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The influence of natural and anthropogenic secondary sources on the glyoxal global distribution

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Abstract. Glyoxal, the smallest dicarbonyl, which has recently been observed from space, is expected to provide indications on volatile organic compounds (VOC) oxidation and secondary aerosol formation in the troposphere. Glyoxal (CHOCHO) is known to be mostly of natural origin and is produced during biogenic VOC oxidation. However, a number of anthropogenically emitted hydrocarbons, like acetylene and aromatics, have been positively identified as CHOCHO precursors. The present study investigates the contribution of pollution to the CHOCHO levels by taking into account the secondary chemical formation of CHOCHO from precursors emitted from biogenic, anthropogenic and biomass burning sources. The impact of potential primary land emissions of CHOCHO is also investigated. A global 3-dimensional chemistry transport model of the troposphere (TM4-ECPL) able to simulate the gas phase chemistry coupled with all major aerosol components is used.

The secondary anthropogenic contribution from fossil fuel and industrial VOCs emissions oxidation to the CHOCHO columns is found to reach 20–70% in the industrialized areas of the Northern Hemisphere and 3–20% in the tropics. This secondary CHOCHO source is on average three times larger than that from oxidation of VOCs from biomass burning sources. The chemical production of CHOCHO is calculated to equal to about 56 Tg y^{-1} with 70% being produced from biogenic hydrocarbons oxidation, 17% from acetylene, 11% from aromatic chemistry and 2% from ethene and propene. CHOCHO is destroyed in the troposphere primarily by reaction with OH radicals (23%) and by photolysis (63%), but it is also removed from the atmosphere through wet (8%) and dry deposition (6%). Potential formation of secondary organic aerosol through CHOCHO losses on/in aerosols and clouds is neglected here due to the significant uncertainties associated with the underlying chemistry. The global annual mean CHOCHO burden and lifetime in the model domain are estimated to be 0.02 Tg (equal to the global burden seen by SCIAMACHY over land for the year

2005) and about 3 h, respectively. The model results are compared with satellite observations of CHOCHO columns. When accounting only for the secondary sources of CHOCHO in the model, the model underestimates CHOCHO columns observed by satellites. This is attributed to an overestimate of CHOCHO sinks or a missing global source of about 20 Tg y^{-1}. Using the current primary emissions of CHOCHO from biomass burning together with the anthropogenic combustion sources of about 7 Tg y^{-1} leads to an overestimate by the model over hot spot areas.

Final Revised Paper (PDF, 2731 KB) Supplement (285 KB) Discussion Paper (ACPD)

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