Comparison of satellite observation of lower tropospheric ozone with model simulation over East Asia

*Sachiko Hayashida[1], Makoto Deushi[2], Mizuo Kajino[2], Kazuyo Yamaji[3], Thomas Sekiyama[2], Takashi Maki[2]

1.Faculty of Science, Nara Women's University, 2.Meteorological Research Institute, 3.Graduate School of Maritime Sciences, Kobe University

The lower tropospheric ozone enhancement over Central and Eastern China (CEC) was reported by Hayashida et al. (2015) by using the Ozone Monitoring Instrument (OMI) multiple-layer product retrieved by Liu et al. (2010). However, to clarify the enhancement in the concentration of the lowermost ozone using spaceborne measurements, it is necessary to understand the effect of ozone variation in the upper troposphere and lower stratosphere (UT/LS) because of large smoothing errors in the retrieval scheme. In this study, a scheme was developed to eliminate the artificial effect of UT/LS ozone enhancement on lower tropospheric ozone retrieval using OMI. By applying the UT/LS screening scheme for June 2006, we removed the effect of the UT/LS ozone enhancement on the artificial effect on the lower tropospheric ozone. Even after UT/LS screening, we were able to show a clear enhancement in the lower tropospheric ozone over CEC in June 2006 and confirmed the conclusion derived by Hayashida et al. (2015). After screening the UT/LS effect, we compared satellite measurements with model simulations for O_3 by using MRI-CCM2 (Deushi and Shibata, 2011). The observed O_3 enhancement over CEC in June 2006 was reproduced very well by the model simulations. The effects of emissions from the open crop residue burning (OCRB) in the North China Plain on lower tropospheric ozone were also examined by utilizing the emission inventory developled by K. Yamaji (Yamaji et al., 2010). In the scale of the vertical resolution of OMI observation, the difference between the O_3 with and without the OCRB effect was not very large (about 1 DU).

Acknowledgements
This study is a collaborative work with Dr. Xiong Liu, Harvard University. S. Hayashida was supported by a Grant-in-Aid from the Green Network of Excellence, Environmental Information (GRENE-ei) program, MEXT, Japan. We express our gratitude to Ms. Satoko Kayaba and Dr. Akiko Ono for their help with the data analysis.

References

Deushi M, Shibata K (2011): PMG, 62:1-46. doi:10.2467/mripapers.62.1
Hayashida S, et al. (2015): ACP, 15:9865-9881. doi:10.5194/acp-15-9865-2015
Liu X, et al.(2010): ACP, 10 (5):2521-2537. doi:10.5194/acp-10-2521-2010
Yamaji K et al. (2010): ACP, 10 (15):7353-7368.doi:10.5194/acp-10-7353-2010

Keywords: ozone, satellite, model simulation, pollution

©2016. Japan Geoscience Union. All Right Reserved.

Estimation and trend analysis of the tropospheric baseline ozone and carbon monoxide concentrations at Mt. Happo

*Sachiko Okamoto[1], Hiroshi Tanimoto[1], Hideki Nara[1]

1.National Institute for Environmental Studies

A large increase in tropospheric O_3 concentrations was observed during spring for the period from 1998 to 2006 at Mt. Happo Observatory (36.7°N, 137.8°E, 1840 m asl), which is one of the Acid Deposition Monitoring Network in East Asia (EANET) stations (Tanimoto, 2009). The increase in the springtime O_3 reproduced by a regional chemistry-transport model incorporating the updated anthropogenic emissions inventory in East Asia can only explain about half of the observed O_3 increase (Tanimoto et al., 2009). Observational datasets at mountain sites have been utilized for estimation of baseline levels of greenhouse gases and aerosols because remote mountainous sites are regarded as having little direct influence from local or regional sources/sinks (e.g., Parrish et al., 2012). However, observations at ground-based stations can often be influenced by local sources. Therefore, the data selection is often an essential part of the analysis for estimation of regional representative baseline levels.
In this study, we estimated baseline and polluted concentrations of O_3 (1998–2014) and CO (1996–2014 and 2013–2014) at Mt. Happo by using the statistical method “REBS (Robust Extraction of Baseline Signal)” (Ruckstul et al., 2012), which is based on the robust local regression. Then, we analyzed these long-term trends. CO concentration (2013–2014) is significantly lower than CO concentration (1996–2004), and the degree is larger in summer and autumn than in spring. Recent baseline CO concentrations are decreasing in all season except for spring. Spring baseline concentration is slightly increasing. Polluted CO concentrations are decreasing in all season, and the degrees are larger than those of baseline concentration. On the other hand, O_3 concentrations show decreasing trends with maximum in the mid-2000s in all season. The rate of decrease is characterized by a spring maximum and summer minimum. Both baseline and polluted O_3 concentrations show decreasing trends in all season and both rates of decrease are the same degree. Hence it is considered that the decrease of springtime CO concentration at Mt. Happo is mainly caused by the decrease of polluted CO concentration, and that the decrease of O_3 concentration is caused by the decreases of baseline and polluted O_3 concentrations.

Keywords: Ozone, Carbon monoxide, Baseline level

©2016. Japan Geoscience Union. All Right Reserved.

Long-term observations of black carbon at Fukue Island during 2009–2015: Rates of emissions from East Asia and removal

*Yugo Kanaya[1,2], Xiaole Pan[3], Takuma Miyakawa[1,2], Yuichi Komazaki[1], Fumikazu Taketani[1,2], Itsushi Uno[3], Yutaka Kondo[4]

1.Department of Environmental Geochemical Cycle Research, Japan Agency for Marine-Earth Science and Technology, 2.Institute of Arctic Climate and Environment Research, Japan Agency for Marine-Earth Science and Technology, 3.Kyushu University, 4.National Institute of Polar Research

Black carbon is a component of atmospheric fine aerosol particles contributing to global warming. However, its emission strengths and removal rates have not been sufficiently understood. We have conducted long-term observations of black carbon mass concentrations using a COSMOS instrument since 2009 at Fukue Island (32.75°N, 128.68°E), western Japan, to provide information on the emission strengths of important source regions in East Asia and on wet removal rate constraints. The annual average mass concentration was 0.36 μg m^{-3}, with distinct seasonality; high concentrations were recorded during autumn, winter, and spring, and were caused by Asian continental outflows, which reached Fukue Island. Statistical analysis of the observed ΔBC/ΔCO ratio was separately made for two classes of data with and without a wet removal effect, using the accumulated precipitation along a backward trajectory (APT) for the last 3 days as an index. The emission ratios estimated from observations with zero APT (5.2–6.9 ng m^{-3} ppb^{-1}) varied over the six air mass origin areas; the higher ratios for South Central East China and South China indicated the relative importance of domestic emissions and/or biomass burning sectors. The BC/CO emission ratios adopted in the bottom-up Regional Emission inventory in Asia (REAS) version 2 (6.5–23 ng m^{-3} ppb^{-1}) over the continent were significantly higher; the ratios needed to be reduced by 60% for China and by a factor of 3.5 for Korea, although the ratio for Japan was in an acceptable range. The wintertime enhancement of the BC emission from China, predicted by REAS2, was verified for air masses from South Central East China. Wet removal of BC was clearly identified as a decrease in the ΔBC/ΔCO ratio against APT. The transmission efficiency (TE), defined as the ratio of the ΔBC/ΔCO ratio with precipitation to that without precipitation, was fitted reasonably well by a stretched exponential decay curve against APT. The dependence on APT was almost similar among the air mass types. An accumulated precipitation of 15 mm halved the BC mass concentration. This expression of wet removal and the emission constraint for East Asia help to test and improve chemical transport and/or climate model simulations.

Keywords: aerosol particles, trans-boundary air pollution, climate Effect, budget analysis, process analysis

©2016. Japan Geoscience Union. All Right Reserved.

Budget analysis of aerosols in China: interannual variation in aerosol concentration and outflow

*Tsuyoshi Yamashita[1], Kengo Sudo[1]

1.Graduate School of Environmental Studies, Nagoya University

Aerosols and their precursor emitted from polluted regions strongly influence climate and atmospheric environment not only at local areas but also in remote areas due to long-range transport. Therefore, it is needed to investigate temporal variations of aerosol concentration in source region and associated ouflow from the region in order to evaluate impacts of air pollutants on climate and atmospheric environment. In this study, we evaluate main factors of changes in aerosol concentrations in the China region (18°N-46°N, 104°E-123°E) and aerosol outflow there focusing on black carbon (BC) and sulfur oxides (SOx) by budget analysis of aerosols using a global chemical climate model CHASER. As a consequence, we found that aerosol outflow from the China region has a seasonal peak in winter and the outflow across the east boundary in the region and the inflow across the west boundary both have peaks on March. Our analysis also shows that variations of net zonal outflow largely contribute to the interannual variability of outflow from the China region. We also found that aerosol outflow largely controls the interannual variation of aerosol concentration in the China region (deposition in the region makes only a small contribution to it). Additionally, our sensitivity experiments with BC emission which has a positive radiative forcing and climate impacts, indicate that the domestic sources in the China region and long-range transport from India account for 75% and 12% of the tropospheric BC burden in the China region, respectively. About a half of the BC inflow across the west boundary in the China region is contributed from BC emitted from India. BC emitted from the China region and from India contribute by about 60% and 20% respectively to the total BC outflow across the east boundary. This study, therefore, suggests that BC transported from South Asia such as India influence variabilities of BC concentration in China, and this BC can be farther transported to Japan and the North Pacific regions.

Keywords: aerosol, long-range transport, black carbon, China

©2016. Japan Geoscience Union. All Right Reserved.

Return to the decade-ago level of tropospheric nitrogen dioxide pollution in East Asia

*Hitoshi Irie[1], Takuya Muto[1], Syuichi Itahashi[2], Jun-ichi Kurokawa[3], Itsushi Uno[4]

1.Chiba University, 2.Central Research Institute of Electric Power Industry, 3.Asia Center for Air Pollution Research, 4.Kyushu University

Long-term (2005-2015) tropospheric nitrogen dioxide (NO_2) column data recorded by the satellite-borne Ozone Monitoring Instrument (OMI) in East Asia were analyzed to investigate annual trends quantitatively and their potential causes. We found an evident decrease in the NO2 level over China after 2011 and then a return to the 2005 level in 2015. The grid-basis trend analysis implies that the rapid decrease occurred on a provincial or larger spatial scale and was likely due to a nationwide action such as the widespread use of denitrification units. Other prominent features were seen in Japan. Despite a significant substitution from nuclear to thermal power after 2011 as a consequence of a massive earthquake off the Pacific coast of northern Japan, the NO_2 level continued to decrease for both periods (2005-2011 and 2011-2015). The decrease contributed to a return to the decade-ago level of tropospheric NO_2 pollution in East Asia.

Keywords: NO2, trend, East Asia, OMI

©2016. Japan Geoscience Union. All Right Reserved.

Rabi-crop CO_2 uptake inferred from CONTRAIL measurements over Delhi, India

*Taku Umezawa[1], Yosuke Niwa[2], Yousuke Sawa[2], Toshinobu Machida[1], Hidekazu Matsueda[2]

1.National Institute for Environmental Studies, 2.Meteorological Research Institute

Recent studies propose that growing agriculture has altered atmospheric CO_2 variations and the global carbon cycle. In this study, we show a clear evidence of significant impact of Indian wintertime (*rabi*) agriculture (mainly wheat) on the regional carbon budget based on high-frequency atmospheric CO_2 measurements onboard commercial airliners over Delhi, India. While a general increasing gradient toward the ground was observed throughout December–April, we have frequently observed sharp decreases near the ground during January–March. In this period, CO_2 concentration at altitudes below 2 km was at seasonal stagnation. Meteorology in the season infers influence from neighboring croplands with patchy urban areas located upwind. We conclude that the observed CO_2 decrease is attributable to active uptake by *rabi*-crop growing in the season and that the uptake is comparable in magnitude to urban CO_2 emissions from the Delhi metropolitan area.

Keywords: CO2, rabi crop, aircraft measurements

©2016. Japan Geoscience Union. All Right Reserved.

Temporal variations of the atmospheric CO_2 concentration and $d^{13}C$ at Ny-Ålesund, Svalbard

*Daisuke Goto[1], Shinji Morimoto[2], Shigeyuki Ishidoya[3], Shuji Aoki[2], Takakiyo Nakazawa[2], Akie Yuba[1]

1.National Institute of Polar Research (NIPR), 2.Tohoku University, 3.National Institute of Advanced Industrial Science and Technology

Long-term measurements of the atmospheric CO_2 concentration and its carbon isotope ratio ($d^{13}C$) have been used for partitioning CO_2 sinks into the terrestrial biosphere and the ocean. However, the CO_2 sinks estimated from $d^{13}C$ suffer with uncertainties in isotopic disequilibrium flux between the atmosphere and the ocean and between the atmosphere and the terrestrial biosphere (so-called isoflux). For a better understanding of the global carbon cycle, we have been carrying out the systematic observation of the atmospheric CO_2 concentration and $d^{13}C$ at Ny-Ålesund (78.93°N, 11.83° E), Svalbard since 1991 by weekly air sampling with subsequent analysis in NIPR. Here, we will present the observational results of CO_2 concentration and $d^{13}C$ for 1991–2013 and 1996–2013, respectively. The $d^{13}C$ data before 1996 were removed from our analysis due to experimental and sample quality problems (Morimoto et al., 2001).

The CO_2 concentrations show a clear seasonal cycle with peak-to-peak amplitude of about 17 ppmv, which reaches the maxima in late April to early May and the minima in late August, superimposed on a secular increase with an average rate of 2.0 ppmv/yr for the period of 1996–2013. On the other hand, the $d^{13}C$ decreases secularly at an average rate of −0.018 ‰/yr, and varies seasonally in opposite phase with the CO_2 concentration. We have also maintained atmospheric $d(O_2/N_2)$ measurements at Ny-Ålesund since 2001 (Ishidoya et al., 2012). Using the atmospheric $d(O_2/N_2)$ and CO_2 concentration records, the terrestrial and oceanic CO_2 sinks are estimated to be 1.7 ±0.8 GtC/yr and 2.2 ±0.7 GtC/yr, respectively, for the 13-year period (2001–2013). Using these values of CO_2 sinks and the $d^{13}C$ record, the average isofulx for the period of 2001–2013 is estimated to be 99 ± 28 Gt ‰/yr.

References

Ishidoya et al. (2012) Oceanic and terrestrial biospheric CO_2 uptake estimated from atmospheric potential oxygen observed at Ny-Ålesund, Svalbard and Syowa, Antarctica. *Tellus B*, 64, 18924, http://dx.doi.org/10.3402/tellusb.v64i0.18924.

Morimoto et al. (2001) Temporal variations of atmospheric CO_2 concentration and carbon isotope ratio in Ny-Alesund, Svalbard, *Mem. Natl Inst. Polar Res.*, Spec. Issue, 54, 71–79.

Keywords: atmospheric CO2, carbon isotope ratio, O2/N2 ratio

©2016. Japan Geoscience Union. All Right Reserved.

Diffusive separation of the lower atmosphere suggested by Ar/N_2, delta^{15}N of N_2, delta^{18}O of O_2 observed at Ny-Ålesund, Svalbard.

*Shigeyuki Ishidoya[1], Daisuke Goto[2], Satoshi Sugawara[3], Shinji Morimoto[4], Shuji Aoki[4], Shohei Murayama[1], Takakiyo Nakazawa[4]

1.National Institute of Advanced Industrial Science and Technology (AIST), 2.National Institute of Polar Research, 3.Miyagi University of Education, 4.Tohoku University

Recent technical advances has made it possible to observe a molecular diffusive separation of the atmosphere based on high precision measurements of the composition of atmospheric major components. In the middle to lower stratosphere, Ishidoya et al. (2013) reported the existence of observable gravitational separation based on the measurements of stratospheric air samples collected using a balloon-borne cryogenic air sampler. In the lower atmosphere, Adachi et al. (2006) reported the diffusive separation of Ar and N_2, mainly due to thermal diffusion, in the center of a wide desert during the nighttime when vertical temperature inversions are generated. To examine whether the diffusive separation of the atmosphere is also detectable near the surface in polar region, air samples collected at Ny-Ålesund, Svalbard (79°N, 12°E) have been analyzed for delta(Ar/N_2), delta(O_2/N_2), delta^{15}N of N_2, delta^{18}O of O_2 and delta^{40}Ar by using a mass spectrometer (Ishidoya and Murayama, 2014) since January 2013. It was found that delta^{15}N and delta^{18}O show small but significant seasonal cycles, with the seasonal maxima and minima in winter and summer, respectively. The peak-to-peak amplitudes of the respective seasonal cycles of delta^{15}N and delta^{18}O were about 2 and 4 per meg. On the other hand, no significant seasonal cycle was seen in delta(Ar/N_2). If we assume the seasonal cycles of delta^{15}N and delta^{18}O are attributed mainly to gravitational separation in a temperature inversion layer during polar night in winter and corrected the delta(Ar/N_2) for the separation by subtracting 12 x (delta^{15}N + delta$^{18}O/2)/2$ (delta$(Ar/N_2)_{cor}$), then the delta$(Ar/N_2)_{cor}$ show clear seasonal cycle with a maximum in August. The peak-to-peak amplitude of the seasonal delta$(Ar/N_2)_{cor}$ cycle is about 25 per meg, and the appearance time of seasonal maximum agrees with that of the sea surface temperature around Ny-Ålesund. These results suggest that gravitational separation is observable near the surface at Ny-Ålesund. Our suggestion would be supported by Keeling et al. (2004) who reported the delta(Ar/N_2) observed in the polar region may be detectably enriched near the ground by gravitational separation or thermal diffusion under condition of strong surface inversions.

References

Adachi, Y. et al. (2006) Diffusive separation of the lower atmosphere, *Science*, 311, 1429.

Ishidoya, S. et al. (2013) Gravitational separation in the stratosphere –a new indicator of atmospheric circulation. *Atmos. Chem. Phys.*, 13, 8787–8796, www.atmos-chem-phys.net/13/8787/2013/, doi:10.5194/acp-13-8787-2013.

Ishidoya, S. & Murayama, S. (2014) Development of high precision continuous measuring system of the atmospheric O_2/N_2 and Ar/N_2 ratios and its application to the observation in Tsukuba, Japan. *Tellus B*, 66, 22574, http://dx.doi.org/ 10.3402/tellusb.v66.22574.

Keeling, R. F. et al. (2004) Measurement of changes in atmospheric Ar/N2 ratio using a rapid-switching, single-capillary mass spectrometer system, *Tellus B*, 56, 322–338.

Keywords: diffusive separation of the atmosphere, Ar/N2 ratio, delta15N of N2, delta18O of O2, vertical temperature inversions

©2016. Japan Geoscience Union. All Right Reserved.

©2016. Japan Geoscience Union. All Right Reserved.

Seasonal cycle of aerosol size distribution at Syowa Station, Antarctica

*Keiichiro Hara[1], Chiharu Nishita-Hara[2], Kazuo Osada[3], Masanori Yabuki[4], Takashi Yamanouchi[5]

1.Fukuoka University, 2.Fukuoka University, FIT, 3.Nagoya Univ., GSES, 4.Kyoto Univ., RISH, 5.NIPR

Atmospheric aerosols are related closely to the climate change through direct and indirect effects. Number concentrations and size distributions of aerosols are one of the most important properties. Recently, aerosol size distributions have been measured even in the Antarctic regions during summer (e.g., Virkkula et al., 2007; Asmi et al., 2010; Pant et al., 2011; Park et al., 2004). These studies focused on the respect of new particle formation. However, only a few wintering-measurements of aerosol size distributions in fine - ultrafine modes have been made in the Antarctic regions (Ito, 1993; Hansen et al., 2009; Järvinen et al., 2013). This study aims to understand seasonal cycle of aerosol size distribution and new particle formation at Syowa Station, Antarctica. For measurement of size distribution in ultrafine - fine modes (Dp: 5–168 nm), a scanning mobility particle sizer (SMPS: 3936-N-25; TSI Inc.) was used from February 2004 - December 2006 at Syowa Station, Antarctica. The scanning time for one scan was set to 5 min in SMPS measurement. Local contaminated data were filtered using CN data (aerosol monitoring data) and wind data (observed by JMA). Log-normal fitting was used to compare modal structure of aerosol size distributions in this study. All daily-mean aerosol size distributions were fitted using the following lognormal modes: fresh nucleation mode (Dp<10 nm), aged nucleation mode (Dp=10-25 nm), 1st Aitken mode (Dp=25-50 nm), 2nd Aitken mode (Dp=50-100 nm), and accumulation mode (Dp>100 nm). Aerosol size distribution showed 2 –4 modal structures from early September –end-March. Fresh nucleation mode appeared occasionally in March-April, and August-November, and rarely in December-February and May-July. The number concentrations in aged nucleation –2nd Aitken modes increased in summer. In contrast, most of size distribution showed 1 - 2 modal structures during April - August. Strong mono-modal distribution appeared in 2nd Aitken and/or accumulation modes under/after the storm conditions in the winter –early spring. This seasonal feature might be associated with seasonal cycles of (1) the concentrations of condensable vapors linked to photochemical reactions and biogenic activity, and (2) sea-salt particles released from sea-ice surface in the winter - spring. Here, we discuss and characterize seasonal cycle of aerosol size distribution in ultrafine –fine modes in the Antarctic coasts.

Keywords: aerosols, size distribution, Antarctica

©2016. Japan Geoscience Union. All Right Reserved.

Seasonal variation of the concentration of black carbon and the size distribution of the surface snow sampled in the Syowa station in the Antarctica

*Takeshi Kinase[1], Kazuyuki Kita[2], Kumiko Goto-Azuma[3,4], Yoshimi Ogawa[3], Yutaka Kondo[3,5], Moteki Nobuhiro[5], Sho Ohata[5], Tatsuhiro Mori[5], Masahiko Hayashi[6], Keiichiro Hara[6], Masataka Shiobara[3,4], hiroto kawashima[7]

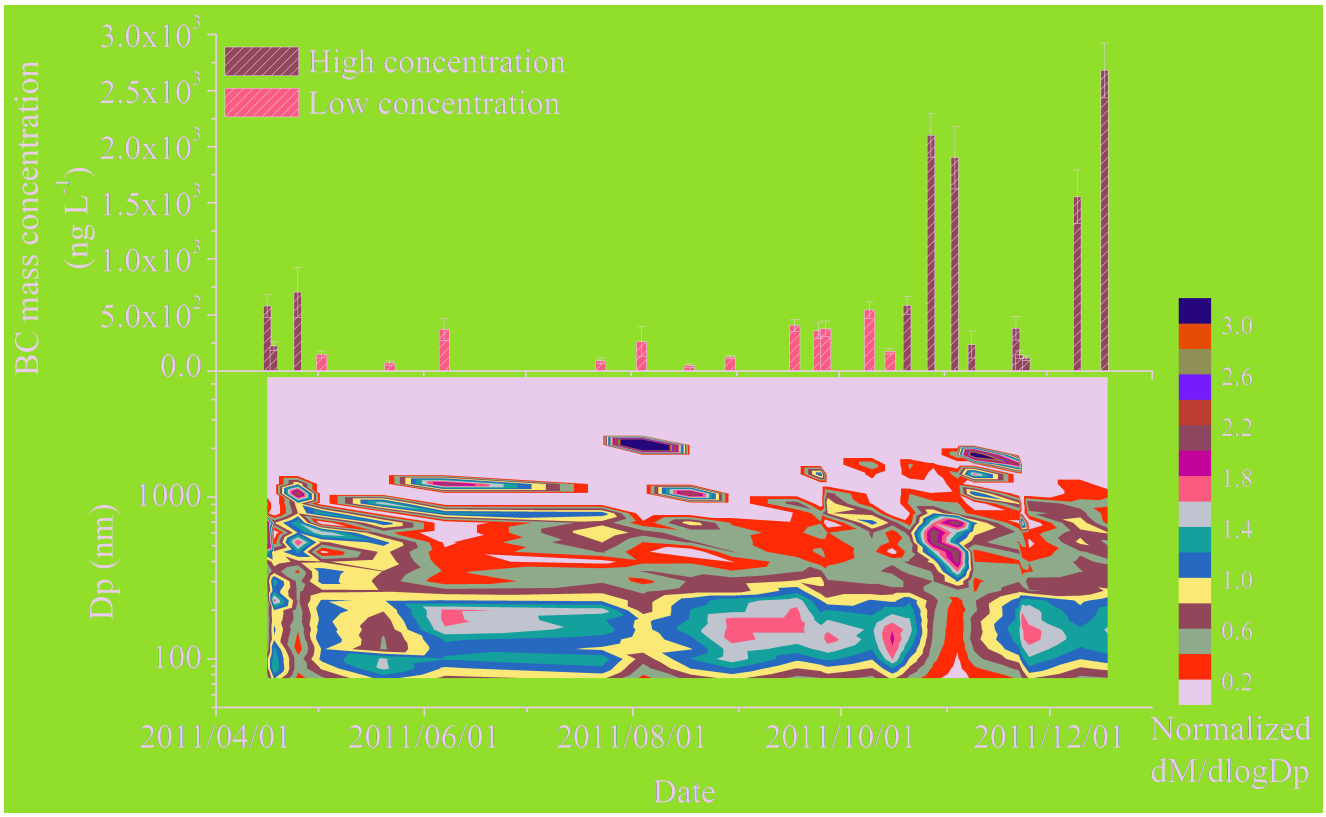
1.Graduate School of Science and Technology, Ibaraki University, 2.College of Science, Ibaraki University, 3.National Institute of Polar Research, 4.SOKENDAI(The Graduate University of Advanced Studies), 5.Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo, 6.Department of Earth System Science, Faculty of Science, Fukuoka University, 7.Department of Management Science and Engineering, Faculty of Systems Science & Technology, Akita Prefectural University

Black carbon (BC) strongly absorbs the radiation, leading to large climate effects. BC deposited on the snow/ice packs also leads to positive radiative forcing, reduces the snow/ice albedo. The snow albedo is estimated considering snow grain size and impurities, to clarify the size distribution of BC particles in snow is important. But the technique for measurement of the size distribution of BC in snow is developed in recent, the size distribution of BC is not understood sufficiently, especially Antarctica.
In this study, 29 snow samples were obtained at Antarctica, Syowa station, with the Japanese Antarctic Research Expedition 52 (JARE52, 2010-2012), measured for the BC concentration and its size distribution in snow and discussed. The BC concentration and its size distribution are measured by the Single Particle Soot Photometer (SP2) with an improved technique for measuring the size distribution wider (Mori et al., 2016), and ions and pH are measured at the each samples. The atmospheric BC concentration was measured by an Aethalometer during sampling period.
Snow samples were scooped from the surface of the snow to 250cc glass bottles directly, had been kept below -20 °C until the measurement. October 2011, relatively continent snows were sampled on the traverse route for the Mizuho station by JARE52. Based on reports of Kinase et al. (JpGU2015) and Kinase et al. (2016), snow samples were distributed to 3 of 20 cc glass bottles using a ceramic knife, melted in refrigerator, sonicated 15 minutes, and mixed to an 1 bottle. 30cc LDPE bottles were obtained for measurements of ions and the pH.
From results, the averaged BC concentration in snow samples were 591.6 ±714.1 (ng L^{-1}), and we found the seasonal variation, low in winter (May to September) and high in other seasons. Mizuho route samples were higher than Syowa. These results were little higher than previous studies which were obtained in other period, other locations and other method, but agreed on an order. Also we could find the seasonal variation of the size distribution, small particles were mainly in winter, but large particles were found in summer. But this measurement had not done in previous, we could not compare with other studies. Concentrations of ions and pH had no same variations.
For the atmospheric BC concentration, the sample air was heated to 300 ºC for decreasing effects by the volatile particles, but the effect of non-volatile particles remained. Hence, the measurement of the atmospheric BC had a large uncertain, it was high during winter and low during summer. Hara et al. (2008) showed that there are two processes of transporting atmospheric BC around Syowa, by the blizzard during winter and by the katabatic wind during summer. This result indicates that the dominant transporting/deposition process of atmospheric BC would change in each season.
Therefore, the atmospheric BC had the opposite trend to BC in snow, the atmospheric BC concentration would not effect to the BC in snow directly. The seasonal variation of BC in snow would depend on other process, such as the deposition process and others.

©2016. Japan Geoscience Union. All Right Reserved.

Moreover, the radiation had the same trend to the concentration and the size distribution of BC in snow, the radiation would effect for the seasonal variation of BC in snow. Heating and melt/refreeze cycle would change the size distribution (Kinase et al., (JpGU2015) ; Kinase et al., (2016)). The radiation would lead the heating of ice nuclei and surface snow, this indicates the seasonal variation of the concentration and the size distribution of BC would be related to the seasonal variation of the radiation. More studies of BC in snow are needed in the future, such as a long-term monitoring, the variation of the water cycle and deposition efficiency of BC to the snow, and the heating effect for the snow and ice nuclei.

Keywords: Antarctica, Syowa station, snow, Black carbon, size distribution, seasonal variation



©2016. Japan Geoscience Union. All Right Reserved.

Volatility measurements of SOA formed from α-pinene ozonolysis and data interpretation

*Kei Sato[1], Yuji Fujitani[1], Satoshi Inomata[1], Yu Morino[1], Kiyoshi Tanabe[1], Akinori Takami[1]

1.National Institute for Environmental Studies

The volatility basis-set (VBS) model employs the secondary organic aerosol (SOA) volatilities evaluated from yield curve measurements. To verify the VBS model, we evaluated the volatilities of dry α-pinene ozonolysis SOA not only from yield curve measurements but also from thermodenuder-AMS measurements, LC-MS and PTR-MS chemical composition analysis, and external dilution chamber measurements. We evaluated also the uncertainty of volatility determined by each experimental metod. The results of thermodenuder-AMS measurements, chemical composition analysis, and external dilution chamber measurements showed that lower-volatility organic compounds are present in SOA particles compared with the results of yield curve measurements. The thermal and dilution properties of oligomers, produced in the particle phase by heterogenious reactions, will strongly affect the total properties of SOA particles under dry conditions.

Keywords: Secondary organic aerosol, Heterogeneous reaction, Oligomer formation

©2016. Japan Geoscience Union. All Right Reserved.

Experimental study on the heterogeneous reaction of gaseous OH radical with aqueous DMSO: Determination of the $CH_3SO_3^-$/ SO_4^{2-} production ratio

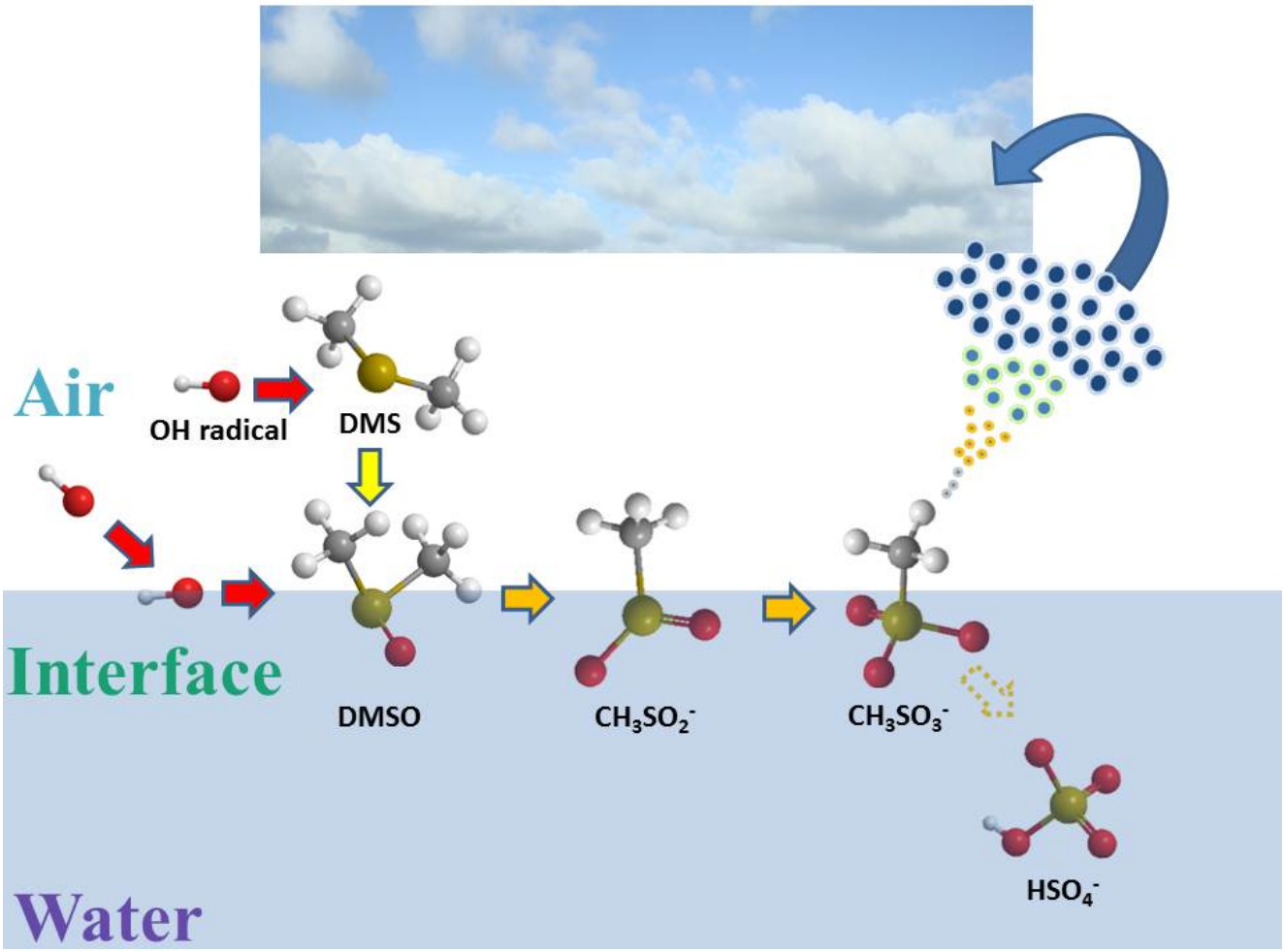
*Shinichi Enami[1]

1.The Hakubi Center, Kyoto university

The oxidation of dimethyl sulfide (DMS) emitted from ocean (~ 45 Tg S per year) is a global source of cloud condensation nuclei. Hydrophobic DMS is mostly oxidized in the gas-phase into H_2SO_4(g) + DMSO(g) (dimethyl sulfoxide), whereas water-soluble DMSO is oxidized into SO_4^{2-} + $CH_3SO_3^-$ (methane sulfonate) on water surfaces. R = $CH_3SO_3^-/SO_4^{2-}$ ratios therefore indicate the extent of DMSO heterogeneous oxidation if R_{het} = $CH_3SO_3^-/SO_4^{2-}$ for DMSO(aq) + ·OH(g) were determined. Here, products and intermediates of the oxidation of aqueous DMSO initiated by gas-phase hydroxyl radicals, OH(g), at the air-water interface were directly detected by mass spectrometry in a novel setup under various experimental conditions. Exposure of millimolar DMSO aqueous microjets to ~ 10 ns OH(g) pulses from the 266 nm laser flash photolysis of O_3(g)/O_2(g)/H_2O(g)/N_2(g) mixtures yielded an array of interfacial intermediates/products, including $CH_3SO_3^-$ and HSO_4^-, that were unambiguously and simultaneously identified in situ by mass spectrometry. We determined R_{het} = 2.7 from the heterogeneous OH-oxidation of DMSO on aqueous aerosols for the first time. The nearly quantitative production of H_2SO_4(g) (that leads to SO_4^{2-}) in the oxidation of DMSO in the gas-phase versus the R_{het} ~ 2.7 value determined at the air-water interface means that R = $CH_3SO_3^-$/nss-SO_4^{2-} variations in the aerosol, particularly in remote locations, should arise from the competition between the gas-phase versus the heterogeneous DMSO oxidation pathways. The present study reveals that interfacial OH-oxidation processes play a more significant role in the generation and growth of atmospheric aerosol over ocean than previously envisioned.

Keywords: Aerosol, Photochemical aging, Climate change, heterogeneous reaction, marine chemistry

©2016. Japan Geoscience Union. All Right Reserved.



©2016. Japan Geoscience Union. All Right Reserved.

Development of a compact, simple and precise PM2.5 sensor and its applications

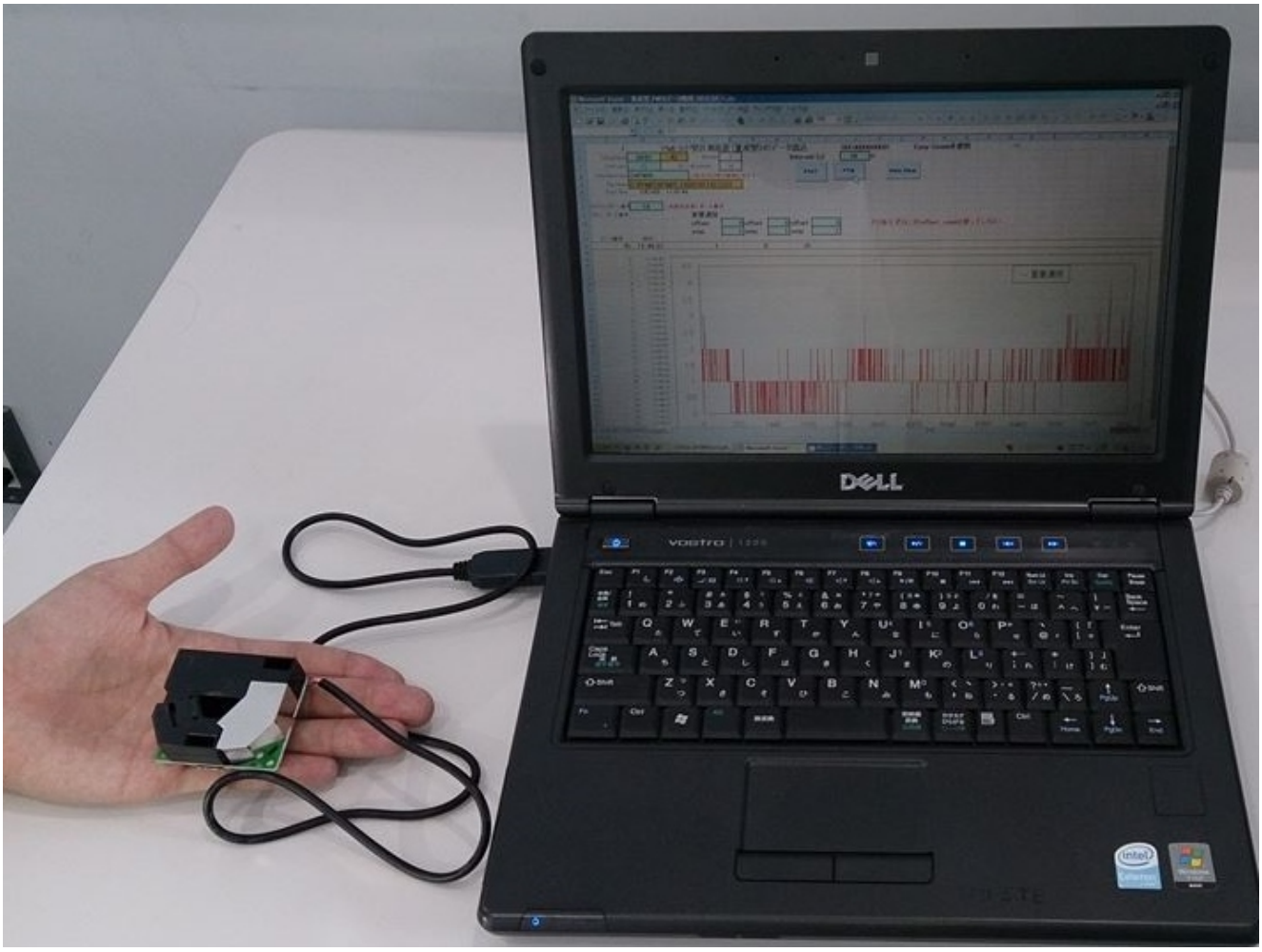
*Yutaka Matsumi[1], Tomoki Nakayama[1]

1.Institute for Space-Earth Environmental Research, Nagoya University

PM2.5 is the concentration of small particles floating in the air with the diameter size of less than 2.5 micrometer. The nation-wide interests about PM2.5 in Japan have increased since the PM2.5 concentration in Beijing, China became extra-ordinary high in 2013-2015. The high concentrations of PM2.5 in China influence not only the environmental conditions in China but also those in Japan through the long-range transportation across the border. The regulation for the PM2.5 concentration in Japan is defined in units of mass concentration. The upper limit values of the regulation are 15 microgram per cubic meter for one-year average and 35 microgram per cubic meter. For instruments for PM2.5 measurements, beta attenuation monitoring (BAM) and filter-based gravimetric method (TEOM) have been used in public observational stations and in environmental research stations. However, those instruments are very expensive and need accumulation time of about half a day for PM2.5 measurements. We have developed a compact and low-cost PM2.5 instrument in collaboration with Panasonic Corporation.
The new compact, palmtop PM2.5 instrument consists of a LED light source and photodiode detecting aerosol particle light scattering. To increase the precision of measurements, the sizes of individual aerosol particles are estimated from the intensities of the scattering light intensity and the PM2.5 mass concentrations are calculated. The PM2.5 measurement results for ambient air using the compact PM2.5 instrument indicated high correlation factor of > 0.8 with the results obtained by simultaneous measurement using a BAM instrument (Thermo, Sharp 5030).
Many applications of the compact, low-cost and simple PM2.5 instrument have been developed. In urban area, many instruments can be installed with high densities. Local PM2.5 sources in the urban areas can be detected with the PM2.5 instruments. 2D and 3D measurements in the atmosphere can be measured by installing the PM2.5 instruments on automobiles, drones (multicopters). Especially, the new PM2.5 instruments are suitable for the measurements in developing countries. Some of developing countries suffer from serious environmental problems of extremely high PM2.5 concentrations and their health effects. The PM2.5 observations in the developing countries have difficulty to install valuable and delicate PM2.5 instruments because of many serious difficulties about space, electric supply, dust, temperature, roof leaks, insects, safety, transportation, maintenance access, standard-gas supply and so on. The new PM2.5 instruments can be installed and operated in those conditions. The PM2.5 instruments widely distributed in high PM2.5 concentration area are suitable for epidemiological studies.
In this presentation, we will present the features of the compact PM2.5 instrument, and also present the new applications such as measurements on vehicles and in the developing countries. We have provided the measurement system which consist of a palmtop sensor and a computer with the USB cable connection. We have also provided the outdoor stand-alone system with small CPU and USB memory for year-long measurements. We will accept proposals of new applications of our compact PM2.5 instrument, and discuss the possibilities of collaborative work for the applications.

Keywords: Compact PM2.5 sensor, Instrument development, Atmospheric aerosol

©2016. Japan Geoscience Union. All Right Reserved.



©2016. Japan Geoscience Union. All Right Reserved.

Tar ball particles from biomass burning smoke

*Kouji Adachi[1]

1.Meteorological Research Institute

Tar ball is a spherical organic particle emitted from biomass burning and is brown carbon that influences climate. Tar ball is abundant in ambient atmosphere and sometime dominates in aged biomass burning smoke. However, its effect on climate remains uncertain because both its composition and shape, which can be determined using microscopic technique, are necessary to identify tar ball. This study uses a transmission electron microscope with an energy-dispersive X-ray spectrometer (TEM-EDS) to analyze tar balls and aims to reveal its abundance in wild biomass-burning smoke and its micro-physical properties.
The samples were collected from wild fires in North America during Biomass Burning Observation Project (BBOP) 2013 aircraft campaign. BBOP campaign is an aircraft-based field campaign to study the near-field evolution of particulate emissions from biomass burning from July to October 2013. Aerosol particles from wildfires in the Western US (Idaho, Oregon, and Washington) and from agricultural burns in the Mississippi Embayment (Arkansas) were sampled. From these samples, number fractions of tar ball were measured from TEM images.
Tar balls primarily originated from wildfires and were lack in agricultural-burning smoke. They were abundant in relatively aged smoke (>several hours from emission), and the number fractions could reach more than half of all aerosol particles with aerodynamic diameter between 100 and 700 nm. Samples with relatively high tar ball fractions were focused, and the bulk optical and chemical compositions within the smoke with many tar balls will be discussed. Abundances and optical properties of tar balls shown in this study are useful to evaluate their effects on the global climate.

Keywords: aerosol, Transmission electron microscope, climate, tar ball, biomass burning

©2016. Japan Geoscience Union. All Right Reserved.

Observational study of wet removal process of black carbon particles

*Tatsuhiro Mori[1], Sho Ohata[1], Moteki Nobuhiro[1], Makoto Koike[1], Yutaka Kondo[2]

1.Graduate school, Tokyo University, 2.National Institute of Polar Research

Black carbon (BC) particles absorb visible solar radiation and heat the atmosphere. An improved understanding of wet removal process is important because it strongly influences temporal and spatial distribution of BC in atmosphere. There are two categories of removal mechanisms: nucleation scavenging and impaction scavenging. The former refers to the mechanisms that BC particles are incorporated into cloud droplets by serving as cloud condensation nuclei (CCN), while the latter refers to scavenging via impaction with cloud droplets or rain droplets. Theoretically, the efficiencies of these mechanisms depend on BC sizes. However, their relative contribution to the removal of BC has never tested by direct observation.
In this study, we observed relationships between size-dependent removal efficiency (RE) and size-dependent CCN activity in Tokyo during summer 2014 and 2015. The size-dependent RE was determined by measuring both size-resolved BC number concentrations in air and in rainwater. The size-dependent CCN activity was estimated by measuring coating thickness and hygroscopicity of BC particles.
Out of 32 rain events during observation period, the strong size-dependent RE was successfully explained by the size-dependent CCN activity for 29 rain events, indicating that nucleation scavenging was the dominant removal process of BC particles. For the other rain events, impaction scavenging might be also effective for larger BC particles.

Keywords: Black carbon, wet deposition

©2016. Japan Geoscience Union. All Right Reserved.

Aerosol single scattering albedo comparison between SKYNET and AERONET

*Pradeep Khatri[1], Hitoshi Irie[1], Tamio Takamura[1]

1.Center for Environmental Remote Sensing, Chiba University

SKYNET and AERONET are two aerosol observing networks in the world. Though aerosol optical thickness (AOT) between them are reported to agree fairly well, their aerosol single scattering albedo (SSA) values have some difference. To explore the reasons behind inconsistent SSA between SKYNET and AERONET, we analyzed collocated observation data of SKYNET and AERONET of four sites, Chiba (Japan), Pune (India), Valencia (Spain), and Seoul (Korea). SKYNET and AERONET algorithms are found to produce nearly same SSAs for similarity in input data, suggesting that SSA differences between them are primarily due to quality of input data due to different calibration and/or observation protocols as well as difference in quality assurance criteria. The most plausible reason for systematically overestimated SKYNET SSAs is found to be underestimated calibration constant for sky radiances determined from the disk scan method in SKYNET, though the disk scan method is noted to produce stable wavelength dependent values in comparison to those determined from the integrating sphere used by AERONET. Aerosol optical thickness (AOT) difference between them can be the next important factor for their SSA difference, if AOTs between them are not consistent. Difference in surface albedos between SKYNET and AERONET while analyzing data can also bring SSA difference between them, but the effect of surface albedo is secondary. The aerosol non-sphericity effect is found to be less important for SSA difference between these two networks.

Keywords: aerosol, single scattering albedo, SKYNET

©2016. Japan Geoscience Union. All Right Reserved.

Analysis of individual aerosol particles collected at the top of Mt. Fuji in 2014 and 2015

*Sena Doi[1], Yoko Iwamoto[1], Kazuhiko Miura[1]

1.Tokyo University of science

Atmospheric aerosol particles impact on climate effects by scattering and absorbing solar radiation directly, and by changing radiative properties of clouds indirectly as cloud condensation nuclei (CCN). Climate effects of atmospheric aerosol depend to a large extent on physicochemical properties of individual particles. Morphological features and mixing states of individual particles in the free troposphere are important to investigate aging of particles during long-range transport and its potential impacts on climate. To elucidate morphological features and chemical composition of individual particles according to their sources and weather conditions, we collected aerosol samples at the summit of Mt. Fuji (35.36°N, 138.73°E, 3776 m a.s.l) located in the free troposphere during summer in 2014 and 2015.
The aerosol samples were obtained using a cascade impactor (the 50 % cut-off diameters of the stages were 0.25 μm and 1.0 μm) on carbon-coated collodion films for 1 hour at flow rate of about 0.55 L/min. To measure the heights of individual particles, particles were coated with Pt / Pd alloy at a shadowing angle of arctan 0.5. Size and chemical composition of individual particles were analyzed using a transmission electron microscope (TEM) equipped with an energy-dispersive X-ray (EDX) analyzer. In this study, 11 samples were obtained and analyzed. The backward trajectories and the average relative humidity (RH) along the trajectories were computed using the HYSPLIT trajectory model (https://ready.arl.noaa.gov/HYSPLIT_traj.php).
In this study, the particles were classified into 9 types (Figure1) based on their morphological features (Ueda *et al.*, 2011). Most of particles were classified as *eroded, dome-like or cluster* particles. In this study, particles classified as the *cluster* particles were included in 6 samples. The *cluster* particles were comprised of some units of *spherical* or *coccoid* particles. It is reported that the *cluster* particles are formed under low RH and rich in sulfur (Ueda. *et al.*, 2011). In this study, however the *cluster* particles were formed under high RH condition. Analysis based on backward trajectories indicates that the sizes of individual particles that constituted *cluster* particles depend on sources and weather conditions. Therefore, the formation mechanism of cluster particles has to be investigated further.

Keywords: Atmospheric aerosol, TEM, Individual particles analyses

©2016. Japan Geoscience Union. All Right Reserved.

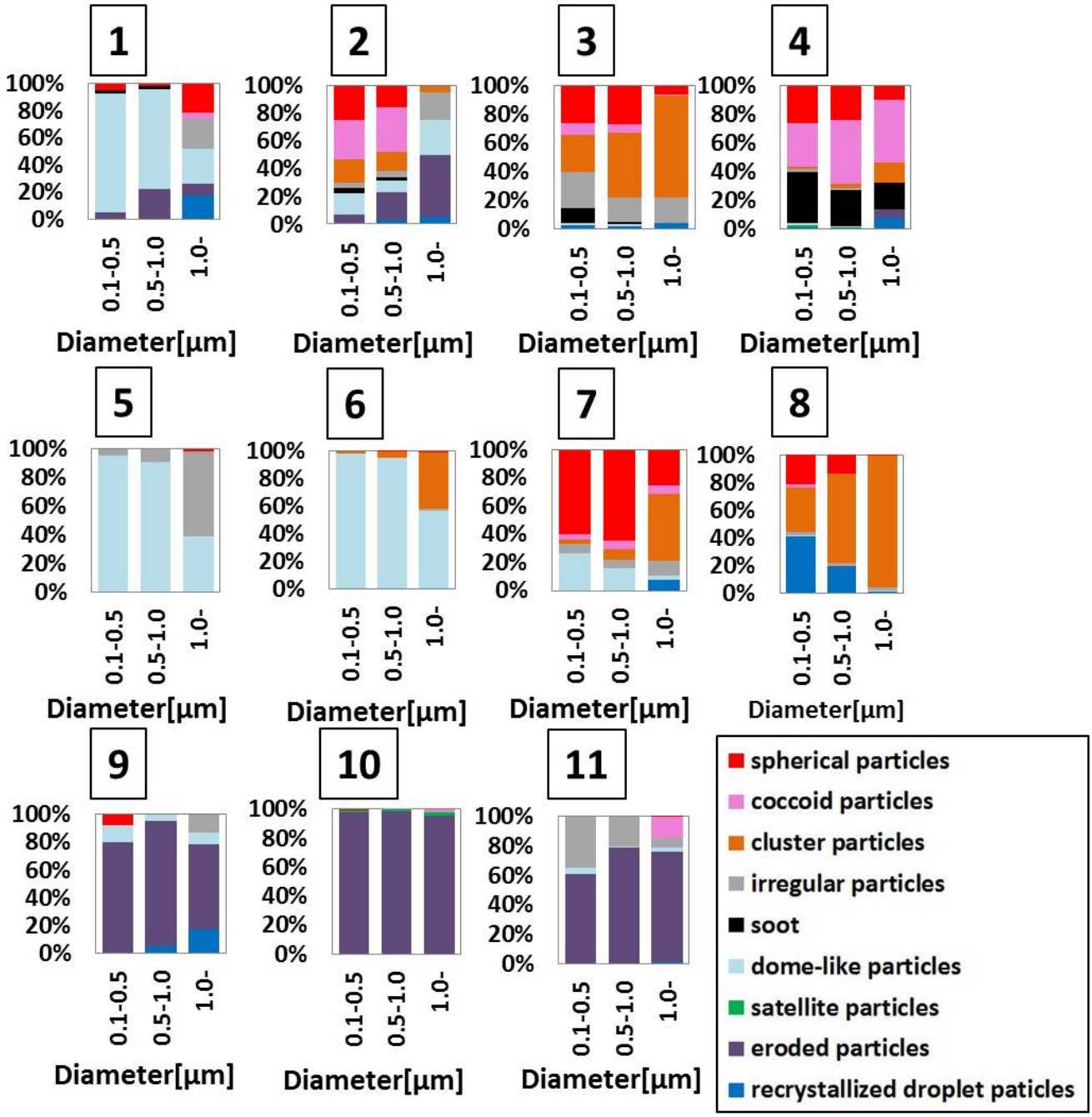


Figure. 1 Size-segregated number proportions of morphological types of particles for samples.

©2016. Japan Geoscience Union. All Right Reserved.

Off-Line Analysis of the Hygroscopicity of Water-Soluble Particulate Matter in the Urban Air of Nagoya

*YUUA NAKMAMURA[1], Miyazaki Yuzo[2], Chen Qingcai[1], Mochida Michihiro[1]

1.Graduate School of Environmental Studies, Nagoya University, 2.Institute of Low Temperature Science, Hokkaido University

Atmospheric aerosol particles are involved in the cloud formation process in the atmosphere by acting as cloud condensation nuclei (CCN). Whether the particles act as CCN are governed by the chemical composition as well as the size and the condition of the water-vapor supersaturation. The presence of water-soluble matter (WSM) in the particles is essential for the CCN activation. Because the composition of water-soluble fraction is complex in terms of the water-soluble organic matter (WSOM), it is difficult to understand the relationship between the composition of aerosol particles and their CCN activity. In this study, we investigated the hygroscopicity of the WSM and the WSOM in urban aerosols over Nagoya, based on the collection of aerosol samples on filters and the analysis of the CCN activity of the water soluble components in the laboratory. The relationship between the O/C ratio of the WSOM and their hygroscopicity was also examined.

Atmospheric aerosol particles with aerodynamic diameters smaller than 0.95 μm ($PM_{0.95}$) were collected on filters in the Higashiyama Campus, Nagoya University, Nagoya, Japan, from 11 to 31, August 2013. Ten aerosol samples were collected, and aerosol components on the filters were extracted with water. Particles were generated by atomizing an extract solution, and they were classified by size using a differential mobility analyzer (DMA). Whereas the classified particles were counted using a condensation particle counter (CPC), CCN-active particles among the classified particles were counted using a CCN counter (CCNC). Four different water-vapor supersaturation (SS) conditions of 0.13%, 0.27%, 0.47%, 0.90% were applied to investigate the CCN activity of the particles. Activation diameters were determined from size-resolved CCN fractions, and then the hygroscopicity parameter $\kappa$of WSM (κ_{WSM}) were calculated. The mass concentrations of WSOM in the sampled atmospheric aerosols were obtained from the analysis of WSOC using a total organic carbon analyzer and the OM/OC ratios derived from the mass spectra collected using a high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). The hygroscopic parameter $\kappa$of the WSOM (κ_{WSOM}) was calculated using the κvalue of the generated particles and the chemical composition data. Two different methods, i.e., a regression-extrapolation method and a Zdanovskii, Stokes, and Robinson (ZSR) method for respective samples, were used to obtain the κ_{WSOM} values.

The κ_{WSM} ranged from 0.34 to 0.51 with a mean of 0.44. The κ_{WSOM} calculated from the regression-extrapolation method were in the range from 0.23 to 0.28 for four respective SS conditions. From the ZSR method for respective samples, the κ_{WSOM} were calculated to be on average 0.16 - 0.28. Whereas no clear relationship was observed for the κ_{WSOM} derived from the regression-extrapolation method and the SS conditions, the higher the SS condition was, the higher the κ_{WSOM} derived from the ZSR method for respective sample was. Further investigation about the cause of the difference of the κ_{WSOM} derived from these two methods is necessary. In the analysis of the studied ten aerosol samples, no clear relationship between the O/C ratio and the κ_{WSOM} was found (r: -0.21).

©2016. Japan Geoscience Union. All Right Reserved.

Individual particle analysis of marine aerosols collected over the Pacific Ocean and its marginal seas

*Momoka Yoshizue[1], Yoko Iwamoto[1], Kouji Adachi[2], Shungo Kato[3], Kazuhiko Miura[1], Mitsuo Uematsu[4]

1.Department of Physics, Faculty of Science Division I, Tokyo University of Science, 2.Meteorological Research Institute, 3.Faculty of Urban Environmental Sciences, Tokyo Metropolitan University, 4.Atmosphere and Ocean Research Institute, The University of Tokyo

Sea-salt particles are produced by bubble bursting processes at the sea surface. Chemical compositions of sea-salt particles are modified when they react with acidic substances such as sulfuric acid (H_2SO_4), nitric acid (HNO_3) and methanesulfonic acid (MSA) in the atmosphere. The sources of acidic substances include anthropogenic pollutants, volcanic eruptions and dimethyl sulfide (DMS) from marine biota. If the acidic substances react with sea-salt particles, they are easily scavenged from the atmosphere. So the lifetime of acid substances in the atmosphere will be reduced and they are unlikely to be cloud condensation nuclei. Therefore cooling effect of clouds might be smaller than expected in the environment where sea-salt particles are abundant. In this study, difference in chemical compositions of sea-salt particles among ocean regions was revealed based on individual particles analyses of marine aerosols collected over the Pacific and its marginal seas. Additionally, source of acidic substances that modified sea-salt particles was discussed.

Sampling of marine atmospheric aerosol particles was carried out during KH-13-7 cruise (2013/12/11~2014/2/12) and KH-14-3 Leg2 cruise (2014/7/17~8/11) in the Pacific and its marginal seas. Individual particles were analysed using a transmission electron microscope and an energy dispersive X-ray spectrometer.

In most ocean regions, unmodified sea-salt particles accounted for more than 80% of the analysed particles. However, sulfate particles accounted for more than 85% in the sample collected around Guam islands. High concentrations of radon and number of aerosol particles, along with analysis of backward trajectories suggested that polluted air masses originated from the Asian continent came to the observation area. The aerosols collected around the Aleutian Islands included large number of sulfate and modified sea-salt particles. A Na-Cl-S ternary diagram indicates that sea-salt particles were modified with MSA produced from DMS oxidation or H_2SO_4. Air masses from a volcano transported from the Kamchatka were also the potential source of H_2SO_4 around the Aleutian island.

Keywords: marine atmospheric aerosol, sea-salt, sulfate, volcano, dimethyl sulfide, methanesulfonic acid

©2016. Japan Geoscience Union. All Right Reserved.

Evaluation of performance of simulated secondary air pollutants by using air quality models for the Kanto area in summer 2011

*Kazuyo Yamaji[1], Hirokazu Uede[1], Satoru Chatani[2], Hiroshi Hayami[3], Hikari Shimadera[4]

1.Kobe University, 2.NIES, 3.CRIEPI, 4.Osaka University

Emissions and concentrations of primary atmospheric pollutants (eg. NOx and VOCs) have been decreased in the Kanto area owing to conduct their emission regulations. Nevertheless, atmospheric pollutants such as particulate matters (PM) with aerodynamic diameter less than 2.5 μm (PM2.5) and photochemical oxidants (Ox) have still remained high concentration levels and their air quality standards of Japan have not been attained at most monitoring stations (Ministry of the Environment of Japan (MOE), 2015). For forming effective air pollution control strategies, currently, we raise expectations for applying air quality models reproducing complicated physical and chemical processes of both of primary and secondary pollutants.
The urban air quality model inter comparison study in Japan (UMICS) was started to improve performances of air quality models (eg. Chatani et al., 2014, Shimadera et al., 2014). UMICS showed some critical problems immanent in the air quality models. For example, the models tended to overestimate NO_3^- but to underestimate OA, although simulated PM2.5 concentrations were reasonable with comparing to observations at Kanto area (Shimadera et al., 2014). In terms of O_3, the models reproduced well the diurnal and inter-diurnal variations in the O_3 concentrations at most observational stations in Kanto area but tended to overestimate nighttime O_3 and to underestimate daytime O_3 at several observational stations (Morino et al., 2010), therefore these models might have a risk failing to predict some of high pollution events.
In order to find the causes of discrepancies between the simulated and observed concentrations of secondary pollutants, constituents of PM2.5 and O_3, in this study, air quality simulations were performed using the Weather Research and Forecasting (WRF) model for a meteorological model and the Community Multi-scale Air Quality (CMAQ) model system for a chemical transport model under the following different model settings. Meteorological analysis data (FNL/NCEP and MSM/JMA) with both different temporal and spatial resolutions were used as for input data of meteorological simulations by the WRF model, respectively. Additionally, the updated JEI-DB (JATOP Emission Inventory Data Base) was used for input emission information for the CMAQ model. Performances of these models under different settings were evaluated by comparing with observed concentrations (O_3, PM2.5, and constituents of PM2.5) of secondary pollutants at Kanto area, which were provided by UMICS and MOE. These results are also compared with the previous studies (eg. Shimadera et al., 2014).

Keywords: Air quality model, secondary pollutants, Urban Air quality

©2016. Japan Geoscience Union. All Right Reserved.

Development of a global aerosol model using a two dimensional bin method

*Hitoshi MATSUI[1,2,3], Naga Oshima[4]

1.Nagoya University, 2.Cornell University, 3.Japan Agency for Marine-Earth Science and Technology, 4.Meteorological Research Institute

Atmospheric aerosols play an important role in Earth's climate system through radiation and cloud/precipitation processes. Estimates of these aerosol impacts on climate remain highly uncertain, and they are one of the largest uncertainties in predicting climate change. Accurate estimations of these effects through a model require good representations of number concentration, size distribution, and mixing state of aerosols. However, many of existing three-dimensional aerosol models do not represent these aerosol parameters sufficiently. In our previous studies, we developed an aerosol model, the Aerosol Two-dimensional bin module for foRmation and Aging Simulation (ATRAS), that can explicitly calculate aerosol number concentration, size distribution, and mixing state with a two-dimensional bin representation. The ATRAS model was implemented into a regional three-dimensional model WRF-chem, and we have shown the importance of aerosol simulations using a model that can treat detailed aerosol processes and parameters [Matsui et al., 2014; Matsui, 2016a, 2016b]. In this study, we developed a new aerosol model based on the ATRAS model and implemented it to a global climate model CAM5.

The computational cost is one of the most important factors in the development of a global aerosol model using a two-dimensional bin representation because global and long-term simulations are necessary in a global modeling study. To reduce the computational cost, we developed a new aerosol box model by improving the source codes of all aerosol microphysical and chemical processes in the ATRAS model and by reducing the number of aerosol variables considerably. These changes reduced the computational cost of aerosol microphysical and chemical processes by 90% compared with the original ATRAS model with keeping the accuracy of simulations for aerosol number concentration, size distribution, and mixing state.

We implemented this new box model to CAM5 and conducted five-year test simulations. The model simulations were validated through the comparison with the original aerosol model in CAM5 (MAM) and various surface and aircraft measurements of aerosols. We also calculated global distributions of some aerosol parameters that can be estimated globally for the first time by using our detailed aerosol model.

In the presentation, we would like to show the concepts and results of the new box model and the results of global model simulations such as the comparison with MAM simulations and measurements and sensitivity simulations.

References:

Matsui, H. et al. (2014), Atmos. Chem. Phys., 14, 10315-10331.
Matsui, H. (2016a), J. Geophys. Res. Atmos., 121, doi:10.1002/2015JD023998.
Matsui, H. (2016b), J. Geophys. Res. Atmos., 121, doi:10.1002/2015JD023999.

Keywords: Aerosol, Global aerosol model, Two-dimensional bin model, Number concentration, Size distribution, Mixing state

©2016. Japan Geoscience Union. All Right Reserved.

Estimating global budget of formaldehyde and BVOCs emission using satellite observations and global chemistry transport model

*Kengo Sudo[1,2], Yuki Isono[1], Akihiko Ito[3,2], Kazuyuki Miyazaki[2]

1.Graduate School of Environmental Studies, Nagoya University, 2.Japan Marine-Earth Science and Technology, 3.National Institute of Environmental Studies

This study evaluates global distribution and budget of atmospheric formaldehyde using satellite measurements and global chemistry-transport model simulations, particularly focusing on the roles of global emission of biogenic VOCs (BVOCs) and methane oxidation. Formaldehyde (HCHO) is chemically produced by oxidation of methane (CH_4) and volatile organic compounds (VOCs) in the atmosphere, and hence its global budget can be used for deriving methane concentrations and VOCs emissions. For simulating global HCHO, this study uses a global chemistry-transport model CHASER (MIROC-ESM version) which considers detailed chemistry in the troposphere and stratosphere with an on-line aerosol simulation including production of particulate nitrate and SOA. We use the NCEP reanalysis data (FNL) for constraining the model's meteorology. Anthropogenic and biomass burning emissions are specified using the EDGAR-HTAP2 and MAC inventories, respectively. For a base emission of BVOCs, we employ calculation by the land ecosystem/trace gas emission model VISIT (Ito et al., 2008) and MEGAN (Guenther et al., 2006) for 2000-2012. In this study, global emission of BVOCs is first estimated employing a series of emission sensitivity simulations by CHASER in combination with the global and regional HCHO distributions derived from the OMI satellite observation. As a result, the global emission amount of isoprene (a major component of BVOCs) is estimated at 300 - 400 TgC yr^{-1}, suggesting that the current estimates as by the VISIT and MEGAN land-ecosystem models (> 500 TgC yr^{-1} for isoprene) are probably overestimated. For the global budget of HCHO, the model with global isoprene emission of 400 TgC yr^{-1} estimates a major contributions from the CH_4 oxidation (ca. 66%) to the global production of HCHO, which is followed by oxidation of BVOCs (ca. 21%) and anthropogenic and biomass burning related VOCs (ca. 13%). The CH_4 contribution to the global HCHO production, inferred from the OMI HCHO observation, is about 71%, significantly larger than the above-estimated value, suggesting that global isoprene emission may be less than 300 TgC yr^{-1} or that anthropogenic VOCs emissions in the current inventories may be overestimated.

Keywords: BVOCs, Isoprene, Formaldehyde, OMI satellite observation, Chemistry-transport model

©2016. Japan Geoscience Union. All Right Reserved.

Contribution of plant-associated microorganisms as global sinks of atmospheric hydrogen

*Manabu Kanno[1], Philippe Constant[2], Hideyuki Tamaki[1], Yoichi Kamagata[1]

1.National Institute of Advanced Industrial Science and Technology, 2.Centre INRS-Institut Armand-Frappier, Canada

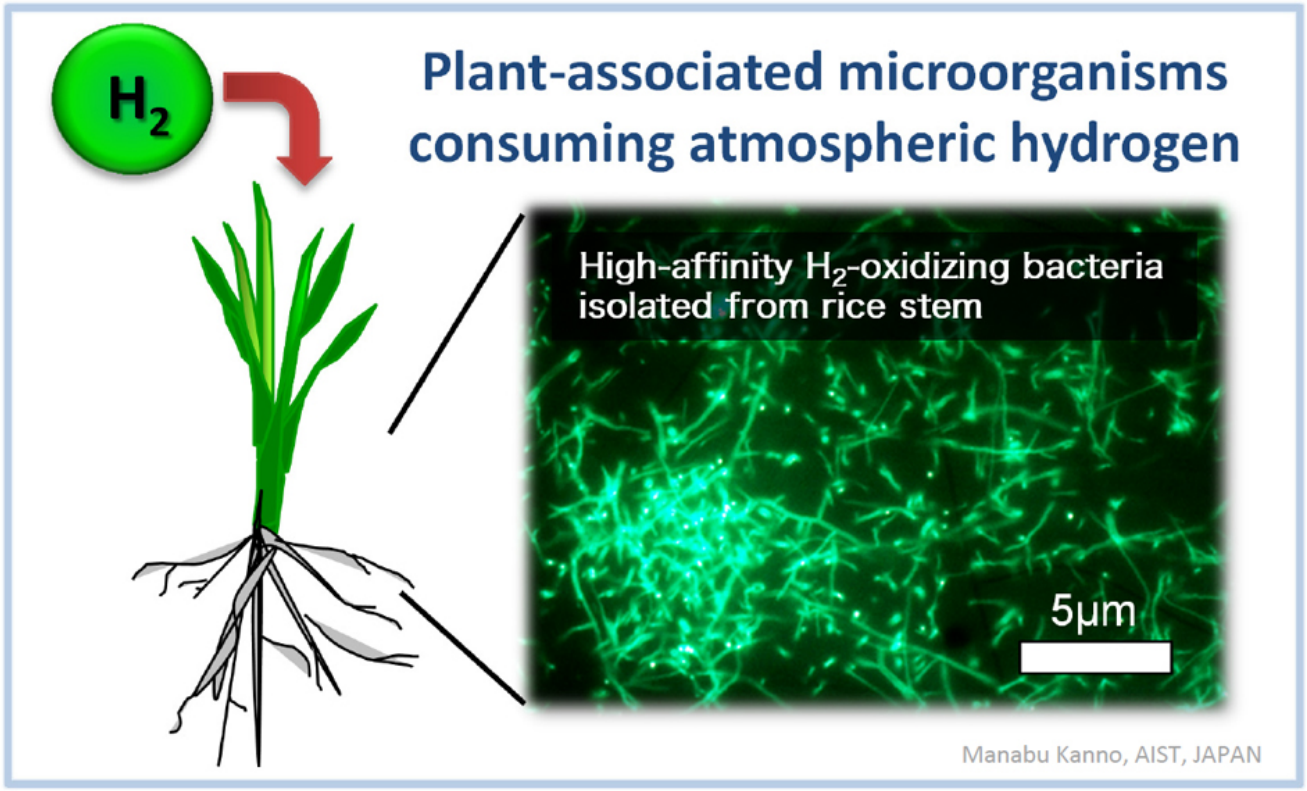
Hydrogen (H_2) is an important constituent of the atmosphere, with a typical mixing ratio of 0.530 parts per million by volume (ppmv). Rising H_2 emissions under a future H_2-based economy are concerned to increase the atmospheric burden of H_2, resulting to the indirect influence of the lifetime of greenhouse gas CH_4, an alteration of temperature and ozone loss in the stratosphere. Thus, mitigation of H_2 emission is of critical importance for atmospheric chemistry. The most part (~80%) of tropospheric H_2 is consumed by microorganisms in soil. A recent literature survey of H_2 flux measurements unveiled that soil H_2 uptake is responsible for the loss of 40 to 90 Tg yr^{-1}. Recently, high-affinity H_2-oxidizing bacteria possessing novel hydrogenase have been found as important contributors to the soil H_2 uptake. Although previous experiments using molecular tritium reported the occurrence of significant H_2 uptake activity in vegetation, there has been no report on the identification and diversity of the responsible microorganisms. This study aimed to verify the existence of plant-associated bacteria possessing the ability to consume atmospheric H_2.

We first investigated the presence of *hhyL* gene in various plant species. The *hhyL* gene, which encodes for the large subunit of the novel group of hydrogenase, has been generally used as a functional biomarker to evaluate the distribution, taxonomic diversity, and abundance of high-affinity H_2-oxidizing bacteria. In total, 42 *hhyL* gene sequences were successfully detected in all tested herbaceous plants, indicating a wide distribution of high-affinity H_2-oxidizing bacteria in plants. It is noteworthy that the abundance levels of *hhyL* gene detected in plants were comparable to those detected in soil. High-affinity H_2-oxidizing bacteria were isolated from inside herbaceous plant tissues. Among 145 isolates, 7 *Streptomyces* strains were shown to possess *hhyL* gene. The H_2 uptake activity was evaluated by gas chromatography. All the isolates reduced H_2 concentration to less than 0.530 ppmv, demonstrating the ability to consume H_2 at ambient level. Sterile plant seedlings were inoculated with selected isolates to verify their ability to penetrate and disseminate in plant tissues and scavenge atmospheric H_2 in plant. After four weeks of seedling inoculation, an internalization of the bacteria in plant tissues was visualized by fluorescence *in situ* hybridization imaging. H_2 oxidation rates measured in plant fractions ranged from 1079 to 3472 pmol $g_{(dw)}^{-1}$ h^{-1}. These rates are comparable to the previously observed activity of atmospheric tritium uptake in other plants. Importantly, atmospheric H_2 is not oxidized in aseptically grown plants, clearly showing that plant-associated bacteria was responsible for H_2 loss. H_2 uptake activity per bacterial cell was comparable between plant and soil, demonstrating that both environments are favorable for the microbial-mediated H_2 uptake.

In conclusion, this study demonstrated the occurrence of plant-associated high-affinity H_2 -oxidizing bacteria and their ability to consume atmospheric H_2 on plant surface or inside plant tissues. From a global perspective, herbaceous and woody plant biomass represent approximately 64 Pg, and 736 Pg, respectively. Considering that high-affinity H_2-oxidizing bacteria may be present and active in these plants, the contribution of plant-associated bacteria deserves more attention to better understand the global cycling of atmospheric H_2.

Keywords: biogeochemistry, microbial ecology, tropospheric H2 cycle, vegetation

©2016. Japan Geoscience Union. All Right Reserved.



©2016. Japan Geoscience Union. All Right Reserved.

Estimating secondary formation of atmospheric HONO using triple oxygen isotopes as tracers

*Fumiko Nakagawa[1], Ray Nakane[1], Urumu Tsunogai[1], Kengo Sudo[1], Izumi Noguchi[2], Takashi Yamaguchi[2]

1.Graduate School of Environmental Studies, Nagoya University, 2.Hokkaido Institute of Environmental Sciences

The photolysis of nitrous acid (HONO) has been recognized as a potentially important source of OH radicals, which is known as a major oxidant in the atmosphere removing reductive trace gases such as methane and NMHCs. Atmospheric HONO originates from both primary sources (direct emissions) and secondary sources (chemical formation in the atmosphere), however, their contributions to atmospheric HONO production have not been well understood. Here, we determined a triple oxygen isotope of HONO because $\Delta^{17}O$ value of HONO produced via “secondary formation” is expected to have highly positive values as those of O_3 ($\Delta^{17}O$ = +30 ±10‰), while no $\Delta^{17}O$ anomaly ($\Delta^{17}O$ = 0‰) should be observed for HONO which is emitted directly from various sources on the ground, making it possible to quantify their contribution to the atmospheric HONO production with $\Delta^{17}O$ measurement. Periodical sampling of atmospheric HONO was carried out once a month starting from December, 2014 at Hokkaido Institute of Environmental Sciences, Sapporo, Japan. The sample collection period was fixed to one week with a flow rate of 10 L/min. $\Delta^{17}O$ value of HONO was determined by combining sensitive determination method on isotope compositions of NO_2^- (Komatsu *et al.*, 2008; Tsunogai *et al.*, 2010) with filter-pack method (Noguchi *et al.*, 2007) in which to collect HONO as NO_2^-. The result of daily mean $\Delta^{17}O$ value of HONO ranged from +6.9‰ to +10.7‰ through the observation periods. $\Delta^{17}O$ value of HONO showed higher value on the day time than night time. The ratios of HONO derived from secondary formation in Sapporo was almost constant throughout the year (day and night : 34±2%, day : 66±8%, night : 21±2%) leading to conclusion that direct emissions are dominant HONO sources in Sapporo.

Keywords: HONO, tripple oxygen isotopes, secondary formation, direct emission, atmosphere

©2016. Japan Geoscience Union. All Right Reserved.

Validation of MODIS MCD64A1 burned area in boreal Eurasia

*Chunmao Zhu[1], Hideki Kobayashi[1], Yugo Kanaya[1], Masahiko Saito[1]

1.Japan Agency for Marine-Earth Science and Technology

Biomass burning plays an important role in affecting vegetation dynamics, biogeochemical cycle of carbon, nitrogen and other elements, atmospheric chemistry and the climate. Boreal Eurasia is one of the most important regions where large areas are burned every year. Numerous particles and greenhouse gases are emitted from these fires. These pollutants could be transported to the industrialized region in East Asia, the western North Pacific and the Arctic. Atmospheric transport of fire-emitted pollutants to the Arctic and the consequential deposition is believed to accelerate the arctic warming. To better understand the effect of fires in boreal Eurasia on the arctic, an accurate assessment of burned area from the boreal Eurasia is necessary.
The Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 5.1 direct broadcast monthly burned area product MCD64A1 is widely used for global burned area mapping. MCD64A1 data was also used for the estimation of global fire emissions such as Global Fire Emissions Database, Version 4 (GFED4). However, uncertainties in burned area estimations could be introduced due to the “moderate resolution” character of MCD64A1 (~500m). Therefore, a comparison of the burned areas of MCD64A1 with those generated from higher resolution satellite products could provide basic and crucial information for its accuracy assessment and further applications. However, there are few studies on the validation of MCD64A1, especially in the boreal Eurasia.
In this work, we used Landsat 7 surface reflectance, along with a few commercial satellite scenes from WorldView, GeoEye and RapidEye as the reference scenes to derive the burned areas in a burning season in 2012 (mostly July to September). A wide range of ecotypes over wide geographic regions spanning from the western Russia/Kazakhstan to the eastern Siberia were covered. Each of these burned areas was compared with the corresponding region of MCD64A1 for the same periods. Our preliminary results indicated that MCD64A1 could well capture the large fires, while those less than 100 ha are prone to be undetected. We also found that MCD64A1 tends to underestimate the burned area in general. Based on the error statistics, we suggested the accuracy levels and precautions for applications in each ecotype.

©2016. Japan Geoscience Union. All Right Reserved.

Evaluation of GOSAT/TANSO-FTS TIR CH_4 data using NICAM-TM and aircraft CH_4 data

*Ryosuke Nonogaki[2,1], Naoko Saitoh[2], Ryoichi Imasu[3], Kei Shiomi[4], Yosuke Niwa[5], Shuji Aoki[6], Shinji Morimoto[7], Toshinobu Machida[8], Hidekazu Matsueda[5], Yousuke Sawa[5], Kazuhiro Tsuboi[5]

1.The Graduate School of Advanced Integration Science, Chiba university, 2.Center for Environmental Remote Sensing, 3.Atmosphere and Ocean Research Institute, The University of Tokyo, 4.JAXA, 5.Meteorological Research Institute, 6.Center for Atmospheric and Oceanic Studies, Graduate School of Science, Tohoku University, 7.Graduate School of Science, Tohoku University, 8.National Institute for Environmental Studies

Greenhouse gases Observing SATellite (GOSAT) is the first satellite that was dedicated to the global observations of CO_2 and CH_4, and was launched on January 23, 2009. CH_4 profiles can be retrieved from the thermal infrared (TIR) band of Thermal and Near-infrared Sensor for Carbon Observation Fourier Transform Spectrometer (TANSO-FTS) on board the GOSAT. In this study, we compared CH_4 data from the TIR band of TANSO-FTS with CH_4 data from Nonhydrostatic ICosahedral Atmospheric Model-based Transport Model (NICAM-TM) [Niwa et al., 2011] and aircraft measurements to evaluate the quality of the TIR CH_4 data. First, we compared TIR, NICAM-TM, and CONTRAIL/GRENE CH_4 data [Sawa et al., 2015] on the isentropic surfaces in the upper troposphere and lower stratosphere. Second, we compared TIR CH_4 data with JMA aircraft CH_4 measurement data over Minamitorishima [Niwa et al., 2014] in the upper and middle troposphere. Here, we adopted a distance between TANSO-FTS and the aircraft measurement locations within ±3 degree and a time difference between the two observations within ±3 days as criteria for the comparisons.
From the isentropic analysis, we found that the CONTRAIL/GRENE CH_4 concentrations showed a large seasonal variation in the lower stratosphere, while the TIR CH_4 data had higher concentrations there than the CONTRAIL/GRENE data and showed a relatively small seasonal variation; the seasonal variation of the NICAM-TM CH_4 data were smaller than that of the TIR data. From the profile comparisons over Minamitorishima, we found that the TIR and aircraft CH_4 data agreed to each other within 30 ppb at around 6 km in winter and spring, while their differences increased to 30 –50 ppb in summer. We also investigated the impact of the coincident criteria on the comparisons results. Besides, we analyzed latitudinal distribution of TIR and aircraft CH_4 data in the upper troposphere between Atsugi and Minamitorishima.

Acknowledgements
CONTRAIL/GRENE flask sampling data over Siberia was conducted under the GRENE Arctic Climate Change Research Project. Aircraft measurements between Atsugi and Minamitorishima were conducted by the Japan Meteorological Agency.

Keywords: GOSAT, CH4, evaluation

©2016. Japan Geoscience Union. All Right Reserved.

The impact on CH_4 retrieval of GOSAT/TANSO-FTS TIR band from the uncertainty of the continuum absorption

*Yamada Akinori[1], Naoko Saitoh[1], Ryoichi Imasu[2], Akihiko Kuze[3], Kei Shiomi[3]

1.Center for Environmental Remote Sensing, Chiba University, 2.Atmosphere and Ocean Research Institude, University of Tokyo, 3.Japan Aerospace Exploration Agency

The thermal infrared (TIR) band of Thermal and Near-infrared Sensor for Carbon Observation Fourier Transform Spectrometer (TANSO-FTS) onboard Greenhouse Gases Observing Satellite (GOSAT) observes CH_4 profile at wavenumber range from 1210 cm^{-1} to 1360 cm^{-1} including CH_4 ν_4 band. The current retrieval algorithm (V1.0) uses MT CKD continuum model (Mlawer et al., 2012) to calculate optical depth of continuum absorption for forward spectrum. The temperature coefficient of the water self continuum of MT CKD model overestimated about 30% at the CH_4 retrieval band compared to that of BPS continuum model (Paynter and Ramaswamy, 2011). We study the impact on CH_4 retrieval from the uncertainty of the continuum absorption.

We used 713 spectra observed by GOSAT/TANSO-FTS TIR band from March 2010 to September 2011. These observations satisfy following criteria. The coincidence criteria of distance and time between GOSAT-TIR observations and HIPPO aircraft observations (Wofsy et al., 2012) are 300 km and 72 hours, respectively, and the degrees of freedom for signal are greater than 0.7. We calculated forward spectrum using retrieved profile by LBLRTM V12.2 (Clough et al., 2005) and convolution of the forward spectrum and sinc function as an instrumental line shape function to set to low wavenumber resolution.

The spectral residual (forward spectrum - GOSAT-TIR observation spectrum) had fine structure from -20 K to 20 K, which was made from line absorption. We calculated moving average with window width of 11 channels to separate continuum from line absorption. The average of the moving-averaged residual were -2.0 K. According to Saitoh et al. (2009), 1 K difference of surface temperature make 5% bias at the altitude of 400 mbar in CO_2 profile, there is a potential for bias in CH_4 profile. There exist H_2O self continuum, H_2O foreign continuum, CO_2 continuum, and O_2 continuum in this band. The wavenumber dependencies of H_2O self continuum and CO_2 continuum are small in this band, while that of H_2O foreign continuum and O_2 continuum are large.

We calculated forward spectra using 10% larger continuum absorption coefficient than MT CKD model to know the impact on brightness temperature spectrum from continuum absorption. The brightness temperature changed about -0.00001 K and 0.01 K when we used 10% larger CO_2 continuum absorption and 10% larger H_2O self continuum absorption, respectively. The differences on brightness temperature were much smaller than the averaged residual. We are trying comparing GOSAT-TIR CH_4 profile retrieved using 10% bias continuum absorption and other continuum model with HIPPO profile.

References:

Mlawer, E. J., et al. (2012) Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences 370, 2520–2556.

Paynter, D. and Ramaswamy, V. (2011) JGR 116, D20302.

Saitoh, N., et al. (2009) JGR 114, D17305.

Wofsy, S. C., et al. (2012) http://dx.doi.org/10.3334/CDIAC/hippo_010.

©2016. Japan Geoscience Union. All Right Reserved.

Development of a new match-up method of the satellite and ground-based greenhouse gases data by trajectory analysis for the GOSAT-2 data validation

*Takahiro Nakatsuru[1], Isamu Morino[1], Osamu Uchino[1], Yukio Yoshida[1], Makoto Inoue[2]

1.National Institute for Environmental Studies, 2.Akita Prefectural University

The GOSAT (Greenhouse gases Observing SATellite) was launched in January 2009 and has been operating jointly by MOE, JAXA and NIES. GOSAT is the world's first spacecraft designed specifically to measure the concentrations of carbon dioxide and methane, the two in the major greenhouse gases. Currently, GOSAT-2 is under development. Many satellite data have been compared to ground-based data, which have higher accuracy and precision. Satellite and ground-based instruments generally observe target quantity with different spatial scale/position and different time. Therefore, it is important to find more plausible coincidence criteria between them to validate satellite data.
The SWIR (Short Wavelength InfraRed) surface scattered solar spectra observed by the TANSO-FTS (Thermal And Near infrared Sensor for carbon Observation -Fourier Transform Spectrometer) onboard GOSAT is used to retrieve column-averaged dry-air mole fractions of carbon dioxide and methane (XCO_2 and XCH_4). TCCON (Total Carbon Column Observing Network) is a ground-based observation network using Bruker IFS 120HR/125HR high-resolution fourier transform spectrometers. The GOSAT data, XCO_2 and XCH_4, has been validated using TCCON data. However, GOSAT and TCCON data are not completely spatio-temporally matched due to characteristics of the satellite orbit.
For example, one of the co-location methods uses a geometric distance between GOSAT and TCCON data to obtain matched data (Geophysical co-location method, e.g., Morino et al., 2011). Validation of GOSAT requires statistically significant match-up number but there is not enough match-up number with geophysical co-location method. In the case of XCO_2, various methods for increasing the number of match-up data have been developed: the same potential temperature field at 700-hPa as a proxy for equivalent latitude for CO_2 gradients (Keppel-Aleks et al., 2011, Wunch et al., 2011), and the same concentration field predicted or assimilated with the atmospheric transport model (Guerlet et al., 2013). For the time gap, it has been used the same day or within the time range of GOSAT overpass time because GOSAT has three-day revisit and sun-synchronous orbit with a local time around 13h at descending. Increasing the number of match-up data in consideration of the gaps of time and space is important for advanced validation.
In this study, we develop a new match-up method utilizing the forward and backward trajectory analyses from GOSAT observation position by HYSPLIT model. The new method is applicable to other gases not only XCO_2 using flow of air masses. We show the result of analyzed bias variation with the spatio-temporal gap and the effectiveness of the developed method together with results using other match-up methods.

Keywords: GOSAT, TCCON, match-up

©2016. Japan Geoscience Union. All Right Reserved.

Continuous measurement of CO_2 and CH_4 concentration from a tower network (JR-STATION) over Siberia

*Motoki Sasakawa[1], Toshinobu Machida[1], Noritsugu tsuda[2], Keiichi Katsumata[1], Mikhail Arshinov[3], Denis Davydov[3], Alexandrov Fofonov[3]

1.National Institute of Environmental Studies, 2.Global Environmental Forum, 3.Institute of Atmospheric Optics, Russian Academy of Sciences, Siberian Branch, Russia

Continuous measurements of CO_2 and CH_4 concentration have been carried out with a tower network in Siberia (JR-STATION: Japan-Russia Siberian Tall Tower Inland Observation Network) in order to study the spatial and temporal variations of CO_2 and CH_4 in the forest, steppe, and wetland regions and estimate the distribution of the flux over this huge area (Sasakawa et al., 2010) where only a few atmospheric investigations were made.

The JR-STATION consists of 6 towers (Figure) located at Berezorechka (BRZ) since 2002, at Karasevoe (KRS) since 2004, at Demyanskoe (DEM) and Noyabrsk (NOY) since 2005, at Azovo (AZV) since 2007, and at Vaganovo (VGN) since 2008. Air samples taken at two heights (~85 m) on each tower were analyzed with an NDIR (LI-COR, LI-820) for CO_2 and a SnO_2 semiconductor sensor (Suto et al., 2010) for CH_4 after passing through the line with a glass water trap, a Nafion membrane dryer (PERMA PURE, MD-050-72F-2), and a magnesium perchlorate. Measurement precision was ±0.3 ppm for CO_2 and ±5 ppb for CH_4.

Both CO_2 and CH_4 concentration showed clear diurnal variation during summer mainly due to the diurnal variation of the PBL height (Sasakawa et al., 2012), which is pronounced in inland continental locations such as Siberia. Sasakawa et al. (2013) reported that daytime (13:00-17:00 LST) mean data observed at the towers can capture the characteristic of CO_2 concentration in the PBL well during dormant season, and growing season (June-August) with a negative bias of -2.4±0.8 ppm (80 m inlet). This bias is characteristic of close ground observation at high source/sink region. Using the daytime mean, spatial and temporal variations in annual and decadal scale were obtained for Siberia; e.g. the CO_2 drawdown in the summer of 2010 in West Siberia was much shallower than in 2009. This result is consistent with the report that carbon uptake at Eurasia in the summer of 2010 was reduced because of the heat wave in Eurasia driving biospheric fluxes and fire emissions (Guerlet et al., 2013).

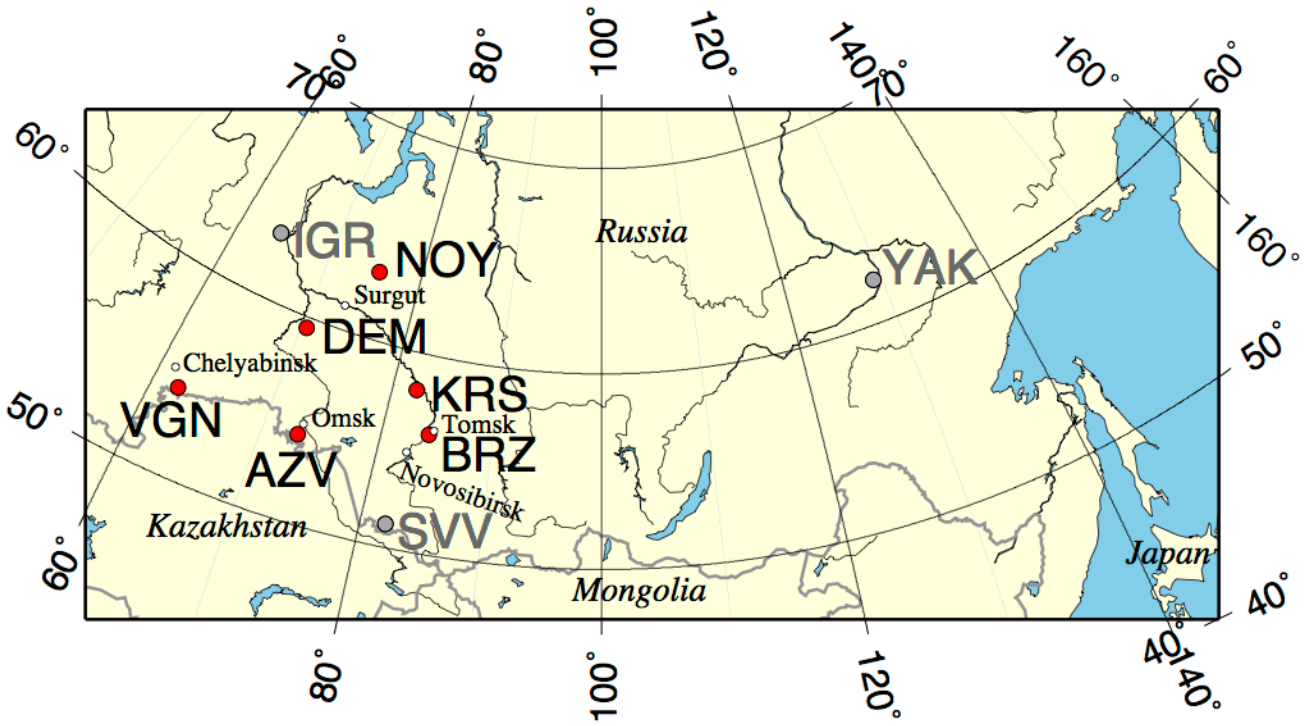
We have started to install a Cavity Ring-Down Spectroscopy (CRDS; Picarro inc., G2401) to renew the antiquated system. Branching before the magnesium perchlorate, sample air flowed into the CRDS through a Nafion dryer (PERMA PURE, MD-050-72S-1). The flow rate was controlled at the same value for the original path (35 ml/min) with a mass flow controller. The water vapor was kept at <0.01 % in this condition, which value is good enough for water correction of the CRDS (Nara et al., 2012). The CO_2 concentration in compressed dry natural air measured with the CRDS (391.85±0.04 ppm) showed good agreement with that measured with the NDIR (391.72±0.18 ppm). We will present preliminary atmospheric data observed with this modified system at KRS in the presentation.

References

Guerlet, S. et al., Geophys. Res. Lett. 40, 2378-2383, 2013.
Nara, H. et al., Atmos. Meas. Tech. 5, 2689-2701, 2012.
Sasakawa, M. et al., Tellus 62B, 403-416, 2010.
Sasakawa, M. et al., Tellus 64B, doi:10.3402/tellusb.v64i0.17514, 2012.
Sasakawa, M. et al., J. Geophys. Res. 118, 1-10, doi:10.1002/jgrd.50755, 2013.
Suto, H. et al., J. Atmos. Ocean. Tech. 27, 1175-1184, 2010.

©2016. Japan Geoscience Union. All Right Reserved.

Keywords: Siberia, Carbon dioxide, Methane, tower observation, CRDS



©2016. Japan Geoscience Union. All Right Reserved.

Estimation of the CO_2 source by measuring oxygen and carbon isotopes in atmosphere

*Yu Hoshina[1], Yasunori Tohjima[1], Yukio Terao[1], Keiichi Katsumata[1], Hitoshi Mukai[1], Toshinobu Machida[1], Yumi Osonoi[1]

1.National Institute for Environmental Studies

Atmospheric carbon dioxide (CO_2) concentrations observed in urban and continental areas and the surrounding areas often show short-term elevations on a timescale from several hours to several days. These variations are considered to be attributed to the CO_2 emissions from biotic activities and burning of fossil fuels (coal, oil and natural gas). If the contribution rate from each CO_2 source is clearly determined, the uncertainty of the CO_2 emissions estimated from atmospheric inversion calculations would be reduced. In this study, we develop a method to evaluate the contribution rate from individual sources based on measurements of carbon stable isotope (^{13}C), radiocarbon (^{14}C) and O_2 concentration as well as CO_2 concentration in the atmosphere. The $-O_2:CO_2$ exchange ratios of the fossil fuel burning are different for the fuel types because the ratios are stoichiometrically related to the elemental compositions of the individual fuels. The ^{14}C measurements is useful to separate the fossil fuel emissions from the biotic emissions because the fossil fuel-derived CO_2 contain no ^{14}C. In addition, values of ^{13}C depend on source of CO_2. Therefore, the combination of CO_2 measurements with O_2, ^{13}C and ^{14}C measurements allow us to estimate the contribution rates of the individual CO_2 source.
In this presentation, we show preliminary results of the atmospheric measurements which were conducted at Tsukuba in July-August 2015 to assess the usefulness of the above method. In the experiment, the atmospheric CO_2 and O_2 concentrations were continuously measured, and the air samples were collected in the glass flasks to measure the carbon isotopes. The relations between CO_2 concentrations and $\Delta^{14}C$ for the observed CO_2 change suggest that 60–70% of the CO_2 change are attributed to the fossil fuel-derived CO_2. Taking into account of the $-O_2:CO_2$ ratio for land biotic processes of 1.1, we can obtain the $-O_2:CO_2$ ratio for the fossil fuel component of the observed changes of 1.37–1.41, which is close to the exchange ratio for oil burring (R=1.44). The relation between CO_2 and ^{13}C is also consistent with the above result.

©2016. Japan Geoscience Union. All Right Reserved.

Temporal variation of the total column of ethane observed with FTIR at Tsukuba

*Isao Murata[1], Hideaki Nakajima[2,3], Isamu Morino[2]

1.Graduate School of Environmental Studies, Tohoku University, 2.National Institute for Environmental Studies, 3.Council for Science, Technology and Innovation, Cabinet Office, Government of Japan

The total columns of C_2H_6 have been observed with a high-resolution Fourier transform spectrometer at Tsukuba, Japan since 1999. SFIT2 spectral fitting program was used to derive the total column from 2 spectral windows in 3 micrometer region.
C_2H_6 is the second major hydrocarbon and contribute to global warming and air pollution indirectly. The main sources are anthropogenic ones such as natural gas, biofuel, and biomass burning. But the measurements at Jungfraujoch [Franco et al., 2015] show that the emissions may be underestimated from the comparison with simulation results.
Preliminary retrieved total column of 1.5 -3.5 x 10^{16} molecule/cm^{-2} at Tsukuba in 2012 is consistent with that of previous study [Zhao et al., 2002] at Hokkaido and is larger than the simulation result. The seasonal variation which shows maximum in spring and minimum in autumn is also consistent with those of previous studies. This is due to the increase of the destruction by the reaction with OH in summer.

Keywords: FTIR, Greenhouse Gas, ethane

©2016. Japan Geoscience Union. All Right Reserved.

Development of a real-time measurement device of atmospheric carbon monoxide combined with mid-infrared wavelength modulation spectroscopy

*Chikara Hashimoto[1], Yusuke Kindaichi[1], Kenichi Tonokura[1]

1.graduate school of frontier sciences

Carbon monoxide (CO) is emitted from incomplete combustion of fossil fuels and biomass. It affects the concentration of CO_2 and CH_4 through the reactions with hydroxyl radicals. The major method of the measurements of CO is NDIR. This method is highly sensitive and highly stable. There are a lot of data of the concentration of CO measured with NDIR, but they show only hourly average. In this study, we developed a mid-IR laser absorption spectrometer that uses a 4.57 μm quantum cascade laser with wavelength modulation spectroscopy (WMS).
The absorption line for the measurements of CO was at 2190.02 cm^{-1}. The laser was scanned at 1 Hz. The beam was collimated with CaF_2 lens and introduced into the cell. The optical path length was 29.91 m. The signal from a photodetector was processed by the lock-in amplifier.
To assess signal stability and detection limit, 1.02 ppm CO gas was introduced into the WMS system. From the Allan variance plot, an optimum integration time of 145 s was derived. Averaging 145 times, the precision (1σ) of the measurement was 6.5 %.
For the measurement of the detection limit of this instrument, we collected the signal of different CO mixing ratio and made the calibration curve. From the slope and the averaged baseline deviation, the detection limit (SNR = 2) was found to be 0.094 ppm.
Measurements of outside air were conducted on the Kashiwa campus of The University of Tokyo over October 24-27, 2015.During the measurements of ambient air, the calibration was performed every 6-9 hours.
Outside air measurements were conducted over October 24-27, 2015. The CO mixing ratio during the measurements ranged from 0.097 ppm to 1.8 ppm. On the night of October 24, the concentration of CO increased continuously. The wind brew from south-southwest. In that direction, there is Keihin industrial area, so the emission from the factories in the area may have contributed to the CO mixing ratio increase. On October 25 the wind brew from northwest throughout the day. There are no big industrial areas in that direction. Car entrance was restricted because the campus was opened to the public. These may be the reasons the concentration of CO was steady and low.
We performed in situ measurements of CO mixing ratios in ambient air with a near-IR laser absorption spectrometer using WMS. We successfully detected CO mixing ratio change in the ambient air instantly.

©2016. Japan Geoscience Union. All Right Reserved.

Direct measurement of photochemical ozone production rate at the Field Museum Tamakyuryo in summer 2015

*Yasuhiro Sadanaga[1], Shio Kawasaki[1], Hiroshi Tsurumaru[2,3], Ramasamy Sathiyamurthi[2], Yosuke Sakamoto[2], Ito Kensuke[2], Fujii Tomihide[2], Shungo Kato[4], Tomoki Nakayama[5], Yutaka Matsumi[5], yoshihiro nakashima[6], Kazuhide Matsuda[6], Yoshizumi Kajii[2,7]

1.Osaka Prefecture University, 2.Kyoto University, 3.Now at Kanazawa University, 4.Tokyo Metropolitan University, 5.Nagoya University, 6.Tokyo University of Agriculture and Technology, 7.National Institute for Environmental Studies

Photochemical production processes of ozone are non-linear with respect to concentrations of ozone precursors such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs). In addition, ozone concentration variations are influenced by meteorological factors such as transport and deposition as well as photochemistry. It is useful if the meteorological and photochemical factors can be divided to discuss ozone budgets. In order to discuss only “photochemical” factors for ozone production, a direct measurement system of a photochemical ozone production rate has been recently developed. Actually, this system measures oxidant ($Ox = O_3 + NO_2$) production rate (P-L(Ox)). The use of Ox can ignore the concentration variations of O_3 by the reaction of O_3 with NO.
Details of a P-L(Ox) were described in JPGU Meeting 2015, Briefly, this system has “reaction” and “reference” chambers. The reaction and reference chambers are made of quartz and Pyrex, respectively. In addition, an outer wall of the reference chamber is coated with a UV-cut film. Both the chambers were put in an outside to be exposed directly to sunlight. Ambient air is introduced into both the chambers. Photochemical reactions proceed to generate Ox in the reaction chamber while Ox is not generated in the reference chamber. The difference of Ox concentrations (Δ Ox) in air from the two chambers is the photochemical net Ox production in the reaction chamber. The P-L(Ox) is obtained by dividing ΔOx by a mean residence time of air in the reaction chamber. Ox concentrations were obtained as follows. Ozone in Ox is converted into NO_2 by the reaction of O_3 with large excess of NO, and then the NO_2 concentration is measured by a cavity attenuated phase shift spectroscopy technique.
The field campaign was performed at the Field Museum Tamakyuryo (FM Tama) of the Tokyo University of Agriculture and Technology from July 20 to August 10, 2015. Total OH reactivity, photolysis frequencies (e.g. $J(O^1D)$), meteorological parameters, O_3, CO, NO_x, VOCs concentrations, and so on as well as P-L(Ox) were observed simultaneously. In this presentation, the main factor of ozone concentration increases (photochemical or meteorological factor), and relationship among P-L(Ox), $J(O^1D)$ and concentrations of ozone precursors were discussed. In addition, P-L(Ox) variations by addition of NO or isoprene into this instrument were investigated on August 3 and 7, and the results of this experiment will also be briefly reported.

Keywords: Ozone, Photocheimcal production rate, Nitrogen oxides, Volatile organic compounds

©2016. Japan Geoscience Union. All Right Reserved.

Intercomparisons of water vapor vertical column amount and vertical distribution observed by ground-based remote sensing techniques

*Satoshi Onuma[1], Hitoshi Irie[2], Pradeep Khatri[2]

1.Graduate School of Advanced Integration Science, Chiba University, 2.Center for Environmental Remote Sensing, Chiba University

Water vapor plays a key role in determining the Earth's climate, accounting for about 60% of natural greenhouse effects. Ground-based remote sensing techniques are thought to be useful for regular continuous observations of water vapor vertical column amount (Precipitable Water Content; PWC) and vertical distribution in the troposphere. However, their quantitative evaluation is limited. Under such circumstances, simultaneous observations using three ground-based microwave radiometers (WVR1125, MP1502, MP1504) were conducted on December 22-31, 2015. In addition, simultaneous observations using a set of four Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) instruments directing toward different azimuth angles (called 4AZ-MAXDOAS) and a CIMEL sunphotometer of NASA AERONET were also conducted. First, we focused on seven days from December 25 to 31, when the weather was relatively stable, to estimate PWC differences using WVR1125 as a standard. Comparisons between the three microwave radiometers showed relatively large systematic differences by about 20%, due to uncertainty in calibration constant. On the other hand, the differences of CIMEL and 4AZ-MAXDOAS from WVR1125 were as small as below 10%. Based on these results, we analyzed data taken on December 22-24 and found that large differences, which cannot be explained by uncertainty in the calibration constant, occurred during and after the rain. This was thought to be due to interferences by raindrops in air and on a radome of the microwave radiometer. This effect lasted in 12 hours after rains stopped. In addition, we found that just before rainfall, MAX-DOAS PWCs were much smaller than those of microwave radiometer. This is suggested to be due to advection of water-rich air into heights (7-8 km), to which the sensitivity of MAX-DOAS observation is weak.

Keywords: Water vapor, microwave radiometer, MAX-DOAS, CIMEL

©2016. Japan Geoscience Union. All Right Reserved.

Causes leading to enhancements in sulfur dioxide concentration observed by MAX-DOAS in Kyusyu

*Takaya Okuzaki[1], Hitoshi Irie[1]

1.Chiba University

We performed continuous observations of sulfur dioxide (SO_2) using the Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) at Kasuga in Kyushu, Japan. Spectra measured at 310-320 nm were analyzed with the DOAS method to retrieve mean SO_2 concentrations for a 0-1 km layer with a horizontal scale of about 10 km. This spatial scale is expected to provide useful inputs for data assimilation. This study focuses on continuous data acquired in July-August 2014. In the period, seven days were identified as the SO_2 level was high (>5 ppbv). Back trajectory analysis indicates that there were two categories; the one from a volcanic origin and the other from a continental origin. A high SO_2 plume observed on July 8 was found to originate from Sakurajima volcano. Plumes with SO_2 higher than 10 ppbv observed on July 30 and August 7 and 13 were from the Aso volcano. High SO_2 observed on August 14 could be affected by both volcanoes. On the other hand, high SO_2 plumes observed on July 12 did not pass over volcanoes but over China for about 1 day. Very high SO_2 of 20 ppbv was observed on July 21. This is suggestted to be due to a rapid transport of high SO_2 plumes from South Korea.

Keywords: SO2, MAX-DOAS, valcano

©2016. Japan Geoscience Union. All Right Reserved.

Development of photovoltaic-driven MAX-DOAS system (Eco-MAXDOAS)

*Hitoshi Irie[1], Tomomichi Kato[1]

1.Chiba University

It is recognized that the Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) technique is suitable for routine observations of atmospheric constituents as its setup is simple, power consumption is low, and fully automated long-term operation without absolute radiometric calibration is possible. In addition, it is also a powerful technique with abilities enabling simultaneous measurements of aerosols and their gaseous precursors, such as nitrogen dioxide (NO_2) and sulfur dioxide (SO_2). To exploit its further applications, we are developing the photovoltaic-driven MAX-DOAS system, called Eco-MAXDOAS. By utilizing the Eco-MAXDOAS, we expect to make multi-component air quality observations possible in areas with problems on stable power supply. For this development, we removed a temperature controller to reduce the power consumption. Instead, a shutter was introduced just before the entrance slit of the spectrometer. With this modification, it was made possible to take dark count measurements more often than for the normal MAX-DOAS instrument by closing the shutter between observations at different elevation angles. We tested the Eco-MAXDOAS and found that the spectrometer temperature varied by less than ±0.3 degrees for 3 minutes interval of changing elevation angles. Using dark count data taken before and after scattered sun light observations, the SNR was estimated to be about 10,000. This supports that analysis for a differential absorption as small as 10^{-4} (0.01%) is possible. In addition, we compared the aerosol and NO2 data retrieved from Eco-MAXDOAS and MAX-DOAS observations, and we confirmed that the two retrieved values were usually almost the same but occasionally showed significant differences. To investigate the cause, we devised the 4AZ-MAXDOAS system, a set of 4 MAX-DOAS instruments directed toward different azimuth angels of north, south, east, and west. The observation with the 4AZ-MAXDOAS confirmed that the data can show significant differences depending on the azimuth angle. We concluded that the differences were caused by the spatial inhomogeneity of atmospheric components, supporting the consistency between Eco-MAXDOAS and MAX-DOAS data.

Keywords: MAX-DOAS, PV, NO2, HCHO

©2016. Japan Geoscience Union. All Right Reserved.

Development of a particle production method for calibration of a nanoparticle composition analyzer

*Koichi Murakami[1], Hiroki Otake[1], Nobuyuki Takegawa[1]

1.Graduate School of Science and Engineering, Tokyo Metropolitan University

1. Introduction

Atmospheric aerosols play an important role in global climate change and air pollution. Online measurements of chemical composition of ultrafine particles (nanoparticles) is important for better understanding of the formation processes of aerosols. Production of well-defined test aerosol particles is necessary to evaluate the performance of an online instrument (e.g., aerosol mass spectrometry). The purpose of this study is to develop a system for the production of test particles and to understand factors affecting the size distributions of test particles for stable operation and control.

2. Experimental Apparatus

The particle production method is based on homogeneous nucleation and subsequent growth of nuclei using oleic acid as a condensable material. The benefits of using oleic acid include: (1) it has moderately low saturation vapor pressure at room temperature, and evaporative loss can be neglected after particles are formed; (2) oleic acid particles are in liquid-phase and thus bounce of particles upon high velocity impact is negligible; and (3) it is a non-toxic material and easy to use. The equipment consists of three main sections; an evaporator of oleic acid, a condenser tube, and a size distribution analyzer. The evaporator produces oleic acid vapor by heating a liquid reservoir. In the condenser tube, homogeneous nucleation takes place by supersaturation of oleic acid vapor due to large temperature difference between the evaporator and condenser. The size distributions of formed nanoparticles are measured using a scanning mobility particle sizer (SMPS). The dependence of the size distribution on the evaporator temperature and flow rate was measured with and without an activated carbon denuder. The flow rate affects the saturation ratio, residence time in the tube, and flux of oleic acid vapor transported downstream. The activated carbon denuder affects the concentration of condensable vapor in the tube.

3. Results

Key parameters of the particle size distribution (peak diameter, width, and number of modes) showed complex dependence on the evaporator temperature, air flow rate, and presence of the activated carbon denuder. Specifically, the flow-rate dependence of the peak diameter with the activated carbon denuder exhibited a trend opposite to that without the denuder. An optimal size distribution for nanoparticle experiments (peak diameter of 40-50 nm) was obtained at a relatively higher flow rate in the presence of the activated carbon denuder. Possible mechanisms of the change in the size distributions are discussed based on simple theoretical calculations.

Keywords: Aerosol, Nanoparticle

©2016. Japan Geoscience Union. All Right Reserved.

New particle formation in different atmospheric environments: Comparison of Kyoto and Tokyo-Tama observations

*Yuki Kuruma[1], Tomoki Nakayama[1], Yutaka Matsumi[1], Hiroshi Tsurumaru[2], Ramasamy Sathiyamurthi[2], Yosuke Sakamoto[2], Manabu Irie[2], Akira Ida[2], Shungo Kato[3], yoshihiro nakashima[4], Kazuhide Matsuda[4], Yoshizumi Kajii[2,5]

1.Nagoya University, 2.Kyoto University, 3.Tokyo Metropolitan University, 4.Tokyo University of Agriculture and Technology, 5.National Institute for Environmental Studies

New particle formation (NPF) is an important source of atmospheric aerosol particles, and may influence to regional/global climate and air quality through a variety of physical and chemical processes. Therefore, understanding of these processes is important. The NPF events have been observed in many different atmospheric environments (e.g., urban, forest, and mountains) and laboratory experiments. But their formation mechanisms are still poorly understood. In this study, field observations of aerosol particle number distributions and gaseous precursors were conducted at two sites: one in an urban area in Kyoto city, and the other in an observational site of Field Museum Tamakyuryo (FM Tama) of the Tokyo University of Agriculture and Technology in Tokyo, in summer to investigate factors controlling NPF.
Observations were performed at the Yoshida campus of Kyoto University, Kyoto (between 19 August and 11 September, 2013), and at the FM Tama, Tokyo (between 24 July and 8 August, 2015). Particle size distributions were measured with a scanning mobility sizer (SMPS; TSI) in both sites. Volatile organic compounds (VOCs) concentrations were measured using a proton transfer reaction mass spectrometer (PTR-MS; IONICON). Other gaseous components such as sulfur dioxide (SO_2) and ozone (O_3) were also measured simultaneously. In this study, we categorized the observed NPF events as a burst of nucleation mode particle (below 30 nm particle diameter) with (Case 1) or without (Case 2) subsequent particle growth, based on Maso *et al.*[1)].
The NPF events corresponding to increases in SO_2 or VOC concentrations were observed at least 7 times during the observation period at Kyoto. On the other hand, no NPF events were observed in the 16 observation days at FM Tama. Although no significant differences in total particle surfaces and SO_2 concentrations in two sites were observed, isoprene concentrations in FM Tama were significantly higher than monoterpenes concentrations during daytime. In this presentation, differences in the particle size distributions and concentrations of possible precursor gaseous between NPF and non-NPF events will be discussed.
References
1) M. Dal Maso, M. Kulmala, I. Rippinen, R. Wagner, T. Hussein, P. P. Aalto, K. E. J. Lehtinen: *Boreal Env. Res.*, 10, 323 (2005).

Keywords: new particle formation, aerosol particle number distribution, sulfur dioxide, volatile organic compounds

©2016. Japan Geoscience Union. All Right Reserved.

Observation of new particle formation in Noto peninsula

*Hiroyuki Hyono[1], Atsushi Matsuki[2], Kento Kinouchi[1]

1.Graduate School of Natural Science & Technology Kanazawa University, 2.Institute of Nature and Environmental Technology, Kanazawa University

Introduction
The process by which new particles (Ultrafine particles having diameter of the range from several nm to several tens of nm) are formed from precursor gases in the atmosphere is one of the main factors that affect the number size distribution of aerosol particles. Since new particle formation is a first step for the aerosol particles to take shape and eventually act as Cloud Condensation Nuclei (CCN), it has important implications in understanding contributions of aerosols on the climate. Recently, studies in various observation sites have pointed out conditions favorable for the new particle formation (NPF). For example, if there are a lot of existing particles in atmosphere, NPF is less likely to occur (Kerminen et al.,2001), because gas molecule preferentially condense to existing particle. However, there have been few reports so far on the seasonality of NPF in coastal East Asia.
Method
We selected Noto Ground-based Research Observatory (NOTOGRO), located at the tip of the Noto Peninsula as observation site mainly for the following reasons. Firstly, the station is away from any densely populated areas such as Kanazawa city, where one can expect less anthropogenic effect from nearby cities. Secondly, we may be able to observe influence caused by the unique monsoon facing the coast of the Japan Sea. Number size distributions of aerosol particles were measured using a scanning mobility particle sizer (SMPS) system that consists of a differential mobility analyzer (DMA, Model 3081) and a condensation particle counter (CPC, Model 3776). The field measurement was conducted from September 2012 to October 2014. In addition, SO_2 gas was continuously measured using (Model, 43B SO_2 Analyzer), from which we calculated H_2SO_4 proxy (Petäjä et al.,2009).
Result&Discussion
Analysis of aerosol number size distributions confirmed that NPF events occurred frequently throughout the year. From two years of observation, some similarities were found in the seasonality of the occurrence frequency. We focused on the variation of SO_2 gas which is considered as the main precursor of aerosol particles. Comparison with the number size distributions revealed that, the values of the H_2SO_4 proxy were high in most cases when NPF events occurred. The daytime maximum values of the H_2SO_4 proxy were extracted and compared among the days with and without NPF. As a result, the values of H_2SO_4 proxy were relatively higher when the event occurred. These results suggested that the conditions favorable for the formation of sulfuric acid vapor are the major factors controlling the NPF in the studied region.

Keywords: aerosol, new particle formation

©2016. Japan Geoscience Union. All Right Reserved.

Measurement of ambient particles using a newly developed polar nephelometer -Relationship between scattering angular distributions and chemical compositions-

*Maho Nakagawa[1], Tomoki Nakayama[1], Hiroshi Sasago[1], Yuki Kuruma[1], Hikari Yai[2], Shuhei Ogawa[2], Yange Deng[2], Michihiro Mochida[2], Yutaka Matsumi[1]

1. Graduate School of Science and Institute for Space-Earth Environmental Research, Nagoya University, 2.Graduate School of Environmental Studies, Nagoya University

Angular distribution of scattering is one of the important optical properties contributing to the radiation balance in the Earth's atmosphere. Therefore, accurate description of the single-scattering properties of aerosol particles is required. The angular distribution of light scattering is an important optical property. In addition, because the angular distribution of scattering depend on size, shape, and refractive index of particles, the angular distribution for individual aerosol particles may provide useful information to estimate mixing state and type of particles. In the present study, applicability of the angular distribution for individual particles to the estimation of mixing state and type of particles was examined by comparing the temporal variations of the angular distributions with those of chemical compositions for ambient particles.
We have developed a new polar nephelometer, which can measure angular distribution of the optical light scattered by an individual particle.[1)] Laser beam at 532 nm from a 300 mW YAG laser was used as light source. The laser beam intersects with a stream of aerosol particles introduced with a sheath flow using a double pipe. There are 21 photodiode detectors arrayed in each plane, totaling 42. Detector apertures were placed to limit sensing angles and minimize background light scattered from walls. In this system, the angular distributions for an incident light polarized parallel and perpendicular to the scattering plane can be measured simultaneously.
Ambient particles were measured at the Higashiyama campus of Nagoya University (35°09¢N, 136°58¢E, 60 m above sea level) located in an urban area in Nagoya, Japan, from July 1st to 7th, 2015. After being dried by a diffusion dryer with silica gel, ambient particles were introduced alternately to a heated line (maintained at 300°C) or the bypass line to measure, respectively, ambient particles directly and after the evaporation of volatile materials under high temperature conditions. The lines were switched every 15 min using two-way ball valves. Then, the particles were passed through a differential mobility analyzer (DMA) and introduced into the polar nephelometer. The DMA voltage was switched to select the nearly monodispersed particle with a mobility diameter 300 or 500 nm every 30 min. Most of organics, sulfate, nitrate, and ammonium are expected to be vaporized at temperatures below 300°C.[2)] Therefore, black carbon (BC) particles are expected to be mainly measured when particles were passed through the heated line. During the observation, mass concentrations of BC were measured using a particle soot absorption photometer (PSAP) combined with a heated inlet (maintained at 300°C). The mass concentrations organics, sulfate, nitrate, and ammonium were measured using a time-of-flight aerosol mass spectrometer (AMS). In the presentation, relationship between the temporal variations of scattering angular distributions of individual particles and those of chemical compositions will be discussed.

Reference

1) Nakagawa, M., Nakayama. T., Sasago, H., Ueda, S., Venables. D. S., Matsumi, Y., Design and Characterization of a Novel Single-Particle Polar Nephelometer, *Aerosol Sci. Technol.* (in press).
2) Nakayama, T., Ikeda, Y., Sawada, Y., Setoguchi, Y., Ogawa, S., Kawana, K., Mochida, M., Ikemori, F., Matsumoto, K., Matsumi, Y., Properties of light-absorbing aerosols in the Nagoya urban area, Japan, in August 2011 and January 2012: Contributions of brown carbon and lensing effect, *J. Geophys. Res. Atmos.*, 119, 12721-12739 (2014).

©2016. Japan Geoscience Union. All Right Reserved.

Keywords: scattering angular distribution, polar nephelometer, chemical compositions

©2016. Japan Geoscience Union. All Right Reserved.

Analysis of the Mixing State of Airborne Particles using a Tandem Combination of Laser-induced Fluorescence and Incandescence Techniques

*Fumikazu Taketani[1], Yugo Kanaya[1], Takayuki Nakamura[2], Naoki Takeda[2], Hiroyuki Koizumi[2], Takuma Miyakawa[1,3], Xiaole Pan[1], Moteki Nobuhiro[3], Noriyuki Hirayama[2], Nobuyuki Takegawa[3,4]

1.Japan Agency for Marine-Earth Science and Technology, 2.Fuji Electric Co. Ltd., 3.University of Tokyo , 4.Tokyo Metropolitan University

We have developed a novel system for real-time measurement of the mixing state of aerosol particles using a tandem combination of laser-induced fluorescence (LIF) and incandescence (LII) techniques. The tandem analysis system comprises two chambers connected in series; particles are analyzed with LIF in the first chamber and LII in the second chamber. We analyzed identical particles using the two methods as judged by the time intervals of detection in the two chambers. This system provides information on the mixing state of fluorescent compounds and black carbon in single particles. Ground-based measurements of ambient particles were performed in Tokyo during October 26–29, 2012. We analyzed 43,881 particles with optical diameters greater than 0.4 mm. The fractions of particles with fluorescent composition, black carbon, and both were 14.2%, 2.3%, and 0.3%, respectively, which indicates the presence of internal mixtures of black carbon and fluorescent species in the ambient air for the first time. Mixtures of biological materials (estimated from fluorescence patterns) and black carbon were also detected. The fluorescence patterns of single particles with and without black carbon were almost identical, suggesting that particles with both black carbon and fluorescent composition might be formed by aggregation in ambient air.

Keywords: Black carbon, Fluorescent particles, Mixing state

©2016. Japan Geoscience Union. All Right Reserved.

Trend analyses of aerosol optical thickness over Japan using long-term remote sensing observations and their evaluation

*Takehiro Hatakeyama[1], Hitoshi Irie[2], Pradeep Khatri[2]

1.Graduate School of Advanced Integration Science, Chiba University, 2.Center for Environmental Remote Sensing, Chiba University

Aerosols in the atmosphere are an important factor affecting the Earth's climate. However, their formation mechanism is complex, and especially in East Asia, rapid changes in emissions of their precursor gases are thought to be occurring in recent years, so that annual trends in aerosol levels have been poorly understood. In this study, we examined the trends in aerosol optical thickness (AOT) at 500nm over Japan using sky radiometer data acquired over a long time period at four sites in Japan (Chiba, Fukuejima Island, Cape Hedo, Miyakojima Island) under the framework of the SKYNET international ground-based observation network. To conduct quantitative trend analyses, sky radiometer measurements were evaluated by simultaneous observations with a CIMEL Sun photometer of NASA/AERONET and a PFR radiometer of WMO/GAW at Chiba in November-December 2015. The mean differences between the CIMEL sun photometer and sky radiometer and between the PFR radiometer and sky radiometer were 0.003 ±0.006 and -0.007±0.004, respectively. Based on these results, we analyzed the annual trends of AOT at the four sites in Japan and found that AOT decreased at a rate of about (0.005-0.02)/year (2-6%/year) on average in recent years. These trends were found to be consistent with those estimated from MODIS data. To understand the cause of the observed decreasing trends, an additional analysis was conducted using Angstrom exponent data. AOTs for smaller aerosol particles exhibited similar decreasing trends, suggesting that at least at Chiba the observed decreasing trends were due to reduction in domestic anthropogenic aerosols. On the other hand, analysis of MODIS AOT data over China showed decreasing trends, except for a limited area around the Shandong province, for 2009-2015. In this presentation, we will discuss in detail potential factors causing decreasing trends in AOT over Japan, with a consideration of impacts by such aerosol variations in China.

Keywords: Aerosol, Sky radiometer, CIMEL, PFR, MODIS

©2016. Japan Geoscience Union. All Right Reserved.

Study of aerosols and clouds using sky radiometer of SKYNET

*Pradeep Khatri[1], Hitoshi Irie[1], Tamio Takamura[1]

1.Center for Environmental Remote Sensing, Chiba University

Long-term observations of aerosol chararacteristics over four typical SKYNET sites (Chiba, Fukuejima, Miyakojima, and Hedo) within Japan are analyzed to clarify the seasonal dependent characteristics of aerosols of different origins and their impacts on atmospheric heat budget. We found optically thicker aerosols with significant amount of light absorbing coarse-mode particles in the spring season. Such light-absorption phenomenon of coarse-mode aerosols is found to be the effect of not only mixed light-absorbing aerosols such as black-carbon, but also due to the effect of aerosol size. The aerosol radiative forcings at the surface and top of the atmosphere in the spring season can be roughly two times of the values in the winter season. We further present our new retrieval method of cloud parameters from sky radiometer of SKYNET, and discuss the optical characteristics of clouds obtained from the sky radiometer of SKYNET and Moderate Resolution Imaging Spectroradiometer (MODIS). Our analysis suggests that MODIS cloud optical depth (COD) may be underestimated, which in turn may lead to overestimate calculated shortwave flux.

Keywords: SKYNET, aerosol, cloud

©2016. Japan Geoscience Union. All Right Reserved.

Analysis of temporal evolution of Angstrom coefficients derived from spectrometric measurements of New Year 2013 aerosols in Manila Observatory (14.67N, 121.07E)

*JohnPaolo Durana Miranda[1,2], Daniel Luis Bautista[1,2], Nofel Lagrosas[1,2]

1.Manila Observatory, 2.Ateneo de Manila University

Spectrometric measurements were done during New Year of 2013 at Manila Observatory (14.67N, 121.07E) for the purpose of characterizing optical properties of New Year aerosols. Intensity measurements from xenon lamp located ~150m from the spectrometer were gathered from 22:00 (local time) of 31 December 2012 to 09:00 (local time) of 01 January 2013, using a USB 2000 Ocean Optics Spectrometer. The sources of New Year aerosols were from burning firecrackers and firework activities from nearby residential areas. From 22:00 to 23:30, sporadic fireworks were observed and the frequency of burning and firework activities increased as the New Year approached. Data were acquired every 1 minute to look at the temporal evolution of optical characteristics of aerosols. The aerosol optical thickness were derived from 8 wavelengths: 387.3, 400.0, 440.0, 472.1, 600.0, 616.2, 764.3, and 823.1 nm. The reference intensity used to measure aerosol optical thickness was the measured intensity at 22:07 when the air could be considered as relatively clean even though isolated burning of firecrackers was observed. The aerosol optical thickness at a particular wavelength is computed from the negative of the logarithm of the ratio of the intensity and the reference intensity. The Angstrom coefficient, which is a measure of the dominance of fine or coarse particles, is obtained from the slope of the line fitting the logarithm of optical thickness and the logarithm of wavelength. Higher (>2) and lower (<1) Angstrom coefficients correspond to the dominance of fine and coarse particles, respectively. Figure 1 shows the Angstrom coefficient values from 23:30 of 31 December 2012 to 02:30 of 01 January 2013. The computed Angstrom coefficients before 23:30 and after 03:00 were not valid because there are no clear differences between the measured intensities and the reference intensity. From 23:30 to 1:00, Angstrom coefficient values noticeably increased and this could be attributed to the increase in firework activities near and just after New Year. The maximum intensity of firework activities is at around midnight. The spike of Angstrom coefficient at 00:30 can be attributed to aerosols transported to the place of measurement from a source where intense firework activities at midnight happened. The visibility at 550nm increased up to 5 km and stayed constant until 12:30. After that, the visibility started to decrease gradually up to 1 km until 01:45. The Angstrom coefficient is roughly constant at a value of 1.5 from 01:00 to 01:30 even though the visibility started decreasing to 1 km. From 01:30 to 02:30, a gradual decrease of Angstrom coefficient up to ~1.0 is observed. This can be attributed to increasing relative humidity in this time interval. Increase in relative humidity can bring about an increase in aerosol size by coagulation (Seinfeld and Pandis, 2006). This suppresses and increases the number of fine and coarse particles in the atmosphere, respectively. This would imply that aerosols would take less space to block light and this is manifested in the increase in visibility (up to 2.5 km) during this time interval. Even though fine particles were generated during the firework activities, the measured Angstrom coefficient values are still relatively low indicating a possible bimodal size distribution of aerosols in the atmosphere (Schuster et al, 2006).

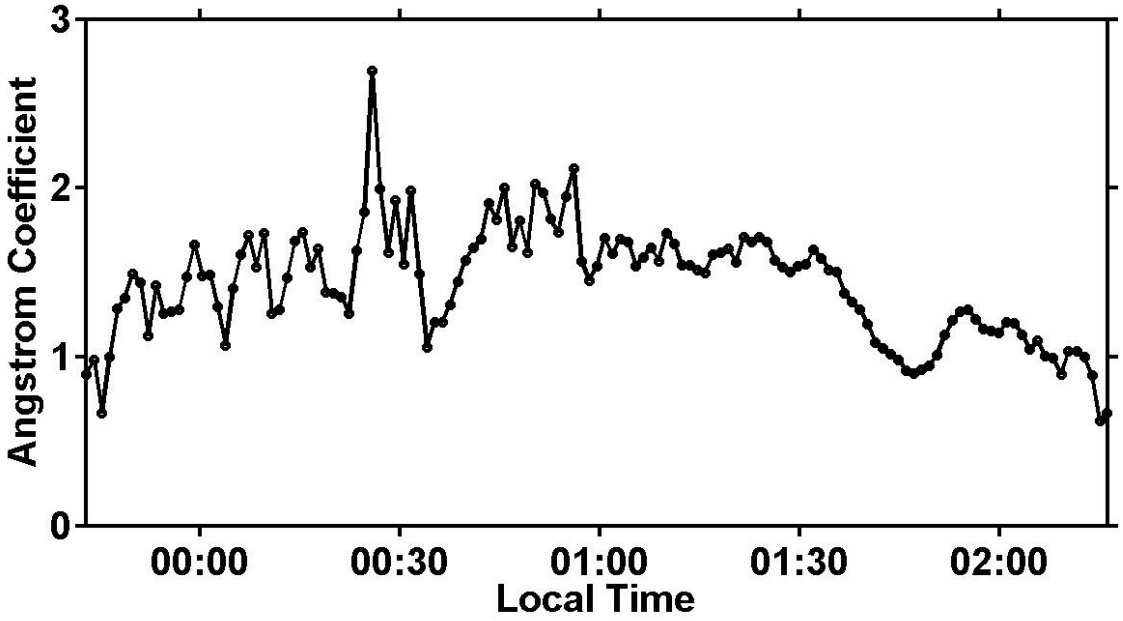
References:

Seinfeld, J. and Pandis, S. (2006). *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change 2nd ed.* Wiley-Interscience, USA.

Schuster, G., Dubovik, O., Holben, B. (2006). *Angstrom exponent and bimodal aerosol size distributions.* J. Geophys. Res, 111, D7.

©2016. Japan Geoscience Union. All Right Reserved.

Keywords: Angstrom coefficient, Aerosols, Optical thickness, Relative Humidity



©2016. Japan Geoscience Union. All Right Reserved.

Derived volume size distribution function from 2012 New Year spectrometric measurements in Manila Observatory (14.64N, 121.07E)

*Daniel Luis Bautista[1,2], John Paolo Miranda[1,2], Nofel Lagrosas[1,2]

1.Manila Observatory, 2.Ateneo de Manila University

Optical properties of New Year aerosols are characterized by spectrometric measurements performed in Manila Observatory during the 2012 New Year celebration. Data were obtained using an Ocean Optics USB2000 spectrometer from 20:00 (local time) of 31 December 2011 to 05:00 of 01 January 2012. Local time is 8 hours ahead of coordinated universal time (UTC). A xenon lamp was used as light source and was located ~150m from the spectrometer. Sources of these aerosols came from fireworks and burning of firecrackers from residential areas near Manila Observatory. Between 20:00 and 23:30, sporadic fireworks and firecracker burning were observed. The intensity and frequency of firework activities increased as midnight approached. Data were collected every 20s to look at temporal evolution of aerosol optical properties during this time interval. In this study we derive the aerosol volume size distribution function using the parametric inversion method of Kaijser (1983). In this method, we selected 8 wavelengths (387.30, 400.00, 440.00, 470.30, 500.30, 530.00, 550.10, 600.00nm) to derive the volume size distribution functions. Optical depths from these wavelengths were derived using a reference intensity obtained at 20:00 (local time) of 31 December 2011 when the air was considered relatively clean even with the occurrence of sporadic firework activities. A least-square minimization process was implemented between the measured optical depths and computed optical depths using Mie theory and assuming a 33 bimodal log-normal distribution functions with geometric mean radii between 0.003 to 1.2um and standard deviation of 2. The result of the least-square minimization process outputs the coefficients of the log-normal volume size distribution functions. This allowed the log-normal volume size distribution functions of the aerosols to be determined and plotted to indicate changes over time. Fig. 1 shows the temporal development of derived aerosol volume size distribution from 20:00 (local time) of 31 December 2011 to 05:00 of 01 January 2012. From 20:00 to 22:00, the aerosols exhibit a unimodal volume size distribution centered at 0.3um. This can be attributed to background urban aerosols, i.e., without any contamination from aerosols contributed by firecrackers and fireworks. From 22:00 to 00:30, small size aerosols with radius 0.04um started to emerge due to increasing firework activities which intensified until 00:00 creating a bimodal volume size distribution. From 00:00 to 00:30, urban background aerosols have developed into larger aerosols of radius 0.8um due to high relative humidity. From 00:30 to 03:00, the intensities measured by the spectrometer were very low because of low visibility conditions. During this time period, there was an abundance of smoke lingering in the atmosphere as a result of the firework activities and relatively low wind speed conditions. The measured low light intensities from the xenon lamp caused the inversion process to fail for the data set during this time interval. At 03:00 up to 04:30, when the visibility slightly improved, the data obtained by the spectrometer provided sufficiently high light intensities for the inversion process to succeed and yield a relatively constant mean radii for both modes, indicating constant relative humidity. From 04:30 to 05:00 the volume size distribution decreases significantly suggesting a sudden clearing of the atmosphere. With these observations, we have shown the possibility of obtaining the aerosol volume size distribution functions under heavy aerosol loading in the atmosphere. In the future, we intend to improve this method to show aerosol growth under increasing relative humidity conditions.

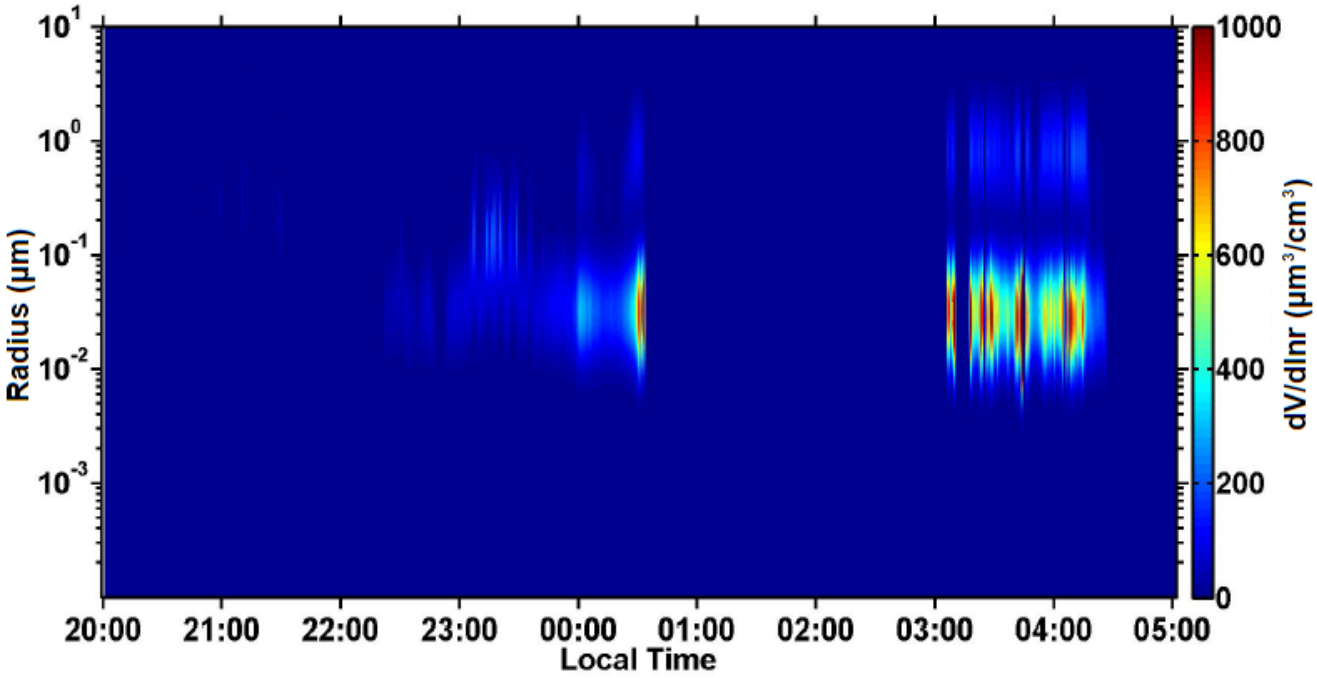
Reference:

Kaijser, T. (1983). A simple inversion method for determining aerosol size distributions. *J. Comp.*

©2016. Japan Geoscience Union. All Right Reserved.

Phys., 52, 80-104.

Keywords: Volume size distribution, Aerosols, Parametric inversion, Spectrometer



©2016. Japan Geoscience Union. All Right Reserved.