Comparison between global models and measurements of trace gases during TROCCINOX

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Abstract

Airborne trace gas measurements carried out over southern Brazil during TROCCINOX-1 with the *Falcon* aircraft are compared to results from three global models: ECHAM, MATCH and TM4. The agreement between the models, with different parameterizations for lightning-produced NO_X (=NO+NO_2), and the measurements is investigated along single flight tracks. A new parameterization based on the mass flux in the updrafts [*Grewe et al.*, 2001; *Kurz and Grewe*, 2002] shows promising results in comparison to the more commonly used parameterization based on the cloud top height [*Price and Rind*, 1992]. The most realistic model results for the total amount of lightning-produced NO_X on the global scale were achieved with 5 Tg(N) yr^{-1}.

1. Introduction

Nitrogen oxide plays an important role in the atmosphere since it controls the production and destruction of ozone. For global models the implications of more realistic NO source strengths are therefore of great importance. In the upper troposphere it has been suggested that the largest contribution to the NO budget (1-20 TgN yr^{-1}) comes from lightning-produced NO [e.g. *Huntrieser et al.*, 1998, 2002]. The majority of these estimates are based on measurements carried out in the mid latitudes. However, observations from the LIS and OTD satellites indicate a high lightning frequency in tropical regions. The field measurements carried out over Brazil during TROCCINOX therefore offer an excellent opportunity to improve global estimates of lightning-produced NO_X. Recent simulations with the MATCH model show an increase in upper tropospheric ozone in the range of 30-45% due to lightning-produced NO_X in tropical regions [*Labrador et al.*, 2004].

2. Measurements onboard the Falcon aircraft

During 14 flights in February and March 2004 both the fresh and aged outflow from deep convective cells over southern Brazil was investigated. The anvil outflow was mainly penetrated at flight altitudes between 8.0 and 12.5 km. Airborne in-situ measurements of CO, O_3, NO, NO_y, and J(NO_2) were performed. NO_X was estimated from the photostationary state based on the photochemical equilibrium between NO, NO_2, O_3 and J(NO_2). In *Figure 1 (left)* the average vertical NO_X profile during TROCCINOX is shown. The mean NO_X mixing ratios observed in the outflow from thunderstorms over Brazil (0.1-0.7 nmol mol^{-1}) are in a similar range as observed in the outflow from European thunderstorms, however the mean anvil height is elevated by 1-2 km. The largest differences between the vertical profiles are found in the boundary layer which is much more polluted over Europe.

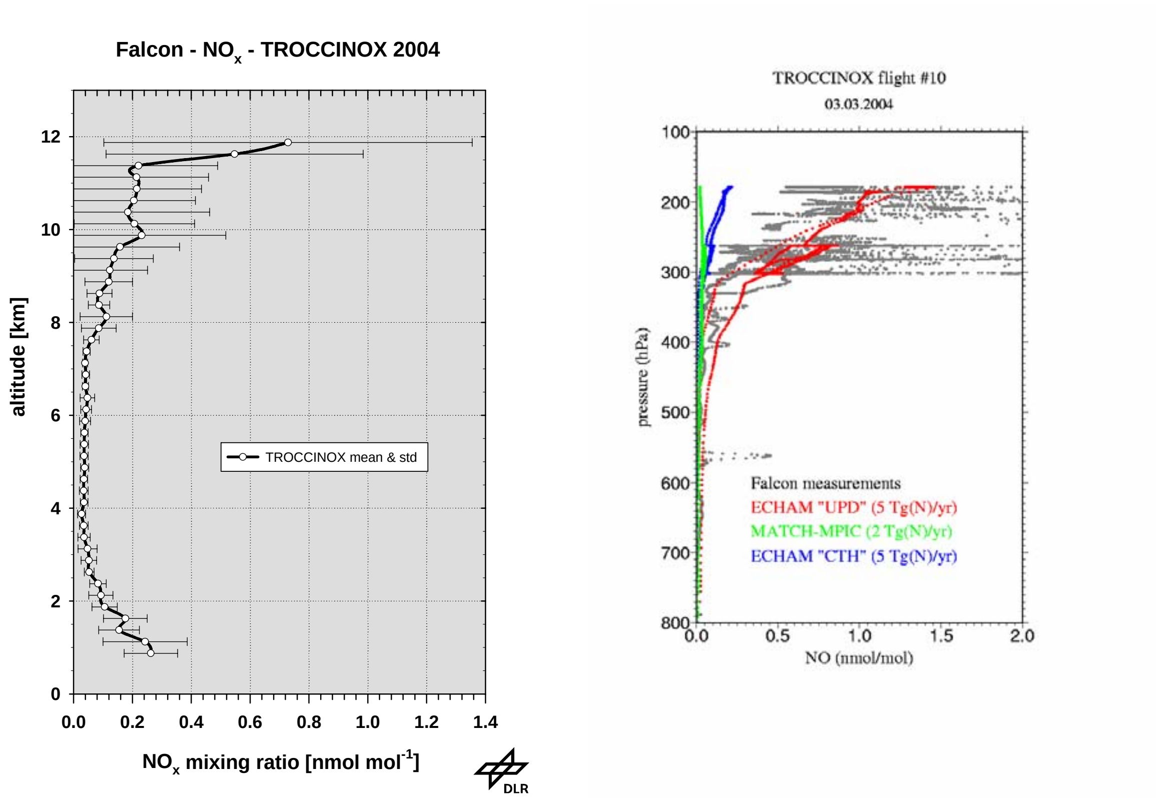

Figure 1. *(left) Mean vertical NO_X profile during TROCCINOX. (right) Vertical NO profiles: Comparison between model results and measurements for flight 10 (March 3, 2004).*

3. Description of models

For the model comparison with measurements three global models were used in this study (*Table 1*). Simulations with the TM4 model (not shown) suggest that the upper tropospheric NO_X budget over Brazil in February 2004 is clearly dominated by the contribution from lightning-produced NO_X.

Table 1.

Model	**Horizontal resolution**	**Vertical resolution**	**Time step**	**Convection scheme**	**Lightning-NO_X parameterization, CTH= cloud top height UPD = updraft**
ECHAM5-MESSy (NUDGE/CHEM) Kurz/Grewe/Jöckel/ Sander, DLR-Oberpfaffenhofen, MPI-Mainz	T63 ~1.9° x 1.9°	19 vertical layers	20 min.	Tiedke convection	CTH or UPD, 5 Tg(N) yr^{-1}
MATCH-MPIC Labrador/Lawrence, MPI-Mainz	T42 ~2.8° x 2.8°	42 vertical layers	30 min.	Zhang convection	CTH, 2 Tg(N) yr^{-1}
TM4 Meijer/Velthoven, KNMI	2° x 3°	32 vertical layers	30 min.	Tiedke convection	convective precipitation, 6.5 Tg(N) yr^{-1}

4. Comparison between model results and measurements

Seven mainly long-range flights were selected for the model-measurements comparison of NO, CO and O_3 mixing ratios (Flight 2, 4, 6, 9, 10, 11, 13). Mean NO mixing ratios (black line in *Fig. 2 upper*, 20 min. mean) during the flights in the upper troposphere varied between 0.1 and 1 nmol mol^{-1} indicating influence from lightning-produced NO_X on some of the days (especially F9-11). In general the MATCH run with 2 Tg(N) yr^{-1} showed too low values for NO in comparison to the measurements (*Fig. 2 upper*). Rather good agreement was achieved with the new ECHAM "UPD" parameterization and 5 Tg(N) yr^{-1}, especially for days with widespread tropical convection like March 3 (Flight 9-10). However, for some flights like Flight 4 a large disagreement between model results and measurements was observed. A closer look to the situation (comparison ECHAM updraft mass flux and cloud position in satellite image, not presented here) showed that the convective system moved faster eastwards than indicated by the ECMWF wind analyse used in the model.

Mean CO mixing ratios (black line in *Fig. 2 lower left*) during the flights in the upper troposphere varied around 100 nmol mol^{-1}. For CO the agreement between the TM4 model and the measurements was excellent in most cases (except Flight 9 and 13). MATCH slightly overestimated CO and the ECHAM model was partly too low and partly too high. During Flight 9 and 13 the CO mixing ratios were enhanced over a wide area (~1 hour of the flight) and reached values between 100-150 nmol mol^{-1}. In addition NO, NO_y and ozone mixing ratios were elevated (1, 3 and 60 nmol mol^{-1}, resp.). A more detailed analysis of Flight 13 (March 7) showed that the large enhancements in the trace gases were mainly caused by emissions from recent fires. Over Paraguay and SW Brazil widespread fires are seen in e.g. MODIS images from March 7 (not shown). At the same time a huge mesoscale convective system (MCS) lifted the pollutants to the upper troposphere which was observed in the *Falcon* measurements in a cloud free area several 100 km downwind of the MCS (not shown). Current burning areas are not included in the models (only climatology) which may explain the large disagreement between models and measurements for some of the flights.

Mean O_3 mixing ratios (black line in *Fig. 2 lower right*) during the flights in the upper troposphere were strongly variable from day to day with values between 25 and 75 nmol mol^{-1}. Concerning O_3 all models show rather large differences to the measurements and in general the model results are too high. The TM4 model is in most cases closest to the measurements. The highest ozone mixing ratios (~75 nmol mol^{-1}) were measured on Flight 11 (March 4) in aged convective outflow from thunderstorms active the previous day.

In addition to the comparison of measurements and model results along the flight tracks also the vertical distributions were compared. In *Fig. 1 right* a comparison for NO is shown. Essential for the parameterization of lightning-produced NO_X is also how it is distributed vertically in the model [e.g. *Pickering et al.*, 1998]. For the selected case a good agreement with the ECHAM "UPD" parameterization and 5 Tg(N) yr^{-1} was achieved.

5. Conclusions

A first comparison between global models and measurements for TROCCINOX flights performed in February and March 2004 has been presented. The measured trace gases (NO, CO, and O_3) show rather large variations from day to day depending on the penetration of deep convective outflow containing lightning-produced NO_X and emissions from fires. During some of the flights widespread areas of enhanced CO, NO and O_3 were observed in the upper troposphere which were not well simulated by the models. It was concluded that most likely emissions from fires, uplifted by MCS over Paraguay and SW Brazil, were transported downwind and penetrated by the aircraft. In near future the behaviour of the models will be studied in detailed sensitivity studies and compared to observed lightning.

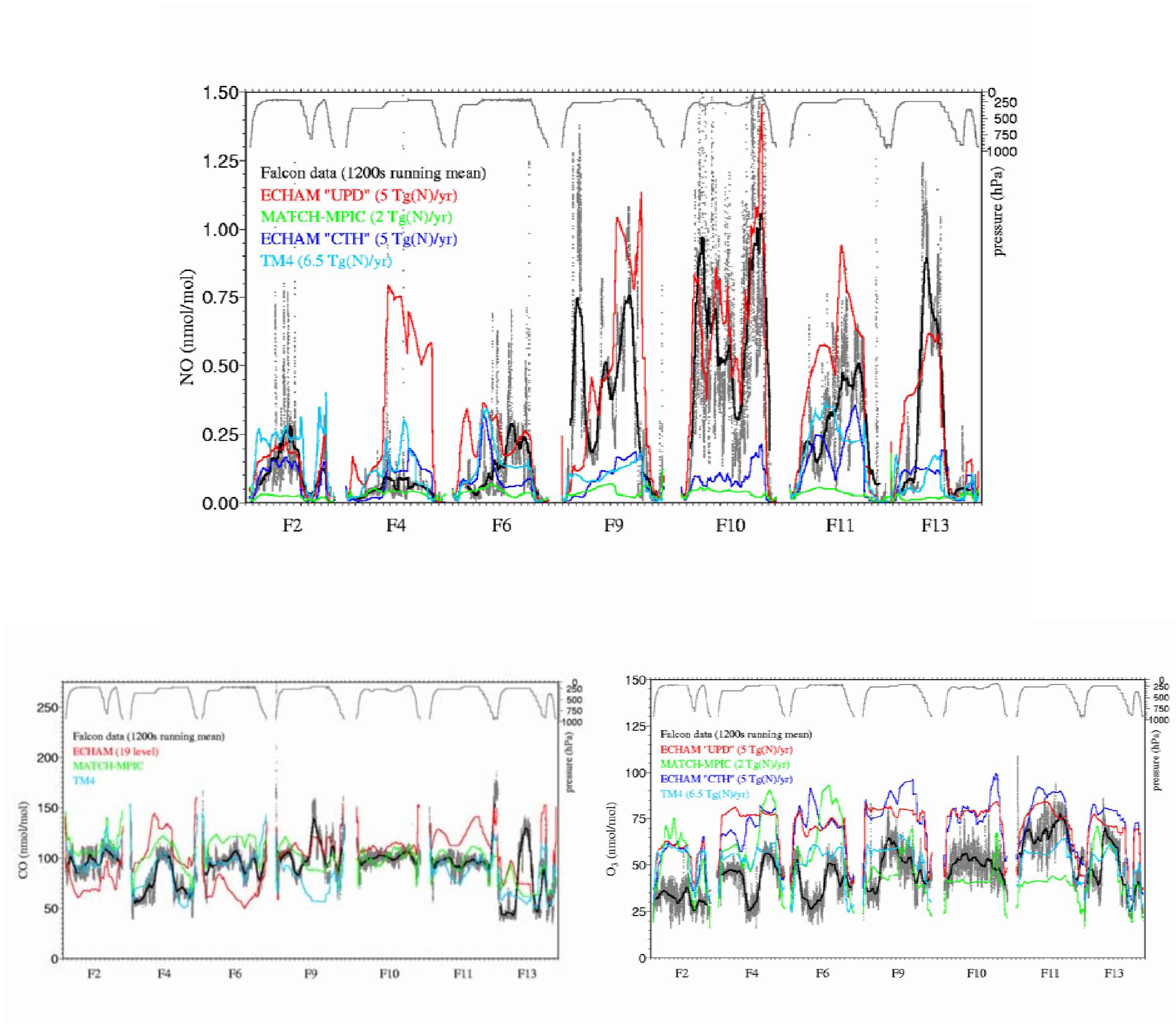

Figure 2. *Comparison between model results (MATCH, ECHAM and TM4) and Falcon trace gas measurements NO (upper), CO (lower left) and O_3 (lower right) for the TROCCINOX flights F2, 4, 6, 9, 10, 11 and 13.*

References

Kurz, C., and V. Grewe, Lightning and Thunderstorms, Part 1: Observational Data and Model Results. *Meteorologische Zeitschrift*, *11*, 6 (10.1127/0941-2948/2002/0011-0379}, 379-393, 2002.

Grewe, V., Impact of Lightning on the Global Chemistry. *8th Scientific Assembly of the IAMAS 2001, International Association of Meteorology and Atmospheric Sciences*, Innsbruck, A, 10-18 July 2001, IAMAS 2001, p. 119, 2001.

Huntrieser, H., H. Schlager, Ch. Feigl, and H. Höller, Transport and production of NO_X in electrified thunderstorms: Survey of previous studies and new observations at midlatitudes, *J. Geophys. Res.*, 103, 28247-28264, 1998.

Huntrieser, H., Ch. Feigl, H. Schlager, F. Schröder, Ch. Gerbig, P. van Velthoven, F. Flatøy, C. Théry, A. Petzold, H. Höller, and U. Schumann, Airborne measurements of NO_X, tracer species and small particles during the European Lightning Nitrogen Oxides Experiment, *J. Geophys. Res.*, 107, doi: 10.1029/2000JD000209, ACH 5-1 - ACH 5-24, 2002.

Labrador, L. J. , R. von Kuhlmann, and M. G. Lawrence, The effects of lightning-produced NO_x and its vertical distribution on atmospheric chemistry: sensitivity simulations with MATCH-MPIC, *Atmos. Chem. Phys. Discuss.*, *Vol. 4*, pp 6239-6281, 6-10-2004.

Pickering, K. E., Y. Wang, W. K. Tao, C. Price, J.-F. Müller, Vertical distributions of lightning NO_X for use in regional and global chemical transport models, *J. Geophys. Res.*, *103*, 31203-31216, 1998.

Price, C., and D. Rind, A simple lightning parameterization for calculating global lightning distributions, *J. Geophys. Res.*, *97*, 9919-9933, 1992.