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Atmospheric Lifetimes of Halogenated Hydrocarbons: Laboratory Measurements and Improved Estimations from an Analysis of Modeling Results.

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Reactions with hydroxyl radicals and photolysis are the main processes dictating a compound's residence time in the atmosphere for a majority of trace gases. In case of very short-lived halocarbons their reaction with OH dictates both the atmospheric lifetime and active halogen release. Therefore, the accuracy of OH kinetic data is of primary importance for the comprehensive modeling of a compound's impact on the atmosphere, such as in ozone depletion (i.e., the Ozone Depletion Potential, ODP) and climate change (i.e., the Global Warming Potential, GWP), each of which are dependent on the atmospheric lifetime of the compound.

Atmospheric modeling provides total lifetimes for a number of compounds as well as their partial lifetimes due to specific photochemical removal process (reactions with OH in the troposphere, reactions with OH in the stratosphere, reactions with $O(^1D)$, and UV photolysis), and partial lifetimes associated with the atmospheric removal regions (troposphere and stratosphere). We have analyzed these results in an effort to find a correlation useful for estimating the lifetimes of other atmospheric trace gases based only on laboratory data of their photochemical properties. Based on this analysis, we suggest an improved semi-empirical approach for deriving a "best" value of the total atmospheric lifetime due to photochemical removal processes based on laboratory derived photochemical properties of a compound, which is consistent with both empirically derived tropospheric lifetime of Methyl Chloroform and results of rigorous atmospheric modeling. These aspects will be illustrated in this presentation for a variety of atmospheric trace gases.

The ability to conduct high accuracy laboratory determinations of OH reaction rate constants over the temperature range of atmospheric interest, thereby decreasing the uncertainty of input kinetic data to 2-3% will be demonstrated as well.