

SESSION 5

TROPOSPHERIC OBSERVATIONS AND ANALYSES





2011 NDACC Symposium

Network for the Detection of Atmospheric Composition Change



Oral Session

5-1 Time-series of biomass burning products from ground-based FTIR measurements at Reunion Island (21°S, 55°E) and comparisons with the model IMAGES.

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Reunion Island (21°S, 55°E) is one of the few subtropical, southern hemisphere stations of the ground-based Network for the Detection of Atmospheric Composition Change (NDACC). Furthermore, Reunion Island is situated in the Indian Ocean, at 2000 km from southeast Africa and only 700 km from Madagascar. It is therefore a good location to observe the transport of biomass burning products from these regions. Ground-based Fourier transform infrared (FTIR) solar absorption observations are sensitive to a large number of biomass burning products. We present the retrieved time-series of acetylene (C₂H₂), ethane (C₂H₆), formic acid (HCOOH), methanol (CH₃OH), formaldehyde (HCHO) and hydrogen cyanide (HCN) from FTIR measurements at Reunion Island, from August to October 2004, from May to October 2007, and from May 2009 to December 2010. The measurements in 2007 and 2009-2010 allow the observation of seasonal variability. The comparisons of these ground-based data with the simulations by the model IMAGES are discussed.



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Oral Session

5-2 New results on validation of EOS-Aura OMI NO₂ data using ground-based NO₂ measurements at Zvenigorod, Russia

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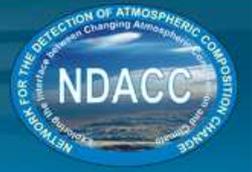
We present the results of comparison between Ozone Monitoring Instrument (OMI) data of NO₂ measurements aboard the NASA EOS-Aura satellite and correlative ground-based twilight measurements at Zvenigorod station located 40 km west to Moscow in 2004-2011. Compared quantities are unpolluted column and tropospheric column amounts of NO₂ which are standard products of OMI measurements. Vertical profiles of NO₂ are retrieved from ground-based observations at Zvenigorod, which allows determination of NO₂ contents in required atmospheric layers. We compare the OMI unpolluted column NO₂ amounts to the NO₂ contents in the vertical atmospheric column above the atmospheric boundary layer retrieved from ground-based measurements. The OMI tropospheric column NO₂ contents are compared to the NO₂ contents in layer 0-10 km retrieved from ground-based measurements. The NO₂ columns observed by our ground-based instrument have been interpolated to the time of OMI measurements using a one-dimensional photochemical model. The unpolluted NO₂ columns from OMI measurements are generally less than those from ground-based measurements by $(0.05 \pm 0.02) \times 10^{15}$ molecules/cm², or $(1.8 \pm 0.6) \%$, with the mean square root discrepancy 0.45×10^{15} molecules/cm² (~17%). The correlation coefficient between all the OMI and ground-based unpolluted NO₂ columns is 0.92. It is contributed substantially by the NO₂ annual cycle. However the monthly dependent correlation coefficients are less than 0.5. On average, the OMI unpolluted NO₂ columns underestimate ground-based measurements in summer (by up to 0.3×10^{15} molecules/cm², or 7%, in June) and overestimate in winter (by about 0.1×10^{15} molecules/cm², or 10%). The tropospheric NO₂ columns derived from OMI measurements are on average by $(1.6 \pm 0.3) \times 10^{15}$ cm⁻², or approximately 40%, less than those derived from ground-based measurements. The difference is largest in winter. The correlation coefficient between these data is about 0.2 for the entire period, being larger in winter. Surprisingly, the monthly correlation coefficients between the tropospheric columns can approach 0.6-0.7, the values that are larger than the maximum monthly correlation coefficient between the unpolluted NO₂ columns. More detail analysis shows that the NO₂ contents derived from OMI measurements are sensitive to cloudiness. The correlation coefficient between the OMI and Zvenigorod tropospheric NO₂ columns calculated for the entire period of observations increases with decreasing cloud fraction approaching 0.45 under cloudless conditions. Unlike this, the correlation coefficient between the unpolluted columns decreases with decreasing cloud fraction down to the value 0.86 under cloudless conditions. Further analysis of OMI and Zvenigorod data involves spectral and cross-

spectral techniques. The dominant mode of variability of the unpolluted and tropospheric NO₂ columns is the annual variation. The annual cycles of the unpolluted columns derived from OMI and ground-based measurements are highly coherent with each other with zero phase shift. However the annual cycle of the OMI tropospheric NO₂ columns lags behind the annual cycle of the ground-based tropospheric NO₂ columns by about 3.5 months (approximately by a quarter of the annual period). According to ground-based measurements, the annual maximum of tropospheric column NO₂ in the neighborhood of Zvenigorod is observed in winter, which is explained by more intensive transport of pollution to the area during this season, but in early autumn according to OMI data. The approximate orthogonality of these annual variations results in the afore-noted small value of the correlation coefficient between the OMI and ground-based tropospheric NO₂ columns. The correspondence of the phases of variations in the tropospheric NO₂ columns of smaller time scales is generally better. Among these variations are the quasi-fortnightly and quasi-bimonthly variations. Concerning the unpolluted NO₂ columns we would mention variations with periods of about 10 and 20 days, phase shift between the variations in OMI and ground-based Zvenigorod data being close to zero but the spectral coherence between the OMI and ground-based NO₂ data at these periods being not as high as at the annual period. The results presented show that care should be taken under analysis of the NO₂ variability using satellite data. With tropospheric data, an incorrect conclusion may be even drawn for the NO₂ annual cycle.



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Oral Session

5-3 Correlating boundary-layer NO₂ with column measurements: Observing ground-level NO₂ from space

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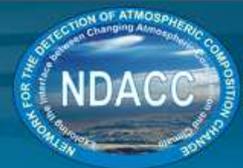
NO₂ is one of seven pollutants in the United States for which EPA has established national ambient air quality standards (NAAQS). NO₂ plays a key role in the formation of O₃ and is of significant importance due to O₃'s oxidative capacity and its influence on radical chemistry (e.g. OH, HO₂, CHO, etc.). NO₂ is also significantly associated with mortality (Stieb et al., 2008) and thereby has far reaching effects on human health, crop yield/photosynthesis, atmospheric oxidative capacity, and radiation balance either directly or via its role in ozone/radical chemistry. Major sources of NO_x (NO + NO₂) include motor vehicles, power plants, fossil fuel combustion, burning biomass fuel, and natural sources (e.g. wildfires). However, surface measurements of "true" NO₂ are very sparse across the United States as most NO₂ instruments use a molybdenum-oxide converter that is incapable of distinguishing between NO₂ and NO_y. Furthermore, the spatial coverage required for air-pollution monitoring can only be achieved via satellite; though, to date, the relationship between column and in-situ observations has remained obscure. Current satellite-based NO₂ instruments lack the temporal and spatial resolution required for monitoring trace-pollutant species on timescales appropriate for regulation enforcement or chemical forecasting. The upcoming GEO-CAPE mission will be in geo-stationary orbit over the United States with improved resolution allowing hourly city-scale NO₂ observations. Translating these column observations into boundary-layer mole fractions provides a direct NO₂ data set that can significantly improve the understanding of emission, chemical transportation, effectiveness of control strategies, and predictive capabilities. Herein we present data collected since 2009 at NASA's Langley Research Site in Hampton, VA. NO₂ column densities were recorded approximately every 2 minutes using a ground-based column instrument (Pandora), and are compared to surface NO₂ measured with an LED-based NO₂ photolytic converter instrument operated by U.S. EPA. Pandora has been previously shown to be in excellent agreement with OMI. Pandora NO₂ column values had the stratospheric component removed (from OMI), with the residual tropospheric NO₂ assumed to be predominantly in the boundary layer due to its short photolytic lifetime. The EDAS40 model was used to convert these column values to boundary-

layer mole fractions (ppb) throughout the day, as the boundary layer changed. Correlation analysis shows this boundary-layer correction factor accounts for up to 70% of the variability between the two observation techniques, with an average difference of ≈ 2 ppb, indicating the above assumptions to be valid most of the time. Similar methods may be useful in comparing column data to ground-based air-quality networks over the globe.



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Oral Session

5-4 MAX-DOAS observations and retrievals: From marine halogen oxides emissions to air quality studies

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We present a range of examples where the MAX-DOAS (Multi-AXis Differential Optical Absorption Spectroscopy) technique has been used to retrieve partial columns in the lower troposphere and the corresponding surface mixing ratios of a variety of trace gases. The examples range from shipboard observations of marine boundary layer halogen oxides in the tropics and mid-latitudes to air quality studies investigating the NO₂ abundances at several locations.

The MAX-DOAS technique uses scattered sunlight received from multiple viewing angles to obtain information of the abundance and altitude distribution of the trace gas of interest. The slant columns retrieved for the different viewing angles are then converted into a profile using an optimal estimation technique and a Monte Carlo radiative transfer model (NIMO) as forward model to deal with multiple scattering in the atmosphere. Ground-based MAX-DOAS is highly sensitive to absorbers in the lowest 1-2 km and partial columns retrieved for up to 4 km are discussed.

The following examples will be presented to show-case the use and application of the MAX-DOAS technique for a range of different research topics:

A) The results from car-based MAX-DOAS measurements of halogen oxide (IO and BrO) along the Malaysian (60N - 10N) and New Zealand (36oS - 46oS) coastline during 2009 to 2011 are discussed together with shipboard observations made during a cruise from Singapore to New Zealand (10N - 41oS) and a second cruise off the coast of New Zealand (approx. 41o - 45oS). First results indicate a small but persistent presence of IO. IO is a highly reactive radical affecting tropospheric chemistry; it is a very efficient agent in ozone destruction and is furthermore involved in the formation of fine particles.

BrO, however, is for most of the observation sites and periods clearly below the measurement threshold.

B) Bromine explosion events in the Antarctic have been observed since 1998 near Scott Base on Ross Island (78oS). Bromine availability in the lowermost troposphere in turn affects the deposition of mercury, a bio-toxin, from the atmosphere to the surface. It also depletes ozone very sufficiently and can lead to the total destruction of surface ozone.

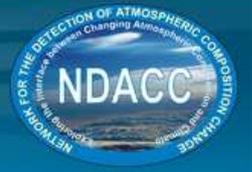
The MAX-DOAS technique allows us to identify these bromine explosion events clearly and to investigate their temporal evolution.

C) MAX-DOAS measurements of NO₂ have been made at several locations such as Auckland (37°S) and Alexandra (45°S) in New Zealand, Singapore (1°N) and Cabauw (52°N) in the Netherlands. The altitude distribution over the lowest 4 km and the diurnal behaviour is investigated.



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Oral Session

5-5 Is There a Hole in the Global OH Shield Over the Tropical Western Pacific Warm Pool?

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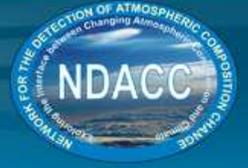
Hundreds of organic species are emitted into the atmosphere mostly from biogenic processes. The rapid breakdown by reactions with OH radicals prevents most of them from reaching the stratosphere. Hence, the omnipresent layer of OH in the troposphere shields the stratosphere from these emissions and is particularly relevant for those species that do not photolyse efficiently. The dominant source of OH in clean tropical air are reactions involving ozone. Hence the OH concentration is closely coupled to ozone abundances. Biogenic halogenated species, biogenic species containing sulphur and perhaps anthropogenic SO₂ emissions play an important role in the stratospheric composition and ozone chemistry. Changes in their abundance and tropospheric breakdown processes provide an ozone climate feedback mechanism. The dominant source of OH in clean tropical air is the photolysis of ozone at wavelengths shorter than about 340nm producing O(1D) radicals followed by the reaction $O(1D) + H_2O \rightarrow OH + OH$. This couples the OH concentration and hence the oxidizing capacity of tropospheric air closely to the concentration of ozone. The area of the Western Pacific warm pool is known to be key for troposphere to stratosphere exchange (e.g. Newell & Gould-Stewart, JAS, 1981; Fueglistaler et al., JGR, 2004). Vertical profiles of tropospheric ozone or OH from that part of the Pacific are not available so far. Measurements from the south east edge of the warm pool area during the Central Equatorial Pacific Experiment in 1993 (Kley et al., Science, 1996) and individual profiles from the station Samoa in the same geographical area (Solomon et al., GRL, 2005) showed extremely low ozone concentrations in the marine boundary layer and at tropopause level but still significant amounts of ozone in most of the free troposphere, where most of the oxidation of biogenic species occurs. We report ship based ozonesonde measurements from the center of the Western Pacific warm pool in October 2009. During a 2500km portion of the ship track between 10S and 15N we found ozone concentrations below the detection limit of the sondes throughout the troposphere. Based on comprehensive CTM modelling we show that the observations suggest the existence of a

pronounced minimum in the tropospheric OH column well correlated with the region where most of the vertical transport of air into the stratosphere occurs. The consequences for the tropospheric lifetimes of chemical species in that key geographical area and implications for the transport of emissions from that area into the stratosphere will be discussed. The results highlight the importance of setting up longer term observational capabilities in the westernmost part of the tropical Pacific.



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Poster Session

5P-1 Towards global routine measurements of high-resolution in-situ NO₂ profiles

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A new instrument, measuring high-resolution NO₂ profiles, has recently been developed. This disposable instrument is light in weight and has low power consumption, which makes it ideal for launch on a weather balloon, similar to the well-known ozone sondes. The measurement principle is based on the chemiluminescent reaction of NO₂ with luminol, where a photo-diode array is used to detect the light signal. Comparison of the NO₂ sonde with a NO_x analyzer with photolytic converter shows that NO₂ variations of 1 ppbv are well captured. The design and calibration of the NO₂ sonde are currently optimised and adapted to allow future serial production. The target precision is 10% or 0.8 ppbv, the vertical resolution is ~5m, determined by the vertical speed of the sonde. The aim is to have the instrument available on the market within a few years. Around twenty flights have been performed in the last two years. The measured NO₂ profiles are compared to model output of Air Quality models. The measured NO₂ profiles regularly show enhancements of NO₂ above the boundary layer with respect to the models. Possible reasons for this have been investigated in detail, using auxiliary meteorological measurements and models. These findings can have an impact on our understanding of the nitrogen cycle, the transport of air pollution, and the production of tropospheric ozone. Regular measurements of high resolution NO₂ profiles are necessary to monitor air pollution above the planetary boundary layer. Also for the validation of tropospheric NO₂ retrievals from satellites, regular measurements of NO₂ profiles are needed. Several research groups aim to use these satellite observations within air quality forecast systems, e.g. in the context of the GMES project MACC. But the uncertainty in the satellite derivations is large, about 30-40% of the tropospheric amount. One of the major sources of uncertainty is the assumed vertical NO₂ profile shape in the troposphere, which is actually quite variable over a day, due to chemical and dynamical processes. Currently, an accurate profile shape can be obtained by in-situ instruments on aircrafts and large balloons. An aircraft can take

several profiles by ascending and descending through the troposphere, and, depending on local regulations, sometimes down to the surface. However, both aircraft and balloon measurements are quite expensive and therefore sparse. What is needed is a global network of stations where routinely NO₂ profiles are measured to verify assumptions used in satellite retrievals and hence improve the air-quality forecasts. The sonde can be used for operational measurements, synchronized with satellite overpasses for operational and long-term satellite validation, in dedicated satellite validation campaigns, and for the calibration of other validation instruments, like MAX-DOAS.



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Poster Session

5P-2 On the use of the MAXDOAS technique for the validation of tropospheric NO₂ column measurements from satellite

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The MultiAxis DOAS (MAXDOAS) technique has been recently developed as a new remote sensing tool for the monitoring of tropospheric pollutants by means of the differential optical absorption spectroscopy (DOAS) method. In complement to zenith-sky DOAS instruments commonly used over the last decade for stratospheric monitoring and satellite validation, e.g. as part of the Network for the Detection of Atmospheric Composition Change (NDACC), MAXDOAS instruments have an increased sensitivity towards atmospheric absorbers present close to the surface. The MAXDOAS are designed to allow the quasi simultaneous observation of the scattered sun light in a range of different line-of-sight (LOS) directions from the horizon to the zenith, and through adequate retrieval process, the near-surface concentration of atmospheric pollutants like NO₂ can be determined, as well as their integrated tropospheric and stratospheric column abundances. Owing to these capabilities, the MAXDOAS technique represents a promising technique for the validation of tropospheric NO₂ column measurements from satellite instruments such as SCIAMACHY, OMI and GOME-2. In this work, we summarize the experience acquired at BIRA-IASB with tropospheric NO₂ validation using the MAXDOAS technique. Results are based on long term measurements performed at the semi-rural site of the Observatoire de Haute Provence (OHP, Southern France) since 2007 but also on more recent high quality observations carried out in the highly polluted Beijing area (China, since June 2008), as well as in the remote high altitude station of the Jungfrauoch (Switzerland, since July 2010). For the latter two sites, the potential of combined exploitation of MAXDOAS, zenith-sky DOAS and direct-sun observations is investigated. The strengths and limitations of the MAXDOAS technique for satellite validation are highlighted and illustrations of achievements recently obtained are given with particular emphasis on the OMI and GOME-2 instruments.



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Poster Session

5P-3 Two Sites in Central Mexico: IR Solar Absorption Measurements

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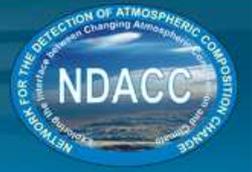
Wolfgang Stremme Universidad Nacional Autonoma de Mexico

Data showing atmospheric transport phenomena, dynamics and emissions of gases from two stations in the Center of Mexico will be presented. Since 2007 solar absorption measurements have been made from the University Campus in Mexico City (19.33, -99.18, 2260 m a.s.l.) with an FTIR spectrometer at moderate spectral resolution. In May 2009 an observatory with sun-tracker and a Bruker Vertex 80 (0.06 max. resolution) spectrometer was installed and is fully operational. A complementary high altitude site in Altzomoni (19.12,-98.65, 4000 m a.s.l.) is located only 60 km to the south east and within a large national park between two volcanoes (one is active). The Universidad Nacional Autonoma de Mexico (UNAM) in collaboration with the Karlsruhe Institut für Technologie (KIT) is setting-up a high resolution instrument (Bruker HR 120) to be installed in Altzomoni before the end of 2011. The stations have distinct characteristics: one is above the boundary layer most of the time and has the scientific objectives of the NDACC; the other submerged in a mega city, focuses on smaller-scale phenomena such as surface emissions, tropospheric ozone-chemistry, boundary layer dynamics and horizontal transport. The opportunity of having two similar experiments close together but with a >1700 m altitude difference opens a wide range of possibilities. Examples supporting this as well as several results of the studies performed so far in both locations will be presented.



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Poster Session

5P-4 Ozone tropospheric and stratospheric trends (1995-2011) at six ground-based FTIR stations (34°S to 79°N)

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Five ground-based stations in Western Europe, from 79°N to 28°N, all part of the Network for the Detection of Atmospheric Composition Change (NDACC), have joined their efforts to homogenize and optimize the retrievals of ozone profiles from FTIR (Fourier transform infrared) solar absorption spectra¹. Using the optimal estimation method, distinct vertical information can be obtained in four layers: ground—10 km, 10—18 km, 18—27 km, and 27—42 km, in addition to total column amounts. A bootstrap resampling method has been implemented to determine annual partial and total column trends². Vigouroux et al. (2008)¹ applied this method to the ozone data and discussed the trends of the total columns and of the partial columns in the above four layers, over the period 1995-2004. The updated trends for the 1995-2009 period has been published in the WMO 2010 report³. Here, we present the updated trends for the 1995-mid-2011 period, for the five European stations but also for a southern hemisphere station, Wollongong (34°S), which recently joined this ozone network.

¹ Vigouroux, C., De Mazière, M., Demoulin, P., Servais, C., Hase, F., Blumenstock, T., Kramer, I., Schneider, M., Mellqvist, J., Strandberg, A., Velasco, V., Notholt, J., Sussmann, R., Stremme, W., Rockmann, A., Gardiner, T., Coleman, M., and Woods, P.: Evaluation of tropospheric and stratospheric ozone trends over Western Europe from ground-based FTIR network observations, ACP, 8, 6865-6886, 2008.

² Gardiner, T., Forbes, A., Woods, P., De Mazière, M., Vigouroux, C., Mahieu, E., Demoulin, P., Velasco, V., Notholt, J., Blumenstock, T., Hase, F., Kramer, I., Sussmann, R., Stremme, W., Mellqvist, J., Strandberg, A., Ellingsen, K., and Gauss, M.: Method for evaluating trends in

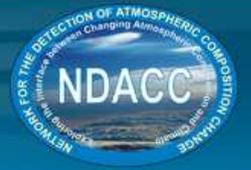
greenhouse gases from ground-based remote FTIR measurements over Europe, *ACP*, 8, 6719-6727, 2008.

3 Douglass, A., and Fioletov, V. (Coordinating Lead Authors), Godin-Beekmann, S., Müller, R., Stolarski, R., Webb, A., Arola, A., Burkholder, J., Burrows, J., Chipperfield, M., Valverde Canossa, J., Cordero, R., David, C., den Outer, P., Diaz, S., Flynn, L., Hegglin, M., Herman, J., Huck, P., Janjai, S., Janosi, I., Krzyscin, J., Liu, Y., Logan, J., McKenzie, R., Matthes, K., Muthama, N. J., Petropavlovskikh, I., Pitts, M., Rex, S. R. M., Salawitch, R., Sinnhuber, B.-M., Staehelin, J., Strahan, S., Tourpali, K., and Vigouroux, C.: Stratospheric Ozone and Surface Ultraviolet Radiation, Chapter 2 in *Scientific Assessment of Ozone Depletion: 2010*, Global Ozone Research and Monitoring Project–Report No. 52, 516 pp., World Meteorological Organization, Geneva, Switzerland, 2011.



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5P-5 Comparison of NDACC Column CH₄ Observations with 3-D Model Results from the Transcom-CH₄ Study

Chris Wilson, Martyn Chipperfield, Manuel Gloor Within the framework of the Transcom-CH₄ 3-D model intercomparison study (Patra et al., 2011), we performed global chemical transport model (CTM) simulations of CH₄ from the late 1980s until the present day. As part of Transcom, results from these simulat

Within the framework of the Transcom-CH₄ 3-D model intercomparison study (Patra et al., 2011), we performed global chemical transport model (CTM) simulations of CH₄ from the late 1980s until the present day. As part of Transcom, results from these simulations were evaluated by comparison with surface in-situ observations. In this work we also compare the model output with column observations from the NDACC (and TCCON) network. The NDACC observations complement the surface data by providing an integrated comparison over the depth of the troposphere.