

Time and space variations of the O₂/N₂ ratio observed over the western North Pacific using a cargo aircraft C-130H

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The atmospheric O₂/N₂ ratio ($d(O_2/N_2)$) has been observed at many ground-based stations since the early 1990s to elucidate the global CO₂ budget (e.g. Manning and Keeling, 2006), however, airborne observations of $d(O_2/N_2)$ in the free troposphere are very limited (e.g. Ishidoya et al., 2012). In this study, the air samples collected using a cargo aircraft C-130H in service between Atsugi Base (35.45 N, 139.45 E) and Minamitorishima (MNM; 24.28 N, 153.98 E) have been analyzed for $d(O_2/N_2)$, Ar/N₂ ratio ($d(Ar/N_2)$), $d^{15}N$ of N₂, $d^{18}O$ of O₂ and $d^{40}Ar$ to clarify time and space variations of the $d(O_2/N_2)$ in the mid-troposphere.

The observations onboard the C-130H are conducted once per month, and 24 air samples are collected into 1.7 L Titanium flasks during the level flight at an altitude about 6 km and descent toward MNM (Tsuboi et al., 2012). 6 air samples are also collected into the similar flasks at the ground surface in MNM around the same time period with the C-130 H observation. The total of 30 air samples are analyzed for CO₂, CH₄, N₂O and CO concentration at Japan Meteorological Agency, then analyzed for $d(O_2/N_2)$, $d(Ar/N_2)$, $d^{15}N$ of N₂, $d^{18}O$ of O₂ and $d^{40}Ar$ at AIST since May 2012.

The $d^{15}N$ of N₂, $d^{18}O$ of O₂ and $d^{40}Ar$ are known to be almost constant in the troposphere, and the $d(Ar/N_2)$ shows slight seasonal cycle at the ground surface with the peak-to-peak amplitude of 10-30 per meg (e.g. Casser et al., 2008). However, the $d(Ar/N_2)$, $d^{15}N$ of N₂, $d^{18}O$ of O₂ and $d^{40}Ar$ from the C-130H observations were found to be significantly different from the surface values at MNM. Especially, the $d(Ar/N_2)$ of the air samples collected during the level flight were higher by about 800 per meg than the surface values. Such the large variations in the $d(Ar/N_2)$ were considered to be due to some sort of the artificial fractionations of Ar and N₂. Therefore, we examined the relationships between $d(Ar/N_2)$, $d^{18}O$ of O₂ and $d^{40}Ar$ and $d^{15}N$ of N₂ to clarify the cause of the fractionation. The obtained relationships were highly consistent with those expected from the fractionation due to the thermal diffusion (Ishidoya et al., 2013), which would be attributed to the branching of flow paths (e.g. Bender et al., 2005) of the ambient air supplied from the jet engine to pressurize the cabin of the C-130H. Taking these facts into consideration, we corrected the $d(O_2/N_2)$ obtained from the C-130H observation for the fractionation by using an experimentally-determined relationship of the $d(Ar/N_2)/d(O_2/N_2)$ due to the thermal diffusion and the measured values of the $d(Ar/N_2)$. Because the $d(Ar/N_2)/d(O_2/N_2)$ ratio due to the thermal diffusion and the measurement precision of the $d(Ar/N_2)$ were about 4.5 and +5 per meg, respectively, the uncertainty of $d(O_2/N_2)$ associated with the correction was estimated to be about +1 per meg. This uncertainty is smaller enough than +4.8 per meg (+1 ppm) of the precision required for the precise observation of the atmospheric $d(O_2/N_2)$. Therefore, it is suggested that variations of the $d(O_2/N_2)$ in the free troposphere are observable using the C-130H by applying the correction method.

The corrected $d(O_2/N_2)$ and CO₂ concentration varied seasonally almost in opposite phase at all heights. The average Atmospheric Potential Oxygen (APO = O₂ + 1.1 x CO₂) (Stephens et al., 1998) during the observation period (May - December 2012) decreased with increasing altitude, which implied the net O₂ outgassing from the ocean around MNM for the period. In the presentation, we will also discuss the characteristic variations of the $d(O_2/N_2)$ observed in the free troposphere based on the analyzed results of comparisons of the $d(O_2/N_2)$ with the CO₂, CH₄, N₂O and CO concentration as well as the backward trajectories for the observation dates. The correction method employed in this study will make it possible to observe the $d(O_2/N_2)$ precisely using air samples collected without special sampling techniques to reduce the fractionations of the molecules.

Keywords: aircraft observation, atmospheric O₂/N₂ ratio, Atmospheric Potential Oxygen (APO), correction method for fractionation of O₂ and N₂