

Data composite of airborne sulfur dioxide measurements in the upper troposphere

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Sulfur dioxide (SO₂) 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 SO₂ 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 SO₂ 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 SO₂ distribution at mid-latitudes, tropical and polar regions. Median SO₂ background mixing ratios in the UT range between 10-25 ppt, however, also areas with strongly enhanced SO₂ mixing ratios in the UT were observed, associated with events of new particle formation. The SO₂ sources and transport pathways for these specific observations will be discussed.

Keywords: sulfur dioxide, aircraft measurements, new particle formation

Temporal characteristics of CH₄ vertical profiles observed over Surgut (Novosibirsk) from 1993 (1997) to 2014 in West Siberian Lowland

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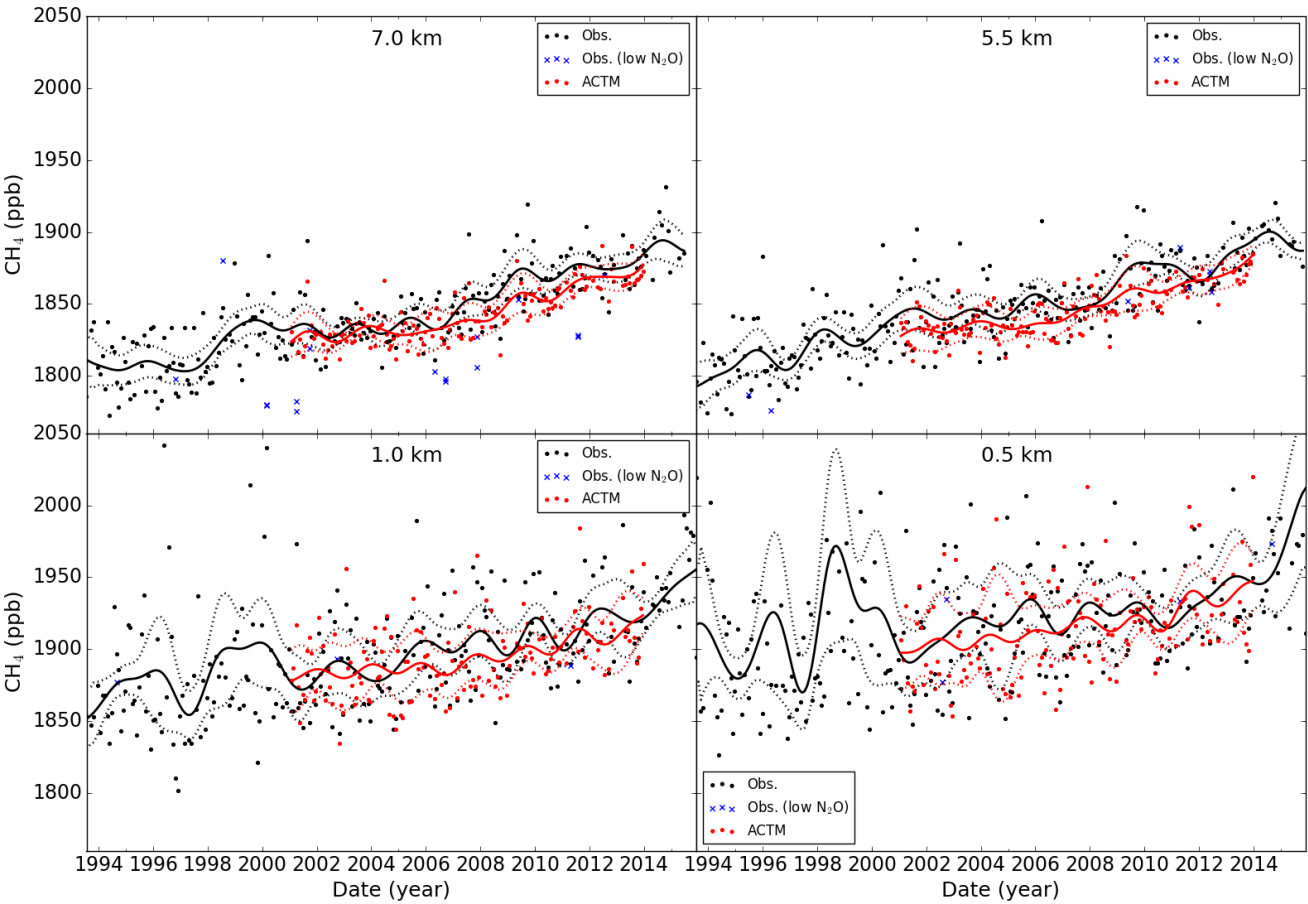
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We have carried out monthly flask sampling with aircraft over the boreal wetlands in Surgut and a pine forest nearby Novosibirsk, both located in West Siberian Lowland (WSL). Vertical profiles of methane (CH₄) concentration were measured up to about 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 CH₄ emissions from the wetlands in the WSL likely have exceeded zonal-mean CH₄ 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 CH₄ gradient and its variability. The vertical gradient at Surgut has been decreasing; the annual mean CH₄ 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 CH₄ vertical gradient appeared at Novosibirsk. An atmospheric chemistry-transport model simulation captured the observed decrease in the vertical CH₄ gradient at Surgut, when CH₄ 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 CH₄ vertical profile over West Siberia enabled us to detect variation in its emissions from the WSL and the surrounding land regions.

キーワード：メタン、航空機観測、西シベリア低地帯

Keywords: Methane, Aircraft observation, West Siberian Lowland



Climatological variations of tropospheric CO₂ over the Asia-Pacific region observed by the CONTRAIL commercial airliner measurements

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We present spatial and temporal variations of tropospheric CO₂ over the Asia-Pacific region analyzed from 10 years of the CONTRAIL commercial airliner measurements. The CONTRAIL flights with the high-frequency CO₂ measurements have covered large part of the Asia-Pacific region. Here we address climatological variations of CO₂ 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 CO₂ 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 CO₂ emissions up to the free troposphere. (2) The region-wide CO₂ decrease is obvious across the Asia-Pacific region and it is principally composed of CO₂ drawdowns originating in two distinct regions: boreal Eurasia and South Asia. We observed seasonally largest variability of CO₂ 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 CO₂ 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 CO₂ 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 CO₂ fluxes in East Asia in spring and those in South Asia in late summer.

Keywords: Carbon dioxide, Aircraft observation, Asian monsoon

北半球大気中 N_2O アイソトポキクル比の最近の経年変動

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 $^{15}\text{N}/^{14}\text{N}$ and $^{18}\text{O}/^{16}\text{O}$ ratios but also site-specific $^{15}\text{N}/^{14}\text{N}$ 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 latitude and two sites in the high latitude 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 ($\delta^{15}\text{N}^{\text{bulk}}$) 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 $\delta^{15}\text{N}^{\text{bulk}}$ has been enhanced since around 2010 at all the three sites, and that vertical gradient of the mixing ratio and $\delta^{15}\text{N}^{\text{bulk}}$ 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.

キーワード：温室効果気体、対流圏、安定同位体解析

Keywords: Greenhouse gas, Troposphere, Stable isotope analysis

Emissions of CO₂, CO, and CH₄ 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 (CO₂, CH₄, 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 CO₂, CO, and CH₄ during the CO enhancement events. Strong correlation was found between these gases, and the calculated emission ratios of CO/CO₂ suggested large contribution of peat forest fires to the observed enhancements (CO/CO₂ = 135 ppb/ppm). We determined the emission factors (EF) of CO₂, CO, and CH₄ from the peat forest fires using the emission ratios of CO/CO₂ and CH₄/CO₂, showing the EFs of 1722, 132.0, and 6.27 (g/kg) for CO₂, CO, and CH₄, 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 CO₂, CO, and CH₄ respectively. These EFs were consistent with each other except CH₄, suggesting that the EF used in GFED4S is overestimated for CH₄ from peat fires.

キーワード：温室効果気体、東南アジア、バイオマス燃焼

Keywords: Greenhouse gases, Southeast Asia, Biomass burning

北太平洋とその縁辺海で採取された海洋性エアロゾルの個別粒子分析 Individual Particle Analysis of Marine Aerosols Collected over the North Pacific and its Marginal Seas

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大気中のエアロゾル粒子は太陽放射を吸収・散乱することや、雲核として働き雲を形成することにより気候
に影響を与える。しかしながら、大気エアロゾルの挙動についての科学的理解度は低い（IPCC, 2013）。本研
究では、日本近海と外洋で採取したエアロゾル粒子に対して電子顕微鏡を用いた個別粒子分析を行い、海域に
よる粒子の組成や形態の違いを明らかにすることを目的としている。

試料採取はKH-13-7 航海（太平洋：2013/12/11-2014/2/12）、KH-14-3 Leg2 航海（太平
洋：2014/7/17-8/11）、KS-16-8 航海（日本近海：2016/7/5-7/13）並びにMR16-06 航海（ベーリング
海・北極海：2016/8/22-10/5）において行った。採取した個々のエアロゾル粒子に対して、透過型電子顕微
鏡を用いた形態観察と、エネルギー分散型X線分析器を用いた元素分析を行った。

外洋において採取したサンプルは変質していない海塩粒子によって80%以上が占められていた。一方、八丈
島沖において採取した2サンプルのうち1サンプルは変質していない海塩粒子によって大部分を占められていた
が、もう1サンプルは変質海塩と硫酸塩によって約90%以上を占められていた。これは梅雨前線の南下に伴う
汚染気塊の流入が原因であると考えられる。講演では、北極海等において得られたサンプルの分析結果につい
ても報告する予定である。

キーワード：大気エアロゾル、海塩粒子、硫酸塩、硝酸塩、TEM、EDX

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

衛星からの対流圏NO₂カラム濃度に対するMAX-DOAS観測からの検証と融合解析

Evaluation of satellite-based tropospheric NO₂ vertical column density using MAX-DOAS observations and their integrated analysis

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自動車・発電所などの人為発生源や、森林火災・雷放電から大気中へ放出される窒素酸化物(NO_x)の主要な成分であるNO₂は、現在もっとも感度よく宇宙から観測できる大気汚染マーカー分子として位置付けられ、最近では、衛星からNO₂濃度の空間分布や10年スケールの変動がとらえられるようになってきた。しかしながら、その対流圏鉛直カラム濃度の報告値は、都市周辺部では真値の約半分にとどまるなど、大きな負のバイアスを持っていることが指摘され (Kanaya et al., ACP 2014)、その値が持つ意味を正確に理解することが難しかった。本研究では、OMI衛星センサ観測に対しDOMINOv2アルゴリズムで導出されたNO₂カラム濃度に関し、以前よりも詳細な検証を行い、またそれを通じて、高度分布の仮定やエアロゾルがもたらす影響を評価した。具体的には、横須賀 (35.32°N, 139.65°E)で2007-2014年に実施された地上MAX-DOAS観測を真値として用い、衛星観測からのアベレージングカーネルを適用してから衛星データと比較することにより、バイアスの大部分が改善することがわかった。このことは、高度分布の仮定がもたらすバイアスが大きいことを意味する。DOMINOv2では、全球スケールの粗い解像度の数値モデルTM4から高度分布形状を借用している。この場合、都市部での導出の際には地表付近の濃度の高まりを十分に考慮できず、エアマスファクター決定の際に重みが過小評価されることによって、低バイアスが起ることがわかった。MAX-DOASから得られた、真値に近い高度分布形状に置き換えると、衛星からの対流圏カラム濃度は約2.2倍にも増大し、MAX-DOASから得られた対流圏カラム濃度とも一致度が向上した。地表付近濃度も同様に増加し、行政モニタリングでの値に近づくことがわかった。以上の解析から、高度分布を適切に考慮することで、衛星データからより信頼度の高いカラム濃度が導出可能で、またあわせて、地表付近濃度も適切に推定しうることがわかった。一方で、エアロゾルによるシールド効果については、cloud fractionが0.03以下のデータを抽出したときに顕著に表れること、その傾向は、エアロゾルの効果を陽に取り入れたアルゴリズムPOMINOによるOMIの解析結果と整合的であることがわかった。

キーワード : remote sensing、vertical profile、aerosol

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 (NO₂), formaldehyde (HCHO), glyoxal (CHOCHO), and water vapor (H₂O) 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 H₂O. 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 NO₂ volume mixing ratios for 4 different viewing directions were as small as ~2 ppbv. Similarly, those differences for HCHO, CHOCHO, and H₂O were estimated to be as small as ~1 ppbv, 60 pptv, and ~0.2%, 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.

キーワード : MAX-DOAS, NO₂, HCHO, CHOCHO, H₂O

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 (NO₂, SO₂, O₃, CHOCHO, HCHO and H₂O). 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 (NO₂, SO₂, O₃, 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 NO₂ 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

GOSAT-TIRによる大気中アンモニア濃度分布の推定

Estimation of atmospheric ammonia distribution from GOSAT-TIR

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大気中アンモニアは食糧生産や廃棄物、バイオマスバーニングなどの様々な発生源から排出されており、その排出量は人間活動の規模の拡大に伴って飛躍的に増加している。アンモニアは比較的活性な物質であるため、他の物質との反応によってPM2.5を生成すること、CCNとなって放射収支に影響を与えること、また、湖沼や海に融解して富栄養化をもたらすことなど、多岐にわたり影響を及ぼすことが知られている。しかし、その大気中での寿命は短く、全球規模での濃度分布やその変化を正確に評価することは難しい。近年、衛星搭載高分解能スペクトルセンサーの打ち上げにより、熱赤外波長域のアンモニアの吸収帯を利用して、全球濃度分布の推定が可能となった。Clarisse et al. (2009)ではIASIのデータを用いて、アンモニアの吸収帯での輝度温度差をアンモニアの気柱量に線形に変換することで、初めてアンモニアの全球濃度の推定に成功している。また、IASI以外でもTES, AIRS, CrISといった高波数分解能スペクトルセンサーによるデータを用いて濃度導出が行われている。ただし、これらのセンサーが搭載されているプラットフォームは回帰日数が16日と比較的長いいため、短時間での変動を捉えることは困難である。また、インベントリを考慮して物質輸送モデルによって推定された時空間変動と観測の結果には差異があることも報告されている(Shephrad et al., 2011)。今回用いるGOSATは回帰周期が3日と上記のものに比べて短いため、より短時間のイベントを捉えられる可能性がある。本発表ではGOSATによるTIRデータを用いた気柱濃度推定方法と解析結果を紹介する。用いた手法は非線形のMaximum a posteriori Method (MAP法; Rogers, 2000)であり、U. S. standardのアンモニア鉛直濃度プロファイルを用いた仮定し、それに係る係数を推定パラメーターとした。アンモニアのリトリーバルに与える影響が大きいと思われる気温プロファイルと水蒸気プロファイルはGOSAT-TIRデータ自体から事前に推定することで誤差の低減を図った。暫定的な解析結果はインド北部や中国東部で高い濃度を示しており、この結果はこれまでの報告と整合的である。発表では全球の解析結果を示す。

キーワード：衛星リモートセンシング、熱赤外放射、大気微量成分

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

2007, 2011, 2016年の南極昭和基地におけるFTIR観測による大気微量成分の変動

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

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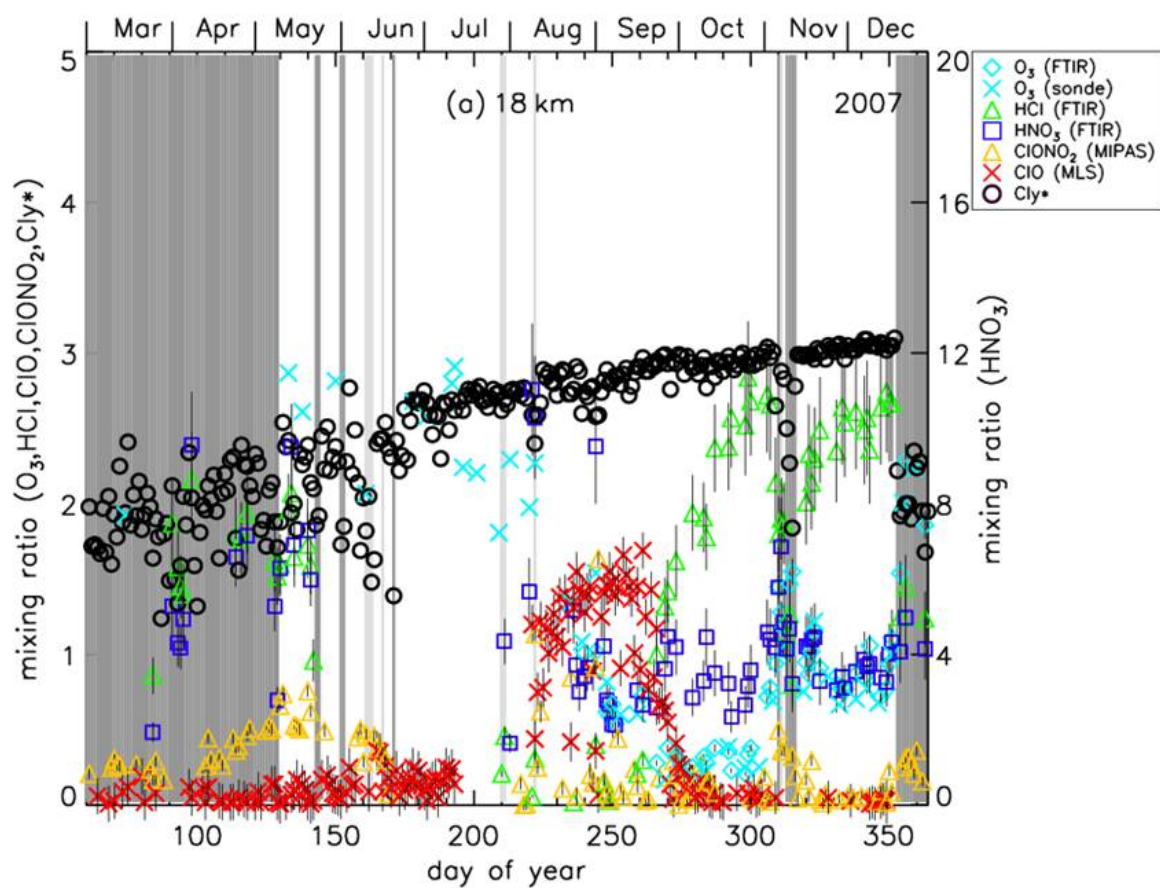
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南極昭和基地において、2007年よりBruker社製IFS-120M型FTIRを観測棟に設置し、太陽赤外線を光源に用いた大気微量成分の観測を行っている。観測は毎年ではなく、FTIR観測専門の隊員が現地で越冬している年に行っている。これまで、2007年（第48次越冬隊）、2011年（第52次越冬隊）、2016年（第57次越冬隊）の3年間の観測を行ってきた。FTIRによる観測は、越冬開始後昭和基地に太陽光が出ている3月～5月、及び8月～12月に行うことが可能である。FTIRによって観測が可能な主な大気微量成分は、 O_3 , HNO_3 , HCl , $ClONO_2$ などである。今回は、これらFTIRで観測された大気微量成分の他に、昭和基地におけるオゾンゾンデ観測、及び人工衛星MLSによる ClO , HCl 、同じく人工衛星MIPASによる $ClONO_2$ の観測結果も併せて解析を行った。解析は高度別に混合比を導出することが可能であり、今回はそのうち18 kmと22 kmに注目して結果を解析した。その結果、昭和基地における冬季にあたる6月初めごろから HCl 濃度が減少を始めることが判った。これは、昭和基地上空におけるPSCの出現によるものである。昭和基地上空に太陽光が戻ってくる8月初めから ClO 濃度が上昇を初め、9月前半にピークになる。そのころまだ HCl 濃度はほとんどゼロのままである。9月中旬から10月にかけて、 ClO 濃度の減少とともに、 $ClONO_2$ 濃度の増加と HCl 濃度の増加がみられる。これらどちらのリザーバーにより多くの Cl が回復するかは、冬によって、また硬度によって異なっていることが判った。また、 O_3 濃度は8月末から減少を始め、10月に最低となりオゾンホールが形成される。これら、南極基地上空において、地上観測によって Cl 濃度のパーティショニングの様子が観測されたのは世界初である。さらに、 Cl パーティショニングの変動の様子が、北極におけるそれとは異なる変動を示すことが確認された。これらは、南極上空成層圏のオゾン濃度が、オゾンホールのため北極上空より低いためであると考えられる。FTIRと衛星による Cl パーティショニングの様子は、3次元化学気候モデルMIROC3.2の結果と比較された。いくつかの化学種では絶対値に系統的な差が見られたが、相対的な変動は化学気候モデルで比較的良く再現されることが判った。講演では、2016年の成層圏水蒸気観測の結果についても報告する予定である。

キーワード：フーリエ変換赤外分光器、昭和基地、塩素化合物、オゾン、オゾンホール

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



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 relying 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 NH₃ molecule [6-8]. Our calculated half-widths of NH₃ 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 ν_1 band of NH₃, 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 ^oP doublets in the ν_4 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 ($\delta^{13}\text{C}$) of diacids dicarboxylic acids ($\text{C}_2\text{-C}_{11}$), oxoacids ($\text{C}_2\text{-C}_9$) and α -dicarbonyls ($\text{C}_2\text{-C}_3$) 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 $\delta^{13}\text{C}$ 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

主要な大気金属エアロゾル種としてのナノ凝集体酸化鉄粒子

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

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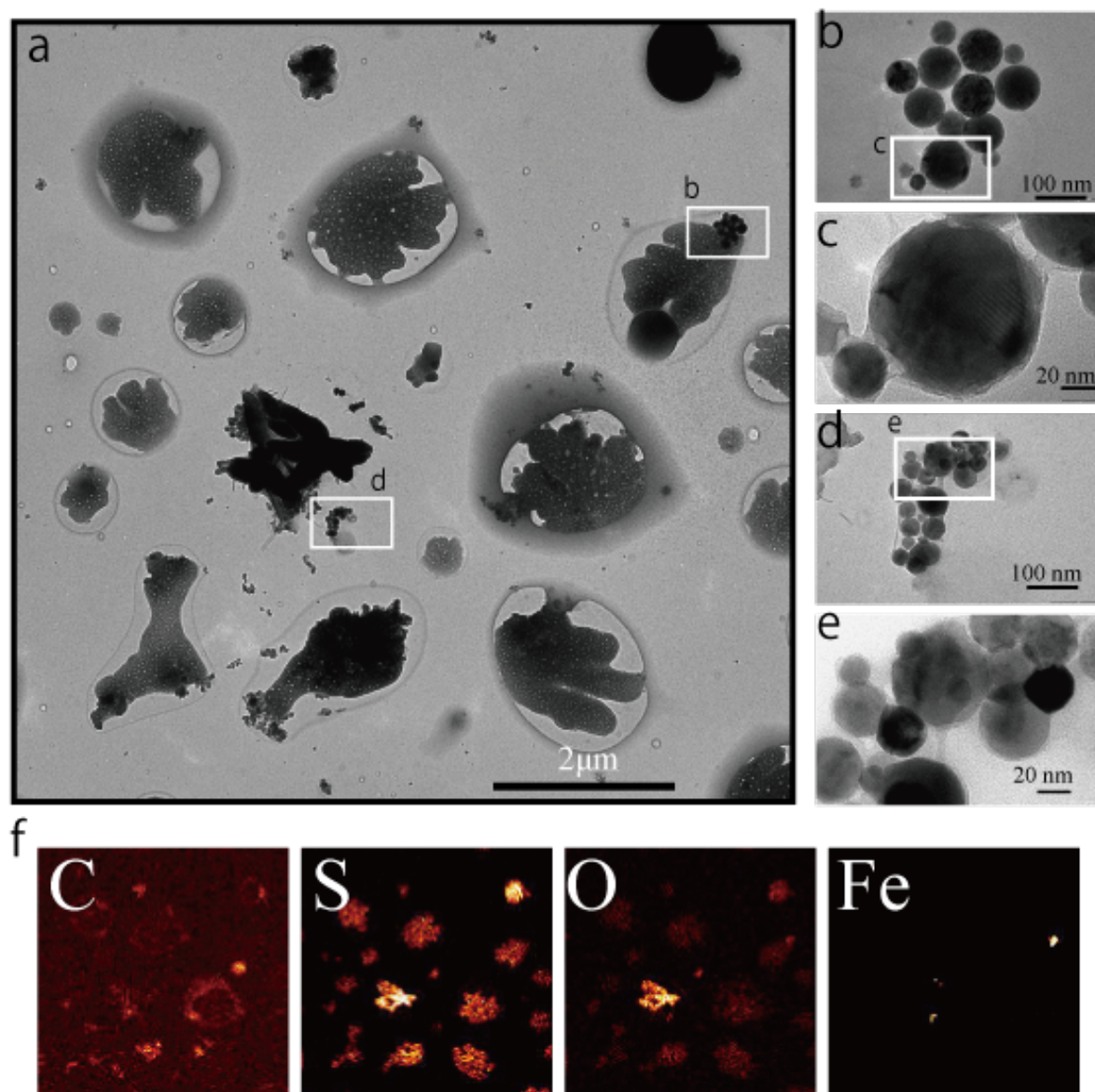
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Iron-oxide particles possibly contribute to climate, if they absorb light; marine environment, if they dissolve to water; and human health, if they have toxicity. However, little is known about the influences of iron-oxide particles in atmosphere to 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.

キーワード：透過型電子顕微鏡、SP2、磁鉄鉱、東京

Keywords: Transmission electron microscope, Single-particle soot photometer, Magnetite, Tokyo



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/m³ ranged from 1.47-25.17 ng/m³ 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 occurred 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.

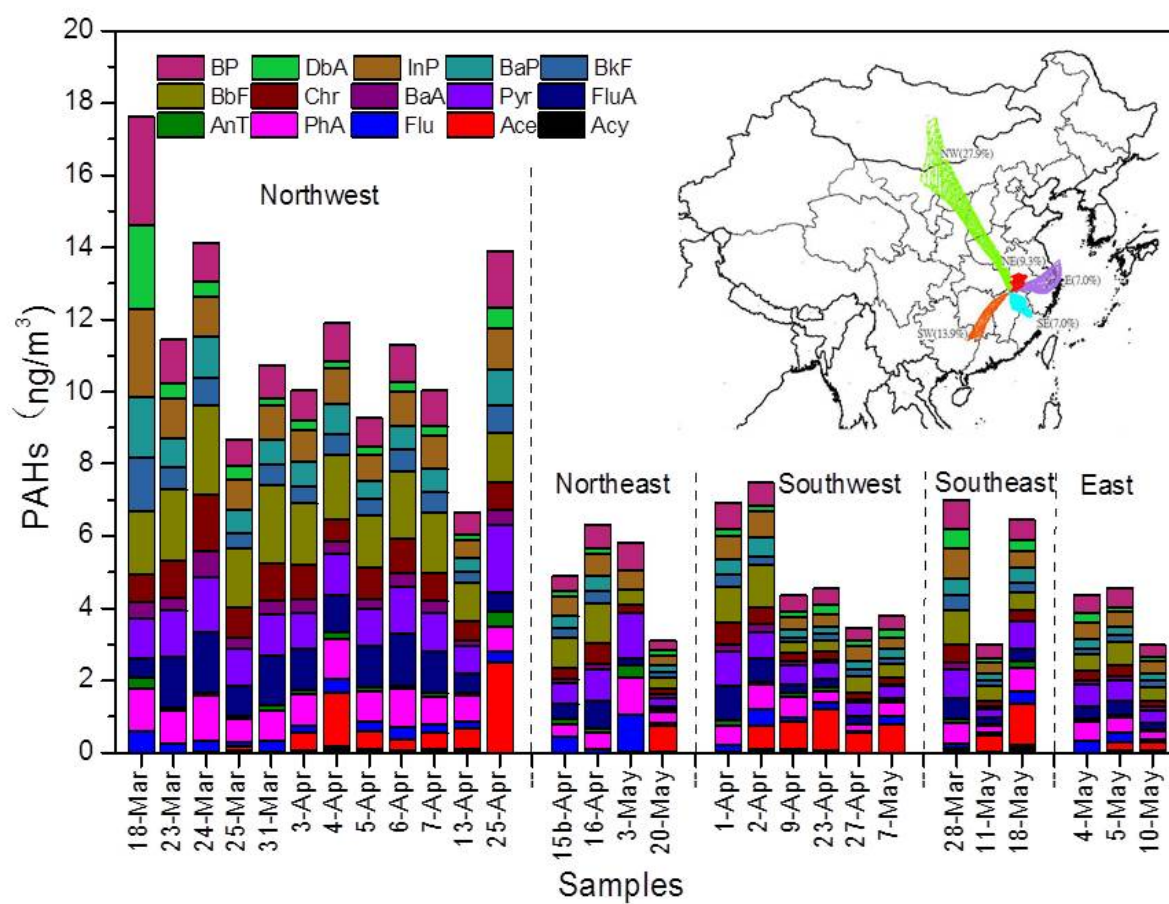
The backward trajectories simulated by HYSPLIT (the Hybrid Single Particle Lagrangian Intergrated Trajectory) model were mainly originated from five directions. The air mass from northwest, northeast, southwest, southeast and east accounted for 27.9%, 9.3%, 13.9%, 7.0% and 7.0%, respectively. Figure 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/m³, 6.65-17.60 ng/m³) under the influence of northwestern air mass. The PAHs levels were similar when air mass from northeast (5.08 ng/m³) and southwest (5.03 ng/m³). When sample were originated from the sea in the east, the PAHs concentrations were at lowest level (3.97 ng/m³). 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 $PM_{2.5}$.

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Keywords: PAHs distributions, Transport, Meteorology, Mount Lushan



2016年春季の韓国で観測されたエアロゾル混合状態の変動 Variability of Mixing State of Aerosols observed at a Surface Site in Korea in the Spring of 2016

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エアロゾルは直接・間接効果により地球の放射収支に影響を与える(IPCC 2013)。これらの効果はエアロゾルの化学組成や混合状態に強く依存する。特に硫酸塩、硝酸塩、有機物がブラックカーボン (BC) とどのような混合状態 (内部混合、外部混合) にあるかがエアロゾルの直接・間接効果を推定する上で重要なパラメーターとなる(Bond and Bergstrom 2006, Adachi et al. 2010)。しかし、実大気中でBCと混合している化学組成をリアルタイムで定量した研究例はほとんどなかった。この研究の目的は、アジア下流域でのBC混合状態別の化学組成を明らかにすることである。

本研究では、韓国において2016年3月1日から4月7日の期間に実大気観測を行った。観測ではレーザー誘起白熱光-質量分析計(LII-MS, Miyakawa et al. 2014)が用いられた。LII-MSはレーザー誘起白熱光分析計 (LII) と質量分析計 (MS) をタンデムに組み合わせた分析手法である。LII部では1024nmのレーザー共振器を用いることにより、BCを内部に含む粒子が効率的に揮発され、白熱光信号が検出される。その後サンプル空気はMS部に連続的に導入され、エアロゾルの化学組成が定量される。LIIレーザーのONとOFFを切り替えることで、BCを内部に含む粒子 (内部混合) と含まない粒子 (外部混合) の化学組成を分けて定量することができる。

観測期間においてPM₁粒子の主要な成分は硝酸塩、硫酸塩、有機物であった。質量濃度の平均では硝酸塩が卓越しており、内部混合の割合では硫酸塩が他の物質に比べて高かった。また、硝酸塩と硫酸塩の外部・内部混合の比較においては実験誤差以上の系統的な差異は見られなかった。発表では化学組成の時間変動とその考察について詳細を述べる。

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キーワード：エアロゾル、化学組成、ブラックカーボン混合状態

Keywords: Aerosol, Chemical composition, BC mixing state

気象研究所地球システムモデルによるブラックカーボンの空間分布と放射効果

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

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大気中の多くのエアロゾル成分が太陽放射を散乱する特性のみを持つのに対し、ブラックカーボンは太陽放射を効率的に吸収し、大気を加熱する。このためブラックカーボンが気候システムに果たす役割は非常に重要であると認識されている。しかしながら、従来の気候モデルによるブラックカーボンの空間分布や放射効果の推定には、未だ大きな不確定性が含まれている。

気象研究所では、気象研究所地球システムモデルMRI-ESM1に数多くの改良を実施することで、第6期結合モデル比較計画CMIP6に向けた新しいバージョンのモデルMRI-ESM2を開発した。この中で、本研究では、従来の気候モデルが含む問題点を克服するために、ブラックカーボンに関する表現については、大きく3つの改良を実施した。第一に、ブラックカーボンが疎水性から親水性へと変換される変質過程（aging）については、従来は一定値の時定数（1.2日）を用いた表現であるのに対し、本研究では物理化学法則に基づき変換速度を表現するパラメタリゼーションを導入することで、大気環境に応じた変換速度を表現できるようにした。第二に、エアロゾルの湿性沈着過程については、従来はエアロゾルの積雲対流による鉛直輸送と降水による除去過程を独立して扱うのに対し、本研究では積雲対流パラメタリゼーションにおいて、エアロゾルが降水除去を経ながら鉛直輸送されるように、鉛直輸送と除去過程を整合的に扱う表現にした。第三に、エアロゾルの放射過程については、従来はブラックカーボンと他エアロゾル成分の内部混合を考慮しないのに対し、本研究では親水性ブラックカーボンについては硫酸塩エアロゾルとの内部混合を仮定することで、被覆による光吸収の増大効果（レンズ効果）を扱うようにした。

MRI-ESM2を用いて、2008-2015年の期間について、計算を実施した。本研究では、水平解像度は約110 km (TL159)、鉛直解像度は80層（上端0.01 hPa）として、現実的な気象場と海面水温を与える再現計算を行った（海洋モデルを結合した計算は実施しない）。

モデル計算結果と地上・航空機観測との比較を行った。北極域の地上においては、従来の変質過程を用いた計算では、観測されたブラックカーボン濃度を過小評価し、季節変化を再現することができなかったのに対し、MRI-ESM2では、ブラックカーボン濃度の季節変化の再現性が向上した。また、従来の湿性沈着過程を用いた計算では、上部・中部対流圏中でブラックカーボン濃度を過大評価したのに対し、MRI-ESM2では、高度分布の再現性が向上した。これらのモデル感度実験を組み合わせた結果から、北極域でのブラックカーボンの季節変化を決める上では、変質過程が重要であるのに対し、上部・中部対流圏中のブラックカーボン濃度を決める上では、積雲対流に伴うエアロゾルの湿性沈着過程が重要な役割を果たすことが示唆された。

大気上端における全球平均のブラックカーボンの直接放射強制力を推定したところ、本研究では、約0.2 W m⁻²と推定された。また、従来の手法を用いたモデル計算結果との比較から、ブラックカーボンの直接放射強制力は、内部混合に伴うレンズ効果により約40%増大し、変質過程の改良により約20%増大することが示唆された。

キーワード：エアロゾル、ブラックカーボン、気候モデル、変質過程、湿性沈着、放射効果

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⁻ + IO₃⁻) 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⁻, Mg²⁺, 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 Mg²⁺, K⁺, Ca²⁺, and halogens (Cl⁻, Br⁻, and I) in frost flowers were associated with sea-salt fractionation by precipitation of mirabilite (Na₂SO₄ 10H₂O), hydrohalite (NaCl 2H₂O), 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 (Mg²⁺/Cl⁻, K⁺/Cl⁻, Ca²⁺/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 SO₄²⁻ 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 MgCl₂ and MgSO₄. 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.

キーワード：海塩粒子、ハロゲン、フロストフラワー

Keywords: Sea-salt aerosols, Halogens, Frost flower

沖縄県辺戸岬における2015年秋季集中観測時の硫酸塩の発生源寄与評価 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.

キーワード：硫酸塩、発生源寄与、大気質モデル、長距離輸送

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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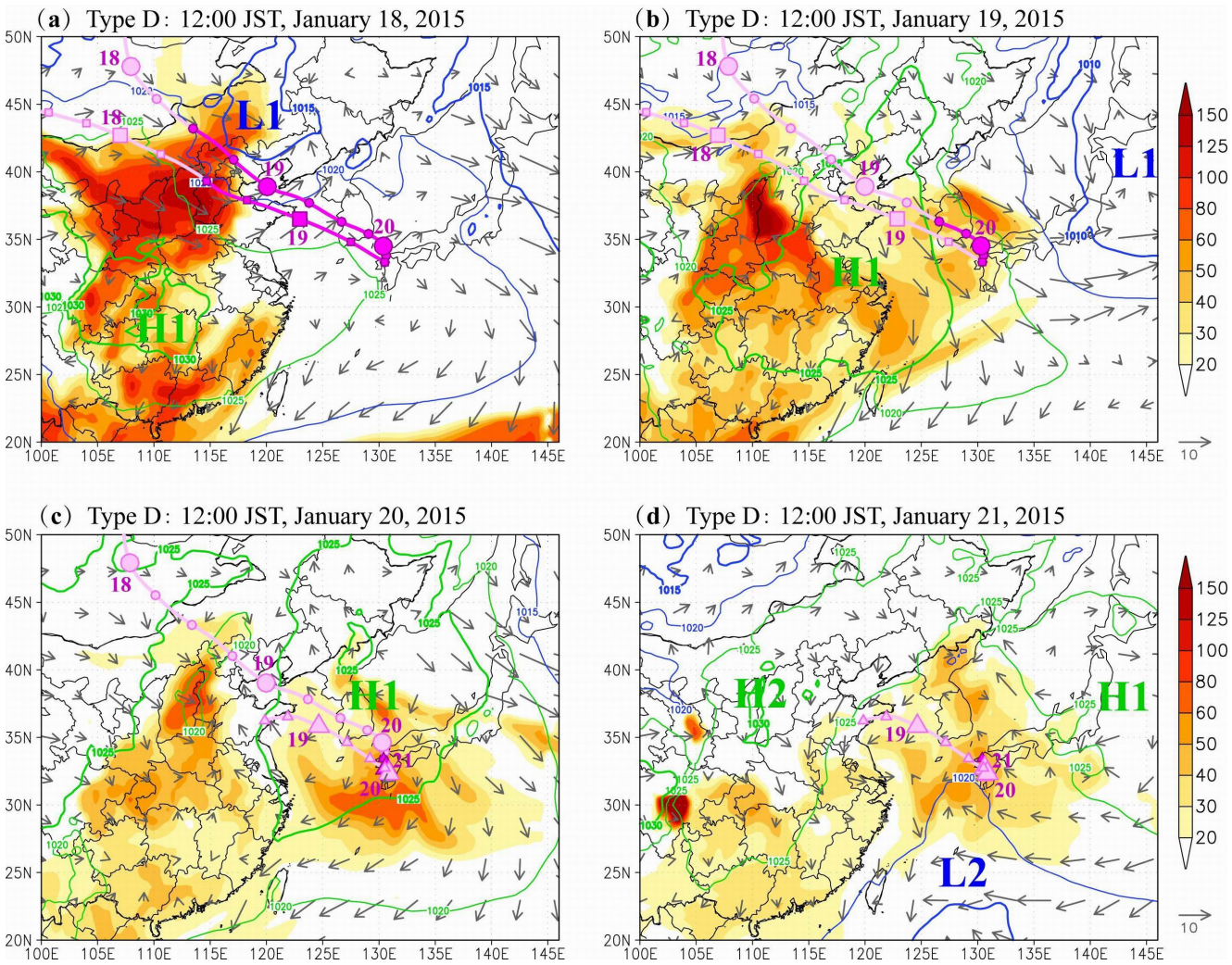
Two regional chemical transport models (NAQPMS and CMAQ) were used to simulate several episodes of high PM_{2.5} concentration observed in January 2015 over China and Japan. Simulation results from both models reasonably explained observed PM_{2.5} levels, as well as the variation observed within three 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 fNO₃⁻ concentrations relative to fSO₄²⁻ (type N), indicating the importance of NO₃⁻ LRT in winter. The second episode showed fSO₄²⁻ concentrations, which were ~3.4-fold greater than fNO₃⁻ (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, SO₄²⁻ 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 cNO₃⁻ concentrations and high cNO₃⁻/fNO₃⁻ 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 fSO₄²⁻ levels, indicating that current models may miss certain emissions of SO₂ and mechanisms promoting the conversion of SO₂ to SO₄²⁻.

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



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: $R^2=0.81$, $RMSE=0.13$, and about 80% AOD retrieval results within the Expected error (EE) of $(0.20 \cdot AOD_{AERONET} \pm 0.05)$. 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.

八方尾根における春季対流圏オゾンの1998年から2016年にかけての長期変動

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

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対流圏オゾンは、地球温暖化、対流圏酸化能および大気質を考える上で、最も重要な微量気体のひとつである。過去数十年の間、東アジアからの大気汚染物質の排出は急激に増加しており、特に中国はその中でも最大の排出国であると考えられている。最近の衛星による対流圏二酸化窒素カラム濃度データの解析に基づく最近の研究では、中国からの窒素酸化物の排出量が高止まりしている可能性が示唆されている。また、前駆物質の排出量だけでなく、気候の変動も対流圏オゾンの濃度変動を決める重要な要素の一つであり、数年から十年規模の気候振動が大気輸送に影響を与え、汚染源からの流入を変化させるという結果も報告されている。本発表では、1998年から2016年の連続観測データから、長野県白馬村の八方尾根での春季対流圏オゾンの長期変動について述べる。

八方尾根における春季オゾン濃度は、1998年から2007年にかけて急激に増加し続けた。これは経済成長に伴う中国東部からのオゾン前駆体排出の増加によるものと考えられる。その後、八方尾根におけるオゾン濃度は横ばいの傾向が続いており、2008年と2012年には二度大きく減少していた。排出量及び気象データの解析から、この二度の大きなオゾン濃度減少は、それぞれ異なる要因によって生じたと考えられた。一つ目の要因は、気象場の変動による中国中東部からの寄与の減少である。日本の東の太平洋上の気圧偏差が減少し、チベット高原周辺の気圧偏差が増加した時に、西向きの風が強まることで、中国中東部からの汚染気塊が八方尾根へ輸送されやすくなり、観測されるオゾン濃度も高くなっていたことが推察された。もう一つの要因は、中国東部からのオゾン前駆体排出量の減少であることが示唆された。

キーワード：オゾン

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 $6 \mu\text{g m}^{-3}$ from 2009 ($35.3 \mu\text{g m}^{-3}$) to 2012 ($41.5 \mu\text{g m}^{-3}$) was recorded (Gupta and Mohan, 2015) in Delhi and in summer 2015, exceedance of O₃ levels ranged from 92% - 97%. Tropospheric ozone is produced by a cycle of reactions involving two basic pollutants NO_x 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 NO_x 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 NO_x in ozone formation model simulated VOC to model simulated NO_x ratios were studied through scatter plots. Ozone production is considered to be VOC limited at low VOC to NO_x ratio i.e. less than about 4 to 1 and for high ratio greater than about 15 to 1 the region is classified as NO_x 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 NO_x ratio, WRF/Chem, Megacity Delhi

国内5箇所の地上観測サイトにおける大気中アルゴン濃度の季節および年々変動

Seasonal and interannual variations in the atmospheric Ar/N₂ ratio observed at five ground based stations in Japan

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Atmospheric Ar/N₂ ratio is a unique tracer to detect the spatiotemporally-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 beginning 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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キーワード：大気中Ar/N₂比、海洋貯熱量、季節変動、年々変動

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

酸素・二酸化炭素濃度比による大気CO₂濃度変動成分の起源推定法の開発 Estimation of CO₂ contributions from fossil fuel consumption based on the atmospheric O₂ and CO₂ continuous measurements

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Atmospheric carbon dioxide (CO₂) 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 CO₂ emissions from biotic activities and burning of fossil fuels. Partitioning the contribution rate between the individual CO₂ sources would be useful to reduce the uncertainty of the CO₂ emissions estimated from atmospheric inversion calculations. The ratio of oxygen (O₂) consumption to CO₂ 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 CO₂ and O₂ concentration at Tsukuba in February 2015, and evaluated the individual contributions of the biotic and fossil fuel-derived emissions to the short-term CO₂ 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 CO₂ 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 ¹⁴CO₂, 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 CO₂ estimated from $-O_2/CO_2$ ratios and ¹⁴CO₂ 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 ¹⁶O/¹⁸O isotopes, (2) a OH-radical scavenger, (3) variable O₃ 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.

キーワード：エアロゾル、テルペン、VOC、オゾン、表面、界面

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 their 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

高分解能光学顕微鏡によって可視化された氷の表面融解現象 Surface melting on ice surfaces visualized by advanced optical microscopy

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氷は地球上に存在する結晶の中で最も多いものの1つであるため、その相転移現象は自然界に大きな影響を及ぼす。相転移現象の1つである表面融解が起こると、氷融点の0℃以下であっても氷表面は疑似液体層と呼ばれる薄い液体で濡れている状態となる。疑似液体層の有無は氷表面での化学反応に影響を及ぼすため、大気化学分野にとっても重要である。

近年、我々はオリンパス（株）と共同でレーザー共焦点微分干渉顕微鏡を開発し、高さ0.37 nmの氷の単位ステップ [1]や疑似液体層 [2]を可視化することに成功した。疑似液体層の直接観察によると、従来の薄膜液層が氷全体を覆う描像ではなく、2種類の形態を持った疑似液体層が部分的に氷表面を覆う様子であることがわかった [2,3]。また、疑似液体層の出現条件は温度だけではなく、周辺大気の水蒸気量 [4,5]とガス成分 [6]に依存することもわかった。

例えば塩化水素ガスの存在は疑似液体層の出現を非常に促進させるため、塩化水素ガス無しでは疑似液体層が出現しない-15 ~ -1.5℃の温度条件でも疑似液体層を観察することができた（ただし-15℃以下の実験は未だ行ってないため下限は不明） [6]。また、この液滴は蒸発によって消えることはないことと液滴挙動の水蒸気量依存性から、塩化水素ガスが液滴に溶解し塩酸液滴となっていることが予測された。

加えて、過飽和条件ではこの塩酸液滴は氷の中に埋没してしまい、氷を蒸発させると同場所から再度液滴が出現することがわかった。これらの結果は、氷結晶がガス成分を流体包有物として多量に保持できる可能性を示唆している。

[1] Sazaki et al. (2010) *PNAS* **107**, 19702.

[2] Sazaki et al. (2012) *PNAS* **109**, 1052.

[3] Asakawa et al. (2015) *Cryst. Growth Des.* **15**, 3339.

[4] Asakawa et al. (2015) *PNAS* **113**, 1749.

[5] Murata et al. (2016) *PNAS* **113**, E6741.

[6] Nagashima et al. (2016) *Cryst. Growth Des.* **16**, 2225.

キーワード：氷、表面融解、疑似液体層、塩化水素ガス、高分解能光学顕微法

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

鉱物エアロゾルの氷晶核能の評価：鉱物種による違いと変質過程に着目して

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

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過冷却水からなる液相の雲内では、氷晶核として働く大気エアロゾルの働きにより氷晶が形成される。特に、大気中における存在量などから、鉱物ダストが氷晶形成において最も重要だと考えられている。しかし、鉱物ダストが氷晶核として有効に機能する機構に関してはまだ十分に理解されていない。そこで本研究では異なる鉱物種の氷晶核能を評価し、鉱物学的な特性との関連性を明らかにすることを目的とした。本研究ではコールドフロート法を用いて、特に氷晶核能が高い長石類を中心に鉱物ダストの氷晶核能を測定した。その結果、長石が高い氷晶核能を示す原因として、秩序性、固溶体形成の有無、陽イオン種の3つの要因が関与している可能性が示唆された。加えて、長石に硫酸を付加すると氷晶核能が低下し、粘土鉱物のそれに近づくことが分かった。以上の結果は、実際の大気中における鉱物ダストの氷晶核としての挙動を理解する上で重要な示唆を与えるものである。