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Tropospheric photooxidation of CF₃CH₂CHO and CF₃(CH₂)₂CHO initiated by Cl atoms and OH radicals

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Abstract. The absolute rate coefficients for the tropospheric reactions of chlorine (Cl) atoms and hydroxyl (OH) radicals with CF₃CH₂CHO and CF₃(CH₂)₂CHO were measured as a function of temperature (263–371 K) and pressure (50–215 Torr of He) by pulsed UV laser photolysis techniques. Vacuum UV resonance fluorescence was employed to detect and monitor the time evolution of Cl atoms. Laser induced fluorescence was used in this work for the detection of OH radicals as a function of reaction time. No pressure dependence of the bimolecular rate coefficients, k_{Cl} and k_{OH} , was found at all temperatures. At room temperature k_{Cl} and k_{OH} were (in 10⁻¹¹ cm³ molecule⁻¹ s⁻¹): $k_{\text{Cl}}(\text{CF}_3\text{CH}_2\text{CHO}) = (1.55 \pm 0.53)$; $k_{\text{Cl}}(\text{CF}_3(\text{CH}_2)_2\text{CHO}) = (3.39 \pm 1.38)$; $k_{\text{Cl}}(\text{CF}_3\text{CH}_2\text{CHO}) = (0.259 \pm 0.050)$; $k_{\text{Cl}}(\text{CF}_3(\text{CH}_2)_2\text{CHO}) = (1.28 \pm 0.24)$. A slightly positive temperature dependence of k_{Cl} was observed for CF₃CH₂CHO and CF₃(CH₂)₂CHO, and $k_{\text{OH}}(\text{CF}_3\text{CH}_2\text{CHO})$. In contrast, $k_{\text{OH}}(\text{CF}_3(\text{CH}_2)_2\text{CHO})$ did not exhibit a temperature dependence over the range investigated. Arrhenius expressions for these reactions were:

$$k_{\text{Cl}}(\text{CF}_3\text{CH}_2\text{CHO}) = (4.4 \pm 1.0) \times 10^{-11} \exp\{-(316 \pm 68)/T\} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$$

$$k_{\text{Cl}}(\text{CF}_3(\text{CH}_2)_2\text{CHO}) = (2.9 \pm 0.7) \times 10^{-10} \exp\{-(625 \pm 80)/T\} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$$

$$k_{\text{OH}}(\text{CF}_3\text{CH}_2\text{CHO}) = (7.8 \pm 2.2) \times 10^{-12} \exp\{-(314 \pm 90)/T\} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$$

The atmospheric impact of the homogeneous removal by OH radicals and Cl atoms of these fluorinated aldehydes is discussed in terms of the global atmospheric lifetimes, taking into account different degradation pathways. The calculated lifetimes show that atmospheric oxidation of CF₃(CH₂)_xCHO are globally dominated by OH radicals, however reactions initiated by Cl atoms can act as a source of free radicals at dawn in the troposphere.

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