Retrieval of ultraviolet skylight radiances and O_3 slant column densities from balloon-borne limb spectrometer

Fuqi Si (司福祺)¹, Pinhua Xie (谢品华)¹, Cheng Liu (刘 诚)², Jianguo Liu (刘建国)¹, Yujun Zhang (张玉钧)¹, Ke Dou (奚 科)¹, and Wenqing Liu (刘文清)¹

¹Key Laboratory of Environmental Optics and Technology, Anhui Institute of Optics and Fine Mechanics,

Chinese Academy of Sciences, Hefei 230031

²Satellite Group Mainz-Heidelberg, Max Planck Institut für Chemie, Joh.-Joachim-Becher-Weg 27, D-55128 Mainz, Germany

Received August 8, 2007

With a novel light-weight and absolutely calibrated ultraviolet (UV) spectrometer, UV skylight radiances and O_3 slant column densities are measured by balloon-borne limb measurements in Xinjiang area, China. UV skylight radiances measured at the height of 31 km are compared with the results from Modtran in the wavelength range from 290 to 420 nm. O_3 slant column densities are derived from radiance spectra in the Huggins bands (320 – 335 nm) using differential optical absorption spectroscopy method. And the parameter exhibits a good correlation with the same value simulated by radiative transfer model (Tracy). The O_3 profile simultaneously measured by an O_3 sonde is used as input in Tracy calculations. The O_3 sonde is launched on the same balloon.

OCIS codes: 280.1120, 300.1030, 300.6540. doi: 10.3788/COL20080607.0541.

In recent years several space-borne limb scattering instruments were launched to remotely sense the Earth's atmosphere. The instruments included the shuttle ozone limb sounding experiment/limb ozone retrieval experiment (SOLSE/LORE)^[1] flown on the space shuttle mission STS-87 in 1997, optical spectrograph and infrared imaging system (OSIRIS)^[2] on the Swedish-led Odin satellite launched in February 2001, scanning imaging absorption spectrometer for atmospheric chartography (SCIAMACHY)^[3] on ESA's ENVISAT launched in March 2002, as well as NASA's SAGE III on the Russian Meteor-3M satellite.

SCIAMACHY onboard European space agencies environmental research satellite ENVISAT is a grating spectrometer consisting of 8 channels measuring in the ultraviolet (UV), visible, and near infrared wavelength region of $240 - 2380 \text{ nm}^{[4]}$. The satellite operates in a near polar, sun-synchronous orbit at an altitude of 800 km and a local equator crossing time at approximately 10:00 am. The typical ground pixel size of SCIAMACHY is 30 (along track) ×60 (km) (across track), thus being a substantial improvement to the large footprint of the predecessor instrument GOME onboard ERS-2. SCIAMACHY is designed to measure sunlight that is either transmitted, reflected or scattered by the earth atmosphere or surface. For this purpose it has 3 viewing geometries, Nadir, Limb and Occultation.

In this letter, we report the development of UV measurement with an absolutely calibrated UV spectrometer for limb viewing. Combining with these measurements, the monitoring of absolute UV skylight radiance becomes available. And by applying differential optical absorption spectroscopy (DOAS)^[5-8], it is allowed to infer slant column densities for all gases absorbing in UV, such as O₃, and possibe NO₂, BrO, OCIO in future.

Figure 1 shows the schematic of instrument. A telescope with an aperture diameter of 40 mm and 1° field

of view (FOV) captures scattering light. By the optical fiber, the scattering light goes into the entrance slit (50 μ m wide) of a charge-coupled device (CCD) spectrometer (Ocean Optics, HR2000). The spectrometer with the dimension of 148.6 × 104.8 × 45.1 (mm) has a mechanically stable, crossed Czerny-Turner design with fixed grating. The CCD array consists of 2048 elements and has high sensitivity for the wavelength range of 290 – 420 nm. The resulting spectral resolution is 0.4 nm.

During the experiment, the CCD gate duration is changed automatically depending on the last signal value. Between successive gate periods there is a time lag of 7 ms during which each spectral data is sent to a computer through the universal serial bus. At every measured cycle the computer also obtains the orientation information from the electronic compass via RS232. For measuring the absolute radiance, the system was calibrated with a 1000-W tungsten halogen lamp in the laboratory. The spectrometer's temperature is stabilized by heating film surrounding the spectrometer during the whole campaign.

To retrieve column densities from the observed spectra, the well known DOAS is applied. The method is based on Lambert-Beer law



Fig. 1. Instrument for measuring UV radiance and trace gases.

$$I(\lambda) = I_0 e^{-\int \sigma(\lambda) c(x) ds},$$
(1)

where I, I_0 are the detected intensity and the reference intensity, respectively. The absorption cross section $\sigma(\lambda)$ is one characteristic of the individual trace gases. The integrated concentration along the light path $\int c(x) ds$ is called slant column density (SCD) and is the result of the DOAS analysis.

The field campaign took place in Xinjiang area, China, during July 2006. The balloon was hooked on at 10:00 local time. Two hours later, it reached maximum height of 31 km. After two hours floating, the load was separated from the balloon at 14:00. Figure 2 shows UV radiances calculated from Modtran and balloon-borne limb measurements from 290 to 420 nm. The Fraunhofer line is evident, and we use it for wavelength calibration. Good correlation can be found between balloon-borne system and Modtran. Since American standard atmospheric parameters were used for calculating UV radiances rather than those for measurement conditions, the radiances of balloon-borne system and Modtran do not have the same value.

In this campaign, the spectral retrieval of O_3 is performed in the wavelength interval of 320 - 335 nm. The cross section at T = 223 K from Voigt^[9] was used for ozone calculation. The high resolution cross section was convolved with recorded line spectra of a mercury lamp. A spectrum correcting for the Ring effect^[10] is included in the considered fitting routine^[11]. A limb spectrum measured at balloon float altitude is used as Fraunhofer reference spectrum, for which the residual trace gas absorptions are expected to be minimal. Figure 3 shows an example of DOAS fit for a spectrum taken on July 11, 2006 at 10:04. The O_3 SCD for this example retrieval is 1.14×10^{19} mol/cm².

The O_3 SCDs retrieved from the measured spectra were compared with those simulated by radiative transfer model (Tracy). The O_3 profile simultaneously measured on an O_3 sonde was used as input in Tracy calculation. The O_3 sonde was launched on the same balloon. In this study, the Monte Carlo radiative transfer model (Tracy) was used for the O_3 SCD simulation. Tracy model was developed by Heidelberg University in 2003^[12,13]. It solves the radiative transfer equation by backward Monte Carlo simulations in a fully spherical, three-dimensional (3D) and refractive atmosphere. In particular, it may



Fig. 2. UV radiance from 290 to 420 nm. (a) Calculated from Modtran; (b) balloon-borne limb measurements.



Fig. 3. An example of the DOAS evaluation of ozone in the wavelength interval of 320 - 335 nm for a spectrum taken on July 11, 2006 at 10:04. Shown in the figure are the retrieved optical densities of O₃, Ring (thick lines) and the residual structure. The measured (thick line) and the Fraunhofer spectra (thin line) are shown in the top graph.



Fig. 4. Comparison of measured and modeled O₃ SCDs.

simulate the measured SCD of trace gases under consideration, such as temperature (T) and pressure (p), ozone and aerosol profiles. In this simulation, the profiles of T, p and ozone come from real test and aerosol profile from American standard atmosphere. Figure 4 shows the comparison between limb measured and forward modeled O₃ SCDs by using the ozone profile measured from the ozone sonde which works on the same load. The measured and simulated O₃ SCDs are in agreement with each other well.

In summary, we present a novel balloon-borne UV spectrometer for limb UV radiance and trace gas measurements. For our data we find reasonably good agreement between radiances simulated by Modtran and obtained from limb UV balloon measured at float altitude of 31 km. Retrieved O_3 SCDs form radiance spectra in the Huggins bands (320 – 335 nm) using DOAS method, and exhibit good agreement with simulated radiance transfer

model. In the future, more trace gases, such as NO_2 , BrO, and OCIO will be addressed, and profiles of these trace gases can be also calculated with optimal estimation method^[14].

This work was supported by the National High Technology Research and Development Program of China ("863" Program) (No. 2006AA06A303 and 2007AA12Z109) and the Knowledge Innovation Project of Chinese Academy of Sciences (No. kzcx1-yw-06-01). F. Si's e-mail address is sifuqi@aiofm.ac.cn.

References

- R. D. McPeters, S. J. Janz, E. Hilsenrath, T. L. Brown, D. E. Flittner, and D. F. Heath, Geophys. Res. Lett. 27, 2597 (2000).
- C. von Savigny, C. S. Haley, C. E. Sioris, I. C. McDade, E. J. Llewellyn, D. Degenstein, W. F. J. Evans, R. L. Gattinger, E. Griffioen, E. Kyrölä, N. D. Lloyd, J. C. McConnell, C. A. McLinden, G. Mégie, D. P. Murtagh, B. Solheim, and K. Strong, Geophys. Res. Lett. **30**, 1755 (2003).
- H. Bovensmann, J. P. Burrows, M. Buchwitz, J. Frerick, S. Noël, V. V. Rozanov, K. V. Chance, and A. H. P. Goede, J. Atmos. Sci. 56, 127 (1999).
- 4. J. Skupin, S. Noël, M. W. Wuttke, M. Gottwald, H.

Bovensmann, M. Weber, and J. P. Burrows, Adv. Space Res. **35**, 370 (2005).

- B. Zhou, W. Liu, F. Qi, Z. Li, and Y. Cui, Acta Opt. Sin. (in Chinese) 22, 957 (2002).
- R. Kan, W. Liu, Y. Zhang, J. Liu, F. Dong, M. Wang, S. Gao, and D. Chen, Acta Opt. Sin. (in Chinese) 26, 67 (2006).
- F. Si, J. Liu, P. Xie, Y. Zhang, W. Liu, K. Hiroaki, C. Liu, and T. Nobuo, Acta Opt. Sin. (in Chinese) 26, 961 (2006).
- A. Li, P. Xie, W. Liu, J. Liu, T. Yu, K. Dou, and Y. Lin, Acta Opt. Sin. (in Chinese) 27, 1537 (2007).
- 9. S. Voigt, J. Orphal, K. Bogumil, and J. P. Burrows, J. Photochem. Photobiol. A: Chem. 143, 1 (2001).
- 10. J. F. Grainger and J. Ring, Nature 193, 762 (1962).
- S. Solomon and A. L. Schmeltekopf, J. Geophys. Res. 92, 8311 (1987).
- C. von Friedeburg, "Derivation of trace gas information combining differential optical absorption spectroscopy with radiative transfer modelling", PhD. Thesis (Ruperto-Carola University of Heidelberg, 2003) p.59.
- G. Hönninger, C. von Friedeburg, and U. Platt, Atmos. Chem. Phys. 4, 231 (2004).
- C. D. Rodgers, Inverse Methods For Atmospheric Sounding (World Scientific, Singapore, 2000) p.65.