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Data Set Description:

PI:	Michel Van Roozendael
Instrument:	UV-visible spectrometer
Site(s):	Harestua, 60N, 10E
Measurement Quantities:	O3 and NO2 total columns, stratospheric BrO vertical profiles and columns

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Instrument Description:

In the period from 1994 until 1997, several experimental zenith-sky spectrometers have been operated during winter-spring campaigns. Starting in 1998, a continuous monitoring program was started. Two generations of instruments were successively deployed, respectively from 1998 until 2013, and from 2012 until now.

BIRA zenith-sky spectrometer version 1 (1998-2013):

This system consisted of two commercial grating spectrometers (ORIEL MultiSpec, 12 cm focal length) mounted outdoor inside a protection case thermally regulated and continuously flushed with dry nitrogen. Light from the zenith sky was collected through hemispherical quartz domes and directed towards the entrance of each spectrometer using a depolarising quartz-fibre bundle, which also served as entrance slit (200 micron width). In the actual configuration (see below), spectra were recorded using 1024 pixels cooled diode-array detectors covering respectively the region from 400 to 560 nm with a resolution of 1.2 nm (Spectrometer 1), and the region from 320 to 395 nm with a resolution of 0.7 nm (Spectrometer 2). Two computers (PC) controlled the acquisition of the spectra. Measurements were performed from sunrise to sunset up to a solar zenith angle (SZA) of 96°, with nominal acquisition sequences of 5 min. Exposure time was automatically optimised as a function of the sky light intensity. Dark current data were measured once each day after sunset. The data collection and its transfer to the laboratory in Brussels were obtained through use of a data logger that was interrogated several times a

day by a NILU-based service. Spectra were then transferred onto the Nadir database where they could be ftp-downloaded to Brussels using internet connection.

BIRA zenith-sky spectrometer version 2 (2012 -> now):
This instrument is a custom-build dual-channel system consisting of an outdoor optical head directed to the zenith, and an indoor spectrometric unit. Two optical fibers with rectangular terminations link the optical head with the two spectrometers. The first spectrometer from Newport (MS260i 1/4m), covers the UV region (290-380 nm) with a grating of 1200 grooves/mm blazed at 350 nm, leading to a spectral resolution of 0.4 nm FWHM. A bandpass filter (U340 HOYA) is used to block the visible light at the entrance slit of the spectrometer hence reducing stray-light at UV wavelengths. This spectrometer is equipped with a thermoelectrically-cooled (-50 deg C) back illuminated UV-enhanced CCD detector from Princeton Instruments (model PIXIS 2KBUV) featuring 2048x512 pixels. The second channel uses a Horiba Micro HR spectrometer covering the spectral range from 418 to 550 nm at a resolution of 0.6 nm FWHM. It is also equipped with CCD detector from Princeton Instruments (model Spec-10:100B) cooled to -50 deg C. The whole system is mounted inside a thermally regulated container to minimize thermal stress on mechanical and optical parts. Data acquisition is controlled by computer and data are pushed daily to BIRA by ftp, allowing for daily processing of the NO2, O3 and BrO columns.

Algorithm Description:
NO2, ozone, BrO and OClO vertical and/or slant column densities are retrieved by the method of differential optical absorption spectroscopy. In the following description, we concentrate more particularly on the NDACC products, NO2 and ozone total columns and stratospheric BrO vertical profiles and columns

A/NO2 and ozone total columns

NO2 is analysed in the 425-500 nm spectral window, and ozone from 450 to 548 nm (in the Chappuis bands), using the spectral analysis software suite developed at IASB-BIRA. In brief the method consists in fitting, using non-linear least-squares routines, the log-ratio of two atmospheric spectra to a set of molecular absorption cross-sections measured in the laboratory. The actual algorithm includes a number of advanced refinements, among which capabilities for precise characterisation of both wavelength calibration and spectral resolution of the instrument and calculationof the Ring effect based on Raman scattering modelling. NO2 and O3 vertical columns are derived using the new NDACC recommendations available at http://uv-vis.aeronomie.be/groundbased/.
NO2 slant columns are retrieved using absorption cross-sections measured at 220°K, and converted to vertical columns using climatological air mass factors (AMFs). For ozone columns, look-up tables of AMFs based on the TOMS V8 O3 profile climatology are used (see also Hendrick et al., 2011). Mean twilight vertical columns are obtained by averaging individual measurements between 86 and 91° SZA.

B/Stratospheric BrO vertical profiles and columns

BrO is analyzed in the 336-359 nm wavelength range using the Fleischmann et al. (2004) BrO cross sections and including the spectral signatures of NO2, O3, O4, OClO, and the Ring effect. Stratospheric BrO vertical profiles and corresponding columns are retrieved by applying an OEM-based profiling

technique to sunrise and sunset BrO slant columns (Hendrick et al., 2007 and 2009). BrO slant columns are evaluated using daily reference spectra, the effective residual amount of BrO in the reference spectra being directly fitted by the profiling algorithm, making the retrieval only sensitive to the stratosphere. The forward model includes a stacked box photochemical model in order to reproduce the rapid variation of BrO at twilight and therefore the retrieved profiles can be photochemically corrected in a transparent way to any SZA. HDF data files contain vertical profiles and columns corresponding to 10h45 and 13h15 local time, and to 80°, 85°, and 90°SZA at sunrise and sunset, allowing an effective coverage of the BrO diurnal variation. It should be noted that this BrO data set is best suitable for validation purpose and process studies. For trend analysis, data should be filtered as documented in Hendrick et al., 2007. Users interested in trend analysis are invited to contact F. Hendrick at BIRA-IASB (franch@oma.be).

Expected Precision/Accuracy of Instrument:
A/NO2 and ozone total columns

The error budget of the measurements is obtained by considering error sources affecting the determination of the slant column densities (SCD), the residual amount in the reference spectrum (R), and the air mass factor (AMF). Fitting errors derived from the least-squares analysis typically give very small uncertainties of the order of 3.1014 molec/cm2 for NO2 SCDs and 5 DU for O3 SCDs. However results from intercomparisons exercises (e.g. OHP, 1996) show that state-of-the-art instruments hardly agree to better than a few percents, even using standardised analysis procedures, which indicates that the actual accuracy on SCDs is limited by remaining uncontrolled instrumental and/or analysis factors. More conservatively, and including uncertainties of absorption cross-sections and their temperature dependencies, we quote an uncertainty of the order 5% for NO2 SCDs, and 2% for O3 SCDs. The accuracy on R is mostly limited by the method used to derive the vertical column at the time of the reference spectrum acquisition (we use a Langley-plot approach). The contribution from this error source to the total error budget is generally small (typically 1-2%), although it may become significantly larger for NO2 when very low abundances are to be monitored. In most conditions, the major contribution to the error budget of both NO2 and O3 total columns is the AMF calculation which requires appropriate modelling of the diffuse radiance in the nadir direction. Published studies indicate that the sensitivity of the AMF to stratospheric profiles of pressure, temperature and the constituent itself accounts for an uncertainty of 10 % maximum for NO2, and 4 % for O3. In the case of NO2, much larger errors can be obtained when tropospheric NO2 is produced or transported above the station. Such “pollution events” are usually easily detected by inspection of the SZA dependency of the NO2 SCDs and are filtered out in the analysis process. In summary we estimate the total accuracy on vertical columns to be in most cases better than 15% for NO2, and better than 5 % for O3.

B/Stratospheric BrO vertical profiles and columns

A description of the error budget can be found in Hendrick et al. (2007). In brief, the error analysis is performed by considering the retrieval errors (smoothing error, retrieval noise, and forward model parameter error) and the systematic error related to the uncertainty on the BrO cross sections. Given these error sources, the uncertainty on the stratospheric BrO columns is estimated to about 20%. The BrO HDF data files contain the total systematic and random uncertainties.

Instrument History:

starting date: 1994/01/21
ending date: 1994/04/27
spectrometer 1:
Oriel MultiSpec, grating 600 gr/mm, 320-640 nm, FWHM 1.3 nm
Entrance slit 100 microns, detector EGG-Reticon 1024 (-40 deg C)
spectrometer 2:
Oriel MultiSpec, grating 1200 gr/mm, 335-450 nm, FWHM 0.7 nm
Entrance slit 100 microns, detector EGG-CCD 1024x256 (-60 deg C)

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starting date: 1994/11/09
ending date: 1995/04/27
spectrometer 1:
Oriel MultiSpec, grating 1200 gr/mm, 400-560 nm, FWHM 0.8 nm
Entrance slit 100 microns, detector EGG-Reticon 1024 (-40 deg C)
spectrometer 2:
Oriel MultiSpec, grating 2400 gr/mm, 340-395 nm, FWHM 0.6 nm
Entrance slit 200 microns, detector EGG-CCD 1024x256 (-60 deg C)

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starting date: 1995/08/02
ending date: 1996/05/20
spectrometer 1:
Oriel MultiSpec, grating 1200 gr/mm, 400-560 nm, FWHM 0.8 nm
Entrance slit 100 microns, detector EGG-Reticon 1024 (-40 deg C)
spectrometer 2:
Oriel MultiSpec, grating 2400 gr/mm, 340-405 nm, FWHM 0.6 nm
Entrance slit 200 microns, detector EGG-Reticon 1024 (-40 deg C)

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starting date: 1996/11/25
ending date: 1997/08/23
spectrometer 1:
Oriel MultiSpec grating 600 gr/mm, 300-600 nm, FWHM 1.4 nm
Entrance slit 100 microns, detector EGG-Reticon 1024 (-40 deg C)
spectrometer 2:
Oriel MultiSpec, grating 1200 gr/mm, 320-470 nm, FWHM 0.7 nm
Entrance slit 100 microns, detector EGG-Reticon 1024 (-40 deg C)

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starting date: 1998/01/13
ending date: 2013/05/27
spectrometer 1:
Oriel MultiSpec, grating 1200 gr/mm, 400-560 nm, FWHM 1.2 nm
Entrance slit 200 microns, detector PI-Reticon 1024 (-40 deg C)
spectrometer 2:

Oriel MultiSpec, grating 2400 gr/mm, 320-395 nm, FWHM 0.7 nm
Entrance slit 200 microns, detector PI-Hammamatsu 1024 (-40 deg C)

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starting date: 2012/11/14
ending date: --
spectrometer 1:
Newport/ORIEL MS127, grating 1200 gr/mm, 418-554 nm, FWHM 0.6 nm
Entrance slit 100 microns, Princeton Instruments Spec-10:100B (-50 deg C)
spectrometer 2:
Newport/ORIEL MS260i 1/4m, grating 1200 gr/mm, 290-380 nm, FWHM 0.4 nm
Entrance slit 100 microns, detector Princeton Instruments PIXIS 2KBUV (-50 deg C)