Towards a more complete quantification of the global carbon cycle

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Supplemental Information

Calculating the Transfer of Organic Compounds to the Oceans

Here we have estimated the production of organic carbon compounds originating from partial oxydation of methane and non-methane volatile organic compounds (NMVOCs) and their transfer to the oceans. We have calculated dry and wet deposition of the volatile organic compounds (VOCs) simulated in a global chemistry-climate model. We used the National Institute of Water and Atmospheric Research – United Kingdom Chemistry and Aerosol (NIWA-UKCA) model (O'Connor *et al*., 2014; Zeng *et al*., 2015), which contains a fairly detailed VOC oxidation methanism. Dry deposition follows the formulation of Wesely (1989), and wet deposition depends on rainfall rates and Henry's Law coefficients (Giannakopoulos *et al*., 1999).

We have adopted the model simulations used in the study by Zeng *et al.* (2015), which includes primary emissions of carbon monoxide (CO), ethane (C_2H_6), ethene (C_2H_4), propane (C_3H_8), propene (C_3H_6), formaldehyde (HCHO), acetaldehyde (CH_3CHO), acetone (CH_3COCH_3), methanol (CH_3OH), isoprene (C_5H_8), monoterpene ($C_{10}H_{16}$), and a lumped compound accounting for other NMVOCs that are not explicitly represented in the chemical mechanism. In this simulation, methane mixing ratios at the surface are prescribed at 1758 ppb, which is equivalent to annual emissions of ~450 MtC yr^{-1}. Isoprene and monoterpene emissions are 448 and 126 MtC yr^{-1}, respectively, and emissions of other VOCs account for ~155 MtC yr^{-1}. The simulation is based on the conditions in 2006.

For species that are subject to dry and wet deposition, we partitioned the ocean flux as follows. We used the modelled global distribution of dry deposition fluxes to the Earth's surface of each species, and accounted for deposition to the ocean using the model's land-sea mask information. Total dry deposition to the land and the ocean were then calculated by integrating the respective fluxes over the land and the ocean. In the model version used here, wet depositon fluxes were output as zonally averaged 2-dimensional fields. Therefore, we needed to partition the global wet deposition fluxes to the ocean using 3-dimensional global distributions of the species and weighted them by the global distribution of total precipitation rates.

For comparison, there have been previous studies of the global methanol budget that derived a wide range of possible deposition estimates (see Jacob *et al*., 2005 and references therein), due mainly to the large uncertainties in terrestrial methanol sources. Those models used estimated total terrestrial emissions of 114–320 Mt(CH_3OH) yr^{-1}, which resulted in total dry deposition of 24–120 Mt(CH_3OH) yr^{-1}, of which 0.3–50 Mt(CH_3OH) yr^{-1} were estimated to have been deposited on the oceans. In addition, total wet deposition was estimated to be 9–50 Mt(CH_3OH) yr^{-1}, which was not partitioned between ocean and the land deposition in these studies.The terrestrial methanol emissions used in our model simulation (103 Mt(CH_3OH) yr^{-1}, or 39 MtC yr^{-1}) was therefore near the lower end of the estimates used in previous studies, while the estimated source from atmospheric production was 34 Mt(CH_3OH) yr^{-1}, or 13 MtC yr^{-1}, which is similar to the estimates obtained by Jacob *et al.* (2005). Our deposition estimates are also comparable to the estimates of Jacob *et al.* (2005) after taking account of the different total terrestrial emission estimates used in the two studies.

References

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