



OMI-derived European NO_x emissions in WRF-Chem: impacts on summertime surface ozone

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Ozone is a secondary air pollutant that is of concern to human and ecosystem health. Regional air quality models are used to predict ozone concentrations and to assess its environmental impacts. These models however suffer from unexplained biases over Europe. The formation of ozone in European summer critically depends on nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$), which catalyze the oxidation of reactive hydrocarbons that leads to ozone production. We hypothesize that uncertainties in NO_x emissions contribute to the underestimation of peak ozone concentrations in European summers. We tested this by calculating satellite-constrained NO_x emissions based on NO_2 columns from the OMI instrument.

We set up a simulation with the regional coupled meteorology-chemistry model WRF-Chem for July 2015 over Europe. An evaluation against in situ observations from the Airbase network shows that mean ozone and its spatial distribution are reproduced satisfactorily. However, similar to previous studies, our WRF-Chem setup underestimates the spread in observed maximum daily 8-hour averaged ozone concentrations (MDA8 O_3), a metric for peak ozone concentrations. Particularly, MDA8 O_3 values exceeding $86 \mu\text{g m}^{-3}$ are underestimated. WRF-Chem simulated surface and column NO_2 generally show an underestimation compared to observations in regions with relatively small fossil fuel and industrial emissions, which suggests underestimated emissions from (agricultural) soils.

We derived top-down surface NO_x emissions using a mass-balance approach based on emission-scaling using the relative difference between OMI and WRF-Chem NO_2 columns. The procedure also accounts for feedbacks through OH, NO_2 's dominant daytime oxidant. This resulted in 67% higher NO_x emissions across Europe. A simulation with these increased NO_x surface emissions reduced the model's mean bias with respect to average NO_2 concentration by $0.39 \mu\text{g m}^{-3}$ (50%), and improved the coefficient of determination from 0.70 to 0.86. Monthly averaged simulated ozone locally increases by $>10 \mu\text{g m}^{-3}$ in areas where NO_x emissions increase strongly. With respect to the a priori simulation, simulated MDA8 O_3 has an improved spatial distribution expressed by an increase in r^2 from 0.40 to 0.52 and reduced mean bias (-14.2 to $-7.8 \mu\text{g m}^{-3}$). The largest increases are seen for high MDA8 O_3 values, but their magnitudes remain underestimated.

Overall, our findings demonstrate that assessments of European summer air quality, in particular surface ozone concentrations, benefit from constraining surface NO_x emission estimates by satellite NO_2 column observations.