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The use of satellite observations for estimating and reducing uncertainties in NO_x emission data employed in an air quality model

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It has been proposed recently [1] that available satellite observations can be used for improving NO_x emission data prescribed in air quality models, which feature much higher spatial resolution than typical global models. Specifically, it has been shown that the use of composite tropospheric NO_2 columns derived from GOME and SCIAMACHY measurements in the inverse modelling scheme may result in considerable (several tenths of percent) reduction of uncertainties in spatial distribution (with the resolution of 0.5 degree) of anthropogenic NO_x emission data for Western Europe.

The main objectives of our new inverse modelling study are (i) improving available NO_x emission estimates for Eastern Europe, Middle East and Northern Africa, (ii) examining the possibilities to improve simulations of near surface ozone concentrations using available satellite data and the inverse modelling technique, (iii) checking consistency of inverse modelling results for Western Europe obtained with different inverse modelling algorithms and different measurement data. We use tropospheric NO_2 columns derived from SCIAMACHY measurements in summer of 2003 and a continental scale chemistry transport model CHIMERE in the framework of an original inverse modelling scheme.

One of the crucial problems of the inverse modelling of atmospheric emissions relates to insufficient information on the uncertainties in observational data, model results and a priori emissions data. Indeed, the inverse modelling consists in specifying observational constraints to the a priori estimates of parameters. The actual “strength” of such constraints is determined by the above uncertainties. If, for example, model errors assumed in an inverse modelling study are underestimated (or neglected), then a posteriori emissions may even be more uncertain than the a priori ones. A key feature of our approach is the self-consistent evaluation of main parameters of the inverse modelling scheme based on observational data. Specifically, in the present study, we estimate the uncertainties of anthropogenic and biogenic emissions by minimising the differences between near surface ozone concentrations simulated by CHIMERE and those measured in the EMEP stations, whereas the total uncertainties in NO_2 columns (due to errors both in the model and measurements) are estimated in parallel as a function of their magnitudes by considering the differences between the simulated and measured tropospheric NO_2 columns.

The results concerning Western Europe confirm major findings of our previous study [1], in which we used different satellite data and employed EMEP measurements of nitrogen dioxide (instead of ozone) concentrations. In particular, the a priori emissions prescribed in CHIMERE based on the EMEP data are probable to be persistently overestimated over Great Britain, Netherlands and northern Germany but underestimated over Spain, southern France and northern Italy. We found also that the uncertainties in a priori emissions for other considered regions are significantly larger than in emissions for Western Europe. In particular, our results indicate that the a priori emissions are strongly underestimated in Middle East and Ural regions. The comparison of a number of statistics (such as RMSE, R^2, empirical regression coefficients, threshold statistics, biases) quantifying the agreement between ozone concentrations measured in EMEP stations and those simulated by CHIMERE with the a priory, a posteriori and randomly perturbed emissions shows unambiguously that the model performance is better with a posteriori emissions, although the relative changes in ozone statistics are not large (from 10 to 20 percent). The magnitudes of the changes are consistent with the sensitivity of ozone concentrations to changes of spatial structure of emissions on “fine” scales in test runs. Much larger improvements in the model performance are expected when NO_x emissions are adjusted together with hydrocarbon emissions, whose uncertainties probably covariate. We expect also that further reduction of uncertainties in NO_x emissions can be obtained after optimising their temporal variations.

1. I.B. Konovalov, M. Beekmann, A. Richter, J.P. Burrows, Inverse modelling of the spatial distribution of NO_x emissions on a continental scale using satellite data, Atmos. Chem. Phys. Discuss., 5, 12641-12695, 2005.

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