

**Long-term, comprehensive observation of long-lived greenhouse gases and short-lived climate pollutants in the Asia-Oceania Regions (Abstract of the Final Report)**

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### 1. Introduction

While carbon dioxide (CO<sub>2</sub>) is one of the most important greenhouse gases, other trace species including methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), anthropogenic halocarbons, tropospheric ozone (O<sub>3</sub>), 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 (CO) 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. In 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 and nitrous oxide. 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.



**Figure 1.** Typical routes for Japan–South East Asia (FTW, right panel) and Japan–Oceania (TF5, left panel) voyages

### 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

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, and 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 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.

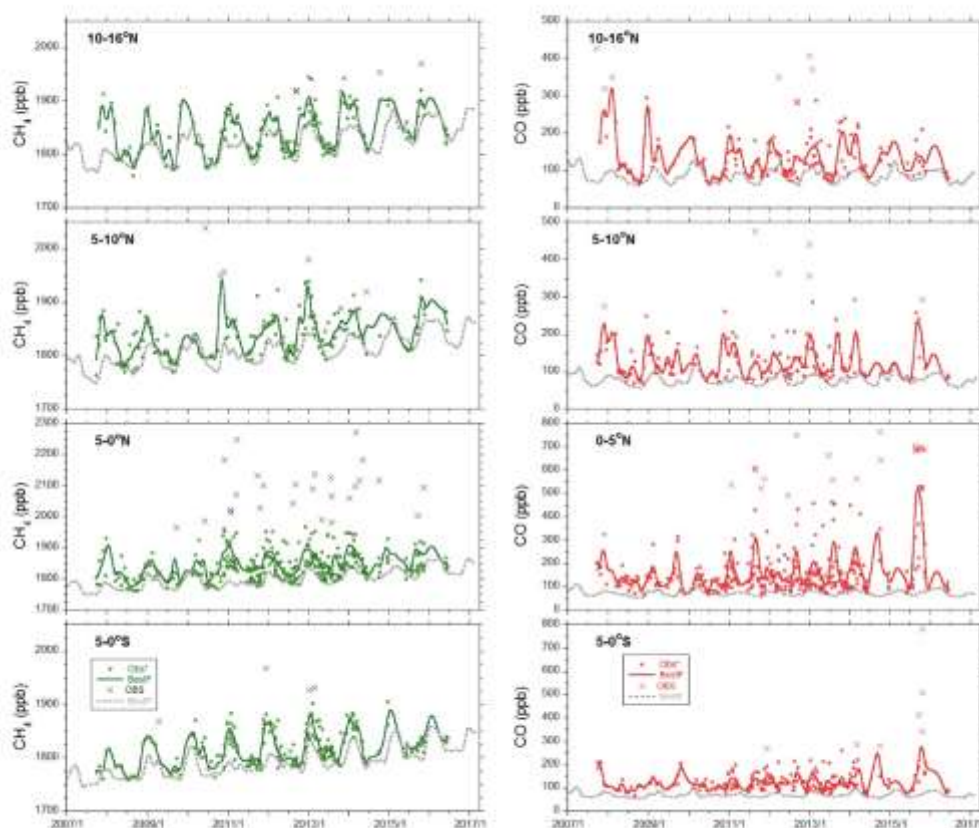
#### (3) Continuous measurement system aboard VOS

We deployed infrared and ultraviolet absorption-based systems for continuous measurements of ozone and carbon monoxide, respectively, and have added a PM/optical black carbon monitor (OBC) recently. 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 stable 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 Southeast Asia/Oceania regions

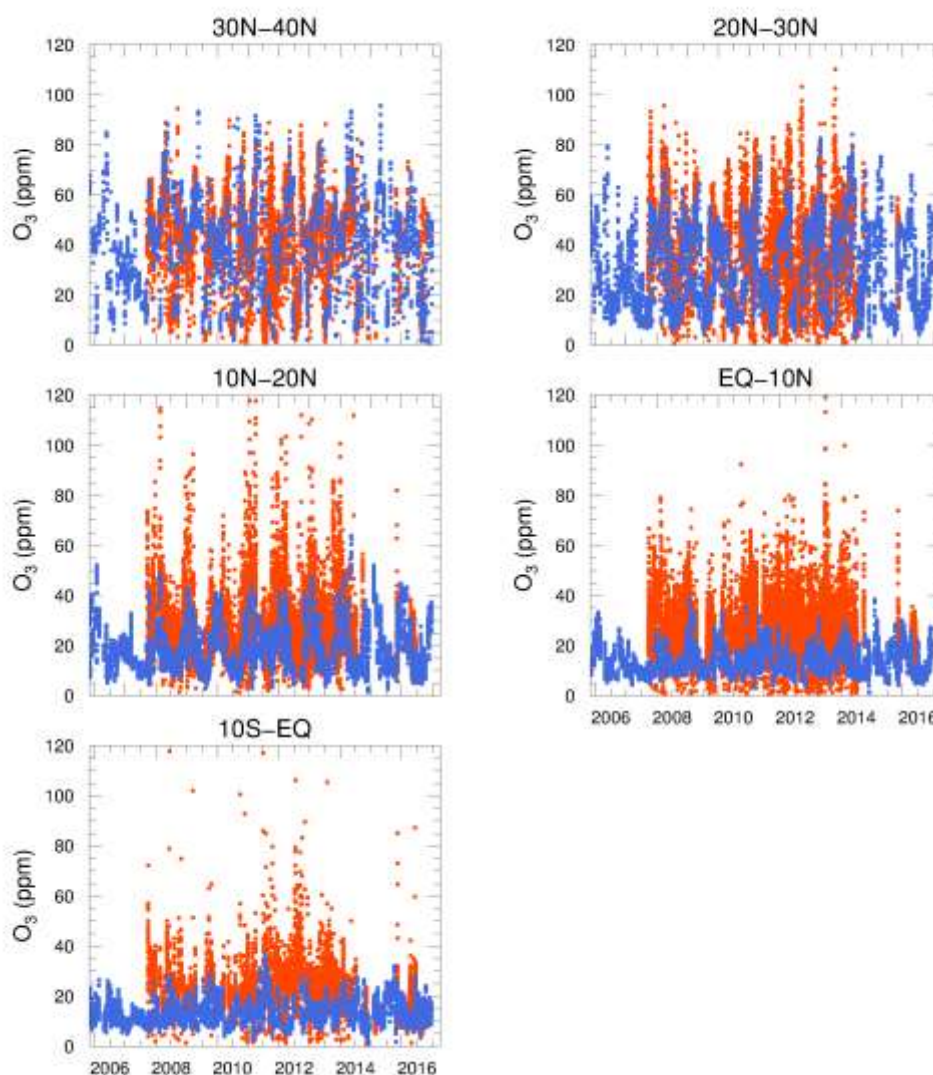
Figure 2 shows the concentrations of methane and carbon monoxide observed in Southeast Asia and western North Pacific. The concentrations in Southeast Asia were substantially enhanced compared to those observed in the western Pacific (Japan-Oceania cruises), which is close to the global background level. This suggests that the effect from continental sources of Southeast Asia was substantial. The source for methane, carbon monoxide, and nitrous oxide is likely specific to the tropical zone in this area.



**Figure 2.** Temporal variations of (left) methane and (right) carbon monoxide at 10-16 degN, 5-10 degN, 0-5 degN, and 0-5 degS, based on the analysis of flask sampled collected on Japan-Oceania (dashed lines) and Japan-Southeast Asia (solid circles and solid lines) cruises.

##### (2) Short-lived climate pollutants in Southeast Asia/Oceania regions

Figure 3 shows the concentrations of tropospheric ozone observed in Southeast Asia and western North Pacific. The tropospheric ozone levels in Southeast Asia were enhanced, compared to the western Pacific. In particular, the ozone levels at tropics are substantially enhanced. This strongly suggests that the photochemical production of ozone in Southeast Asia is positive while negative over the western Pacific, resulting in overall higher concentrations in Southeast Asia.

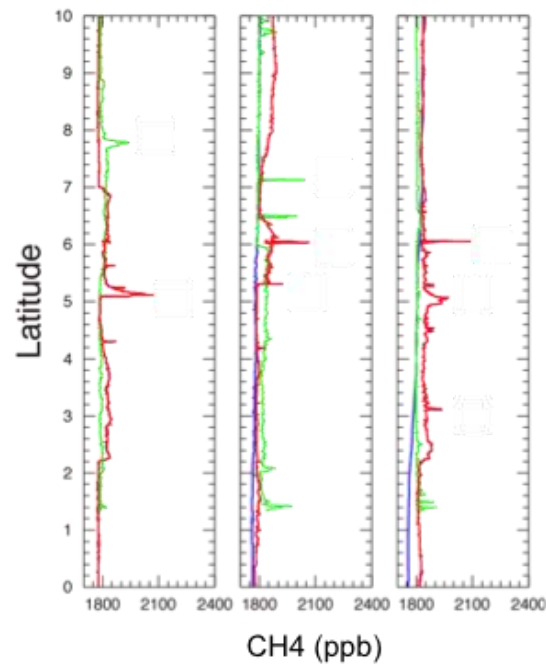


**Figure 3.** Temporal variations of ozone at 30-40 degN, 20-30 degN, 10-20 degN, 0-10 degN, and 0-10 degS, based on the continuous measurements onboard Japan-Oceania (blue dots) and Japan-Southeast Asia (red dots) cruises.

### (3) Emissions of methane from oil and gas platforms in Southeast Asia

Based on continuous measurements onboard a commercial cargo vessels, we detected intensive methane emissions from offshore platforms off the east coast of the Malay Peninsula and the northwest coast of Borneo (Figure 4). We used the observed peaks to estimate the emission rates of methane from offshore platforms. We also identified 112 offshore platforms in the Southeast Asian region (defined as: 15°N–10°S, 90°–140°E); thus, the resultant regional total emission rate is calculated as 3.3 kg s<sup>-1</sup>. The total regional annual emission of CH<sub>4</sub> from offshore platforms in the Southeast Asian region is estimated to be about 0.1 Tg y<sup>-1</sup>, associated with an uncertainty range of 0.02–0.32 Tg y<sup>-1</sup> (the median values of the lower and upper limits). EDGAR reports that annual CH<sub>4</sub> emissions from oil and gas production in 12 Southeast Asian countries (Brunei, Cambodia, Lao, Myanmar, Malaysia, the Philippines, Singapore, Thailand, Timor, Vietnam, Indonesia, and Papua New Guinea) were about 3.7 Tg y<sup>-1</sup> for 2010, which corresponds to about 1% of the 335 Tg y<sup>-1</sup> global anthropogenic CH<sub>4</sub> emissions<sup>18</sup>. Offshore CH<sub>4</sub> emissions account for about 8% (0.29 Tg y<sup>-1</sup>) of the total emissions from oil and gas production in the Southeast Asian region. Despite the large uncertainty inherent in the mass balance approach, our estimate displays relatively good agreement with that by EDGAR. However, we note substantial differences in the locations of the offshore platforms between the EDGAR inventory and those

determined by DMSP satellite observation. The distributions of point sources of CH<sub>4</sub> are an important uncertainty in the existing inventories. The relative contributions of the offshore CH<sub>4</sub> emissions to the regional CH<sub>4</sub> emissions in Southeast Asia are estimated to be about 3% for the oil and gas production sector (both offshore and onshore) (3.7 Tg y<sup>-1</sup>, as estimated by EDGAR), and about 0.2% for the anthropogenic sources (63 Tg y<sup>-1</sup>, as estimated by EDGAR).



**Figure 4.** Latitudinal distribution of 1-min mean methane mole fractions between 10°N and the equator along the Southeast Asian shipping routes.