# Data composite of airborne sulfur dioxide measurements in the upper troposphere

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

Keywords: sulfur dioxide, aircraft measurements, new particle formation

 

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

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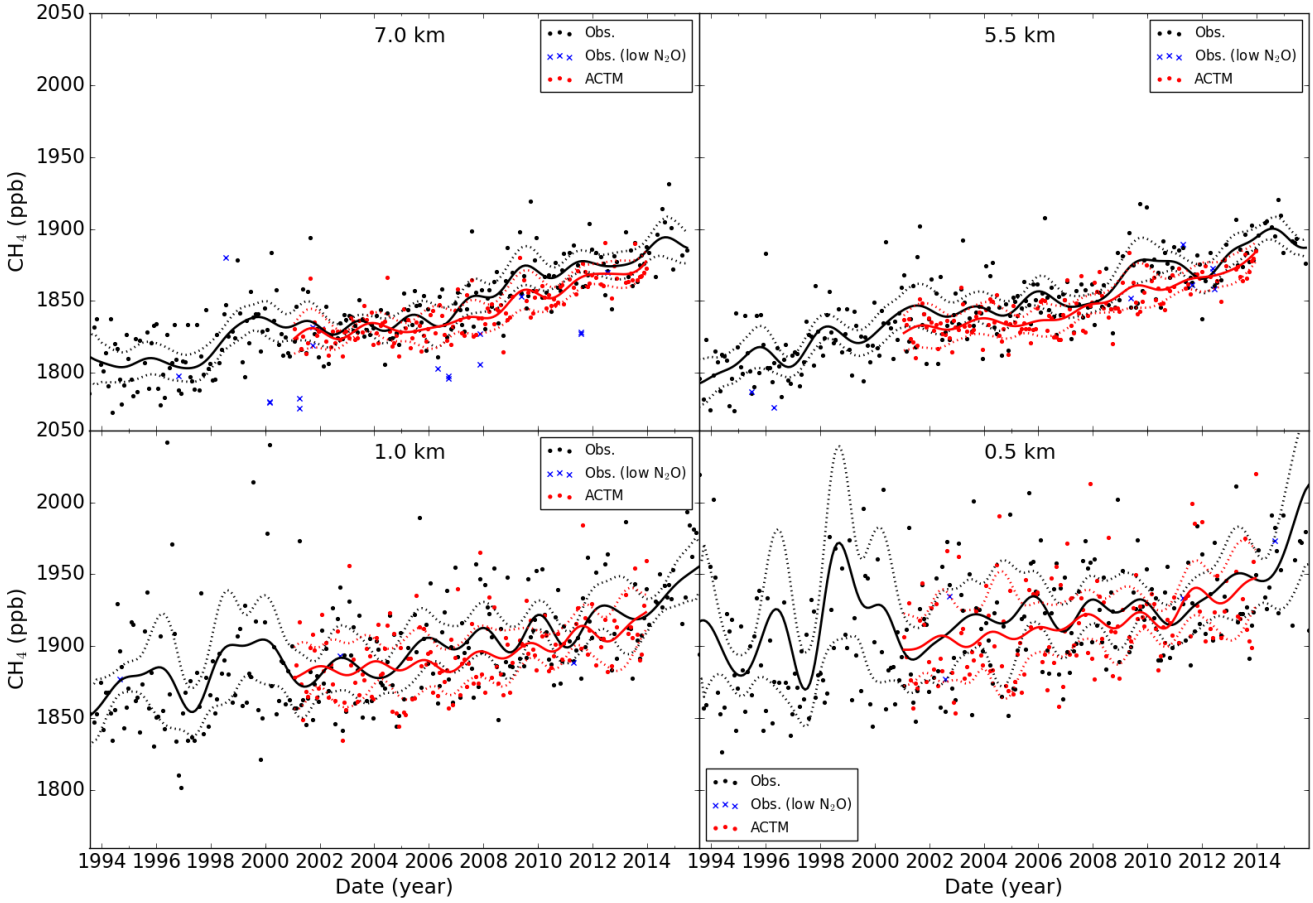
We have carried out monthly flask sampling with aircraft over the boreal wetlands in Surgut and a pine forest nearby Novosibirsk, both located in West Siberian Lowland (WSL). Vertical profiles of methane ($CH_4$) concentration were measured up to about 7 km since 1993 at Surgut and 1997 at Novosibirsk.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

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 $CO_2$ over the Asia-Pacific region observed by the CONTRAIL commercial airliner measurements

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

Keywords: Carbon dioxide, Aircraft observation, Asian monsoon

 

# 北半球大気中$N_2O$アイソトポキュ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 $^{15}N/^{14}N$ and $^{18}O/^{16}O$ ratios but also site-specific $^{15}N/^{14}N$ ratio in asymmetric NNO molecule, are regarded as useful parameters to infer the origin and production–consumption mechanisms of $N_2O$, and to estimate its global budget. Previous studies on $N_2O$ trapped in the firn in polar ice sheet revealed the secular trend of isotopocule ratios, but there have been only a few reports on long-term monitoring of atmospheric $N_2O$ isotopocule ratios in the Northern Hemisphere.

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

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

 

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

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

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$NO_2$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 $NO_2$ 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(NOx)の主要な成分である$NO_2$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$NO_2$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$NO_2$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、低バイアスが起こることがわかった。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 ($NO_2$), formaldehyde (HCHO), glyoxal (CHOCHO), and water vapor ($H_2O$) on a spatial scale of about 10 km. We found that daily 4AZ-MAXDOAS data for a 0-1 km layer show the significant spatial inhomogeneity, even for $H_2O$. However, the observed spatial inhomogeneity as well as the temporal inhomogeneity can be lowered by taking averages over longer time. We found that the differences among monthly-mean $NO_2$ volume mixing ratios for 4 different viewing directions were as small as ~2 ppbv. Similarly, those differences for HCHO, CHOCHO, and $H_2O$ were estimated to be as small as ~1 ppbv, 60 pptv, and ~0.2%v, respectively. It is suggested that for measurements made at a single site, averaging data over time can lower effects of both spatial and temporal inhomogeneities on the estimate of the representative values for a grid adopted by a model and for a pixel measured by a satellite instrument.

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 ($NO_2$, $SO_2$, $O_3$, CHOCHO, HCHO and $H_2O$). The MAX-DOAS data of AEC and its vertically-integrated quantity, i.e., the Aerosol Optical Depth (AOD), were compared to those of the co-located AD-Net (Asian Dust and aerosol lidar observation network) LIDAR (Light Detection and Ranging) and SKYNET sky radiometer measurements, respectively. Aerosol measurements from all the three platforms showed similar seasonal variations with enhanced aerosol loading during the dry season (October-May). This enhancement was associated with biomass burning, which is a pronounced event in this region according to literature reports. This was further supported by satellite observations (MODIS), which show high correlations with MAX-DOAS data with an $R^2$ of 0.71. The seasonal variation of trace gases ($NO_2$, $SO_2$, $O_3$, CHOCHO, and HCHO) showed similar patterns with enhanced concentrations during the dry season, consistent with the influence of biomass burning. In the dry season, the CHOCHO/HCHO ratio was estimated to be 0.025±0.017, which is in the range of the literature values reported for rural sites. The Ozone Monitoring Instrument (OMI) tropospheric $NO_2$ data also showed similar seasonal variations but tended to be biased high by 27%. It is thus expected that such unique results from multi-component observations with MAX-DOAS will be useful for evaluating satellite data and models for the Southeast Asia region.

Keywords: MAX-DOAS, Aerosols, AOD

 

# 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を仮定し、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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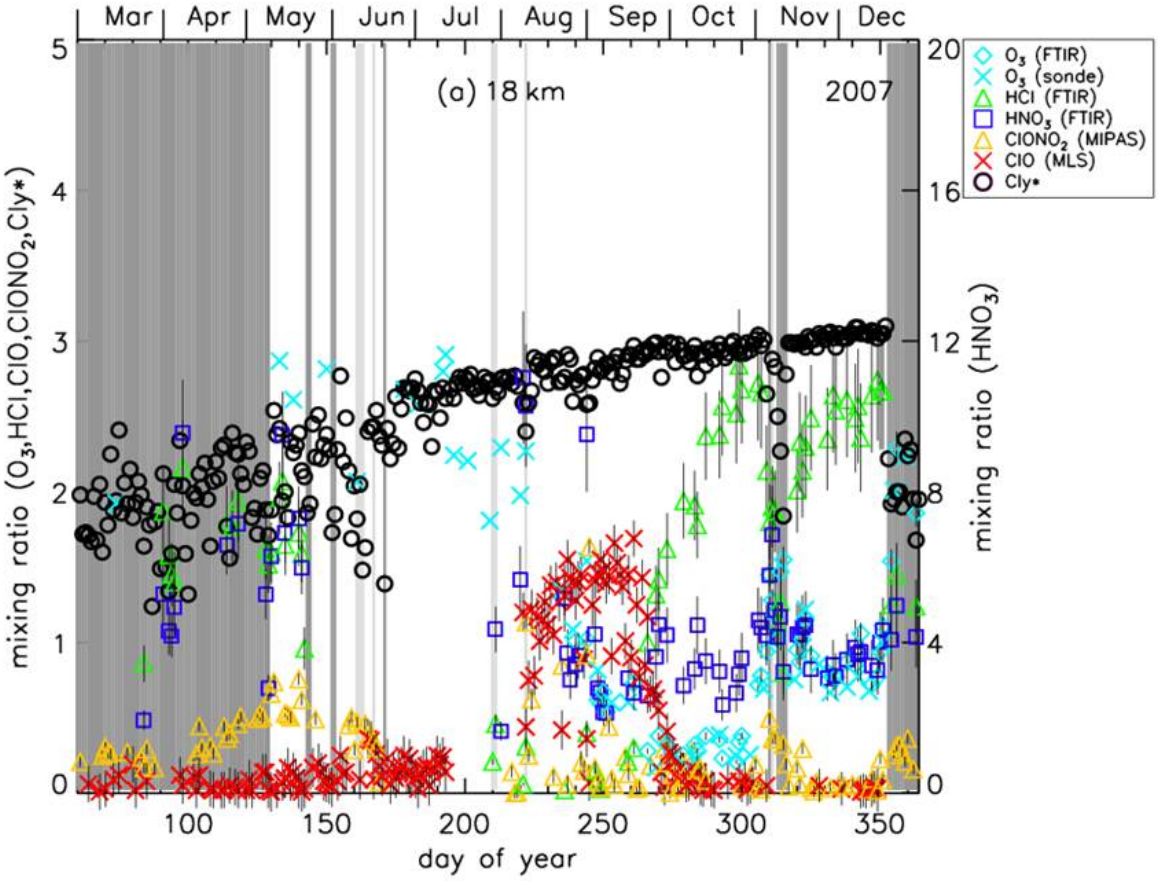
南極昭和基地において、2007年よりBruker社製IFS-120M型FTIRを観測棟に設置し、太陽赤外線を光源に用いた大気微量成分の観測を行っている。観測は毎年ではなく、FTIR観測専門の隊員が現地で越冬している年にのみ行っている。これまで、2007年（第48次越冬隊）、2011年（第52次越冬隊）、2016年（第57次越冬隊）の3年間の観測を行ってきた。FTIRによる観測は、越冬開始後昭和基地に太陽光が出ている３月～5月、及び8月～12月に行うことが可能である。FTIRによって観測が可能な主な大気微量成分は、$O_3$, $HNO_3$, HCl, $ClONO_2$などである。今回は、これら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Cly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Cly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、Cly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Cly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, Cly, 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 relaying on the isolated line approximation, the semi-classical Robert-Bonamy formalism, which has been widely used to calculate half-widths and shifts for decades, fails in calculating the off-diagonal matrix elements. As a result, in order to simulate atmospheric spectra where effects from line mixing are important, semi-empirical fitting or scaling laws such as the energy corrected sudden (ECS) and infinite order sudden (IOS) models are commonly used. Recently, we have found that in developing semi-classical line shape theories, to rely on the isolated line approximation is not necessary [1]. By eliminating this assumption, we have developed a more capable formalism that enables one not only to reduce uncertainties of calculated half-widths and shifts, but also to calculate the whole relaxation matrix. Thanks to this progress, one can address the line mixing based on interaction potentials between two interacting molecules. We have applied this formalism for Raman and infrared spectra of linear molecules [1-4], asymmetric-top molecule [5], and symmetric tops with inversion symmetry such as the $NH_3$ molecule [6-8]. Our calculated half-widths of $NH_3$ lines in the parallel and perpendicular bands match measurements very well. Furthermore, the method has been applied to the calculation of the shape of the Q branch and of some R manifolds in the nu1 band of $NH_3$, for which an obvious signature of line mixing effects has been experimentally demonstrated. Similarly, the formalism very well reproduces off diagonal elements measured in some of $^pP$ doublets in the nu4 band. In summary, comparisons with measurements show that predictions from the new formalism accurately match the experimental line shapes.

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

 

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

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

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

 

# 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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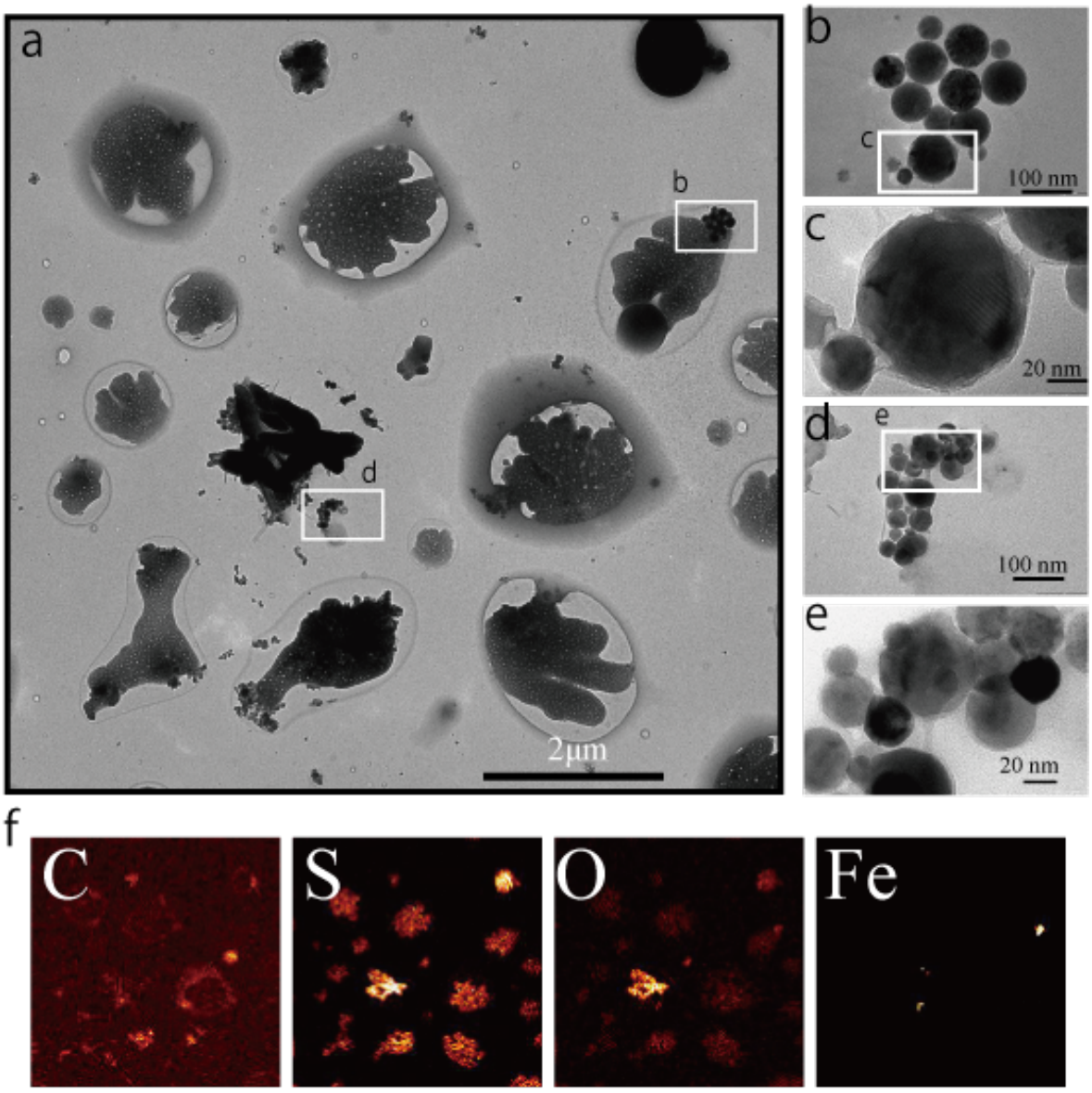
Iron-oxide particles possibly contribute to climate, if they absorb light; marine environment, if they dissolve to water; and human health, if they have toxicity. However, little is known about the influences of iron-oxide particles in atmosphere to these important issues. This study examines iron-oxide 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/$m^3$ ranged from 1.47-25.17 ng/$m^3$ during the sampling time. The PAHs level at Mount Lushan was at a medium level comparing to other sites around the world. PAHs mass in $PM_{2.5}$ were 160.24 ug/g ranging from 63.86 to 427.97 ug/g. The predominant compounds were BbF (benzo[b]fluoranthene), Pyr (pyrene) and BP (benzo[g,h,i]perylene). In terms of aromatic-ring PAHs distributions, 6-ring and 4-ring were predominant, accounting for 27.9% and 24.2% of the total. It is due to that high molecular weight (HMW) PAHs have lower volatility and easier to attach in the particles comparing to low molecular weight (LMW) PAHs.

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

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

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

 

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

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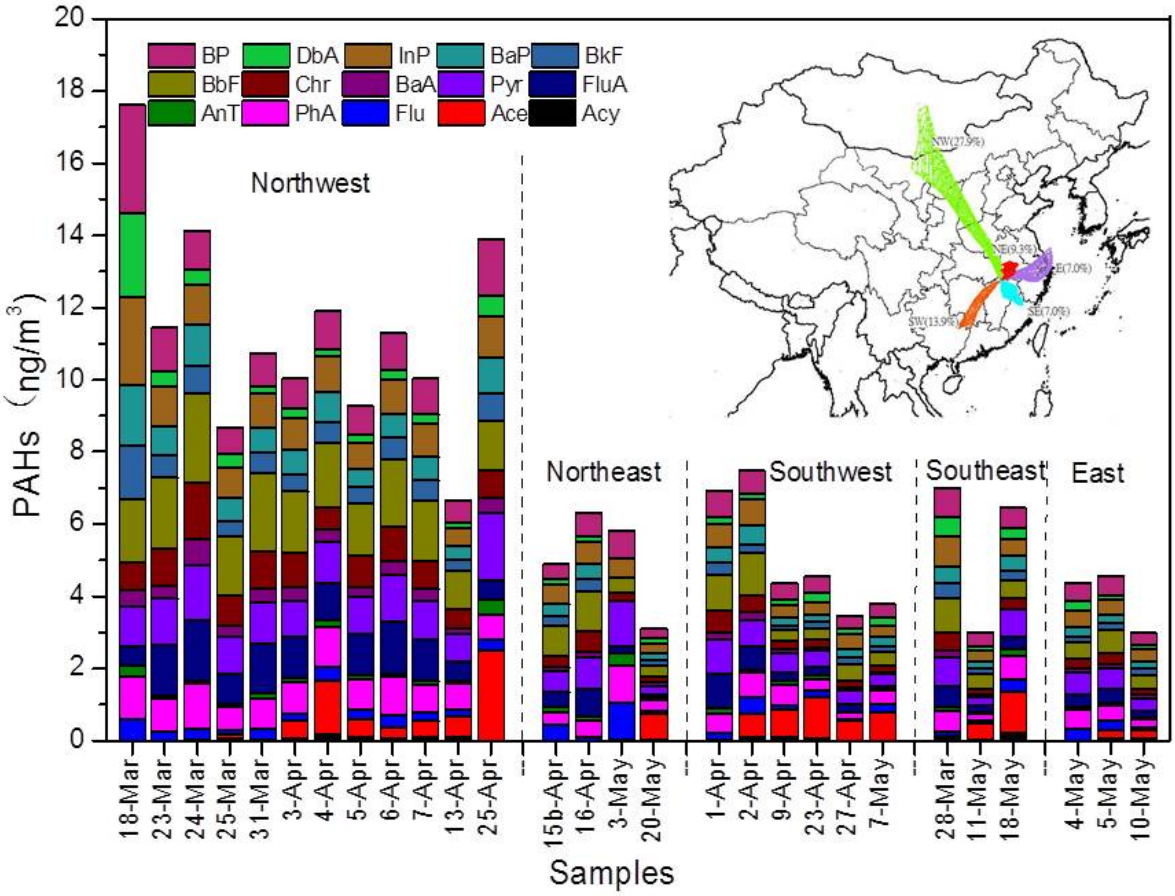
Keywords: PAHs distributions, Transport, Meteorology, Mount Lushan



# 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$PM_1$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 ($I^-$ + $IO_3^-$) were analyzed with IC-MS and ICP-MS, respectively. Individual aerosol particles were observed and analyzed using SEM-EDX.
Results show that water-soluble frost flower and brine constituents were sea salt constituents (e.g., $Na^+$, $Cl^-$, $Mg^{2+}$, and $Br^-$). Concentration factors of sea-salt constituents of frost flowers and brine relative to seawater were 1.14–3.67. Sea-salt enrichments of $Mg^{2+}$, $K^+$, $Ca^{2+}$, and halogens ($Cl^-$, $Br^-$, and I) in frost flowers were associated with sea-salt fractionation by precipitation of mirabilite ($Na_2SO_4$ $10H_2O$), hydrohalite (NaCl $2H_2O$), and sylvite (KCl). Comparison between sea-salt ratios in brine and frost flower implied that precipitation of mirabilite and hydrohalite proceed in slush layer and then the residual brine were migrated vertically onto frost flowers in our research conditions. Molar ratios of sea-salts ($Mg^{2+}/Cl^-$, $K^+/Cl^-$, $Ca^{2+}/Cl^-$, and $Br^-/Cl^-$) changed gradually with aging and growth of frost flowers and sea-salt fractionation under colder conditions. Furthermore, sea-salt fractionation was associated with not only surface air temperature but also sea-ice thickness which related to heat conduction from seawater. In contrast to Br- enrichment in frost flower with the aging, changes of $I/Cl^-$ ratio in frost flowers, however, were not clear. Iodine release from frost flowers might be more likely to proceed relative to $Br^-$ release. Aerosol number concentrations, particularly in coarse mode, were increased considerably by release from the sea-ice surface under strong wind conditions. Sulphate depletion by sea-salt fractionation was found to be slight in sea-salt aerosols because of heterogeneous $SO_4^{2-}$ formation on sea-salt particles. Mg was enriched in coarse and fine sea-salt particles collected on sea-ice area. Mg in sea-salt particles was in the form of $MgCl_2$ and $MgSO_4$. Strong Mg enrichment might be more likely to proceed in fine sea-salt particles. Mg enrichment in sea-salt particles was enhanced under colder conditions. In addition, ikaite-like and mirabilite-like particles identified in the atmosphere only near new sea ice are close to the sea-ice margin. Thus, Ikaite-like and mirabilite particles might be released from initial sea-ice before freezing over, and Mg-rich sea-salt particles might be released from sea-ice surface with frost flowers.

 

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.

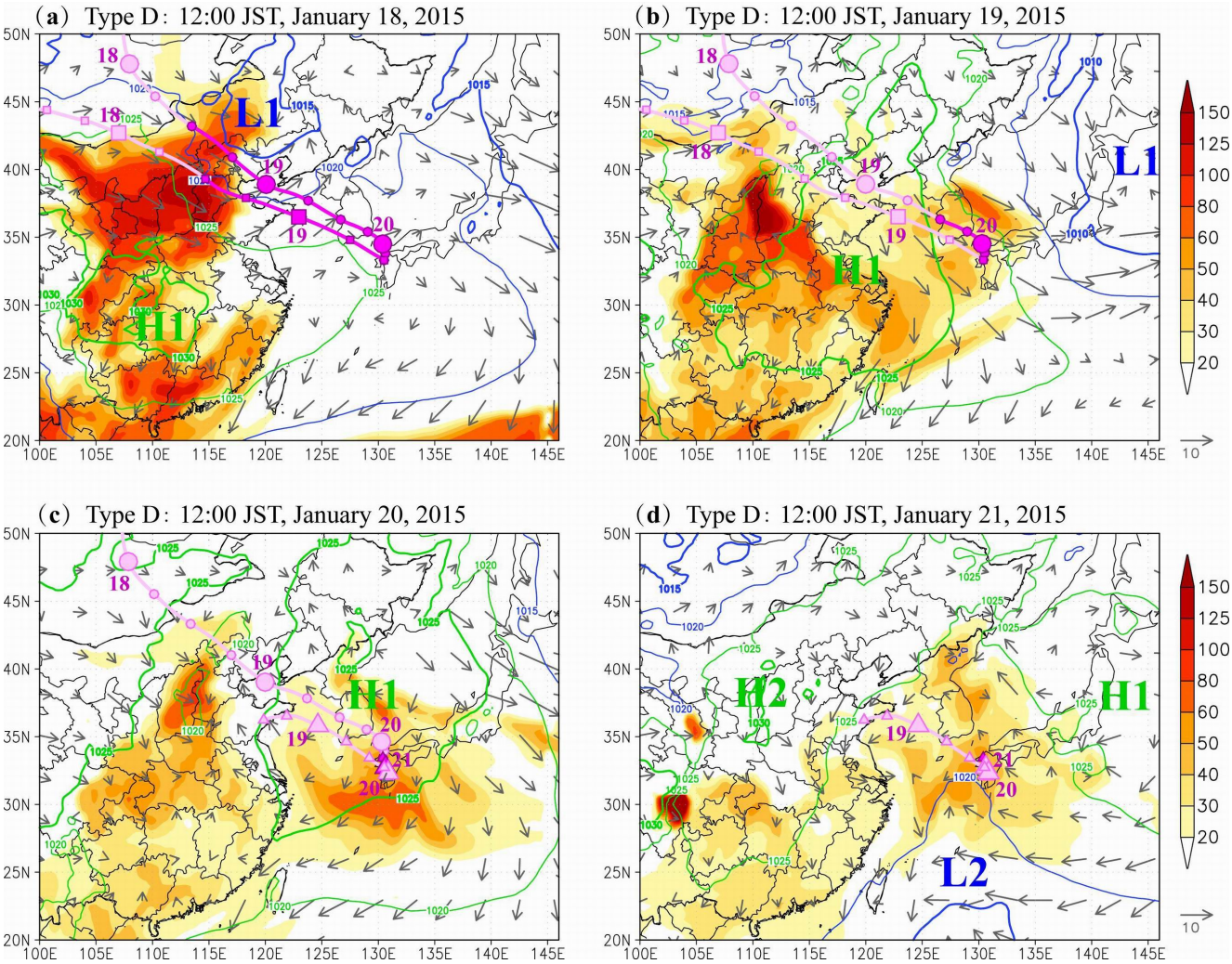
Based on the model results and the synergetic aerosol observations in Fukuoka, Japan, three types of LRT of air pollutants were observed. The first episode showed increased $fNO_3^-$ concentrations relative to $fSO_4^{2-}$ (type N), indicating the importance of $NO_3^-$ LRT in winter. The second episode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

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

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

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



# Joint retrieval of aerosol optical depth and surface reflectance over land using geostationary satellite data

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The Advanced Himawari Imager (AHI) aboard Himawari-8 geostationary satellite provides high-frequent observations with big coverage, multiple spectral channels, and high spatial and spectral resolution. With these characteristics, the AHI data have significant advantages to monitor the air quality and estimate the aerosol properties. In this study, AHI data were used to develop an algorithm for the joint retrieval of aerosol optical depth (AOD) over land and land surface bidirectional reflectance. Instead of constructing a Look-Up-Table (LUT) and pre-estimating the surface reflectance to retrieve the AOD, the atmosphere properties and surface bidirectional reflectance were retrieved simultaneously using an optimal estimate method. The algorithm uses an Earth-atmosphere model, which couples the atmospheric radiative transfer model and surface bidirectional reflectance factor (BRF) model. Utilizing the character that the surface reflectance properties are much more stable than atmosphere aerosol, our retrieval is based on the two basic assumptions: the surface bidirectional reflective properties is invariant during a short time window while aerosol properties (e.g. AOD, AE) change. Optimal estimate method is employed to calculate the AOD and surface BRF. Detailed analysis and validation about the retrieval results were conducted using ground-based measurements (AErosol RObotic NETwork (AERONET) sites) and satellite product (MODIS C6 aerosol product). The validation of the AOD with AERONET measurements shows a high correlation coefficient: R2=0.81, RMSE=0.13, and about 80% AOD retrieval results within the Expected error (EE) of (0.20*$AOD_{AERONET}$ ±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 $\mu g\ m^{-3}$ from 2009 (35.3 $\mu g\ m^{-3}$) to 2012 (41.5 $\mu g\ m^{-3}$) was recorded (Gupta and Mohan, 2015) in Delhi and in summer 2015, exceedance of $O_3$ levels ranged from 92% - 97%. Tropospheric ozone is produced by a cycle of reactions involving two basic pollutants NOx and VOCs. Chemical regional transport models such as WRF/Chem are used extensively for modeling of ozone concentration. As ozone production has distinctive daytime and nighttime chemistry, Gupta and Mohan (2015) recommended that for Delhi model implementation shall be made for policy decisions cautiously with due consideration to the magnitudes of ozone levels. In order to implement control strategies it is conducive to understand whether the study area is NOx or VOC limited. This study focuses on the WRF/Chem model to simulate ozone concentration for megacity Delhi during summer conditions for three consecutive years. To understand the role of VOC and NOx in ozone formation model simulated VOC to model simulated NOx ratios were studied through scatter plots. Ozone production is considered to be VOC limited at low VOC to NOx ratio i.e. less than about 4 to 1 and for high ratio greater than about 15 to 1 the region is classified as NOx limited. There are various sources that contribute to VOC emissions such as vehicular pollution, refueling stations, industrial hubs, diesel generator sets etc. In Delhi overall about 26 to 54% of VOCs are due to the emissions from diesel internal combustion engine (Srivastava et al., 2005). Srivastava et al. (2005) reported source contribution to total VOC for a traffic junction in Delhi as 43% from diesel exhaust, 24% from gasoline, 19% from evaporative and 14% from other sources. As the ozone production is VOC limited for megacity Delhi, WRF/Chem model can be further used to apply control strategies. The response of reduction in VOC emissions from transport sector on ozone production can be modeled in order to formulate effective mitigation measures involving both VOC and ozone reduction.

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

 

# 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/$N_2$ ratio observed at five ground based stations in Japan

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

Acknowledgements
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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$CO_2$濃度変動成分の起源推定法の開発
# Estimation of $CO_2$ contributions from fossil fuel consumption based on the atmospheric $O_2$ and $CO_2$ continuous measurements

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

 

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

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

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 ther mechanisms of formation. We will discuss the implications of our findings on the adverse health effects induced in healthy and diseased subjects.

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

 

# 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け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。

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

 

# イソプレンと一酸化窒素の混合気体試料に対するオゾン反応性全量計の応答特性

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

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生物起源揮発性有機化合物 BVOCs は、対流圏オゾン $O_3$ や二次有機エアロゾル SOA の前駆体として注目されている。多くの BVOCs は分子内に C=C 二重結合を持つため、$O_3$ に対する反応性を有する。発表者はこれまでに、BVOCs 包括把握のためのオゾン反応性全量 $R_{O3}$（$k_i$[$VOC_i$]の総和）の測定装置を構築してきた [1-4]。BVOCs 試料の $R_{O3}$ は、BVOCs のオゾン反応に伴うオゾン減少量を高精度に測定することで決定できる。これまでの研究の結果、$R_{O3}$ 全量計の検出下限 $6.3 \times 10^{-5}$ $s^{-1}$ を実現した ($S/N$=3, 60-s 平均値, 反応時間 50 s) [4]。本測定装置を実大気などの多成分混合試料の測定に用いるには、装置特性を詳細に検証する必要がある。たとえば、一酸化窒素 NO はさまざまな大気試料中に有意に含まれるうえ、オゾン反応が速いため、オゾン反応性測定に対する NO の寄与は無視できない。Mogensen ら [5] は、森林大気に関するモデル計算研究において、オゾン反応性に対する NO の寄与の重大性を示している。一般には、大気試料中の NO 濃度は化学発光法 NO 計により容易に測定可能である。したがって、NO 濃度に対する $R_{O3}$ 全量計の応答特性を十分に把握しておくことで、NO 濃度の測定結果を用いて $R_{O3}$ 測定結果に占める NO+$O_3$ 反応の寄与を決定し、$R_{O3}$ に占める BVOCs+$O_3$ の寄与を正しく定量できるようになる。本研究では、イソプレン標準試料と NO 標準試料の混合ガス試料を用意して、これに対する $R_{O3}$ 計の応答特性を調べる実験を試みて、BVOCs 試料に対する NO の標準添加法による $R_{O3}$ への NO 寄与分の決定の可能性について検証した。実験の結果、イソプレン標準試料 (A) および NO を添加した試料 (B, C, D) について NO 濃度に対するオゾン反応性 $R_{O3}$ の実測値をプロットしたところ、Fig.1 を得た。図に示すように、NO 添加イソプレン試料(B, C, D) に関する回帰直線の $y$ 切片は、NO 添加前のイソプレン標準試料 (A) と完全に一致した。NO 標準添加法による $R_{O3}$ 測定値に占める NO と BVOCs の寄与の切り分けが十分に妥当であることを、実験的に確認した。すなわち、BVOCs 試料の $R_{O3}$ を測定するとき、実大気など共存 NO が無視できない場合には、NO 濃度も測りつつ、濃度既知 NO 試料を混合する標準添加 $R_{O3}$ 測定も実施すればよいことがわかった。今後は、実際の森林大気や植物放出試料の $R_{O3}$ 測定に NO 標準添加法を用いる試みが重要となろう。

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キーワード：生物起源揮発性有機化合物、窒素酸化物、標準添加法
Keywords: biogenic VOCs, nitrogen oxides, standard addition method

 

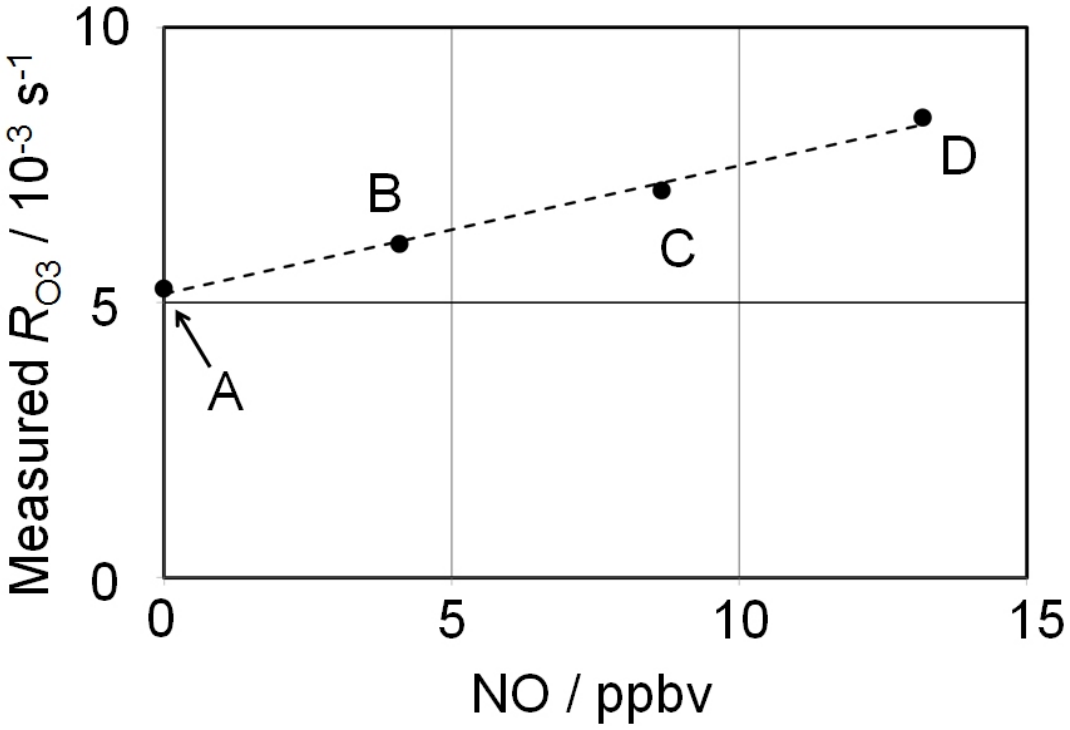


Fig.1 Observed relationship between NO mixing ratio and measured ozone reactivity. A: isoprene standard sample. B, C, D: NO-added isoprene samples. Regression line (dashed line): slope $2.3 \times 10^{-4}$ $s^{-1}$ $ppbv^{-1}$, intercept $5.2 \times 10^{-3}$ $s^{-1}$.

 

# 波照間島および落石岬で観測された大気中$N_2O$濃度のトレンド、季節変動、および短期変動の解析

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

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

キーワード：大気中N2O、温室効果ガス、季節変動、短期変動
Keywords: atmospheric N2O, greenhouse gas, seasonal variation, short-term variation

 

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

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

キーワード：一酸化二窒素　一酸化炭素、流跡線解析、輸送、下部対流圏、北東アジア
Keywords: N2O CO, trajectory analysis, transport, lower troposphere, northeast Asia

 

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

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Carbon monoxide (CO) is emitted from incomplete combustion of fossil fuels and biomass. It affects the concentration of $CO_2$ and $CH_4$ through the reactions with hydroxyl radicals. The major method of the measurements of CO is NDIR. This method is highly sensitive and highly stable. There are a lot of data of the concentration of CO measured with NDIR, but they show only hourly average. In this study, we observed CO concentration in the atmosphere with a mid-IR laser absorption spectrometer that uses a 4.69 $\mu$m quantum cascade laser with wavelength modulation spectroscopy (WMS).
The absorption line for the measurements of CO was at 2131.63 $cm^{-1}$. The laser was scanned at 1.10 Hz. The beam was collimated with $CaF_2$ lens and introduced into the cell. The optical path length was 29.91 m. The signal from a photodetector was processed by the lock-in amplifier.
To assess signal stability and detection limit, 1.02 ppm CO gas was introduced into the WMS system. The precision (1 $\sigma$) of the measurement for 7 hours was 4.18 ‰.
For the measurement of the detection limit of this instrument, we collected the signal of different CO mixing ratio and made the calibration curve. From the slope and the averaged baseline deviation, the detection limit (SNR = 2) was found to be 10 ppb.
Measurements of outside air were conducted on the Kashiwa campus of The University of Tokyo over February 8-11, 2017. During the measurements of ambient air, the calibration was performed every 6-9 hours.
Outside air measurements were conducted over February 8-11, 2017. The CO mixing ratio during the measurements ranged from 0.11 ppm to 0.60 ppm. In the daytime of February 10, the wind brew from northwest. There are no big industrial areas in that direction. So the CO concentration was low and steady. There are some spikes in CO concentration. These may be attributed to cars passing near the observation point.
We performed in situ measurements of CO mixing ratios in ambient air with a near-IR laser absorption spectrometer using WMS. We successfully detected CO mixing ratio change in the ambient air instantly.

 

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

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Continuous measureements of $CO_2$ and $CH_4$ concentration have been carried out with a tower network in Siberia (JR-STATION: Japan–Russia Siberian Tall Tower Inland Observation Network) in order to study the spatial and temporal variations of $CO_2$ and $CH_4$ and estimate the distribution of the flux over this vast area (Sasakawa *et al.,* 2010, 2012, 2013) where only a few atmospheric investigations were made.
The JR-STATION consists of 6 towers (Figure) located at Berezorechka (BRZ) since 2002, at Karasevoe (KRS) since 2004, at Demyanskoe (DEM) and Noyabrsk (NOY) since 2005, at Azovo (AZV) since 2007, and at Vaganovo (VGN) since 2008. Air samples taken at two heights (5-85 m) on each tower were analyzed with an NDIR (LI-COR, LI-820) for $CO_2$ and a $SnO_2$ semiconductor sensor (Suto *et al.,* 2010) for $CH_4$ after passing through the line with a glass water trap, a Nafion membrane dryer (PERMA PURE, MD-050-72F-2), and a magnesium perchlorate. Measurement precision was ±0.3 ppm for $CO_2$ and ±5 ppb for $CH_4$.
We will discuss the long-term variations in $CO_2$ and $CH_4$ observed with the present system. From the year of 2015, we installed a Cavity Ring-Down Spectroscopy (CRDS; Picarro inc.) at KRS, DEM, and NOY. We thus validate the recent data with the data by the CRDSs.

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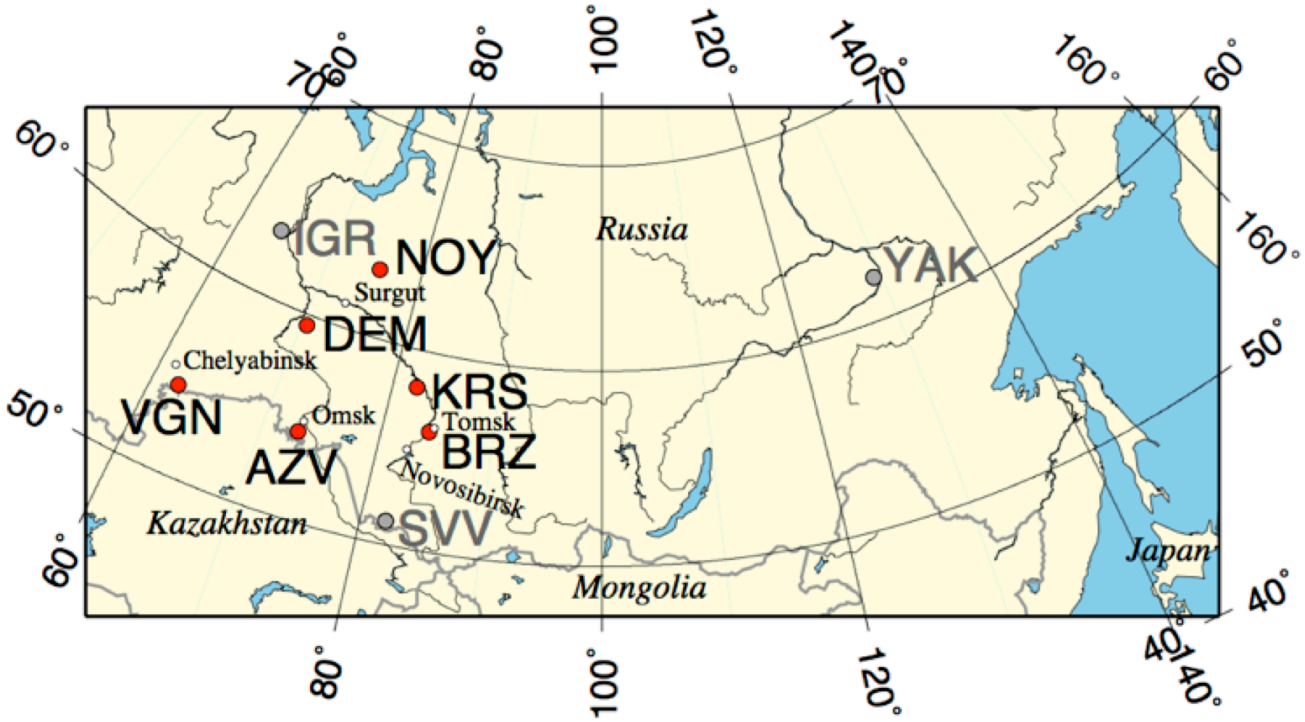
キーワード：二酸化炭素、メタン、タワー観測、西シベリア低地帯
Keywords: Cabon Dioxide, Methane, Tower measurement, West Siberian Lowland



# Estimating vertical fluxes of ozone within the atmospheric boundary layer

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Investigation of the vertical distribution of ozone within the atmospheric boundary layer (ABL) was carried out by use of AN-2 light aircraft as a research platform. Vertical fluxes of ozone and their direction from the ground to the free-tropospheric level were calculated based on the in situ measurement data. Research flights have been performed over the greenhouse gas monitoring station located in a background area (56.1–56.4 N, 84.2–84.8 E) in the vicinity of abandoned village of Berezorechka (West Siberia). The schedule of diurnal flights was as follows: the first one just after the sunrise; the second one at noon; the third one 2-3 hours after noon, when a well-developed turbulence is observed; and the last one just before the sunset. A total of 10 diurnal cycles of measurements were undertaken.
Analysis of the obtained data showed that the rate of ozone influx from upper layers of the atmosphere is 3-10 times less than the ozone production rate in the ABL. Average rate of ozone influx from the free troposphere was about 1 mg $m^{-3}$ $h^{-1}$, but ozone production rate in the ABL was about 5 mg $m^{-3}$ $h^{-1}$, so the major part of ozone is formed by photochemical reactions that occur within the ABL and only 20 % of its content is determined by the influx from the free troposphere. The vertical profiles of the ozone fluxes have shown that their maximum values are observed at heights from 200 to 600 m AGL. The height of the maximum depends on the season: in winter it is lower than 200-300 m, and in summer the maximum is observed at 500-600 m. The value of the ozone flux maximum also depends on the season and varies from 1 $\mu$g $m^{-2}$ $s^{-1}$ in winter to 4.2 $\mu$g $m^{-2}$ $s^{-1}$ in spring.

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

Keywords: Atmosphere, Ozon, Distribution

 

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

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Studying the formation and dynamics of ozone in the atmosphere is important due to several reasons. First, the contribution of tropospheric ozone to the global greenhouse effect is only slightly less than that of water vapor, carbon dioxide, and methane. Second, tropospheric ozone acts as a strong poison that has negative effects on human health, animals, and vegetation. Third, being a potent oxidizer, ozone destroys almost all materials, including platinum group metals and compounds. Fourthly, ozone is formed in situ from precursors as a result of photochemical processes, but not emitted into the atmosphere by any industrial enterprises directly.
In this work, we present some results of the study aimed at the revealing relationship between ozone formation rate and surface air temperature in the background atmosphere. It has been found that this relationship is nonlinear. Analysis of the possible reasons showed that the nonlinear character of this relationship may be due to a nonlinear increase in the reaction constants versus air temperature and a quadratic increase in the concentration of hydrocarbons with increasing temperature.
This work was supported by the Russian Foundation for Basic Research (grant No 17-05-00374).

Keywords: Atmosphere, Ozon, Temperature

 

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

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

キーワード：オゾン、成層圏、重力波、ロスビー波
Keywords: Ozone, Stratosphere, Gravity wave, Rossby wave

 

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

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

This research was supported by Science and Technology Research Partnership for Sustainable Development (SATREPS), Japan Science and Technology Agency (JST)/Japan International Cooperation Agency (JICA).

キーワード：成層圏、オゾン、DIAL、MLS、MIROC、化学輸送モデル
Keywords: stratosphere, ozone, DIAL, MLS, MIROC, CTM

 

# 北海道陸別観測所高分解能フーリエ変換型赤外分光器による対流圏・成層圏微量成分の長期観測

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

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名古屋大学宇宙地球環境研究所は1995年に北海道陸別町（43.46˚N, 143.77˚E, 380 m a.s.l.）に高分解能フーリエ変換型赤外分光器（FTIR）（Bruker IFS120M）を、2014年には同地に国立環境研究所が高分解能FTIR（Bruker IFS120/5HR）を設置し、大気微量分子による太陽光吸収スペクトルの観測を共同して行っている。観測は波長2~15 mmの領域で行い、0.0035 $cm^{-1}$の波数分解能でスペクトルを取得している。これまでに私たちは解析ソフトウエアSFIT4 (version 0.944)を用い、大気組成変化モニタリングネットワーク赤外グループ（NDACC/IRWG）で標準化された解析パラメータを適用して対流圏・成層圏微量成分の高度分布解析を行った。高度グリッドや微量成分の初期推定値等の解析パラメータの標準化により、各NDACC観測地点間での解析精度を均一化し、全球での微量成分のトレンド解析やモデル等のデータとの比較を容易にすることが目的である。解析の補助データとして、気温・気圧分布はNCEP再解析データ、水蒸気以外の微量分子の初期推定値としてWACCM V6による1980~2020年の40年予測計算の2000年における年平均値を用い、水蒸気については観測スペクトルを用いて事前に高度分布解析を行い、それを初期推定値として他の大気微量成分解析に用いた。これまでに11種（$O_3$、HCl、HF、$HNO_3$、$ClONO_2$、$CH_4$、$C_2H_6$、$N_2O$、CO、HCN、$CCl_4$）の微量分子についてカラム全量及び高度分布の解析を進め、1995年から2016年までの時間変動を得た。各微量分子の長期変動に着目すると、成層圏$O_3$カラム量には明瞭なトレンドの変化は見られないのに対し、対流圏$O_3$カラム量には2000年代以降、有意な減少が見られた。また2000年代以降にHClやClONO2のカラム量が減少トレンドを示すなど、概ね全球的な傾向と一致していることがわかった。

発表ではこれらの要因について議論するとともに、得られた微量成分ごとの時間変動の特徴等について報告する。

キーワード：大気組成、赤外分光観測、長期トレンド
Keywords: Atmospheric composition, Infrared spectroscopy, Trends

 

# 2015年初冬に北海道陸別で観測されたCOカラム平均混合比（XCO）の急増イベントの原因解析

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

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国立環境研究所では2014年から北海道陸別町（43.46˚N, 143.77˚E, 380 m a.s.l）に設置した高分解能FTIR（Bruker IFS120/5HR）を用いて、全量炭素カラム観測ネットワーク(Total Carbon Column Observing Network、TCCON)として近赤外域での太陽光吸収スペクトル観測を行い、大気中微量成分（$CO_2$、$CH_4$、COなど）のカラム平均混合比を求めている。これまでの観測から各微量成分の様々な時間変動が得られているが、我々はデータの中でCOカラム平均混合比（XCO）が短期間に増減するイベントに着目した。特に、2015年初冬にはXCOが11月13日に増加し始め、30日に季節平均値の約4.5倍となり、12月9日に季節平均値に戻るという増加・減少が見られた。このとき$CO_2$（$XCO_2$）や$CH_4$（$XCH_4$）にも同じような変動が見られることから、燃焼起源の気塊の流入の影響が推測された。

そこで我々は、この変動が見られる期間について流跡線解析を行った。解析にはFLEXPART（Stohl et al. 2005）を用い、気象場データにNCEP CFSv2-6時間解析データを使用した。それぞれの日において1km、5km、7kmの高度で2週間の後方流跡線計算を行うと、中国東北部からの影響と遠方からの影響が示唆される結果が得られた。そこでこれら結果をもとに、MODISによるhotspotデータから後方流跡線から推定されるCOの放出場所と日時の範囲のものをすべて取り出し、領域ごとにそれからの前方流跡線計算を行った。計算の結果、中国東北部起源のCOの流入がその他遠方の影響の約10倍大きいことがわかり、観測されたXCOの急増は中国東北部の燃焼起源の気塊流入によることが確認できた。

発表では、流跡線解析の詳細とともに観測期間中に見られる他の急増イベント例との比較、他の微量成分変動との関連について議論する。

キーワード：近赤外域太陽吸収スペクトル、対流圏一酸化炭素、流跡線解析
Keywords: near-infrared solar absorption spectra, tropospheric carbon monoxide, trajectory analysis

 

# FTIRで観測されたつくばにおけるエタンの近年の増加およびその後の減少

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

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東北大学と国立環境研究所では、国立環境研究所所有の高分解能フーリエ変換型赤外分光計(FTIR)を用いて、つくばにおいて1998年12月より大気微量成分の地上観測を行っている。今回は昨年に引き続いて$C_2H_6$カラム全量の経年変化を報告する。$C_2H_6$は$CH_4$に次いで多く存在する炭化水素類であり、化学反応過程も類似した部分が多い。そのため、$C_2H_6$が$CH_4$の濃度に影響することによる間接的な温室効果がある。また、汚染大気中のオゾン生成に寄与する他、$C_2H_6$の酸化過程で生成するアセトアルデヒド($CH_3CHO$)と$NO_2$からPeroxyacetyl nitrate(PAN)が生成され、これが窒素酸化物の長距離輸送に寄与することから、越境大気汚染にも寄与する。発生源は天然ガス、バイオ燃料、バイオマス燃焼等人為起源が主であり、そのため地表付近の濃度は北半球では数ppbv程度である一方、南半球では数百pptvといった値が報告されている。

FTIR観測の国際的グループであるNDACC/IRWGではこのような$C_2H_6$の変動要因を解明するため、世界各地のFTIR観測の結果を総合して解析することになり、我々もつくばの観測結果を提供している。解析にはロジャーズ法を用いたスペクトルフィッティングプログラムSFIT4を使用し、Franco et al., [2015]と同様のパラメータで行っている。ただし、解析に用いる波数領域は3μm付近の①2976.66 - 2977.059 $cm^{-1}$と②2983.20 - 2983.50 $cm^{-1}$のふたつで、Franco et al. [2015]で用いているもう一つの領域③2986.45 - 2986.85 $cm^{-1}$は用いない。これは③の領域には$H_2O$の強い吸収線があり、日本のような高湿度地域では解析に適さないためである。

解析は2001年5月-2016年12月について行い、日平均したカラム全量の経年変化を調べたところ、2001-2008年の期間では若干の減少傾向(-0.4%/year程度)であったものが2009-2013年にかけて増加(2.2%/year程度)し、2014-2016年に再び減少(-0.6%/year程度)する様子が見られた。原因に関してはまだ調査中であるが、2014年以降の減少については原油価格の下落に伴う原油生産量の増加が$C_2H_6$の発生源である天然ガスやバイオ燃料の生産量の減少を招き、その結果$C_2H_6$の発生量が減少した、といったことが影響しているようである。

キーワード：フーリエ変換型分光計、温室効果気体、エタン
Keywords: FTIR, Greenhouse Gas, ethane

 

# MAX-DOASによって観測された九州の二酸化硫黄濃度変動の要因解析
# Causes leading to enhancements in sulfur dioxide concentration observed by MAX-DOAS in Kyusyu

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代表的な大気汚染物質のひとつである二酸化硫黄($SO_2$)は、酸性雨やエアロゾルの前駆気体としての役割を通じて、人体や生態系、気象などに影響を及ぼすため、その濃度変動の要因を解明することは重要である。これまで$SO_2$の観測は主に地表濃度に焦点が当てられ実施されてきたが、これをエアロゾルの予測モデルへのデータ同化等の応用研究に利用する上では空間代表性に留意する必要がある。そのような背景の下、本研究では、アジア大陸からの長距離輸送や火山ガスなど様々な影響を受けることが考えられる九州地方(福岡県春日市)において多軸差分吸収分光法(Multi-Axis Differential Optical Absorption Spectroscopy : MAX-DOAS)と呼ばれる地上リモートセンシング手法による連続観測を行った。波長域310-320nmの測定スペクトルをDOAS法で解析し、高度0-1km層内の$SO_2$平均濃度を水平スケール10km程度で導出した。はじめに、2014年7-8月において日最大値が5ppbvを超える日を高濃度日の基準にしたところ、7日間を特定することができた。これらの日を対象にバックトラジェクトリー解析を行ったところ、火山起源と大陸起源の2つのケースに分類することができ、火山起源については阿蘇山および桜島、大陸起源については中国や韓国からの長距離輸送の影響が示唆された。この結果を踏まえ、解析対象期間を2014年1月-2016年9月に延ばして、火山起源および大陸起源の影響について定量的な解析を行った。この期間の日最大値の平均値は3.3ppbv、日最大値の中央値は2.1ppbvであった。まずは火山起源の影響について、空気塊が火山を通過したと判断する範囲の大きさを変えた場合で日最大値の平均値の比較を行ったところ、火口中心近くを通過してきたほうが高濃度を示すという傾向が見られた。阿蘇山の影響を受けた場合の日最大値の平均値は約6ppbv、桜島は約4ppbvであり、阿蘇山の影響の大きさが示唆された。次に大陸起源の影響について、日最大値の平均値と、中国・韓国上空での空気塊の滞留時間および中国・韓国から輸送されてくるまでの時間の長さとの関係についてそれぞれ比較を行ったが、相関は見られなかった。また、大陸からの空気塊が到達した日における日最大値の平均値は約2.4ppbvであった。都市部周辺からの空気塊が到達した場合を見ても同様の結果であった。これらの結果より、九州において大陸の影響は火山の影響よりもが小さいことが示唆された。本講演では2016年10月8日に発生した阿蘇山の爆発的噴火の影響についての考察も含め、解析対象期間をさらに延ばした結果の議論を行う。

キーワード：二酸化硫黄、MAX-DOAS、火山
Keywords: SO2, MAX-DOAS, valcano

 

# GOSAT/TANSO-FTS SWIRバンド、TIRバンド及びNICAM-TMの温室効果ガス気柱平均濃度データの比較解析

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

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GOSAT（Greenhouse gases Observing SATellite）は2009年1月23 日に打ち上げられた温室効果ガス観測技術衛星であり、GOSAT搭載のTANSO-FTS（Thermal And Near infrared Sensor for carbon Observation –Fourier Transform Spectrometer）の熱赤外（TIR）バンドから二酸化炭素及びメタンの鉛直濃度分布が、短波長赤外（SWIR）バンドから気柱平均濃度$XCO_2$及び$XCH_4$が導出できる[Saitoh et al., 2009; Yoshida et al., 2011]。本研究では、GOSATのTIRバンドから導出された二酸化炭素及びメタンの鉛直濃度分布データ（Version 1）[Saitoh et al., 2016]から$XCO_2$、$XCH_4$を計算し、SWIRバンドの$XCO_2$、$XCH_4$データ（Version 2.21）及び大気輸送モデルNICAM-TM（Nonhydrostatic ICosahedral Atmospheric Model-based Transport Model）[Niwa et al., 2011]の$XCO_2$データと比較し、それぞれのデータの特徴を調べた。なお、航空機観測データとの比較に基づいて評価されたTIRバンドの二酸化炭素データのバイアスを補正した上で$XCO_2$を計算した。

TANSO-FTSのTIRバンドとSWIRバンドの$XCO_2$データ、TANSO-FTSの濃度導出の際の先験情報（アプリオリ）であるNIES-TM05モデル[Saeki et al., 2013]の$XCO_2$データ、NICAM-TMの$XCO_2$データについて、濃度緯度分布の比較を行った。TIRバンドの$XCO_2$は、サハラ砂漠地域を除く北半球陸域ではSWIRバンドの$XCO_2$より若干小さく、逆に南半球陸域では若干高い傾向が見られた。サハラ砂漠地域においては、TIRバンドの昼間の$XCO_2$データがSWIRバンド及びNICAM-TMの$XCO_2$データよりも極端に低い値を取っていることから、昼間のTIRバンドのリトリーバル処理時に設定しているサハラ砂漠地域での地表面パラメータに問題があることが示唆された。一方、地上に強いソースがないハワイ周辺では、TIRバンドとSWIRバンドの$XCO_2$データが1%以内で一致していた。

キーワード：GOSAT、XCO2、XCH4
Keywords: GOSAT, XCO2, XCH4

 

# GOSAT/TANSO-FTS TIR スペクトルを用いた地表面温度及び地表面射出率の同時推定

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

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温室効果ガス観測技術衛星GOSAT（Greenhouse gases Observing SATellite）に搭載されているTANSO-FTS（Thermal And Near infrared Sensor for carbon Observation –Fourier Transform Spectrometer）は、短波長赤外バンド及び熱赤外（TIR）バンドで二酸化炭素とメタンのカラム量と鉛直濃度分布を観測することができる[Yoshida et al., 2011; Saitoh et al., 2009]。TIRバンドのスペクトルから気体濃度を導出する際、地表面パラメータの誤差は気体濃度の導出精度を悪化させることがわかっている[Saitoh et al., 2009]。そこで、本研究では、TANSO-FTSのTIRスペクトルから地表面射出率及び地表面温度の同時推定を試みた。まず初めに、地表面射出率の波長依存性が相対的に小さいことを利用してAIRS （Atmospheric Infrared Sounder）の高波長分解能観測スペクトルから地表面パラメータの同時推定を試みた先行研究［松井・森山, 2008］に従って、TANSO-FTSのTIRスペクトルから地表面パラメータを同時推定する手法を検討した。さらに、TANSO-FTSのTIRバンドの実観測スペクトルに開発した手法を適用した。本研究では、地表面射出率の特徴が比較的わかっている海面に注目し、推定手法の有効性を調査した。

800 $cm^{-1}$から1000 $cm^{-1}$の理論スペクトルから、連続吸収を考慮していない場合の大気の透過率が0.95以上となる波数チャンネルを「スムースパート」とし、このチャンネルセットをもとに地表面パラメータの推定を行った。初めに、中緯度の海上のTANSO-FTSのTIRバンドの観測スペクトルについて、地表面温度幅を286 Kから310 Kとし、その範囲内で1 K刻みで地表面温度を変えていき、最も地表面射出率が波長方向に滑らかになる地表面温度を算出した。その結果、地表面温度は294 Kとなり、観測地点の海面温度（293.4 K）と近い値を示した。また、同時推定された900 $cm^{-1}$における地表面射出率は0.97となり、Aster Spectral Libraryの海水射出率0.99と近い値となった。

さらに、開発した手法を2013年1月1–3日の海上（10–20˚N, 160–170˚E）の複数のTIRバンドの観測スペクトルに適用した。我々の手法で導出した地表面温度と近傍のMODISの海面温度を比較したところ、両者の差が小さい観測地点では同時導出された地表面射出率はAster Spectral Libraryの海水射出率と概ね近い値になった。しかし、地表面温度と海面温度に大きな差が見られる観測地点では、同時導出された地表面射出率も海水射出率から大きな隔たりが見られた。講演では、導出結果の精度評価、他の季節・領域での推定結果、地表面パラメータを導出する際の各種パラメータを変更した際の導出結果への影響についても報告する予定である。

キーワード：GOSAT、熱赤外バンド、地表面温度、地表面射出率
Keywords: GOSAT, thermal infrared band, surface temperature, surface emissivity

 

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

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Large areas are burned every year in the boreal Eurasia. Pollutants emitted from fires in the Boreal Eurasia could be transported to the Arctic and the consequential deposition could accelerate the Arctic warming. However, there are so far limited knowledges on the wildfire pattern in the boreal Eurasia and the causes. Our purposes are to clarify the temporal and spatial pattern of wildfire and to investigate the factors affecting wildfire occurrence in the boreal Eurasia during 2005-2014.
Focusing on a domain in 50–75˚N, 30–180˚E, we divided the boreal Eurasia into 15 sub-regions and investigated the wildfire occurrences based on the Moderate Resolution Imaging Spectroradiometer MCD64A1 burned area product. Lands were burned mainly (96%) in >100ha size, although with large small fires (<100ha) counts (58%). Interannually, large land losses (>30 million hectares) occurred in 2008, 2010, 2012 and 2014 over the whole domain. Seasonally, wildfires occurred since April, peaked in July to August, and continued until October. Spatially, six high fire-prone regions were identified locating at the southwestern Russia, Kazakhstan, southwest Siberia, central Siberia, eastern Siberia and the Far East. Furthermore, we investigated the relations of burned area with climatic indices such as temperature, precipitation and soil drought index (Palmer Drought Severity Index, PDSI) in each sub-region. It was found that the burned area in southwest Russia, Kazakhstan, west Siberia had positive relation with temperature ($p < 0.05$). On the other hand, burned area had negative relations with precipitation and PDSI ($p < 0.05$) in most of the fire-prone regions. These results indicate that wildfire occurrence were fostered by the dry soil and air conditions. Furthermore, burned area is likely related to snow melting date, to which early snow melting date caused larger burned area. Our study implies that under a warmer world, wildfires in the boreal Eurasia tend to be severer and mega-fires more frequent.

 

# ガス状有機物と一次・二次エアロゾルとの不均一反応による粒子内有機組成変化の実時間測定

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

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揮発性有機化合物（VOC）の光酸化反応で生成する二次有機エアロゾル（SOA）は、人への健康被害が懸念される他、気象場の変化によって地域スケールでの水循環等や将来の気候にも影響を及ぼすことが懸念されている。これらの影響を定量的に評価していくには、SOAの生成過程や化学組成の理解が必要であり、具体的にはVOCの大気酸化過程で生成する半揮発性有機化合物（SVOC）の特定、そのSVOCのガス相・粒子相分配の定量化、SOA中の成分の把握、及び粒子内での変質過程の解明が必要である。最近我々は、室内実験において二次有機エアロゾル中のアルデヒドが絡んだオリゴマー等のリアルタイム計測に成功した。このことにより、ガスー粒子間の反応でオリゴマーを生成する過程のモデル化が可能になると期待される。そこで本研究では、ディーゼル排出粒子や炭化水素の酸化反応で生成する二次有機エアロゾルにガス状アルコール、アルデヒド、カルボン酸、ヒドロペルオキシドを添加し、粒子の成長と粒子内でのオリゴマー等の変化を調べる実験を行った。

キーワード：陽子移動反応質量分析法、オリゴマー、不均一反応、二次有機エアロゾル、ディーゼル排出粒子
Keywords: Proton transfer reaction-mass spectrometry, Oligmer, Heterogeneous reaction, Secondary organic aerosol, Diesel exhaust particle

 

# 原子間力顕微鏡を用いた個別雲凝結核の吸湿特性評価
# Hygroscopic characterization of individual cloud condensation nuclei with atomic force microscope

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大気エアロゾルの多くは吸湿性を持ち、水蒸気を取り込み雲凝結核（CCN）として働くことで間接的に気候に影響を与える。吸湿性パラメーターκはエアロゾル粒子の吸湿性を示す単一パラメーターである（Petters and Kreidenweis., 2007）。κを求めるには、相対湿度・乾燥粒径・吸湿成長因子（gf）・気相液相界面の表面張力の正確な測定が必要である。その測定のため、従来の研究では、主に吸湿性タンデム微分型静電分級装置（HTDMA）が広く用いられてきた。

HTDMAの観測から求められるのκは、大多数の粒子の平均を反映しており、必ずしも極端な吸湿性を示す一部の粒子の存在を反映しない（Morris et al., 2016）。または、液滴の表面張力を直接的に測定する手法がなかったために、主に表面張力を純水かバルク溶質のそれ過程してκを導出してきた点などが問題として挙げられる。

我々は原子間力顕微鏡（AFM）を用いて、単一液滴の表面張力とgfをそれぞれ直接測定し、原理上、個別粒子のκを直接的に求める手法の確立を目指している。本研究では、塩化ナトリウム（NaCl）のgfと表面張力を測定した。この実験で得られたgf (80% RH) は1.73±0.35となり、先行研究（Morris et al., 2016）の値と標準偏差内で一致する。表面張力の導出においては、その算出に必要な保持力が平均19±3 nN となり、先行研究（Morris et al., 2015）で報告されている30 nN よりも僅かに低くなった、この差は双方の研究で使用されたカンチレバーの形状の違いによるものだと考えられる。この実験で得たgf、乾燥粒径、表面張力から導出される吸湿性パラメーターκは1.10となった。この値は先行研究（Koehler et al., 2006; Clegg and Wexler., 1998）で報告されているκの値とよく一致したことから、この手法が妥当なκを導出できる可能性を示した。将来的には、この手法を用いて様々な表面活性を示す物質、無機塩や有機種を混合した物質などを分析し、最終的には実際の大気エアロゾル物質に適用する予定である。

 

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

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

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

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

 

# 東京スカイツリーで測定された都市上空の雲凝結核の特性
# Characteristics of Cloud Condensation Nuclei over the Urban Areas Measured at Tokyo Skytree

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大気エアロゾルが気候に与える影響は直接効果と間接効果（雲調整効果）がある。直接効果とはエアロゾルが太陽光を直接散乱・吸収する効果、間接効果とは雲を形成したときに雲凝結核（CCN）として働き、雲の光学特性や寿命を変化させる効果である。放射強制力に与える雲調整効果に関しては科学的理解度が低く、大きな不確かさを持っている(IPCC, 2013)。したがって、多くの地域でエアロゾルのCCN特性を測定することが必要となっている。世界的に定点観測の少ない都市上空のCCN特性を調査するため、本研究では、東京スカイツリーにおいてCCNを観測した。

観測は、2016年6月3日~6月30日、東京スカイツリー458 m（35.71˚N , 139.81˚E）地点において行った。雲凝結核計( CCNC )、走査型移動度粒径分布測定器 ( SMPS )を用いてCCN数濃度 ($N_{CCN}$) とCN数濃度 ($N_{CN}$) を同時に測定した。得られた結果から、CCN活性比を求めることができる。本研究では拡散ドライヤーを通して大気試料の相対湿度を30%以下にして4段階の過飽和度（SS%）の$N_{CCN}$、$N_{CN}$とその粒径分布を測定し、$\kappa$-ケーラー理論(Petters and Kreidenweis, 2007)を用いて吸湿性パラメータ（$\kappa$）を算出した。またFog Monitorで測定した霧粒数濃度（$N_{Fog}$）、霧粒の有効半径（$R_{eff}$）とあわせて解析を行った。東京スカイツリーで得られた結果を同様の観測が行われた富士山頂の観測データ（渡辺, 2015）と比較した。

東京スカイツリーでは富士山頂と比べると全体的に$N_{CCN}$は大きく、$\kappa$は小さいことが確認できた。また気塊の輸送起源によるCCN特性のはっきりとした傾向は見られなかった。

霧雨を含まない雲で覆われている期間のデータを用い、CCN特性について調べた結果、$N_{Fog}$と$N_{CCN}$との間に正の相関はなかった。 また、Twomey効果(Twomey,1959)として知られる微物理的関係のように、$R_{eff}$と$N_{CCN}$との間に負の相関はなかった。これは、東京スカイツリーでは$N_{CCN}$の変動が小さかったためと考えられる。過飽和度0.12%における$R_{eff}$と$N_{CCN}$の関係を富士山頂の結果 と比較したところ、富士山頂で得られた$R_{eff}$と$N_{CCN}$との間の負の相関の一部とみなすことができた。

キーワード：雲凝結核数濃度、凝結核数濃度、有効半径、霧粒数濃度
Keywords: cloud condensation nuclei number concentration, condensation nuclei number concentration, effective radius, fog droplet number concentrations

 

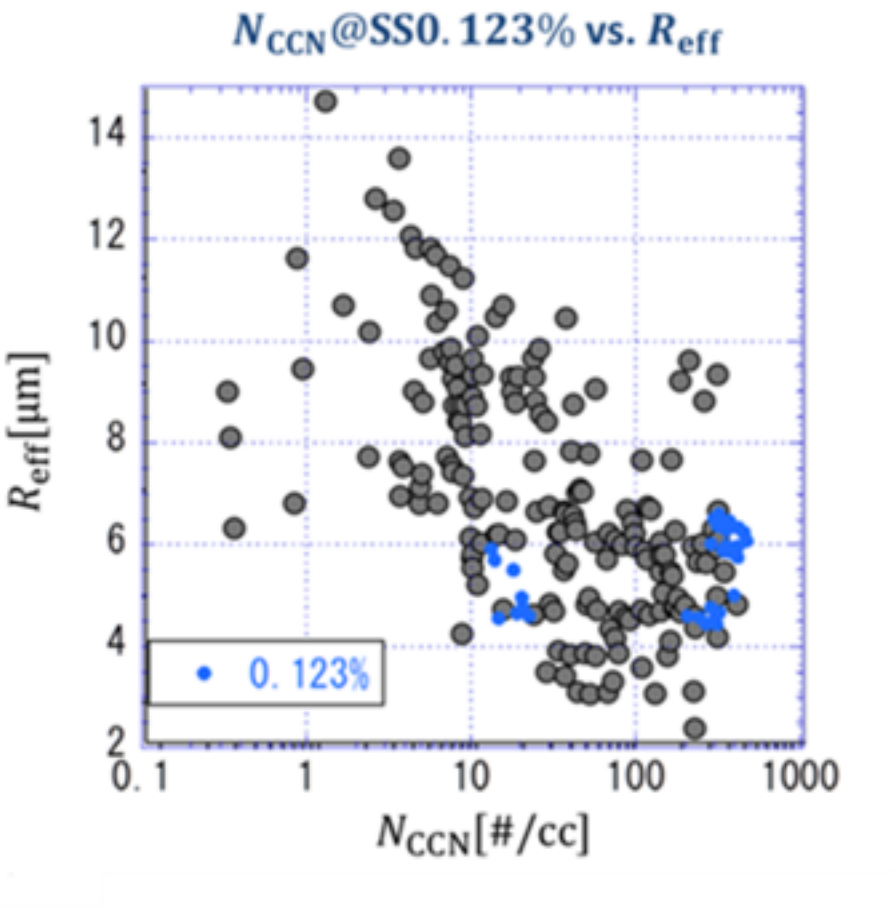


図 $R_{eff}$と$N_{CCN}$@SS0.123%（青）、
$N_{CCN}$@SS0.15%（渡辺, 2015）（黒）の関係

 

# Wet removal mechanisms of black carbon observed in Tokyo and Okinawa

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Quantitative understanding of wet removal process of black carbon (BC) is important because it controls temporal and spatial distribution of BC in the atmosphere. There are two categories of wet removal mechanisms: nucleation scavenging and impaction scavenging. Theoretically, the removal efficiencies of both mechanisms depend on microphysical properties (size($D_{BC}$), coating thickness, and hygroscopicity) of BC. However, the relative contribution of these mechanisms to the removal of BC and the key BC microphysical property controlling the wet removal efficiency of BC have never been tested by direct observation. In this study, we conducted ground-based observations of BC in air and in rainwater in Tokyo and Okinawa, using a new observational method (Mori et al., 2016). We compared the observed size-dependent wet removal efficiency of BC particles ($RE(D_{BC})$, defined as the ratio of the measured size distributions of BC in rainwater to that of BC in ambient air) with the size-dependent number fractions of BC particles scavenged by nucleation ($F_{ccn}(D_{BC})$) and impaction ($F_{imp}(D_{BC})$) mechanisms. The $F_{ccn}(D_{BC})$ and $F_{imp}(D_{BC})$ were estimated from the observed microphysical properties of BC in the air before precipitation starts and the observed droplet size and intensity of precipitation.
The size dependence of $RE(D_{BC})$ showed remarkable differences for the observed 42 precipitation events (31 events in Tokyo and 11 events in Okinawa). The size dependence of $RE(D_{BC})$ for BC-containing particles with $D_{BC} > 100$ nm was successfully explained by the size dependence of $F_{ccn}(D_{BC})$, whereas the contribution of impaction mechanism can be dominant for particles with $D_{BC} < 100$ nm. For $D_{BC} > 100$ nm, when BC particles have relatively thick coating (shell/core ratio > 1.2), the $RE(D_{BC})$ depended little on $D_{BC}$ and the major fraction of BC were removed via nucleation scavenging. On the other hand, when BC particles are nearly bare or have less coating, the $RE(D_{BC})$ highly depended on $D_{BC}$, and $RE$ (i.e. $F_{ccn}$) is more sensitive to the hygroscopiticy of coating materials and the maximum supersaturation of water vapor that BC particles would experience during moist convection process. Our results show that the coating thickness of BC is the key parameter controlling wet removal of BC, and indicate that for accurate simulation of vertical transport of BC from the boundary layer to the free troposphere, detailed modeling of microphysical properties of BC and atmospheric supersaturation is required especially for BC particles with thin coatings.

キーワード：ブラックカーボン、湿性除去、地上観測
Keywords: Black carbon, Wet removal mechanism, Field observation

 

# 西部北太平洋域での船上ブラックカーボン観測：2016年1月における輸送イベント解析

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

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

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