Validation of MODIS MCD64A1 burned area in boreal Eurasia

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Biomass burning plays an important role in affecting vegetation dynamics, biogeochemical cycle of carbon, nitrogen and other elements, atmospheric chemistry and the climate. Boreal Eurasia is one of the most important regions where large areas are burned every year. Numerous particles and greenhouse gases are emitted from these fires. These pollutants could be transported to the industrialized region in East Asia, the western North Pacific and the Arctic. Atmospheric transport of fire-emitted pollutants to the Arctic and the consequential deposition is believed to accelerate the arctic warming. To better understand the effect of fires in boreal Eurasia on the arctic, an accurate assessment of burned area from the boreal Eurasia is necessary.

The Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 5.1 direct broadcast monthly burned area product MCD64A1 is widely used for global burned area mapping. MCD64A1 data was also used for the estimation of global fire emissions such as Global Fire Emissions Database, Version 4 (GFED4). However, uncertainties in burned area estimations could be introduced due to the “moderate resolution” character of MCD64A1 (~500m). Therefore, a comparison of the burned areas of MCD64A1 with those generated from higher resolution satellite products could provide basic and crucial information for its accuracy assessment and further applications. However, there are few studies on the validation of MCD64A1, especially in the boreal Eurasia.

In this work, we used Landsat 7 surface reflectance, along with a few commercial satellite scenes from WorldView, GeoEye and RapidEye as the reference scenes to derive the burned areas in a burning season in 2012 (mostly July to September). A wide range of ecotypes over wide geographic regions spanning from the western Russia/Kazakhstan to the eastern Siberia were covered. Each of these burned areas was compared with the corresponding region of MCD64A1 for the same periods. Our preliminary results indicated that MCD64A1 could well capture the large fires, while those less than 100 ha are prone to be undetected. We also found that MCD64A1 tends to underestimate the burned area in general. Based on the error statistics, we suggested the accuracy levels and precautions for applications in each ecotype.

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

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

Acknowledgements
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Keywords: GOSAT, CH4, evaluation

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

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

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

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

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

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

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The GOSAT (Greenhouse gases Observing SATellite) was launched in January 2009 and has been operating jointly by MOE, JAXA and NIES. GOSAT is the world’s first spacecraft designed specifically to measure the concentrations of carbon dioxide and methane, the two in the major greenhouse gases. Currently, GOSAT-2 is under development. Many satellite data have been compared to ground-based data, which have higher accuracy and precision. Satellite and ground-based instruments generally observe target quantity with different spatial scale/position and different time. Therefore, it is important to find more plausible coincidence criteria between them to validate satellite data.

The SWIR (Short Wavelength InfraRed) surface scattered solar spectra observed by the TANSO-FTS (Thermal And Near infrared Sensor for carbon Observation –Fourier Transform Spectrometer) onboard GOSAT is used to retrieve column-averaged dry-air mole fractions of carbon dioxide and methane (XCO_2 and XCH_4). TCCON (Total Carbon Column Observing Network) is a ground-based observation network using Bruker IFS 120HR/125HR high-resolution fourier transform spectrometers. The GOSAT data, XCO_2 and XCH_4, has been validated using TCCON data. However, GOSAT and TCCON data are not completely spatio-temporally matched due to characteristics of the satellite orbit.

For example, one of the co-location methods uses a geometric distance between GOSAT and TCCON data to obtain matched data (Geophysical co-location method, e.g., Morino et al., 2011). Validation of GOSAT requires statistically significant match-up number but there is not enough match-up number with geophysical co-location method. In the case of XCO_2, various methods for increasing the number of match-up data have been developed: the same potential temperature field at 700-hPa as a proxy for equivalent latitude for CO_2 gradients (Keppel-Aleks et al., 2011, Wunch et al., 2011), and the same concentration field predicted or assimilated with the atmospheric transport model (Guerlet et al., 2013). For the time gap, it has been used the same day or within the time range of GOSAT overpass time because GOSAT has three-day revisit and sun-synchronous orbit with a local time around 13h at descending. Increasing the number of match-up data in consideration of the gaps of time and space is important for advanced validation.

In this study, we develop a new match-up method utilizing the forward and backward trajectory analyses from GOSAT observation position by HYSPLIT model. The new method is applicable to other gases not only XCO_2 using flow of air masses. We show the result of analyzed bias variation with the spatio-temporal gap and the effectiveness of the developed method together with results using other match-up methods.

Keywords: GOSAT, TCCON, match-up

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

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

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

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

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

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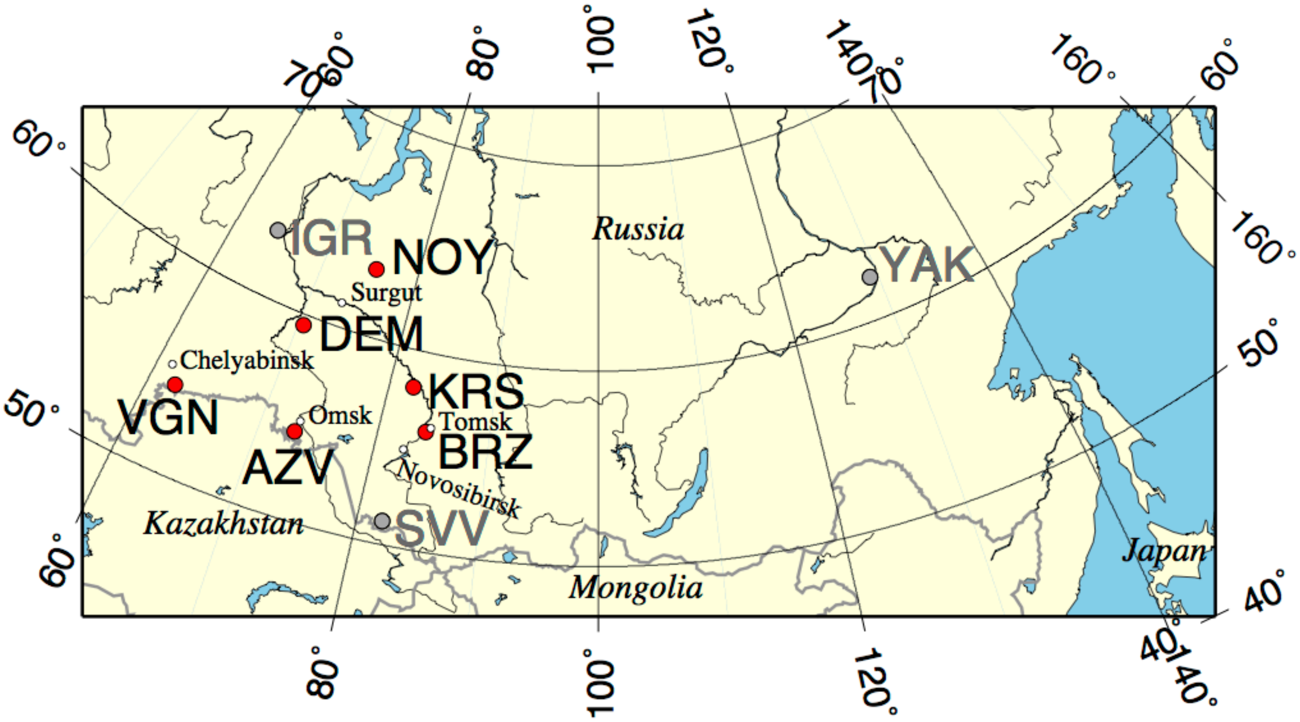
Keywords: Siberia, Carbon dioxide, Methane, tower observation, CRDS



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

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

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

Keywords: FTIR, Greenhouse Gas, ethane

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

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

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

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

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

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

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

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. This study focuses on continuous data acquired in July-August 2014. In the period, seven days were identified as the SO_2 level was high (>5 ppbv). Back trajectory analysis indicates that there were two categories; the one from a volcanic origin and the other from a continental origin. A high SO_2 plume observed on July 8 was found to originate from Sakurajima volcano. Plumes with SO_2 higher than 10 ppbv observed on July 30 and August 7 and 13 were from the Aso volcano. High SO_2 observed on August 14 could be affected by both volcanoes. On the other hand, high SO_2 plumes observed on July 12 did not pass over volcanoes but over China for about 1 day. Very high SO_2 of 20 ppbv was observed on July 21. This is suggestted to be due to a rapid transport of high SO_2 plumes from South Korea.

Keywords: SO2, MAX-DOAS, valcano

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

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

Keywords: MAX-DOAS, PV, NO2, HCHO

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

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1. Introduction

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

2. Experimental Apparatus

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

3. Results

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

Keywords: Aerosol, Nanoparticle

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

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New particle formation (NPF) is an important source of atmospheric aerosol particles, and may influence to regional/global climate and air quality through a variety of physical and chemical processes. Therefore, understanding of these processes is important. The NPF events have been observed in many different atmospheric environments (e.g., urban, forest, and mountains) and laboratory experiments. But their formation mechanisms are still poorly understood. In this study, field observations of aerosol particle number distributions and gaseous precursors were conducted at two sites: one in an urban area in Kyoto city, and the other in an observational site of Field Museum Tamakyuryo (FM Tama) of the Tokyo University of Agriculture and Technology in Tokyo, in summer to investigate factors controlling NPF.

Observations were performed at the Yoshida campus of Kyoto University, Kyoto (between 19 August and 11 September, 2013), and at the FM Tama, Tokyo (between 24 July and 8 August, 2015). Particle size distributions were measured with a scanning mobility sizer (SMPS; TSI) in both sites. Volatile organic compounds (VOCs) concentrations were measured using a proton transfer reaction mass spectrometer (PTR-MS; IONICON). Other gaseous components such as sulfur dioxide (SO_2) and ozone (O_3) were also measured simultaneously. In this study, we categorized the observed NPF events as a burst of nucleation mode particle (below 30 nm particle diameter) with (Case 1) or without (Case 2) subsequent particle growth, based on Maso *et al.*(1).

The NPF events corresponding to increases in SO_2 or VOC concentrations were observed at least 7 times during the observation period at Kyoto. On the other hand, no NPF events were observed in the 16 observation days at FM Tama. Although no significant differences in total particle surfaces and SO_2 concentrations in two sites were observed, isoprene concentrations in FM Tama were significantly higher than monoterpenes concentrations during daytime. In this presentation, differences in the particle size distributions and concentrations of possible precursor gaseous between NPF and non-NPF events will be discussed.

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

Observation of new particle formation in Noto peninsula

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Introduction

The process by which new particles (Ultrafine particles having diameter of the range from several nm to several tens of nm) are formed from precursor gases in the atmosphere is one of the main factors that affect the number size distribution of aerosol particles. Since new particle formation is a first step for the aerosol particles to take shape and eventually act as Cloud Condensation Nuclei (CCN), it has important implications in understanding contributions of aerosols on the climate. Recently, studies in various observation sites have pointed out conditions favorable for the new particle formation (NPF). For example, if there are a lot of existing particles in atmosphere, NPF is less likely to occur (Kerminen et al.,2001), because gas molecule preferentially condense to existing particle. However, there have been few reports so far on the seasonality of NPF in coastal East Asia.

Method

We selected Noto Ground-based Research Observatory (NOTOGRO), located at the tip of the Noto Peninsula as observation site mainly for the following reasons. Firstly, the station is away from any densely populated areas such as Kanazawa city, where one can expect less anthropogenic effect from nearby cities. Secondly, we may be able to observe influence caused by the unique monsoon facing the coast of the Japan Sea. Number size distributions of aerosol particles were measured using a scanning mobility particle sizer (SMPS) system that consists of a differential mobility analyzer (DMA, Model 3081) and a condensation particle counter (CPC, Model 3776). The field measurement was conducted from September 2012 to October 2014. In addition, SO_2 gas was continuously measured using (Model, 43B SO_2 Analyzer), from which we calculated H_2SO_4 proxy (Petäjä et al.,2009).

Result&Discussion

Analysis of aerosol number size distributions confirmed that NPF events occurred frequently throughout the year. From two years of observation, some similarities were found in the seasonality of the occurrence frequency. We focused on the variation of SO_2 gas which is considered as the main precursor of aerosol particles. Comparison with the number size distributions revealed that, the values of the H_2SO_4 proxy were high in most cases when NPF events occurred. The daytime maximum values of the H_2SO_4 proxy were extracted and compared among the days with and without NPF. As a result, the values of H_2SO_4 proxy were relatively higher when the event occurred. These results suggested that the conditions favorable for the formation of sulfuric acid vapor are the major factors controlling the NPF in the studied region.

Keywords: aerosol, new particle formation

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

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Angular distribution of scattering is one of the important optical properties contributing to the radiation balance in the Earth’s atmosphere. Therefore, accurate description of the single-scattering properties of aerosol particles is required. The angular distribution of light scattering is an important optical property. In addition, because the angular distribution of scattering depend on size, shape, and refractive index of particles, the angular distribution for individual aerosol particles may provide useful information to estimate mixing state and type of particles. In the present study, applicability of the angular distribution for individual particles to the estimation of mixing state and type of particles was examined by comparing the temporal variations of the angular distributions with those of chemical compositions for ambient particles.

We have developed a new polar nephelometer, which can measure angular distribution of the optical light scattered by an individual particle.1) Laser beam at 532 nm from a 300 mW YAG laser was used as light source. The laser beam intersects with a stream of aerosol particles introduced with a sheath flow using a double pipe. There are 21 photodiode detectors arrayed in each plane, totaling 42. Detector apertures were placed to limit sensing angles and minimize background light scattered from walls. In this system, the angular distributions for an incident light polarized parallel and perpendicular to the scattering plane can be measured simultaneously.

Ambient particles were measured at the Higashiyama campus of Nagoya University (35°09¢N, 136°58¢E, 60 m above sea level) located in an urban area in Nagoya, Japan, from July 1st to 7th, 2015. After being dried by a diffusion dryer with silica gel, ambient particles were introduced alternately to a heated line (maintained at 300°C) or the bypass line to measure, respectively, ambient particles directly and after the evaporation of volatile materials under high temperature conditions. The lines were switched every 15 min using two-way ball valves. Then, the particles were passed through a differential mobility analyzer (DMA) and introduced into the polar nephelometer. The DMA voltage was switched to select the nearly monodispersed particle with a mobility diameter 300 or 500 nm every 30 min. Most of organics, sulfate, nitrate, and ammonium are expected to be vaporized at temperatures below 300°C.2) Therefore, black carbon (BC) particles are expected to be mainly measured when particles were passed through the heated line. During the observation, mass concentrations of BC were measured using a particle soot absorption photometer (PSAP) combined with a heated inlet (maintained at 300°C). The mass concentrations organics, sulfate, nitrate, and ammonium were measured using a time-of-flight aerosol mass spectrometer (AMS). In the presentation, relationship between the temporal variations of scattering angular distributions of individual particles and those of chemical compositions will be discussed.

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Keywords: scattering angular distribution, polar nephelometer, chemical compositions

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

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

Keywords: Black carbon, Fluorescent particles, Mixing state

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

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

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

Study of aerosols and clouds using sky radiometer of SKYNET

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

Keywords: SKYNET, aerosol, cloud

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

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

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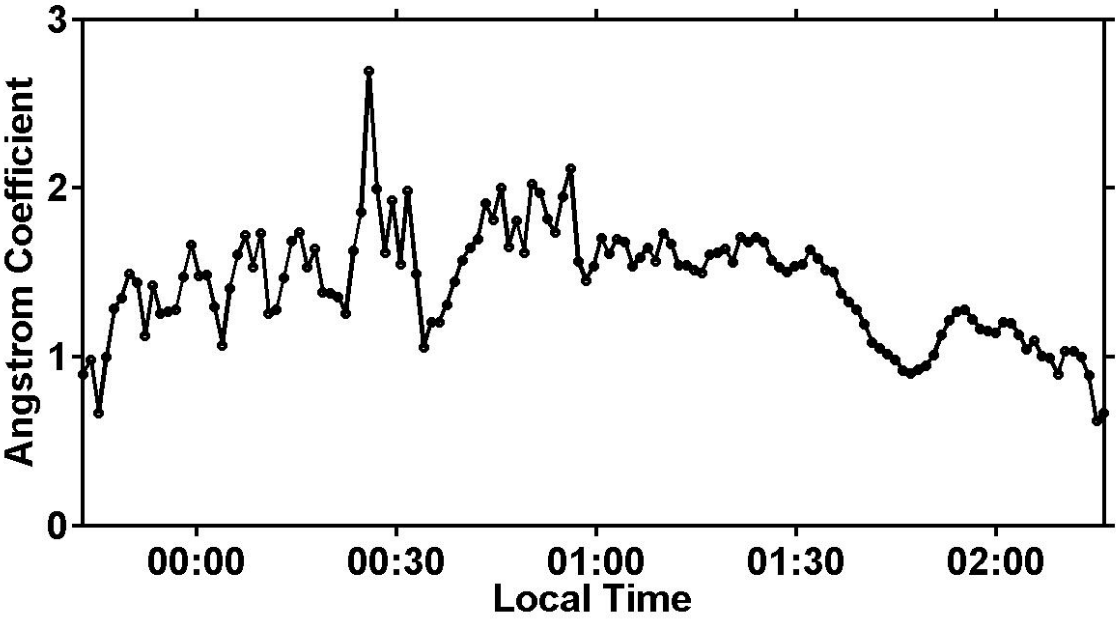
Keywords: Angstrom coefficient, Aerosols, Optical thickness, Relative Humidity



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

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

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