Study on distribution of trace greenhouse gasses over the Western Pacific and their sources (Abstract of the Final Report)

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1. Introduction

Greenhouse gases (GHGs) other than CO_2 are mainly trace gases, such as methane, nitrous oxide, hydrofluorocarbons, perfluorocarbons and sulfurhexafluoride, and these are accounted as greenhouse gases in terms of Kyoto Protocol. These atmospheric concentrations are smaller than that of CO_2, however, the sum of these radiative forcing is nearly half of CO_2, because each of these gases has high global warming potential (GWP).

The emissions of these trace greenhouse gasses may increase, especially in Asian region in the future, however, emission inventories of these gasses are not well established in Asia. Therefore, source identification and its emission estimation for these gasses in the Asian region must be done with high priority. In this study observational source identification or estimation of emission strength are tried by means of latitudinal atmospheric observation using ship routes over the western Pacific ocean. To calculate the emission strength, studies of life time and its decline mechanisms of these gasses in the atmosphere are also necessary.

National Institute for Environmental Studies has been operating an atmospheric CO_2 monitoring program using commercial cargo ship operated between Japan, Australia and New Zealand. The same platform can be used fortunately for this study measuring trace greenhouse gases, but new analytical instruments were necessary to be developed for the ship observation and sampling of these trace species. Such observational estimation of source strength can be compared to the emission inventories in the Asian region in the future.

2. Research Objective

(1) Latitudinal distribution of methane and nitrous oxide

The sampling of the atmosphere for the measurement of CH_4 and N_2O will be continued to reveal the latitudinal distribution in the western Pacific region and the secular increasing trend of these gases is to be extracted. Methane will be produced by agricultural activity and biomass burning in Asia. The influence of these emission sources could be detected from these distribution comparing with the data from the existing monitoring station. The cargo ship based observation is an economic way to have meaningful latitudinal concentration gradient for the atmospheric species, easier than the establishment of a large number of ground based stations. On the other hand, N_2O has various sources in both land area and the ocean, however, the understanding of its latitudinal distribution and seasonality is poor.

(2) Latitudinal distribution of ozone and carbon monoxide

Although tropospheric ozone (O_3) is one of the most important greenhouse gases, observational data of tropospheric O_3 in remote oceanic air masses have been very limited so far, because of availability of long-term platforms. Carbon monoxide (CO) is emitted from combustion sources, and is often used as an indicator of anthropogenic activities and biomass burning for air masses characterization. CO itself is also very important as it reacts with OH radicals, a key player for oxidation capacity in the troposphere. A so-called bottom-up emission inventory has a large uncertainty due to its multiple sources, including oxidation of methane and non-methane hydrocarbons, combustion of fossil fuels, biomass burning (agricultural waste burning, forest fires), and bio-fuels. Continuous and dense observational data particularly over remote oceanic regions are needed to better constrain contribution of tropospheric O_3 to global warming, and emissions inventory of CO. In this study we make continuous measurements of surface O_3 and CO onboard cargo-ship in operation between Japan and Australia/New Zealand.

(3) HFC, PFC, SF_6 measurement

Very limited data are available for the spatial distribution and time variation of fluorocarbons such as HFCs, PFCs and SF_6, which included in the target greenhouse gases for emission reduction within Kyoto Protocol. In this study, we started sampling for the measurement of HFCs, PFCs, SF_6, CFCs, HCFCs and some other anthropogenic halocarbons over the western Pacific using the ship of opportunity to know the year-to-year change, seasonal change and north-south trend of their background concentration. The emission inventory could be checked by their latitudinal distribution and/or concentration ratio. The measurement of these less reactive greenhouse gases also contributes the understanding of air mass exchange between both hemispheres.

3. Results

(1) Methane and N_2O over the Pacific

The latitudinal distribution and its secular trend were analyzed from the observational data obtained by the measurement by ship board atmospheric sampling. The north-south gradient of CH_4 has not showed much change during our observation from 1993, however, the annual increasing rate significantly decreased from 2000. As the major CH_4 emission sources exist in the northern hemisphere, increasing rate decrease may give change in latitudinal distribution. The less significant change in the latitudinal gradient is contradiction. The seasonal change of CH_4 in the atmosphere was extracted and shown in Fig. 1. The seasonality of atmospheric CH_4 changes at 10 degree S in latitude. The northern and the southern halves are mirror image, both hemispheric seasonal variation show the summer decrease of atmospheric CH_4 by the photo oxidation.

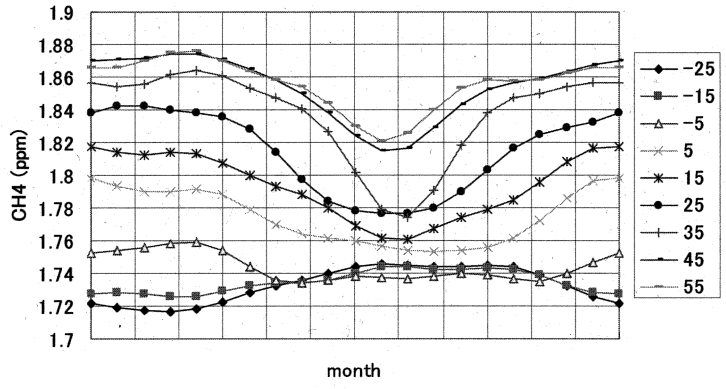

Fig. 1 Seasonal variation of atmospheric CH_4 observed by the sampling and measurement program using cargo ship in the western Pacific. The concentration is for a standard year of 2005.

The increasing trend of the N_2O is more continuous than that of CH_4. Recent increasing rate is 0.7 ppb/y and shows larger rate in the northern hemisphere and smaller in the southern hemisphere. Increasing rates in 1997, 2000 and 2003-2004 were larger than the other periods. The increasing rate change has

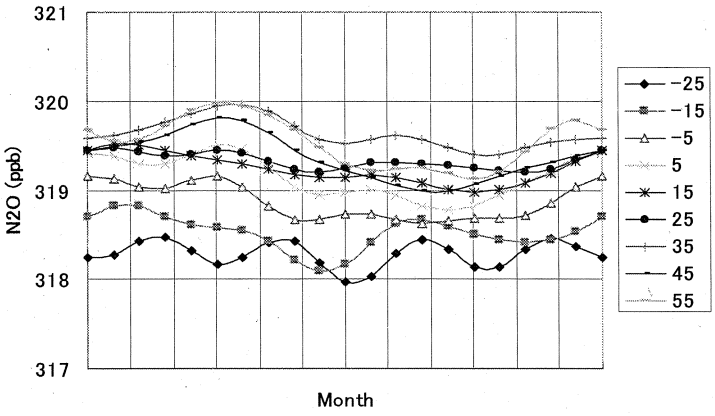

Fig. 2 Seasonal variation of atmospheric N_2O observed by the sampling and measurement program using cargo ship in the western Pacific. The concentration is for a standard year of 2005.

relationship with that of CO_2. The larger increasing rate of N_2O appears just before or just later of the CO_2 increasing rate increase. The relationship suggests the N_2O emission change with ENSO cycles. The large increase event in 2003-2004 covers the whole latitude and must be resulted by large emission. However the historically large ENSO event of 1998 did not result the large change in the trend of atmospheric N_2O. The seasonal change of N_2O in the atmosphere was extracted and shown in Fig. 2. The north-south gradient is clearly indicated however the seasonality shows very complex feature. In the northern hemisphere, N_2O concentration shows maximum in April and minimum in September.

(2) Latitudinal distribution of ozone and carbon monoxide

In addition to ongoing measurements of O_3, a continuous CO instrument was operated on the M/V Trans Future 5 since November 2005. Spatial and temporal variations of CO have been measured concurrently with O_3. Both O_3 and CO data were obtained in the wide latitudinal band in both Northern and Southern Hemispheres between Japan and New Zealand.

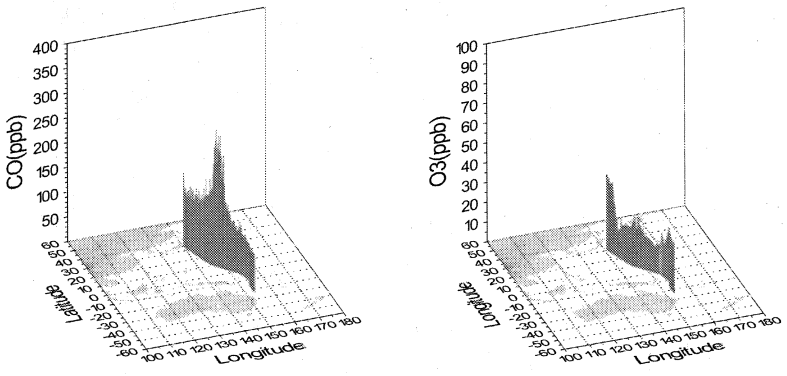

Fig.3 Latitudinal distributions of ozone (left) and carbon monoxide (right) over the Western Pacific Ocean

In general, good correlative behaviors of O_3 to CO in the Northern Hemisphere are observed, suggesting common sources, likely combustion sources following photochemical O_3 production. Poor correlative behaviors observed in the Southern Hemisphere suggest uncommon sources between O_3 and CO.

Fig.3 shows latitudinal distributions of O_3 and CO between Japan and New Zealand, depicting large enhancement of CO in the tropical regions in fall of 2006. Back trajectories analysis coupled with hot spots data showed that the air masses come from Southeast Asia, where severe biomass burning occurred in this season of 2006.

(3) Halocarbon observation

Air samples were collected over western Pacific between 30°N and 30°S using volunteer ships, M/V Fuji Transworld and M/V Transfuture 5. At each cruise, 7-8 samples were collected in a 6-L Silico Can at ambient pressure, or were collected in a 3-L ECB canister at 0.2 MPa. After being transported to the laboratory, samples were analyzed using pre-concentration/capillary GC-MS for HFCs (except for HFC-32 and HFC-23) and HCFCs.

The whole datasets obtained for HFC-134a, HCFC-1452b and HFC-152a are shown as

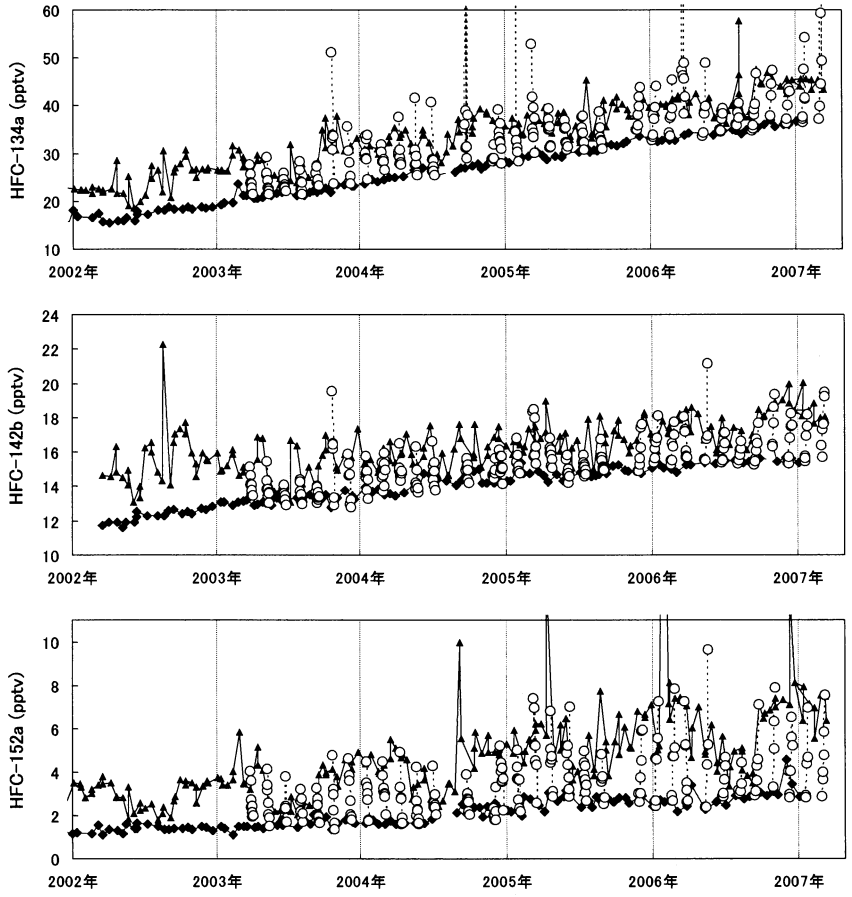

Fig. 4 Time series of (1) HFC-134a, (2) HCFC-142b, (3) HFC-152a.
○: Western Pacific (30°N-20°S), ◆: Cape Grim, ▲: Hateruma Island

time-series in Fig. 5, together with the periodic monitoring data from Hateruma Island (24.05° N, 123.7° E) and Cape・Grim (40.41° S、144.64° E) where grab sampling has been done semi-monthly for years. It can be seen that the concentration of these compounds from the cruises in the present study lie mostly between the plots from Hateruma Island and from Cape Grim. The concentration of HFC-134a (Fig. 4-1), which is widely used for car air conditioners, has increased rapidly, and its annual median value was changed from 26.0 ppt (FY 2003) to 30.8 ppt (FY 2004) to 35.3 ppt (FY 2005) to 38.8 ppt (FY2006) with annual increase rate of 10%~15%. HCFC-142b (Fig. 4-2) and HCFC-141b have also shown gradual increase; from 13.8 ppt (FY 2003) to 15.0 ppt (FY 2004) to 15.7 ppt (FY 2005) to 16.5 ppt (FY 2006), and 17.3 ppt (FY 2003) to 17.8 ppt (FY 2004) to 18.2 ppt (FY 2005) to 18.5 ppt (FY 2006), respectively. HFC-152a concentration (Fig. 4-3) in the northern hemisphere showed a significant seasonal variation, that amplitude has been increased year by year. Such a seasonal change of HFC-152a can be explained by its uneven distribution in NH and SH due to short lifetime (1.4 yr). The annual median value of HFC-152a concentrations in wintertime has increased from 3.0ppt (FY 2003) to 3.6 ppt (FY 2004) to 4.1 ppt (FY 2005) to 4.5 ppt (FY 2006). Among the highly volatile halocarbons, SF_6 and HFC-23 were more abundant in NH, while PFCs concentrations showed no significant difference in NH and SH. The present study has shown that the latitudinal variation of the halocarbons are reflecting their emission trend, and therefore the halocarbon monitoring over western Pacific should be continued to assess the impact of halocarbon greenhouse gases.

4. Conclusion

The atmospheric increasing of CH_4 and N_2O were observed by sampling on board cargo ship over the western Pacific. Recently, the increasing rate of CH_4 decreased and its concentration has been nearly stable from 2000. A slight decreasing trend was started to be observed in the southern hemisphere. The concentration variability in the mid-latitude in the northern hemisphere is large, however, it is suggested that the concentration increasing coincidently with the large CO_2 increasing of 2003. It might be due to the Asian and/or Siberian emission change in the mid- and high-latitude in the northern hemisphere. The CH_4 concentration is caused by complex effect of variability in the emission sources. Hence, effort of continuous atmospheric monitoring is necessary. The increasing rate of N_2O was 0.7 ppb/y for the observation period. It is suggested an unknown emission source in the equatorial region in N_2O from its latitudinal distribution. Even the inter annul change in the increasing rate is small, however, large increasing rates were observed in 1997 and 2004. The increasing rate change might have relationship between ENSO cycles. The possible emission source in equatorial region, such as ocean or tropical soil, needs further study for their N_2O emission variability. The atmospheric transportation change might be another possible mechanism of N_2O increasing rate variability.

Distinct latitudinal gradient and its seasonal dependence are observed for O_3 in both hemispheres. For example, springtime maxima in O_3 are observed over western Pacific. Latitudinal gradient in O_3 show steep decreasing trend from mid-latitudes to tropics in the northern hemisphere, and gradual increasing trend from tropics to mid-latitudes in the southern hemisphere. In the northern hemisphere, variability of O_3 and CO is positively correlated to each other, suggesting that O_3 and CO have a common origin (i.e., O_3 formation takes place due to O_3 precursors emitted from combustion sources, including anthropogenic activities and/or biomass burning). In contrast, in the southern hemisphere, O_3 and CO is anti-correlated to each other, indicating that sources are different for the two species. This includes, for example, natural origin such as stratospheric influences. Simultaneous measurements of O_3 and CO thus provide an opportunity to discuss about origins of O_3 in detail. Remarkable enhancement in CO observed in October 2006 around tropics seems due to biomass burning activities in Southeast Asia. In contrast to severe biomass burning events in 1997, enhancement in O_3 is small, implying that factors controlling O_3 formation are not similar to those in 1997. The satellite-derived CO data over southeast Asian region clearly captured CO plumes originating over Indonesia and being transported eastward, impacting to the cargo-ship observations. Differences between cargo-ship and satellite measurements highlight the importance of cargo-ship observation in helping validation and improvement of the precision satellite retrieval.

Observed fluorocarbon species showed latitudinal concentration gradient high in the northern hemisphere and low in the southern hemisphere. Species having large increasing rate has larger inter-hemispheric concentration difference. It observed that species of low atmospheric life time and increasing concentration showed increasing north-south difference, which is caused by the limited transportation of fluorocarbons from northern hemisphere to southern hemisphere. HCFC-32, which is newly developed for refrigeration media and has been measured by the improved method from Feb. 2006, showed very large increasing rate of 20%/y. Inter-hemispheric concentration differences of PFCs and their seasonal changes are small. The latitudinal distribution of fluorocarbons, especially of long atmospheric life time, is controlled by the source intensity. It needs continuous monitoring of their latitudinal distribution to constrain its emission inventory.

Reference

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