Global NO$_x$ emission estimates using OMI NO$_2$ data and ensemble-based data assimilation

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A data assimilation system has been developed to analyze global NO$_x$ emissions using OMI tropospheric NO$_2$ column data and a global chemical transport model CHASER. The data assimilation system employs an ensemble Kalman filter approach to analyze the surface emission of NO$_x$. OMI tropospheric NO$_2$ column data retrieved at KNMI with its averaging kernel and retrieval error information are used to optimize daily NO$_2$ emissions at every model grid point with a horizontal resolution of 2.8 degree, during the year 2005-2006. A super observation approach is employed to fill spatial-scale gaps between the model and OMI data, which provides representative data for data assimilation. In the data assimilation system, surface emissions and its ensemble perturbations are analyzed using OMI data during data assimilation step, providing an ensemble of emissions used for next forecast step. Then, concentrations of chemical species are simulated using the updated emission at forecast step. This approach allows to accumulate observation information through data assimilation cycle and to represent indirect relationship (caused by chemical and transport processes) between the emission and tropospheric column concentration.

The data assimilation results are validated by comparing analyzed NO$_2$ concentrations with independent data, tropospheric columns obtained from SCIAMACHY and vertical profiles obtained during INTEX-B aircraft campaign. We have confirmed that spatiotemporal variations in NO$_2$ concentration are more realistically represented by the data assimilation than the model simulation, by correcting a priori emission obtained based on bottom-up estimations. The data assimilation tends to increase NO$_x$ emissions over Southern and Western Europe, Eastern China, Eastern and Southern USA, and Central Africa, and to decrease it over Northern and Eastern Europe, and Japan, during both winter and summer. OnF (or OmA) analysis revealed great reductions in both bias and mean difference by the data assimilation, especially over polluted areas. Comparisons with vertical profiles measured from INTEX-B imply that the assimilation of OMI tropospheric NO$_2$ column data provides a distinct improvement in the NO$_2$ analysis with a slight improvement in the ozone analysis in the lower troposphere. These results demonstrate improvements in NO$_x$ emissions by the data assimilation. Moreover, the augmentation of chemically related species (e.g., ozone, HNO$_3$) into a state vector and/or multi-species data assimilation may be able to further improve the chemical balance of analyzed fields and NO$_x$ emission estimates. The impacts of augmenting various species and of assimilating OMI NO$_2$ data obtained from different retrievals and other chemical species data (e.g., satellite ozone data) will be further discussed.