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Emission rate and chemical state estimation by 4-dimensional variational inversion

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Abstract. This study aims to assess the potential and limits of an advanced inversion method to estimate pollutant precursor sources mainly from observations. Ozone, sulphur dioxide, and partly nitrogen oxides observations are taken to infer source strength estimates. As methodology, the four-dimensional variational data assimilation technique has been generalised and employed to include emission rate optimisation, in addition to chemical state estimates as usual objective of data assimilation. To this end, the optimisation space of the variational assimilation system has been complemented by emission rate correction factors of 19 emitted species at each emitting grid point, involving the University of Cologne mesoscale EURAD model. For validation, predictive skills were assessed for an August 1997 ozone episode, comparing forecast performances of pure initial value optimisation, pure emission rate optimisation, and joint emission rate/initial value optimisation.

Validation procedures rest on both measurements withheld from data assimilation and prediction skill evaluation of forecasts after the inversion procedures. Results show that excellent improvements can be claimed for sulphur dioxide forecasts, after emission rate optimisation. Significant improvements can be claimed for ozone forecasts after initial value and joint emission rate/initial value optimisation of precursor constituents. The additional benefits applying joint emission rate/initial value optimisation are moderate, and very useful in typical cases, where upwind emission rate optimisation is essential. In consequence of the coarse horizontal model grid resolution of 54 km, applied in this study, comparisons indicate that the inversion improvements can rest on assimilating ozone observations only, as the inclusion of NO_x observations does not provide additional forecast skill. Emission estimates were found to be largely independent from initial guesses from emission inventories, demonstrating the potential of the 4D-var method to infer emission rate improvements. The study also points to the need for improved horizontal model resolution to more efficient use of NO_x observations.

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