Data composite of airborne sulfur dioxide measurements in the upper troposphere

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Sulfur dioxide (SO_2) is a key aerosol precursor gas, however, only during recent years high-precision in-situ data could be obtained for the upper troposphere (UT) using airborne chemical ionization mass spectrometry. Data summaries of SO_2 will be presented from a large number of campaigns performed with the research aircraft Falcon and HALO during the years 2005 to 2015 covering a geographical region from 83°N to 65°S and 105°W to 135°E and altitudes up to 15 km. The SO_2 data were gridded onto a 5° latitude by 5° longitude horizontal grid with a 1-km vertical resolution. The data composites provide information about the SO_2 distribution at mid-latitudes, tropical and polar regions. Median SO_2 background mixing ratios in the UT range between 10-25 ppt, however, also areas with strongly enhanced SO_2 mixing ratios in the UT were observed, associated with events of new particle formation. The SO_2 sources and transport pathways for these specific observations will be discussed.

Keywords: sulfur dioxide, aircraft measurements, new particle formation

Temporal characteristics of CH_4 vertical profiles observed over Surgut (Novosibirsk) from 1993 (1997) to 2014 in West Siberian Lowland

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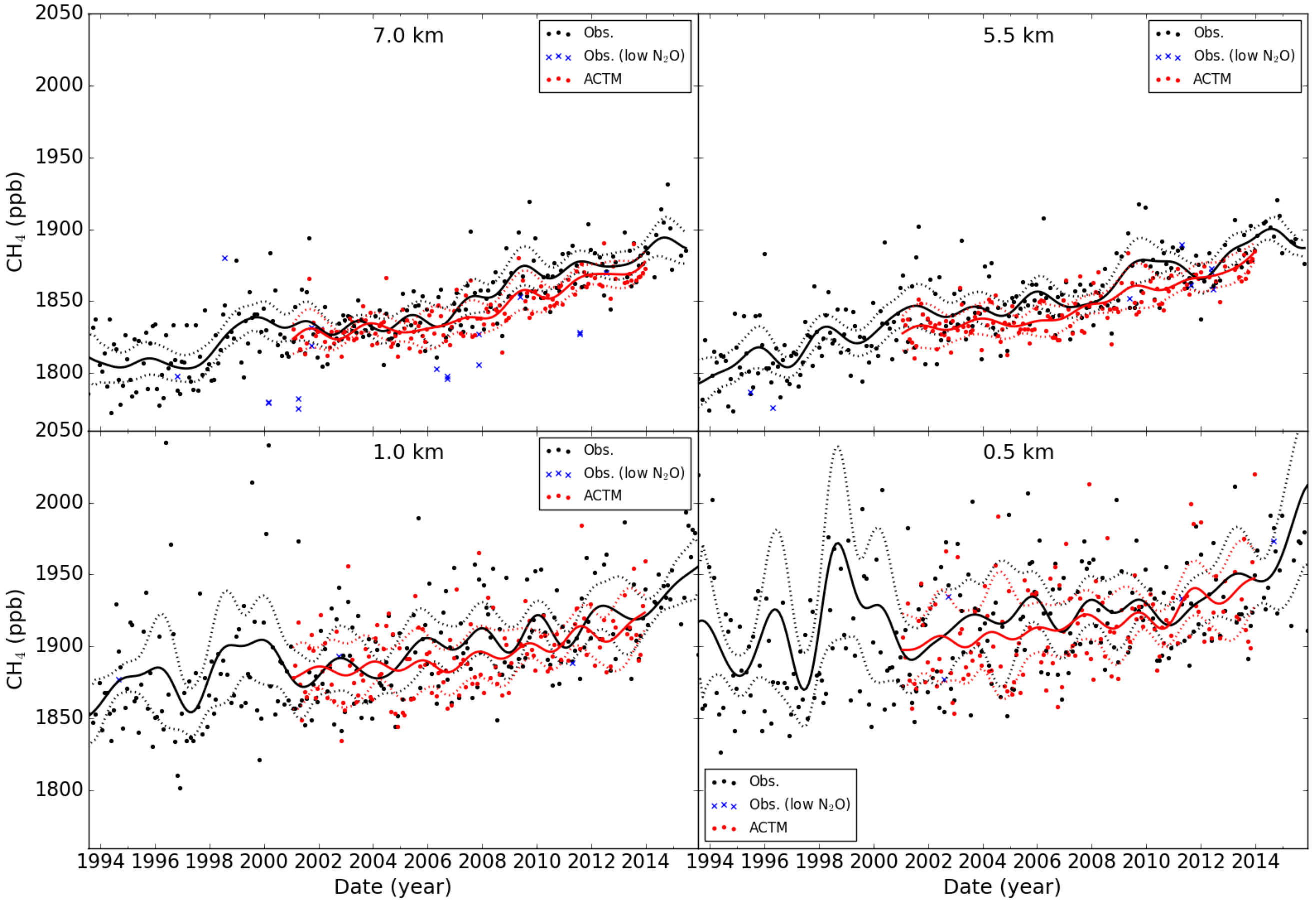
We have carried out monthly flask sampling with aircraft over the boreal wetlands in Surgut and a pine forest nearby Novosibirsk, both located in West Siberian Lowland (WSL). Vertical profiles of methane (CH_4) concentration were measured up to about 7 km since 1993 at Surgut and 1997 at Novosibirsk. Temporal variation at each altitude of both sites exhibited increasing trend with stagnation during 2000-2006 as observed globally from ground-based measurement network (Figure). In addition to winter maximum as seen at other remote sites in northern mid to high latitudes, a maximum was observed in summer, particularly in lower altitudes over the WSL. Large CH_4 emissions from the wetlands in the WSL likely have exceeded zonal-mean CH_4 loss by reaction with OH radical during summer, leading to the summer maximum. Methane emissions from different parts of the Earth's surface, atmospheric transport, and chemical loss produce the vertical CH_4 gradient and its variability. The vertical gradient at Surgut has been decreasing; the annual mean CH_4 difference between 5.5 km and 1.0 km decreased from 63±9 ppb during 1994-2000 to 37±8 ppb during 2009-2013. On the other hand, no clear decline in CH_4 vertical gradient appeared at Novosibirsk. An atmospheric chemistry-transport model simulation captured the observed decrease in the vertical CH_4 gradient at Surgut, when CH_4 emission from Europe was decreased but increased from the regions south of Siberia, e.g., the populated Asian nations and tropical land. At Novosibirsk, the influence of the European emission was relatively small. Our results also suggest that the regional emissions around the WSL did not change significantly over the period of our observations. Long-term monitoring of CH_4 vertical profile over West Siberia enabled us to detect variation in its emissions from the WSL and the surrounding land regions.

Keywords: Methane, Aircraft observation, West Siberian Lowland


7.0 km
5.5 km
1.0 km
0.5 km
Obs.
Obs. (low N_2O)
ACTM
CH_4 (ppb)
1800
1850
1900
1950
2000
2050
1994 1996 1998 2000 2002 2004 2006 2008 2010 2012 2014
Date (year)

Climatological variations of tropospheric CO_2 over the Asia-Pacific region observed by the CONTRAIL commercial airliner measurements

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We present spatial and temporal variations of tropospheric CO_2 over the Asia-Pacific region analyzed from 10 years of the CONTRAIL commercial airliner measurements. The CONTRAIL flights with the high-frequency CO_2 measurements have covered large part of the Asia-Pacific region. Here we address climatological variations of CO_2 from the boundary layer to the upper troposphere across the Asia-Pacific region toward comprehensive understanding of influence of Asian surface fluxes under the varying seasonal meteorology. Highlights of this study are summarized as follows. (1) Seasonally elevated and highly variable CO_2 is observed in East Asia to the North Pacific in spring. This is likely explained by active passage of eastward-tracking synoptic systems that sweeps the continental East Asia and uplifts the region’s CO_2 emissions up to the free troposphere. (2) The region-wide CO_2 decrease is obvious across the Asia-Pacific region and it is principally composed of CO_2 drawdowns originating in two distinct regions: boreal Eurasia and South Asia. We observed seasonally largest variability of CO_2 in the UT north of 40° N, likely due to heterogeneous spatial distributions of biospheric fluxes combined with sporadic convective vertical transport over the continent. Our observations also characterized distinct CO_2 depletion in the upper troposphere over South Asia as a result of strong South Asian biospheric uptakes and confinement in the Asian summer monsoon anticyclone. The development and decay of the anticyclone remarkably contributes to distributing CO_2 over the Asia-Pacific region. (3) As the cyclonic activity decays and the monsoon anticyclone develops from spring to summer, CONTRAIL measurements with highest data density over East Asia to the North Pacific serve as better constraints to CO_2 fluxes in East Asia in spring and those in South Asia in late summer.

Keywords: Carbon dioxide, Aircraft observation, Asian monsoon

Recent trend of atmospheric nitrous oxide isotopocules in the Northern Hemisphere

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Nitrous oxide (N_2O) is one of the increasing greenhouse gases in the troposphere and is the most important stratospheric ozone-depleting gas emitted in the 21st century. In the Northern Hemisphere, sources of atmospheric N_2O include human activity in Europe, Russia, Asia, and North America (e.g., agriculture), biomass burning (forest fires), oceans, and future climate change might cause substantial change in such sources. Isotopocule ratios of N_2O, which include not only elemental $^{15}N/^{14}N$ and $^{18}O/^{16}O$ ratios but also site-specific $^{15}N/^{14}N$ ratio in asymmetric NNO molecule, are regarded as useful parameters to infer the origin and production–consumption mechanisms of N_2O, and to estimate its global budget. Previous studies on N_2O trapped in the firn in polar ice sheet revealed the secular trend of isotopocule ratios, but there have been only a few reports on long-term monitoring of atmospheric N_2O isotopocule ratios in the Northern Hemisphere.

We have been measuring mixing ratio and isotopocule ratios of N_2O at one site in the low lattitude and two sites in the high lattitude of the Northern Hemisphere. At Hateruma, a southwestern island of Japan (24°N, 124°E), monthly air samples are collected into glass flasks at 46 m above sea level, and N_2O isotopocule analysis has been conducted since 1999. At Novosibirsk in the western Siberia, Russia (55°N, 83°E), monthly samples are collected at altitudes of 500 m and 7000 m by aircraft, and N_2O isotopocule data have been obtained since 2005. At Churchill, northern Canada (59°N, 94°W), surface air samples are collected biweekly, and N_2O isotopocule analysis has been conducted bimonthly since 2011.

Results show that the bulk nitrogen isotope ratio ($d^{15}N^{bulk}$) are decreasing at the similar rate (about $-0.04‰\ yr^{-1}$) as reported by firn-air analysis while the N_2O mixing ratio are increasing (about $0.8\ ppbv\ yr^{-1}$) at the three sites. This suggests isotopically light N_2O sources such as agriculture are still contributing to the increase in the atmospheric N_2O. Detailed analysis of the time series reveals that year-to-year variation of the mixing ratio and $d^{15}N^{bulk}$ has been enhanced since around 2010 at all the three sites, and that vertical gradient of the mixing ratio and $d^{15}N^{bulk}$ over Novosibirsk has been also variable recently. Cause of these findings will be discussed with respect to temporal change in N_2O flux and isotopic signature of surface sources and change in atmospheric circulation including troposphere-stratosphere exchange, with the aid of atmospheric model simulation.

Keywords: Greenhouse gas, Troposphere, Stable isotope analysis

Emissions of CO_2, CO, and CH_4 from peat forest fires on Sumatra Island in non El-Niño year 2013

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We observed substantial enhancements of atmospheric trace gases and aerosols (CO_2, CH_4, CO, PM2.5, PM10, and black carbon) in summer of 2013, with continuous instruments onboard the NIES voluntary observing ships sailing in the Southeast Asia region. The enhancements were observed off the east coast of the Malay Peninsula and in the straits of Malacca along the shipping route, associated with prominent enhancements in CO. The 6-year climatology (2008 –2013) of the monthly mean CO mixing ratios in these areas shows the maximum in June, followed by moderate but relatively high values in following months of July and August. Our analysis combining in-situ measurements, satellite observations, and an air trajectory analysis showed that the observed enhancements were due mainly to the intensive biomass burnings that occurred in the central Sumatra. We examined the quantitative relationships between the observed CO_2, CO, and CH_4 during the CO enhancement events. Strong correlation was found between these gases, and the calculated emission ratios of CO/CO_2 suggested large contribution of peat forest fires to the observed enhancements (CO/CO_2 = 135 ppb/ppm). We determined the emission factors (EF) of CO_2, CO, and CH_4 from the peat forest fires using the emission ratios of CO/CO_2 and CH_4/CO_2, showing the EFs of 1722, 132.0, and 6.27 (g/kg) for CO_2, CO, and CH_4, respectively. These EFs were compared with those calculated from the current emission inventory (GFED version 4S), which are 1671, 144.5, and 11.95 (g/kg) for CO_2, CO, and CH_4 respectively. These EFs were consistent with each other except CH_4, suggesting that the EF used in GFED4S is overestimated for CH_4 from peat fires.

Keywords: Greenhouse gases, Southeast Asia, Biomass burning

Individual Particle Analysis of Marine Aerosols Collected over the North Pacific and its Marginal Seas

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Atmospheric aerosol particles can play a vital role in the climate change, because they can absorb and scatter solar radiation, and form clouds by working as cloud condensation nuclei. However, scientific understanding of atmospheric aerosol behavior is not sufficient (IPCC, 2013) due to the lack of knowledge. In this study, chemical compositions and morphology of marine aerosol particles were characterized based on individual particle analysis collected over the North Pacific and its marginal seas.

Samplings of atmospheric aerosol particles were carried out during KH-13-7 cruise (the Pacific Ocean: 2013/12/11-2014/2/12), KH-14-3 Leg2 cruise (the Pacific Ocean: 2014/7/17-8/11), KS-16-8 cruise (the sea near Japan: 2016/7/5-7/13) and MR16-06 (the Bering Sea and the Arctic Ocean: 2016/8/22-10/5). Individual particles were analyzed using a transmission electron microscope and an energy dispersive X-ray spectrometer.

Sea-salt particles without apparent reactions with other components accounted for more than 80% in the samples collected over the open seas. On the other hand, one sample out of the two samples collected near Hachijo-jima was accounted about 90% by modified sea-salt particles and sulfates. This result indicates that the pollutants from the Asian continent were transported to the observed area by the transition of the Baiu front. In the presentation, we are planning to report on the samples collected over the Arctic Ocean as well.

Keywords: Atmospheric aerosol, Sea-salt, Sulfate, Nitrate, TEM, EDX

Evaluation of satellite-based tropospheric NO_2 vertical column density using MAX-DOAS observations and their integrated analysis

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Tropospheric NO_2, a major component of nitrogen oxides (NOx) emitted from anthropogenic sources (cars and power plants) and also from forest fires and lightning, is now regarded as a pollution marker detectable from space with a highest sensitivity. Temporal and spatial variations in the NO_2 density have been depicted by satellite observations and even their long-term trend is discussed. However, the tropospheric NO_2 vertical column density (TropoNO2VCD) reported from satellite observations may be biases low; they can be only half of the true value near the urban region (Kanaya et al, 2014). As such, accurate understanding of the TropoNO2VCD values derived from satellites is still poor. In this study, we re-examined the TropoNO2VCD product from OMI/Aura satellite derived with DOMINOv2 algorithm in terms of the influence from assumptions in the vertical profile and co-existing aerosol effect. Here, ground-based MAX-DOAS observations of TropoNO2VCD made at Yokosuka (35.32°N, 139.65°E) during 2007-2014 were regarded as true values and were compared with satellite values. Applying averaging kernels from satellite observations to the MAX-DOAS profiles removed most of the low bias, suggesting that the assumption in the vertical profile shape was critical. In the DOMINOv2 algorithm, NO_2 vertical profile shapes were borrowed from TM4 model simulations with a coarse spatial resolution; it tended to underestimate weight of NO_2 present near the surface in urban regions and this fact was overlooked when the tropospheric air mass factor (AMF) was determined, resulting in the low bias. When AMF was re-determined by borrowing MAX-DOAS based vertical profile shape, instead of that of TM4, the OMI-based TropoNO2VCD increased by as much as a factor of 2.2 in average and showed better agreement with MAX-DOAS observations. Similarly, the corresponding near-surface concentrations increased and showed better consistency with the values from ground-based monitoring. In summary, utilizing correct vertical profile shapes enabled reasonable estimation of TropoNO2VCD and surface-level concentrations from satellite. On the other hand, shielding effect from co-existing aerosols was found important when cloud fraction was 0.03 or lower. The tendency was consistent with the analysis results of OMI using POMINO algorithm, taking aerosol effect into account explicitly.

Keywords: air pollution, nitrogen dioxide, algorithm

Deployment of the 4AZ-MAXDOAS system at Chiba, Japan: the potential to investigate the spatial inhomogeneity of atmospheric components

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Ground-based observation data utilized for the model evaluation have been often obtained at sites, where target physical parameters, such as concentration of trace gas, are thought to be homogeneously distributed on a model grid spatial scale. However, the spatial distribution should depend on the physical parameter. More importantly, there tends to be the lack of sufficient test regarding the spatial homogeneity, particularly around sites, which are usually categorized as urban or suburban sites. Here, we present the new observation system, the 4-different-azimuth-viewing Multi-Axis Differential Optical Absorption Spectroscopy (4AZ-MAXDOAS) system. The continuous observation by the 4AZ-MAXDOAS system has been conducting in Chiba University, Chiba, Japan (35.63N, 140,10E) since November 2014, to investigate spatial distributions of atmospheric components such as nitrogen dioxides (NO_2), formaldehyde (HCHO), glyoxal (CHOCHO), and water vapor (H_2O) on a spatial scale of about 10 km. We found that daily 4AZ-MAXDOAS data for a 0-1 km layer show the significant spatial inhomogeneity, even for H_2O. However, the observed spatial inhomogeneity as well as the temporal inhomogeneity can be lowered by taking averages over longer time. We found that the differences among monthly-mean NO_2 volume mixing ratios for 4 different viewing directions were as small as ~2 ppbv. Similarly, those differences for HCHO, CHOCHO, and H_2O were estimated to be as small as ~1 ppbv, 60 pptv, and ~0.2%v, respectively. It is suggested that for measurements made at a single site, averaging data over time can lower effects of both spatial and temporal inhomogeneities on the estimate of the representative values for a grid adopted by a model and for a pixel measured by a satellite instrument.

Keywords: MAX-DOAS, NO2, HCHO, CHOCHO, H2O

Characterization of aerosols and trace gases in Phimai, Thailand using MAX-DOAS measurements.

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We present the Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) aerosol and trace gas measurements performed in Southeast Asia, at Phimai, Thailand (15.18°N,102.56° E). Our MAX-DOAS instrument has been operating there since September 2014. Aerosol and trace gas vertical profiles were retrieved using the Japanese MAX-DOAS profile retrieval algorithm version 2 (JM2), a multi-component profile retrieval algorithm based on the optimal estimation method. The components retrieved are Aerosol Extinction Coefficient (AEC) at 357 and 476 nm and 6 trace gases (NO_2, SO_2, O_3, CHOCHO, HCHO and H_2O). The MAX-DOAS data of AEC and its vertically-integrated quantity, i.e., the Aerosol Optical Depth (AOD), were compared to those of the co-located AD-Net (Asian Dust and aerosol lidar observation network) LIDAR (Light Detection and Ranging) and SKYNET sky radiometer measurements, respectively. Aerosol measurements from all the three platforms showed similar seasonal variations with enhanced aerosol loading during the dry season (October-May). This enhancement was associated with biomass burning, which is a pronounced event in this region according to literature reports. This was further supported by satellite observations (MODIS), which show high correlations with MAX-DOAS data with an R^2 of 0.71. The seasonal variation of trace gases (NO_2, SO_2, O_3, CHOCHO, and HCHO) showed similar patterns with enhanced concentrations during the dry season, consistent with the influence of biomass burning. In the dry season, the CHOCHO/HCHO ratio was estimated to be 0.025±0.017, which is in the range of the literature values reported for rural sites. The Ozone Monitoring Instrument (OMI) tropospheric NO_2 data also showed similar seasonal variations but tended to be biased high by 27%. It is thus expected that such unique results from multi-component observations with MAX-DOAS will be useful for evaluating satellite data and models for the Southeast Asia region.

Keywords: MAX-DOAS, Aerosols, AOD

Estimation of atmospheric ammonia distribution from GOSAT-TIR

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Atmospheric ammonia has the various sources such as food production, wastes, and biomass-burning and their emissions are increasing rapidly with human activities. It is reported that it has the large impacts for the environments, for instance, the formation of particulate matter or CCN, and the eutrophications. However, the accurate estimation of its behavior is difficult because their life-time in the atmosphere is not so long. Space-borne hyper-spectral TIR sounders enable to monitor them globally. Clarisse et al. (2009) had reported the global concentration distribution using the brightness temperature difference at the ammonia absorption band. The similar studies are also reported using the other sounders, TES, AIRS, and CrIS. Although they had captured the seasonal variations of the concentrations, the smaller time-scale events can be hardly detected because of their revisit cycle of 16 days. In addition, it also reported that there are some differences between the results from the satellites and the numerical simulations based on the inventories (Shephard et al., 2011). GOSAT which has the shorter revisit cycle of 3 days has the potential to reveal short time-scale events. The retrieval algorithm was developed based on non-linear Maximum a Posteriori (MAP) method (Rogers, 2000) and the scaling factor to the U. S. standard ammonia profile is estimated as the parameter. The large error causes, temperature and water vapor profiles are pre-estimated from GOSAT-TIR data. The preliminary results show the high concentrations in the northern India and the eastern China consistently with the previous researches and the global distribution will be presented.

Keywords: satellite remote sensing, thermal infrared radiation, atmospheric minor constituents

Temporal evolution of minor species observed with ground-based FTIR at Syowa Station, Antarctica in 2007, 2011, and 2016

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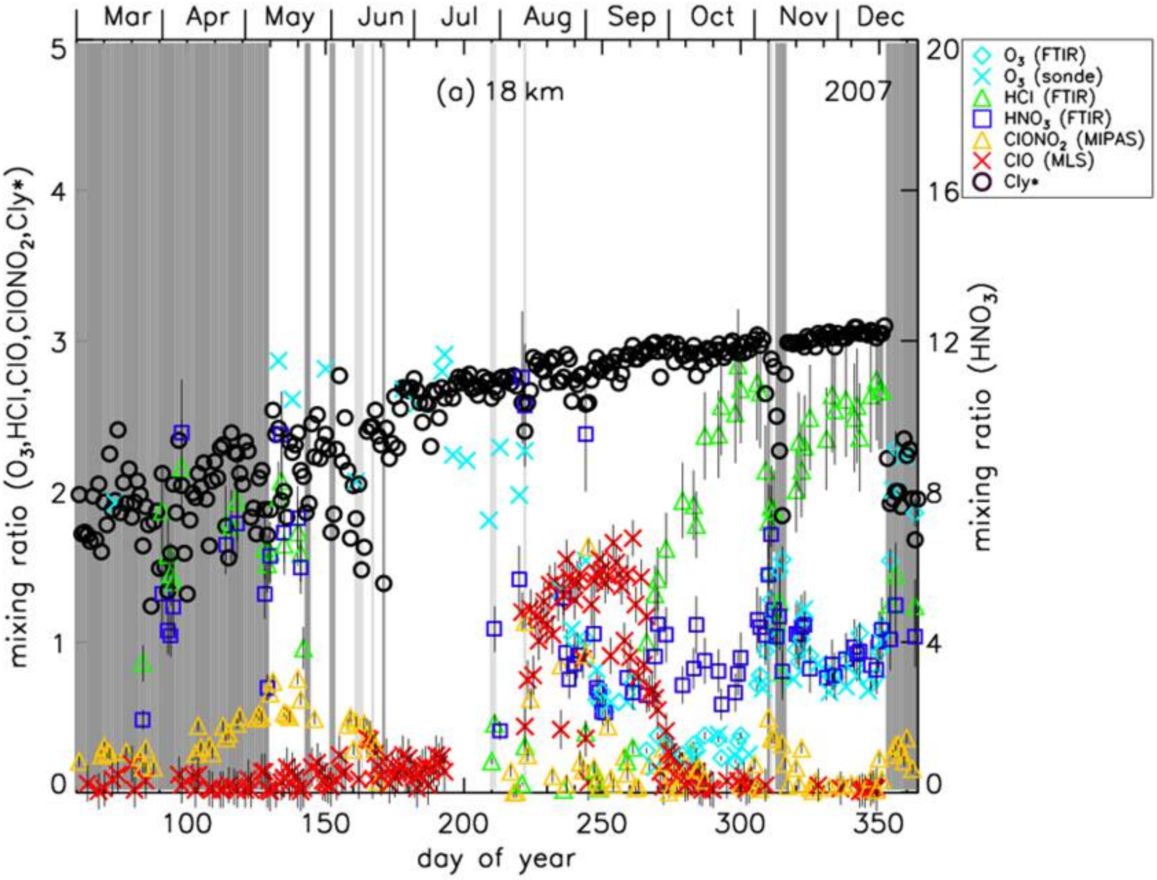
Vertical profiles of O_3, HNO_3, HCl, and $ClONO_2$ were retrieved from solar spectra taken with a ground-based Fourier-Transform infrared spectrometer (FTIR) installed at Syowa Station, Antarctica (69.0S, 39.6E) from March to December, 2007, September to November, 2011, and October to December, 2016. We analyzed temporal variation of these species combined with ClO data taken by Aura/MLS, and $ClONO_2$ data taken by Envisat/MIPAS satellite sensors at 18 and 22 km over Syowa Station. In early July, polar stratospheric clouds (PSCs) started to be formed over Syowa Station. With the return of sunlight at Syowa Station in early July, $ClONO_2$ and HCl showed depleted values while ClO showed enhanced values. At all three altitudes (18 and 22 km), when ClO concentrations started to decline in early September, HCl started to increase rapidly, while the increase in $ClONO_2$ was gradual. The Cly partitioning between HCl, $ClONO_2$, and ClO showed difference at different altitudes. At the altitudes of 18 km, where ozone was almost depleted, ClO and HNO_3 amounts are low, so conversion to HCl was favored rather than $ClONO_2$. Whereas, at 22 km, sufficient ozone still remained, at an amount that $ClONO_2$ formation from ClO and NOy species continued to occur at this altitude. This is the first in the world to observe O_3 destruction and recovery process of reservoir chlorine (HCl and $ClONO_2$) after disappearance of PSCs using ground-based FTIR in the Antarctic.

Keywords: FTIR, Syowa Station, Cly, ozone, ozone hole


Mar
Apr
May
Jun
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Aug
Sep
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Dec
(a) 18 km
2007
5
4
3
2
1
0
20
16
12
8
4
0
mixing ratio (O3,HCl,ClO,ClONO2,Cly*)
mixing ratio (HNO3)
O3 (FTIR)
O3 (sonde)
HCl (FTIR)
HNO3 (FTIR)
ClONO2 (MIPAS)
ClO (MLS)
Cly*
100
150
200
250
300
350
day of year

Theoretical Studies of Spectroscopic Line Mixing in Remote Sensing Applications

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The phenomenon of collisional transfer of intensity due to line mixing has an increasing importance for atmospheric monitoring. From a theoretical point of view, all relevant information about the collisional effect on line shapes is contained in the relaxation matrix whose diagonal elements are the half-widths and shifts of individual lines while the off-diagonal elements correspond to line interferences. For simple systems such as those consisting of diatom-atom or diatom-diatom, accurate fully quantum calculations based on sophisticated and realistic interaction potentials are feasible. However, fully quantum calculations become unrealistic for more complex systems. Meanwhile, due to relaying on the isolated line approximation, the semi-classical Robert-Bonamy formalism, which has been widely used to calculate half-widths and shifts for decades, fails in calculating the off-diagonal matrix elements. As a result, in order to simulate atmospheric spectra where effects from line mixing are important, semi-empirical fitting or scaling laws such as the energy corrected sudden (ECS) and infinite order sudden (IOS) models are commonly used. Recently, we have found that in developing semi-classical line shape theories, to rely on the isolated line approximation is not necessary [1]. By eliminating this assumption, we have developed a more capable formalism that enables one not only to reduce uncertainties of calculated half-widths and shifts, but also to calculate the whole relaxation matrix. Thanks to this progress, one can address the line mixing based on interaction potentials between two interacting molecules. We have applied this formalism for Raman and infrared spectra of linear molecules [1-4], asymmetric-top molecule [5], and symmetric tops with inversion symmetry such as the NH_3 molecule [6-8]. Our calculated half-widths of NH_3 lines in the parallel and perpendicular bands match measurements very well. Furthermore, the method has been applied to the calculation of the shape of the Q branch and of some R manifolds in the nu1 band of NH_3, for which an obvious signature of line mixing effects has been experimentally demonstrated. Similarly, the formalism very well reproduces off diagonal elements measured in some of ^{P}P doublets in the nu4 band. In summary, comparisons with measurements show that predictions from the new formalism accurately match the experimental line shapes.

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Keywords: Line mixing, relaxation matrix, line shape parameters, pressure broadened half-widths and shifts

Seasonal and decadal variations in stable carbon isotope ratios of low molecular weight dicarboxylic acids and related compounds in the remote marine aerosols from the western North Pacific

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Oxalic and other dicarboxylic acids comprise an important fraction of water-soluble organic aerosols in various environments. To better understand long-term atmospheric changes in the western North Pacific, we collected marine aerosol samples in 2001-2014 on weekly basis at a remote island, Chichijima (27° 04'E; 142°13'N), which is located in the boundary of westerly and easterly wind regimes. Here we present seasonal and decadal trends in stable carbon isotopic compositions ($d^{13}C$) of diacids dicarboxylic acids (C_2-C_{11}), oxoacids (C_2-C_9) and a-dicarbonyls (C_2-C_3) in the Chichijima aerosols. These compounds were determined using gas chromatography (GC) and GC/MS techniques after BF_3/n-butanol derivatization and using a GC/isotope ratio/MS technique.
Concentrations of total diacids fluctuated in a range of 10-600 ngm^{-3} with winter/spring maxima and summer minima. The maximum concentrations in winter/spring can be explained by a combination of enhanced emissions of polluted aerosols and their precursors in the Asian Continent and enhanced atmospheric transport to the North Pacific due to the intensified westerly winds in winter/spring. The concentrations of diacids seemed to increase from 2001 to 2008 and then decrease toward recent years, as supported by major ion analyses. Recent decline of diacids concentrations may be associated with the changes in the anthropogenic emissions due to the air quality improvement in Asian countries.
Stable carbon isotopic compositions determined for 2006 showed relatively high $d^{13}C$ values of oxalic acid (-22‰ to -4‰). The isotopic ratios were found to increase from winter to summer. Significant enrichment of ^{13}C in oxalic acid in summer should be associated with photochemical aging of organic aerosols in the marine atmosphere, caused by kinetic isotopic fractionation during the photo degradation of oxalic acid. We will discuss the decadal changes in the stable carbon composition of oxalic acid in relation to the atmospheric oxidation capability in the western North Pacific.

Keywords: Marine aerosols, Oxalic acid, Stable carbon isotope ratio, Seasonal and decadal trends

Aggregated, iron-oxide nanoparticles as a major metal atmospheric aerosol

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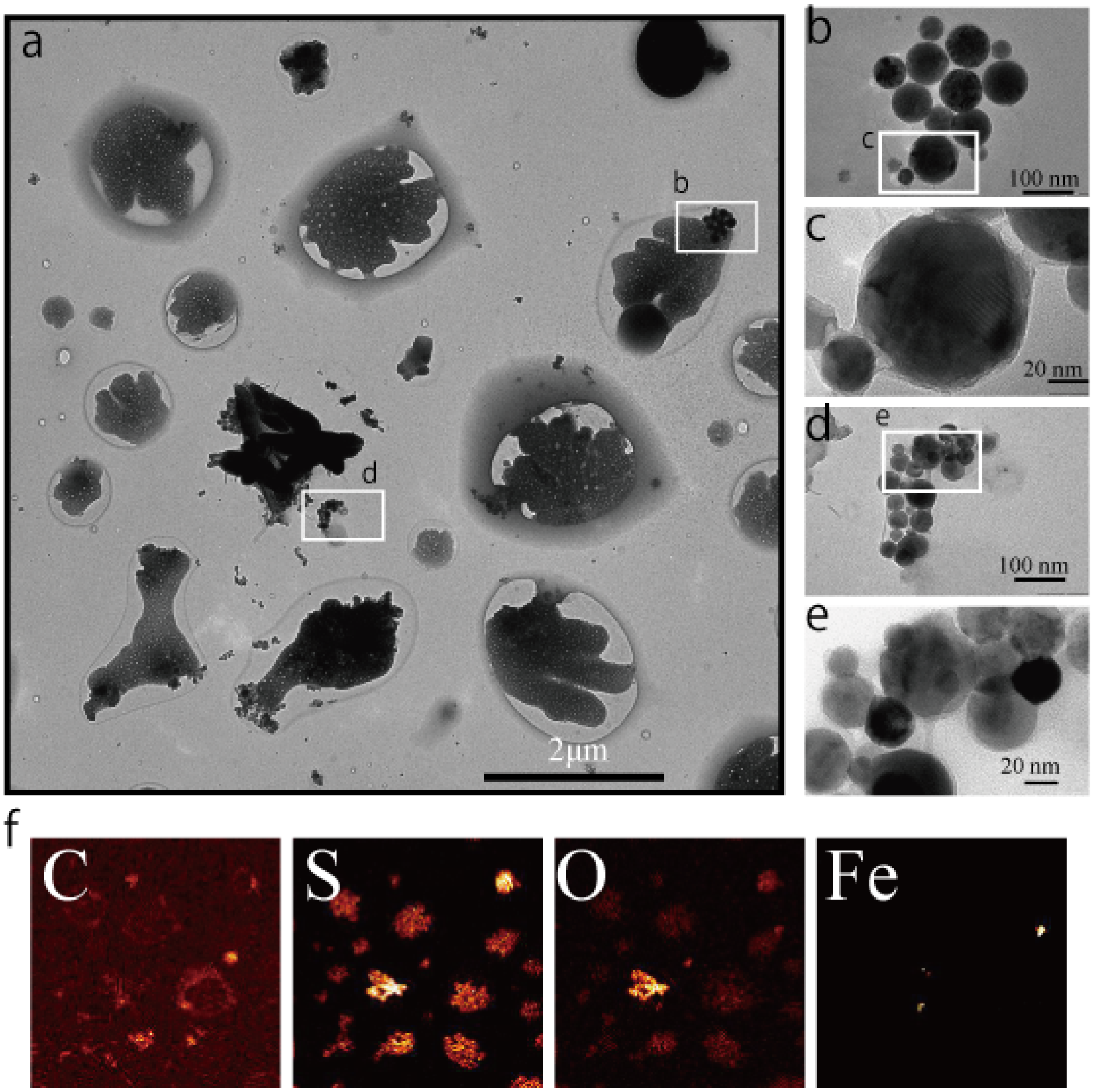
Iron-oxide particles possibly contribute to climate, if they absorb light; marine environment, if they dissolve to water; and human health, if they have toxicity. However, little is known about the influences of iron-oxide particles in atmosphere to these important issues. This study examines iron-oxide particles by using transmission electron microscopy (TEM) and single-particle soot photometer (SP2) from samples collected from Tokyo, Japan as well as those from other locations. TEM and SP2 use fundamentally different detection techniques for the iron-oxide particles. TEM allows characterization of the morphological, chemical, and structural features of individual particles, whereas SP2 optically measures the number, size, and mixing states of iron-oxide particles. By using both techniques, we aim to understand the occurrence, abundance, and chemical and physical properties of iron-oxide particles in atmosphere.

In this presentation, we mainly focus on the results by using TEM. TEM revealed that there are substantial amount of iron-oxide particles in aerosol samples with smaller than one micrometer, consistent to the results from SP2. These iron-oxide particles are mostly aggregates of iron-oxide particles with less than one hundred nanometer, similar to soot particles, suggesting that they are emitted from anthropogenic sources. They mainly consist of iron oxide although manganese, chromium, nickel, and other elements are occasionally detected. Electron-energy loss spectroscopy (EELS) analysis shows that these iron-oxide particles are magnetite, which absorbs light and has potential toxicity. Our results suggest that iron-oxide particles could be abundant in the aerosols having anthropogenic sources and may have an important contributions to climate, marine environment, and human health.

Keywords: Transmission electron microscope, Single-particle soot photometer, Magnetite, Tokyo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PAHs in $PM_{2.5}$ at high altitude in southern China: Meteorology, Emissions and Transport

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$PM_{2.5}$ samples were collected for polycyclic aromatic hydrocarbons (PAHs) analysis from March 18 to May 20 in 2012 at Mount Lushan (1165m), where is located in southern China. The sampling site was located between the boundary layer and troposphere, an ideal site for learning the influences of long-range transport on PAHs distributions. The volume concentrations of the measured PAHs were 6.98 ng/m^3 ranged from 1.47-25.17 ng/m^3 during the sampling time. The PAHs level at Mount Lushan was at a medium level comparing to other sites around the world. PAHs mass in $PM_{2.5}$ were 160.24 ug/g ranging from 63.86 to 427.97 ug/g. The predominant compounds were BbF (benzo[b]fluoranthene), Pyr (pyrene) and BP (benzo[g,h,i]perylene). In terms of aromatic-ring PAHs distributions, 6-ring and 4-ring were predominant, accounting for 27.9% and 24.2% of the total. It is due to that high molecular weight (HMW) PAHs have lower volatility and easier to attach in the particles comparing to low molecular weight (LMW) PAHs.

Meteorological conditions including temperature and humidity could affect the PAHs distributions. Temperature had a negative correlation with PAHs concentrations, i.e. when the temperature was higher, the PAHs concentrations become lower. This is due to the volatility of PAHs. Humidity had no significant relationship with PAHs concentrations. When the wind is strong, PAHs concentrations were lower. Strong wind would accelerate air motion, therefore PAHs were not easy to attach to particulates. Foggy and rainy weather occured during the sampling campaign. Selected samples before and after the special weather were analyzed. The concentrations of LMW PAHs were much higher than the HMW PAHs before fog or rain. For the PAHs concentrations, they are lower obviously after fog and rain. This suggested that foggy and rainy day lower pollutant levels and played a role in cleaning air.

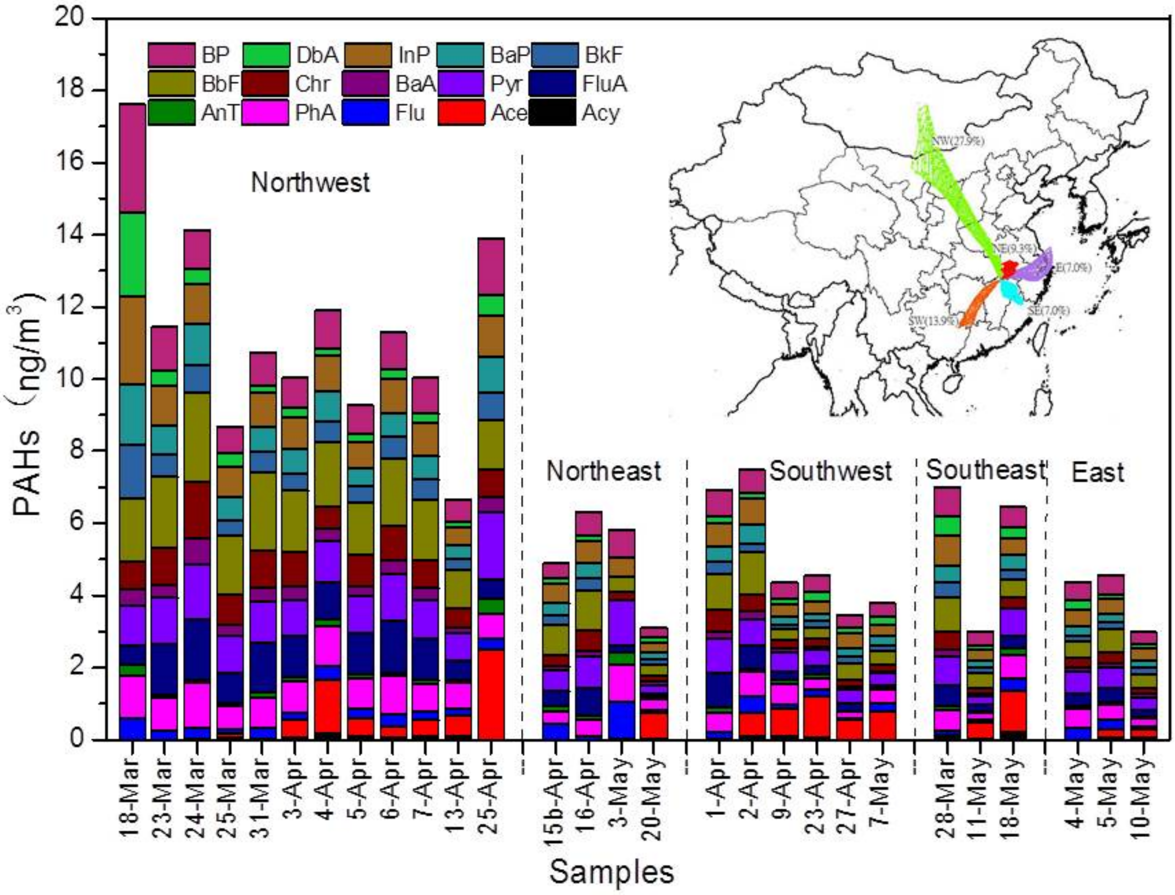
The backward trajectories simulated by HYSPLIT (the Hybrid Single Particle Lagrangian Intergrated Trajectory) model were mainly originated from five directions. The air mass from northwest, northeast, southwest, southeast and east accounted for 27.9%, 9.3%, 13.9%, 7.0% and 7.0%, respectively. Figure 1 suggested that Mount Lushan was mainly influenced by air mass from northwest during the sampling. The total concentration of PAHs were highest (11.31 ng/m^3, 6.65-17.60 ng/m^3) under the influence of northwestern air mass. The PAHs levels were similar when air mass from northeast (5.08 ng/m^3) and southwest (5.03 ng/m^3). When sample were originated from the sea in the east, the PAHs concentrations were at lowest level (3.97 ng/m^3).In addition, when Mount Lushan was mainly affected by northwestern air mass, most of the PAHs species were much higher except for AnT (anthracene) and Flu (fluorene). The concentration of AnT was similar (low level) in every sample during the sampling time and Flu concentration was the highest when affected by air mass from northeast. Thus, the air mass from northwest carried large amounts of pollutants to Mount Lushan and the long-range transport influenced the PAHs distributions.

Another important factor that affected PAHs distributions was the emissions. Ratio analysis AnT/(AnT+PhA), FluA/(FluA+Pyr), BaA/(BaA+Chr) and InP/(InP+BP) and PCA (principal components analysis) were used to identify the emission sources of PAHs in $PM_{2.5}$. It is suggested that the main emission sources were mainly from pyrolysis of petroleum fuel (vehicle exhaust) and biomass (coal) combustion. Many factories and highways existing in the north and southwest of Mount Lushan can well explain the main source of the PAHs. The PAHs distributions were mainly influenced by long-range

transport and emission sources. Temperature, wind, fog and rain could also affect PAHs distributions in $PM_{2.5}$.

We gratefully acknowledge the National Natural Science Foundation of China (21177073) and Mount Lushan Meteorological Station.

Keywords: PAHs distributions, Transport, Meteorology, Mount Lushan



Variability of Mixing State of Aerosols observed at a Surface Site in Korea in the Spring of 2016

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Aerosols have large influence on the radiative balance of the Earth’s atmosphere by scattering or absorbing solar visible radiation (direct effect) and by altering cloud microphysical properties (indirect effect) (IPCC, 2013). These effects can significantly depend on the chemical composition and mixing state of aerosol particles. Specifically, the mixing state of sulfate, nitrate, and organics with black carbon (BC) is a key parameter for estimating the aerosol direct and indirect effects (Bond and Bergstrom 2006, Adachi et al. 2010). However, there have been few studies on real-time measurements of chemical compositions mixed with BC in the atmosphere. The purpose of this study is to investigate the chemical composition of aerosol particles classified by the mixing state with BC in Asian outflow.

Ambient measurements were conducted in Korea from March 1 to April 7, 2016. A laser induced incandescence-mass spectrometric analyzer (LII-MS, Miyakawa et al. 2014) was used for the measurements. The LII-MS consists of a series of laser induced incandescence (LII) and mass spectrometric (MS) detectors. In the LII section, BC containing particles can be efficiently vaporized and emit incandescence signals by rapid heating in a near-infrared laser cavity. The sample air is subsequently introduced into the MS section to quantify chemical compositions of aerosol particles. The classification of aerosol particles with respect to the mixing state with BC (internal and external mixture) can be achieved by turning on and off the LII laser.

Nitrate, sulfate, and organics were the major species of PM_1 aerosols. On average, nitrate was most abundant and the fraction of internal mixture for sulfate was higher than that for the others. We found no systematic difference exceeding experimental errors between external and internal mixtures for nitrate and sulfate. Details of the temporal variation of chemical composition will be presented and discussed.

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Keywords: Aerosol, Chemical composition, BC mixing state

Spatial distributions and radiative effects of black carbon with MRI Earth System Model

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Most aerosol components have only the scattering property of solar radiation; however, black carbon (BC) aerosols efficiently absorb it and lead to heating of the atmosphere. Because of these effects, the role of BC particles in the climate system has been recognized to be particularly important. However, there remain large uncertainties in estimates of the spatial distributions of BC and its radiative effects in current climate models.

Recently, Meteorological Research Institute (MRI) developed a new version of MRI Earth System Model (MRI-ESM2) for participating in the Coupled Model Intercomparison Project Phase 6 (CMIP6). In this study, we modified treatments of BC in MRI-ESM2. We applied a new BC aging parameterization, which enables the representation of variations of the conversion rate from hydrophobic BC to hydrophilic BC depending on atmospheric conditions, although the original approach assumes the constant conversion rate of 1.2 days. We developed a new treatment of wet removal of aerosols in cumulus parameterization scheme, which enables a simultaneous treatment of vertical transport and wet removal of aerosols in cumulus convection in a consistent manner, although the original approach does not. We represented enhancement of BC light absorption due to coatings assuming internal mixing with sulfate, although the original approach does not take into account the coatings on BC.

We performed the MRI-ESM2 calculations for 2008-2015 with nudging towards the meteorological data (JRA-55) and sea surface temperature (not coupled with the ocean model in this study). Comparisons with the surface measurements showed that MRI-ESM2 improves the predictions of seasonal variations of BC over the Arctic, although the original constant-rate aging approach underestimates the BC concentrations and does not reproduce the seasonal variations. Comparisons with several aircraft measurements showed that MRI-ESM2 improves the vertical profiles of BC in the upper and middle troposphere, although the original wet removal approach largely overestimates BC mass concentrations. These results suggest that the seasonal variations of BC over the Arctic were primarily controlled by aging processes and wet removal by cumulus convection played an important role for BC concentrations in the upper and middle troposphere.

The annually and globally averaged direct radiative forcing by BC at the top of atmosphere was estimated to be approximately 0.2 W m^{-2} in this study. Comparisons with calculations using the original approaches suggest that the forcing increased by 40% due to enhancement of BC light absorption with coatings and increased by 20% due to improvement in representations of BC aging processes.

Keywords: Aerosol, Black carbon, Climate model, Aging process, Wet deposition, Radiative effects

Frost flowers and sea-salt aerosols over seasonal sea-ice areas in north-western Greenland

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Sea-salts and halogens in aerosols, frost flowers and brine play an important role in atmospheric chemistry in polar regions. Sea-salt fractionation proceeds on new and young sea ice. For that reason, sea-salt ratios in sea-salt particles (or aerosols) released from sea-ice areas differ from those of bulk seawater ratio, because of sea-salt fractionation on frost flower and in brine. Sea-salt fractionation can engender modification of aerosol hygroscopicity, which is closely related to phase transformation, heterogeneous reactions, and abilities of cloud condensation nuclei and ice nuclei. To elucidate the atmospheric impact of fractionated sea-salt particles, and their relation between sea-salt particles in the atmosphere and frost flowers on sea ice, one must ascertain their (1) chemical properties (e.g., concentrations, ratios, and pH) of frost flowers and brine, and (2) the physical and chemical properties of aerosols (e.g., size distribution, constituents, and mixing states) above seasonal sea ice with frost flowers. In spite of the importance, simultaneous observations and measurements of aerosols and frost flowers over seasonal ice areas with frost flower appearance have not been reported for polar regions.

Simultaneous sampling and observations of frost flowers, brine, and aerosol particles were conducted on several types (fresh - aged) of sea-ice and frost flowers around Siorapaluk in north-western Greenland during December 2013 –March 2014. Sea-salt constituents in frost flowers were determined with ion chromatograph (IC). Br- and iodine (I^- + IO_3^-) were analyzed with IC-MS and ICP-MS, respectively. Individual aerosol particles were observed and analyzed using SEM-EDX.

Results show that water-soluble frost flower and brine constituents were sea salt constituents (e.g., Na^+, Cl^-, Mg^{2+}, and Br^-). Concentration factors of sea-salt constituents of frost flowers and brine relative to seawater were 1.14–3.67. Sea-salt enrichments of Mg^{2+}, K^+, Ca^{2+}, and halogens (Cl^-, Br^-, and I) in frost flowers were associated with sea-salt fractionation by precipitation of mirabilite (Na_2SO_4 $10H_2O$), hydrohalite (NaCl $2H_2O$), and sylvite (KCl). Comparison between sea-salt ratios in brine and frost flower implied that precipitation of mirabilite and hydrohalite proceed in slush layer and then the residual brine were migrated vertically onto frost flowers in our research conditions. Molar ratios of sea-salts (Mg^{2+}/Cl^-, K^+/Cl^-, Ca^{2+}/Cl^-, and Br^-/Cl^-) changed gradually with aging and growth of frost flowers and sea-salt fractionation under colder conditions. Furthermore, sea-salt fractionation was associated with not only surface air temperature but also sea-ice thickness which related to heat conduction from seawater. In contrast to Br- enrichment in frost flower with the aging, changes of I/Cl^- ratio in frost flowers, however, were not clear. Iodine release from frost flowers might be more likely to proceed relative to Br^- release. Aerosol number concentrations, particularly in coarse mode, were increased considerably by release from the sea-ice surface under strong wind conditions. Sulphate depletion by sea-salt fractionation was found to be slight in sea-salt aerosols because of heterogeneous SO_4^{2-} formation on sea-salt particles. Mg was enriched in coarse and fine sea-salt particles collected on sea-ice area. Mg in sea-salt particles was in the form of $MgCl_2$ and $MgSO_4$. Strong Mg enrichment might be more likely to proceed in fine sea-salt particles. Mg enrichment in sea-salt particles was enhanced under colder conditions. In addition, ikaite-like and mirabilite-like particles identified in the atmosphere only near new sea ice are close to the sea-ice margin. Thus, Ikaite-like and mirabilite particles might be released from initial sea-ice before freezing over, and Mg-rich sea-salt particles might be released from sea-ice surface with frost flowers.

Keywords: Sea-salt aerosols, Halogens, Frost flower

Estimation of sulfate aerosol sources during an intensive field campaign in October–November, 2015 at Cape Hedo, Okinawa

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Air quality in Asia is a major regional-to-global environmental problem, and under such circumstances, several collaborative international experiments have been conducted over the western Pacific region. To investigate the transformation of aerosols during long-range transport (LRT) is necessary for promoting our understanding of regional air pollution and climate change. An intensive observation campaign at Cape Hedo, Okinawa, Japan (CHAAMS) was conducted from late October to early November 2015. The location of CHAAMS is suitable for capturing the atmospheric pollutants via LRT. During this period, sulfate (SO_4^{2-}) was the dominant aerosol component, and the air quality model can capture the observed meteorological conditions and SO_4^{2-} variation. By using the air quality model with the tagged tracer method, the sources of high SO_4^{2-} concentration were estimated. On October 27, when the westerly wind was dominant, the main source was anthropogenic SO_2 emissions in China. On November 1, when the northerly wind prevailed, the impact of volcanoes in western Japan was significant and the conversion ratio from SO_2 to SO_4^{2-} was lowest, at less than 70%, due to the faster transport. During the latter part of the campaign, the northerly to easterly winds were prominent, and the impacts of Korea, Japan, and ship to SO_4^{2-} observed at CHAAMS were also obvious. On November 4, when the contributions from Korea, Japan, and ship were the highest, the conversion ratio was also the highest, at greater than 95% due to long-range transport. The modeled sources of volcanoes and ship emissions corresponded well with the observed coarse-mode SO_4^{2-} and V/Mn ratio, respectively. It was demonstrated that the mutual evaluation of sources from model and observations enable to estimate SO_4^{2-} sources with higher confidence.

Keywords: Sulfate Aerosol, Source contribution, Air quality model, Long range transport

Modeling the Long-Range Transport of Particulate Matters During Winter in East Asia using NAQPMS and CMAQ

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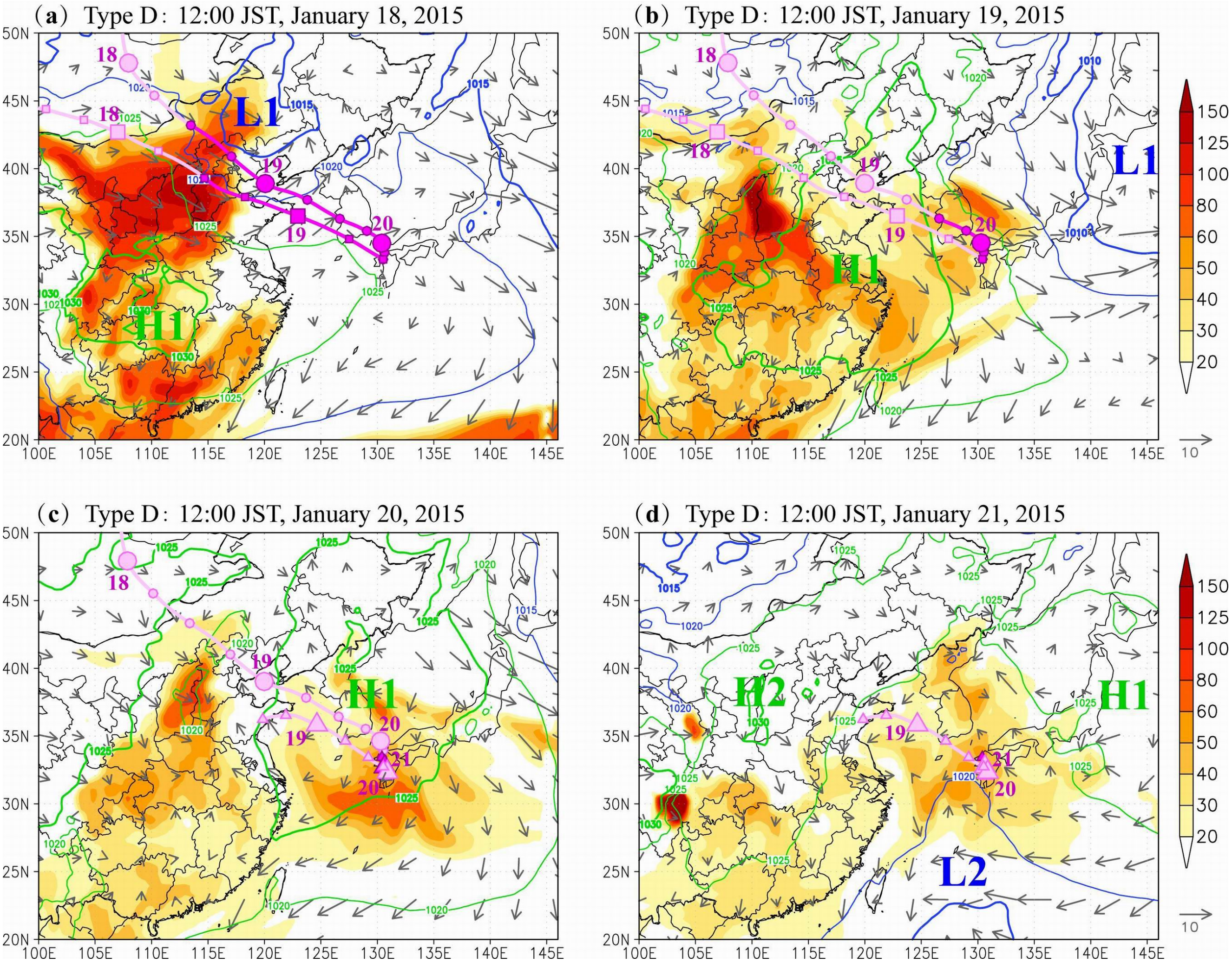
Two regional chemical transport models (NAQPMS and CMAQ) were used to simulate several episodes of high $PM_{2.5}$ concentration observed in January 2015 over China and Japan. Simulation results from both models reasonably explained observed $PM_{2.5}$ levels, as well as the variation observed within three sites in both China and Japan. Some bias existed between these models due to differences in frameworks, including model domains, horizontal resolution, vertical layers, and emissions.

Based on the model results and the synergetic aerosol observations in Fukuoka, Japan, three types of LRT of air pollutants were observed. The first episode showed increased fNO_3^- concentrations relative to fSO_4^{2-} (type N), indicating the importance of NO_3^- LRT in winter. The second episode showed fSO_4^{2-} concentrations, which were ~3.4-fold greater than fNO_3^- (type S). The third episode showed high dust concentrations mixed with anthropogenic pollutants (type D), indicating that the LRT of dust was also important in winter, as well as spring.

Both models reasonably explained variations in aerosol components during episodes N and S. Simulated spatial distribution variations indicated the outflow of fSIA from continental Asia to western Japan, consistent with the corresponding $PM_{2.5}$ peak at Qingdao and over Japan. During episode S, RH was significantly higher than episode N, therefore, SO_4^{2-} formed quickly due to aqueous-phase reactions under high RH conditions.

During episode D, mineral dust transported from continental Asia was quickly transported to downwind regions, stagnating over the south of Japan for three days (See Figure). Measurements showed high cNO_3^- concentrations and high cNO_3^-/fNO_3^- ratio during episode D. These findings were well reproduced by the NAQPMS model after considering heterogeneous reactions on dust particles, which indicates the importance of heterogeneous processes for the LRT of dust and anthropogenic pollutants over East Asia. During this period, both models underestimated fSO_4^{2-} levels, indicating that current models may miss certain emissions of SO_2 and mechanisms promoting the conversion of SO_2 to SO_4^{2-}.

Keywords: Secondary inorganic aerosol, Dust, Air quality model, Long-Range Transport, East Asia


(a) Type D: 12:00 JST, January 18, 2015
(b) Type D: 12:00 JST, January 19, 2015
(c) Type D: 12:00 JST, January 20, 2015
(d) Type D: 12:00 JST, January 21, 2015
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Joint retrieval of aerosol optical depth and surface reflectance over land using geostationary satellite data

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The Advanced Himawari Imager (AHI) aboard Himawari-8 geostationary satellite provides high-frequent observations with big coverage, multiple spectral channels, and high spatial and spectral resolution. With these characteristics, the AHI data have significant advantages to monitor the air quality and estimate the aerosol properties. In this study, AHI data were used to develop an algorithm for the joint retrieval of aerosol optical depth (AOD) over land and land surface bidirectional reflectance. Instead of constructing a Look-Up-Table (LUT) and pre-estimating the surface reflectance to retrieve the AOD, the atmosphere properties and surface bidirectional reflectance were retrieved simultaneously using an optimal estimate method. The algorithm uses an Earth-atmosphere model, which couples the atmospheric radiative transfer model and surface bidirectional reflectance factor (BRF) model. Utilizing the character that the surface reflectance properties are much more stable than atmosphere aerosol, our retrieval is based on the two basic assumptions: the surface bidirectional reflective properties is invariant during a short time window while aerosol properties (e.g. AOD, AE) change. Optimal estimate method is employed to calculate the AOD and surface BRF. Detailed analysis and validation about the retrieval results were conducted using ground-based measurements (AErosol RObotic NETwork (AERONET) sites) and satellite product (MODIS C6 aerosol product). The validation of the AOD with AERONET measurements shows a high correlation coefficient: R2=0.81, RMSE=0.13, and about 80% AOD retrieval results within the Expected error (EE) of (0.20**AODAERONET* ±0.05). The retrieved AOD were also compared with MODIS Collection 6 AODs and it shows high consistency. All comparison and validation demonstrated that the algorithm has the ability to estimate hourly aerosol optical depth with high accuracy over land.

Long-term trend of springtime tropospheric ozone at Mt. Happo, Japan, 1998–2016

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Tropospheric ozone is one of the most important trace gases in the Earth’ s atmosphere because it plays a central role in global warming, oxidizing capacity of the troposphere, and air quality at local, regional and global scales. During the past decades the emissions of pollutants from East Asia have been rapidly increasing. In particular China is the largest and dominant contributor to the Asian emissions. However, recent several studies based on the analysis of satellite tropospheric nitrogen dioxides data inferred possible peaking out of nitrogen oxides emissions in China. Along with the emissions, climate plays an important role in controlling the variations in tropospheric ozone. Some studies suggested that interannual to decadal climatic variations modulate the airflow from polluted regions. We revisited and updated the long-term trend of tropospheric ozone at Mt. Happo, Japan, based on continuous measurements for the period from 1998 to 2016. We focused on the springtime ozone concentration and possible influences by the continental outflow from East Asia. Since 1998 the springtime ozone concentration has shown a large increase until 2007, very likely caused by the increase in the emissions of ozone precursors associated with economic growth in eastern China, as evidenced from satellite observations of nitrogen dioxides. After the monotonic increase until 2007, the ozone level has been flattened associated with substantial drops in 2008 and 2012. And the recent ozone levels have been sustained until 2016. We concluded that two decreases were induced by two different factors. One is the decrease of contribution from central eastern China because of the changes in meteorological fields. When the anomaly of pressure over the Pacific Ocean east of Japan decreases and the anomaly surrounding the Tibetan Plateau increases, polluted air masses from central eastern China tend to be transported to Mt. Happo by the strong westerly component. The other factor is the decrease in emissions of ozone precursors from eastern China, which is the phenomenon observed by satellite.

Keywords: ozone

Sources and effects of atmospheric nitrous acid in the marine boundary layer

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Nitrous acid (HONO) is an important reservoir of hydroxyl (OH) radicals in the atmospheric boundary layer. However, its sources are still not well understood. As few HONO observations have been performed in marine areas, we conducted measurements at two coastal sites, Tuoji Island in North China and Hong Kong, to investigate the sources and effects of HONO in the marine boundary layer. Compared with urban sites, HONO concentrations were low in marine sites. However, they were still significantly larger than that could be explained by the mechanisms in photostationary state (PSS). Through case study, results have indicated an interesting phenomenon that HONO may be produced faster on sea surface than on land surface. And further studies should be carried out to confirm this finding. In addition, HONO plays an influential role in atmospheric oxidative capacity and air quality in coastal regions.

Keywords: Nitrous acid (HONO), Sources, Effects, Marine boundary layer

Assessment of Sensitivity of Tropospheric Ozone Production to NO_x and VOCs Emissions Using WRF/Chem Model for Megacity Delhi

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An increase in ozone build up in the recent years has been witnessed in Delhi, the capital city of India which is a cause of alarm due to the detrimental effects of ozone on public health. Regular monitoring of ozone concentrations revealed that an increase of 6 $\mu g\ m^{-3}$ from 2009 (35.3 $\mu g\ m^{-3}$) to 2012 (41.5 $\mu g\ m^{-3}$) was recorded (Gupta and Mohan, 2015) in Delhi and in summer 2015, exceedance of O_3 levels ranged from 92% - 97%. Tropospheric ozone is produced by a cycle of reactions involving two basic pollutants NOx and VOCs. Chemical regional transport models such as WRF/Chem are used extensively for modeling of ozone concentration. As ozone production has distinctive daytime and nighttime chemistry, Gupta and Mohan (2015) recommended that for Delhi model implementation shall be made for policy decisions cautiously with due consideration to the magnitudes of ozone levels. In order to implement control strategies it is conducive to understand whether the study area is NOx or VOC limited. This study focuses on the WRF/Chem model to simulate ozone concentration for megacity Delhi during summer conditions for three consecutive years. To understand the role of VOC and NOx in ozone formation model simulated VOC to model simulated NOx ratios were studied through scatter plots. Ozone production is considered to be VOC limited at low VOC to NOx ratio i.e. less than about 4 to 1 and for high ratio greater than about 15 to 1 the region is classified as NOx limited. There are various sources that contribute to VOC emissions such as vehicular pollution, refueling stations, industrial hubs, diesel generator sets etc. In Delhi overall about 26 to 54% of VOCs are due to the emissions from diesel internal combustion engine (Srivastava et al., 2005). Srivastava et al. (2005) reported source contribution to total VOC for a traffic junction in Delhi as 43% from diesel exhaust, 24% from gasoline, 19% from evaporative and 14% from other sources. As the ozone production is VOC limited for megacity Delhi, WRF/Chem model can be further used to apply control strategies. The response of reduction in VOC emissions from transport sector on ozone production can be modeled in order to formulate effective mitigation measures involving both VOC and ozone reduction.

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Keywords: Ozone, VOC to NOx ratio, WRF/Chem, Megacity Delhi

Seasonal and interannual variations in the atmospheric Ar/N_2 ratio observed at five ground based stations in Japan

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Atmospheric Ar/N2 ratio is a unique tracer to detect the spaciotemporally-integrated air-sea flux or ocean heat content (OHC), since the variations in surface Ar/N2 ratio are driven by air-sea Ar and N2 fluxes principally due to changes in solubility in seawater. The relative temperature dependence of the solubility of Ar is larger than that of N2, so that the atmospheric Ar/N2 ratio increases with increasing ocean temperature. We have started systematic measurements of the Ar/N2 ratio by using a mass spectrometer (Ishidoya and Murayama, 2014) at Tsukuba (36°N, 140°E) and Hateruma Island (24°N, 124°E), Japan since 2012 and at Cape Ochi-ishi (43°N, 146°E), Takayama (36°N, 137°E) and Minamitorishima Island (24°N, 154°E), Japan since 2013. Not only clear seasonal Ar/N2 cycles with summertime maxima were found at all stations, but also clear interannual variations were seen from the 5-years data at Tsukuba and Hateruma; gradually increased from 2012 to the begenning of 2015 and did not show a significant increase/decrease since then. The seasonal amplitudes and appearances of maxima and minima of the Ar/N2 cycles were larger and earlier, respectively, at coastal stations at Hateruma, Ochi-ishi and Minamitorishima than those at inland sites at Tsukuba and Takayama. The peak-to-peak seasonal amplitudes were in the range of 15-45 per meg, which were comparable to or larger than those reported by past studies (Keeling et al., 2004; Cassar et al., 2008). The interannual variations of Ar/N2 ratio at Tsukuba and Hateruma were generally in phase with those in the global OHC reported by NOAA/NODC (updated from Levitus et al., 2012), which suggests our observational results reflect wide-area averaged information of ocean temperature.

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Keywords: atmospheric Ar/N2 ratio, ocean heat content, seasonal variation, interannual variation

Estimation of CO_2 contributions from fossil fuel consumption based on the atmospheric O_2 and CO_2 continuous measurements

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Atmospheric carbon dioxide (CO_2) concentrations observed in urban areas and the surroundings often show short-term elevations on a time scale ranging from several hours to a few days. These variations are considered to be attributed to the CO_2 emissions from biotic activities and burning of fossil fuels. Partitioning the contribution rate between the individual CO_2 sources would be useful to reduce the uncertainty of the CO_2 emissions estimated from atmospheric inversion calculations. The ratio of oxygen (O_2) consumption to CO_2 emission for fossil fuel burning ($-O_2/CO_2$) ranges from 1 to 2, depending on the elemental composition of the fossil fuel. The average $-O_2/CO_2$ exchange ratio for the fossil fuel consumed in Japan is more than 1.4 while that for land biotic process is 1.1. Therefore, the difference in the exchange ratios could allow us to determine the contributions from the individual sources. In this study, we started a continuous observation of the atmospheric CO_2 and O_2 concentration at Tsukuba in February 2015, and evaluated the individual contributions of the biotic and fossil fuel-derived emissions to the short-term CO_2 variations based on the observed $-\Delta O_2/\Delta CO_2$ changing ratios. The observed $-\Delta O_2/\Delta CO_2$ changing ratios show clear seasonal variation with minimum in summer and maximum in winter, ranging from 1.0 to 1.6. Taking into account of the average $-O_2/CO_2$ exchange ratio of 1.58 for the fossil fuel consumption in Tsukuba, we obtained the contribution ratios of fossil fuel burning to the CO_2 variations were more than 70% in winter and 0–30% in summer. To validate the above estimations based on the $-O_2/CO_2$ exchange ratio, we conducted flask samplings of air 6 times at the different months during the observation period and measure $^{14}CO_2$, which is known as good indicator of the burning of fossil fuels. Note that 5 flask samples were usually collected for each sampling at an interval of 3 hours to detect the diurnal variations. The contribution rates of the fossil fuel CO_2 estimated from $-O_2/CO_2$ ratios and $^{14}CO_2$ agree each other within the uncertainties.

Heterogeneous reactions of gaseous ozone with aqueous sesquiterpenes: The roles of Criegee intermediates

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We report the detection of intermediates/products generated on fresh surfaces of alpha-humulene and beta-caryophyllene solutions in water/acetonitrile mixtures exposed to gaseous ozone for ~ 10 microsecond. We focus on the identification of intermediates/products and their mechanisms of formation via negative ion online electrospray mass spectrometry complemented with the use of (1) H/D and $^{16}O/^{18}O$ isotopes, (2) a OH-radical scavenger, (3) variable O_3 number densities, and (4) n-alkyl carboxylic acid additions. We will discuss how our results provide direct evidence on the distinct reactivity of Criegee intermediates at air/aqueous interfaces.

Keywords: Criegee, Terpenes, BVOC, ozone, surface, interface

Inhaled ozone reactions with endogenous antioxidants and surfactants on the surface of lung lining liquid films

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We report the detection, via online electrospray ionization mass spectrometry, of the intermediates/products generated on the surface of surfactant protein B aqueous solutions exposed to gaseous ozone for 10 microseconds in the presence/absence of ascorbic acid/ascorbate as antioxidant. These experiments simulate how inhaled ozone reacts with typical components of the air-aqueous interface of human respiratory tract lining fluids. We found dramatic changes in the species detected at pathological acidic pH ˜4 vs normal physiological pH 7 conditions. We will provide clues on the identity of the species generated in each case and on ther mechanisms of formation. We will discuss the implications of our findings on the adverse health effects induced in healthy and diseased subjects.

Keywords: Inhaled ozone, Surfactant protein B , Health effects of air pollution on healthy and diseased subjects

Surface melting on ice surfaces visualized by advanced optical microscopy

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Ice is one of the most abundant crystals on the earth, and hence its phase transitions exert enormous influence on the global environment. Surface melting of ice crystals is one of such phase transitions: ice crystal surfaces are covered with thin liquid layers, so called quasi-liquid layers (QLLs), even below the melting point (0˚C). Since QLLs enhance various chemical reactions in ice clouds, presence or absense of QLLs is also important for atomospheric chemistry.

Recently, we and Olympus Engineering Co., Ltd. have developed laser confocal microscopy combined with differential interference contrast microscopy (LCM-DIM) by which we succeeded in the direct visualization of 0.37-nm-thick elementary steps [1] and QLLs [2] on ice for the first time. The direct visualizations revealed that QLLs with two types morphologies partially cover the ice surface [2,3] although QLLs had been thought to cover the whole ice surface uniformly. And we found that the appearance of QLLs depended on not only the temperature but also water vapor pressures [4,5] and species of ambient gases [6].

For example, we found that hydrogen chloride (HCl) gas strongly induced the appearances of QLLs. The droplet shape QLLs were observed in the temperature range of -15.0 ˜ -1.5˚C, where no QLL appears in the absence of HCl gas [6]. These results indicates that HCl gas adsorbed on ice crystal surfaces probably changed the surface structure of ice crystals and then induced the subsequent melting of ice surfaces. The long-term (one-hour) existence of the droplet QLLs under the undersaturated conditions and their behavors strongly suggests that the droplet QLLs were thermodynamically-stable HCl solutions.

In addition, we found that the HCl induced droplets were embedded into ice crystals by growth of ice crystals and the embedded droplets appeared again by evapolation of ice crystals. These results shows the possibility that ice crystals can store large amount of gas components as fluid inclusions.

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Keywords: Ice, Surface melting, Quasi-liquid layer, Hydrogen chloride gas, Advanced optical microscopy

Evaluation of ice nucleating activity of mineral aerosols: importance of mineralogy and aging process

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Ice crystals may be formed in the super-cooled liquid phase clouds with the aid of aerosol particles that act as ice nuclei. Among various aerosol particles that act as ice nuclei, mineral dusts are considered most ubiquitous due to their abundance in the earth’ s atmosphere. However, the mechanism by which mineral dusts act as efficient ice nuclei is not well understood. The purpose of this study is to narrow down the mineralogical factors that determine their initial ice nucleation activity. This study employed a cold-float technique to evaluate ice nucleation activity of mineral dusts with various mineralogical compositions. We show that there are potentially three major mineralogical factors controlling the ice nucleating activity of feldspar mineral group, namely, the cation type, solid solution state and ordering structure. In addition, we found that the ice nucleating activity of feldspar drops and shows similar activity as the clay minerals following treatment by sulfate acid, suggesting atmospheric aging may slow down the ice nucleation by the most efficient mineral types (e.g. K-feldspar). These results have important implications in the ice nucleating activity of mineral dusts in the actual atmosphere.

Response of total ozone reactivity analyzer to mixture of gaseous isoprene and NO

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Biogenic volatile organic compounds (BVOCs) have been focused on as precursors of tropospheric ozone (O_3) and secondary organic aerosols. Various species of BVOCs have C=C double bonds and can react with O_3. To capture BVOCs comprehensively, a total ozone reactivity (R_{O3}, the sum of $k_i[VOC_i]$) analyzer has been developed [1-4]. R_{O3} of sample BVOCs can be determined when decrease of O_3 due to BVOCs+O_3 is precisely monitored. In our previous studies, the detection limit of the analyzer reached 6.3×10^{-5} s^{-1} (S/N=3, 60-s average, 50-s reaction) [4]. To apply the analyzer to field observations where the samples consist of multiple compounds, characteristics of the analyzer should be explored further. For example, nitric oxide, NO, which exists significantly everywhere in the troposphere, can react with ozone rapidly. When BVOCs are captured as R_{O3}, NO can be detected simultaneously. A model study reported that, even when R_{O3} in the forest atmosphere is focused, contribution of NO to R_{O3} can be critical [5]. Generally, NO concentration can be captured easily utilizing an NO analyzer. Thus, if dependence of the output of R_{O3} analyzer on NO concentration is understood, contribution of NO to observed R_{O3} can be separated and R_{O3} due to BVOCs can be accurately quantified. In this study, mixtures of isoprene and NO standard gases were prepared and response of R_{O3} analyzer to the mixtures was experimentally captured in order to explore the possibilities of the standard addition method of NO to BVOCs samples. As a result, among the isoprene standard sample (A) and NO-added isoprene samples (B, C, D), a strong correlation was observed between the concentration of NO and the measured R_{O3} as shown in Fig.1. The intercept of the regression line agreed well with the observed R_{O3} of isoprene standard sample (A). It was experimentally confirmed that, when NO addition was conducted, the contribution of BVOCs (isoprene in the figure) to R_{O3} could be determined as the intercept of the regression line between NO concentration and measured R_{O3}. Consequently, the method of standard addition to separate the contribution of NO from observed R_{O3} was established. Evaluation of BVOCs as R_{O3} is possible even when NO coexists in the sample. As a next step, application of this method to R_{O3} observations for studying BVOCs emission from real plants and/or ambient samples is promising.

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Keywords: biogenic VOCs, nitrogen oxides, standard addition method

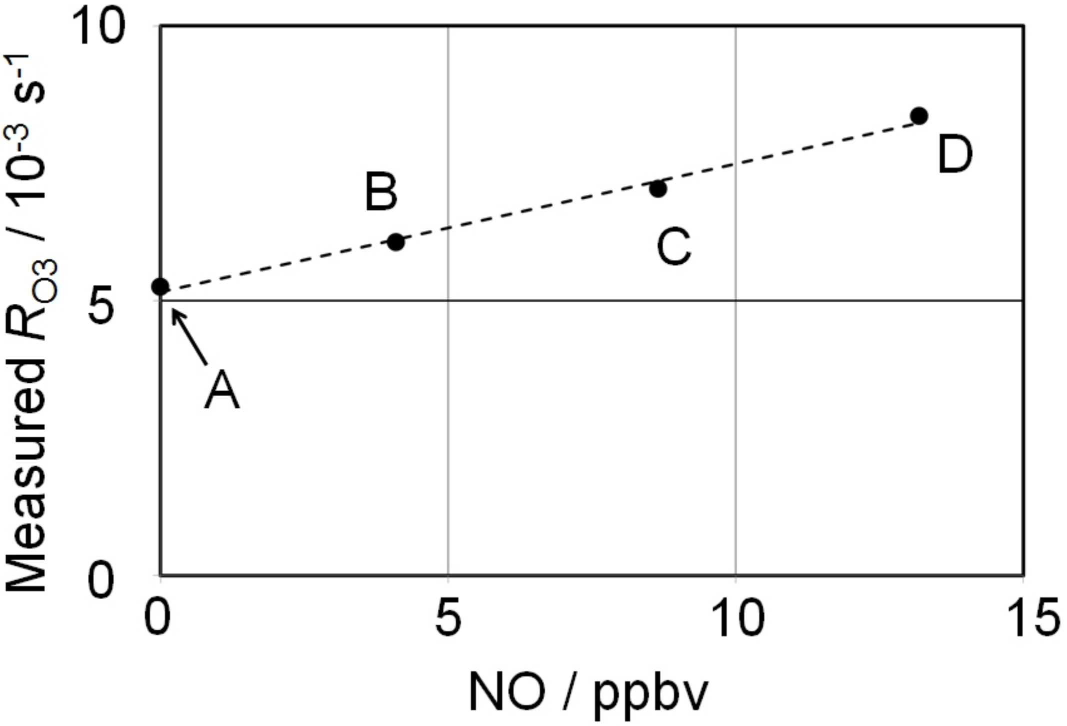

Fig.1 Observed relationship between NO mixing ratio and measured ozone reactivity.
A: isoprene standard sample. B, C, D: NO-added isoprene samples.
Regression line (dashed line): slope 2.3×10^{-4} s^{-1} $ppbv^{-1}$, intercept 5.2×10^{-3} s^{-1}.

Analyses of trends, seasonal variations, and short-term variations of atmospheric N_2O concentrations observed at Hateruma Island and Cape Ochi-ishi

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Nitrous oxide (N_2O) is one of the important anthropogenic greenhouse gas in the atmosphere, having a GWP 265 times that of CO_2 for a 100-year timescale. To enhance understanding of the global N_2O cycle, the National Institute of Environmental Studies (NIES) has been carrying out in-situ observations of the atmospheric N_2O at Hateruma Island (HAT; lat. 24.1°N, long. 123.8°E) since March 1996 and at Cape Ochi-ishi (COI; lat. 43.2°N, long. 145.5°E) since June 1999 by using gas chromatographic systems. In this study, the trends, the seasonal variations, and the synoptic-scale variations of the atmospheric N_2O observed at both sites are examined. The N_2O concentrations at both sites steadily increased at an almost same rate of about 0.8 ppb/yr. Although the interannual variations in the growth rates seem to be rather small, relatively large increasing rates larger than 1.1 ppb/yr were observed for HAT in 1999-2000 and for HAT and COI in 2014-2015. The average seasonal cycles show the maxima in April for both sites and the minima in July for HAT and in September for COI. The peak-to-peak amplitudes of the average seasonal cycles are about 0.5 ppb and 0.8 ppb for HAT and COI, respectively. It should be noted that the seasonal amplitude for HAT seems to increase temporally at a rate of 0.012±0.007 ppb/yr ($p=0.1$) although that for COI doesn’t show a significant long-term change. In addition to the seasonal variations, synoptic scale variations are often observed especially at HAT during a period from November to March, when the polluted air masses are predominantly transported from the continental regions. The standard deviations of the detrended and deseasonalized time series of N_2O for the winter 5-month period for HAT show significant temporal increase. The EDGAR inventory estimation (v4.2, FT2012) shows that the anthropogenic N_2O emissions from China increase about 40% during 1996-2012. Therefore, the increasing trends in the amplitudes of the seasonal and short-term variations at HAT may reflect the anthropogenic emission increase in China.

Keywords: atmospheric N2O, greenhouse gas, seasonal variation, short-term variation

Short-term variations of N_2O and CO mixing ratios observed at suburb of Sendai from November to December 2016

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Nitrous oxide (N_2O) is one of the most important greenhouse gas, and carbon monoxide (CO) has a significant role in atmospheric chemistry through reactions with hydroxyl radical (OH) in the atmosphere. The both gases are released from anthropogenic and natural origins and should be monitored with high accuracy to understand their dynamics. Atmospheric N_2O and CO mixing ratios over Aoba-yama, suburb of Sendai, Japan had been observed from November to December 2016 by a continuous measurement system for N_2O and CO, that was a newly developed in Tohoku University using Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS). The baseline mixing ratios of N_2O and CO mixing ratios were ~331 ppb and ~150 ppb, respectively, during the measurement period. Further, short-term and sporadic increases of N_2O and CO were also frequently observed. By means of meteorological analyses including backward trajectory calculations, those short-term variations are interpreted as being affected by local emissions near the observation site in some cases, or by regional-scale transport of air masses in other cases. The overall tendency for trajectories suggests that air masses transported from northeast China have relatively low mixing ratios of N_2O and CO, while those from near Japan have relatively high mixing ratios of them.

Keywords: N2O CO, trajectory analysis, transport, lower troposphere, northeast Asia

Real-time measurement of atmospheric carbon monoxide combined with mid-infrared wavelength modulation spectroscopy

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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 observed CO concentration in the atmosphere with a mid-IR laser absorption spectrometer that uses a 4.69 μm quantum cascade laser with wavelength modulation spectroscopy (WMS).
The absorption line for the measurements of CO was at 2131.63 cm^{-1}. The laser was scanned at 1.10 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. The precision (1 σ) of the measurement for 7 hours was 4.18 ‰.
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 10 ppb.
Measurements of outside air were conducted on the Kashiwa campus of The University of Tokyo over February 8-11, 2017. During the measurements of ambient air, the calibration was performed every 6-9 hours.
Outside air measurements were conducted over February 8-11, 2017. The CO mixing ratio during the measurements ranged from 0.11 ppm to 0.60 ppm. In the daytime of February 10, the wind brew from northwest. There are no big industrial areas in that direction. So the CO concentration was low and steady. There are some spikes in CO concentration. These may be attributed to cars passing near the observation point.
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.

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

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Continuous measureements 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 and estimate the distribution of the flux over this vast area (Sasakawa *et al.,* 2010, 2012, 2013) 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 (5-85 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.
We will discuss the long-term variations in CO_2 and CH_4 observed with the present system. From the year of 2015, we installed a Cavity Ring-Down Spectroscopy (CRDS; Picarro inc.) at KRS, DEM, and NOY. We thus validate the recent data with the data by the CRDSs.

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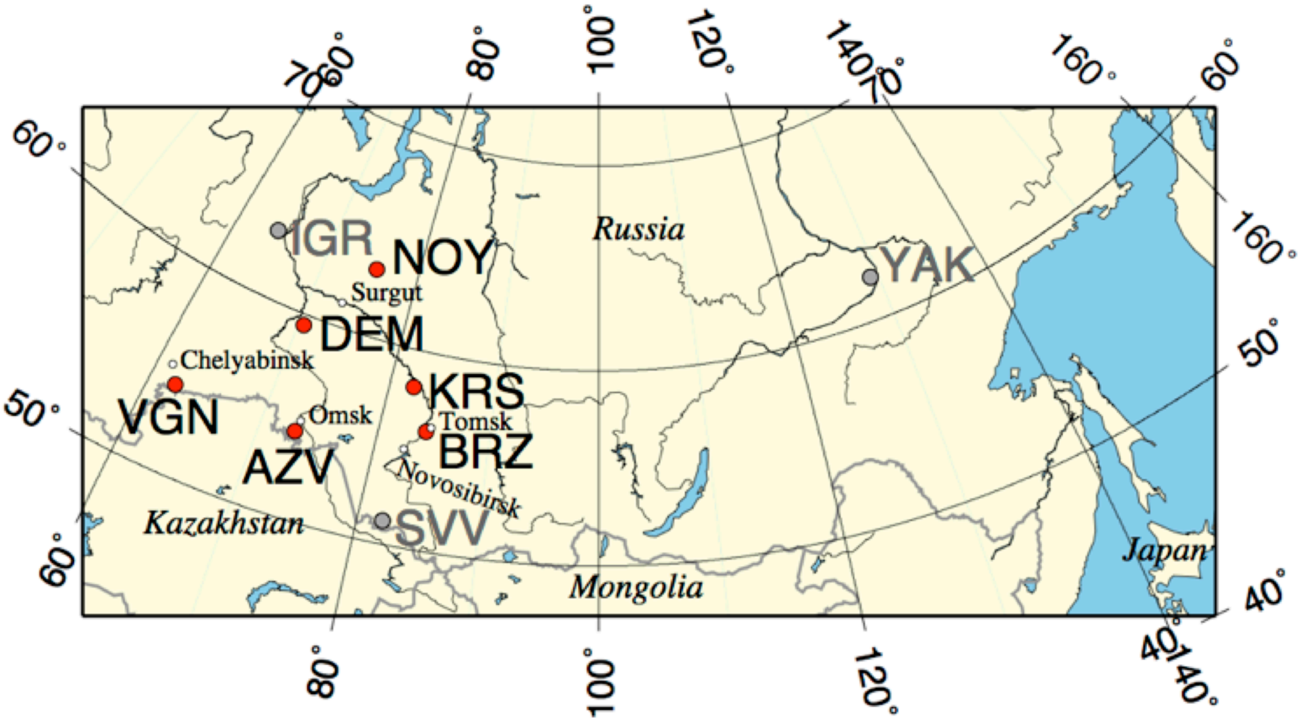
Keywords: Cabon Dioxide, Methane, Tower measurement, West Siberian Lowland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Omsk
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BRZ
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Novosibirsk
Kazakhstan
SVV
Russia
YAK
Mongolia
Japan
60°
70°
80°
100°
120°
140°
160°
50°
40°

Estimating vertical fluxes of ozone within the atmospheric boundary layer

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Investigation of the vertical distribution of ozone within the atmospheric boundary layer (ABL) was carried out by use of AN-2 light aircraft as a research platform. Vertical fluxes of ozone and their direction from the ground to the free-tropospheric level were calculated based on the in situ measurement data. Research flights have been performed over the greenhouse gas monitoring station located in a background area (56.1–56.4 N, 84.2–84.8 E) in the vicinity of abandoned village of Berezorechka (West Siberia). The schedule of diurnal flights was as follows: the first one just after the sunrise; the second one at noon; the third one 2-3 hours after noon, when a well-developed turbulence is observed; and the last one just before the sunset. A total of 10 diurnal cycles of measurements were undertaken.

Analysis of the obtained data showed that the rate of ozone influx from upper layers of the atmosphere is 3-10 times less than the ozone production rate in the ABL. Average rate of ozone influx from the free troposphere was about 1 mg m^{-3} h^{-1}, but ozone production rate in the ABL was about 5 mg m^{-3} h^{-1}, so the major part of ozone is formed by photochemical reactions that occur within the ABL and only 20 % of its content is determined by the influx from the free troposphere. The vertical profiles of the ozone fluxes have shown that their maximum values are observed at heights from 200 to 600 m AGL. The height of the maximum depends on the season: in winter it is lower than 200-300 m, and in summer the maximum is observed at 500-600 m. The value of the ozone flux maximum also depends on the season and varies from 1 μg m^{-2} s^{-1} in winter to 4.2 μg m^{-2} s^{-1} in spring.

This work was supported by the Russian Foundation for Basic Research (grant No 17-05-00374).

Keywords: Atmosphere, Ozon, Distribution

THE RELATIONSHIP BETWEEN OZONE FORMATION AND AIR TEMPERATURE IN THE ATMOSPHERIC SURFACE LAYER

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Studying the formation and dynamics of ozone in the atmosphere is important due to several reasons. First, the contribution of tropospheric ozone to the global greenhouse effect is only slightly less than that of water vapor, carbon dioxide, and methane. Second, tropospheric ozone acts as a strong poison that has negative effects on human health, animals, and vegetation. Third, being a potent oxidizer, ozone destroys almost all materials, including platinum group metals and compounds. Fourthly, ozone is formed in situ from precursors as a result of photochemical processes, but not emitted into the atmosphere by any industrial enterprises directly.

In this work, we present some results of the study aimed at the revealing relationship between ozone formation rate and surface air temperature in the background atmosphere. It has been found that this relationship is nonlinear. Analysis of the possible reasons showed that the nonlinear character of this relationship may be due to a nonlinear increase in the reaction constants versus air temperature and a quadratic increase in the concentration of hydrocarbons with increasing temperature.

This work was supported by the Russian Foundation for Basic Research (grant No 17-05-00374).

Keywords: Atmosphere, Ozon, Temperature

Characteristics of atmospheric wave-induced laminae observed by ozonesonde at the southern tip of South America

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We have performed ozonesonde measurements at Punta Arenas (53.14°S, 70.88°W) in Chile and Río Gallegos (51.61°S, 69.29°W) in Argentina since 2014 to investigate vertical structures in the edge region of polar vortex as well as to validate ozone profiles measured by a differential absorption lidar and a millimeter-wave radiometer located at Río Gallegos. In such a southern part of South America (Patagonia area), the National Meteorological Service of Argentina also has made regular ozonesonde measurements at Ushuaia (54.85°S, 68.31°W) in Argentina since 2008. In these ozone concentration profiles, small-scale fluctuations of the ozone volume mixing ratio with a layer of a few kilometers are frequently found. The lamina structure is formed by vertical displacement of isopleths due to gravity waves and by isentropic advection of a filament of vortex air due to Rossby wave breakings. In the Patagonia area, notable gravity waves are generated on the leeward of the Andes. The polar vortex could contribute to both the generation of the gravity waves by a spontaneous adjustment mechanism and the formation of large meridional ozone gradient associated with lamina due to the horizontal advection. To distinguish between the effects of the vertical displacement and the horizontal advection, we investigated correlation between the ozone and potential temperature fluctuations [Pierce and Grant, GRL, 1998; Thompson et al., JGR, 2011], which were obtained by applying a high-pass filter of 3 km to the ozone and temperature profiles. Monthly fractions of the ozone fluctuation resulting from the gravity waves and Rossby waves were derived for the altitude ranges of tropopause height to 15 km, 15–20 km, and 20–25 km. Additionally, we investigated the effects of the ozone fluctuation on total ozone column for the outer side, the edge region, and the inner side of the polar vortex.

Keywords: Ozone, Stratosphere, Gravity wave, Rossby wave

Comparison of ozone profiles from DIAL, MLS, and chemical transport model simulations over Río Gallegos, Argentina in the 2009 spring

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This paper evaluates the agreement of ozone profiles from the ground-based DIfferential Absorption Lidar (DIAL), the satellite-borne Aura Microwave Limb Sounder (MLS), and the 3-D chemical transport model simulations (CTM) over the South Patagonian Atmospheric Observatory (OAPA, 51.6°S, 69.3°W) in Río Gallegos, Argentina for the period from September to November 2009. We focus on this period, because a persistent ozone decrease for three weeks was found over the area around the southern tip in South America. Such the long-lasting decrease over the area was unusual (e.g., de Laat et al., GRL, 2010; Wolfram et al., Ann. Geophys., 2012). To compare the ozone profiles from DIAL with MLS retrievals, the averaging kernel matrix of MLS and the a priori profiles are used for converting the high-resolution DIAL profiles to the retrieval pressure levels of MLS. The comparison was done for pressure levels between 86 hPa and 5 hPa with the coincidence criteria of <500 km spatially and <24 hrs temporally. CTM used here incorporates a chemical module into MIROC3.2-GCM using horizontal winds and temperature nudged toward ERA-Interim data (e.g., Akiyoshi et al., JGR, 2016). The result shows a good agreement between DIAL and MLS with mean differences of ±0.1 ppmv (n=180), except for the 86 hPa level. CTM also agrees to DIAL with the mean differences of ±0.3 ppmv (n=23) between 56 hPa and 10 hPa. The root-mean-square differences increase with increasing altitudes from ˜0.5 ppmv at the 32-56 hPa levels to 1.3 ppmv at the 5 hPa level for both the two comparisons. Both of the two comparisons give mean differences of 0.5 to 0.7 ppmv at the 83 hPa level. It seems that DIAL tends to underestimate the ozone values only for this lower altitude region or some small scale differences in the ozone field cause the biases.

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Keywords: stratosphere, ozone, DIAL, MLS, MIROC, CTM

A long-term monitoring of trace gases in troposphere and stratosphere retrieved from high-resolution FTIR measurements at Rikubetsu in Hokkaido, Japan

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The Institute for Space-Earth Environmental Research (ISEE) of Nagoya University had carried out measurements of solar absorption spectra using a ground-based Fourier Transform InfraRed Spectroscopy (FTIR) instrument (Bruker IFS120M) at Rikubetsu observatory (43.46°N, 143.77°E, 380 m a.s.l.) in Hokkaido since 1995. In 2014, the National Institute for Environmental Studies (NIES) installed another high-resolution FTIR instrument (Bruker IFS 120/5HR), and started the cooperative measurements with ISEE. The spectrum is obtained in mid-infrared band with a resolution of 0.0035 cm^{-1}. The vertical distribution of trace gases in troposphere and stratosphere is retrieved from the measured spectrum by using the SFIT4 (version 0.944) software with uniform retrieval parameters recommended by the Infrared Working Group of the Network for the Detection of Atmospheric Composition Change (NDACC/IRWG). This is for the purpose of obtaining the homogeneous retrievals at the NDACC measurement sites. We use an initial profile of pressure and temperature taken from the NCEP Reanalysis dataset and those of the molecular species except for water vapor taken from the results of the WACCM V6 40-yrs run. The vertical profile of water vapor is retrieved from the spectrum before the retrievals of the other species. Using SFIT4, we have retrieved the total column amounts and vertical profiles of O_3, HCl, HF, HNO_3, $ClONO_2$, CH_4, C_2H_6, N_2O, CO, HCN, CCl_4, and find the various temporal variations and the long-term trends from 1995 to 2016. Although the observed partial column of stratospheric O_3 does not show any trend with significance, that in the troposphere seems to decrease in 2000s. In addition, after 2000, we find negative trends in the time series of HCl and $ClONO_2$ total columns, being consistent with the global results.
In the presentation, we report the features of temporal variations of the species in stratosphere and troposphere as well as the details of the retrieval.

Keywords: Atmospheric composition, Infrared spectroscopy, Trends

Detection and analysis of the short-term increases of column-averaged dry air mole fraction of CO (XCO) observed at Rikubetsu, Hokkaido in 2015 early winter

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The National Institute for Environmental Studies (NIES) has observed solar absorption spectra in near-infrared region with a high-resolution FTIR (Bruker IFS120/5HR) set up at Rikubetsu in Hokkaido (43.46°N, 143.77°E, 380 m a.s.l) as a site of the Total Carbon Column Observing Network (TCCON) since 2014, and retrieved the column-averaged dry air mole fractions of various species such as CO_2, CH_4 and CO. Although seasonal and short-term variations of the their species were observed, we focus on the short-term events that CO column-averaged mixing ratio (XCO) increased and decreased. In 2015 early winter, the observed XCO started increasing on 13 November, and became about 4.5 times larger than the seasonal mean value on 30. After that, on 9 December, it returned to the mean value. The same features also appear in CO_2 (XCO_2) and CH_4 (XCH_4), suggesting that this was caused by an air mass inflow from combustion sources. In order to understand the relation between the XCO enhancement and emission sources, we made trajectory analysis during the period of this event by using FLEXPART (Stohl et al. 2005) and NCEP CFSv2-6hour analysis dataset. In each day, we made the back-trajectory calculations in 2 weeks at the height of 1, 5 and 7 km, respectively. The results show that the air mass mainly comes from the North-east China and far distant regions. To evaluate influence on XCO in each region, we picked up the hotspots in the regions estimated above from the MODIS hotspot dataset, and made the forward-trajectory calculations from them. The result is that influence of the emission from the North-east China region was about 10 times larger than those in other regions. Thus, the observed increase of XCO is like to be the effect of air mass inflow from combustion source of the North-east China.

In this presentation, we discuss the comparison with other XCO-increasing events during the observation period and the relation among the species as well as the details of trajectory analysis.

Keywords: near-infrared solar absorption spectra, tropospheric carbon monoxide, trajectory analysis

Recent increase and subsequent decrease of the total column of ethane observed with FTIR at Tsukuba

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The total columns of C_2H_6 have been observed with a high-resolution Fourier transform spectrometer at Tsukuba, Japan since 2001. SFIT4 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.

Temporal variation of retrieved total column of C_2H_6 shows a little bit decrease (-0.4%/year) from 2001 to 2008, increase (2.2%/year) from 2009 to 2013, and again decrease (-0.6%/year) from 2014 to 2016. The last trend from 2014 may due to decrease in emission (natural gas and biofuel) related to the increase in the production of fossil fuel due to the fall of the price.

Keywords: FTIR, Greenhouse Gas, ethane

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

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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. First, we focused on 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 (the Aso volcano and the Sakurajima volcano) and the other from a continental origin (China and Korea). Based on these results, we performed quantitative analysis using data obtained from January 2014 to September 2016. The mean and the median of the daily maximum SO_2 concentrations in the period were 3.3 and 2.1 ppbv, respectively. We investigated the dependence of the observed SO_2 level on the size of the range to judge whether the observed air masses passed over volcanoes and found that the higher SO_2 concentrations were observed in air masses that passed closer to volcanoes. The mean of the daily maximum SO_2 values affected by the Aso volcano was about 6 ppbv, which is larger than that for the Sakurajima volcano (4 ppbv), suggesting that the influence of the Aso volcano was larger. To consider the continental effect, we analyzed relations between SO_2 and the length of residence time over China or Korea and between SO_2 and the time required for reaching Kasuga from China or Korea. However, no significant correlation was observed. In this case, the mean of the daily maximum SO_2 values was about 2.4 ppbv. The results were unchanged even if only influences of urban areas in China or Korea were considered. These results suggest that the impact of long-range transport of air pollution from China or Korea on enhancements in SO_2 concentration was smaller than that of volcanoes in Kyusyu. In this presentation, we will also discuss results of analysis including the explosive eruptions of the Aso volcano occurred on 8 October, 2016.

Keywords: SO2, MAX-DOAS, valcano

Comparisons of column-averaged dry-air mole fractions of greenhouse gases among GOSAT/TANSO-FTS SWIR, TIR, and NICAM-TM data

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Greenhouse gases Observing SATellite (GOSAT) was launched on 23 January, 2009. Thermal and Near-infrared Sensor for Carbon Observation Fourier Transform Spectrometer (TANSO-FTS) on board the GOSAT has SWIR and TIR bands and can observe column-averaged dry-air mole fractions of CO_2 and CH_4 (XCO_2 and XCH_4) in the SWIR bands [Yoshida et al., 2011] and CO_2 and CH_4 vertical profiles in the TIR band [Saitoh et al., 2009]. In this study, we calculated XCO_2 and XCH_4 values from the TIR CO_2 and CH_4 profiles, and then compared them with XCO_2 and XCH_4 data of the SWIR bands and Nonhydrostatic ICosahedral Atmospheric Model-based Transport Model (NICAM-TM) [Niwa et al., 2011]. Before calculating the TIR XCO_2 values, we applied bias-correction values evaluated based on the comparisons of aircraft CO_2 data.

We compared latitudinal distributions of XCO_2 among TANSO-FTS TIR, SWIR, NICAM-TM, and a priori (NIES-TM05) data [Saeki et al., 2013]. TIR XCO_2 data over the land in the Northern Hemisphere except the Sahara desert were slightly smaller than SWIR XCO_2 data and, in contrast, slightly larger over the land in the Southern Hemisphere. Over the Sahara desert, TIR XCO_2 data in the daytime were considerably smaller than SWIR and NICAM-TM XCO_2 data, which suggests that surface parameters used in the TIR retrieval had some problems. Over Hawaii where there is no strong CO_2 source, TIR XCO_2 data agreed with SWIR XCO_2 data to within 1% on average.

Keywords: GOSAT, XCO2, XCH4

Simultaneous estimate of surface temperature and surface emissivity from GOSAT/TANSO-FTS TIR spectra

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Thermal and Near-infrared Sensor for Carbon Observation Fourier Transform Spectrometer (TANSO-FTS) on board Greenhouse gases Observing SATellite (GOSAT) can observe column amounts and vertical profiles of CO_2 and CH_4 from short-wave infrared (SWIR) and thermal infrared (TIR) bands, respectively [Yoshida et al., 2011; Saitoh et al., 2009]. Uncertainties in surface parameters could affect retrieved CO_2 concentrations in the TIR retrieval [Saitoh et al., 2009]. In this study, we tried to estimate surface temperature and surface emissivity simultaneously from TIR spectra of GOSAT/TANSO-FTS, following the method proposed by Matsui and Moriyama [2008]. Their method utilized the relatively moderate wavelength dependence of surface emissivity compared to atmospheric gas absorption lines, and was applied to thermal infrared spectra with a high wavelength resolution obtained with Atmospheric Infrared Sounder (AIRS). We modified their method for the application to TANSO-FTS TIR spectra, and applied it to the spectra obtained over the oceans where the wavelength-dependence of surface emissivities is relatively known to examine the utility of the method.

We here defined wavelength channels with a transmittance above 0.95 without considering any continuum as a “smooth part” in the spectral range between 800 and 1000 cm^{-1}, and then estimated surface parameters on the basis of the “smooth part” channels. First, we applied the method to a TIR spectrum obtained over the ocean in mid-latitudes. We calculated surface emissivities by changing a surface temperature by 1 K in the range from 286 K to 310 K, and adopted the surface temperature when the calculated surface emissivities became smoothest with respect to wavelength. The estimated surface temperature was 294 K, which was close to the nearby MODIS sea surface temperature (SST) value (293.4 K). The simultaneously-estimated surface emissivity at 900 cm^{-1} was 0.97, which was also close to the sea surface emissivity of Aster Spectral Library, 0.99.

Furthermore, we applied our developed method to several TANSO-FTS TIR spectra obtained over the ocean (10–20°N, 160–170°E) on 1–3, January, 2013. We compared surface temperatures estimated by our method and nearby MODIS SST data. When their differences were small, the simultaneously-estimated surface emissivies were quantitatively close to the sea surface emissivities of Aster Spectral Library. However, when their differences were relatively large, the structures of the simultaneously-estimated surface emissivies were far from those of the sea surface emissivities. In the presentation, we will discuss the accuracy of our estimates of surface parameters, results of other seasons and regions, and the effect of changing several parameters in our method on estimated surface parameters.

Keywords: GOSAT, thermal infrared band, surface temperature, surface emissivity

Size-dependent wildfire occurrences in the boreal Eurasia and driving factors

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Large areas are burned every year in the boreal Eurasia. Pollutants emitted from fires in the Boreal Eurasia could be transported to the Arctic and the consequential deposition could accelerate the Arctic warming. However, there are so far limited knowledges on the wildfire pattern in the boreal Eurasia and the causes. Our purposes are to clarify the temporal and spatial pattern of wildfire and to investigate the factors affecting wildfire occurrence in the boreal Eurasia during 2005-2014.

Focusing on a domain in 50–75°N, 30–180°E, we divided the boreal Eurasia into 15 sub-regions and investigated the wildfire occurrences based on the Moderate Resolution Imaging Spectroradiometer MCD64A1 burned area product. Lands were burned mainly (96%) in >100ha size, although with large small fires (<100ha) counts (58%). Interannually, large land losses (>30 million hectares) occurred in 2008, 2010, 2012 and 2014 over the whole domain. Seasonally, wildfires occurred since April, peaked in July to August, and continued until October. Spatially, six high fire-prone regions were identified locating at the southwestern Russia, Kazakhstan, southwest Siberia, central Siberia, eastern Siberia and the Far East. Furthermore, we investigated the relations of burned area with climatic indices such as temperature, precipitation and soil drought index (Palmer Drought Severity Index, PDSI) in each sub-region. It was found that the burned area in southwest Russia, Kazakhstan, west Siberia had positive relation with temperature ($p < 0.05$). On the other hand, burned area had negative relations with precipitation and PDSI ($p < 0.05$) in most of the fire-prone regions. These results indicate that wildfire occurrence were fostered by the dry soil and air conditions. Furthermore, burned area is likely related to snow melting date, to which early snow melting date caused larger burned area. Our study implies that under a warmer world, wildfires in the boreal Eurasia tend to be severer and mega-fires more frequent.

Time-resolved measurements of organic compounds in particles during heterogeneous reaction of volatile organic compounds with primary and secondary aerosols

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Volatile organic compounds (VOCs) undergo oxidation in the gas phase to yield semi-volatile organic compounds (SVOCs) and secondary organic aerosols (SOAs), which directly and indirectly affect the global climate as well as human health. Quantification of the impacts of SOAs requires understanding their chemical composition and processes of formation as well as mass yields. In particular, because a systematic underestimation of simulated SOA production increases with air mass ageing, speciation of the SVOCs produced by gaseous oxidation is essential. In addition, information about the partitioning of each SVOC between the gaseous and condensed phases as well as the reactions of the condensed SVOCs within the particulate phase is important for the description of SOA formation. Recently, we succeeded in a time-resolved measurement of organic compounds in particles by using proton transfer reaction mass spectrometry. In this study, changes of organic compounds in diesel exhaust particles and SOAs by an addition of some kinds of VOCs through heterogeneous reaction were investigated.

Keywords: Proton transfer reaction-mass spectrometry, Oligmer, Heterogeneous reaction, Secondary organic aerosol, Diesel exhaust particle

Hygroscopic characterization of individual cloud condensation nuclei with atomic force microscope

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Majority of atmospheric aerosols takes up water and can eventually act as cloud condensation nuclei (CCN). How these particles interact with water therefore, regulates the climate and hydrological cycle. The hygroscopicity parameter κ is a single parameter that represents hygroscopicity of aerosol particles (Petters and Kreidenweis., 2007). In order to physically derive κ, it is necessary to obtain precise measure of the relative humidity, dry CCN size, hygroscopic growth factor (gf) and surface tension of the solution/air interface. Previously, κ of a given aerosol population has been derived using a hygroscopic tandem differential mobility analyzer (HTDMA).Although HTDMA has been widely accepted as a powerful tool to measure aerosol hygroscopic behavior, the measurement reflects an overall average of numerous particles and does not necessarily preserve extremes of the distribution. Therefore, it is also important to carefully examine particle hygroscopic growth at an individual particle level (Morris et al., 2016). Another limitation is that, previous works mainly assumed the surface tension of pure water or bulk solution, since there was no conventional method for directly measuring the surface tension of the droplets. Here, we present our attempts to combine methods to directly measure the surface tension and gf of a single droplet by atomic force microscopy (AFM), which in principle allow us to directly derive κ of individual particles. In this study, we measured gf and surface tension of sodium chloride (NaCl). The gf (80% RH) derived in this experiment (1.73±0.35) coincides with the value of the previous work (Morris et al., 2016) within the standard deviation. In terms of the surface tension, the retention force required for deriving the surface tension was found to be 19±3 nN, which is slightly smaller than the reported value of 30 nN (Morris et al., 2015), the difference can be attributed to the different shape of the cantilever used in both studies. Hygroscopicity parameter κ was derived from gf, dry particle diameter and surface tension obtained in this experiment and found to be 1.10. The fact that current value coincided with the values obtained in the previous study (Koehler et al., 2006; Clegg and Wexler., 1998), supports the validity and potential of this new method. In the future, other substances having various surface activities and mixing states (mineral salt and organic species) will be tested using this method, and further applied to actual atmospheric aerosol particles.

Impact of aerosol composition on the oxidation of bisulfite during the reactive uptake of nitrogen dioxide on aqueous aerosols

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Multiphase chemistry plays a vital role in global atmosphere. The importance of multiphase chemistry in the lower troposphere has been recently underscored by the severe haze-fog pollution episodes experienced over China megacities. A key finding is that despite reduced photochemistry under the haze, the oxidation of sulfur dioxide (SO_2) into sulfate aerosol remains unabated in the presence of low levels of ozone. The main oxidant under such conditions is the nitrogen dioxide (NO_2) emitted by motor vehicles and other combustion sources. The dark conversion of bisulfite (HSO_3^-) into bisulfate (HSO_4^-) by NO_2 has been ascribed to a heterogeneous process taking place on the neutral aerosol prevalent in Northern China. Previous studies in our laboratory, however, have shown that anions catalyze the disproportionation of NO_2 into NO_2^- + NO_3^-.This suggests that coexisting anions in aerosol and aerosol acidity could impact the heterogeneous oxidation of bisulfite during the gaseous NO_2 uptake on aqueous aerosols.

This work explores the contribution of aerosol composition to the oxidation of HSO_3^- during the reactive uptake of NO_2 on the surface of aqueous solutions. The aqueous aerosol surface is generated by microjets containing chloride ion (Cl^-) or formic acid (HCOOH) as proxies of ambient inorganic and organic components. The surface is instantaneously exposed to NO_2(g) followed by in situ anion analysis via online electrospray ionization mass spectrometry. The heterogeneous reaction occurs in ~1nm interfacial layers of the aqueous aerosol under NO_2 exposures similar to atmospheric conditions. The competition between the disproportionation process controlled by aerosol anion compositions, and the oxidation of bisulfite will be followed by the HSO_4^-/NO_3^- ratio. We will review other significant pathways for the oxidation of bisulfite under atmospherically relevant condition. This work addresses how the reactive uptake of gaseous pollutants affects the aerosol composition, thereby contributing to understand the mechanisms underlying urban pollution in haze-fog episodes.

Keywords: Haze-fog pollution, nitrogen dioxide, sulfate, sulfur dioxide, China megacities

Characteristics of Cloud Condensation Nuclei over the Urban Areas Measured at Tokyo Skytree

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Atmospheric aerosols influence the Earth’ s radiation budget directly through absorption and scattering of solar radiation, and indirectly by modifying the properties of clouds. However, climate models still face large uncertainties as a result of insufficient understanding of aerosol-cloud interactions[1]. Thus, further studies on aerosol-cloud interaction are needed to improve climate models and to make better predictions of climate change. To investigate cloud condensation nuclei (CCN) characteristics over the urban areas, where few fixed-point observations are conducted in the world, CCN properties of atmospheric aerosols were observed at the Tokyo Skytree (TST) in Tokyo.

Measurements were carried out at TST (35.71°N, 139.81°E, 458m,a.g.l), from 3 to 30 June 2016. In this study, aerosol number size distribution, condensation nuclei (CN) number concentrations (N_{CN}), CCN number concentrations (N_{CCN}) with four supersaturations (SS%) were measured. Fog droplet size distributions and fog droplet number concentrations (N_{Fog}) were also measured. Using these data, we obtained the hygroscopic parameter (κ) and the effective radius (R_{eff}) of fog droplets.The results obtained at TST were compared with observation data of the summit of Mt. Fuji[2] where similar observations were made.

The average value of N_{CCN} at TST was larger than that of the summit of Mt. Fuji.[2]. The average value of κ at TST was smaller than that of the summit of Mt. Fuji . The CCN properties of aerosols did not vary significantly with air mass origins. As a result of the CCN characteristics of the period covered with clouds without drizzle, there was no positive correlation between the N_{Fog} and N_{CCN}. Also, there was no negative correlation between R_{eff} and N_{CCN} like the microphysical relationship known as the Twomey effect[3]. This is considered to be due to the small variation of N_{CCN} at TST. The relationship between R_{eff} and N_{CCN} under SS0.12% could be regarded as part of the negative correlation between R_{eff} and N_{CCN} obtained at the summit of Mt. Fuji .

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Keywords: cloud condensation nuclei number concentration, condensation nuclei number concentration, effective radius, fog droplet number concentrations

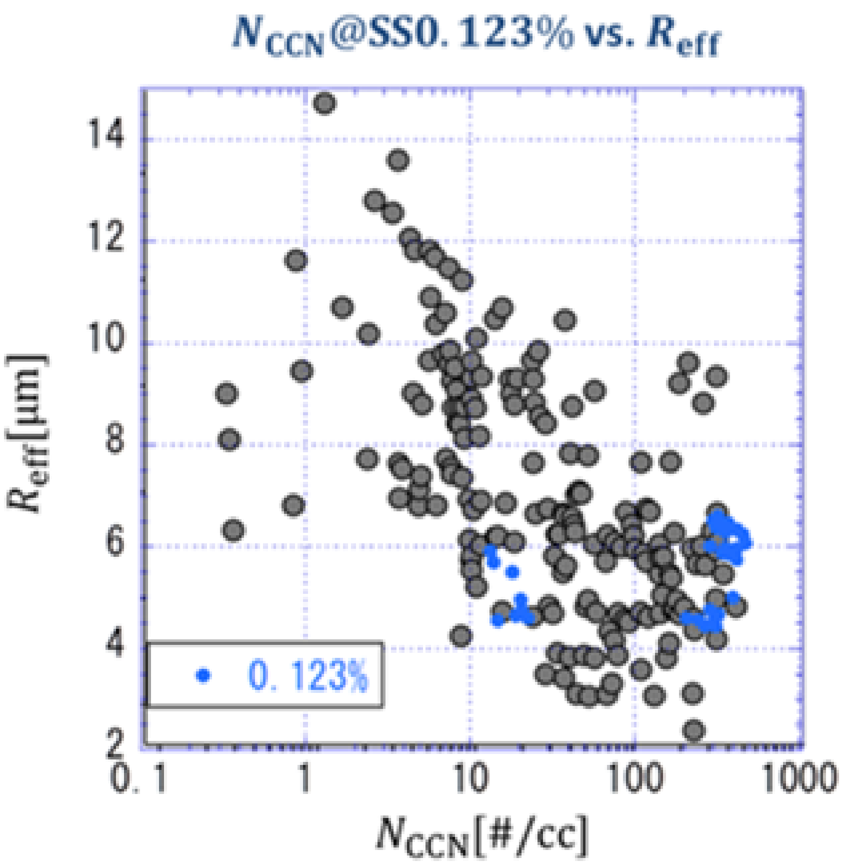

図　R_{eff}とN_{CCN}@SS0.123%（青）、
N_{CCN}@SS0.15%（渡辺, 2015）（黒）の関係

Wet removal mechanisms of black carbon observed in Tokyo and Okinawa

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Quantitative understanding of wet removal process of black carbon (BC) is important because it controls temporal and spatial distribution of BC in the atmosphere. There are two categories of wet removal mechanisms: nucleation scavenging and impaction scavenging. Theoretically, the removal efficiencies of both mechanisms depend on microphysical properties (size(D_{BC}), coating thickness, and hygroscopicity) of BC. However, the relative contribution of these mechanisms to the removal of BC and the key BC microphysical property controlling the wet removal efficiency of BC have never been tested by direct observation. In this study, we conducted ground-based observations of BC in air and in rainwater in Tokyo and Okinawa, using a new observational method (Mori et al., 2016). We compared the observed size-dependent wet removal efficiency of BC particles ($RE(D_{BC})$, defined as the ratio of the measured size distributions of BC in rainwater to that of BC in ambient air) with the size-dependent number fractions of BC particles scavenged by nucleation ($F_{ccn}(D_{BC})$) and impaction ($F_{imp}(D_{BC})$) mechanisms. The $F_{ccn}(D_{BC})$ and $F_{imp}(D_{BC})$ were estimated from the observed microphysical properties of BC in the air before precipitation starts and the observed droplet size and intensity of precipitation.

The size dependence of $RE(D_{BC})$ showed remarkable differences for the observed 42 precipitation events (31 events in Tokyo and 11 events in Okinawa). The size dependence of $RE(D_{BC})$ for BC-containing particles with $D_{BC} > 100$ nm was successfully explained by the size dependence of $F_{ccn}(D_{BC})$, whereas the contribution of impaction mechanism can be dominant for particles with $D_{BC} < 100$ nm. For $D_{BC} > 100$ nm, when BC particles have relatively thick coating (shell/core ratio > 1.2), the $RE(D_{BC})$ depended little on D_{BC} and the major fraction of BC were removed via nucleation scavenging. On the other hand, when BC particles are nearly bare or have less coating, the $RE(D_{BC})$ highly depended on D_{BC}, and RE (i.e. F_{ccn}) is more sensitive to the hygroscopiticy of coating materials and the maximum supersaturation of water vapor that BC particles would experience during moist convection process. Our results show that the coating thickness of BC is the key parameter controlling wet removal of BC, and indicate that for accurate simulation of vertical transport of BC from the boundary layer to the free troposphere, detailed modeling of microphysical properties of BC and atmospheric supersaturation is required especially for BC particles with thin coatings.

Keywords: Black carbon, Wet removal mechanism, Field observation

Ship-based observation of black carbon at North Pacific Ocean: Analysis of transportation event at January on 2016

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Ship-based measurement for black carbon (BC) and carbon monoxide (CO) was conducted on board the R/V Mirai from 18 at North Pacific Ocean (12.9 °N, 130.5 °E) to 24 at Yokohama Port (35.4°N, 139.7°W) January 2016. Measurements of atmospheric BC particles and CO were conducted using a single particle soot photometer (SP2) instrument and CO analyzer, respectively. Ambient particles and gases were sampled on the flying bridge, ~18m above sea level. From 18 to 19 January 2016 (around 20°N and 135° E), high concentrations of BC and CO were observed. It was suggested the contribution from the Asian continent air mass by backward trajectory analysis. In spite of almost same source, it was indicated that different BC size distribution and mixing state at air mass in 18 and 19 January plumes, suggesting the air masses would undergo different removal processes. In the presentation, we are going to discuss the comparison with CO and other event cases in the cruise.

Keywords: Black carbon, single particle, Ship-based measurement