Thunderstorms: Generator and exhaust for pollutants

H. Huntrieser

Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, 82234 Wessling, Germany

Thunderstorms and lightning have an important impact on chemistry in the atmosphere, especially on the nitrogen oxides and ozone budget in the upper troposphere. Polluted air masses in the boundary layer may be ingested by thunderstorms, rapidly uplifted, and emitted within the anvil outflow in the upper troposphere. The strongest natural source of nitrogen oxides in the upper troposphere originates however from lightning-induced nitrogen oxides (LNOx). In the past 15 years, we studied LNOx in a number of airborne field experiments accompanied with model simulations. Airborne in situ measurements were performed in thunderstorms over Europe, Brazil, Australia, and West Africa.

1. Introduction

Almost 200 years ago, von Liebig [1] first suggested that lightning plays a major role in the global nitrogen cycle. A flash of lightning induces a hot channel of air. When it expands, we hear thunder. In this hot lightning channel (~30,000 K), the nitrogen (N_2) and oxygen (O_2) molecules of the air are dissociated into single N and O atoms. During the rapid cooling of the channel, these atoms and molecules may recombine to form NO (Zel´dovich mechanism). First airborne measurements in thunderstorms measuring NO were performed by Dickerson et al. [2]. The sum of nitric oxide (NO) and nitrogen dioxide (NO_2), known as mononitrogen oxides (NO_x), is quasi-conserved during the upward transport in thunderstorms.

Up until recently, a global LNOx nitrogen-mass source strength varying from 1-20 Tg a^{-1} was given by the World Meteorological Organization (WMO), whereas for model simulations a value of 5 Tg a^{-1} was frequently used. In a review paper by Schumann and Huntrieser [3] summarizing three decades of research activities on this topic (considering theoretical and laboratory studies as well as surface-, airborne-, and satellite-based measurements), the wide range of the LNOx source strength was slightly reduced. The best estimate of the annual global LNOx source and its uncertainty range was estimated to 5±3 Tg a^{-1}, which is about 10 % of the total NO mass emitted into the atmosphere.

NO plays an important role in the production of ozone (O_3). It acts as a catalyst during the photochemical oxidation of hydrocarbons and carbon monoxide (CO) producing O_3. On the other hand, ozone plays an essential role in atmospheric chemistry since it determines the oxidizing capacity of the troposphere and acts as an important greenhouse gas. Therefore, to increase understanding about the climate impact of NO_x, it is a crucial task to reduce the large uncertainties in estimates of the global LNOx source.

2. Field experiments

Between 1996 and 2006, the Deutsches Zentrum für Luft- und Raumfahrt (DLR) performed a number of airborne field experiments in thunderstorms focusing on LNOx [4-9]. The measurements were carried out over almost all continents (Europe, South America, Australia and Africa). The DLR thunderstorm field experiments mainly consisted of in situ trace gas measurements aboard an aircraft, ground-based measurements with radar monitoring the dynamic evolution and microphysical composition of the thunderstorms, and ground-based measurements with a lightning detection network.

For measurements in the thunderstorm anvil cloud, the DLR research aircraft Falcon is a suitable platform due to its robust construction and its elevated operating altitude up to ~13 km, covering the main anvil outflow region. The standard in situ trace gas instrumentation during these campaigns is mainly used to measure nitrogen oxides, CO, O_3, and carbon dioxide (CO_2), with a high temporal resolution of 1 s for most instruments.

The most suitable tracers indicative of boundary layer air mass transport to the anvil outflow region are CO (with a lifetime of ~2-3 months in the troposphere) and CO_2 (with a lifetime of the order of 30-95 years). For fresh transport from the boundary layer, O_3, with a lifetime in the order of days to months, can partly also be used as tracer. Measurements in thunderstorms have shown that the mixing ratios of tracers in the main outflow region can occasionally be in the same range as measured at the top of the boundary layer, which indicates a very fast and undiluted vertical transport, a “mirroring” of trace gas compositions [5].

3. Results and conclusion

Extensive airborne field experiments performed by DLR in the past few years indicate that most thunderstorms in tropical regions produce less LNOx than previously assumed [7-9]. The overall mean for the annual global LNOx N-mass production rate, estimated from airborne measurements with the DLR in Falcon in selected tropical and subtropical thunderstorms, is ~2 and 4 Tg a^{-1}, respectively. These results indicate a slightly lower global LNOx source strength compared to the value of 5±3 Tg a^{-1} given by Schumann and Huntrieser [3] as the best estimate of the annual global LNOx production rate based on available literature up to that date. In general, thunderstorms in tropical regions have flash rates similar to those observed in subtropical and mid-altitude regions; however, the NO production rate per flash was estimated to be lower. It is suggested that the vertical wind shear may influence the overall flash length by separating the charged regions in the thunderstorms differently. In tropical regions, the predominantly weak wind shear may induce shorter flash lengths in general, resulting in less LNOx production per flash. Furthermore, the ice and graupel volume mass within a thundercloud seem to be crucial parameters for the evolving flash length.

Model simulations indicate that local NO sources in the upper troposphere are distinctly more efficient in producing O_3 compared to sources in the boundary layer [10]. LNOx therefore has an important climate impact, since it contributes up to ~35-40 % of the total greenhouse effect resulting from O_3 production by the different atmospheric NO sources.

4. References

[1] von Liebig, J., 1827: Une note sur la nitrification. *Ann. Chem. Phys.*, **35**, 329-333.

[2] Dickerson, R. R., G. J. Huffman, W. T. Luke, et al., 1987: Thunderstorms: An important mechanism in the transport of air pollutants. *Science*, **235**, 460-465, doi: 10.1126/science.235.4787.460.

[3] Schumann, U. and H. Huntrieser, 2007: The global lightning-induced nitrogen oxides source. *Atmos. Chem. Phys.*, **7**, 3823-3907, doi:10.5194/acp-7-3823-2007.

[4] Huntrieser, H., H. Schlager, C. Feigl, and H. Höller, 1998: Transport and production of NO_x in electrified thunderstorms: Survey of previous studies and new observations at mid-latitudes. *J. Geophys. Res.*, **103**(D21), 28,247–28,264, doi:10.1029/98JD02353.

[5] Huntrieser, H., Ch. Feigl, H. Schlager, et al., 2002: Airborne measurements of NO_x, tracer species, and small particles during the European Lightning Nitrogen Oxides Experiment. *J. Geophys. Res.*, **107**(D11), 4113, doi:10.1029/2000JD000209.

[6] Huntrieser, H., H. Schlager, A. Roiger, M. Lichtenstern, U. Schumann, C. Kurz, D. Brunner, C. Schwierz, A. Richter, and A. Stohl, 2007: Lightning-produced NO_x over Brazil during TROCCINOX: airborne measurements in tropical and subtropical thunderstorms and the importance of mesoscale convective systems. *Atmos. Chem. Phys.*, **7**, 2987-3013, doi:10.5194/acp-7-2987-2007.

[7] Huntrieser, H., U. Schumann, H. Schlager, H. Höller, A. Giez, H.-D. Betz, D. Brunner, C. Forster, O. Pinto Jr., and R. Calheiros, 2008: Lightning activity in Brazilian thunderstorms during TROCCINOX: implications for NO_x production. *Atmos. Chem. Phys.*, **8**, 921-953, doi:10.5194/acp-8-921-2008.

[8] Huntrieser, H., H. Schlager, M. Lichtenstern, et al., 2009: NO_x production by lightning in Hector: first airborne measurements during SCOUT-O3/ACTIVE. *Atmos. Chem. Phys.*, **9**, 8377-8412, doi:10.5194/acp-9-8377-2009.

[9] Huntrieser, H., H. Schlager, M. Lichtenstern, P. Stock, T. Hamburger, H. Höller, K. Schmidt, H.-D. Betz, A. Ulanovsky, and F. Ravegnani, 2011: Mesoscale convective systems observed during AMMA and their impact on the NO_x and O_3 budget over West Africa. *Atmos. Chem. Phys.*, **11**, 2503-2536, doi:10.5194/acp-11-2503-2011.

[10] Dahlmann, K., V. Grewe, M. Ponater, and S. Matthes, 2011: Quantifying the contributions of individual NO_x sources to the trend in ozone radiative forcing. *Atmos. Env.*, **45** (17), 2860-2868, doi: 10.1016/j.atmosenv.2011.02.071.