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Atmos. Chem. Phys., 5, 1157-1186, 2005

www.atmos-chem-phys.net/5/1157/2005/

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Inversion of CO and NO_x emissions using the adjoint of the IMAGES model

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Abstract. We use ground-based observations of CO mixing ratios and vertical column abundances together with tropospheric NO₂ columns from the GOME satellite instrument as constraints for improving the global annual emission estimates of CO and NO_x for the year 1997. The agreement between concentrations calculated by the global 3-dimensional CTM IMAGES and the observations is optimized using the adjoint modelling technique, which allows to invert for CO and NO_x fluxes simultaneously, taking their chemical interactions into account. Our analysis quantifies a total of 39 flux parameters, comprising anthropogenic and biomass burning sources over large continental regions, soil and lightning emissions of NO_x, biogenic emissions of CO and non-methane hydrocarbons, as well as the deposition velocities of both CO and NO_x. Comparison between observed, prior and optimized CO mixing ratios at NOAA/CMDL sites shows that the inversion performs well at the northern mid- and high latitudes, and that it is less efficient in the Southern Hemisphere, as expected due to the scarcity of measurements over this part of the globe. The inversion, moreover, brings the model much closer to the measured NO₂ columns over all regions. Sensitivity tests show that anthropogenic sources exhibit weak sensitivity to changes of the a priori errors associated to the bottom-up inventory, whereas biomass burning sources are subject to a strong variability. Our best estimate for the 1997 global top-down CO source amounts to 2760 Tg CO. Anthropogenic emissions increase by 28%, in agreement with previous inverse modelling studies, suggesting that the present bottom-up inventories underestimate the anthropogenic CO emissions in the Northern Hemisphere. The magnitude of the optimized NO_x global source decreases by 14% with respect to the prior, and amounts to 42.1 Tg N, out of which 22.8 Tg N are due to anthropogenic sources. The NO_x emissions increase over Tropical regions, whereas they decrease over Europe and Asia. Our inversion results have been evaluated against independent observations from aircraft campaigns. This comparison shows that the optimization of CO emissions constrained by both CO and NO₂ observations leads to a better agreement between modelled and observed values, especially in the Tropics and the Southern Hemisphere, compared to the case where only CO observations are used. A posteriori estimation of errors on the control parameters shows that a significant error reduction is achieved for the majority of the anthropogenic source parameters, whereas biomass burning emissions are still subject to large errors after optimization. Nonetheless, the constraints provided by the GOME measurements allow to reduce the uncertainties on savanna

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burning emissions of both CO and NO_x, suggesting thus that the incorporation of these data in the inversion yields more robust results for carbon monoxide.

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