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日本上空の成層圏下部におけるメタン濃度と炭素同位体比の長期変化
Long-term changes of CH4 concentration and its carbon isotopic ratio in the lower stratosphere over Japan

菅原 敏 [1*] ; 森本 真司 [2] ; 梅澤 拓 [3] ; 青木 周司 [2] ; 中澤 高清 [2] ; 石戸谷 重之 [4] ; 豊田 栄 [5] ; 本田 秀之 [6]
SUGAWARA, Satoshi[1*] ; MORIMOTO, Shinji[2] ; UMEZAWA, Taku[3] ; AOKI, Shuji[2] ; NAKAZAWA, Takakiyo[2] ; ISHIDOYA, Shigeyuki[4] ; TOYODA, Sakae[5] ; HONDA, Hideyuki[6]

[1] 宮城教育大学, [2] 東北大院理大気海洋センター, [3]Max Planck Institute for Chemistry, [4] 産業技術総合研究所, [5] 東京工業大学, [6] 宇宙科学研究所
[1]Miyagi Univ. of Education, [2]CAOS, Tohoku Univ., [3]Max Planck Institute for Chemistry, [4]AIST, [5]Tokyo Institute of Technology, [6]ISAS/JAXA

It is expected that d13C of CH4 provides us with useful information not only about CH4 emissions from biogenic and abiogenic sources but also about its oxidation process in the atmosphere. Therefore, measurements of d13C have been carried out for the major CH4 sources as well as for the background atmosphere. However, the measurements are still insufficient for elucidating the CH4 cycle on the earth's surface. In the stratosphere, CH4 is destroyed by reactions with OH, O(1D) and Cl atom. These destruction processes play an important role in the stratospheric chemistry, but the respective contributions to the CH4 loss and their temporal changes have not been yet well understood quantitatively. Measurements of the isotopic ratios of the stratospheric CH4 are one of the most promising methods to detect possible change of the CH4 destruction processes in the stratosphere on the basis of the different isotopic fractionations occurring in the different reactions. However, only a few measurements have been made so far, due mainly to difficulty of collecting air samples in the stratosphere. Systematic collections of stratospheric air samples have been carried out over Japan since 1985 using a balloon-borne cryogenic sampler. We analyzed the air samples collected in the period of 1994-2010 for concentrations of CH4, N2O, CO2 and SF6, and d13C of CH4. In this study, we report the preliminary results of the long-term change of d13C of CH4 in the stratosphere. Almost linear and compact relationships between CH4 and N2O concentrations were found for the all observations in the different years. CH4 concentration and d13C also showed compact relationships in the lower stratosphere, although those in the mid-stratosphere were less correlated. The tight correlations between CH4 and N2O in spite of the different destruction processes suggest that the ratio of both destruction rates has been kept as almost constant during the transport process in the stratosphere. It is well known that tropospheric CH4 and N2O have been secularly increasing in the recent decades. Such increasing trends should have been propagated into the stratosphere, and the compact relationships between the stratospheric CH4 and N2O would change depending on their increase rates. To elucidate an inter-annual changes of the stratospheric CH4 and its d13C, we employed N2O-loss, instead of the N2O concentration, as an indicator of how the chemical reactions have proceeded during the stratospheric transport. The N2O-loss was calculated as a concentration difference between the tropical troposphere and the stratosphere by considering the mean age of air estimated from CO2 and SF6 concentrations. This procedure eliminates the effect of the secular N2O increase from the relationships between CH4 and N2O, and enables us to detect possible change in the stratospheric CH4. As a result, we found that the CH4 concentration increased at a rate of 4.5+-0.9 ppbv/year in the lower stratosphere during 16 years. This increase rate is consistent with those observed in the troposphere. The same technique was applied to the correlations between CH4 concentration and d13C, and we found no significant changes of d13C in the lower stratosphere. Considering the fact that d13C in the troposphere also does not show a clear trend in a recent decade, our result implies that the relative contributions of the CH4 destruction processes have been unchanged in the lower stratosphere over the observed period.

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