Geophysical Research Abstracts, Vol. 7, 03292, 2005
SRef-ID: 1607-7962/gra/EGU05-A-03292

Inverse modelling of spatially resolved NO_x emissions on a continental scale using tropospheric NO_2 columns from satellite measurements

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The recent important developments in satellite measurements of the composition of the lower atmosphere open a challenging perspective to use such measurements as an independent source of information on sources and sinks of atmospheric pollutants. A particular challenge is to retrieve a spatial structure of emissions with a high resolution that would match a spatial resolution of satellite measurements, which currently is of order of several tenths of kilometres. Because of a much lower spatial resolution of global models, such a task necessitates the implication of regional scale chemistry transport models (CTM).

We present a first, to the best of our knowledge, study that resulted in "top-to-down" evaluations of NO_x emissions with a spatial resolution typical for a continental scale CTM (0.5 degree). The study is based on a novel original method which not only enables a computationally efficient optimisation of a large number (1800 in our case) of model parameters corresponding to the seasonally averaged NO_x emissions with an account of transport, but diminishes the amount of a priori assumptions via a combination of satellite and ground based measurements. The key features of our method are (i) the replacement of a CTM by a set of linear regressions describing the relationships between tropospheric NO_2 columns and NO_x emissions with sufficient accuracy, (ii) the utilisation of measurements of near-ground NO_2 concentrations for estimation of a priori unknown ratio of total uncertainties of modelled and measured NO_2 columns to the uncertainty of a priori "down-to-top" estimates of emissions, and (iii) the evaluation of uncertainties of both a priori and a posteriori emissions by means of a special Monte-Carlo experiment which is based on random sampling of errors of both NO_2 columns and surface concentrations. Using CHIMERE CTM, we apply our method to NO_2 columns derived from GOME measurements. GOME data for 2001 were used because surface EMEP measurements were available for the same year. It is found that in spite of significant measurement and model errors, the inverse modelling procedure leads to substantial reduction of the uncertainty of a priori data for NO_x emissions in Western Europe. It is demonstrated also that the use of NO_x emissions derived from GOME measurements enables improvement of the agreement between the modelled and measured near-ground concentrations of NO_2.