Japan Geoscience Union Meeting 2015
(May 24th - 28th at Makuhari, Chiba, Japan)

AAS21-P01 Room:Convention Hall Time:May 27 18:15-19:30

Coating of black carbon aerosols and increase of their light absorption coefficient observed in Tokyo

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Black carbon (BC) aerosols can influence the climate due to the heating the atmosphere by their strong light absorption (direct effect). Light absorption coefficient of BC is varied with their size, shape and mixing states. BC is often coated with other aerosol component such as organics and sulfate, and the coating is supposed to cause the increase of light absorption coefficient (lens effect). Lens effect has still not fully understood because the coating material and amount in the real atmosphere have not understood quantitatively. Therefore it is important to observe change of light absorption coefficient of BC and their shape and mixing state simultaneously. Our group conducted intensive observation of various parameters of BC, including light absorption coefficient and coating amount, in huge city, Tokyo. In this paper, variation of light absorption coefficient of BC in Tokyo and its relation with BC coating is studied.

Intensive BC observation named as Black Carbon / Carbonaceous Aerosol Removal Experiment (BC-CARE) were conducted from July 28 to August 15 in 2014. The atmospheric BC was sampled and observed at the sixth floor (20m) in Tokyo University, located central Tokyo city area. Light absorption coefficient was measured using the photoacoustic soot spectrometer (PASS). Amplification factor (FA) by coating of BC was evaluated from the ratio of light absorption coefficient in the unheated sampled air to that in the 300 ℃ heated air, where volatile coating materials were removed from BC. BC mass concentration and the ratio of coating thickness with BC diameter (Dp/Dc) was measured using single-particle soot photometer (SP2) simultaneously. During observation period, aerosol particle were regularly sampled and the BC size, shape and mixing states were observed using a transmission electron microscope (TEM).

The significant increase in BC light absorption coefficient with the BC coating was measured on 29th July. Maximum increase was about 80% in this period. In the early half of this period, FA and Dp/Dc values were increased correlatingly from about 1.0 to about 1.8. However, in the latter half of this period, FA values were gradually decreased although Dp/Dc values kept high. Observation with TEM showed that many coating BC particles larger than 1μm in the early half of this period. In the latter half, particle size of coated BC was generally less than 0.5μm. Because the measuring range of particle size of SP2 is less than 1μm and PASS can measure BC about 1μm or more, the difference between the two period may be cause by the contribution of these larger BC particles. However, both PASS and SP2 can measure BC particles less than 0.5μm, these results cannot fully explain the observed difference in FA and Dp/Dc behavior.

Keywords: Black carbon, Light absorption coefficient, Lens effect, Electron microscope

AAS21-P02 Room:Convention Hall Time:May 27 18:15-19:30

Online Measurement of Aerosol Chemical Composition Classified by Black Carbon Mixing State using a LII-MS

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Aerosols have large influences on the radiation budget of the Earth's atmosphere by scattering or absorbing solar visible radiation (direct effect) and by altering cloud microphysical properties (indirect effect) (IPCC, 2013). These effects can significantly depend on the chemical composition and mixing state of aerosol particles. Specifically, the mixing state of sulfate, nitrate, and organics with black carbon (BC) is a key parameter for estimating the aerosol direct and indirect effects.

We have developed a new method to measure aerosol chemical composition classified by the mixing state with BC by combining laser induced incandescence (LII) and mass spectrometric (MS) methods (LII-MS, Miyakawa et al., AST, 48, 853-863, 2014). The purpose of this study is to further evaluate the LII-MS in the laboratory and also to understand the temporal variations of BC mixing state in a suburban area in Tokyo.

The LII-MS consists of a series of LII and MS. In the LII section, BC containing particles introduced into a near-infrared laser cavity can be efficiently vaporized and incandescence signals from the BC particles are detected. The sample air is then introduced into the MS section by a tandem nozzle to measure the mass concentrations of sulfate and nitrate (Takegawa et al., AST, 46, 428-443, 2012). The aerosol composition classified by the BC mixing state is obtained by alternatively switching on and off the LII laser.

Several modifications of the LII-MS hardware and additional experiments have been performed compared to the previous version presented by Miyakawa et al. (2014). First, the control electronics for switching the LII laser has been modified to achieve more stable operation. Second, the cycle of the LII laser on/off and zero-air modes has been modified to improve the quantification of the BC mixing state. Third, the performance of the tandem nozzle, which is one of the key components of the LII-MS, has been tested in the laboratory to investigate the stability and reproducibility of the particle transport efficiency. Ambient measurements have been conducted at the Tokyo Metropolitan University to test the overall performance of the LII-MS and to investigate the temporal variation of the BC mixing state in suburban air. Details of the laboratory experiments and ambient measurements will be presented and discussed.

Keywords: Aerosol, BC mixing state, Laser-induced incandescence, Mass spectrometer

AAS21-P03 Room:Convention Hall Time:May 27 18:15-19:30

Single-particle measurement of iron-containing dust particles using a laser-induced incandescence technique

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1. Introduction

Two major light-absorbing ubiquitous in earth atmosphere, black carbon (BC) and mineral dust, contribute positive climate forcing by absorbing solar radiation in atmosphere and by reducing albedo of snowpack. Light-absorption efficiency of mineral dust has shown to be strongly correlated with iron-content [Moosmueller 2012]. Reliable data for concentration and micro-physical properties (size distribution, mixing state, complex refractive index, etc.) of BC and iron-containing dust particles are important to provide a physical basis in climate simulations.

For statistical sampling and continual observations, we need a fast and real-time technique to measure concentrations and the microphysical properties of these light-absorbing particles. In this study, we experimentally show that the laser-induced incandescence (LII) technique, which has been used only for BC measurement so far, is also effective for identifying and measuring iron-containing dust particles.

2. Laser-induced incandescence (LII)

We use the single-particle soot photometer (SP2, Droplet measurement technology) to measure laser-induced incandescence and light-scattering signals for individual particles. In SP2, sample air containing aerosols are continually introduced into an intra-cavity Nd:YAG laser beam with 1064 wavelength. Refractory light-absorbing particles with boiling higher than ~3000K emit thermal radiation detectable in visible wavelength. In SP2, thermal radiation is measured at two distinct wavelength bands, the blue-band (300-500 nm) and the red-band (580-710 nm), to infer the spectra of incandescent light. As the spectra of thermal radiation shifts shorter wavelength as temperature increases, the blue-band to red-band signal ratio (color ratio) is a proxy of boiling point of the incandescing particle.

3. Identification of incandescing particles

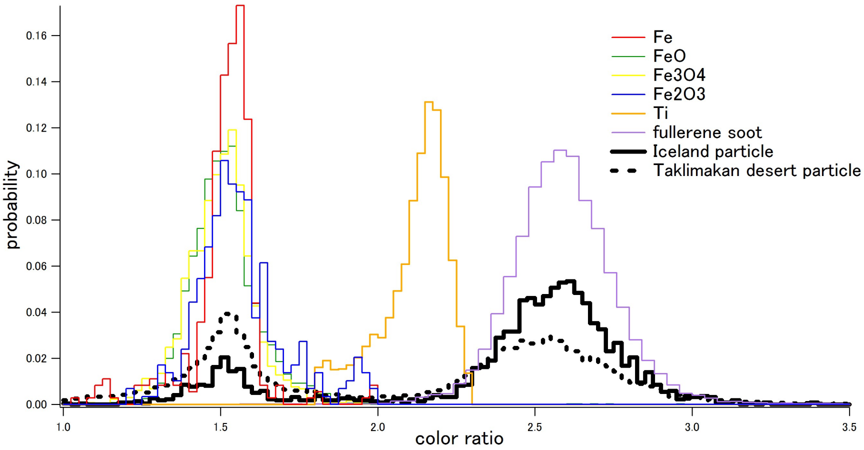
The probability distribution of measured color ratio for various laboratory samples and field dust samples are shown in Figure. For laboratory sample, there are three distinct modes in color ratio distribution: iron and iron oxides (~1.5), titanium (~2.0), and fullerene soot (~2.6) (standard material of BC). Therefore, we can distinguish incandescent materials (iron, iron oxides, titanium, BC) in a particle using the value of color ratio.

Figure also shows the result for two field samples: Iceland particle (provided by Dr. P. Dagsson-Waldhauserova, University of Iceland) and Taklimakan desert particle (provided by Dr. R. Tada (The University of Tokyo)). The color ratio distributions of these samples have two distinct modes with peaks around 1.5 and 2.6, corresponding to iron (and iron oxides) and fullerene soot, respectively. From the observed color ratio, the incandescing particles observed in the two field samples were identified as BC and iron-containing particles.

4. Calibration for iron measurement by LII

We have experimentally determined the relationships between blue-band signal intensity and mass of pure iron particle. This relationship was used as a calibration curve for estimating iron-content in field samples.

Keywords: aerosol, atmospheric chemistry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0.14
0.12
0.10
0.08
0.06
0.04
0.02
0.00
1.0
1.5
2.0
2.5
3.0
3.5
color ratio
Fe
FeO
Fe3O4
Fe2O3
Ti
fullerene soot
Iceland particle
Taklimakan desert particle

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AAS21-P04 Room:Convention Hall Time:May 27 18:15-19:30

Development and preliminary evaluation of an aerodynamic lens for focusing nanoparticles

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Aerosols have significant impacts on the radiation budget of the Earth's atmosphere by direct and indirect effects. Chemical composition of aerosol particles with diameters smaller than 100 nm (nanoparticles) is an important factor for understanding the formation process of aerosols. However, measuring chemical composition of nanoparticles is challenging because of their extremely low mass loadings in ambient air. Aerosol mass spectrometry coupled with an aerodynamic lens (ADL; Liu et al., AST, 22, 293, 1995), which can efficiently deliver aerosol particles into vacuum, is a useful tool for online measurements of aerosol composition. Although the concept of an aerodynamic lens for nanoparticles (nano-ADL) has been reported by previous studies (e.g., Wang et al., IJMS, 258, 30, 2006), the application of nano-ADL to ambient measurements has not been established. We have developed an improved nano-ADL based on the design of ADL for submicron particles (submicron-ADL) that has been used for aerosol mass spectrometry.

Laboratory experiments were performed using a custom-made particle generation system consisting of saturation and condensation tubes for oleic acid vapor. Monodisperse particles generated by a differential mobility analyzer were introduced into the submicron or nano ADL.

The detection of particles was performed using a Faraday cup. The transmission efficiency and particle beam width were measured using a movable knife-edge.

The submicron-ADL showed nearly 100% transmission efficiency for particles larger than 100 nm. On the other hand, the nano-ADL showed nearly 100% transmission efficiency at around 50 nm and decreased with increasing particle diameters, suggesting that the current design is favorable for the sampling of nanoparticles. The application of the nano-ADL to an aerosol mass spectrometry system will be discussed in the presentation.

Keywords: Aerosol, Nano particle, Aerodynamic lens, Mass spectrometry

AAS21-P05 Room:Convention Hall Time:May 27 18:15-19:30

Evaluation of an online analysis method of nitrate aerosols using a particle trap laser desorption mass spectrometer

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Aerosols play important roles in global air quality and climate change. Ammonium nitrate, which is generated via photochemical reaction of NOx, often contributes to a major fraction of fine particles in urban air. The gas-to-particle equilibrium reaction of ammonium nitrate exhibits strong temperature dependency near ambient temperatures, which often yields evaporative loss of particles during sampling.

We have developed a particle trap-laser desorption mass spectrometer (PT-LDMS) for online measurements of aerosol chemical compositions (Takegawa et al., AST, 46, 428, 2012). Factors affecting the quantification of nitrate aerosols, including, have not been fully characterized. The purpose of this study is to evaluate the quantification of nitrate aerosols by the PT-LDMS in the current configuration and also to investigate an optimal condition for ambient sampling.

Laboratory experiments were performed by generating monodisperse ammonium nitrate particles using an atomizer and differential mobility analyzer. The dependence of the sensitivity (defined as ion signals per collected nitrate mass) on the particle trap temperature was measured by altering the temperature of the particle trap holder between 280 and 313 K. The dependence of the sensitivity on the time after particle collection until laser irradiation (exposure time) was also measured. In addition, effects of the mixing state of nitrate particles on the sensitivity were evaluated by atomizing a mixed solution of ammonium nitrate and sulfate.

Preliminary results show that the sensitivity tended to decrease with increasing the exposure time for each temperature condition, which is likely due to evaporative loss of nitrate particles in the vacuum. On the other hand, the sensitivity did not exhibit significant temperature dependency at least for the temperature conditions during the experiments.

Possible mechanisms affecting the loss of nitrate particles, along with the effects of the mixing state on the sensitivity, will be presented and discussed.

Keywords: nitrate aerosols, online analysis method, mass spectrometer

AAS21-P06 Room:Convention Hall Time:May 27 18:15-19:30

Development of photovoltaic-driven atmospheric observation instrument Eco-MAXDOAS

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Atmospheric aerosols are an important factor controlling the Earth's climate. However, their complicated formation mechanisms and effects on climate are poorly understood. Under these circumstances, the MAX-DOAS technique that enables simultaneous measurements of aerosols and their gaseous precursors, such as nitrogen dioxide (NO_2), has been developed by Chiba University. The MAX-DOAS technique can derive the vertical distribution and column amount for aerosols and gases by utilizing the inversion analysis of scattered sunlight spectra measured at multiple viewing elevation angles. When MAX-DOAS observations are made, the line of sight must be clear at all elevation angles selected for the observations. In addition, there is the need for sufficient electric power, limiting the observation site. To solve these limitations, we developed the new instrument, called Eco-MAXDOAS, using the photovoltaics as a power source. For this development, we removed a temperature controller to reduce the power consumption. Instead, a shutter was introduced just before the entrance slit of the spectrometer. With this modification, it was made possible to take dark count measurements more often than for the normal MAX-DOAS instrument by closing the shutter between observations at different elevation angles. We tested the Eco-MAXDOAS and found that the spectrometer temperature changed by less than ± 0.3 degrees for 3 min. interval of changing elevation angles. Using dark count data obtained before and after a scattered sunlight observation was made, the SNR was estimated to be about 10000. This supports that analysis for a differential absorption as small as 10^{-4}(0.01%) is possible. In addition, we conducted continuous observations using 30-W and 60-W solar panels. When the 30-W solar panel was used, the Eco-MAXDOAS terminated in few days. On the other hand, using the 60-W solar panel prolonged the operation time period and the Eco-MAXDOAS worked continuously for over full test observation period of two weeks under usual weather conditions in winter. In this presentation, we also assess the performance for the Eco-MAXDOAS observation by comparing aerosol an NO_2 data retrieved from Eco-MAXDOAS and MAX-DOAS observations.

Keywords: MAX-DOAS, solar power, aerosol, nitrogen dioxide

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Measurement of stable isotope ratios of atmospheric carbon dioxide by wavelength modulation absorption spectroscopy with

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Since the industrial revolution, the concentration of carbon dioxide, a greenhouse gas is increasing every year, it has become a cause of global warming. Carbon dioxide is released from various emission sources and is absorbed to different sinks. Carbon dioxide is circulated among the atmosphere, hydrosphere, and geosphere. For suppression global warming, there is a need to accurately grasp the emissions and removals of carbon dioxide between these reservoirs. Measurement of concentration and stable isotope ratio of carbon dioxide provide us useful information in order to elucidate the carbon cycle. Therefore, we focus on the approach to use the stable isotope as an index. Carbon dioxide has stable isotopes, $^{12}CO_2$, $^{13}CO_2$, and $^{12}C^{18}O^{16}O$, it is known that these isotope ratios are different for each emission source.

As one of conventional measurement techniques of isotope ratios which has been used, there is isotope ratio mass spectrometry (IRMS) technique. Although this technique has a very high measurement precision (0.01-0.1 ‰), it cannot be measured outside the laboratory because the device is a large. Therefore, a laser absorption spectroscopy in recent years has been attracting attention. This technique is excellent in portability since the device can be made compact. Also, because suitable selection of absorption lines prevents interference from other species, the sample can be introduced directly to the device. Therefore, the device can be carried to the location where we want to measure the stable isotope ratio of carbon dioxide, it is possible to perform real-time measurements at high time resolution.

In this study, we constructed a high-precision measuring device of atmospheric carbon dioxide stable isotope ratios, using wavelength modulation absorption spectroscopy with a newly developed 2.8 μm DFB laser and a Heriot type multi-pass reflection cell. In the measurement, absorption lines are selected in terms of being continuous of $^{12}CO_2$, $^{13}CO_2$ and $^{12}C^{18}O^{16}O$ and small interference from water around 2.8 μm. Wavelength modulation absorption spectroscopy allows improved sensitivity and zero background measurement by modulating wavelengths at a high frequency and performing heterodyne detection.

Keywords: carbon dioxide, stable isotope, near-infrared absorption spectroscopy

AAS21-P08 Room:Convention Hall Time:May 27 18:15-19:30

Seasonal variation of Pb stable isotope ratio in PM observed in Yakushima Is.

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A total of 36 sets of PM10 and PM2.5 aerosol particles collected from Yakushima Island during a period from January to June 2013 were determined for atmospheric Pb concentrations. Among these samples, 36 sets of samples representing two seasons, winter and spring, were selected for measuring Pb isotopic compositions to determine the relative contributions of various pollution sources. Results reveal an evident seasonality of high spring and comparatively low winter Pb concentrations, resembling those observed in Beijing, China as well as many East Asian countries. Together with synoptic atmospheric conditions analysis, the seasonal pattern is attributable to the impact of long-range transport of Pb-rich anthropogenic aerosols from the Chinese pollution outflows in the northeast monsoon. Results of206Pb/207Pb and 208Pb/207Pb isotope ratios show a minimum at February, thereafter increasing progressively to March and reaching a maximum at April. From January to June, Pb isotope ratios are quite comparable with those measured in China, especially Dalian and Tianjin. Again this demonstrates Yakushima Island has already been affected by continental pollution of long-range transport during the northeast monsoon season beginning in winter and ending in late spring.

AAS21-P09 Room:Convention Hall Time:May 27 18:15-19:30

Comprehensive analyses of air pollutants at Suzu, Noto peninsula, Japan

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Recent remarkable economic progress in East Asia has increased emissions of air pollutants such as nitrogen oxides, sulfur dioxide, ammonia and volatile organic compounds. Such pollutants are transported over a long distance with photochemical reactions and then arrive at Japan as aged species such as gas phase nitric acid (HNO_3), particulate nitrate (NO_3^-), sulfate (SO_4), peroxy nitrates (PNs), organic nitrates (ONs), ammonium (NH_4^+), organic aerosol (Org) and so on. Many researches on the transboundary pollution in Japan focus on the areas of western Japan near the Asian continent. On the other hand, it is important to investigate the transboundary air pollution in the central Japan area because there are many large cities. In addition, the central Japan is moderately far from the Asian continent, so that more aged air mass from the Asian continent would come at the central Japan. In this research, continuous observations of such air pollutants at Suzu, Noto peninsula, Japan. Suzu is representative remote area and located at the central Japan.

Observations are performed at NOTOGRO (NOTO Ground-based Research Observatory) supersite (37.45^{o}N, 137.36^{o}E) in Suzu. Total odd nitrogen species (NO_y) and total nitrate ($T.NO_3 = HNO_3 + NO_3^-$) were measured by a scrubber difference / NO-O_3 chemiluminescence method. PNs and ONs were measured by a thermal dissociation / cavity attenuated phase shift spectroscopy method. CO, O_3 and SO_2 were observed by non-dispersive IR, UV absorption, and pulsed UV fluorescence methods, respectively. Org, NH_4^+, SO_4 and fine NO_3^- were measured by an aerosol mass spectrometer. SO_4 was also measured by a thermal reduction / pulsed UV fluorescence method.

Results of SO_4 and $T.NO_3$ were reported in this abstract. The air mass origins were classified into five groups; China and Korea (CK) North China (NC), Japan (JP), Russia (RU), and Sea (S), by backward trajectory analyses. Concentrations of air pollutants from CK air mass origin were generally high. In many cases, SO_4 concentrations from JP were lower than those from CK, while $T.NO_3$ concentrations from JP were similar to those from CK. In addition, SO_4 concentrations from CK were very high, but $T.NO_3$ concentrations from CK were not, in August 2013 and June 2014. Many of SO_4 in remote area is present as fine particles while NO_3^- exists as coarse particles mainly. The deposition velocity of coarse particles (ca. 0.03-1.24 cm s^{-1}) is larger than that of fine aerosols (ca. 0.05-0.6 cm s^{-1}). In addition, the deposition velocity of HNO_3 (ca. 1-8 cm s^{-1}) is larger than that of NO_3^-. The lifetime of SO_4 is longer than that of $T.NO_3$, so that SO_4 contributes strongly to the transboundary air pollution at Suzu, in comparison with $T.NO_3$. In this presentation, more detailed results and discussion, including other air pollutants will be described.

Keywords: Long-range transport, Air pollutants, East Asia

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Long-term observation of CCN characteristics at Suzu, Noto peninsula, Japan

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Atmospheric aerosols can play a significant role in regulating radiative properties and lifetimes of clouds by acting as cloud condensation nuclei (CCN). The atmospheric concentrations of CCN are perturbed by major anthropogenic emission sources. This is particularly true in East Asia and its downwind regions which can be considered by far as one of the global hotspots of anthropogenic aerosols. Despite the regional relevance, there are still few reports on the variations of CCN properties in relation to the distinct monsoon and seasonal climate in the region.

In this study, we performed long-term monitoring of CCN activity at the remote coastal site along the Sea of Japan, namely at the tip of Noto peninsula. Such a remote geographical setting is considered ideal for characterizing CCN over extended periods with particular emphasis on the effects of typical seasonal atmospheric transport patterns and occasional outflow of atmospheric pollutants.

The measurement was conducted at the NOTOGRO (acronym for NOTO Ground-based Research Observatory) station in Suzu city (37.45 oN, 137.36 oE) at the tip of a peninsula. The PM10 inlet (14.7 m a.g.l.) provided sample air into the building for the aerosol in-situ measurements. The ambient aerosol was dried by silica-gel before entering into a differential mobility analyzer (DMA, Model 3081, TSI) for size selection. The mono-dispersed aerosol was then guided to a condensation particle counter (CPC, Model 3785, TSI) and a continuous flow thermal gradient CCN counter (CCNC, CCN-100, DMT). The CCNC was operated at four different supersaturation conditions (SS=0.1%, 0.2%, 0.5%, 0.8%). We employed SMCA (Scanning mobility CCN analysis) method for obtaining the activation diameter d_{act} for each SS (Moore et al., Aerosol Sci. Tech., 2010) from which the hygroscopicity parameter κ can be derived (Peters and Kreidenweis, Atmos. Chem. Phys., 2007). The bulk chemical composition of non-refractory submicorometer-sized aerosols was also measured simultaneously by an aerosol chemical speciation monitor (ACSM, Aerodyne Inc.).

The spring and autumn months were characterized by large variation in mass and composition of CCN relevant particles. This was caused by the subsequent arrival of extra-tropical cyclone and anti-cyclone often accompanying transport of polluted continental air-mass. However, unexpectedly high concentrations of fine particles persisted even in summer period, characterized by relatively large contribution of organics. There was a rather good correlation between the abundance of organics (relative to sulfate) in the aerosol bulk chemical composition and the hygroscopicity κ, such that κ values were particularly low during the summer period.

There was a further evidence that the temporal variation in κ value was not in phase with org/inorg mass ratios alone, but also weakly correlated with the organic composition. Analysis of mass spectra from ACSM revealed that the oxidative state of organics also influenced the κ_{org}. These findings highlight the importance of the temporal variations in particle chemistry as well as their aging states for conducting CCN closure in the region.

Keywords: aerosol, CCN, Organics, Long-range transport

AAS21-P11 Room:Convention Hall Time:May 27 18:15-19:30

Enhancement of dimethylsulfide production by anoxic stress in natural seawater

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Dimethylsulfide (DMS) is the dominant reduced sulfur species in the ocean and an important source of aerosols particles and clouds in the marine atmosphere. Marine DMS plays a key role in the climate system of the Earth. A better knowledge of the distribution of marine DMS and its controlling factors is required. Previous field studies have reported the formation of DMS peak upper anoxic layer though the governing processes have not been clearly understood yet. Here we show the first direct evidence for the enhancement of DMS production caused by anoxic stress.

Isotope tracer experiments were made using the oxic and anoxic coastal seawater to quantitatively evaluate DMS production rates in three processes; cleavage of dimethylsulfoniopropionate (DMSP), dimethylsulfoxide reduction and phytoplankton release.

Under the anoxic condition, DMS production was considerably enhanced and DMS consumption was inhibited, resulting in an 8-fold higher rate of gross DMS production than that under the oxic condition. While almost all DMS was derived from DMSP cleavage (99%) under the oxic condition, the DMS production under the anoxic condition was mainly due to direct release of DMS from phytoplankton (63%). These results demonstrate that phytoplankton suffered from anoxic stress emits DMS into the seawater, resulting in a rise in DMS levels. Anoxic stress is indicated to be one of important environmental factors in the dynamics of marine DMS, suggesting the possible global importance due to a ubiquity of anoxic conditions in the coastal oceans.

Keywords: dimethylsulfide, dimethylsulfoniopropionate, dissolved oxygen

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Atmospheric hydorogen mesurements in the western North Pacific

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Molecular hydrogen (H_2) plays a significant role in global atmospheric chemistry due to its role in CH_4 -CO-OH cycling and water vapor source in the stratosphere. The balance of H_2 could change with the implementation of a new H_2 energy carrier. Therefore, it is important to establish its global budget and atmospheric trend (WMO/GAW Report No.197, 2011).

We started atmospheric H_2 measurement at Minamitorishima (MNM) from Nov. 2011. The measurement system using a GC-RGD (gas chromatographs equipped with a reduction gas detector) was installed for sirmulteneouse analyses of H2 and carbon monoxide at 3 stations of MNM, Yonagunijima (YON), and Ryori (RYO) operated by Japan Meteorological Agency (JMA). In this study, high-precision H2 standard gases are prepared to determine the atmospheric concentrations from the output signal of the GC/RGD.

The H_2 concentrations at MNM varied seasonally from 490 ppb to 560 ppb with a yearly mean of about 520 ppb. The H_2 variations often show distinct episodic events with enhanced concentrations on a synoptic scale in winter. This result indicates that H_2 increases are caused by the long-range transport of Asian polluted air masses to the station, suggesting that H_2 is a good tracer for identifying continental air masses in winter season. On the other hand, the H_2 concentrations are higher and stable in summer season. This result indicates that the maritime air masses are dominated, and the influence of soil absorption was small.

Keywords: hydorogen

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Observation of optical and chemical properties of aerosols at a forest site in Kii Peninsula during summer of 2014

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Aerosols scatter and absorb solar radiation and influence to the radiation balance in the atmosphere. Forests are a significant source of both primary biological aerosol particles (PBAPs) and biogenic secondary organic aerosols (BSOAs). In addition, polluted air masses including sulfate and black carbon (BC) particles may also be long range transported to forest areas in Japan. If the BC particles were coated with inorganic and/or organic materials during the long-range transport, the light absorption of BC could be enhanced due to the lensing effect. However, relations of aerosol optical properties including lensing effect with chemical properties of aerosols in Asian forest area have not been well understood. In this work, optical and chemical properties of aerosols were simultaneously measured in a forest site in Japan.

The observations were conducted from 17 July to 3 September 2014 at the Wakayama Forest Research Station, Kyoto University, Japan (34.06N, 135.52E, around 535 m above sea level), which is located in the central part of Kii Peninsula. Ambient particles were sampled from an inlet placed at 6.4 m above ground level. Absorption and scattering coefficients of PM1 particles were measured using two photoacoustic spectrometers (PASS-3 at λ = 405, 532, 781 nm and PAX at λ = 375 nm, DMT) after passing aerosols through a heater controlled at 300 ℃ or a bypass line by switching ball valves every 10 min. By comparing absorption coefficients at 781 nm with and without heating, increase in BC light absorption due to coating can be estimated. Mass concentrations of non-refractory materials were measured using an aerosol mass spectrometer (AMS, Aerodyne Research). Mass concentrations of elemental carbon (EC) and organic carbon (OC) were also measured by thermo-optical technique using a semi-continuous EC/OC analyzer (Sunset Lab.). Size distributions of particles were measured using a scanning mobility particle sizer (SMPS, TSI) and optical particle counters (OPCs, RION and TSI). Aerosol particles were also collected using an impactor for morphological analysis using a transmission electron microscope (TEM). Optical thickness (AOT) and extinction Angstrom exponent of aerosols were also measured using a Skyradiometer (Prede). In the presentation, relation between the obtained optical properties with chemical and physical properties of aerosols will be discussed.

Keywords: Aerosol optical property, Ambient measurement, Forest site, Biogenic SOA, Lensing effect, Photoacoustic spectroscopy

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AAS21-P14 Room:Convention Hall Time:May 27 18:15-19:30

Chemistry with KROME: Dynamical photochemical solver coupled to chemical disequilibrium and sulfur isotopes

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The study of chemical networks of Earth's geological past such as the Archean and Exoplanetary atmospheres requires the resolutions larges number of chemical reactions. This necessity is based on the lack of observational parameters abundant in today's planet Earth or neighbor planets in the solar system. The aim of this work is to construct a planetary atmosphere chemical network solver that relies on a minimal number of observational parameters.

We present here the latest development in our effort to develop such model. Our previous report presented the efficiency of the chemical solver for a large number of chemical species and reaction networks. In this report we present a photochemical dynamic core capable of solving ultraviolet opacities and photo-dissociation reaction rates at each step of the calculation. Additionally the model has been equipped with a set of equations to calculate disequilibrium effects on the chemical network. The stability and robustness of the code has been tested for a large network with more than 500 reactions interlinking more than 40 chemical species. The results obtained so far have been contrasted with the most common chemical codes available in the literature for benchmark.

Keywords: Archean Atmosphere, Sulfur Isotopes

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Urban fog and atmospheric pollution: contrasted effects on pollutants in Lyon (France)

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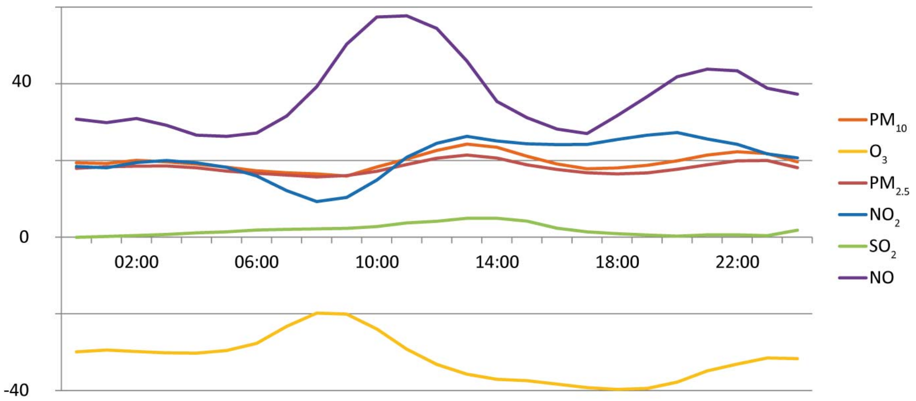
Lyon (500 000 inhabitants), located in the southeastern France, is an industrial city, with a polluted atmosphere and many days of fog. This unfavorable atmospheric situation poses many health problems to people and harms the attractiveness of the city and the development of trade. Greater Lyon has the highest levels of fine particulates and nitrogen dioxide in the Rhone-Alpes region and is prosecuted by the European Union for non-compliance with the Directive of 21 May 2008 concerning the quality of ambient air and cleaner air for Europe. This area is disadvantaged by its geographical situation, with the presence of two major rivers (Rhone and Saone) and many reliefs, influencing the conditions of dispersion of pollutants in the atmosphere and makes the Lyon an atmospherically sensitive city, despite prevailing winds North-South or South-North oriented which tend to favor the dispersion of pollutants In addition, Lyon concentrates pollution from traffic, industrial, and tertiary sectors. In winter, temperature inversions (temperature higher in altitude than on the ground) promote stagnation of pollutants at low altitude. This phenomenon is amplified considerably during episodes of urban mist. As a first step, the evolution of the number of foggy days is studied, and explanatory factors are proposed. The months of October to January are the most affected, with an average of 7 days of fog in December and January, over the period 1949-2013. It is thus noticed a steady decline in the number of foggy days since 1921 until early 2000s, from more than 90 days to only fifteen, followed by stagnation and a slight increase in recent years. This trend is compared to the annual minimum temperatures of Lyon, following an opposite trend, with an increase until the late 2000s, from 6 degrees C to 9 degrees C, followed by a stagnation and a slight decrease. A strong relationship is obtained between these two parameters, but the average wind speed may also explain the decrease, limiting conditions for fog formation, with a more important mixing of the air. Again, it can be noticed an increase in the average wind speed and in the proportion of winds faster than 1 m / s until the early 2000s, and then a decrease. Finally, better controls of industrial air emissions also explain the decrease in the number of days with fog over the long term. In a second step, the concentrations of air pollutants (PM10, PM2.5, 03, NO2, NO and SO2) are studied with and without fog. A station in the center of the city records these pollutants since 2007 continuously, every hour. In 2013, during foggy days and comparatively to clear days, it is found a strong increase in PM 10 and PM 2.5, respectively from 22.3 μg.m-3 and 16.8 μg.m-3 to 39.7 μg.m-3 and 33 μg.m-3, above recommended thresholds. Concentrations of nitric oxide (NO) and nitrogen dioxide (NO2) are respectively 9.0 μg.m-3 and 27.7 μg.m-3 during clear days, passing to 52 8 μg.m-3 and 50.5 μg.m-3 during foggy days, exceeding the recommended values here too. And inversely, as a result of sunlight blocking by water droplets in suspension, there is a decrease in O3, from 42.4 μg.m-3 to 13.9 μg.m-3, while the SO remains stable and very low, at 1.2 μg.m-3(fig. 1). These observations made in 2013 are the same since 2007. Finally, the evolution of the concentration of PM 10 is studied during persisting foggy days, hours after hours. There is a steady and dramatic increase in the concentration with consecutive days of fog, going to an increase of 40 μg.m-3 on the fourth day. In conclusion, air pollution in urban areas under clear skies is already a major concern, but the action of the latter on health is particularly worrying during episodes of urban smog.

Keywords: fog, atmospheric pollution, particulate matter, Lyon, France



Average hourly evolution of pollutant concentrations (µg.m^{-3}) during foggy days, compared to standard days (Lyon city center, France ; 2007-2013)

AAS21-P16 Room:Convention Hall Time:May 27 18:15-19:30

Impacts of weather regimes on PM10 pollution peaks in Rhone-Alpes (France)

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Pollution in particulate matter is a more and more significant problem, especially in urban areas, and widely publicized when regulatory thresholds are exceeded. Health consequences have to be carefully considered, for both short-term and long-term effects. First, this paper describes the spatial and temporal distribution of PM10 concentrations (particulate matter with a diameter inferior to 10 μm) in urban environments in the Rhone-Alpes administrative area, located in South-east France. Pollution monitoring stations are grouped together by ascending hierarchical classification. The goal is to identify similar patterns on a regional scale. Then, this PM10 distribution is analyzed according to synoptic-scale weather regimes. As the classic analysis between anticyclonic and cyclonic types appears insufficient, particulate matter concentrations are related to weather regimes according to Hess-Brezowsky classification system, which is widely used in Western Europe for numerous climate studies. The latter has been chosen after a review of the existing European classifications, and this analysis is one of the first to be made in France. Results show that strong annual disparities are observed amongst the different urban monitoring stations in Rhone-Alpes on both spatial and temporal scales. More precisely, stations in deep alpine valleys and in the regional capital city Lyon are the most polluted ones. Concerning monthly means, the most polluted months are obviously those in winter, due to the emitting sources considered. Steepest gaps are nonetheless better observed on an hourly basis, with two peaks at the end of the morning and at early evening. When analyzed according to the weather regimes, high pressure area over Central Europe weather regimes are prevailing during information and alert thresholds exceedances. More specifically, six regime types (Groswetterlagen) are strongly associated with pollution peaks. Two are cyclonic (WZ and NWZ) and their persistence during several consecutive days leads to a decrease in pollution. They are only associated with pollution peaks because they have represented a large part of global circulation during the last few years. By contrast, the most worrying regime types in regard with pollution peaks are anticyclonic : SWA, WA, HM, and BM. Those last two types, when persisting over several days, lead to a steep increase in pollutant concentrations.

Keywords: particulate matter, pollution peaks, weather regimes, Hess-Brezowsky, Lyon, France

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Height-resolved measurements of the aerosol size distributions in a temperate forest by tower observation system

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Dry and wet depositions are quite important for aerosol particles to be removed from the atmosphere. Also deposition of ammonium, sulfate, and nitrate contained in aerosol particles may contribute to potential acidification and eutrophication of the ecosystems. Both deposition processes are size specific, in particular, dry deposition of aerosol particles depends principally on particle size, atmospheric turbulence and stability and the collecting properties of the surface.

We conducted observational studies measuring the number size distributions of ambient submicron and ultrafine aerosol particles in a deciduous forest, during the summers of 2013 and 2014. The deciduous forest is located in suburban Tokyo, as a part of the experimental forest at the Tokyo University of Agriculture and Technology (Field Museum Tamakyuryo (FM Tama)). The observation site has a 30-m tall tower where we installed aerosol measurement instruments of the Scanning Mobility Particle Sizer and the Optical Particle Counter to explore the vertical profiles of the aerosol size distributions within and above the forest canopy. We report that the size distribution for submicron particles varied significantly with both temporally and vertically, depending on the wind field as well as the relative humidity related to the hygroscopic properties of aerosol particles.

Keywords: atmospheric aerosol, size distributions, tower observation

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Direct measurements of photochemical ozone production rate at a forest area in Japan during summer of 2014

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We developed a direct measurement system of photochemical ozone production rate in order to evaluate ozone concentration variations quantitatively. In fact, this system measures oxidant (Ox = O_3 + NO_2) production rate. The use of Ox can ignore the concentration variations of O_3 due to titration of O_3 by NO. The field campaign was performed at Wakayama, a remote site, in Japan during summer of 2014. Measurement parameters were photochemical net Ox production rate (P-L(Ox)), [O_3], [NO], [NO_2], [RO_2], [VOCs], OH reactivity, photolysis frequencies of various trace species and so on.

The P-L(Ox) measurement system has “reaction” and “reference” chambers. The reaction and reference chambers (17.1-cm inner diameter and 50-cm length) are made of quartz and Pyrex, respectively. Inner walls of both the chambers are coated with clear Teflon films to avoid wall loss of O_3. An outer wall of the reference chamber is coated with a UV-cut film (50% cutoff wave length of 405 nm). Both the chambers were put in an outdoor location to be exposed directly to sunlight. Ambient air is introduced into both the chambers. In the reaction chamber, photochemical reactions proceed to generate Ox. On the other hand, Ox is not generated in the reference chamber. The difference of Ox concentrations (ΔOx) in air from the two chambers is the Ox produced by photochemical reactions in the reaction chamber. The P-L(Ox) is obtained by dividing ΔOx by a mean residence time of air in the reaction chamber. Ox concentrations were obtained as follows. O_3 in Ox is converted into NO_2 by the reaction of O_3 with large excess of NO, and then the NO_2 concentration is measured by a laser-induced fluorescence technique.

The field campaign was conducted at Field Science Education and Research Center, Kyoto University, Wakayama Forest Research Station, in Wakayama Prefecture, Japan. Observation site is in forest area and anthropogenic sources of air pollutants are very low. Observations were conducted from 28 July to 8 August. Most periods of the campaign, O_3 concentrations were approximately 10 ppbv in the daytime. NO_x (= NO + NO_2) concentrations were less than about 1 ppbv throughout the campaign. BVOCs (Biogenic Volatile Organic Compounds) concentrations were high. Results of P-L(Ox) and Ox concentration on 6 August were reported in this abstract. A diurnal variations were observed for P-L(Ox) and Ox concentration, and the maximum rate and concentration were observed around noon. Ox concentration increased in the early morning, while P-L(Ox) was still 0 ppbv h^{-1} and then increased after a few hours. This result shows that O_3 concentration is increased by non-photochemical factors in the early morning. Vertical mixing of air is week at night and O_3 concentration near the surface of the ground decreases by deposition, reactions with olefin and so on. Surface of the ground is gradually warmed by sunlight in the morning and vertical mixing of air is activated. O_3 concentration would increase because of O_3 influx from above. P-L(Ox) achieved a peak value around noon and photochemical O_3 production was active in the daytime. Ox concentration did not increase around noon, however. This suggests that O_3 increase by photochemical production competes against O_3 decrease by non-photochemical factors such as deposition, advection, and reactions of O_3 with olefin.

Keywords: photochemical ozone production rate, oxidant, forest area

AAS21-P19 Room:Convention Hall Time:May 27 18:15-19:30

A global trend map of seasonal total column ozone using a MIROC3.2 nudged Chemistry-Climate Model

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We created a global trend map of seasonal total column ozone using the results of two CCM experiments to examine the influence of CFC on global total column ozone. The model used is the MIROC3.2 Chemistry-Climate Model nudged toward ERA-Interim reanalysis data. We performed two experiments. One experiment (REF-C1SD) uses observed ozone depleting substance (ODS) and greenhouse gas (GHG) concentrations that changes with time. In the other experiment (SEN-fODS1979), ODS concentration is fixed to the 1979 value. The REF-C1SD experiment shows a large decreasing seasonal total column trend globally during the CFC decreasing period (1979-1996). The trends have clear longitudinal structures at winter mid-latitudes and spring polar region.

Comparing the result of the REF-C1SD experiment with that of the SEN-fODS1979 experiment, the contributions of ODS and other processes except for ODS (for example, trends in ozone transport) are analyzed globally. We also show the trend map for the ODS decreasing period (1997-2011).

Keywords: Total Column Ozone, Trend, Chemistry-Climate Model, Global Map, ERA-Interim

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Trend analysis of satellite observed tropospheric NO2 vertical column density over East Asia

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Nitrogen dioxide plays a central role in the atmospheric environment as a toxic substance for respiratory system and precursors of ozone and aerosols. Furthermore, OH concentration is dependent on nitrogen dioxide concentration in the atmosphere. Although Hilboll et al.(2013) showed an increase in NO_2 concentration over Central Eastern China(CEC) until 2011 and a gradual decrease in NO_2 concentration over Japan until 2011, the latest trend until 2014 has not been reported yet. The time period is of interest, because it corresponds to the 12th 5-year-plan regulating NOx emissions in China and the period with the power substitution of thermal power generation for the nuclear power generation in Japan. In this study, we used two satellite datasets from OMI and GOME-2 that have been operationally observing tropospheric NO_2 VCD in recent years until 2014. Tropospheric NO_2 VCD trends in China and Japan were estimated based on the regression analysis for annual mean values. Although an increase in NO_2 VCD occurred at a rate of 7% per year from 2005 to 2011 over Central Eastern China, we found a decrease at a rate of 11% per year from 2011 to 2014. Over Japan, the NO_2 VCD increased at a rate of 4% per year from 2005 to 2011 and decreased from 2011 to 2014 at a rate of 4% per year. In this presentation, we also conduct detail trend analysis on a grid basis to discuss the potential causes for these variations in NO_2 VCD over China and Japan.

Keywords: Nitrogen dioxide, OMI, GOME-2, trend analysis

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CH4, H2O, N2O, and temperature from the mid-troposphere to the stratosphere in the northern mid- and high-latitudes

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Profiles of CH_4 have been retrieved from satellite-borne nadir sensors since 1996 by measuring thermal infrared (TIR) emissions from the Earth's atmosphere. GOSAT/TANSO-FTS has been operated since 2009. Profiles of CO_2 and CH_4 are retrieved from the TIR band of the TANSO-FTS. In this study, we assess data quality for CH_4, H_2O, N_2O, and temperature between the mid-troposphere and the stratosphere, contributing the improvement of our knowledge on CH_4 distributions, for instance, in the western Siberia. For comparisons with the TIR data, we used the solar occultation sensor, ACE-FTS, and the routine aircraft observations in the western Siberia. We found that (1) TIR CH_4 mixing ratios are systematically larger than ACE-FTS CH_4 in the 400-200 hPa levels between January and April in 2010/2011, especially in January, several profiles exceeded 2.0 ppmv of CH_4 at the 300 hPa level. (2) TIR H_2O mixing ratios are larger than ACE-FTS H_2O in the 300-250 hPa levels and the above throughout the period studied. (3) TIR CH_4 at the 7 km (430 hPa) and the 5.5 km (500 hPa) altitudes are in good agreement with those from the aircraft observations from the temporal variation view within the range of variations in TIR CH_4.

Acknowledgements: We thank Kaley A. Walker, University of Toronto, Canada for providing us the Version 3.5 ACE-FTS data through http://ace.uwaterloo.ca. This research was supported by the Environment Research and Technology Development Fund (ERTDF) of the Ministry of the Environment, Japan (A1202). Aircraft observations in the western Siberia are conducted by NIES with the cooperation of Russian Academy of Science (RAS).

Keywords: methane, troposphere, stratosphere, GOSAT, aircraft

AAS21-P22 Room:Convention Hall Time:May 27 18:15-19:30

Comparisons between NICAM-TM and GOSAT/TANSO-FTS TIR CO2 data

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Greenhouse gases Observing SATellite (GOSAT), which was the first satellite for global observations of greenhouse gases, was successfully launched on 23 January 2009. Our recent analysis suggested that CO_2 vertical profiles retrieved from Thermal and Near Infrared Sensor for Carbon Observation (TANSO) - Fourier Transform Spectrometer (FTS) thermal infrared (TIR) band had a negative bias in the middle troposphere. In this study, we globally evaluated the magnitude of the bias through the comparisons between the TIR CO_2 data and Nonhydrostatic Icosahedral Atmospheric Model - based Transport Model (NICAM-TM) CO_2 data [Niwa et al., 2011]. Furthermore, we calculated a correction factor to modify the bias for each latitude band and applied the latitude-dependent correction factors to the TIR CO_2 data on 500 hPa; here, we estimated the correction factors on the basis of comparisons between CO_2 profiles observed over airports by Continuous CO_2 Measuring Equipment (CME) in Comprehensive Observation Network for Trace gases by Airliner (CONTRAIL) project [Machida et al., 2008] and the coincident TIR CO_2 profiles. Then, we analyzed seasonal variations of the modified TIR CO_2 data to check the validity of the correction factors estimated here.

Comparisons of the differences of CO_2 concentrations on 500 hPa and 200 hPa (500 hPa minus 200 hPa) between TIR CO_2 data and NICAM-TM CO_2 data showed that the differences of the TIR CO_2 data were larger than those of the NICAM-TM CO_2 data because of the negative bias of the mid-tropospheric TIR data. The CO_2 differences between the two pressure levels of the TIR data were particularly large (˜8 ppmv) in low latitudes; this characteristic was not seen both in the NICAM-TM CO_2 data and the a priori CO_2 data (NIES-TM05). Next, we applied the latitude-dependent correction factors to the TIR CO_2 data on 500 hPa, and then compared the CO_2 differences on 500 hPa and 200 hPa. In low latitudes (25 ^{o} S-25 ^{o} N), the CO_2 differences between the two pressure levels of the TIR data became closer to the CO_2 differences of the NICAM-TM CO_2 data and the a priori CO_2 data when we applied the correction factor estimated over Bangkok. On the other hand, in northern high latitudes (northern latitudes of ˜40 ^{o} N), most of the CO_2 differences between the two pressure levels of the TIR data were positive unlike the NICAM-TM CO_2 and the a priori CO_2 data when we applied the correction factor estimated over Amsterdam. In boreal summer, surface CO_2 concentrations are lower than middle and upper tropospheric CO_2 concentrations; in that sense, the correction factor applied here was not appropriate in northern high latitudes in summer. These results suggest that the magnitude of the negative bias seen in TIR CO_2 data would vary depending on seasons as well as regions, and therefore, we should estimate a latitude-dependent correction factor for each season.

Furthermore, we compared time series of TIR CO_2 data with those of NICAM-TM CO_2 data and the a priori CO_2 data for several different regions that were categorized in terms of climatic divisions and latitude bands [Niwa et al., 2011]. Our preliminary results showed that the seasonal variations of the TIR CO_2 data in some regions were closer to those of the NICAM CO_2 data than of the a priori CO_2 data. For future work, we should review how to compare the three CO_2 data sets, and then closely analyze the differences in seasonal variations among the three data sets globally.

Acknowledgements

We thank the staff and engineers of Japan Airlines, the JAL Foundation, and JAMCO Tokyo for supporting the CONTRAIL project.

Keywords: CO2, satellite remote sensing, GOSAT

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Validation of GOSAT/TANSO-FTS TIR CO2 profiles using aircraft CO2 data

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Greenhouse gases Observing SATellite (GOSAT) was launched on 23 January 2009 to observe major greenhouse gases such as CO_2 and CH4. Thermal and Near-infrared Sensor for Carbon Observation Fourier Transform Spectrometer (TANSO-FTS) on board the GOSAT can observe CO_2 profiles in the thermal infrared (TIR) region, but the quality of the retrieved CO_2 profile data has not yet been fully validated. In this study, we compared GOSAT/TANSO-FTS TIR CO_2 profiles with aircraft CO_2 data to evaluate their quality. The aircraft data we used were obtained by Comprehensive Observation Network for Trace gases by Airline (CONTRAIL) project and Civil Aircraft for the Regular investigation of the atmosphere Based on an Instrument Container (CARIBIC) project, both of which are commercial airliner projects.

First, we assumed CONTRAIL data obtained during ascending and descending flights over airports as a "CO_2 profile", and then compared TIR CO_2 profiles with the CONTRAIL CO_2 profiles to which the TIR averaging kernel functions were applied. We adopted a distance between the GOSAT observation and the airport within 300 km and a time difference between the two observations within 72 hour as criteria for the comparison. Here, we used the CONTRAIL profile data obtained over the ten airports: Moscow, Amsterdam, Vancouver, Narita, Delhi, Honolulu, Bangkok, Singapore, and Djakarta. We also used CONTRAIL and CARIBIC level flight data to validate the global distributions of TIR upper tropospheric CO_2 data. We divided the level flight aircraft data into several regions, and then compared the averaged aircraft data with the averaged TIR data in each region.

From the CO_2 profile comparisons at each airport, we found the TIR data had a low bias of 1-1.5%. The magnitude of the bias varied depending on seasons and latitudes; in spring and summer in low latitude, the magnitude of the bias was larger than that in autumn and winter in mid and high latitudes. From the upper tropospheric CO_2 comparisons, the TIR data showed better agreements to the aircraft data than the a priori data, and the distribution of the TIR upper tropospheric CO_2 data had a similar pattern to the distribution of the aircraft data. In the poster, we will also report the details of the comparisons using other aircraft data.

Acknowledgements
We thank the staff and engineers of Japan Airlines, the JAL Foundation, and JAMCO Tokyo for supporting the CONTRAIL project. CARIBIC data used in this study were provided by the CARIBIC project. We also thank Dr. C. Brenninkmeijer and other members of the CARIBIC project.

Keywords: GOSAT, validation, CO2, aircraft measurement

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Climatology of spatiotemporal variations of tropospheric CO2 observed by CONTRAIL-CME

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CONTRAIL is the ongoing project that measures atmospheric trace gases during intercontinental flights of Japan Airlines. Atmospheric CO_2 concentration is analyzed using Continuous CO_2 Measuring Equipment (CME) onboard the aircraft. From ˜20 thousands of measurement flights since 2005, extensive number of CO_2 data (˜2 millions) along level-flight and ascent/descent tracks have been obtained, enabling us to well characterize spatiotemporal distributions of atmospheric CO_2 covering large part of the globe especially the Asia-Pacific regions. In this study, we define $\triangle CO_2$ as a deviation from the long-term trend observed at a northern hemispheric baseline station Mauna Loa, Hawaii, to illustrate climatological CO_2 distributions including seasonal and shorter-term variations. For instance, over airports in Japan, $\triangle CO_2$ reaches seasonal maximum at the end of April with higher values near the surface. In this season, high $\triangle CO_2$ spreads east of the Asian continent in the upper troposphere over the northern Pacific. In contrast, seasonal minimum of $\triangle CO_2$ occurs in September with more depletion in the upper troposphere. The summertime low $\triangle CO_2$ in the upper troposphere appears to be more pronounced over the Asian continent than over the Pacific. Likewise, we present seasonal variations of vertical profiles of tropospheric $\triangle CO_2$ over various airports and of spatial distributions in the upper troposphere in large-scale perspective, and discuss them from viewpoints of seasonally varying continental sources/sinks and atmospheric transport.

Keywords: CONTRAIL, CO2, troposphere, seasonal variation, vertical profile