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## 日本上空の成層圏下部におけるメタン長期トレンド Long-term trend of methane concentration in the lower stratosphere over Japan

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Stratospheric methane acts as an important source of stratospheric water vapor and also a sink for chlorine radical. For predicting the future of the stratospheric chemistry and dynamics, as well as for evaluating strategies for limiting or reducing future emission of methane into the atmosphere, it is indispensable to figure out the past methane trend in both of stratosphere and troposphere accurately. Systematic collections of stratospheric air samples have been carried out over Japan since 1985, using a balloon-borne cryogenic sampler. The air samples collected were analyzed for the CH<sub>4</sub> and N<sub>2</sub>O concentrations since 1988. Almost linear and compact relationship between CH<sub>4</sub> and N<sub>2</sub>O concentrations was found for all observations. It is well known that tropospheric N<sub>2</sub>O has been secularly increasing quite monotonously in recent decades. On the other hand, increase rate of tropospheric CH<sub>4</sub> has been varied complicatedly. Despite the fact that both concentrations have been increased independently in troposphere, there is no significant difference between the correlations of stratospheric CH<sub>4</sub> and N<sub>2</sub>O concentrations in each year, at first glance. Considering this fact, the compact relationship obtained over Japan suggests that both CH<sub>4</sub> and N<sub>2</sub>O are destroyed at similar rate in the lower stratosphere during the poleward transport of stratospheric air, although the chemical destruction processes of the two gases are quite different. Therefore, we employed N<sub>2</sub>O-depressions, instead of N<sub>2</sub>O concentrations, for examining correlations with CH<sub>4</sub> concentrations. This method cancels the effects of secular N<sub>2</sub>O increase in stratosphere and enables us to detect possible change in stratospheric CH<sub>4</sub>. Thus we found that stratospheric CH<sub>4</sub> shows a significant increase before 2000 and clear stagnation after 2000. We categorized CH<sub>4</sub> concentration data into different N<sub>2</sub>O depression ranges, and calculated increase rates by applying the curve fitting procedure, taking into account the age of stratospheric air. Average increase rates were calculated to be about 0.4 and 0.1%/year before and after 2000, respectively, in the lower stratosphere. This rate is comparable with the results of 207 ppbv increase in the period of 1978 to 2003 in the lower stratosphere reported by Rohs et al.(2006). However, increase rate before 2000 obtained in this study is much smaller than the result of 1.95 and 0.87 %/year in 1985 and 1994, respectively, by ATMOS/ACE-FTS measurements (Rinsland et al., 2009).

キーワード: メタン, 成層圏, テレンド

Keywords: methane, stratosphere, trend