

Japan Geoscience Union Meeting 2011

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AAS021-P01

Room:Convention Hall

Time:May 23 16:15-18:45

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

Ignacio Pizzo^{1*}, Prabir Patra¹, Masayuki Takigawa¹, Takakiyo Nakazawa², Yousuke Sawa³, Toshinobu Machida⁴, Hidekazu Matsueda³

¹JAMSTEC, ²Tohoku University, ³MRI, ⁴NIES

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's 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 4.8×10^{-7} to 3.45×10^{-7} $\text{kg}_{chem\{CO_2\}} \text{m}^{-2} \text{s}^{-1}$ 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

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

Satoshi Kusakari^{1*}

¹Solar-Terrestrial Environment Laboratory

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 μ 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 μ 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

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Observation of isotopic compositions of CO₂ and H₂O in an urban region

Masahiro Takemura^{1*}, Ryuichi Wada¹, Tomoki Nakayama¹, Yutaka Matsumi¹, Tetsuya Hiyama², Gen Inoue², Naoyuki Kurita³, Yasushi Fujiyoshi⁴, Kenichiro Muramoto⁵, Satoru Takanashi⁶

¹STE laboratory, Nagoya Univ., ²Research Inst. For Humanity and Nature, ³JAMSTEC, ⁴Inst. Low Temp. Sci., Hokkaido Univ., ⁵College of Sci. and Eng., Kanazawa Univ., ⁶FFPRI

It is important to estimate the terrestrial CO₂ cycle, including such factors as emissions, storages and fluxes. Knowledge of the terrestrial CO₂ cycle will help increase understanding of climate change phenomena, and aid in predicting future atmospheric CO₂ concentrations and global temperatures. Using CO₂ isotope compositions is a very powerful method for investigating the sources of atmospheric CO₂. We investigated phenomena of oxygen isotope exchanges between CO₂ 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 CO₂ and H₂O 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 CO₂ and H₂O isotopologues (¹⁶O¹²C¹⁶O, ¹⁶O¹³C¹⁶O and ¹⁸O¹²C¹⁶O for CO₂, D₂O and H₂¹⁸O for H₂O) using infrared absorption laser spectrometers (Aerodyne Inc. for CO₂ and Los Gatos Research Inc. for H₂O). The CO₂ isotope laser spectrometer can measure the isotope ratios (Delta ¹³C, Delta ¹⁸O) of ambient air CO₂ 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: CO₂ isotopes, H₂O isotopes, laser spectroscopy, ecosystem, atmospheric CO₂, urban atmosphere

AAS021-P04

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

Tomoko Shirai^{1*}, Toshinobu Machida¹, Shamil Maksyutov¹, Jiye Zeng¹, Hidekazu Matsueda², Yousuke Sawa², Yosuke Niwa², Kaz Higuchi³

¹NIES, ²MRI, ³York Univ.

The synoptic scale variability of CO₂ over the Narita International Airport (lat 35.8°N, 140.4°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 CO₂ 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 CO₂ 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 CO₂ 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, CO₂ 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 CO₂ 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 CO₂ air mass 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

AAS021-P05

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

Yoshiaki Yokoo^{1*}, Takashi Maki², Kazuyuki Miyazaki³, Kazutaka Yamada⁴, Toshiki Iwasaki¹

¹Tohoku University, ²MRI, ³JAMSTEC, ⁴JMA

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

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

Motoki Sasakawa^{1*}, Toshinobu Machida¹, Noritsugu Tsuda², Kou Shimoyama³, Mikhail Arshinov⁴, Denis Davydov⁴, Alexandrov Fofonov⁴, Oleg Krasnov⁴

¹NIES, ²GEM, ³Hokkaido University, ⁴IAO, RAS, Russia

Figure. Vertical profiles of CO₂ concentration (ppm) observed over the tower.

1. Introduction

To understand the difference in CO₂ behavior between planetary boundary layer (PBL) and free troposphere (FT) over Siberian taiga, we have conducted CO₂ measurements using a small aircraft and a tower at the taiga in West Siberia since 2002. Continuous CO₂ time series at 4 levels (5, 20, 40, and 80 m) were monitored with the tower. Up to 3 km vertical CO₂ 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 Berezhovka village (56°09N, 84°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 CO₂ 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 CO₂ 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

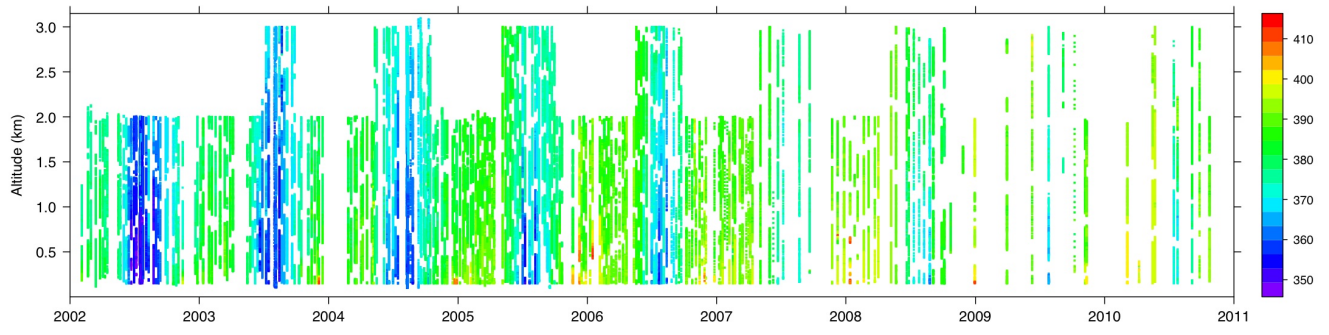
Figure shows the vertical profiles of CO₂ concentration observed by routine flights (261 times) from 2002 to 2010. Carbon dioxide concentrations showed slightly higher in the lower altitude during the dormant season. On the other hand, CO₂ concentration during summer gradually decreased with decrease 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 CO₂ 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 CO₂ 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 CO₂ 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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- Sasakawa et al. (2010a), *Tellus*, 62B, 403-416.
- Sasakawa et al. (2010b), *Atmos. Chem. Phys. Discuss.*, 10, 27759-27776.



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

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AAS021-P07

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

Chika Minejima^{1*}, Yasunori Tohjima¹, Dmitry Belikov¹, Shamil Maksyutov¹

¹NIES

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 O_2 , atmospheric potential oxygen (APO), seasonal cycle, global atmospheric transport model

AAS021-P08

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

Kentaro Ishijima^{1*}, Prabir Patra¹, Toshinobu Machida², Hidekazu Matsueda³, Yousuke Sawa³, Taku Umezawa⁴, Shuji Aoki⁴, Takakiyo Nakazawa⁴

¹JAMSTEC, ²NIES, ³MRI, ⁴Tohoku University

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

Hideo Sagawa^{1*}, YASUKO KASAI¹, Philippe Baron¹, Satoshi Ochiai¹, Jana Mendrok², UT/LS humidity & ice-cloud research team³

¹NICT, ²Lulea University of Technology, Sweden, ³NICT, Lulea Univ., Chalmers Univ., JPL

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

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

Hisashi Yashiro^{1*}, Kengo Sudo², Masayuki Takigawa¹

¹JAMSTEC/RIGC/EBCRP, ²Graduate School of Environmental Studies

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 Langdrif 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 ($O_3 \rightarrow O_2 + O(^1D)$, $O_3 \rightarrow O_2 + O$) 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-dependent variation of quantum yield, and 3) updated ACS of formaldehyde (HCHO) increased the photodissociation rate of both of channels ($HCHO \rightarrow CO + 2HO_2$, $HCHO \rightarrow CO + H_2$), 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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AAS021-P11

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

Katsuyuki Noguchi^{1*}, Itsushi Uno², Toshimasa Ohara³, Andreas Richter⁴, John P. Burrows⁴

¹University of Bremen/Nara Women's Univ., ²Kyushu University, ³NIES, ⁴University of Bremen

We conducted a case study on the transport of tropospheric nitrogen dioxide (NO₂) from China to the East China Sea and Japan by using satellite measurements. In the event, high concentrations of NO₂ ($\sim 1 \times 10^{16} \text{ cm}^{-2}$) were found off the coast of Shanghai and Kyushu. A trajectory analysis suggests that the air parcels with the high concentration of NO₂ found around Kyushu passed over Shanghai about 24 hours before, indicating that the source of the observed NO₂ 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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AAS021-P12

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Development of DOAS O₃ radiometer using D-UV LED

Kazuyuki Kita^{1*}

¹Faculty of Science, Ibaraki University

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

AAS021-P13

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

Sho Ohata^{1*}, Nobuhiro Moteki¹, Takuma Miyakawa¹, Yutaka Kondo¹

¹RCAST, The University of Tokyo

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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AAS021-P14

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

Masahiro Yano^{1*}, Kazuyuki Kita², Naga Oshima³, Nobuhiro Moteki⁴, Nobuyuki Takegawa⁴, Makoto Koike⁵, Yutaka Kondo⁴

¹Graduate School, Ibaraki University, ²Faculty of Science, Ibaraki University, ³Meteorological Research Institute, ⁴Research Center, University of Tokyo, ⁵Graduate School, The University of Tokyo

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: Black Carbon aerosol, CO

AAS021-P15

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

Xiaole Pan¹, Yugo Kanaya¹, Zifa Wang², Hiroshi Tanimoto³, Fumikazu Taketani^{1*}, Hajime Akimoto⁴, Yu Liu², Huabin Dong², Jie Li², Baozhu Ge², Xiquan Wang²

¹JAMSTEC, ²IAP, ³NIES, ⁴ACAP

Strong emissions of pollutants (e.g., BC and CO) in East China due to ever-quicken 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 $\Delta BC/\Delta CO$ 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 $\Delta BC/\Delta CO$ 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, Emission Inventory, Back trajectory

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

Masayuki Takigawa^{1*}, Kohei Ikeda²

¹RIGC/JAMSTEC, ²AORI/Univ. of Tokyo

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

Yuuki Goto¹, Hiroyuki Suzuki¹, Satomi Kagamitani¹, Tomoki Nakayama^{1*}, Yutaka Matsumi¹

¹STEL, Nagoya University

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

Motonori Okumura^{1*}, Junpei Kawamoto¹, Susumu Tohno¹, Yuji Kominami², Takafumi Miyama²

¹Kyoto University, ²FFPRI

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

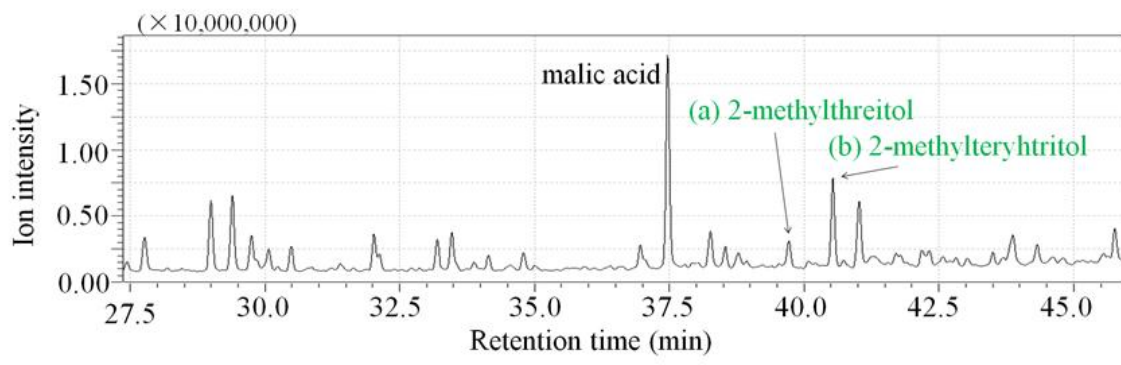
Atmospheric aerosols were collected at a tower above a forest canopy in southern Kyoto, Japan, between 2009 and 2010. YMS is in a mixed temperate forest with *Quercus serrata* (Japanese oak); one of the major tree species in Japan. The oak is a strong isoprene emitter (Okumura et al., 2008). A high-volume 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)trifluoroacetic anhydride (BSTFA, with 1% TMCS) and pyridine. Samples were heated for 2h at 70°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 carboxylic 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.

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Okumura, M. et al., (2008). *J. Agric. Meteorol.*, 64, 49-60.



AAS021-P19

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

SATOSHI IREI^{1*}, Akinori Takami¹, Toshihide Hikida², Akio Shimono²

¹NIES, ²Shoreline Science

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

Kei Sato^{1*}, Yuji Fujitani¹, Takamasa Seta¹, Takashi Imamura¹, Akinori Takami¹

¹NIES

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 NO_x 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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K. Sato, S. Hatakeyama, and T. Imamura, Secondary organic aerosol formation during the photooxidation of toluene: NO_x dependence of chemical composition, *J. Phys. Chem. A*, 39, 9796-9808 (2007).

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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 reactions in atmosphere.

Shohei Hattori^{1*}, Sebastian Danielache¹, Matthew S. Johnson², Henrik G. Kjaergaard², Yuichiro Ueno³, Naohiro Yoshida¹

¹Tokyo Institute of Technology, ²University of Copenhagen, ³Tokyo Institute of Technology

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(³P) 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 OC³²S, OC³³S, and OC³⁴S were measured using labeled samples prepared in the laboratory. The observed cross section of OC³²S 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 OC³³S 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(³P) 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

Sebastian Danielache^{1*}

¹Tokyo Institute of Technology

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 isotopologues. 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