Global distribution pattern of anthropogenic nitrogen oxide emissions: Comparison and integration of tropospheric satellite observations and model calculations

Nitrogen oxide (NO_x, $NO+NO_2$) plays a key role in tropospheric chemistry, for example as an important precursor of ozone. Its distribution is studied with global chemistry transport models, which need surface emission data as input. A widely-used emission inventory is the EDGAR database, where the anthropogenic emissions are estimated using economic data of the individual countries. This database contains large and mostly unknown uncertainties.

In this thesis, satellite data are used in addition to EDGAR to estimate the geographical distribution pattern of anthropogenic NO_x emissions. Due to the short tropospheric lifetime of NO_x ($\approx$ 1 day), its global distribution is highly correlated to the distribution of the emissions, which allows using measurements of tropospheric NO_2 column densities by the satellite instrument GOME as a proxy for anthropogenic emissions in the areas dominated by those emissions. Two GOME evaluations are used in this thesis, one done by Richter & Burrows (IUP Bremen) and one done by Leue et al. (IUP Heidelberg). As yet another proxy for anthropogenic emissions, calibrated satellite measurements of the nighttime lights of the world (Operational Linescan System (OLS), Defense Meteorological Satellite Program, NGDC) are used.

Within this thesis, a method is developed to calculate pattern errors using the correlation coefficients of at least three fields with independent errors (correlation error analysis). The pattern error of a field is defined here as the ratio of the variance of the error contained in that field to the variance of the total field.

At first, the correlation error analysis is applied to the annual mean values of four NO_2 column density fields in those areas dominated by anthropogenic emissions: Two GOME evaluations and two model calculations done with the global chemistry transport model MOZART. The first model calculation was done using the model's standard emission fields which are based on the EDGAR database; in the second calculation, the anthropogenic NO_x emissions were replaced by a source based on the satellite images of the nighttime lights of the world. Since neither the errors of the two GOME evaluations (same instrument, similar evaluation algorithms) nor the errors of the two model calculations (done by the same model) are independent, only error ranges can be given for the column density fields: The pattern errors of the two model calculations range from 18% to 50%, the pattern error of the GOME evaluation by Richter & Burrows ranges from 0% to 39% and the pattern error of the GOME evaluation by Leue et al ranges from 26% to 55%.

For the correlation error analysis of the emission fields, there are three independent sources available: EDGAR, OLS and GOME. To at least partly eliminate the effect of undirected transport, the GOME fields are deconvoluted and risen

to a higher power. This sharpens the patterns of the satellite measurements when interpreted as emissions. If outliers in the source fields are eliminated before applying the correlation error analysis, the pattern errors of the four fields determined in this thesis read as follows: EDGAR anthropogenic: $(27 \pm 5)\%$, light-based NO_x source: $(26 \pm 5)\%$, NO_x source GOME Richter: $(33 \pm 5)\%$, NO_x source GOME Leue: $(45 \pm 5)\%$. So far, the error estimates for EDGAR were rather rough; the error specifications for the GOME fields can help to improve the retrieval algorithms.

Finally, the four emission fields are combined minimizing the pattern error of the combination field. Assuming that the pattern errors that were determined in the areas dominated by anthropogenic emissions are the same in other regions as well, an anthropogenic NO_x emission field with global coverage can be constructed: In the areas dominated by biogenic emissions, only the anthropogenic EDGAR source and the light-based emissions are combined. In the areas dominated by anthropogenic emissions, all four fields are combined where possible. At grid points where one or two fields show outliers or are undefined, only the other fields are combined. The pattern error of the combination field amounts to $(15 \pm 2)\%$, which is a considerable reduction compared to the pattern errors of the four original fields.

The combination field is unique only up to a constant offset and a constant factor, since only the pattern of that field is fixed by the construction. This offset and factor are chosen relative to the anthropogenic NO_x emissions of EDGAR. With this source field a MOZART model calculation is done. The spatial correlation of the annual mean of the tropospheric column densities of this field with either of the GOME evaluations is higher than that of the original model calculations with either the EDGAR or the light-based NO_x emissions.