Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-01 Room:102 Time:May 23 08:30-08:45

Carbon budget estimation by inverse modeling with atmospheric CO2 concentrations from surface and CONTRAIL measurements

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A great deal of understanding of the global and regional carbon budget helps us to perform a reliable prediction of future climate with an earth system model. However, the accuracy of CO2 source/sink estimation by inverse modeling, which is one of the leading methods to estimate regional carbon budget, is not very high because of sparse observational data coverage. The recent evolving aircraft measurements of CO2 in a three-dimensional view are expected to provide new constraints on the estimation of surface CO2 fluxes.

In this study, regionally divided carbon budgets are estimated by inverse modeling using surface measurement networks and aircraft measurements from Comprehensive Observation Network for Trace gases by Airliner (CONTRAIL). The CONTRAIL project has started since late 2005 and a huge amount of atmospheric CO2 data has been obtained covering altitudes between the Earth surface to the upper-troposphere and lower-stratosphere, latitudes between the boreal high-latitudes to the austral mid-latitudes. Monthly mean observational data from GLOBALVIEW-CO2, which mostly consists of surface measurements, and CONTRAIL are used in this inverse analysis. The CONTRAIL data measured both vertically over each airport and horizontally at the cruising altitude are used. The inversion method is based on the Bayesian statistics and the approach of the TransCom 3 is used. To relate atmospheric concentrations to surface fluxes, a three-dimensional transport model is employed. In this study, Nonhydrostatic ICosahedral Atmosphere Model (NICAM)-based transport model (NICAM-TM) is used with prescribed CO2 flux data of fossil fuel emission, respiration/photosynthesis in terrestrial biosphere and atmosphere-ocean exchange. The transport simulations of atmospheric CO2 are performed by low-resolution version of NICAM; the horizontal grid interval is about 240 km. The analyzed period is five years during 2005-2009 and meteorological fields in each year are used to drive the transport model with the nudging method. The inversion setup is similar to TransCom3, but number of flux regions to be estimated is 42. Land regions are divided into 31 according to vegetation types and the same 11 ocean regions as TransCom 3 are used.

A preliminary result of forward simulation with the prescribed fluxes shows that the transport model has good performance for reproducing general features of three-dimensional structure of CO2 observed by CONTRAIL. However, some discrepancies between the simulation and CONTRAIL are found in horizontal gradient even in the upper-troposphere during summer, indicating that aircraft measurements have significant impacts on flux estimates when vertical transport is efficient. The inversion results will be discussed in the presentation.

Keywords: carbon cycle, inversion

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CO2 seasonal distributions in the UT/LS region as observed by CONTRAIL and four transport models

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In the Comprehensive Observation Network for Trace gases by AIrLiner (CONTRAIL) project, high-frequency and wide-ranging CO2 data in the upper troposphere(UT)/lower stratosphere(LS) region have been obtained by Continuous CO2 Measuring Equipment (CME) onboard commercial aircraft operated by Japan Airlines (JAL).

The observed distributions in UT/LS region showed that CO2 isopleths followed the tropopause during the winter and spring. On the other hand, distributions tracked potential temperature surfaces crossing the tropopause in summer, suggesting fast meridional transport of high CO2 from the tropical troposphere. However, it is difficult to show how the tropospheric air masses intrude into the lower stratosphere across the tropopause due to the limited observational data.

Recently, we conducted CONTRAIL transport model intercomparison to improve our knowledge of three-dimensional structures of atmospheric CO2. The distributions in UT/LS region simulated by four global chemical transport models (ACTM, MJ98-CDTM, NICAM-TM, NIES) with common CO2 flux dataset were used to study the transport processes near the tropopause. It was shown that the models reproduced observed CO2 distributions following the tropopause in winter-spring season, although the CO2 gradients across the tropopause are underestimated by approximately 2 ppm between 300 and 350 K in potential temperature surfaces. In summer, the isentropic transport of high CO2 from the upper-troposphere in lower latitudes to the lower-stratosphere in higher latitudes were well simulated by the models. The detailed processes of transport will be analyzed and shown in the meeting.

Keywords: CO2, transport model, UT/LS exchange

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AAS021-03 Room:102 Time:May 23 09:00-09:15

JMA aircraft observation for greenhouse gases using a cargo aircraft C-130H to Minami-torishima

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Japan Meteorological Agency (JMA) started a long-term aircraft observation of greenhouse gases as one of the operational atmospheric monitoring activities in 2011. In cooperation with the Ministry of Defense, a cargo aircraft C-130H is used for the flask sampling observation during a roundtrip flight to Minamitorishima (MNM) once a month. The air samples are collected during a cruising flight at about 6 km as well as a descending to MNM. After the flight, we measure 4 trace gas concentrations of carbon dioxide (CO2), methane (CH4), carbon monoxide (CO), and nitrous oxide (N2O). Before the regular observations, preliminary observation flights using the C-130H were made in 2010 to evaluate a new flask sampling equipment and a high-precision measuring system with the collaboration of Meteorological Research Institute (MRI). In this presentation, the details of newly developed instrumentations for JMA aircraft observation and their performances are introduced.

Air samples are pressurized into the flasks by a manual diaphragm pump to an absolute pressure of about 0.4MPa. To minimize the drifts of trace gas concentrations in the flasks, we specially prepared a 1.7-L titanium flask of which internal surface is coated by silica. The storage tests for the flask samples during several days were repeated to ensure the stability of trace gases until analyses. To avoid the contamination of cabin air, sample air was taken from an air-conditioning blowing nozzle upstream of the recirculation fan. Specially coordinated flights at a low altitude of 1000ft over MNM were made using the C-130H to compare with the ground-based measurements from the MNM monitoring system operated by JMA. From these comparison experiments, it was confirmed that our aircraft sampling procedure was suitable for the precise measurements of trace gases.

JMA/MRI developed a new automated measuring system consisting of a conventional NDIR analyzer (Licor, LI-7000) for CO2 as well as recently advanced spectroscopy instruments of WS-CRDS analyzer (Picarro, G2301) for CH4, VURF analyzer (Aero-Laser, AL5002-AIR) for CO, and off-axis ICOS analyzer (Los Gatos, N2O/CO Analyzer) for N2O. A lot of test runs using standard gases and natural air indicated that higher-precision analyses could be easily achieved rather than before, in stead of the complicated GC systems, although relatively larger amount of sample air is required.

Keywords: aircraft observation, greenhouse gas

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AAS021-04 Room:102 Time:May 23 09:15-09:30

Evaluation of CO2 emission from the Tokyo metropolitan area based on balloon born measurements: Simultaneous observation

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Emissions of CO2 have increased drastically over the past century as a result of the mass consumption of fossil fuels due to the expansion of industrial activities resulting in dramatic increases in atmospheric concentrations of CO2. CO2 emissions from urban area are an important term of global carbon budget, but its estimation is mainly based on inventories of fossil fuel consumption and road traffic. To ascertain the CO2 flux values from urban area, detailed CO2 measurements including vertical distribution are required.

In this study, simultaneous measurements of CO2 vertical profiles using originally developed balloon borne instruments (CO2-sonde) at three sites on January 7, 2011 to evaluate CO2 emission from the Tokyo metropolitan area. The three sites, Isezaki (Gunma Pref.), Ichihara (Chiba Pref.), and Shirako (Chiba Pref.), where the balloon borne instruments were launched, are located upwind, inside, and downwind of the metropolitan area, respectively. The CO2 sensors are based on a non-dispersed infrared absorption spectroscopy technique at the wavelength around 4.3 micrometers. The data of the optical infrared absorption are transmitted through a GPS rawin-sonde (Meisei RS-06G) with temperature, humidity and GPS data every second.

As a result, at lower altitude ($<$1 km), CO2 mixing ratio obtained at Ichihara are higher than those obtained at other sites by 2-7 ppmv, while the three vertical profiles are indistinguishable at free troposphere. These observational data will be used to evaluate CO2 emission from the Tokyo metropolitan area using CO2 transport models and also to validate CO2 total column measurements by the greenhouse gas observing satellite (GOSAT) and a ground base fiber-etalon spectrometer.

Keywords: Carbon dioxide emission, Balloon-borne measurement, Tokyo metropolitan area, Satellite validation

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Real-time measurements of CO2 stable carbon isotope ratio in the atmosphere using wavelength modulation spectroscopy

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1. Introduction

Measurements of the stable isotopes provide important information on the source and history of their compounds in many research areas. Since the isotope ratio changes in relation to the conditions, the real-time measurements are required. It is recognized that the standard measurement technique for the stable isotope ratio is isotope ratio mass spectroscopy (IRMS). The IRMS has high precision in the range from 0.01 to 0.1 per mill. Nevertheless, pre-treatment is needed when a gaseous sample is injected in the mass spectrometer. Therefore, it is difficult to perform the real-time measurements of the isotope ratio. Recently, the laser absorption spectroscopy has been applied to the measurements of the stable isotopes. The isotopomers are easily recognized without interference of other species, when the absorption line is selected appropriately. Using this technique, since the sample gas is just introduced into the sample gas cell without any pre-treatment, the real-time measurements are able to be performed. Several studies using a quantum cascade laser with the direct absorption technique and a DFB laser with wavelength modulation spectroscopy or cavity ring-down spectroscopy have been performed. However, the precision is insufficient. Therefore, in this paper, higher precision measurements of the CO_2 stable carbon isotopes have been performed using WMS with a 2008 nm DFB laser.

2. Experimental

A 2008-nm single-mode DFB laser diode with a typical output power of 10 mW was used as a light source. The beam was introduced into a wedged-window Herriott-type multi-pass cell. The cell had a path length of 29.91 m and a volume of 0.9 L. The transmitted laser beam was focused using an AR-coated CaF_2 lens on an InGaAs photodiode detector. The wavelength of the DFB laser was modulated sinusoidally at 11 kHz using the output of a digital lock-in amplifier. The laser was scanned at 0.77 Hz by changing the injection current, which was controlled using a function generator that supplied a triangle voltage wave. The detected line pairs were $^{12}CO_2$ at 4978.205 cm^{-1} and $^{13}CO_2$ at 4978.023 cm^{-1}. The stable isotope ratio was determined with comparing the measurements $^{13}C/^{12}C$ ratio to the international PDB-standard $^{13}C/^{12}C$. All experiments were performed at 313 K. The pressure in the cell is kept 10 kPa.

3. Results and Discussion

The second-harmonic WMS spectra of the sample CO_2 gas, 379 ppm and -30.5 per mill, are detected. The S/N (Signal to noise ratio) of $^{13}CO_2$ is approximately 66, even though the concentration of $^{13}CO_2$ is 100 times lower than that of $^{12}CO_2$. It was found that this line pair is favorable because of their clearly separated signals and comparable intensities. The continuous stable carbon isotope ratio measurements were performed using this apparatus to investigate long-term stability. The signal was measured during 10 hours with 5 minutes increment. The pressure and temperature were found to be controlled within 0.01 kPa and 0.1 K, respectively, during continuous measurements. In this long-term measurement, the precision ?was achieved to be 0.1 per mill. Then, the continuous stable carbon isotope ratio measurements in ambient air was performed. The CO_2 concentration was also measured with NDIR. The CO_2 concentration profile obtained by our constructed apparatus was good agreement with that obtained with NDIR. The stable carbon isotope ratio of CO_2 was also measured successfully using this system. It was found that its value decreased with increasing CO_2 concentration.

4. Conclusions

The stable carbon isotope ratios of CO_2 were measured using WMS with a 2008 nm DFB laser diode. The precision was achieved to be 0.1 per mill for long-term detection (10 h). The stable carbon isotope ratios of CO_2 was successfully measured using our constructed system.

Keywords: Measurements of carbon isotope ratio, Carbon dioxide, Wavelength modulation spectroscopy, Multi-pass cell, DFB laser

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AAS021-06 Room:102 Time:May 23 09:45-10:00

The vertical profiles of CH_4 observed at Tsukuba with a Fourier transform spectrometer

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Fourier transform spectrometer (FTS) has advantages in its high resolution and the wide wavenumber range. Vertical profiles of some species can be derived from the high-resolution spectra. The vertical profiles and column densities of CH_4 were retrieved from the solar spectra observed at Tsukuba, Japan with SFIT2 spectral fitting program developed by Rinsland et al. (1998). It needs to select an appropriate wavenumber region and the optimization of fitting parameters are also needed. Now we are investigating these parameters in the NDACC/IRWG group and we will reanalyze the vertical profiles and column densities of CH_4. We found that the phase of seasonal variation of the mixing ratios in the lower stratosphere is shifted from those in the troposphere and the temporal variation of total column shows steplike increase in 2007 from preliminary analysis.

Keywords: FTIR, Trace Species, Methane

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Long-term trend of methane concentration in the lower stratosphere over Japan

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Stratospheric methane acts as an important source of stratospheric water vapor and also a sink for chlorine radical. For predicting the future of the stratospheric chemistry and dynamics, as well as for evaluating strategies for limiting or reducing future emission of methane into the atmosphere, it is indispensable to figure out the past methane trend in both of stratosphere and troposphere accurately. Systematic collections of stratospheric air samples have been carried out over Japan since 1985, using a balloon-borne cryogenic sampler. The air samples collected were analyzed for the CH4 and N2O concentrations since 1988. Almost linear and compact relationship between CH4 and N2O concentrations was found for all observations. It is well known that tropospheric N2O has been secularly increasing quite monotonously in recent decades. On the other hand, increase rate of tropospheric CH4 has been varied complicatedly. Despite the fact that both concentrations have been increased independently in troposphere, there is no significant difference between the correlations of stratospheric CH4 and N2O concentrations in each year, at first glance. Considering this fact, the compact relationship obtained over Japan suggests that both CH4 and N2O are destroyed at similar rate in the lower stratosphere during the poleward transport of stratospheric air, although the chemical destruction processes of the two gases are quite different. Therefore, we employed N2O-depressions, instead of N2O concentrations, for examining correlations with CH4 concentrations. This method cancels the effects of secular N2O increase in stratosphere and enables us to detect possible change in stratospheric CH4. Thus we found that stratospheric CH4 shows a significant increase before 2000 and clear stagnation after 2000. We categorized CH4 concentration data into different N2O depression rages, and calculated increase rates by applying the curve fitting procedure, taking into account the age of stratospheric air. Average increase rates were calculated to be about 0.4 and 0.1%/year before and after 2000, respectively, in the lower stratosphere. This rate is comparable with the results of 207 ppbv increase in the period of 1978 to 2003 in the lower stratosphere reported by Rohs et al.(2006). However, increase rate before 2000 obtained in this study is much smaller than the result of 1.95 and 0.87 %/year in 1985 and 1994, respectively, by ATMOS/ACE-FTS measurements (Rinsland et al., 2009).

Keywords: methane, stratosphere, trend

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Latitudinal distribution of APO seasonal cycles and its relation to the meridional circulation in the lower troposphere

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We examine the latitudinal differences in the phasing of the average seasonal cycles of the atmospheric potential oxygen (APO= O{sub}2{/sub}+1.1xCO{sub}2{/sub}), which is based on the atmospheric O{sub}2{/sub} and CO{sub}2{/sub} observations in the western Pacific region. Because APO is invariable with respect to the terrestrial biotic exchanges, its seasonal variations mainly reflect air-sea exchanges of O{sub}2{/sub}. Investigating on the APO gives new insights into the meridional circulation because APO is a tracer from the ocean, which has very different flux distribution from those of land tracers. The seasonal minimum occurs in March and September in the Northern and Southern Hemispheres, respectively, and the latitudinal distribution of the date of the seasonal minimum shows discontinuous change at the equator. Contrary to this, the date of the seasonal maximum smoothly changes across the equator from March at 35 deg. S to July at 15 deg. N and levels off between 15 deg. N and 50 deg. N. The seasonal variation in APO is predominantly driven by the air-sea O{sub}2{/sub} fluxes between 30-60 deg. in both hemispheres because seasonal variation in the air-sea O{sub}2{/sub} fluxes in the equatorial regions are relatively small. Therefore, the seasonal cycles of APO in the tropics depend mainly on the meridional propagation of the atmospheric signals in the lower troposphere. The observational results indicate that the seasonal minima propagate equatorward with little phase lags in both hemispheres and the seasonal maximum propagate equatorward with a substantial phase lag in the Southern Hemisphere. These seasonal differences in the propagation speed could be explained by the strong and weak meridional circulation in winter and summer, respectively, in the lower troposphere. The latitudinal distribution of the date of the seasonal maximum in the Northern Hemisphere may be attributed to the influence of the propagation of the seasonal maximum in the Southern Hemisphere because the seasonal variations in the air-sea O{sub}2{/sub} fluxes is about 2 times larger in the Southern Hemisphere than in the Northern Hemisphere.

Keywords: APO, seasonal cycle, meridional circulation, atmospheric oxygen, air-sea gas exchange

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14C measurements in suburban aerosols from northern Japan: An enhanced production of biogenic organic aerosols in spring

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Here, we report the year-round observation of fossil and modern carbon in total carbon (TC) and water-soluble organic carbon (WSOC) in atmospheric aerosols from Sapporo, northern Japan, based on radiocarbon measurements. We found that modern carbons are more important in both TC and WSOC, except for wintertime, with elevated levels in spring. Interestingly, WSOC showed higher percent modern carbon (pMC) throughout the year, suggesting that WSOC is produced by photochemical oxidation of biogenic volatile organic compounds especially during spring to summer.

Keywords: aerosols, radiocarbon, total carbon, water-solulbe organic carbon

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AAS021-10 Room:102 Time:May 23 11:00-11:15

Significant contribution of isoprene oxidation products in summertime organic aerosols at the summit of Mt. Fuji, Japan

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We investigated the organic molecular compositions and size distributions of the summertime aerosols collected at the summit of Mt. Fuji (3776 m, a.s.l.) using gas chromatography/mass spectrometry. More than 120 organic species were detected in the aerosols and grouped into different compound classes such as aliphatic lipids, sugars, phthalate esters, sterols, hydroxy-/polyacids, and biogenic secondary organic aerosol (SOA) tracers for the photooxidation of isoprene (e.g., 2-methyltetrols), alpha/beta-pinene (e.g., pinic acid), and beta-caryophyllene (beta-caryophyllinic acid). Total concentrations of the identified organics were 76.1-325 ng m-3 (average 183 ng m-3) for the whole-day samples, which are more than 10 times higher than those in nighttime samples (9.28-19.2 ng m-3, average 15.6 ng m-3). Higher concentrations of both primary and secondary organic marker compounds were observed in the whole-day samples, indicating that the mountain venting at Mt. Fuji should act as an efficient pump that uplifts the ground-surface aerosols and their precursors to the free troposphere. Interestingly, isoprene SOA tracers (2.87 ng m-3 in nighttime and 69.2 ng m-3 in whole day) were found to be the most abundant compound class. 2-Methylerythritol and 2-methylthreitol, the well-known isoprene SOA tracers, were detected as the dominant single compounds. Using a tracer-based method, we estimated the concentrations of secondary organic carbon (SOC) derived from isoprene, alpha/beta-pinene, and beta-caryophyllene to be 2.16-51.2 ngC m-3 (15.5 ngC m-3) during the nighttime and 183-954 ngC m-3 (465 ngC m-3) during the whole day. These values correspond to 0.80-31.8% (12.5%) and 21.6-48.9% (31.9%) of the organic carbon (OC) concentrations in nighttime and the whole-day samples, in which isoprene-derived SOC accounts for 80% and 72% of total SOC, respectively. This indicates that a large amount of organic aerosols in the free troposphere should be derived from the oxidation of isoprene emitted from the forest areas on the foothills. Size distributions of the identified organics were unimodal in most cases. Biogenic SOA tracers (e.g., 2-methyltetrols and 3-hydroxyglutaric acid), levoglucosan and malic acid were detected in the fine mode, while sucrose and trehalose that are abundant in airborne pollen and dust aerosols peaked in the coarse mode. This study provides useful information to understand the sources and abundances of organic aerosols over high mountains in East Asia.

Keywords: organic aerosols, Mt. Fuji, levoglucosan, 2-methyltetrols, biogenic VOCs, isoprene

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A field experiment to determine the CCN activity of primary marine aerosols generated from natural seawater

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Oceanic surface waters contain a large amount of organic substances produced by marine biota, which are transferred to the atmosphere as primary marine aerosols (PMA) by bubble bursting processes. The organics potentially play an important role in regulating the physico-chemical properties of the PMA, including the cloud condensation nuclei (CCN) activity. Our current knowledge on the relationship of the organics in PMA and the particle properties is, however, still limited despite recent laboratory/field studies on PMA. To better characterize the CCN activity of PMA associated with the organics, we preformed an experiment of particle production by the bubble bursting of natural seawater.

The experiment was conducted in Maizuru Bay, the Japan Sea, onboard R/V Ryokuyo-Maru, Maizuru Fishery Research Station, Kyoto University. The PMA generator, which floats on the sea, was prepared to produce aerosols by the bursting of air bubbles in natural seawater. The generator is equipped with a bubble-producing glass ball filter, through which compressed dry air was passed, at 30 to 40 cm below the air-sea interface. The bubbles rose to the sea surface and burst inside a 30 L PTFE dome. The generated PMA were transferred to the instruments onboard through PTFE tubing. Dried PMA was introduced to a differential mobility analyzer (DMA) for size selection, and the resulting monodisperse aerosol was transferred to a condensation particle counter and a continuous flow thermal gradient CCN counter to measure the number concentrations of condensation nuclei (CN) and CCN, respectively. The activation diameters (D_{act}) of the PMA at supersaturations ranging from 0.1% to 0.5% were calculated from the CCN to CN ratios. Surface seawater samples were also collected, which were used for the determination of chlorophyll-a (chl-a) concentrations.

The chl-a concentrations inside Maizuru Bay (IMB) were much higher than that outside Maizuru Bay (OMB). The D_{act} of the generated PMA at IMB and OMB were clearly different in particular at higher supersaturations (0.3% and 0.5%); D_{act} of PMA at IMB were larger than that of PMA at OMB. The hygroscopicity parameter kappa (Peters and Kreidenweis, 2007) calculated for the PMA at the highest supersaturation were lower than that of sodium chloride, suggesting that the PMA were the mixture of less hydroscopic organics and sea salts. The results suggest that organics in seawater transferred to the atmosphere as PMA are enriched especially in the ultrafine mode and affect the particle CCN activity.

Reference: Petters, M. D., and S. M. Kreidenweis (2007), A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7(8), 1961-1971.

Keywords: primary marine aerosols, cloud condensation nuclei, organics, natural seawater

Japan Geoscience Union Meeting 2011
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Intensive field observations of trace gases/aerosols in Rudong, China in spring 2010: Objectives and initial results

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Although Central East China is regarded as an emerging region with high emission rates of precursors of ozone and aerosols, available observational data are still limited. We conducted an intensive field campaign observing ozone and its precursors and chemical components/physical and optical parameters of aerosol particles at Rudong (32.26N, 121.37E), Jiangsu, China in May/June 2010 under international collaboration. The location is near the west side of Yellow sea and is away from Shanghai by 100 km and from Rudong city center by 15 km. In this presentation, objectives of the field measurement and initial results are summarized. The main objectives are to characterize typical emission ratios of primary pollutants for this region (including those from biomass burning), to examine the mass closure and point optical closure of aerosols and to obtain useful information to evaluate and improve the capability of the tropospheric chemical transport model to simulate PM2.5 and AOD, to study ozone production rates/controlling factors including the effect of aerosols on ozone photochemistry, and to achieve instrument intercomparisons to improve our observational capabilities. Three distinct periods are clearly found: relatively polluted period (possibly under influence from urban, May 15-28), clean period with easterly wind (May 29? June 9), and post-harvest biomass burning period (June 10- 24). The BC/CO ratio during the biomass burning period was clearly higher than other periods, in accordance with the emission inventory. The features of the BC/CO and other ratios potentially characteristic to emission sources are compared to past observations in China. Within the 1.5 month observation period, we found several cases where the air mass traveled from Rudong to Fukue Island (32.75N, 128.68E), located at the opposite east side of the Yellow Sea, allowing the direct comparison of the observational features at the two sites. At Rudong, photolysis frequencies (J values) important to ozone photochemistry were found to be attenuated significantly by the presence of dense aerosols. The effect of the aerosols on ozone photochemistry was analyzed. The relationship between the increase in the aerosol scattering coefficient with relative humidity and chemical composition of aerosols was analyzed and the results were compared to those for Fukue Island in the previous year. Comparison of black carbon concentrations measured by three different instruments resulted in a magnitude relationship similar to those obtained at different locations (e.g., Fukue Island and Mt. Tai), contributing to reduction of the observational uncertainty or to improvement of the consistency among the existing black carbon concentration data observed by different instruments in East Asia.

Keywords: China, ozone, aerosol, intensive field observation

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PM2.5 variation in Rudong, China and Fukue Island, Japan in spring 2010

Fumikazu Taketani[1*], Yugo Kanaya[1], Xiaole Pan[1], Hisahiro Takashima[1], Hitoshi Irie[1], Yuichi Komazaki[1], Shinji Saito[2], Satoshi Inomata[2], Hiroshi Tanimoto[2], Huabin Dong[3], Baozhu Ge[3], Jie Li[3], Xiquan Wang[3], Hongyan Chen[3], Meigen Zhang[3], Zifa Wang[3]

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We carried out an intensive field campaign observing ozone and its precursors and chemical components/physical and optical parameters of aerosol particles at Rudong (32.26N, 121.37E), Jiangsu, China in May/June 2010. In this presentation, to investigate the PM2.5 mass concentration, their composition and correlation in both Rudong and Fukue Island(32.75N, 128.68E), we observed PM2.5 mass concentration using SHARP monitor and sampled PM2.5 on the quarts filter by high volume air sampler. PM2.5 total mass concentration is monitored every 1 min for both sites. At the Rudong site(May16-June23), 9 or 14-hours PM2.5 samples were collected on the quartz filters using High-volume(500L/min) sampler, while 22-hours PM2.5 samples were collected at the Fukue Island site(May18-25). In the end of May, high PM2.5 mass concentrations were observed in both Rudong and Fukue Island. We are going to discuss mass closure and correlation of the PM2.5 for Rudong and Fukue Island results.

Keywords: PM2.5, aerosol, composition, transportation, mass closure

Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-14 Room:102 Time:May 23 12:00-12:15

Impact of new particle formation on the concentrations of aerosol number and cloud condensation nuclei around Beijing

Hitoshi Matsui[1*], Makoto Koike[2], Yutaka Kondo[1], Nobuyuki Takegawa[1]

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New particle formation (NPF) is one of the most important processes to determine the concentrations of aerosol number (condensation nucleus, CN) and cloud condensation nuclei (CCN) in the atmosphere. In this study, we developed NPF-explicit WRF-chem model with 20 aerosol size bins from 1 nm to 10 um and with activation-type nucleation parameterization, which was recently suggested. This model was applied to Beijing region for the periods during the CARE-Beijing 2006 campaign conducted in August and September 2006.

Model calculations reproduced the timing of NPF (21 days out of 26 days measurement available) and the rapid growth of nucleated particles (NP) up to several-tens nanometers reasonably. NPF was mostly observed and calculated in ”sweeping” periods when 0-3 days after the cold front passages with the inflow of clean air from the north, while there were few NPF events in ”stagnant” periods when Beijing region was influenced from high-pressure system with the accumulation of trace gases and aerosols. The difference of NPF frequency between sweeping and stagnant periods could be explained by the balance of growth rate and condensation and coagulation sink of NP. This result suggests that once the reasonable nucleation rates of cluster formation at 1 nm were given, model calculations can represent the timing of NPF (contrast of ”NPF” and ”no-NPF” days) and further growth up to several-tens nanometers through the theoretical calculations of condensation and coagulation processes.

The contribution of NPF to the CN concentrations larger than 10 nm (CN10) was estimated to be 20% in Beijing in period average. This contribution became maximum in the noontime (12-16 LT): 73% in NPF days and 17% in no-NPF days, respectively. The impact of NPF on CCN concentrations was dependent on supersaturations (S): CCN concentrations were increased (by 100-200% in maximum) at higher supersaturations ($S > 0.2\%$) but decreased (by 50% in maximum) at lower supersaturations ($S < 0.1\%$) by NPF. This is likely because NPF suppresses the increases in size and hygroscopicity of pre-existing particles through the competition of condensable gases between smaller secondary particles and larger pre-existing particles.

Sensitivity calculations were also conducted with the reduction of primary aerosol emissions (black carbon and primary organic aerosol). We will show the sensitivity of CN and CCN concentrations to primary aerosol emissions in the presentation.

Keywords: New particle formation, Aerosol number concentration, Cloud condensation nuclei, Regional three-dimensional model, Mega city

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(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-15 Room:102 Time:May 23 12:15-12:30

Comparison of aerosols among surface measurements, CALIOP data, and the SPRINTARS model(I): Sources of dust at Phimai

Haruo Tsuruta[1*], Eiji Oikawa[1], Syugo Watanabe[1], Toshiro Inoue[1], Daisuke Goto[1], Toshihiko Takemura[2], Nobuo Sugimoto[3], Koichiro Sera[4], Shigeto Sudo[5], Seiichiro Yonemura[5], Yuichiro Shirasuna[6], Koichiro Hirano[6], Tadahiro Hayasaka[7], Teruyuki Nakajima[1]

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For atmospheric aerosols measured at Phimai, Thailand, comparison of chemical and optical properties was performed between the surface measurements and the SPRINTARS model in the previous report at Japan Geoscience Union Meeting 2010. According to the analysis by the field study, dust particles were transported from East China and Indochina, in the early- and the late-dry season, respectively. In contrast, the dust concentration in wet season was usually low compared with that in the dry season, while high concentration of dust was measured, comparable to that in the dry season due to transport of the edge of dust storms in East Asia. The purpose of this study is to clarify if the high dust particles were caused by the local sources or by long range transport. The case study on the episode of high dust concentration during 17-20 June 2008 was made, by comparing the surface data with CALIOP data (http://www-calipso.larc.nasa.gov/data/BROWSE/production/V3-01/), NIES RIDAR data(http://www-lidar.nies.go.jp/Phimai/archives/), and the result of the SPRINTARS model(http://sprintars.riam.kyushu-u.ac.jp/archivej.html). The CALIOP data showed that high dusts were measured during 14-20 June 2008, from Saudi Arabia to Somalia, from Pakistan to Arabian Sea, from Bangladesh to the Bay of Bengal, and the maximum vertical height of the dust layer reached up to 7 km. According to the RIDAR data, high dust aerosols were observed up to the height of 2-3km during 15-17 June 2008 (missing data from 18 June). Furthermore, the backward trajectory analysis by NOAA HYSPRIT MODEL (http://www.arl.noaa.gov/ready/hysplit4.html) showed that the air masses arrived at Phimai on 17-20 June 2008, was transported in the lower troposphere from over the Bay of Bengal a few days after, and in the layer of 2-4km height from the east coast of north Africa one week after. On the other hand, the SPRINTARS model, a high dust layer existed up to the height of 2-3km at least for a week from 14 June 2008, spreading horizontally from the eastern part of North Africa to the Bay of Bengal through west Asia, India, and the edge of which reached Indochina. All these data strongly suggest that the high dust particles at Phimai in the wet season could be caused by the long range transport of dust generated in the desert areas of west Asia, in addition to the local dust.

Keywords: atmospheric aerosol, dust, CALIOP, LIDAR, long range transport, SPRINTARS model

Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-16 Room:102 Time:May 23 12:30-12:45

Simulating black carbon at Syowa station, Antarctica: long-range transport from various source regions

Kengo Sudo[1*], Hiroya Endo[1], Toshihiko Takemura[2]

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This study evaluates long-range transport of black carbon (BC) to the Antarctic region in detail with a chemistry and aerosol coupled global climate model CHASER-SPRINTARS. BC, also called soot, is aerosol species emitted from fossil fuel combustion and biomass burning. BC is one of the most important air pollutants and also causes significant climate impacts with absorbing sunlight and melting snow/glacier. In this study, data derived from the continuous observation at Syowa station (69.0^{o}S, 39.6^{o}E) are used for a representative mass concentration of BC in the Antarctic region. It is found that the current version of the model tends to underestimate BC concentration all year round especially in polar region. This suggests that BC long-range transport process may not be simulated properly in the model, probably due to the model uncertainties in surface emission process and wet deposition process associated with precipitation. To reduce the model underestimation of BC in the Antarctic region, we perform several sensitivity experiments for improving reproducibility of BC long-range transport in the model. In the experiments we reduce the activity as CCN (cloud condensation nucleus) for hydrophobic BC and/or increase the fraction of BC in external mixing for surface emissions (the emission ratio of hydrophobic BC to total BC) to larger. The sensitivity experiments reproduce the observed BC level at Syowa station, but fail to reproduce the observed seasonal cycle of BC with winter high and summer low. Previous studies have suggested that aging process which changes hydrophobic BC to hydrophilic by coating it with water soluble species like sulfate during transport may play an important role in seasonal cycle of BC in remote regions. In this study, we examine the impacts of such aging process on BC in the Antarctic region, newly introducing an aging scheme for BC in the model. As a result, the model with aging process successfully reproduces the seasonal cycle of BC as well as the concentration level at the Syowa station. These sensitivity experiments reveal that long-range transport and subsequent concentrations of BC in remote areas are largely controlled not only by atmospheric transport, but also by BC mixing state and wet-deposition with precipitation and aging effect. In this study, a tagged tracer experiment is also conducted to estimate source region and transport pathway for BC at the Syowa station, Antarctica. The experiment indicates that about 50% of annual mean BC at the Syowa station comes from South America with ˜20% from South Africa and 15-20% from Australia. The model reveals two patterns of transport pathway. One is the case that BC is transported to the Syowa station in the lower troposphere (below 3 km altitude). The other case is that BC is first lifted up to the tropopause regions over source region and is transported toward Antarctica via the upper troposphere and lower stratosphere and eventually reaches to the Syowa station associated with Katabatic winds.

Keywords: Black Carbon, Antarctic, Long-range Transport, Chemical Transport Model

Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-17 Room:102 Time:May 23 14:15-14:30

Process study of volatile organic compounds in surface seawater using PTR-MS

Hiroshi Tanimoto[1*], Sohiko Kameyama[2], Satoshi Inomata[1], Urumu Tsunogai[2], Toru Iwata[3], Kazuhiro Okuzawa[1], Mitsuo Uematsu[4]

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We developed an equilibrator inlet-proton transfer reaction-mass spectrometry (EI-PTR-MS) method for fast detection of dimethyl sulfide (DMS) and volatile organic compounds (VOCs) dissolved in seawater. Dissolved DMS and VOCs extracted by bubbling pure nitrogen through the sample were continuously directed to the PTR-MS. DMS and several other VOCs reached equilibrium with an overall response time of minutes. The detection limit for DMS was 50 pmol L?1 at 5-s integration. The EI-PTR-MS instrument was deployed during a research cruise in the western North Pacific Ocean. For DMS and VOCs, comparison of the EI-PTR-MS results with results obtained by means of membrane tube equilibrator-gas chromatography/mass spectrometry agreed reasonably well on average. EI-PTR-MS captured temporal variations of dissolved DMS and VOCs concentrations, including elevated peaks associated with patches of high biogenic activity. These results demonstrate that EI-PTR-MS was effective for highly time-resolved measurements of DMS and VOCs in the open ocean. Further measurements will improve our understanding of the biogeochemical mechanisms of the production, consumption, and distribution of DMS and VOCs in the ocean surface.

Keywords: volatile organic compounds, DMS, surface seawater, PTR-MS, SOLAS, biogeochemistry

Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-18 Room:102 Time:May 23 14:30-14:45

Eight-component retrievals from ground-based MAX-DOAS observations

Hitoshi Irie[1*], Hisahiro Takashima[1], Yugo Kanaya[1], Folkert Boersma[2], Lou Gast[3], Folkard Wittrock[4], Dominik Brunner[5], Yipin Zhou[5], Michel Van Roozendael[6]

[1]JAMSTEC, [2]KNMI, [3]RIVM, [4]University of Bremen, [5]Empa, [6]BIRA

We attempt for the first time to retrieve lower-tropospheric vertical profile information for 8 quantities from ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) observations. The components retrieved are the aerosol extinction coefficients at two wavelengths, 357 and 476 nm, and NO_2, HCHO, CHOCHO, H_2O, SO_2, and O_3 volume mixing ratios. A Japanese MAX-DOAS profile retrieval algorithm, version 1 (JM1), is applied to observations performed at Cabauw, the Netherlands (51.97N, 4.93E), in June-July 2009 during the Cabauw Intercomparison campaign of Nitrogen Dioxide measuring Instruments (CINDI). Of the retrieved profiles, we focus here on the lowest-layer data (mean values at altitudes 0-1 km), where the sensitivity is usually highest owing to the longest light path. In support of the capability of the multi-component retrievals, we find reasonable overall agreement with independent data sets, including a regional chemical transport model (CHIMERE) and in situ observations performed at the 3- and 200-m height levels of the tall tower in Cabauw. Plumes of enhanced HCHO and SO_2 were likely affected by biogenic and ship emissions, respectively, and an improvement in their emission strengths is suggested for better agreement between CHIMERE simulations and MAX-DOAS observations. Analysis of air mass factors indicates that the horizontal spatial representativeness of MAX-DOAS observations is about 3-15 km (depending mainly on aerosol extinction), comparable to or better than the spatial resolution of current UV-visible satellite observations and model calculations. These demonstrate that MAX-DOAS provides multi-component data useful for the evaluation of satellite observations and model calculations and can play an important role in bridging different data sets having different spatial resolutions.

Keywords: MAX-DOAS, retrieval, multi-component, aerosol, ozone

Japan Geoscience Union Meeting 2011
(May 22-27 2011 at Makuhari, Chiba, Japan)

AAS021-19 Room:102 Time:May 23 14:45-15:00

Regional O3 trend and its chemical linearity in recent anthropogenic emissions change over East Asia

Kazuyo Yamaji[1*], Hitoshi Irie[1], Itsushi Uno[2]

[1]JAMSTEC, [2]Kyushu University

Regional O3 response to perturbations of Chinese anthropogenic emissions is investigated using the brute force method by a regional chemical transport model. Both springtime and summertime ozone responses are unlikely to show nonlinearly in the emissions perturbation of 70%?200% over East Asia. Observed NO2 in the east central China in 2003?2008 is ranging within a factor of 0.82?1.35 of the 2004 level, and that explains recent O3 change stays within the linear O3 response range. As for observed O3 at Japanese remote sites, the O3 response is 11.7 ppbv with respect to a 100% increase at the east central China from NO2 level in 2004. The simulated relationship between O3 and NO2 shows 5.9 ppbv of O3 growth is caused by doubled NO2 from the 2004 level.

Keywords: tropospheric O3, chemical linearity, emissions, East Asia

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AAS021-20 Room:102 Time:May 23 15:00-15:15

CO emissions from biomass burning in Southeast Asia in the 2006 El Nino year: Shipboard and AIRS satellite observations

Hideki Nara[1*], Hiroshi Tanimoto[1], Yukihiro Nojiri[1], Hitoshi Mukai[1], Jiye Zeng[1], Yasunori Tohjima[1], Toshinobu Machida[1]

[1]NIES

[Introduction]

During the dry season (October-November) in the 2006 ENSO year, we observed substantial CO enhancement over the western tropical Pacific by shipboard observations routinely operated between Japan and Australia/New Zealand. In this presentation, we present evidence of CO pollution episodes over the tropical Pacific due to intensive biomass fires in Southeast Asia and north Australia during 2006 El Nino year. We discuss locations of CO emissions from fires followed by long-range transport by combination of in-situ ship measurements, Atmospheric Infrared Sounder (AIRS) satellite observations, and Lagrangian particle dispersion model (FLEXPART).

[Method]

Beginning in November 2005, continuous monitoring of atmospheric trace gases has been initiated by using a commercial cargo vessel, M/V Transfuture 5 (TF5) (owned by Toyofuji Shipping Co. Ltd.). The ship takes regular service among ports of Japan, Australia and New Zealand with the 6 weeks interval, covering subtropical latitudinal region over North and South Pacific. The shipping route of TF5 is shown in Figure. Automated instruments were installed into an observation room for in situ measurements of CO, CO_2 and O_3.

[Result & Discussion]

Abnormal enhancements in CO were observed between 15N and the Equator during the southbound voyage (Episode 1), and around the Equator (Episode 2) during the northbound voyage, which had large impact on the seasonal variations of CO in the tropical Pacific. During Episode 1, AIRS satellite images and Global Fire Emissions Database version 2.1 (GFEDv2.1) suggested that the CO plume originated from biomass burning in Borneo and Sumatra followed by long-range transport to the tropical Pacific region. The amplitude of observed CO enhancement during Episode 2 was much smaller than that during Episode 1. Simulations by FLEXPART showed well consistent results with our analysis, which provides a reasonable interpretation of the data, complimenting in situ and satellite observations.

Correlations of CO to CO_2 and of O_3 to CO observed during two episodes were also examined. Scatter plots of CO versus CO_2 during Episode 1 showed significant correlation (R^2 = 0.60) with a steep dCO/dCO_2 slope (171 +/- 31 ppbv/ppmv at the 95% confidence interval). The dCO/dCO_2 ratio observed was higher than previously reported for savanna and grassland (63 +/- 20 ppbv/ppmv), tropical forest (103 +/- 21 ppbv/ppmv), and slightly higher that Indonesian peatland fires 142.7 ppbv/ppmv, indicating that Indonesian fires including peat soil burning are a dominant factor during Episode 1. Comparison between the dCO/dCO_2 ratio and CO/CO_2 emission ratios from GFEDv2.1 suggests the uncertainty in CO emissions of GFEDv2.1 in Southeast Asia region associated with peatland fires. Significant O_3-versus-CO correlation was observed only for Episode 1 (R^2 = 0.68). The dO_3/dCO ratio (0.05 +/- 0.01 ppbv/ppbv) was considerably smaller than values reported in previous observations in this region, which suggests that net O_3 production was not efficient in the burning plumes transported in the lower troposphere over the western tropical north Pacific. The reason for low dO_3/dCO ratio is not clear, but is likely associated with combustion properties of the peat and/or meteorological condition during the transport, both of which are specific to Southeast Asia region. Further research is needed to understand the low dO_3/dCO ratio in the long-range transport of the burning plumes over western tropical north Pacific in the lower troposphere.

Keywords: Carbon monoxide, Voluntary observing ship, AIRS, Southeast Asia, Biomass burning

Japan Geoscience Union Meeting 2011
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AAS021-21 Room:102 Time:May 23 15:15-15:30

Development of the long-term mass transport model and verification by Chernobyl accident and Sakurajima volcano

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In those days, environmental problems are taken up as the societal issues. Air pollution is one of them. The air pollution cases have been confirmed all over the world. There are some cases that require the long-term prediction and behavior grasp. For example, the radionuclide pollution in Chernobyl and the volcanic gas exposure in the surrounding area. To predict the air pollution and grasp the condition of the air, many models have been studied. Gaussian plume model is the most used among them. However, most models including the model don't assume the long-term prediction.

In this study, we propose the model suited for the long-term prediction, which have not studied enough, and verify the applicability of our model by fitting with measured data.The long-term model in this study is constructed by the governing equation based on the advection equation, and the approximate analytical solution is derived from the mathematical method. The governing equation is the following equation.

$dC/dt+\{(v_x)d/dx+(v_y)d/dy\}C+l_{env}C+l_{decay}C=P(x,y,t)$

where C is the concentration in the air, t is the days since the standard time, and v is each forward advection velocity by wind. l_{env} is a local environmental removal rate, that is, the sum of reaction rate by local environmental kinetics such as chemical reactions of the nuclide with soil, permeation into soil, vegetation uptake, water run off, etc. l_{decay} is the physical decay.

$P(x, y, t)$ is the term expressing the emission of pollutant, and the analytical solution differs according to the definition of $P(x, y, t)$.

In this study, the pollution types were divided into four types. As the emission form, the instantaneous emission and the continuous emission were assumed. As the emission source type, the point source and the plane source were assumed.

Fitting with measured data, P was defined according to the cases and the analytical solution was derived in each cases. Radionuclide diffusion derived from Chernobyl Nuclear Power Plant accident was assumed as the instantaneous emission at the point source. In addition, SO_2 diffusion derived from Sakurajima Volcano was assumed as the continuous emission at the point source. And derivation of the model(the analytical solution) was assumed about the both cases.

In case of the instantaneous emission at the point source, P was defined as the following equation.
$P=d(x)d(y)d(t).$
and the model was derived. Here, d(x) is delta function. As the result, the model was derived as
$C(t)=A\ exp(-l_{decay}\ t)t^{-a}.$
A and a are fitting parameters.
In case of the continuous emission at the point source, P was defined as the following equation.
$P=d(x)d(y)\{P1d(t1)+...+Pnd(tn)\}.$
P was defined as the sum of delta functions according to times of explosion in this case. The model became the equation including the Fourier transform. So the calculation cannot complete by calculating parameters, and the calculation result was calculated by Mathematica.

As a result, the fitting of 4 radionuclides from Chernobyl Nuclear Power Plant accident was successful at 21 observation points within 40 km.The fitting of period was from 3000 days to 5000 days. The fitting was also showed that calculated parameters A and a has a positive correlation totally but a feature of a positive correlation varied by each radionuclide.Moreover, the fitting of SO_2 from Sakurajima Volcano was also successful with the data of Arimura station which can be most affected by Sakurajima Volcano. The period of fitting was from 2002 to 2008.This result showed that the model can calculate the annual oncentration within $\pm$0.005 error.

The above results about two pollution cases shows that the model we proposed was suitable to the long-term prediction of concentration in the air. It is assumed our model can be used in case of the more complicated case such as the continuous emission at the plane source.

Keywords: long-term mass transport model, Chernobyl, Sakurajima volcano, air pollution, dispersion predicting

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AAS021-22 Room:102 Time:May 23 15:30-15:45

Variations of atmospheric radon-222 at Rishiri Island, Japan and traced fetch regions

Chunmao Zhu[1*], Hisayuki Yoshikawa[1], Hidekadzu Matsueda[2], Yosuke Sawa[2], Yosuke Niwa[2], akira WADA[3], Hiroshi Tanimoto[4]

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Measurements of atmospheric ^{222}Rn were made on Rishiri Island (45.1^{o}N, 141.2^{o}E) over the period from December 2008 to December 2010, in order to discuss the fetch regions affecting atmospheric ^{222}Rn concentration and some other gases related to climate change. Atmopheric ^{222}Rn data showed variability of a diurnal, synoptic, and seasonal time scales, which will allow us to evaluate transport and mixing schemes of atmospheric and chemical transport models.

Atmospheric ^{222}Rn concentration indicated a clear diurnal variation in summer, which was characterized of a maximum appearing before the dawn and a minimum in the afternoon, with amplitude of 0.64 Bq m^{-3}. This could be caused by the accumulation of ^{222}Rn emitted from the soil (Rishiri Island) in the stable nocturnal boundary layer, and vertical mixing of surface air with upper air due to convection (Zahorowski *et al.*, 2008). The amplitude of diurnal variation is relatively lower than that of the synoptic influence. No clear diurnal variation occurred during the remaining seasons.

Atmospheric ^{222}Rn concentration in daytime, selected to remove effects of local source on ^{222}Rn concentration, showed a broad minimum in winter and a maximum in summer, with an accompanying significant short-term variability. In February monthly mean of atmospheric ^{222}Rn concentration was 3.23 Bq m^{-3}, and in July 0.95 Bq m^{-3}. In Rishiri Island, short-term variations corresponding to the synoptic influence were large in February as compared with those in July. The amplitude of the seasonal variation (2.28 Bq m^{-3}) was somewhat larger than that of Sado Island located in the Sea of Japan. At Sado Island, maximum ^{222}Rn concentration occurred in winter and minimum in summer, which was the pattern attributable to the onshore-offshore pattern of the Asian monsoons (Chambers *et al.*, 2009).

Backward trajectory analysis of air fetch regions was conducted using extremely low and high radon events. In the annual basis, most (76.7%) of high radon events was observed in winter, of which the air masses originated predominately from 40^{o}N to 60^{o}N of Eurasian continent. 41.7% of low radon events were observed in summer, of which air masses usually originated from the relatively lower latitude of the western North Pacific, and 48.3% of low radon events were observed in spring, most in May. In May, back trajectory analysis showed a pattern that the origin of air masses was in the west North Pacific, and air mass moved westward en route the southernmost of Sea of Okhotsk to the RIO.

Keywords: atmospheric tracer, back trajectory analysis, fetch regions, radon

Japan Geoscience Union Meeting 2011
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AAS021-23 Room:102 Time:May 23 15:45-16:00

Night time radical chemisrty observed by SMILES

YASUKO KASAI[1], Philippe Baron[1*], Hideo Sagawa[1], Donal Murtagh[2], Joachim Urban[2], BrO teamn in SMILES Algorithm team[1]

[1]NICT, [2]Chalmers Institute of Technology

A new generation of super-sensitive submillimeter-wave receivers, employing SIS (Superconductor-Insulator- Superconductor) technology, will provide new opportunities for precise remote sensing observation of minor constituents in the atmosphere. SMILES had been launched at 11/09/2009, and installed on the Japanese Experiment Module (JEM) in the International Space Station (ISS). SMILES is a collaboration project between NICT and JAXA.

Mission objectives of SMILES are:

i) Space demonstration of super-sensitive SIS mixer and 4-K mechanical cooler technology

ii) Demonstration of super-sensitive global observation of atmospheric minor constituents

JEM/SMILES observes the atmospheric species such as O3, H35Cl, H37Cl, ClO, HO2, BrO, HOCl, HOBr, HNO3, CH3CN, Ozone isotope species, H2O, and Ice Cloud with the precisions in a few to several tens percents. We will present the diurnal variation of the minor radical such as BrO, HO2.

Keywords: SMILES, BrO

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AAS021-24 Room:102 Time:May 23 16:00-16:15

Diurnal variation of ClO observed with SMILES

Yu Onodera[1*], Tomohiro Sato[3], Philippe Baron[2], YASUKO KASAI[2], Eric Dupuy[2], Kazuyuki Kita[1], Joachim Urban[4], Donal Murtagh[4], Nathaniel Livesey[5]

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A new generation of super-sensitive submillimeter-wave receivers, employing SIS (Superconductor-Insulator- Superconductor) technology, will provide new opportunities for precise remote sensing observation of minor constituents in the atmosphere. SMILES had been launched at 11/09/2009, and installed on the Japanese Experiment Module (JEM) in the International Space Station (ISS). SMILES is a collaboration project between NICT and JAXA.

SMILES has observed the diurnal observation of ClO, HOCl, HO2, and HCl from October 12, 2009 to April 21, 2010. We will present 1) the result of the evaluation of ClO data quality, including inter-comparison between other satellites measurements and error analysis, and 2) the first simultaneous observations of ClO, HOCl, HO2, and HCl.

Japan Geoscience Union Meeting 2011
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AAS021-P01 Room:Convention Hall Time:May 23 16:15-18:45

Constraints on CO2 fux emissions: reconstructions of in-situ measurements from Lagrangian stochastic inversion

Ignacio Pisso[1*], Prabir Patra[1], Masayuki Takigawa[1], Takakiyo Nakazawa[2], Yousuke Sawa[3], Toshinobu Machida[4], Hidekazu Matsueda[3]

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In order to use high resolution in-situ measurements to constrain regional emissions of carbon dioxide ($¥mathbf{CO_2}$) we use a Lagrangian methodology based on diffusive backward trajectory tracer reconstructions. We use aircraft, ground and tower sites for $¥mathbf{CO_2}$ data, collected during the CONTRAIL campaign, from the MRI/JMA Tsukuba tall tower, nearby the $¥mathbf{CO_2}$ emission hot spot of the Tokyo Bay area and from the World Data Centre for Greenhouse Gases (WDCGG). Advective transport based on ECMWF analyzed meteorological winds and the WRF mesoscale model is characterized by the sensitivity/transition probability (Green?Aos function) allowing direct comparison with observations via the reconstruction of the volume mixing ratio of $¥mathbf{CO_2}$. Sensitivity to simplified boundary layer representations, turbulent mixing representations and meteorological fields was studied and applied to the assessment of publicly available inventory data. Longer time series in remote sites (e.g. the Yonagunijima island) are used to constrain the influence of far field/continental East Asia emissions. Estimated fluxes for the Tokyo Bay Area for the analyzed period in 2007 range between $4.8 ¥times 10^{-7} to $3.45 ¥times 10^{-7} $kg_{¥mathit{chem}\{CO_2\}}$ m^{-2} s^{-1} with significant time variations. We assess the uncertainties in terms of errors associated with the transport and mixing processes in the vicinity of the emission sources.

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AAS021-P02 Room:Convention Hall Time:May 23 16:15-18:45

Development of the precise measurement of carbon dioxide in the atmosphere with optical spectrum analyzer

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Carbon dioxide (CO2) is a greenhouse gas which is most significant effect on the global warming, and therefore, it is necessary to reveal the distribution and variation of CO2 precisely to understand the mechanism of the global warming. Around 20 high-resolution Fourier transform Infra-Red spectrometers (FTIRs), which are one of the instruments provided an accurate measurement of CO2 column-averaged mixing ratio (XCO2), have been operated all over the world. However, it is difficult to extend a measurement site with a high-resolution FTIR due to its cost and size. For more extended measurements of XCO2, a compact and portable instrument with considerable measurement accuracy is highly desired. For this purpose, we have newly developed an instrument measuring XCO2 by using an Optical Spectrum Analyzer (OSA). The instrument measures a CO2 absorption spectrum of the sunlight in a 1.6 ?m band. The sunlight is guided to the OSA through an optical fiber from a solar tracker. The absorption spectrum from 1.569 to 1.576 ?m with a resolution of 0.07 nm is measured. The number of sampling points is 5001, and a scan time is 135 seconds. Continuous measurements of XCO2 are carried out automatically by using a PC running with LabVIEW programs. In this presentation, we show the details of the instrument and measurement results, and will discuss a possible improvement of measurement accuracy.

Keywords: carbon dioxide, precise measurement, optical spectrum analyzer

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AAS021-P03 Room:Convention Hall Time:May 23 16:15-18:45

Observation of isotopic compositions of CO_2 and H_2O in an urban region

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It is important to estimate the terrestrial CO_2 cycle, including such factors as emissions, storages and fluxes. Knowledge of the terrestrial CO_2 cycle will help increase understanding of climate change phenomena, and aid in predicting future atmospheric CO_2 concentrations and global temperatures. Using CO_2 isotope compositions is a very powerful method for investigating the sources of atmospheric CO_2. We investigated phenomena of oxygen isotope exchanges between CO_2 and water vapour. The oxygen isotope exchanges should be happened either on the ground or on the plant leaves, and it will influence the isotopic compositions of CO_2 and H_2O in the atmosphere. We had been deployed in Nagoya from September 1 to September 30, 2010. Ambient air was sampled from the top of the Solar Terrestrial Environment Laboratory building at Nagoya University, which was surrounded by vegetation, but was located in a highly populated urban area of Nagoya. We had successfully measured CO_2 and H_2O isotopologues ($^{16}O^{12}C^{16}O$, $^{16}O^{13}C^{16}O$ and $^{18}O^{12}C^{16}O$ for CO_2, D_2O and $H_2{}^{18}O$ for H_2O) using infrared absorption laser spectrometers (Aerodyne Inc. for CO_2 and Los Gatos Research Inc. for H_2O). The CO_2 isotope laser spectrometer can measure the isotope ratios (Delta ^{13}C, Delta ^{18}O) of ambient air CO_2 in 10-second integration time with a precision of 0.1 permil in real-time. We will discuss the details of the observation result with meteorological data at the meeting.

Keywords: CO_2 isotopes, H_2O isotopes, laser spectroscopy, ecosystem, atmospheric CO_2, urban atmosphere

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Sensitivity of short timescale variability of CO2 over Narita Airport to the magnitude of regional surface fluxes

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The synoptic scale variability of CO_2 over the Narita International Airport (lat 35.8^{o}N, 140.4^{o}E, 43m a.s.l.) was investigated using measurements obtained from frequent observation by commercial aircraft combined with analyses of results from a transport model simulation for the year 2007. The standard deviation (SD) from fitted curves was considered as the metric of synoptic scale variability of CO_2 mixing ratio. Tagged simulation was conducted to evaluate the relative contributions of regional fluxes to the synoptic scale variability over Narita. Tags were put on anthropogenic (FF) and terrestrial biosphere(TB) fluxes from 6 regions (Japan, East Asia, East Russia, Southeast Asia, India, and Himalaya). The result indicated that the major contribution was made by the fluxes in East Asia (mainly China) in the free troposphere (FT) and by those in Japan in the planetary boundary layer (PBL), respectively.

A sensitivity analysis was performed to evaluate the relative influence of transport and flux variations on the CO_2 SD over Narita for 2007. When the FF fluxes from East Asia and those from Japan were doubled, changes in annual mean SD over Narita with height was; 41% and 3% at 9km, 61% and 4% at 5km, 19% and 83% at 0.5km, respectively. This result indicated that SD over Narita was sensitive to transport (synoptic scale meteorological variability) from upwind in FT, but depends largely dependent on the magnitude of local fluxes in the PBL.

Among 12 tagged fluxes, those that made more than 20% difference in monthly CO_2 SD when flux magnitude was doubled were FF fluxes from Japan and East Asia, and TB fluxes from East Asia and East Russia. Throughout the year, CO_2 SD in the PBL and the FT were most sensitive to the FF flux from Japan and East Asia, respectively. In summer, the contribution of the TB flux from East Asia and East Russia to CO_2 SD was increased in the FT, when strong CO2 uptake makes large negative flux. The TB flux from East Russia affected the most at 2-4km altitude in July, and those from East Asia affected at 8-10km altitude in September. This delay corresponded to the seasonality of the sink magnitude within each tracer region. Meteorological analysis and forward/backward trajectories indicated that low CO_2 airmass affected by TB fluxes in East Russia drifted to the northeast and occasionally transported to Narita by northeasterly behind the cold front whereas those affected by TB fluxes in East Asia were often directly convected to the UT by tropical depression etc., and transported to Narita via UT much more rapidly. These transport mechanisms explained the different altitude ranges that were sensitive to the TB fluxes from East Russia and East Asia in summer 2007.

Keywords: CO2, aircraft observation, short timescale variation, carbon cycle, flux, transport

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Sensitivity experiment of vertical diffusional parameter and CO2 surface flux on CO2 transport model MJ98-CDTM

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We intend to carry out data assimilation experiments of CO2 by using a chemical transport model. In general, assimilation results are seriously affected by the performance of the chemical transport. Prior to assimilation experiments, we are studying the performance of the chemical transport model, related to the impact of vertical diffusion and the surface flux.The model used in this experiment is an online chemical transport model directly coupled to an atmospheric general circulation model (MJ98-CDTM), which is developed at Meteorological Research Institute (MRI). The model is integrated with respect to time for five years from 2002 and nudged to the atmospheric reanalyses of JRA-25. CO2 distributions for 2006 are compared with the JMA CO2 analyses.

The chemical transport model parameterizes effects of shallow convections as an enhanced vertical diffusion. The vertical diffusion has large impacts on the distributions of CO2 and it is carefully tuned up through the comparison with JMA CO2 analysis. Further study is needed to assess actual effects of vertical diffusions.

The chemical transport model is subject to the ambiguity of the CO2 surface flux. The so-called CASA data is widely used. CASA data are processed in different ways between MRI and Japan Agency Marine-Earth Science and Technology (JAMSTEC). For this reason, we also examine sensitivity experiments using two CASA fluxes, from MRI and from JAMSTEC. As is expected, CO2 distributions are also sensitive to the replacement of the surface flux. However, both results are still different from the JMA analyses.

Now, we are going to conduct data assimilation experiments using real observation data. In this course, we will examine the sensitivity of data assimilation to the vertical diffusion and the surface flux. The surface flux will be modified from the first guess in the data assimilation. However, it is suspected that the surface flux is affected by the first guess of the climatic value, because coverage of observation data is not dense enough to converge flux values.

Keywords: CO2, data assimilation

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Aircraft and tower measurements of carbon dioxide in the PBL and FT over taiga in West Siberia (2002-2010)

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Figure. Vertical profiles of CO_2 concentration (ppm) observed over the tower.

1. Introduction

To understand the difference in CO_2 behavior between planetary boundary layer (PBL) and free troposphere (FT) over Siberian taiga, we have conducted CO_2 measurements using a small aircraft and a tower at the taiga in West Siberia since 2002. Continuous CO_2 time series at 4 levels (5, 20, 40, and 80 m) were monitored with the tower. Up to 3 km vertical CO_2 profiles above the tower in the fine day were frequently obtained with the aircraft.

2. Method

Carbon dioxide concentrations were measured continuously at the tower located in Berezorechka village (56^{o}09N, 84^{o}20E). Sampled air from 4 levels was dehumidified and then introduced into a NDIR (LI-820, LI-COR; LI-7000 was used until September 2008). Measurement precision is +-0.3 ppm. The more detail information of the system was described in *Sasakawa et al.* [2010a]. Small CO_2 measurement device based on a NDIR (LI-800, LI-COR) equipped with flow and pressure regulation system was developed and installed in the small aircraft (Antonov An-2). Two standard gases are introduced into the NDIR every 5 minutes. Overall measurement precision is estimated to be +-0.3 ppm when we use 2 seconds averaged data. An-2 ascended to 2 km (winter) or 3 km (summer) above the tower and then descended to 0.15 km to obtain the vertical profile of CO_2 concentration. The routine aircraft measurement had been conducted basically in the afternoon with the frequency of 2-4 times per month until 2007 March despite low frequency after then.

3. Results and discussions

Figure shows the vertical profiles of CO_2 concentration observed by routine flights (261 times) from 2002 to 2010. Carbon dioxide concentrations showed slightly higher in the lower altitude during the dormant season. On the other hand, CO_2 concentration during summer gradually decreased with decrease in the altitude during summer, which implies the effect of photosynthesis by vegetation of the taiga during the growing season. The same tendency was observed in the previous years [*Machida et al.,* 2005]. Unfortunately there was no simultaneous tower data in 2010 due to equipment malfunctions.

To compare the temporal CO_2 variation between in the PBL and FT, we defined the PBL height using vertical profiles of temperature, potential temperature, and specific humidity. We found that seasonal variation of PBL height with maximum during summer (-over 3 km). Annual mean CO_2 in the PBL was always higher (approx. 2 ppm) than that in the lower FT, which is mainly due to rectifier effect. The annual means also showed an increase of 11.6 ppm (PBL) and 11.2 ppm (FT) from 2003 to 2009. We will also present diurnal variation of CO_2 vertical profiles up to 3 km observed in the summer of 2002-2004.

Generally daily minimum at the tower was observed in the afternoon when active vertical mixing occurred [*Sasakawa et al.,* 2010b]. Thus, we calculated daytime mean with averaging the data observed in 13:00-17:00 LST to compare with the data from An-2 observation. Daytime mean concentrations observed in the tower (40 and 80 m) agreed well with those in the PBL observed with An-2, which suggests that daytime tower data can be the representative values in the PBL.

References

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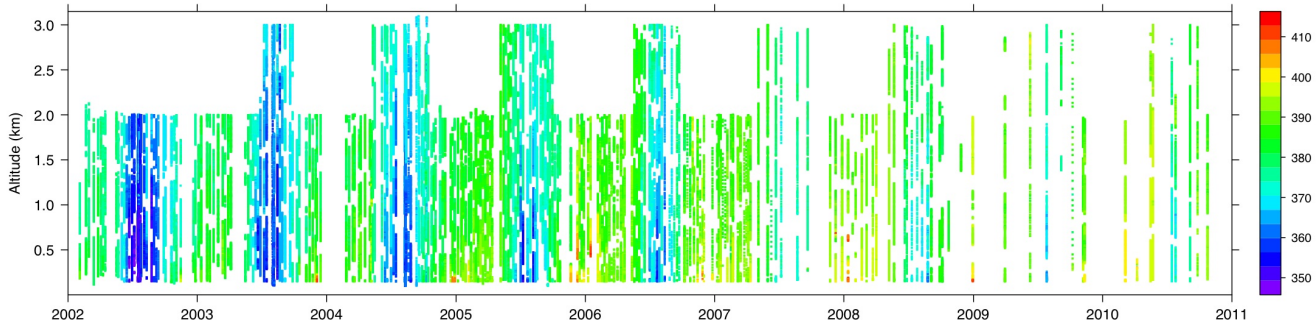

Keywords: Siberia, taiga, tower, PBL, carbon dioxide, aircraft

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AAS021-P07 Room:Convention Hall Time:May 23 16:15-18:45

Comparison of 2 versions of a global atmospheric transport model (NIES99 TM and NIES08 TM) using APO observations

Chika Minejima[1*], Yasunori Tohjima[1], Dmitry Belikov[1], Shamil Maksyutov[1]

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We show comparisons of two versions of an atmospheric global transport model, NIES99 and NIES08, and observations using Atmospheric Potential Oxygen (APO = O_2 + 1.1 CO_2). APO is defined so that it is invariable with the terrestrial biotic exchanges and it is not as sensitive to fossil fuel burning activities as CO_2 or O_2 is. Thus, the main variation is caused by air-sea exchanges of O_2. Therefore, APO has a unique flux distribution and is able to provide a good test to the transport models from different perspectives.

National Institute for Environmental Studies (NIES) has been making observations of CO_2 and O_2 on cargo ships which have been repeating round-trip cruises between Japan and Canada/the United States and between Japan and Australia/New Zealand since December 2001. With this valuable data set, we compare the observations and model results including annual mean distributions and seasonal amplitudes.

For the NIES99 TM and NIES08 TM, the same flux set was used to make transport difference clearly. We used oceanic fluxes of O_2, N_2 and CO_2 , and fossil fuel burning anthropogenic fluxes of CO_2 and O_2. As for the oceanic O_2 and N_2 fluxes, climatological monthly anomalies of Garcia and Keeling [2001] and annual mean oceanic O_2 and N_2 fluxes from the annual-mean ocean inversion studies of Gruber et al. [2001] and Gloor et al. [2001], respectively, are used. As for the oceanic CO_2 fluxes, we use two sets of monthly sea surface CO_2 flux climatology of Takahashi et al. [2002] and Takahashi et al. [2009]. For the anthropogenic CO_2 flux, global fossil fuel CO_2 emission with spatial resolution of 1 deg. x 1 deg. for the year 2006 from CDIAC database is repeatedly used every year in this simulation. The O_2 consumptions associated with the fossil fuel burning is calculated from the fossil fuel CO_2 fluxes and the -O_2:C exchange ratios for the burnings.

In general, NIES99 TM reproduces the seasonal amplitudes, the annual mean values and rectifier effect at northern North Pacific better than NIES08 TM does. NIES08 TM represents APO in the Southern Hemisphere more smoothly because of more balanced tracer transport due to strong mass conservation. We will show the comparisons more in details in this presentation.

Keywords: atmospheric O2, atmospheric potential oxygen (APO), seasonal cycle, global atmospheric transport model

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Source signals in N2O and CH4 variability in the upper troposphere over the western Pacific derived by model simulations

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Multiple greenhouse gases simulations with tagged-tracers are performed to mainly understand surface-source influences on latitudinal-temporal variabilities of nitrous oxide (N2O) and methane (CH4) in the upper troposphere (UT) over the western Pacific observed by the Automatic air Sampling Equipment (ASE) included in the CONTRAIL project. We use greenhouse gases concentration data, which have been obtained almost fortnightly in the altitude range of 9-11 km between Sydney or Brisbane, Australia and Tokyo, Japan for the period Dec 2005 - Mar 2009. An atmospheric general circulation model-based chemistry transport model (ACTM), which is nudged toward the Japanese 25 year ReAnalysis data from the Japan Meteorological Agency (JMA) (JRA-25), is used in this study. Dynamical structure in the tropical UT region in ACTM is reasonably validated by the fact that ACTM simulation of mean latitudinal SF6 gradient for the observation period is almost perfectly consistent with the observation within 0.03ppt, which is much smaller than the measurement precision. For tracing the origins of N2O and CH4 in model, the globe is separated into more than ten of emission regions so that each region emission affects the corresponding tagged-tracers concentration variation on the globe through the atmospheric transport. The tracers simulation results and observation results are detrended, and their seasonal or shorter eventual concentration variabilities are compared. In case of N2O, concentration values around 30N and 30S are largely fluctuated by stratospheric intrusions, which lower N2O concentration in the UT, but some surface source signals can be still detectable especially around 30N by removing data highly affected by the stratosphere. That indicates that N2O around 20-30N seems to be most affected by Middle East and South Asia region, and secondly by East and Southeast Asia region mainly in summer. It is possible that monsoon and Tibetan Plateau work to transport N2O emitted from South Asia to this region in the UT over the western Pacific. Around 10S, N2O seems to be relatively dominated by emissions from Australia, but the degree is not so prominent compared to above two Asian regions for 20-30N. CH4 shows slightly different features of affecting source regions from those of N2O. It is indicated that China and India affect 26-30N region in the UT over the western Pacific in summer and fall, respectively. Meanwhile, region around 10S is dominantly affected by Malaysia-Indonesia-Papua New Guinea emission region. It might be necessary to take into account CH4 loss by OH enhanced in the tropics and transport of CH4 through the region to the UT for better understanding the different emission region affecting around 10S between N2O and CH4.

Keywords: N2O, CH4, upper troposphere, model simulation

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Distribution of the UT/LS water vapour retrieved from the JEM/SMILES observations

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Water vapour in the upper troposphere and lower stratosphere (UT/LS) region plays an important role in the weather and climate system on Earth. Despite its high importance, we are still lacking thorough understanding of the distribution and climatological roles of UT/LS water vapour because of insufficient temporal observational coverage.

New insight into the UT/LS water vapor can be provided by high sensitivity observations with the Superconducting Submillimeter-Wave Limb-Emission Sounder, SMILES: a limb emission sounder attached on the Japanese experiment module (JEM) onboard the International Space Station (ISS). One of the unique characteristics of the SMILES observation is the non-sun synchronous orbit of ISS. By accumulating the data from the several ISS evolutions, diurnal variations of the atmospheric constituents can be analyzed. Although SMILES has no water vapour transitions included in its observation spectral range, a significant contribution of the water vapour opacity of the 620 GHz and 557 GHz transitions comes into the SMILES observed spectra.

We will present the first results of the UT/LS humidity retrieval from the JEM/SMILES measurements focusing on its diurnal variation.

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AAS021-P10 Room:Convention Hall Time:May 23 16:15-18:45

Evaluation of updated photodissociation scheme on CHASER model: the impact to chemical reaction in troposphere

Hisashi Yashiro[1*], Kengo Sudo[2], Masayuki Takigawa[1]

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To improve the accuracy of calculated photodissociation rates (J-values) in global chemical climate/transport model (CHASER), we updated the scheme for estimation of absorption cross section (ACS) and quantum yield (QY), which is coupled with the broadband radiative transfer model in AGCM. The new scheme used data from NASA-JPL recommendations in 2006 (Sander et al., 2006). Based on the method of Langdraf and Crutzen (1997), ACS and QY were weighted by the attenuated solar spectrum and were averaged into the bins. The average value obtained at each grid point of the model was approximated by multivariable polynomial of temperature, pressure, and a partial ozone column. The comparison between the result of old and new scheme shows that 1) the changes of J-values for ozone photodissociation of both two channels (O_3->O_2+O(^{1}D), O_3->O_2+O) are less than 10% in the troposphere but indicate considerable increase in tropics and decrease in polar region, 2) J-values for some ketones increase in free troposphere, reflecting the pressure-depentdent variation of quantum yield, and 3) updated ACS of formaldehyde (HCHO) increased the photodissociation rate of both of channels (HCHO->CO+2HO_2, HCHO->CO+H_2), and as a result, the chemical production rate of CO was increased in the tropical region.

Keywords: atmospheric chemistry, photochemical reaction, chemical climate model, atmospheric radiation

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A case study of the transport of tropospheric nitrogen dioxide from China observed from space

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We conducted a case study on the transport of tropospheric nitrogen dioxide (NO_2) from China to the East China Sea and Japan by using satellite measurements. In the event, high concentrations of NO_2 (~$1x10^{16}$ cm^{-2}) were found off the coast of Shanghai and Kyushu. A trajectory analysis suggests that the air parcels with the high concentration of NO_2 found around Kyushu passed over Shanghai about 24 hours before, indicating that the source of the observed NO_2 is from pollution in that area. The satellite observations will be also compared to results from a regional chemical transport model and data from the ground air-pollution monitoring network in Japan.

Keywords: air pollution, nitrogen dioxide, troposphere, satellite observation, transport

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Development of DOAS O3 radiometer using D-UV LED

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The UV ozone radiometer has been widely used to measure atmospheric ozone concentration for monitoring the environment and for industrial purposes. Because it uses a Hg lamp as a UV source, much Hg waste is produced. Because it used single (253.7 nm) wavelength for the absorbance measurement, it needs frequent zero-level calibration, leading to slow response.

Recently, LED emitting deep UV (D-UV) near 250 nm is developed. By using D-UV LED, a prototype differential absorption radiometer for measuring ozone has been developed. Using two wavelength, 255nm and 285nm (FWHM 15 nm), LEDs with about 30-cm absorption cell, it can measure ozone between 0 to 10 ppm with an accuracy of about 5 ppbv. Now, high-precision and accurate version of this type ozone monitor is produced.

Keywords: ozone, measurement technique, UV LED

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Evaluation of a method for measurement of black carbon particles suspended in rainwater

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Black carbon (BC) aerosols are produced by incomplete combustion of fossil fuels and biomass. They contribute to global warming due to their strong absorption of solar radiation. The distribution of BC is controlled by emission, transport, and wet deposition during transport. BC concentration in rainwater is an important parameter for understanding of the detailed processes of wet deposition of BC. Reliable data of wet deposition flux of BC is also useful in validating representation of the removal processes by three dimensional models used for assessing the impacts of BC on climate.

In previous studies, total mass concentrations of BC in rainwater were mainly measured by a thermal-optical transmittance technique applied to BC particles collected on filters. However, this method is not practical for weak rain or high time-resolved measurements during each rain event because we need more than 100 mL of rainwater to determine the BC mass concentration by this method. It also cannot provide BC size distribution in rainwater, which are important for understanding of the CCN activities of BC. In this study, we evaluated a new method to measure BC particles suspended in rainwater. The new method utilizes an ultrasonic nebulizer and a laser-induced incandescence technique for BC detection (Single Particle Soot Photometer: SP2). We demonstrate that this method is practical for measurements of mass concentrations and size distribution of BC in a small amount (several mL) of rainwater sample.

The rainwater sample is transferred to an ultrasonic nebulizer by a peristaltic pump at a constant flow rate. In the ultrasonic nebulizer, some fraction of the introduced rainwater is converted to small droplets in air flow, and they are heated at 140 C and dried at the downstream. Remaining non-volatile cores of individual droplets in the air flow are introduced into the SP2 for single-particle detections of BC.

In this method, we need laboratory experiments to determine the fraction of BC mass in rainwater sample transferred into SP2. We used a commercially available carbon black AquaBlack001 and AquaBlack162 (Tokai Carbon Co. Ltd.) as laboratory standards of BC aqueous solution for the experiments. Under the optimized operating condition of the nebulizer, we determined the fraction R to be about 9% by using samples of various BC concentrations. The R value was almost independent of BC mass concentration and concentrations of other co-existing solutes. For the given value of R, we can determine the BC mass concentrations in rainwater samples.

Second, we tested the reproducibility of the measurement system by repeated measurements of one rainwater sample. This test also showed that the volume of rainwater required for reliable BC measurement is less than 5 mL. We also tested the temporal stability of a rainwater sample over a few months. The measured mass concentrations of a rainwater sample just after sampling and the same one after a storage in refrigerator over several months agreed to within 14%, indicating a relatively small effect of the degradation of rainwater samples over time.

The size distribution of BC in rainwater can change if multiple BC particles are contained in some of droplets generated by the nebulizer, because of coagulation associated with the evaporation of water droplet. Actually, it was found that the size distribution of BC particles shifted to larger sizes as increase of BC concentration. In order to minimize this artifact, we diluted the rainwater sample with pure water before measurements of size distribution. We derived size distribution of BC in a rainwater sample collected in Tokyo in December 2010 following the method described above. For this sample, the BC size distribution can be approximated by a lognormal function with count and mass median diameters of 98 and 190 nm, respectively.

Keywords: black carbon, aerosol, wet deposition

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Estimation of deposition rate of Black Carbon aerosol during the transportation over sea by aircraft observation

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Black Carbon aerosol (BC) is significant for the climate change because it absorbs the solar radiation to heat surrounding air. In China, a large amount of BC is emitted with anthropogenic activities, and its global influence depends on the deposition rate of BC during the transportation.

In this study, the deposition rate of BC in air masses transported from Asian continent over East-china Sea is estimated. The BC deposition rate is evaluated from the decrease of BC concentration with the transportation time. The transport time and distance are evaluated by the backward trajectory analysis. The decrease of BC concentration by the deposition is evaluated from the ratio of the BC concentration and the increase of CO concentration from the CO background value (d-CO value). The median ratio of BC concentration to d-CO value in the air masses which had passed about 72-hours after leaving the Asian continent shoreline is about 65% smaller than that near the shoreline.

Keywords: Brack Carbon aerosol, CO

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Correlation of BC and CO during biomass burning and urban pollution episodes in eastern China

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Strong emissions of pollutants (e.g., BC and CO) in East China due to ever-quickening industrial development, surging automobile ownership, and intensive seasonal burning of biomass are well known, however, estimation of its emission has large uncertainties. Generally relationship between black carbon (BC) and carbon monoxide (CO) will help improve their emission inventories and the further evaluation of global/regional climate forcing effects. In present work, two field observation studies were performed at background mountain site (Mt.Huangshan 2007˜ 2009), and at coast site (Rudong, Jiangsu province 2010) about 100 km northern to Yangtze River Delta Region respectively. Results from Mt.Huangshan show that annual mean BC concentration was 654.6 +/- 633.4 ng/m3 with maxima in spring and autumn, when biomass was burned over a large area in eastern China. The yearly averaged CO concentration was 446.4 +/- 167.6 ppbv, and the increase in the CO concentration was greatest in the cold season, implying that the large-scale domestic coal/biofuel combustion for heating has an effect. The BC-CO relationship was found to have different seasonal features but strong positive correlation ($R > 0.8$). Trajectory cluster study combined with measurements of urban PM10 concentrations and satellite observations demonstrated that the deltaBC/deltaCO ratio for a plume of burning biomass was 12.4 ng/m3/ppbv and that for urban plumes in eastern China was 5.3 +/- 0.53 ng/m3/ppbv. The field campaign at Rudong site displayed different results with relatively lower deltaBC/deltaCO values of 4.5 +/- 0.2 ng/m3/ppbv for urban plumes and 8.9 +/- 0.3 ng/m3/ppbv in biomass burning influencing episode. This result for urban plumes was quiet similar with value (4.1 ng/m3/ppbv in INTEX-B) from statistical approaches, and transportation and industry were identified as controlling factors of the BC-CO relationship. Large uncertainties still existed for biomass burning smoke, and biomass types (grass, agriculture residues or twigs), combustion condition (inflammation or smoulder) seemed to be essential reasons to explain the discrepancies among the results.

Keywords: Black carbon, carbon monoxide, Emisson Inventory, Back trajectory

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Estimation of radiative impact of soil dust by using WRF/Chem

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We are now trying to include mineral dust aerosols in the model, because mineral dusts have a large impact on the radiation in East Asia in spring. In the present study we use WRF/ARW (Advanced Research WRF) version 3.1.1, and the gaseous and aerosol chemistry is based on RADM2 and MADE/SORGAM, respectively. Dust flux is estimated based on Shaw et al. (2008), and slightly modified to adapt to the East Asia following to Uno et al. (2004). Gravitational settlement of mineral dust is based on GOCART (Goddard Chemistry Aerosol Radiation and Transport) model. Landuse is based on the MODIS land-use data. For the evaluation of the model, we conducted calculation for spring 2006 and compared with ground-based observation in Beijing. The model well captured the increase of PM10 during 17-19 April and 22-24 April, and it was estimated the most part of this PM10 particles were soil dust.

The impact of soil dust on shortwave radiation was also estimated by the model, and found 15W/m2 of decrease was caused by the soil dust during the dust event.

Keywords: Atmospheric Chemistry, soil dust, East Asia, shortwave radiation, transport

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Measurements of wavelength dependence of aerosol light absorption at Nagoya during summer 2010

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Atmospheric aerosol particles affect the Earth's energy radiation balance by scattering and absorbing incident light. The optical properties of black carbon (BC) are changed by coating with sulfate or organic matters (lens effect). In addition to BC, there is a possibility that a part of organic carbon, which is called "brown carbon", absorb solar radiation at UV or short visible wavelength may affect the Earth's energy radiation balance. But it is difficult to estimate contributions of the lens effect and brown carbon using the conventional filter-based photometer because of multiple scattering and change in quality on a filter. In this study, we use three wavelengths photo-acoustic spectrometer (PASS-3) to achieve direct measurement of wavelength dependence of absorption coefficient of ambient aerosols.

Simultaneous measurements of absorption and extinction coefficient were performed at Higashiyama campus of Nagoya University from 25th July to 4th August 2010 using the PASS-3(405,532,781nm) and a originally developed cavity ring-down spectrometer (CRDS:355,532nm). The mass concentrations of elemental carbon (EC) and organic carbon (OC) were measured by thermal optical techniques. The concentration of ozone was also monitored by commercially available O3 detector. Using the obtained data, the contributions of the lens effect and brown carbon to aerosol light absorption has been discussed.

Keywords: aerosol, optical properties, photoacoustic spectroscopy, lens effect, black carbon, brown carbon

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Characterization of Organic Aerosols from Isoprene at a Forested Site in Japan

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Figure 1. GC-MS total ion chromatogram

Introduction

Organic compounds are major components of atmospheric aerosols which affect the global radiative balance by directly scattering sunlight and indirectly as cloud condensation nuclei. A significant fraction of atmospheric organic aerosols are formed through gas-to-particle conversion process with oxidation of precursors such as volatile organic compounds (VOCs) in the gas phase. These are referred to as secondary organic aerosols (SOA). Recently, SOA formation from biogenic volatile organic compounds (BVOCs), especially isoprene has been paid attention. Global isoprene emission is far higher than those of biogenic terpenes and anthropogenic VOCs. Claeys et al. (2004) first identified oxidation products of isoprene in the Amazonian rain forest aerosols. Since then, some studies have been conducted to detect the oxidation products in ambient air samples, however, there is little data in Asian regions. We have measured organic compounds in the aerosols collected at a forest with strong isoprene emitters in Japan.

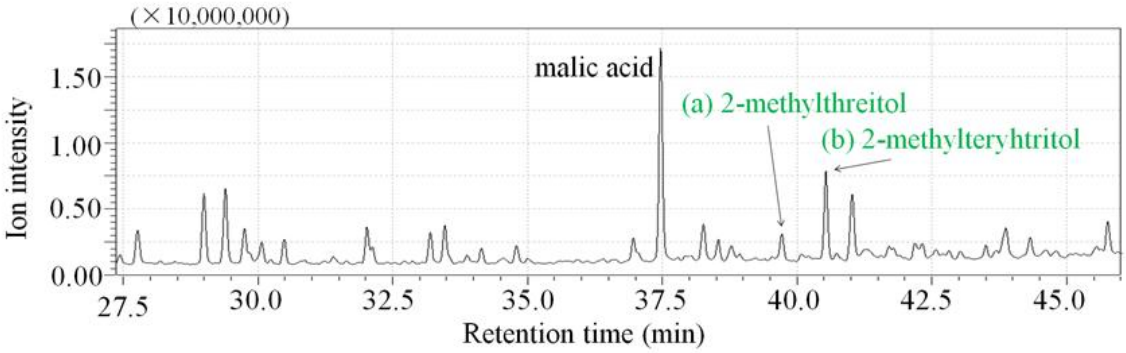
Methods

Atmospheric aerosols were collected at a tower above a forest canopy in southern Kyoto, Japan, between 2009 and 2010. YMS is in a mixed temperate forest with *Quercus serrata* (Japanese oak); one of the major tree species in Japan. The oak is a strong isoprene emitter (Okumura et al., 2008). A high-volume sampler with a PTFE filter was installed in a tower (about 30m above ground) to collect aerosol samples. Organic compounds on a part of the PTFE filter were extracted in a 1:1 dichloromethane/ methanol mixture under ultrasonic agitation. The extracts were filtered through a Teflon filter and the filtrate was concentrated by reducing the volume with a rotary evaporator. The concentrates were dried under a nitrogen stream. The dry residue was derivatized by adding bis(trimethylsilyl)trifluoro- acedimide (BSTFA, with 1% TMCS) and pyridine. Samples were heated for 2h at 70^{o}C to complete the derivatization reaction and then subjected to GC-MS analysis. To determine the quantity of compounds, glycerol and meso-erythritol were used for internal standard substances.

Results and Discussion

We have identified calboxylic acids and tetrol compounds as well as levoglucosan, a tracer of biomass combustion. Based on the retention time data, etc., the detected tetrols were likely to be isoprene SOA tracers (e.g. 2-methyltreitol and 2-methylerythritol), reported in the previous experiments (e.g. Claeys et al., 2004). A strong relationship between temperature and total mass concentration of the tetrols is quite similar to the temperature dependence of isoprene emissions (Okumura et al., 2008), also suggesting that they are isoprene SOA tracers.

Claeys, M. et al., (2004). Science, 303, 1173-1176.
Okumura, M. et al., (2008). J. Agric. Meteorol., 64, 49-60.


(×10,000,000)
malic acid
(a) 2-methylthreitol
(b) 2-methylteryhtritol
Ion intensity
1.50
1.00
0.50
0.00
27.5
30.0
32.5
35.0
37.5
40.0
42.5
45.0
Retention time (min)

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Application of Positive Matrix Factorization to Data of Organic Mass Concentration Obtained by Aerosol Mass Spectrometer

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Due to long-range transport of air pollution from neighbor countries, monitoring air quality in our environment is a public concern. Particularly, understanding of level and composition of airborne particulate matter (or so called aerosols) is very important due to its possible link to climate change and adverse health effect. We have carried out ambient measurements by Aerodyne's aerosol mass spectrometer (AMS) for analysis of chemical composition of aerosols (sulfate, nitrate, ammonium, chloride, and organics) at Fukue island, Nagasaki where long-range transported aerosols from other eastern Asian countries may directly impact the air quality. This time, we analyze the mass spectra data obtained in the spring 2009 by Positive Matrix Factorization for quantitatively better understanding of organic aerosols.

The PMF analysis showed that the observed data are reasonably solved with the factors between two and five. Although the results from the five factorial solution were indicated as "best fit" according to Q-value evaluations, comparison with reference mass spectra indicates that the five factorial solution seemed to unnecessarily break down physically meaningful mass spectra patterns into patterns that were resembled each other. Based on the combination of the Q-value evaluation and the comparison of reference mass spectra, we concluded that a 3 factorial solution is plausible to explain the observations: factors of oxygenated organic aerosol, hydrocarbon-like organic aerosol, which were well correlated with ozone and carbon monoxide respectively, and another which has not been reported to date.

Keywords: AMS, PMF, Aerosol, SOA

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Measurement of oxidants present in secondary organic aerosol using spectrophotometric Iodometry

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Redox reactions proceeding in atmospheric particles and its aqueous solution are believed to affect not only chemical composition of aerosol but also human health; however, the amount of oxidant present in secondary organic aerosol (SOA), which is a major component of atmospheric fine-mode aerosol, remains poorly understood. In this study, we conducted a series of laboratory experiments on the alpha-pinene ozonolysis and the photooxidation of 1,3,5-trimethylbenzene (TMB) under high NOx conditions, and quantified oxidants present in SOA particles produced employing spectrophotometric iodometry (KI method). The ratios of oxidant to the total SOA mass measured for the reactions of pinene and TMB were 0.45 +/- 0.08 and 0.17 +/- 0.03, respectively. The result of pinene agreed with a literature value (0.47 +/- 0.12), and the result of TMB was close to a literature value of toluene (0.16-0.18). On the other hand, a recent study using dithiothreitol (DTT) method has reported that the oxidation potential of SOA formed from the TMB photooxidation is higher than that of SOA formed from the monoterpenes oxidation. The oxidants detected by the KI method are mainly organic peroxides (ROOH and ROOR). In the DTT method, DTT is extremely consumed by chain reactions catalyzed by quinones as well as reactions with organic peroxides. To interpret the oxidation potentials obtained by KI and DTT methods, direct comparisons with toxicity tests of SOA particles would be necessary.

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Keywords: secondary organic aerosol, oxidative stress, cloud process, iodometry, aromatic hydrocarbon, monoterpene

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Sulfur isotope fractionations in carbonyl sulfide sink reations in atmosphere.

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Carbonyl sulfide (OCS), a relatively inert tropospheric sulfur compound is thought to play an important role as a source of background stratospheric sulfate aerosols (SSA). The main sink for OCS in the stratosphere is photolysis which reaches 80% of the total chemical sink at above 20 km; other sinks include reactions with O(^{3}P) and OH. In order to investigate isotope fractionation in OCS sink reactions at present atmosphere, we undertook laboratory experiments. First, we investigated wavelength dependence of kinetic isotope effect in OCS photolysis. The absolute ultraviolet (UV) absorption cross sections of OCS isotopologues OC^{32}S, OC^{33}S, and OC^{34}S were measured using labeled samples prepared in the laboratory. The observed cross section of OC^{32}S is consistent with previous reported cross sections of natural abundance samples. The peak positions for labeled samples were shifted in a systematic way. Isotopologue absorptions cross sections were not only shifted in energy but in intensity. In particular, the OC^{33}S isotopologue had the largest cross section of the measured OCS isotopologues. This finding indicates that OCS photolysis may have a positive mass-independent effect on sulfur in the stratosphere. In addition, relative rate constants of OCS sink reaction with O(^{3}P) and OH were investigated using photochemical chamber.

Keywords: isotope fractionation, photolysis, sulfur cycle, carbonyl sulfide, wavelength dependence

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NMD Fractionation Estimated from SO2 Isotopologues Photolysis UV Spectra

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We report measurements of the ultraviolet absorption cross sections of 32SO2, 33SO2, 34SO2 and 36SO2, recorded using a Bruker vacuum Fourier-transform spectrometer 190 to 220 nm at 293 K with a resolution of 8 cm-1. The samples were produced by combustion of isotopically enriched 32S, 33S, 34S and 36S elemental Sulfur. The spectrum of the 32S, 33S and 34S samples are in agreement with previously published spectra. We conclude that the main source of error is the standard deviation of the measurements themselves while the root-mean-square of other sources of error is in average 20%.The spectra of the isotopically pure species were corrected based on the isotopic composition of the samples used for the measurements. The absorption spectra show rich vibrational structure and the positions and widths of the peaks change with isotopic substitution in a complex fashion.

We present here the application of the first direct laboratory studies of the absorption cross sections of the 32SO2, 33SO2, 34SO2 and 36SO2 isotpologues. These data, together with extrapolated spectra by red shifting are used to derive the photolytic fractionation factors and the mass independent anomaly at different levels of present atmospheric levels (PAL) of oxygen. By comparison of the employed data sets it is discussed the origin of the mass independency factor during the photolysis in the presence of oxygen. Our results show that the mass independency is a combination of red shifting of the heavier isotopes and the change in the absorption cross section which is independent of the isotopomers. The obtained results give a new insight of the possibilities of mass independent fractionations observed in archean rock samples or stratospheric aerosols.

Keywords: SO2, Sulfur isotopes