Time and space variations of Atmospheric Potential Oxygen over the Western North Pacific observed by using a cargo aircraft C-130H

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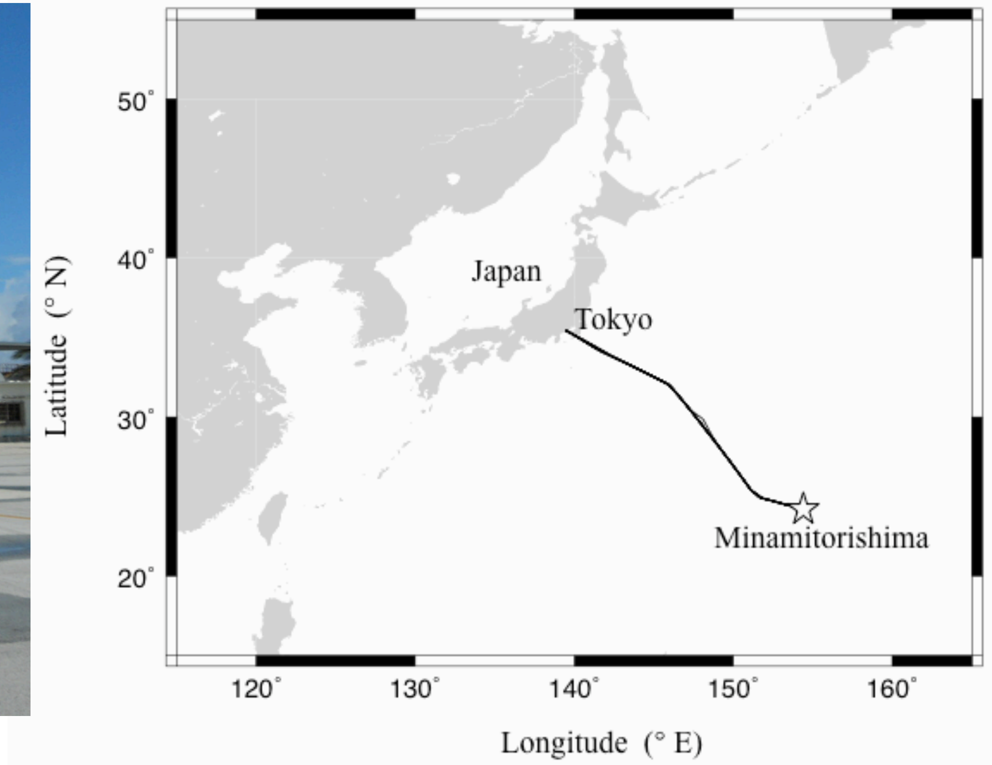
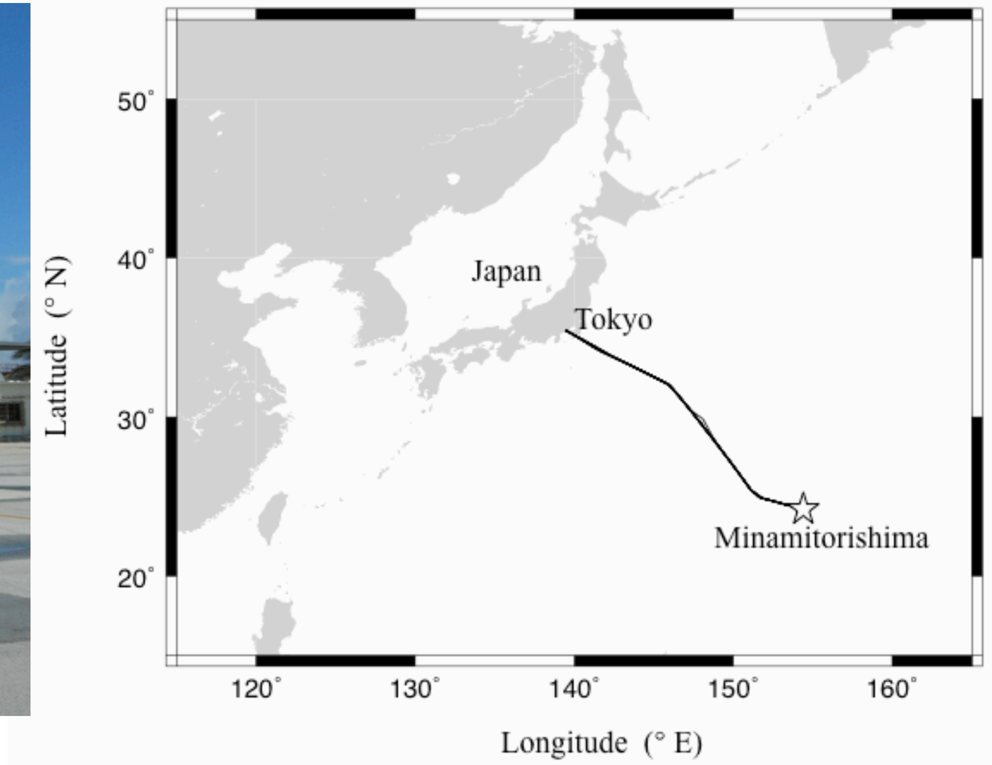
Collection of middle tropospheric air samples over the western North Pacific



- Flight : once per month from Atsugi Base (35.45°N, 139.45°E), Kanagawa, Japan, to Minamitorishima (MNM; 24.28°N, 153.98°E).
- 24 air samples are pressurized into 1.7 L titanium flasks whose inner walls are silica-coated to an absolute pressure of 0.4 MPa.
- A set of 17–20 samples are collected during the level flight (about 6 km a.s.l.) and others are obtained during the descent portion at MNM.

Collection of middle tropospheric air samples over the western North Pacific



- CO_2, CH_4, N_2O and CO concentrations were analyzed at Japan Meteorological Agency (JMA) (Tsuboi et al. 2013 AMT; Niwa et al., 2014 JMSJ)
- <u>$\delta(O_2/N_2)$, $\delta(Ar/N_2)$, stable isotopic ratios of N_2, O_2 and Ar ($\delta^{15}N$, $\delta^{18}O$ and $\delta^{40}Ar$) were analyzed at National Institute of Advanced Industrial Science and Technology (AIST)</u> (Ishidoya & Murayama, 2014 Tellus B; Ishidoya et al. 2014 SOLA)

Collection of middle tropospheric air samples over the western North Pacific

Topics:

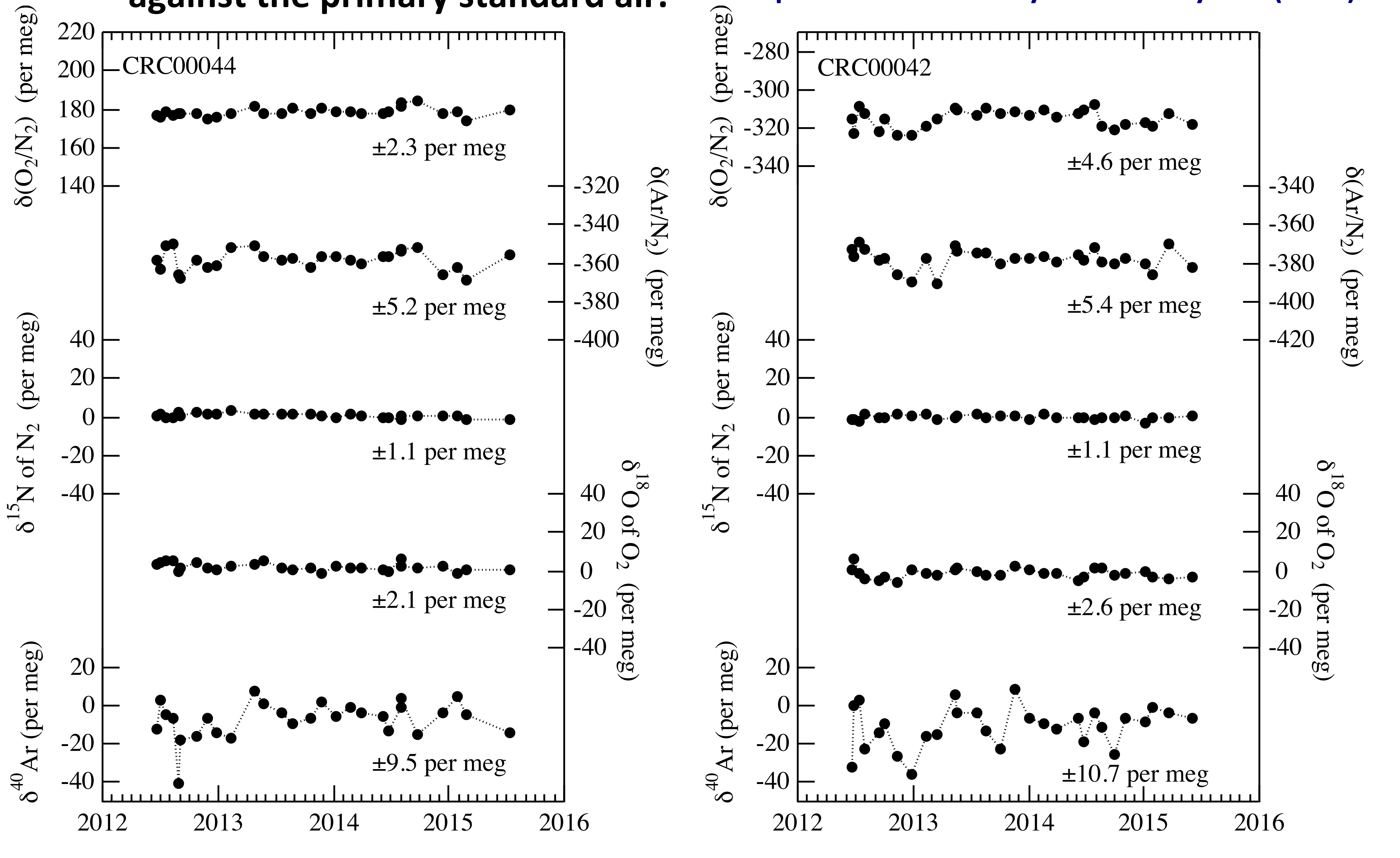
- **Correction of the thermally fractionation superimposed on $\delta(O_2/N_2)$**
- **Latitudinal difference seen in seasonal APO cycles**
- **Altitudinal gradient of annual average APO over MNM**
- **Latitudinal gradient of annual average APO**

Updated from Ishidoya et al. (2014)

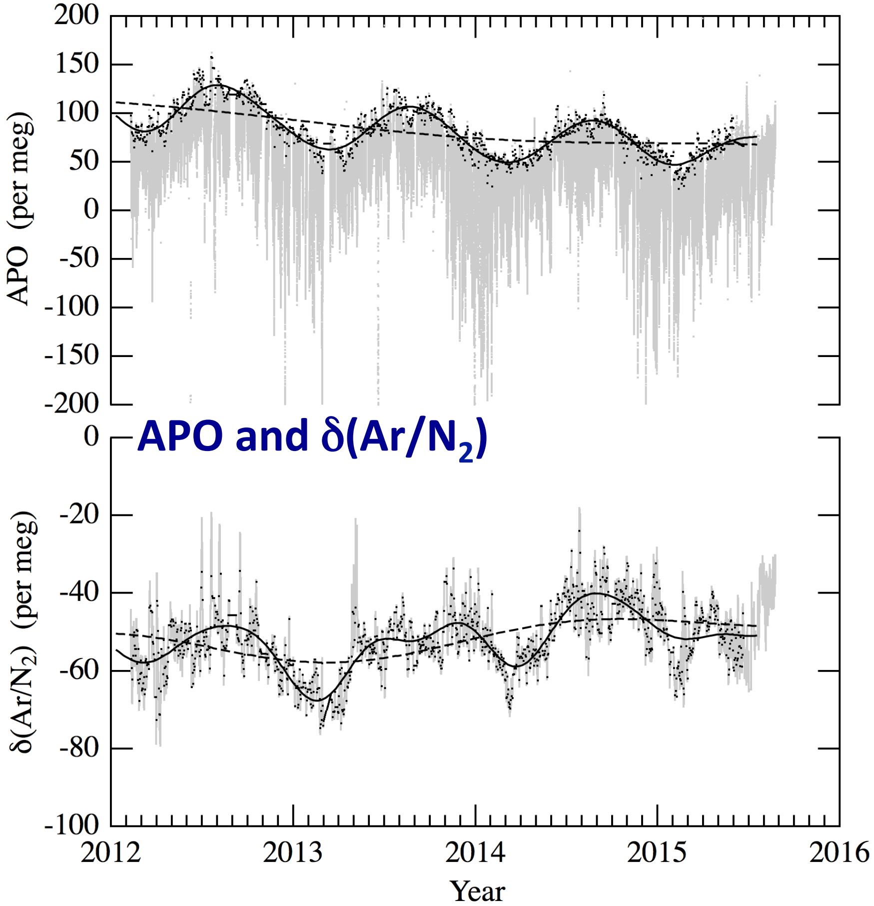
Measurement system of O_2/N_2 and Ar/N_2 ratios, CO_2 concentration and stable isotopic ratios of N_2, O_2 and Ar (the slides presented in GGMT)

Temporal changes in measured values of the working standard air against the primary standard air.

Updated from Ishidoya and Murayama (2014)



Preliminary observational results of O_2/N_2, CO_2 and Ar/N_2 ratios at Tsukuba, Japan (the slides presented in GGMT)



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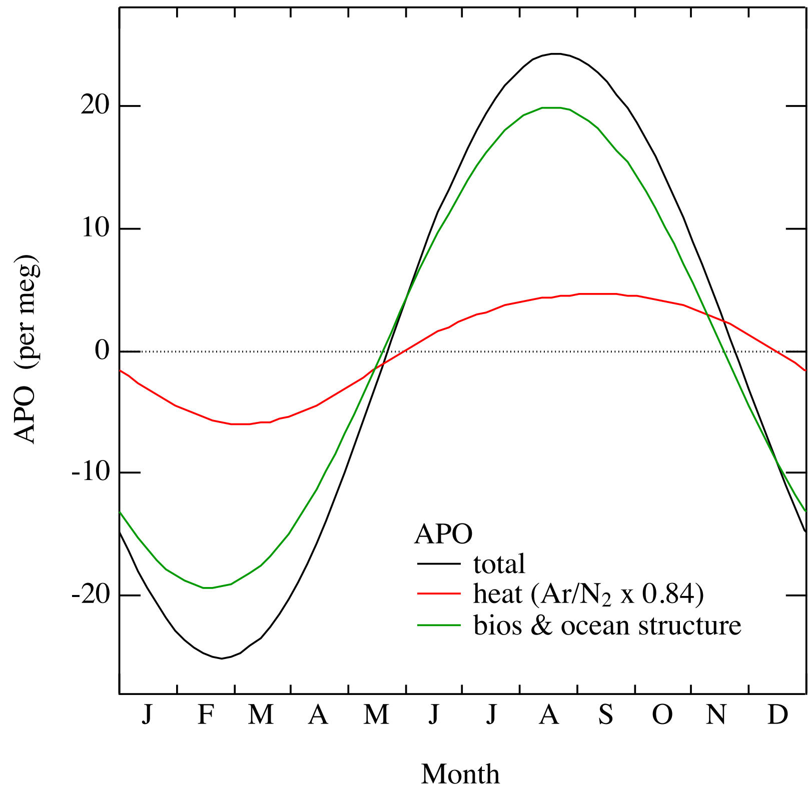
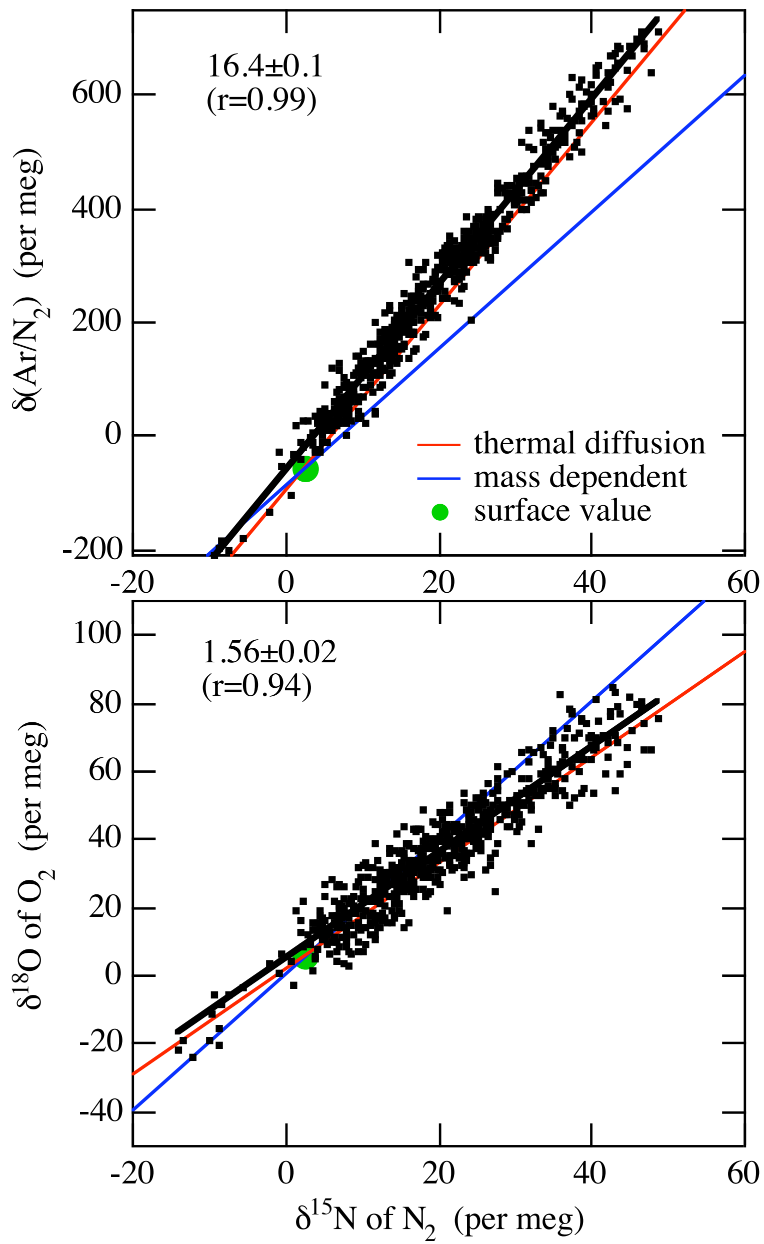


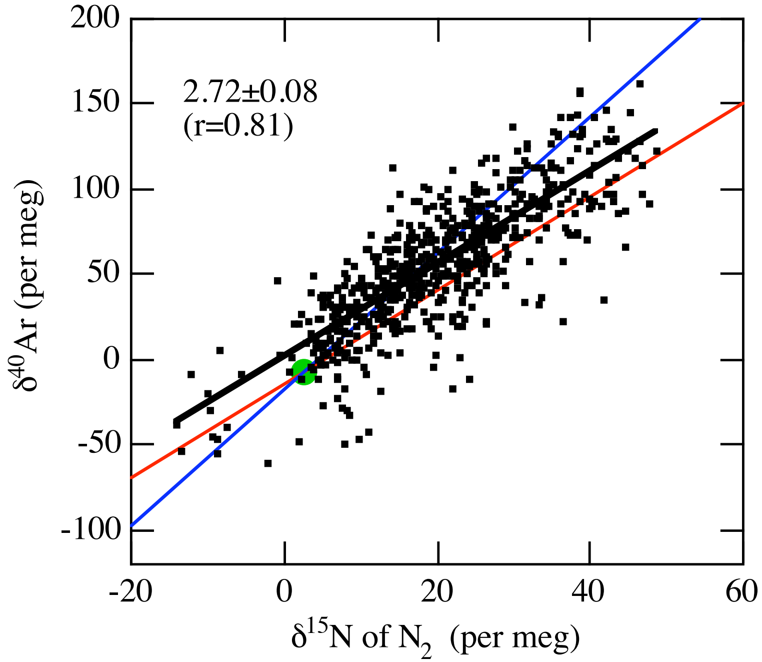
δ(Ar/N_2) varied basically in phase with APO, and both of them showed clear seasonal cycles. APO decreases secularly. δ(Ar/N_2) may increase slightly? (not clear)

APO: 1 minutes mean, δ(Ar/N_2): 20 hours mean

The APO/δ(Ar/N_2) ratio was 3.9 for their seasonal amplitudes. The seasonal APO cycle driven by solubility change accounts for 21% of the observed seasonal APO cycle.

Relationships of the measured δ(Ar/N$_2$), δ^{18}O and δ^{40}Ar with δ^{15}N obtained from the air samples collected on the aircraft

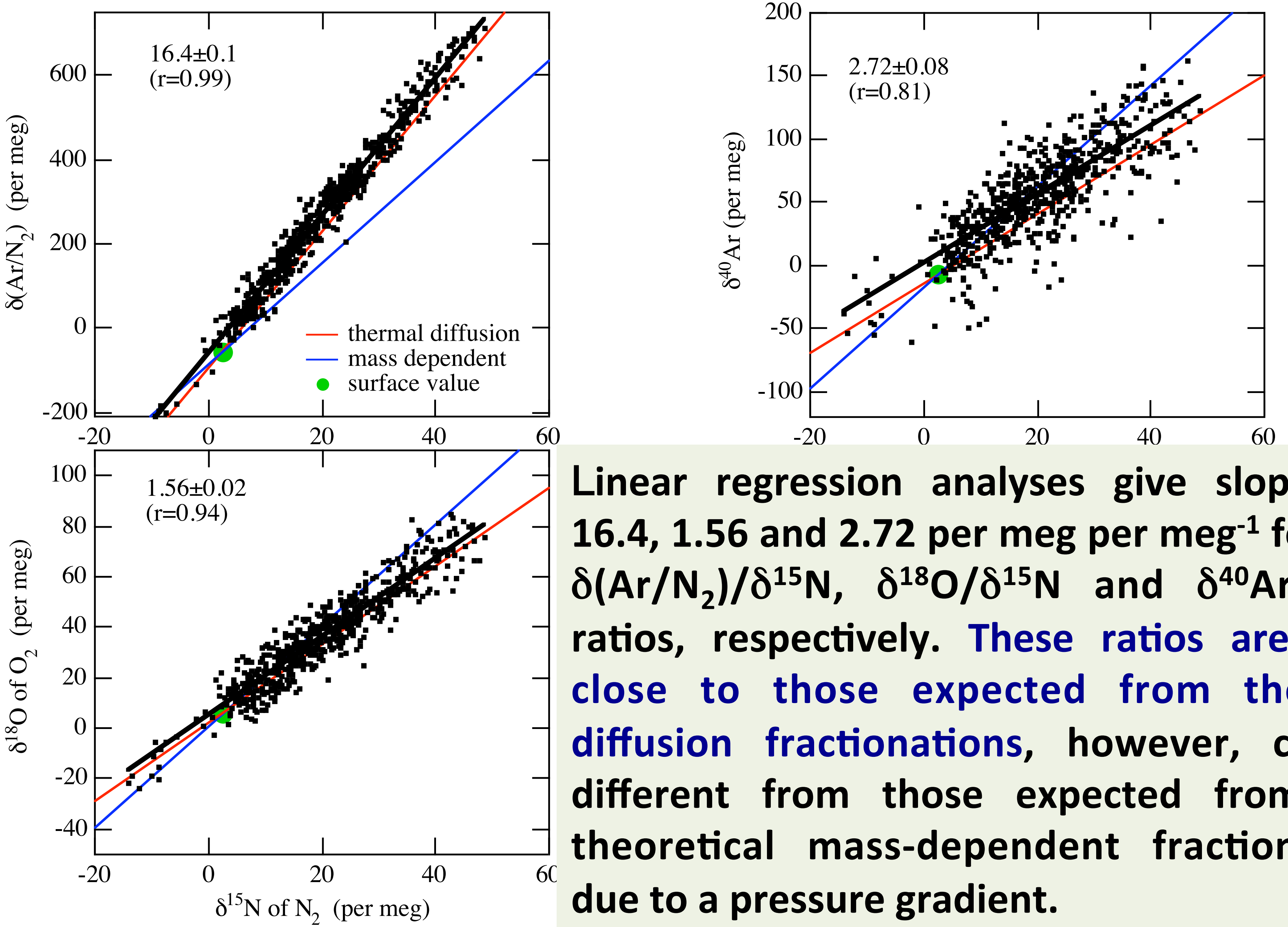




δ^{15}N, δ^{18}O and δ^{40}O (and δ(Ar/N$_2$)) should be almost constant in the troposphere, however, unrealistic large variations of them were observed.

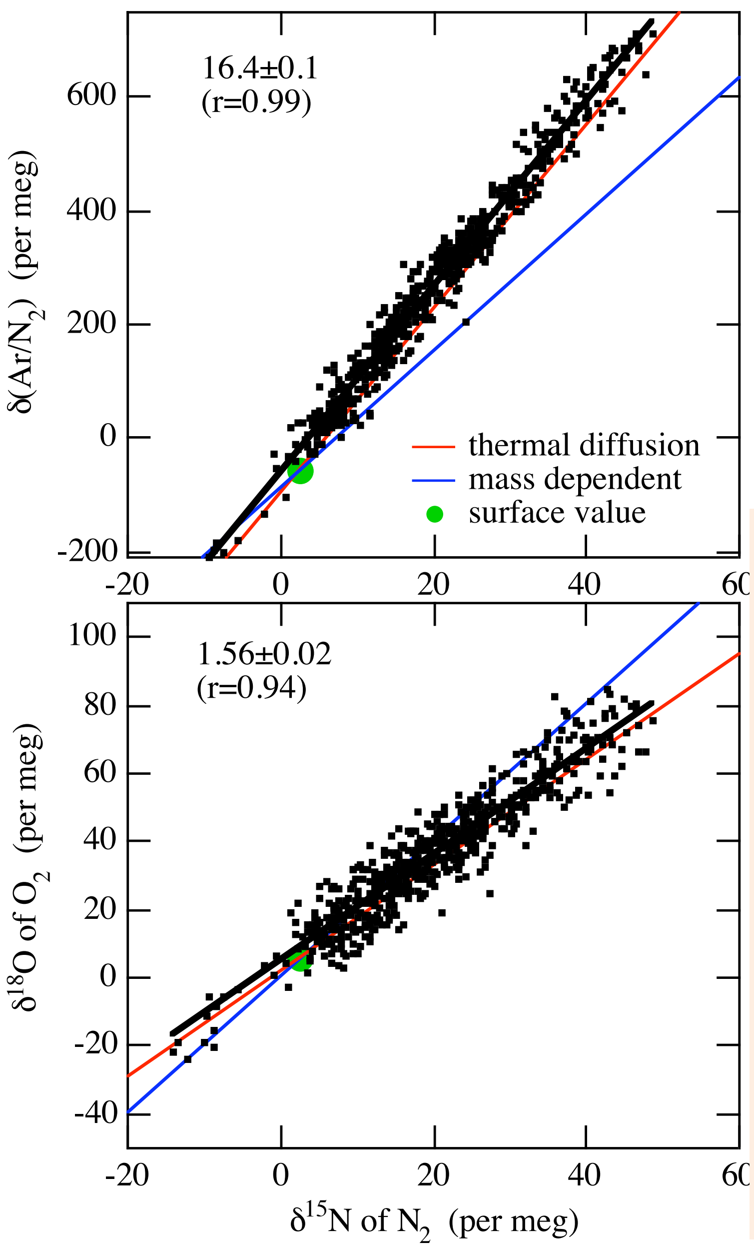
The significant variations are attributable to some kind of artificial fractionations caused by the flask air sampling onboard the C-130H aircraft.

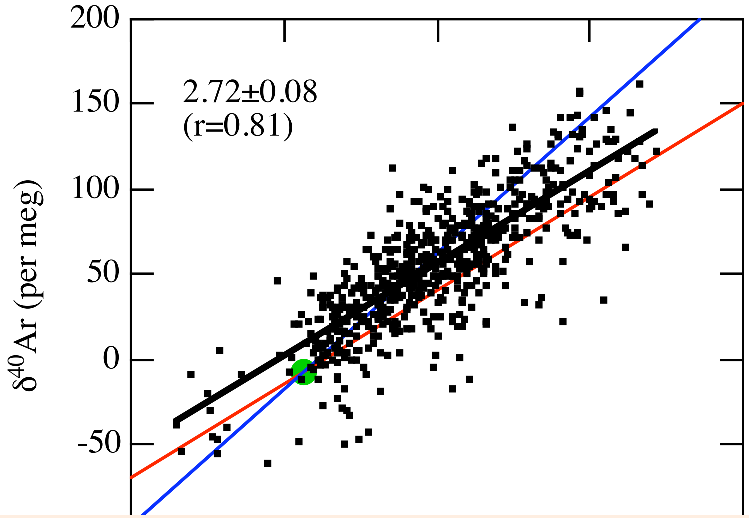
Relationships of the measured δ(Ar/N_2), δ^{18}O and δ^{40}Ar with δ^{15}N obtained from the air samples collected on the aircraft



Linear regression analyses give slopes of 16.4, 1.56 and 2.72 per meg per meg^{-1} for the δ(Ar/N_2)/δ^{15}N, δ^{18}O/δ^{15}N and δ^{40}Ar/δ^{15}N ratios, respectively. These ratios are very close to those expected from thermal diffusion fractionations, however, clearly different from those expected from the theoretical mass-dependent fractionation due to a pressure gradient.

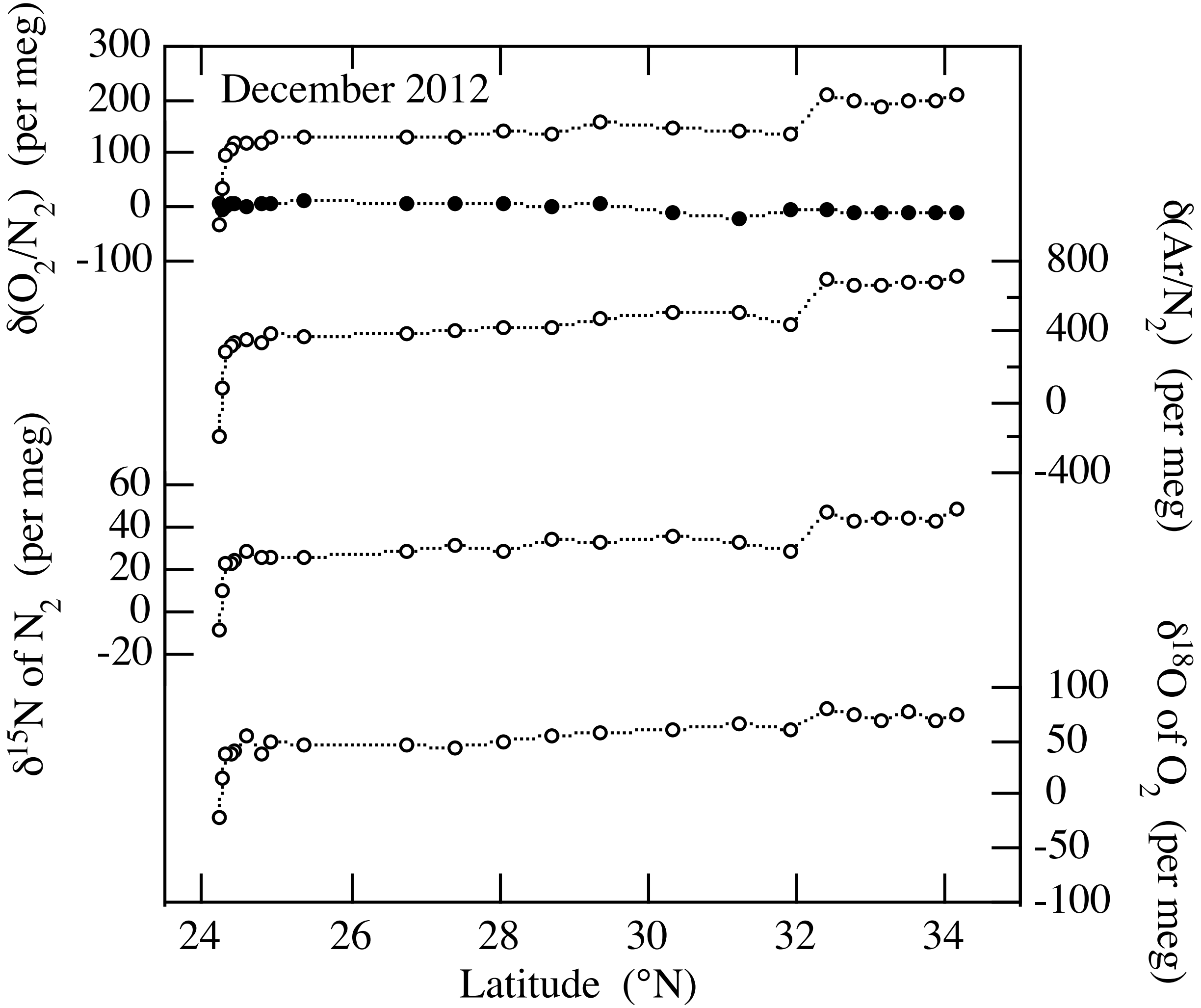
Relationships of the measured $\delta(Ar/N_2)$, $\delta^{18}O$ and $\delta^{40}Ar$ with $\delta^{15}N$ obtained from the air samples collected on the aircraft





Although the mechanism how the thermal diffusion fractionation occurs during the air sampling is not clear, it may be related to the fact that the ambient air, supplied from the jet engine of the C-130H to pressurize the cabin, is split into several branches. Since the air samples are collected through one of the branches, the fractionation of air molecules could be attributed to a temperature gradient at the branches.

Correction of the thermally-diffusive fractionation superimposed on the measured $\delta(O_2/N_2)$ using the simultaneously-measured $\delta(Ar/N_2)$



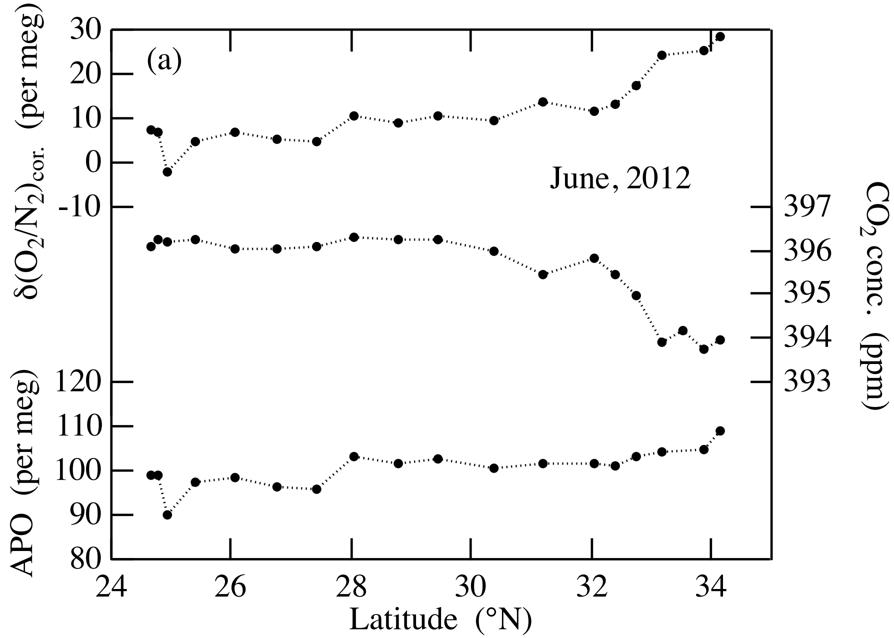
An example of the correction: The flight on December, 2012

○: uncorrected $\delta(O_2/N_2)$
●: corrected $\delta(O_2/N_2)$

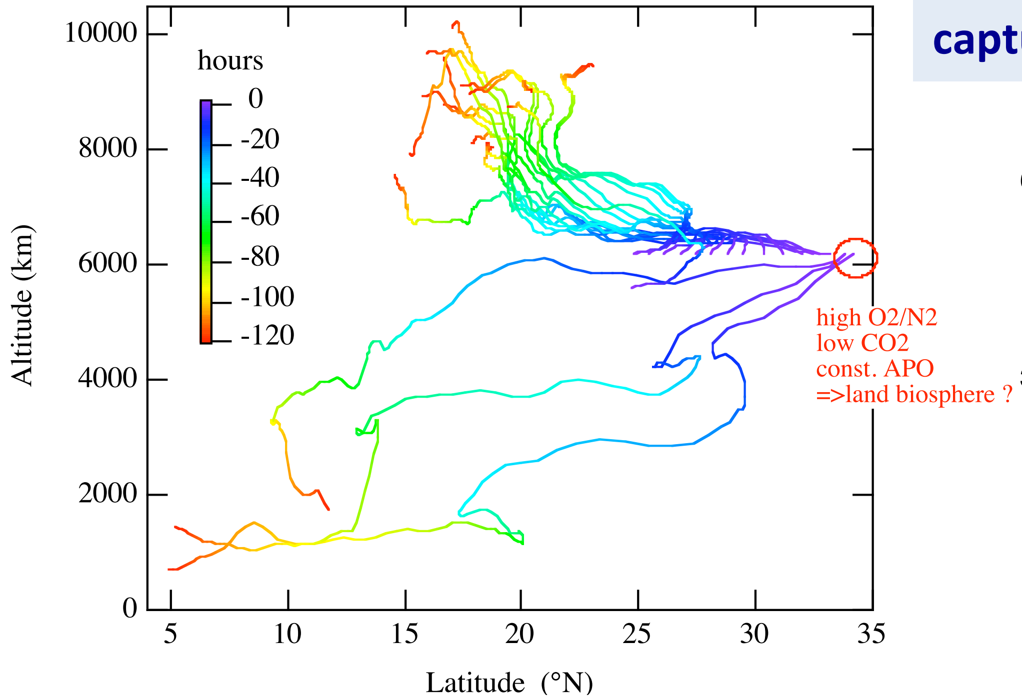
$$\delta(O_2/N_2)_{cor.} = \delta(O_2/N_2)_{meas.} - \alpha_{O_2} \cdot \alpha_{Ar}^{-1} \cdot \Delta\delta(Ar/N_2)_{meas.}$$

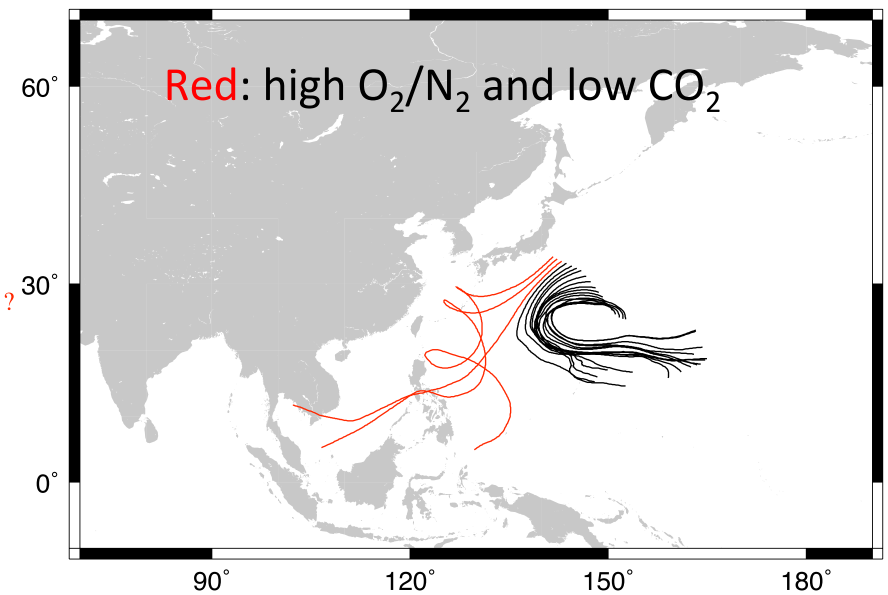
The coefficients α_{O2} = 4.57 and α_{Ar} = 16.2 are the $\delta(O_2/N_2)/\delta^{15}N$ and $\delta(Ar/N_2)/\delta^{15}N$ ratios respectively, determined from our experiment on the thermal diffusion fractionation.

An example of the spatial variations in the corrected $\delta(O_2/N_2)$ (June 2012)

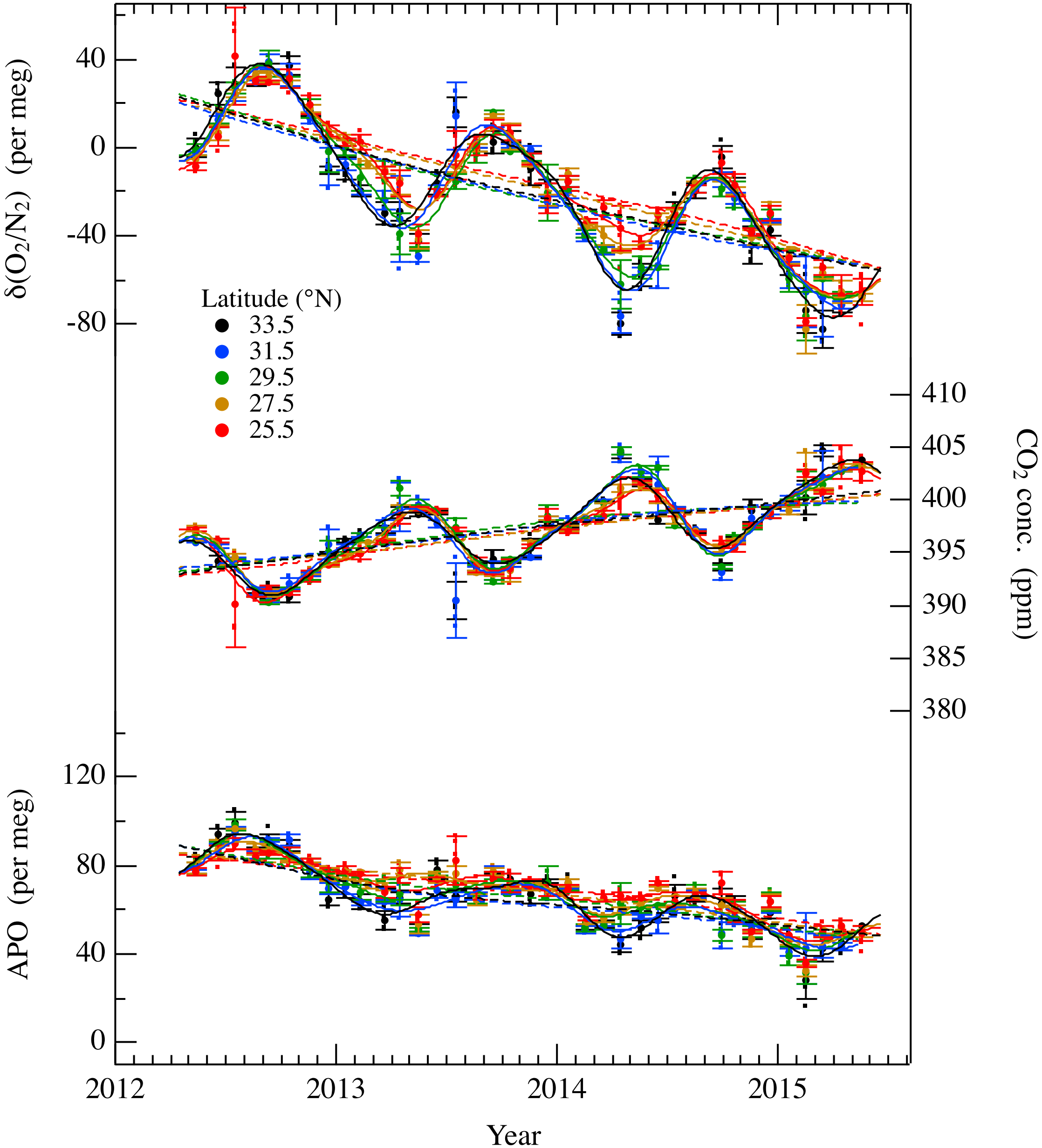


Increasing $\delta(O_2/N_2)$cor. with decreasing CO_2 is observed at higher latitudes north of 33°N. In this region, APO shows no significant changes, suggesting an intrusion of air masses influenced by the terrestrial biospheric activities and/or fossil fuel combustions. This conjecture is based on the backward trajectory analysis using the NOAA-HYSPRIT showing air parcels rising from the equatorial surface region into the middle troposphere north of 33°N. **These results indicate that the $\delta(O_2/N_2)$cor. data are able to capture synoptic-scale variations.**





$\delta(O_2/N_2)_{cor.}$, CO_2 concentration and APO observed at height interval between 5.1–6.9 km at each latitude over the western North Pacific



$\delta(O_2/N_2)$cor. and APO show clear secular decreases, while CO_2 concentrations show secular increases.

$\delta(O_2/N_2)$cor. (CO_2 concentration) shows clear seasonal cycles at all latitudes with summertime maxima and minima (minima and maxima), respectively.

APO also shows clear seasonal cycles, however, its seasonal amplitude decreases significantly toward the lower latitudes.

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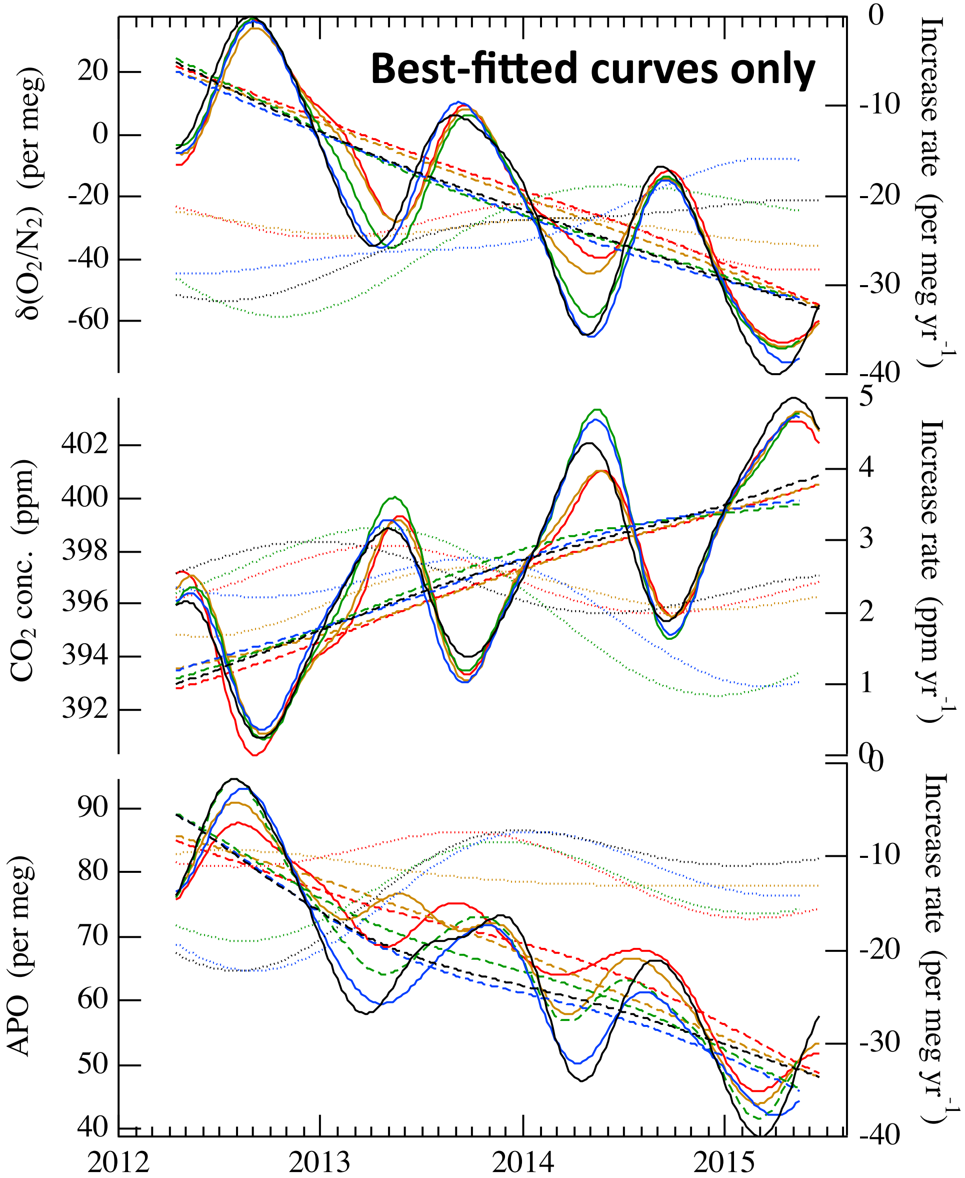
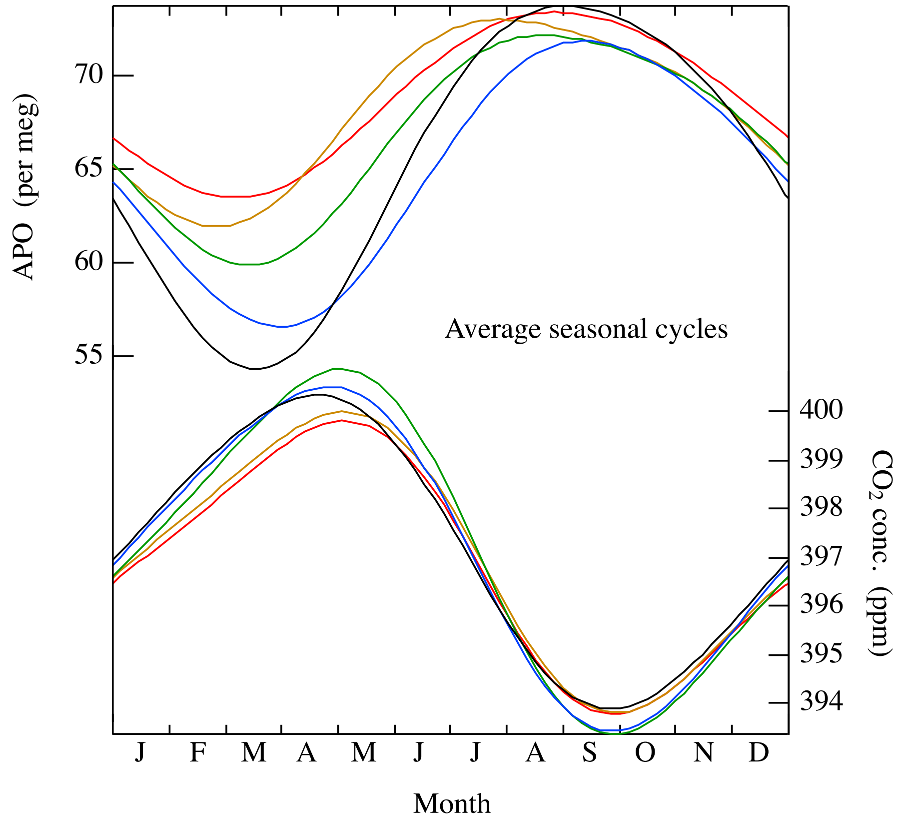




The amplitude of the mid-tropospheric seasonal APO cycle at 33.5°N was found to be twice as large as that observed at 25.5°N, whereas the corresponding latitudinal difference in the seasonal CO_2 amplitude was less than 10%

Comparison between the observed and simulated APO over the western North Pacific

<u>**The atmospheric transport models**</u>

STAG

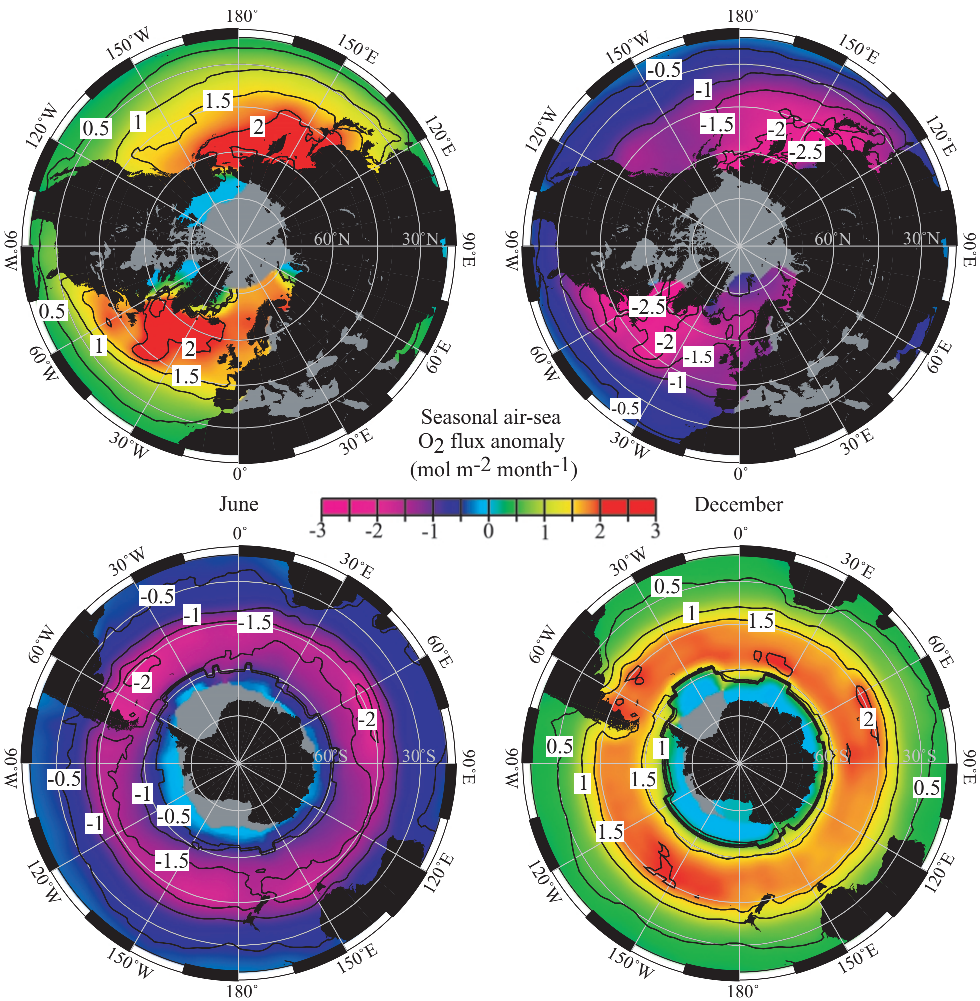
(e.g. Taguchi et a., 2002, Tellus B)

NICAM-TM

(e.g. Niwa et al., 2011, JMSJ)

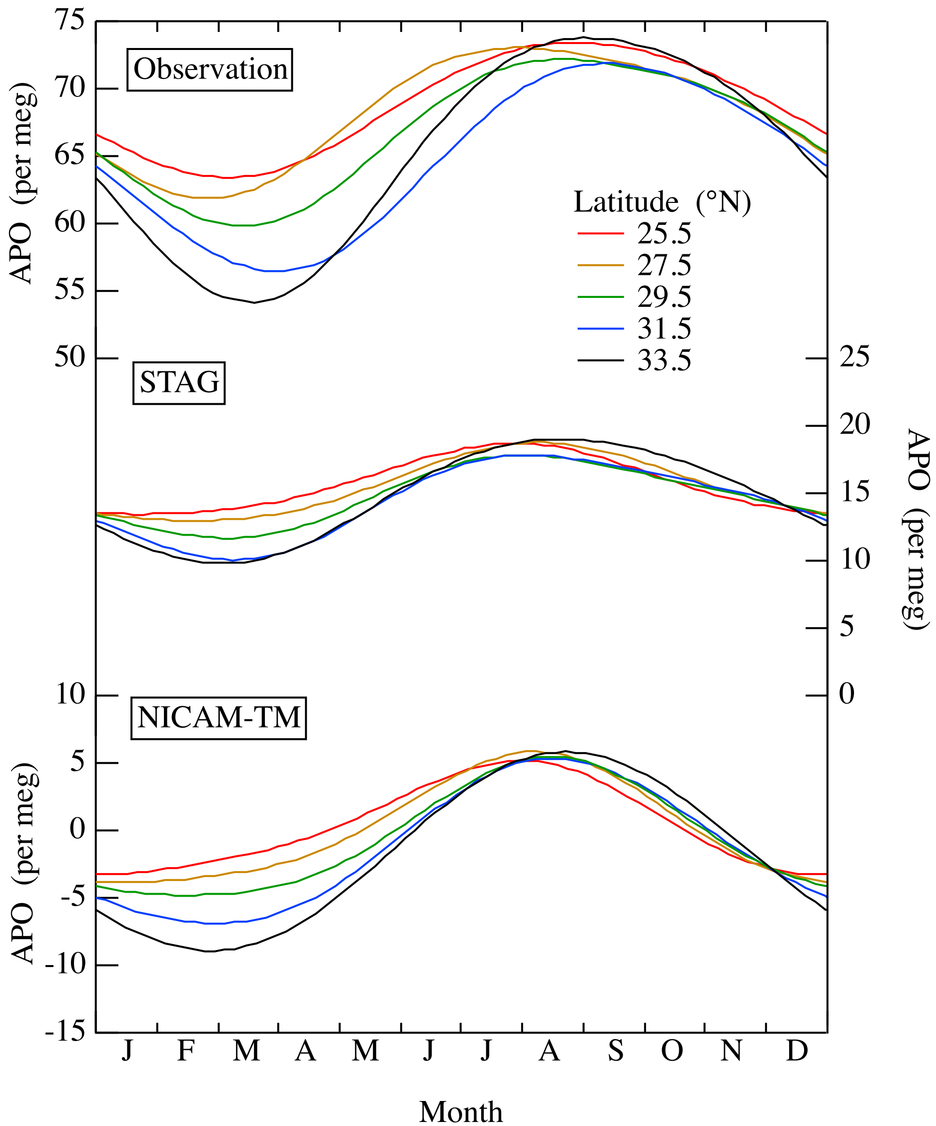
<u>**The O_2 and N_2 fluxes incorporated in the transport models**</u>

TransCom monthly air-sea O_2 and N_2 fluxes climatology (Garcia and Keeling, 2001)



Garcia and Keeling (2001, JGR)

Comparison between the observed and simulated APO over the western North Pacific



The amplitudes of seasonal cycles of the simulated APO underestimate the observed amplitudes. The underestimation is more significant in the simulated APO by STAG than that by NICAM-TM.

=>similar underestimation is also found in the middle to upper troposphere over Sendai-Fukuoka, Japan (Ishidoya et al., 2012).

Both the simulated APO by STAG and NICAM-TM show the significant decrease of the seasonal amplitudes toward the lower latitude.

=>consistent with the observed characteristic.

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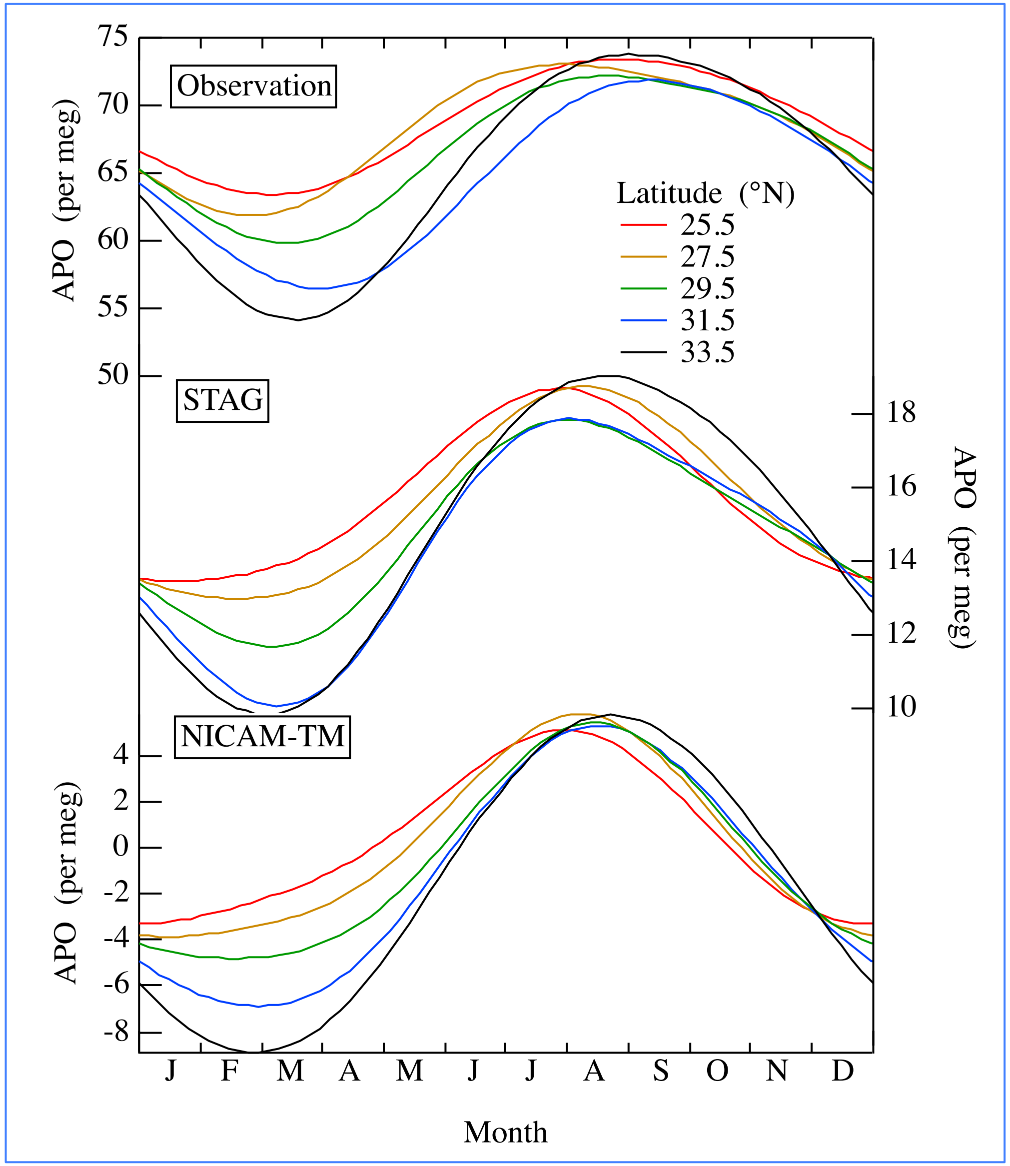

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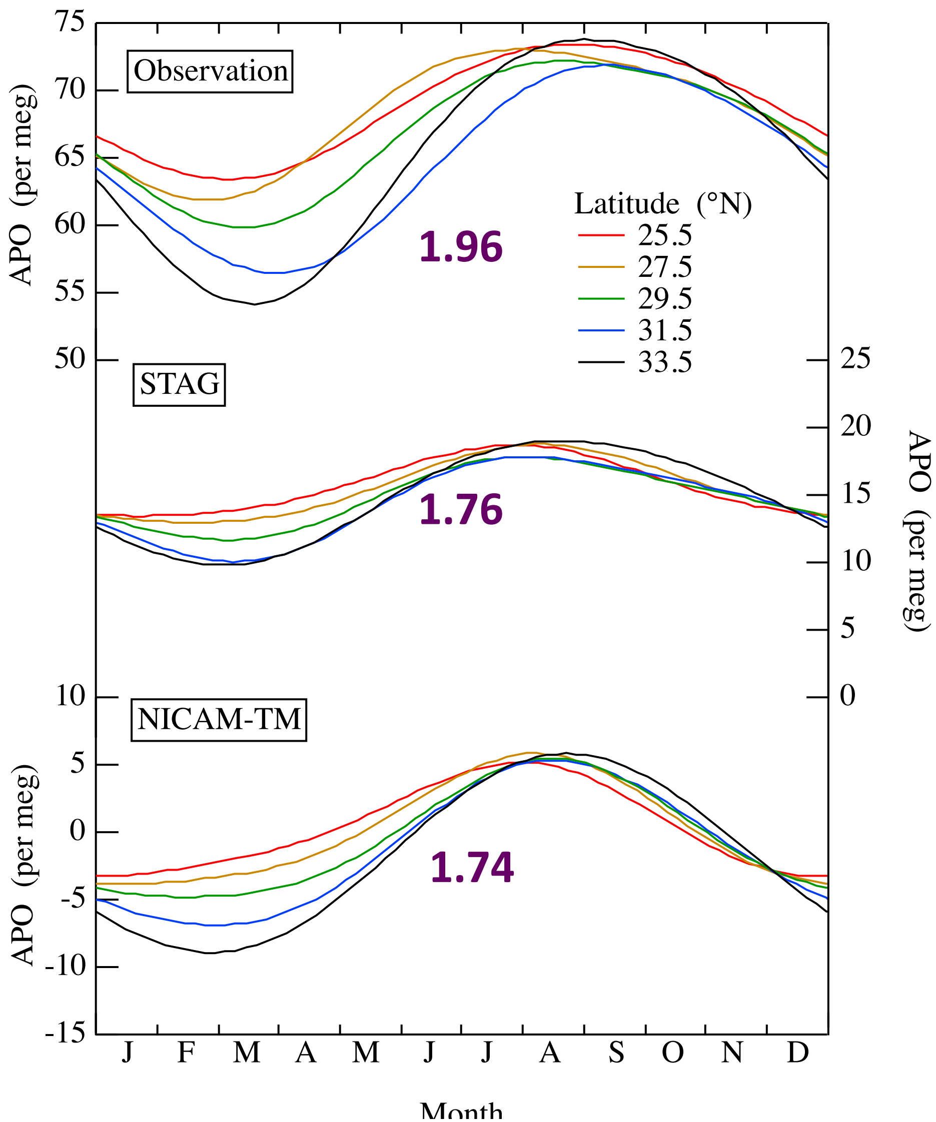
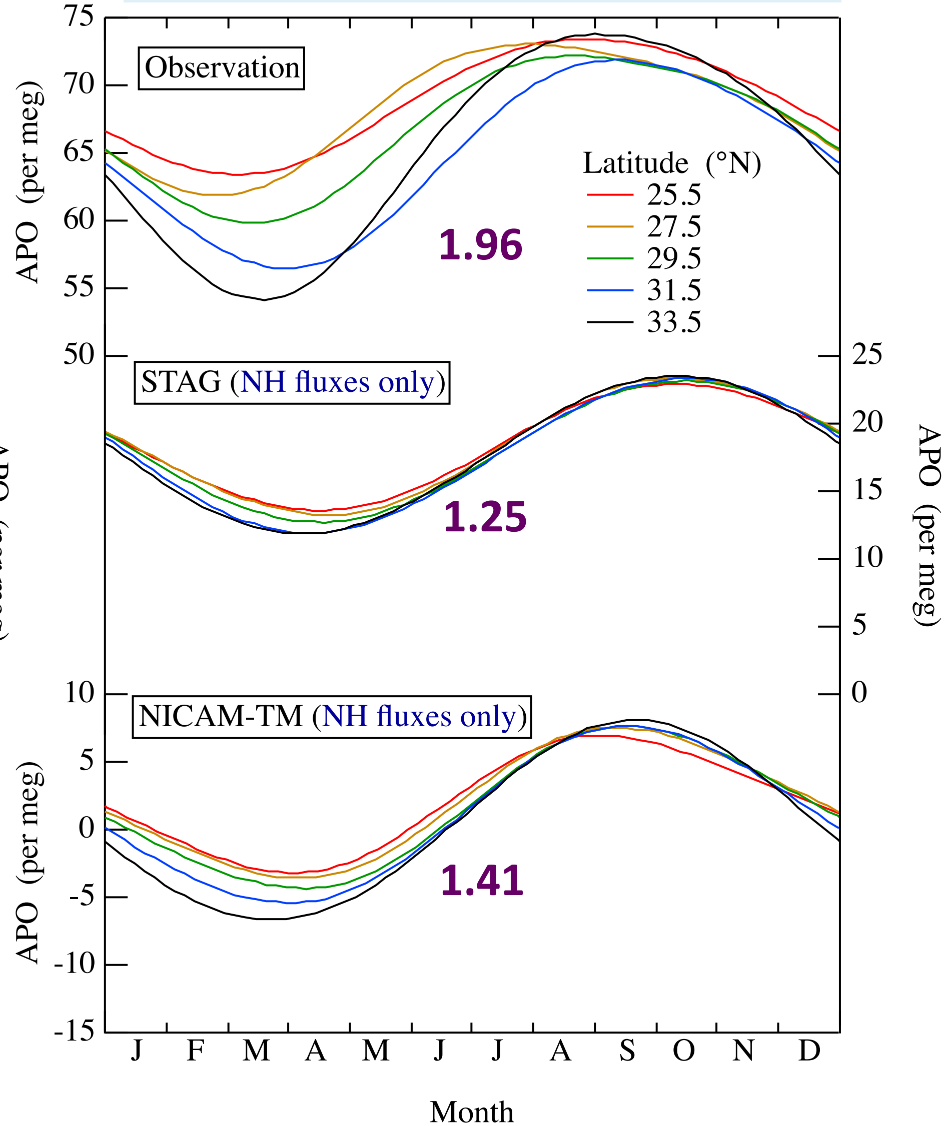
Comparison between the observed and simulated APO over the western North Pacific

Purple: Ratio of the seasonal amplitude of APO at 33.5 to that at 25.5°N
(In case of CO_2: The corresponding ratio is about 1.1)

Simulation using global O_2 and N_2 fluxes



The APO simulated by using Northern hemispheric Fluxes only



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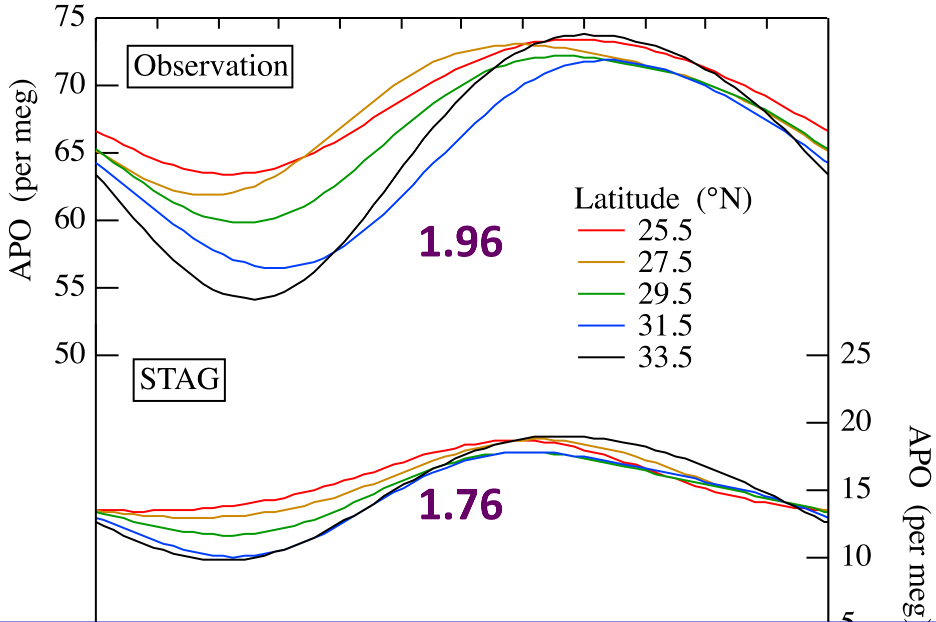

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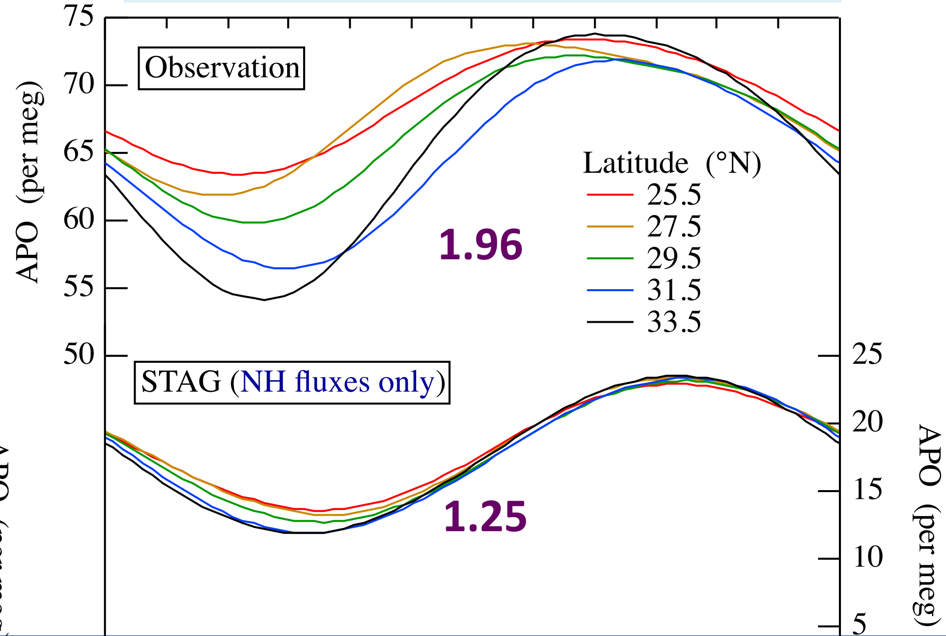

These results suggest that the mid-tropospheric seasonal APO cycle in the northern low latitudes is reduced by a superposition of the anti-phase seasonal APO cycles in the northern and southern hemispheres.

Therefore, it is expected the seasonal APO cycles observed in the troposphere over the subtropical region will be used to evaluate the interhemispheric air mixing.

Comparison be… North Pacific

Purple: Ratio of …
(In case of CO_2: T…

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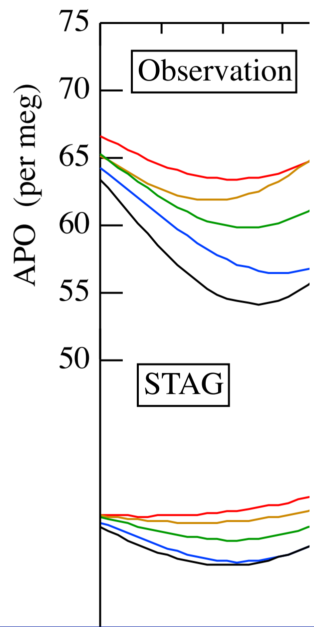

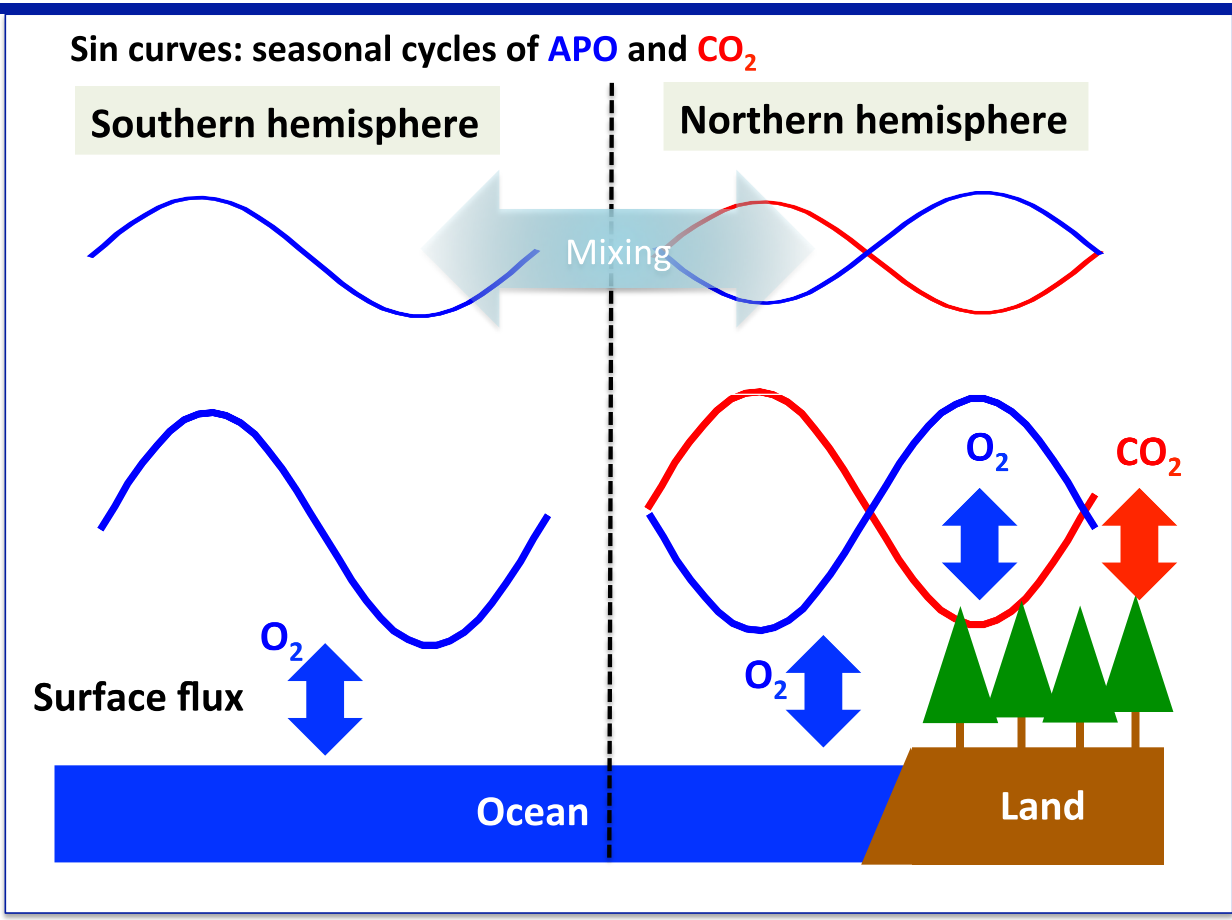

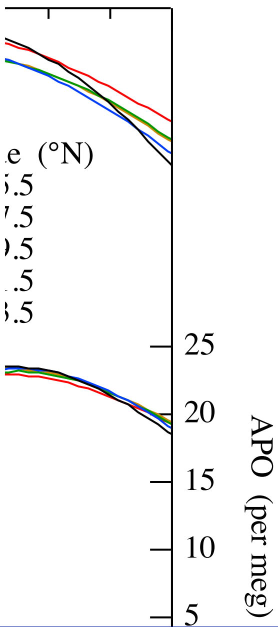

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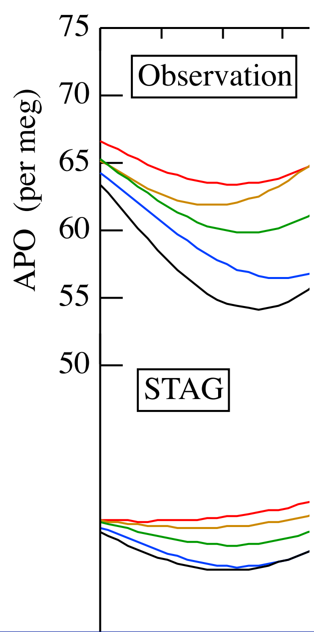
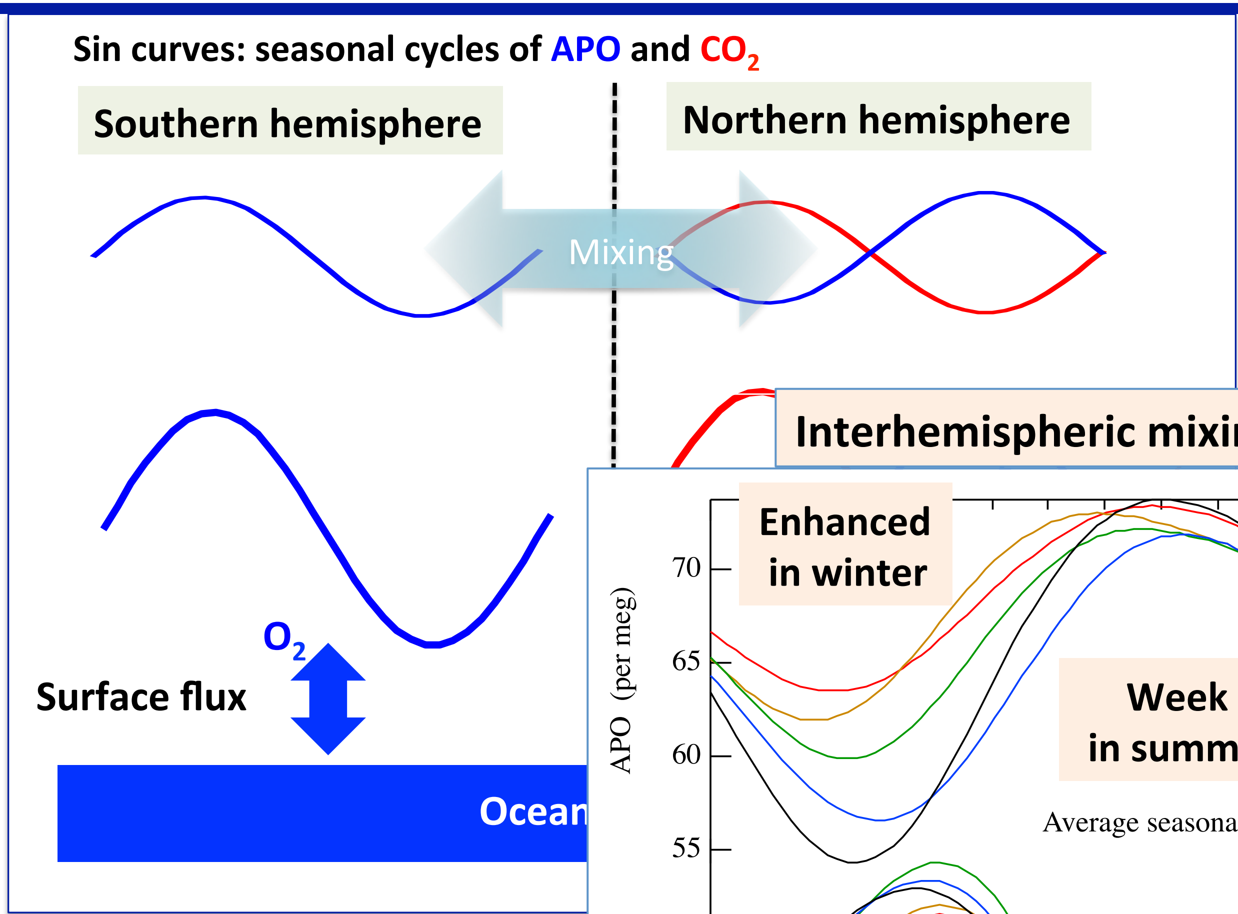
Month

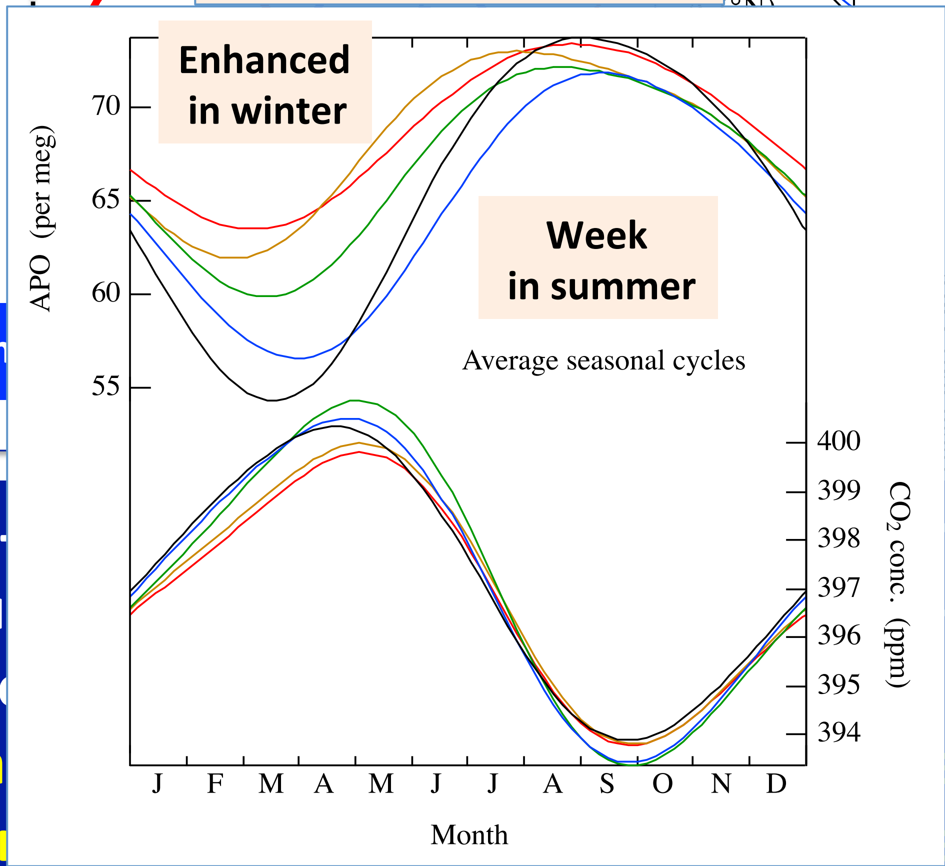
Comparison be… North Pacific

Purple: Ratio of …
(In case of CO_2: T…

Simulation u… …sing …es only

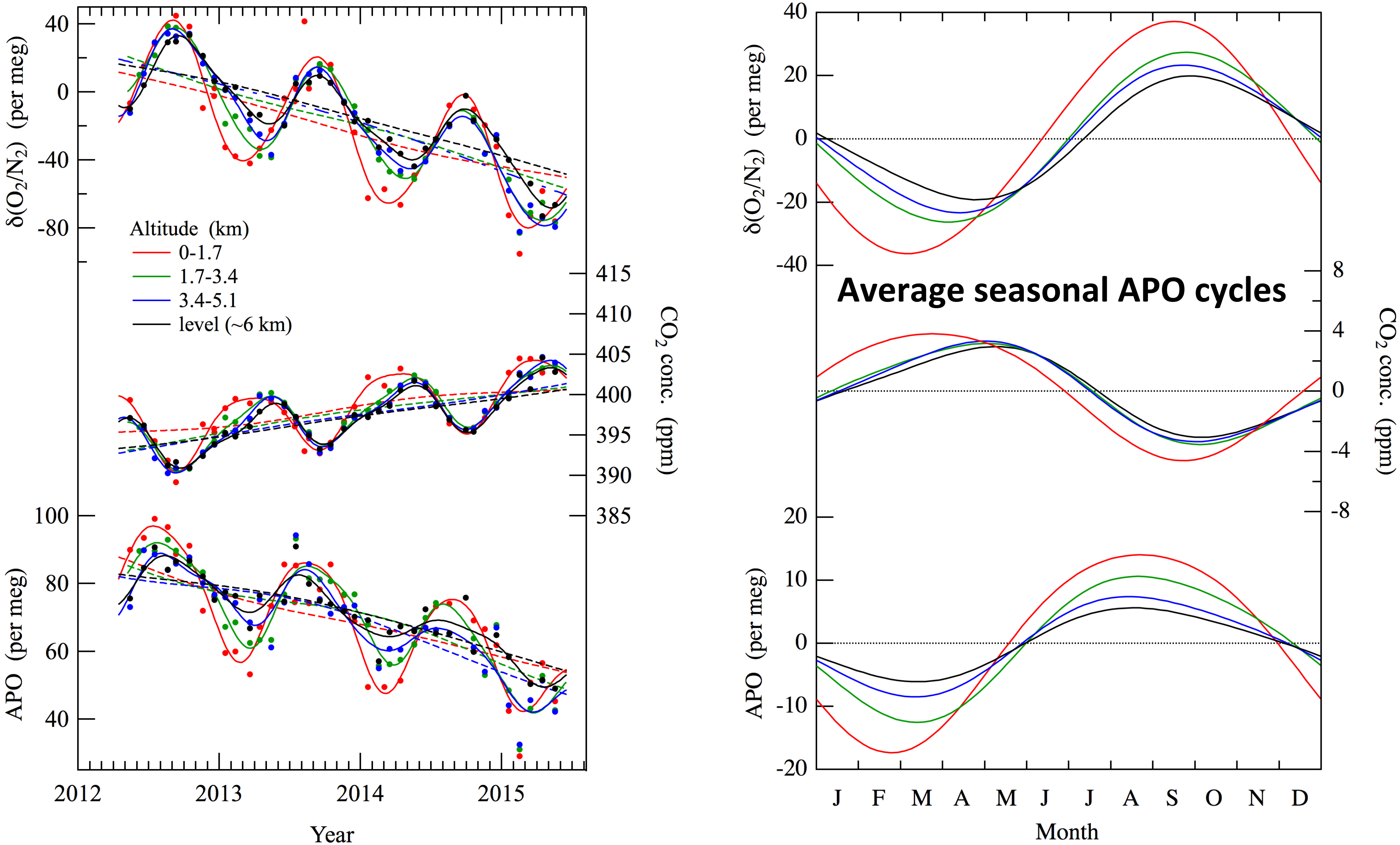






These results suggest that the mid-… the northern low latitudes is redu… phase seasonal APO cycles in the n…
Therefore, it is expected the sea… troposphere over the subtropical … interhemispheric air mixing.

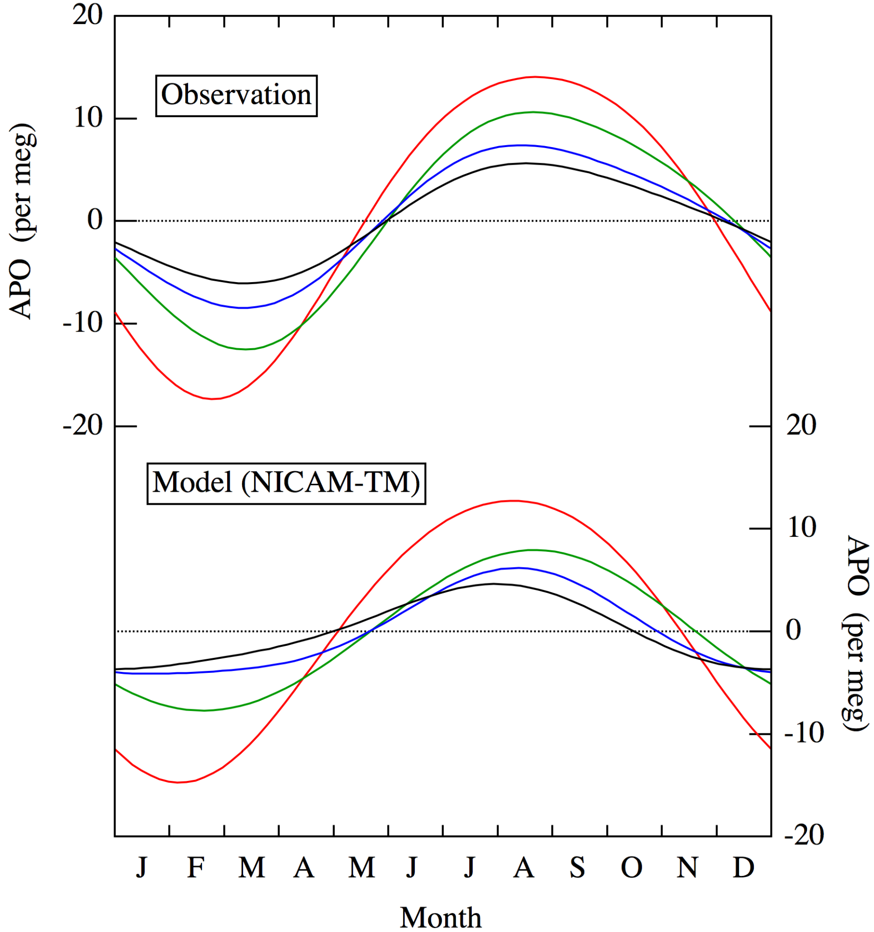
$\delta(O_2/N_2)_{cor.}$, CO_2 concentration and APO observed in the troposphere over Minamitorishima (MNM), Japan.



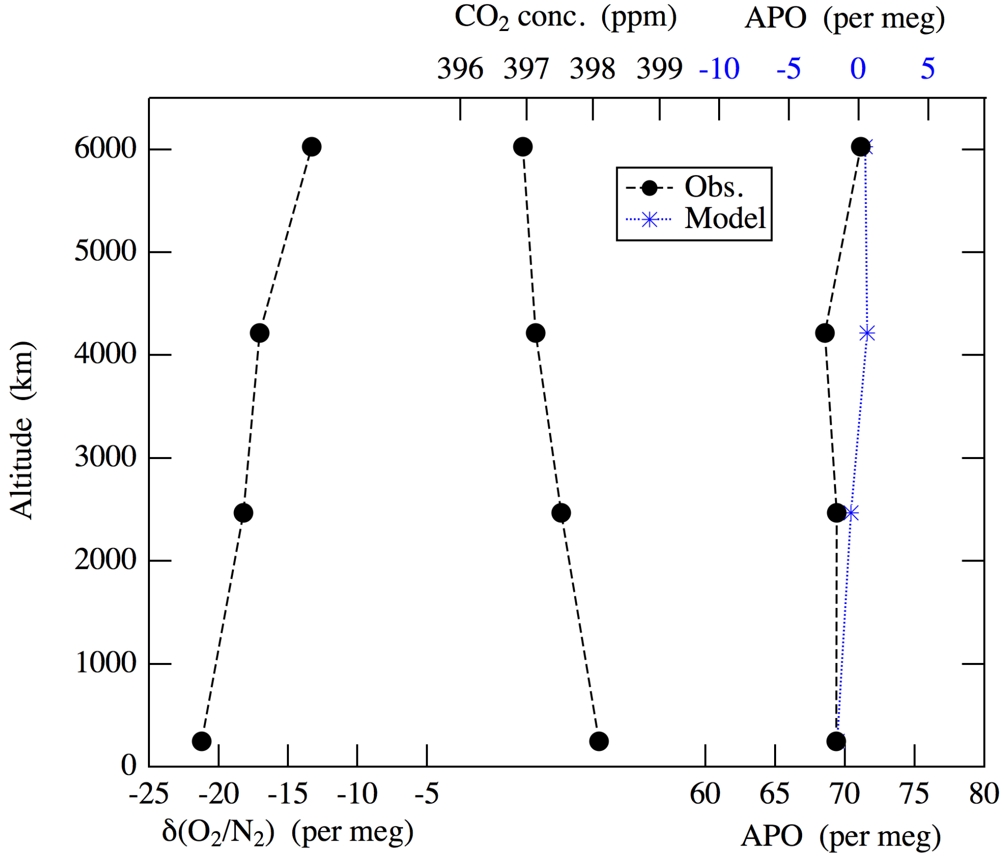
The amplitudes of seasonal $\delta(O_2/N_2)$cor., CO_2 concentration and APO cycles found to decrease with increasing height over MNM (24.28°N).

The observed and simulated APO in the troposphere over MNM

Average seasonal APO cycles

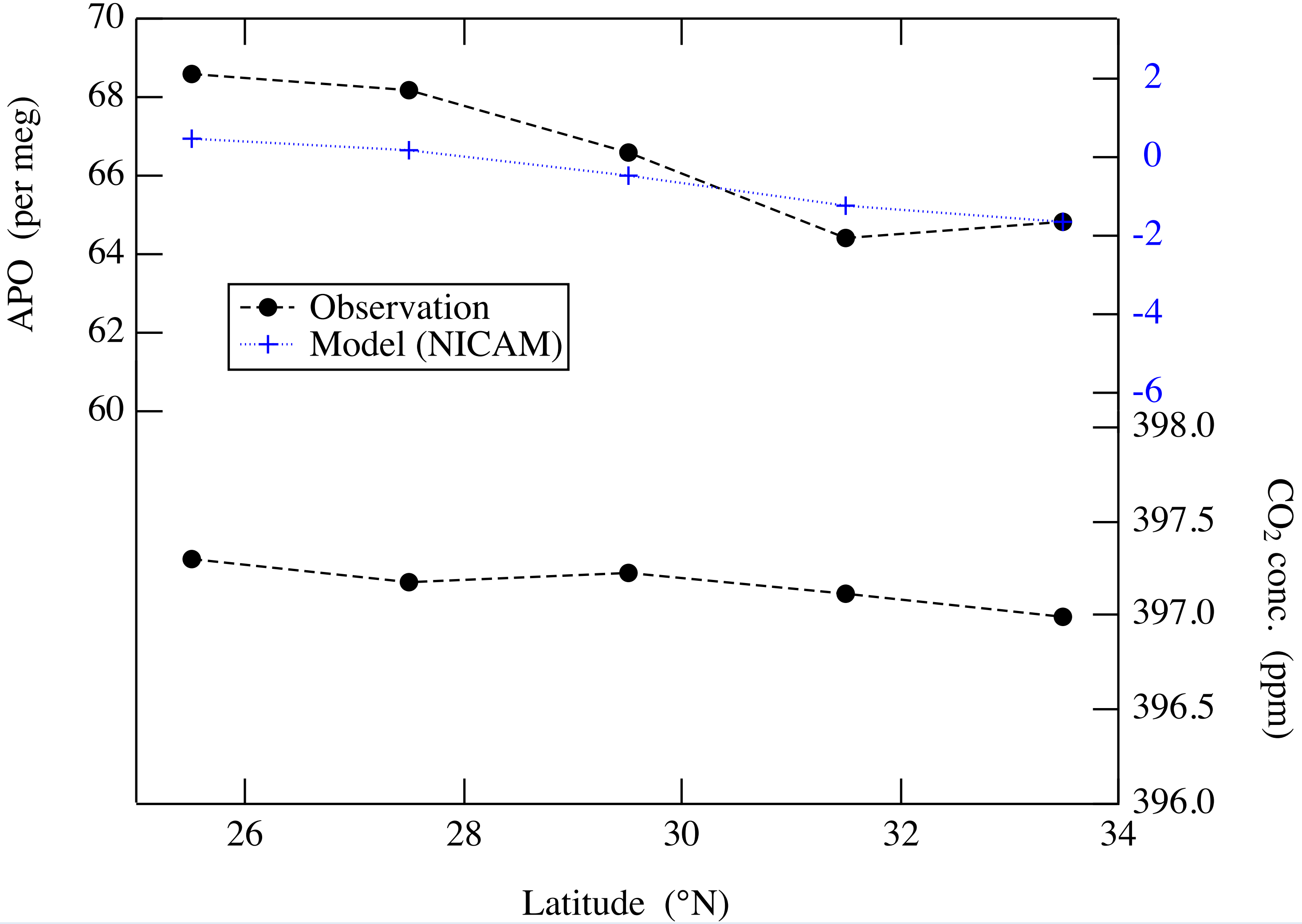


Annual average vertical gradients of δ(O_2/N_2)cor., CO_2 concentration and APO



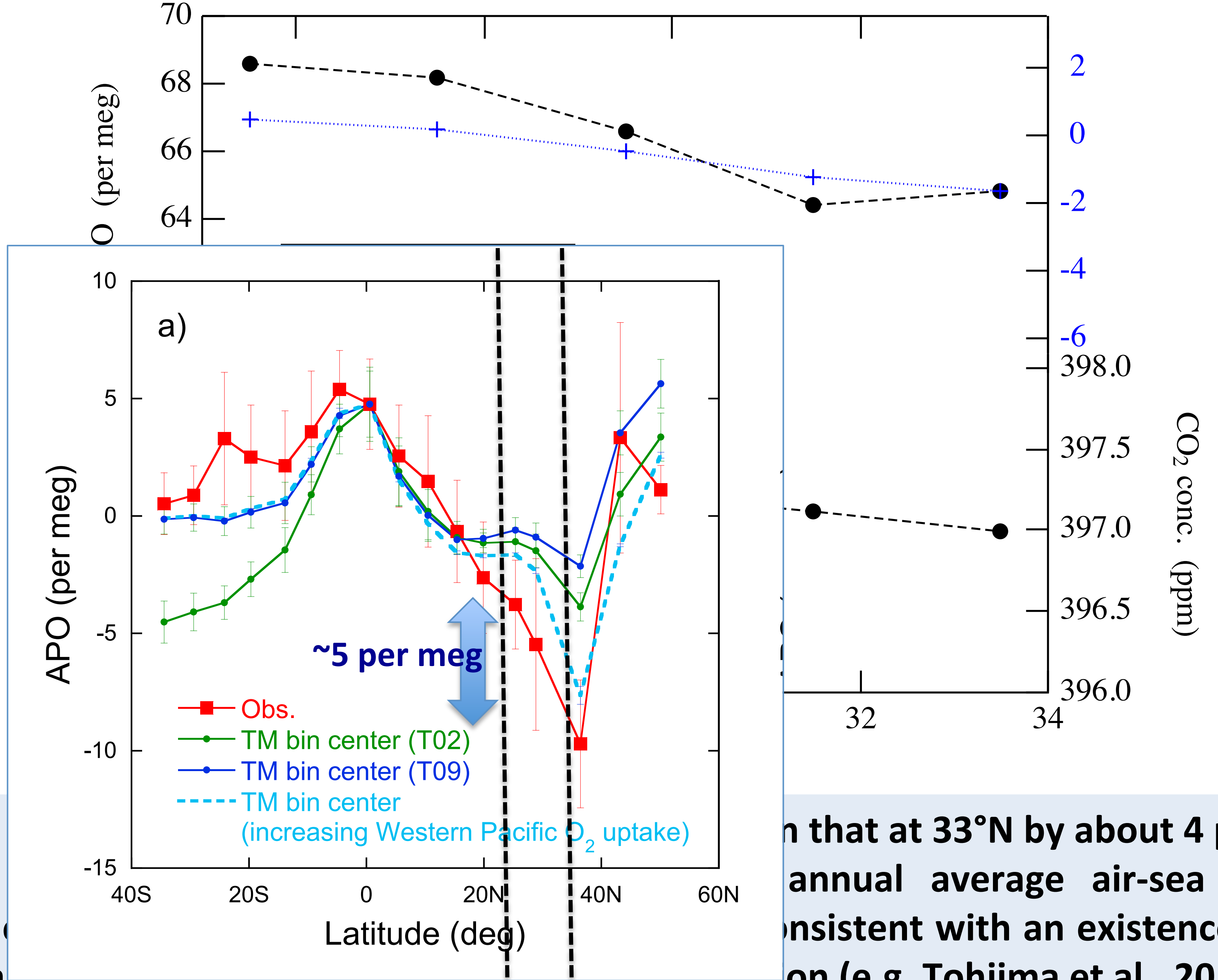
The observed altitudinal decrease of the seasonal amplitudes is relatively well reproduced by the simulated APO by NICAM-TM. Both the annual average vertical gradients in the observed and simulated APO are very small (within 2 per meg), which may suggest that there is no annual average significant source and sink of O_2 in the ocean around MNM.

Latitudinal gradients in annual average APO from 25.5 to 33.5 °N



The annual average APO was higher at 25.5°N than that at 33°N by about 4 per meg, and the APO simulated without considering annual average air-sea O_2 flux underestimates the latitudinal gradient. This is consistent with an existence of sea-to-air annual average O_2 flux in the equatorial region (e.g. Tohjima et al., 2012).

Latitudinal gradients in annual average APO from 25.5 to 33.5 °N



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Concluding Remarks

- We have analyzed the air samples collected in the middle troposphere over the western North Pacific since May 2012. The significant artificial fractionation due to thermal diffusion superimposed on the observed $\delta(O_2/N_2)$ were corrected by using the simultaneously-observed $\delta(Ar/N_2)$. =>This method will enable us to analyze $\delta(O_2/N_2)$ of air samples obtained from various kinds of aircrafts.
- The corrected $\delta(O_2/N_2)$ and the APO showed prominent seasonal cycles superimposed on clear secular downward trends. The amplitude of the mid-tropospheric seasonal APO cycle at 33.5°N was found to be twice as large as that observed at 25.5°N.
- By comparisons between the observed and simulated seasonal APO cycles, it was suggested that the mid-tropospheric seasonal APO cycle in the northern low latitudes is reduced by the anti-phase seasonal APO cycles in the northern and southern hemispheres.
- The difference between the annual average APO at the surface and that in the mid-troposphere are found to be within 2 per meg over Minamitorishima, Japan.
- The annual average APO was higher at 25.5°N than that at 33°N by about 4 per meg, which may reflect an annual average sea-to-air O_2 flux in the equatorial region.