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Atmos. Chem. Phys., 4, 693-699, 2004

www.atmos-chem-phys.net/4/693/2004/

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The impact of anthropogenic chlorine emissions, stratospheric ozone change and chemical feedbacks on stratospheric water

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Abstract. Mixing ratios of water (H₂O) in the stratosphere appear to increase due to increased input of H₂O and methane from the troposphere and due to intensified oxidation of CH₄ in the stratosphere, but many of the underlying mechanisms are not yet understood. Here we identify and quantify three chemical mechanisms which must have led to more efficient oxidation of CH₄ in the stratosphere over the past several decades: 1) The increase in stratospheric chlorine levels due to anthropogenic CFC emissions, 2) the thinning of the stratospheric ozone column and 3) enhanced OH levels in the stratosphere due to increasing H₂O levels themselves. In combination with the increase in tropospheric CH₄ mixing ratios and with solar cycle related variations of upper stratospheric ozone, these effects can explain about 50% of the additional conversion of CH₄ to H₂O as observed throughout the stratosphere. The relative contributions from the individual processes have varied over the past decades.

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Citation: Röckmann, T., Grooß, J.-U., and Müller, R.: The impact of anthropogenic chlorine emissions, stratospheric ozone change and chemical feedbacks on stratospheric water, Atmos. Chem. Phys., 4, 693-699, 2004. [Bibtex](#) [EndNote](#) [Reference Manager](#)

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