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Constraints on OH in a global 3D inversion of methyl chloroform

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The hydroxyl radical (OH) is the primary atmospheric oxidant. In this role, OH is involved in the removal of a wide variety of atmospheric pollutants and greenhouse gases. Despite the central role of OH in atmospheric chemistry, important metrics such as interannual variability and trends in OH on large spatial scales remain poorly constrained. This is mainly due to its low abundance and short lifetime of seconds.

Over the past decades, the anthropogenically emitted methyl chloroform (MCF) has been uniquely qualified as a tracer to indirectly constrain OH on large spatio-temporal scales. However, recent box model studies have shown that OH, as estimated from MCF observations, is still very uncertain^{1,2,3}. This translates for example to large uncertainties in global methane (CH₄) emissions, even if changes in the global CH₄ burden are well-defined. Box model studies however, do not fully capitalize on the MCF measurement network and the gradients therein. Moreover, they may introduce biases due to incorrect or incomplete representation of atmospheric transport.

Here, we present results from a 4DVAR inversion of MCF over the 1998-2018 period, performed in the 3D chemistry-transport model TM5. Starting from typical OH priors, we find adjustments in the OH spatio-temporal distribution that bring the simulated MCF mole fractions closer to observations. Large uncertainties in this improved estimate remain, but we find that no large interannual variability (>2%) and no significant trend in global mean OH are needed to match MCF observations. We do find significant adjustments in the latitudinal gradients of OH (e.g. an increase in tropical OH).

¹ Rigby, M., et al. PNAS (2017), 114.21: 5373-5377

² Turner, A.J., et al. PNAS (2017), 114.21: 5367-5372

³ Naus, S et al. ACP (2019), 19.1: 407-424