

INCREASING ATMOSPHERIC CONCENTRATIONS OF  
LONG-LIVED HALOCARBONS AND  
METHANE (ABSTRACT)

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A great concern has been indicated again about the increasing atmospheric concentrations of long-lived anthropogenic halocarbons regarding the "ozone hole" observed recently over Antarctica, and the increase in atmospheric methane concentration has also received a great attention because of its "greenhouse effect" in addition to the effect by  $\text{CO}_2$ . We have been measuring atmospheric concentrations of those halocarbons ( $\text{CCl}_2\text{F}_2$ ,  $\text{CCl}_3\text{F}$ ,  $\text{CH}_3\text{CCl}_3$ , etc.) and methane ( $\text{CH}_4$ ) in Antarctica as well as in the Northern Hemisphere (N.H.) in order to clarify behaviors of these compounds in the atmosphere and to estimate future trends of concentrations and their effects on the earth's environment.

Surface-level samples in Antarctica have been collected (since 1982) at 500–1000 m NE of Syowa Station into all-stainless steel sample canisters which had been prepared in extremely clean conditions and evacuated for grab-sampling. All the Antarctic samples have been analyzed after 3–15 months at University of Tokyo by ECD gaschromatography (for halocarbons) and by FID gaschromatography (for methane). Averaged concentrations in the mid-latitude N.H. have been obtained (since 1979) by analyzing the samples collected in Hokkaido ( $42\text{--}45^\circ\text{N}$ ) every summer (July–August) and winter (January–February).

The  $\text{CCl}_2\text{F}_2$  and  $\text{CCl}_3\text{F}$  concentrations observed in Antarctica have been 8–10% lower than those observed in Hokkaido in accordance with the predominant emission of these halocarbons in the N.H. and their delayed diffusion into the Southern Hemisphere across the Intertropical Convergence Zone. The concentration of  $\text{CH}_3\text{CCl}_3$  has been about 30% lower in Antarctica in accordance with its relatively short atmospheric lifetime (about 6 years) due to the reaction with tropospheric OH radicals.

The atmospheric concentrations of  $\text{CCl}_2\text{F}_2$  and  $\text{CCl}_3\text{F}$  have been increasing steadily (almost linearly) in both hemispheres (by 4–5% every year). These increments of  $\text{CCl}_2\text{F}_2$  and  $\text{CCl}_3\text{F}$  concentrations correspond to their unchanged or rather increasing releases in the world (total exceeding 700 kt/year) and their extremely long lifetimes in the atmosphere exceeding 80 years. The total Cl concentration in the troposphere controls the amount of Cl introduced into the stratosphere and its present concentration in the atmosphere is calculated to be 3000–3500 pptv with 1000–1500 pptv increase per decade. Under these situations, substantial depletion of stratospheric ozone is anticipated in the future.

The methane concentration was found to increase about 1% a year with small seasonal variations. If the current increase of  $\text{CH}_4$  concentration continues, it will cause a large greenhouse effect (about 1/3 of  $\text{CO}_2$ ), and a large depletion of tropospheric OH radical concentration thus leading to the prolonged lifetimes of many anthropogenic air pollutants (including  $\text{CH}_3\text{CCl}_3$ ) susceptible to OH attack.

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