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ACE-FTS and MIPAS observations of phosgene ($COCl_2$) and comparisons with SLIMCAT chemical transport model calculations

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The majority of chlorine in the atmosphere has arisen from anthropogenic emissions of 'organic' species such as chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs). Due to their long lifetimes, many of these species reach the stratosphere where they break down, liberating chlorine which catalyses the destruction of ozone. The principal degradation products of Cl-containing organic species are carbonyl chloride (phosgene, $COCl_2$), carbonyl chloride fluoride (COClF), and hydrogen chloride (HCl). Of these, phosgene is probably the most notorious, having been used as a chemical weapon in World War I. In the lower stratosphere, where the phosgene mixing ratios peak, the principal sources are the photolysis of carbon tetrachloride (CCl_4) and, to a lesser extent, methyl chloroform (CH_3CCl_3). Smaller contributions arise from very short-lived substances such as CH_2Cl2, $CHCl_3$ and C_2Cl_4. Due to the success of the Montreal Protocol in phasing out the use of CCl_4 and CH_3CCl_3, the abundance of phosgene continues to fall. Observing and understanding phosgene in the stratosphere helps us better understand the chlorine budget, and particularly the atmospheric removal of CCl_4, which has attracted particular interest recently on account of the inconsistency between observations of its abundance and estimated sources and sinks.

This work presents global distributions and trends of $COCl_2$ using data from two satellite limb instruments: the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS), and the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). The ACE-FTS instrument, on board the SCISAT satellite, has been recording solar occultation spectra through the Earth's atmosphere since 2004 and continues to take measurements with only minor loss in performance. ACE-FTS time series are available for a range of chlorine 'source' gases, including CCl_3F (CFC-11), CCl_2F_2 (CFC-12), CHF_2Cl (HCFC-22) and CCl_4, and the chlorine 'product' gases $COCl_2$, COClF and HCl. The MIPAS instrument, onboard ENVISAT (ENVIronmental SATellite), recorded atmospheric limb emissions spectra between 2002 and 2012, with time series available for the key Cl-containing species except HCl. ACE-FTS and MIPAS phosgene observations are compared with the output of SLIMCAT, a state-of-the-art offline three-dimensional chemical transport model (CTM), which contains a detailed treatment of stratospheric chemistry, including the major species in the O_x, NO_y, HO_x, F_y, Cl_y, and Br_y chemical families.