAS FOR IMPROVEMENT IN CURRENT METHODOLOGY

Depending on the methodology of assessing the emission factors, there are still large discrepancies. These have to be settled before any more detailed estimations can be performed. In addition, the chemical conversion processes in lightning need to be better understood, especially with regard to IC lightning.

12 SPATIAL DISAGGREGATION CRITERIA FOR AREA SOURCES

Spatial disaggregation can be performed according to the distribution of lightning and thunderstorm events.

13 TEMPORAL DISAGGREGATION CRITERIA

Temporal disaggregation should be done according to diurnal and annual cycle of thunderstorm / lightning activity.

14 ADDITIONAL COMMENTS

Lightning is not known to be influenced by humans at all, thus it should be considered as a purely natural source.

15 SUPPLEMENTARY DOCUMENTS**16 VERIFICATION PROCEDURES****17 REFERENCES**

- [1] Levy H., Moxim W., Kasibhatla P. (1996). A global three-dimensional time-dependent lightning source of tropospheric NO_x. *J.Geophys.Res.*, 101, 22911-22922.
- [2] Placet M., Battye R., Fehsenfeld F., Basset G. (1990). Emissions involved in acid deposition processes. NAPAP State of Science and Technology Report 1, Chapter 5, pp. 9-19.
- [3] Novak J., Pierce T. (1993). Natural emissions of oxidant precursors. *Water, Air and Soil Pollution*, 67, 57-77.
- [4] Sisterson D., Liaw Y. (1990). An Evaluation of Lightning and Corona Discharge on Thunderstorm Air and Precipitation Chemistry. *Journal of Atmospheric Chemistry*, 10 (1) 83-96.
- [5] Gallardo L., Cooray V. (1996). Cloud cloud-to-cloud discharges be as effective as cloud-to-ground discharges in producing NO_x? *Tellus*, 48B, 641-651.
- [6] Price C., Rind D. (1993). What determines the cloud-to-ground lightning fraction in thunderstorms? *Geophys.Res.Lett.* 20, 463-466.
- [7] Pierce T., Novak J. (1991). Estimating Natural Emissions for EPA's Regional Oxidant Model. Presented at Environmental Protection Agency/Air and Waste Management Association International Specialty Conference on Emission Inventory Issues in the 1990's, Durham, NC., September 9-12, 1991. 14p.
- [8] Turman B., Edgar B. (1982). Global lightning distribution at dawn and dusk. *J. Geophys. Res.* 87, 1191-1206.
- [9] RADIANT Corp. (1996). EIIP Volume 5, Biogenic sources preferred methods. Final report to the Area Sources Committee, Emission Inventory Improvement Program, May 1996.
- [10] Biazar A, McNider R. (1995). Regional estimates of lightning production of nitrogen oxides. *J.Geophys.Res.* 100, 22861-22874.
- [11] Franzblau E., Popp C. (1989). Nitrogen oxides produced from lightning. *J. Geophys. Res.* 94, 11089-11104.

- [12] Lawrence M., Chameides W., Kasibhatla P., Levy H., Moxim W. (1995) Lightning and atmospheric chemistry: the rate of atmospheric NO production. In: Handbook of Atmospheric Electrodynamics, vol. 1, edited by H. Volland, pp. 189-202, CRC Press, Boca Raton, Florida, USA.

18 BIBLIOGRAPHY

19 RELEASE VERSION, DATE AND SOURCE

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AEA Technology
UK
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20 POINT OF ENQUIRY

Any comments on this chapter or enquiries should be directed to:

David Simpson

Norwegian Meteorological Institute (MSC-W)
c/o IVL, PO Box 47086
Dagjammingsgatan 1
S-402 Goteborg
Sweden

Tel: +46 31 7256214
Fax: +46 31 7256290
Email: david.simpson@ivl.se