Study on long-term observation of trace greenhouse gases in the Asia-Oceania Regions (Abstract of the Interim Report)

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1. Introduction

While carbon dioxide is one of the most important greenhouse gases, other trace species including methane, nitrous oxide, anthropogenic halocarbons, tropospheric ozone, and black carbon also contribute to the global warming. The sum of radiative forcing coming from these species is comparable to that of carbon dioxide. Carbon monoxide is emitted from incomplete combustion sources, and is often used as an indicator of anthropogenic activities and biomass burning for characterization of air masses and sources. Carbon monoxide itself is also very important as it reacts with atmospheric OH radicals, a key player for oxidation capacity in the troposphere. East Asia and South East Asia are thought to be a large source region of these species, and the emissions are increasing due to expanding human dimensions and developing socio-economic activities. Testing emission inventories, which are typically built by bottom up approach depending on statistics for fuel consumption, is now feasible with the development of inverse modeling technique. However, observational data is needed for better application of the inversion technique. Observational data of these trace species in the Asia-Oceania regions have been very limited so far, because of limited availability of long-term platforms. Systematic observational data in these regions are particularly needed to better understand the spatial and temporal variations of these species, and to better constrain emissions of these species. I this program, we utilize two volunteer observational ship (VOS) platforms, one in the South East Asian region and the other in the Western Pacific. The VOS observations have advantage to take distribution of atmospheric species with the wide coverage by a single platform under a single calibration standard scale. It costs much less than distributing and arranging large number of ground-based stations.

2. Objectives

Research target species for the long-lived trace greenhouse gases are methane, nitrous oxide, and anthropogenic halocarbons. Atmospheric samples are taken in canisters on board VOS and brought back to the laboratory. Under monitoring programs by the National Institute for Environmental Studies (NIES), samples can be collected in maritime air in the wide latitudinal zones ranging from 53°N to 40°S, to identify their global distributions along with long-term trends. The sampling and analysis programs for many of

the species have been operated since 1992. By this program, we added sampling program on board VOS in South East Asian region, with which continental outflow of the trace greenhouse gases can be sampled. In addition, volatile organic species of natural origin are to be measured as the by-products of gas chromatographic measurement. Distributions of short-lived species, such as ozone and carbon monoxide, are not uniform and have much more temporal and special variety than the long-lived species, because of the short lifetime in the atmosphere. Emissions of carbon monoxide from the East and South East Asia dominate a large part of fraction to the global emissions. Understanding regional emission sources of these species requires continuous atmospheric measurement systems. We combine two VOS in operation en route Japan - South East Asia and Japan - Oceania to detect changes in the emissions of greenhouse gases from East Asia, and its resulting impacts on global warming by comparing background air over open Pacific with rural air over the Asian marginal sea. We aim to fill the gap in the global observing system of greenhouse gases by making frequent samplings and continuous measurements in the Asia-Oceania regions using VOS platforms.

3. Methods

(1) Volunteer Observational Ships (VOS) in Asia-Oceania regions

We install atmospheric sampling and continuous measurement systems on board two cargo ships. The M/V FUJITRANS WORLD is a car carrier from Japan to South East Asian countries. She departs Japan and make every 4 week cruise, visiting Hong Kong, Thailand, Singapore, Malaysia, Indonesia and Philippine. The M/V TRANSFUTURE 5 is a car carrier from Japan to the Oceania region. She departs Japan and make every 6 week cruise, visiting Australia and New Zealand. Figure 1 illustrates typical route of M/V FUJITRANS WORLD and M/V TRANSFUTURE 5.

(2) Atmospheric sampling system aboard VOS

From 2002 to 2005, M/V FUJITRANS WORLD contributed our atmospheric observation program, when she was served to Japan-Oceania route. We were able to use

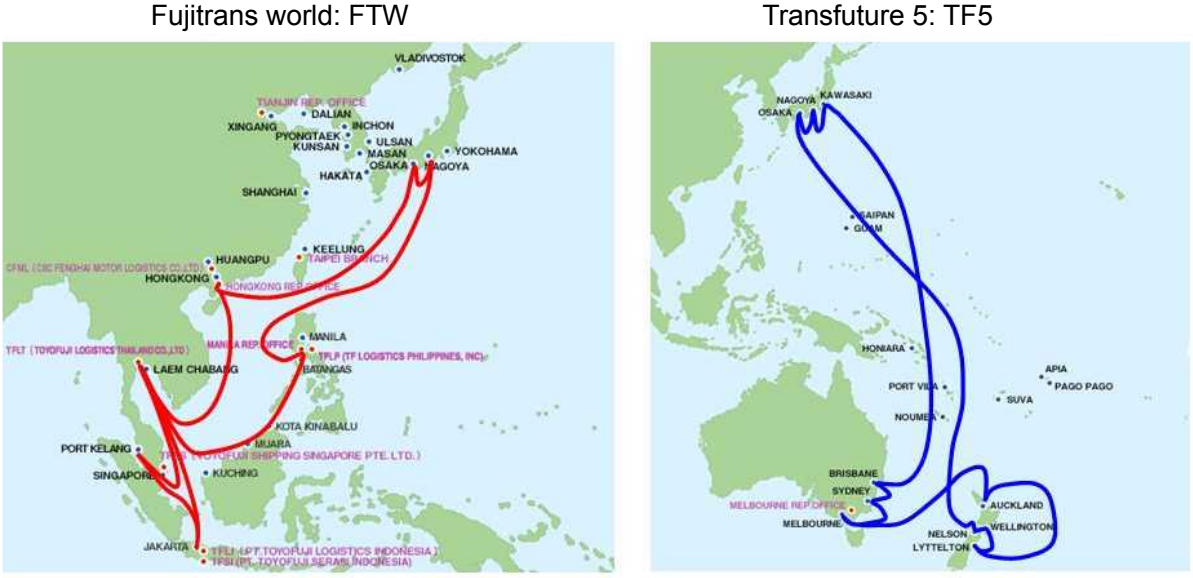

Figure 1. Typical routes for Japan–South East Asia (FTW) and Japan–Oceania (TF5) voyages

pre-installed facilities in her observation laboratory. Many of instruments were re-installed as same as previously. We use a series of seven glass bottles for a cruise, and sampling covers from 15°N to 5°S. The air intake is installed at the compass deck of the ship. Air is drawn by a metal bellows pump and dried with a cryogenic trap. For many species including methane, nitrous oxide, carbon monoxide, glass bottles of 2.5L are automatically pressurized at 1.5 MPa when the ship crosses the latitude set by a GPS navigation system. Stainless steel canisters of 3L are used for air samples of halocarbons and volatile organic compounds. After being transported to the laboratory, the air samples are analyzed with gas chromatographs coupled with a mass spectrometer, an electron capture detector, and a flame ionization detector.

(3) Continuous measurement system aboard VOS

We deployed infrared and ultraviolet absorption-based systems for continuous measurements of ozone and carbon monoxide, respectively, and have added a black carbon monitor in 2008. Operating continuous instruments is challenging since measurement condition is severer in ships than ground-based stations by large changes in temperature, ship-derived vibrations, and short-period for maintenance at calling port in Japan. For carbon monoxide measurement, instability with gas standards at low concentration (tens to hundreds of ppbv) often makes problem in conducting long-term monitoring. Because of wide range of linearity, infrared-absorption based instrument we use in this program make use of single standard gas at high concentration (several ppmv), as high concentration standard gassed are more table for long-term storage. Black carbon instrument is based on detection of both reflection and scattering of infrared beams.

4. Results

(1) Long-lived greenhouse gases in Asia/Oceania regions

Observed concentrations of methane and nitrous oxide in South East Asia were substantially higher than in those observed on board Japan-Oceania cruise, which is close to the global background level (Figure 2). The effect from continental sources of South East Asia was suggested. The source for methane, carbon monoxide, and nitrous oxide is likely specific to the tropical zone in this area. Concentrations of alternative fluorocarbons such as HFC-134a were generally higher over South East Asian than over western Pacific, suggesting that South East Asia is now a big emitter to the global atmosphere.

(2) Short-lived greenhouse gases in Asia/Oceania regions

Tropospheric ozone levels over South East Asia and over the western Pacific (Japan-Oceania cruise) are compared (Figure 3). The ozone levels at the latitude higher than 10°N are comparable. In contrast, the ozone levels at tropics (i.e., 10°N to 5S°) are higher in South East Asia than in he western Pacific. This strongly suggests that the photochemical production of ozone in South East Asia is positive while negative over the western Pacific, resulting in overall higher concentrations in South East Asia. Combined with satellite data and meteorological analysis, carbon monoxide was found to be affected by biomass burning occurred in Indonesia during El Nino year of 2006. The main source region is attributed to peat land in Borneo Island.

(3) Novel instrumentation for continuous measurements of methane and black carbon

Cavity ring-down spectrometry (CRDS) was used for continuous measurement of methane. It was found from laboratory experiments and comparison of CRDS continuous

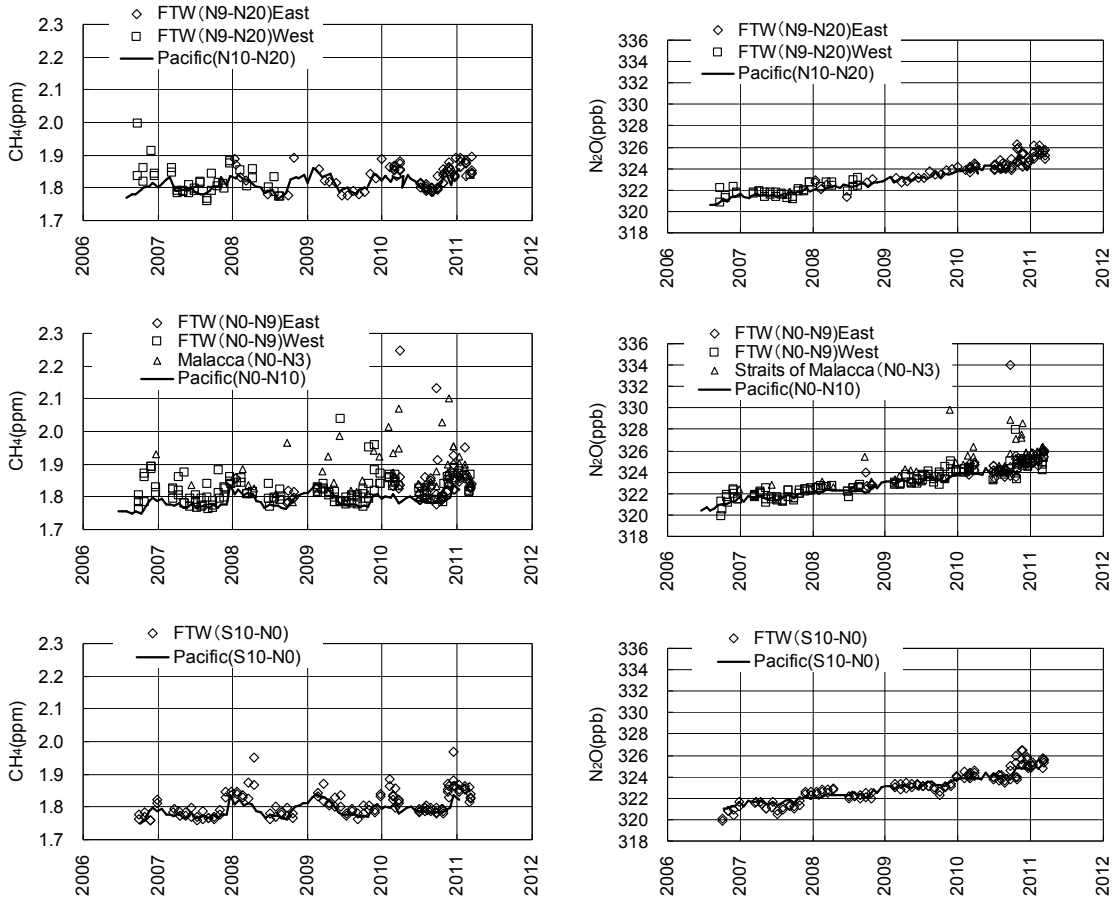

Figure 2. Trends of CH_4 and N_2O in the South East Asia (FTW, Malacca) and the Oceania (Pacific)

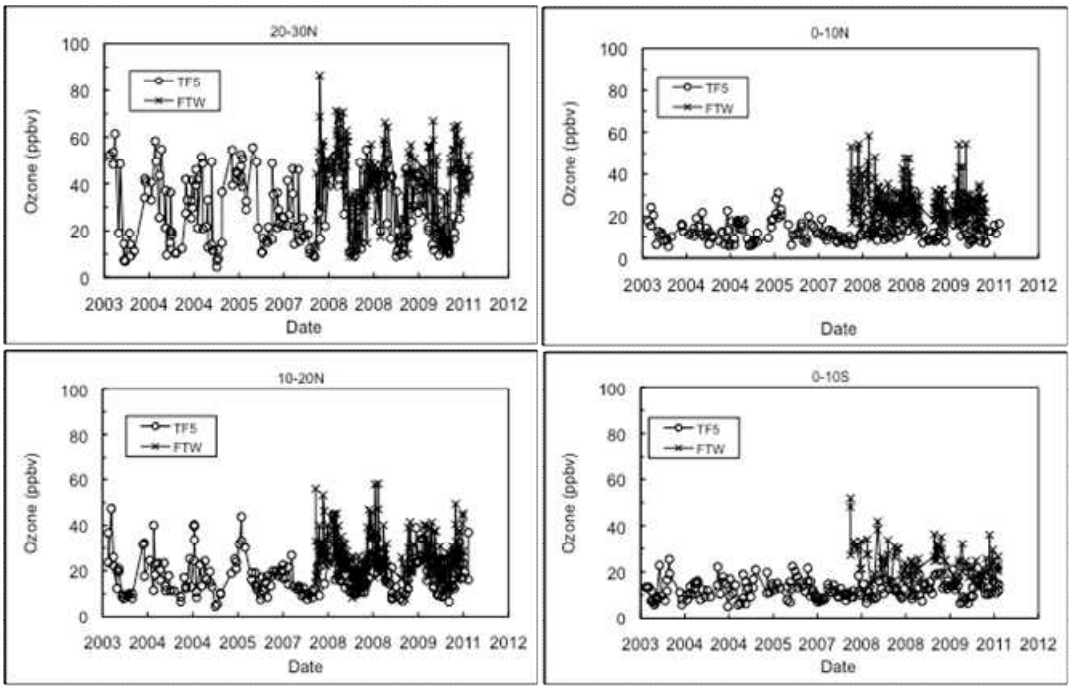

Figure 3. Trends of tropospheric ozone in the South East Asia (FTW) and the Oceania (TF5)

measurements to analysis of bottle samples that CRDS is subject to water vapor interference, particularly when ambient air is humid, as is the case of shipboard sampling.

5. Discussions

It was found that concentration levels of methane and nitrous oxide observed on Japan -South East Asia cruises were much higher than those on Japan-Oceania cruises. The data of multiple species obtained with bottle sampling and subsequent laboratory analyses suggest that there exist some sources for methane, nitrous oxides, and carbon monoxide in this tropical region of the South East Asia. The observed dataset for alternative fluorocarbons suggest that anthropogenic emissions of HFC-134a were strong in South East Asia. Photochemical formation of tropospheric ozone is very active in South East Asia, even over the ocean, due likely to sustained with the co-presence of abundant precursors and sunlight. Highly time-resolved measurements of carbon monoxide revealed the importance of long-range transport of plumes originated from biomass burning in South East Asia. In order to enhance capability of source characterization, further installation of continuous monitors would be especially useful. Full year observation will give more information for the analysis of emissions of these trace species from South East Asian region.