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Top-down NOX emissions over European cities from LOTOS-EUROS simulated and OMI observed tropospheric NO_2 columns using the Exponentially Modified Gaussian approach

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High nitrogen oxides concentrations at the surface (NOX = NO + NO_2) impact humans and ecosystem badly and play a key role in tropospheric chemistry. Surface NOX emissions drive major processes in regional and global chemistry transport models (CTM). NOX contributes to the formation of acid rain, act as aerosol precursors and is an important trace gas for the formation of tropospheric ozone (O_3). Via tropospheric O_3, NOX indirectly affects the production of the hydroxyl radical which controls the chemical lifetime of key atmospheric pollutants and reactive greenhouse gases.
High NOX emissions are mainly observed in polluted regions produced by anthropogenic combustion from industrial, traffic and household activities typically observed in large and densely populated urban areas. Accurate NOX inventories are essential, but state-of the- art emission databases may vary substantially and uncertainties are high since reported emissions factors may differ in order of magnitude and more.
To date, the modelled NO_2 concentrations and lifetimes have large associated uncertainties due to the highly non-linear small-scale chemistry that occurs in urban areas and uncertainties in the reaction rate data, missing nitrogen (N) species and volatile organic compounds (VOC) emissions, and incomplete knowledge of nitrogen oxides chemistry. Any overestimation in the chemical lifetime may mask missing NOX chemistry in current CTM's. By simultaneously estimating both the NO_2 lifetime and concentrations, for instance by using the Exponentially Modified Gaussian (EMG), a better surface NOX emission flux estimate can be obtained.
Here we evaluate if the EMG methodology can reproduce the emissions input from the tropospheric NO_2 columns simulated by the LOTOS-EUROS (Long Term Ozone Simulation-European Ozone Simulation) CTM model. We apply the EMG methodology on LOTOS-EUROS simulated tropospheric NO_2 columns for the period April-September 2013 for 21 selected European urban areas under windy conditions (surface wind speeds > 3 m s-1). We then compare the top-down derived surface NOX emissions with the 2011 MACC-III emission inventory, used in the LOTOS-EUROS model as input to simulate the NO_2 columns. We also apply the EMG methodology on OMI (Ozone Monitoring Instrument) tropospheric NO_2 column data, providing us with real-time observation-based estimates of midday NO_2 lifetime and NOX emissions over 21 European cities in 2013.
Results indicate that the top-down derived NOX emissions from LOTOS-EUROS (respectively OMI) are comparable with the MACC-III inventory with a R2 of 0.99 (respectively R2 = 0.79). For St-Petersburg and Moscow the top-down NOX estimates from 2013 OMI data are biased low compared to the MACC-III inventory which uses a 2011 NOX emissions update.