OPEN PATH MEASUREMENT OF ATMOSPHERIC TRANSMISSION SPECTRA IN THE REGION OF 3000–3450 nm USING TUNABLE MID INFRARED LIDAR

S. Veerabuthiran*, A. K. Razdan, Yogesh Sharma

*Laser Science & Technology Centre, Metcalfe House,
Delhi-110054, India; e-mail: s.veerabuthiran@gmail.com*

This paper reports the open path measurement of atmospheric transmission in the spectral region of 3000–3450 nm using a mid infrared tunable lidar in New Delhi (28.7°N, 77.1°E), India. Results on the measurement of atmospheric transmission in New Delhi are reported for first time, to the best of our knowledge. A tunable infrared lidar system has been developed in-house, which transmits laser radiation with the wavelength interval of 1 nm in the above spectral band into the atmosphere. A 200 mm diameter Cassegrain receiver telescope with mercury cadmium telluride (MCT) detector is used to collect the backscattered radiation from the atmosphere. The reflected laser radiation received from the tower located at a distance of 400 m is used to generate the transmission spectrum. Data recorded during the daytime measurements have been considered in this paper. Transmission spectra measured over several days showed a similar pattern, which confirmed the repeatability of the system performance. The measured spectra are also compared with the SkyCalc transmission model generated using the online database. Preliminary analysis indicated that these spectra had good agreement in general with a small shift in wavelength region. We have also analyzed the HITRAN database line-by-line and identified the possible major absorbing molecules in this band. The sources of air pollutants that absorb in this band and also discussed.

Keywords: *atmospheric transmission, lidar, HITRAN, trace gases.*

ТРАССОВЫЕ ИЗМЕРЕНИЯ СПЕКТРОВ АТМОСФЕРНОГО ПРОПУСКАНИЯ В ОБЛАСТИ 3000–3450 нм С ПОМОЩЬЮ ПЕРЕСТРАИВАЕМОГО ЛИДАРА СРЕДНЕГО ИНФРАКРАСНОГО ДИАПАЗОНА

S. Veerabuthiran*, A. K. Razdan, Y. Sharma

УДК 535.33

*Лазерный научно-технический центр, Меткалф Хаус,
Дели 110054, Индия; e-mail: s.veerabuthiran@gmail.com*

(Поступила 22 августа 2018)

Проведено трассовое измерение атмосферного пропускания в области 3000–3450 нм с использованием перестраиваемого лидара среднего ИК диапазона в Нью-Дели (28.7° с.ш., 77.1° в.д.), Индия. Разработанная перестраиваемая ИК лидарная система посылает в атмосферу лазерное излучение в указанном диапазоне длин волн с интервалом 1 нм. Приемный телескоп Кассегрена диаметром 200 мм с детектором на основе кадмиевого ртутного теллурида используется для сбора излучения, обратно рассеянного из атмосферы. Лазерное излучение, отраженное от вышки, расположенной на расстоянии 400 м, использовано для формирования спектра пропускания. Обсуждаются данные, полученные в ходе дневных измерений. Спектры пропускания мало изменяются на протяжении нескольких дней, что свидетельствует о стабильности работы системы. Измеренные спектры сравниваются с моделью пропускания SkyCalc, созданной с помощью онлайн-базы данных. Предварительный анализ показывает, что указанные спектры в целом хорошо согласуются, хотя имеется небольшой сдвиг по длинам волн. Проанализирована база данных HITRAN и определены основные возможные молекулы, поглощающие в указанном диапазоне частот. Обсуждаются источники загрязняющих атмосферу веществ, которые поглощают в рассматриваемом диапазоне частот.

Ключевые слова: *атмосферное пропускание, лидар, HITRAN, следовые газы.*

Introduction. Studies on atmospheric transmission in the infrared spectral band are very important to assess the performance of many electro-optical devices working under various atmospheric conditions. A large number of electro-optical and laser-based systems work in the infrared band. The quality of the transmitted beam and received signal gets altered due to the presence of atmospheric trace gases and background aerosol. The atmospheric trace gases play an important role in many processes, such as the chemistry of troposphere, biosphere-troposphere interaction, etc. [1, 2]. The increase in concentration of trace gases due to industrial activities, movement of transport vehicles, smog, etc. alters the atmospheric transmission properties through the process of scattering and absorption of radiation. They also degrade the quality of air and health in urban cities [3–5]. In order to understand the optical properties of the atmosphere especially in the area of long-path laser propagation and optical remote sensing, a reliable calculation and accurate modeling is required. Therefore, the monitoring of atmospheric trace gases becomes important for the studies concerned with air pollution, transmission, health effects, and climate change. A large number of air pollutants, toxic chemicals, chemical warfare agent, etc. absorb in the mid IR region as it coincides with the fundamental carbon-hydrogen stretch [6]. A tunable infrared differential absorption LIDAR (DIAL) has been extensively used for remote sensing of trace gases and toxic chemicals present in the atmosphere [7–12]. Tunable lidar systems working in the broad spectral band can be also used to study the property of atmospheric transmission. A single ended infrared lidar operating in the 1400–4000 nm spectral band has been used earlier for remote measurement of atmospheric transmission and trace gases [13].

In this paper, we discuss the results on atmospheric transmission properties measured at a tropical station, New Delhi, India, using an optical parametric oscillator (OPO) laser based differential absorption lidar operating in the spectral region 3000–3450 nm. The reflected laser signal measured at different wavelengths is used to generate the atmospheric transmission spectra. We have also carried out detailed studies using HITRAN molecular database to identify the possible molecules that are responsible for absorption in this band. Our results on atmospheric transmission are in good agreement with SkyCalc model in general. Further, we have also discussed the sources of air pollutants, which absorb significantly in this band. Details of the experimental set up and results are discussed.

System description. A tunable mid infrared laser operating in the spectral band of 3000–3450 nm based lidar system has been developed for remote sensing applications. A schematic of the experimental set up is shown Fig. 1. An optical parametric oscillator (OPO) based tunable laser with pulse energy 5 mJ, pulse width 6 ns, and 10 Hz pulse repetition frequency is used as transmitter. A 200 mm optical telescope integrated with thermoelectrically cooled mercury cadmium telluride (MCT) detector and focusing lens is used as receiver to collect the backscattered radiation from the atmosphere. Laser radiation from the source is folded using mirrors FM1 and FM2 before transmitting into the atmosphere as shown Fig. 1. The return signal is digitized using a panel PC based data acquisition system with 30 MSPS sampling rate and 14 bit analog to digital converter. Signal acquisition and processing software is developed in-house in LabView platform.

The transmitter and receiver are mounted on pan and tilt system, which can scan the atmosphere from 0 to 360° in the azimuth and –10 to 30° in the elevation. The present study uses the data recorded by transmitting laser radiation in the horizontal direction under the topographic configuration.

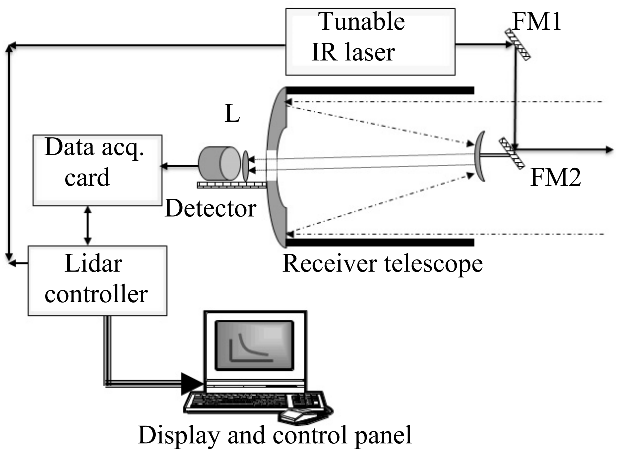

Fig. 1. Schematic of tunable mid infrared lidar system;
FM1: Folding mirror; FM2: Firing mirror; L: Focus lens.

Results and discussion. Firstly, the performance of the system has been calibrated by measuring the absorption signal of the methane gas. The wavelength of the transmitted laser beam was measured using a wavelength meter (M/s. High Finesse, Germany). Methane, one of the greenhouse gases present in the atmosphere with background concentration of about 2–4 ppm, was used for calibration of the system. Methane has the most intense ν_3 stretching vibration band, the central Q branch (3315 nm) which falls within the tunable range of our system [14, 15]. An absorption cell of 50 cm long with CaF_2 window in both sides filled with pure methane was used in calibration testing in the laboratory. The laser radiation in the wavelength band from 3310 to 3326 nm was allowed to pass through the absorption cell and recorded the output signal. Methane gas has weak and strong absorption lines in this band. The experimentally measured signal in this spectral region had been validated with the HITRAN database [11, 16].

The lidar reflected laser signal was collected by transmitting laser radiation in the direction of topographic targets during daytime. The transmitted laser wavelengths and their respective pulse energies were measured independently. The reflected laser signals were normalized with respect to transmitted pulse energies at each wavelength. Signals were averaged over a period of 10 seconds to improve the signal to noise ratio and reduce the noise fluctuation. The reflected signal (peak value) received from the hard targets located at distance of 400 m was used for generating the relative intensity at each wavelength to create the atmospheric transmission spectra in the spectral band from 3000 to 3450 nm. We have transmitted all the wavelengths with an interval of 1nm along the horizontal path up to distance of 400 m (two-way path is 800 m). The transmission spectrum is specific to the experiments carried out at a tropical station, New Delhi. It is the capital of India located in the northern part of India. It is a cosmopolitan city having lots of industrial activities. The lidar laboratory is situated in the central part of the city and near to the outer ring road in which motor vehicle traffic is very high. They emit various air pollutants like methane, carbon dioxide, carbon monoxide, nitrogen dioxide, etc. The recorded data were used for understanding the influence of atmospheric trace gases in the transmission spectra. Data were recorded during the summer months of the season. Figure 2 shows the relative intensity versus the wavelength recorded in spectral band 3000–3450 nm with an interval of 1 nm. The relative intensity measured along the horizontal beam path is considered as the atmospheric transmission. Dark regions of the figure show the wavelengths which have high transmission, whereas transparent region shows the absorption due to background molecules. The wavelength region 3000–3450 nm contains absorption bands of many gases that are present in the atmosphere. The observed meteorological parameters were atmospheric temperature ~35ºC, pressure ~1005 mb, and relative humidity ~26%. The observation period was from 1200 to 1800 h. Based on our data we have identified four regions of interest as mentioned in the Fig. 2. These four regions have strong absorption due to the background molecules. Region 1 has a trough in the region of 3055–3075 nm. Region 2 has two wider troughs in the region 3115–3120 and 3125–3130 nm. Wider troughs are observed in region 3, which extends from 3220 to 3250 nm, in addition to a dip in 3265–3270 nm. Region 4 has a trough in the 3310–3318 and 3330–3338 nm bands. In addition to this, we also observed a trough near the 3015–3055 and 3415–3430 nm bands.

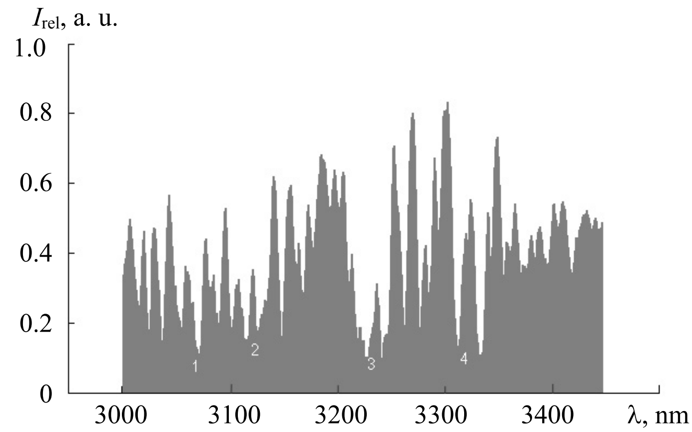

Fig. 2. Relative intensity received for different wavelengths from a target located at distance of 400 m at New Delhi.

We have also simulated the atmospheric transmission spectra in the band of 3000–3450 nm using SkyCalc sky model calculator [17] as shown Fig. 3. The reported simulated transmission values are higher compared to experimentally measured values. However, the pattern over the given wavelength region is similar to that of experimentally measured spectra using lidar. In some region, a minor shift in the wavelengths also observed. Nevertheless, the general agreement of both traces in Figs. 2 and 3 confirms the usefulness of this system for spectroscopic applications. Byer and Endermannt [13] have reported the use of infrared lidar operating in the spectral band 1400–4000 nm for remote measurements of atmospheric trace gases and atmospheric transmission. Return signal measured under cooperative target condition was used to generate the transmission spectra. The reported transmission property in the 3030–3448 nm band is similar to that of our results. Byer and Endermannt have also derived the background concentration of some of the trace gases using their system. Theriault et al. [18] reported results on atmospheric transmission measurements made over a 5.7 km path in the 2800–5500 nm region under a wide range of temperatures and humidities using Fourier interferometric transmissometer (FIT). Our observed results agree well with the reported transmittance in 3000–3350 nm region.

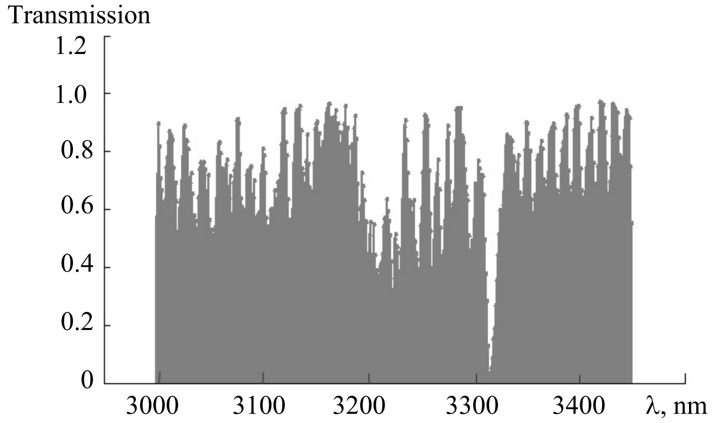

Fig. 3. Simulated atmospheric transmission spectra using SkyCalc model.

Further, we have also analyzed the HITRAN database to identify the possible absorbing molecules in 3000–3450 nm band. The common absorbers of radiation in this band are carbon dioxide (CO_2), ethylene (C_2H_4), nitrogen dioxide (NO_2), nitrous oxide (N_2O), ozone (O_3), methane (CH_4), and water vapor (H_2O). Figure 4 shows the absorption region of the molecules, namely C_2H_4, NO_2, and H_2O, as an example. The water vapor shows the absorption at 3000–3450 nm with the maximum absorption cross section of about 6.1×10^{-25} m^2, whereas NO_2 shows the absorption predominantly at 3400–3450 nm with the maximum absorption cross section of about 2.8×10^{-24} m^2, and C_2H_4 shows dense absorption at 3100–3400 nm with the maximum absorption cross section of about 4.4×10^{-24} m^2. Further analysis, revealed that N_2O molecule had absorption near the 3000–3050 nm region with the maximum absorption cross section of about 2.1×10^{-25} m^2. Ozone has absorption at 3320–3340 nm with the maximum absorption cross section of about 1.5×10^{-25} m^2. This region is totally masked by ethylene with a stronger absorption feature. CO_2 has weaker absorption over the entire band (maximum absorption cross-section ~8.2×10^{-28} m^2). Methane has prominent absorption lines at 3260, 3290, 3300, 3350, and 3420 nm with the maximum absorption cross section of about 1.5×10^{-22} m^2. It is also noticed that methane has dense absorption lines at 3310 to 3330 nm with the maximum absorption cross section of about 8×10^{-23} m^2. Based on the background concentration levels, molecules such as methane and water vapor are found to be the potential absorbers at 3000–3450 nm. Table 1 shows the list of absorbing molecules in this wavelength band.

Various research groups have reported that air polluting chemicals are steadily increasing over Delhi due to industrial activities and transport. Delhi is the largest commercial hub of north India by nature of its geographical location, availability of infrastructure facilities, etc. Based on the nature of activities, a lot of commercial activities take place. The movement of heavy transport vehicles and use of diesel generator sets for power back-up are the major source of polluting gases in these areas. Ghosh et al. [19] reported the total column density and surface level concentration of the greenhouse gases (CO_2, CO, CH_4, H_2O, O_3, and N_2O)

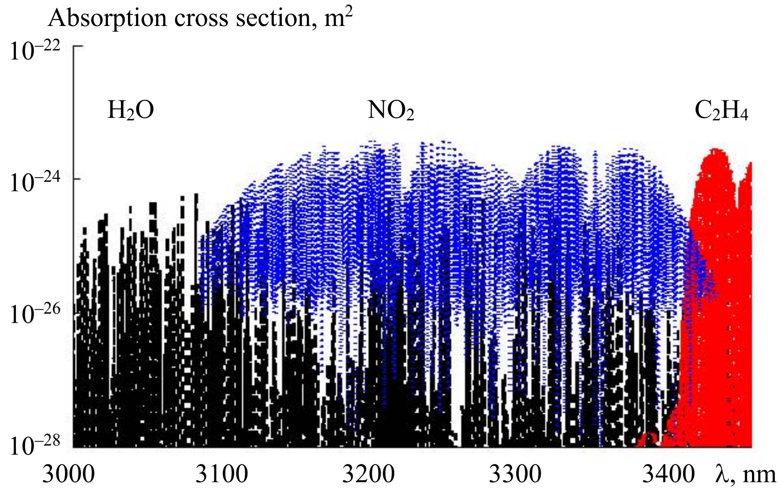

Fig. 4. Absorption lines of H_2O, NO_2, and C_2H_4.

TABLE 1. List of Potential Trace Gases Absorbing in the Wavelength Band 3000–3450 nm

Molecule	Wavelength band, nm	Maximum absorption cross section, m^2
N_2O	3000–3050	2.4×10^{-25}
O_3	3320–3340	1.5×10^{-25}
H_2O	3000–3450	6.1×10^{-25}
NO_2	3400–3450	2.8×10^{-24}
C_2H_4	3100–3400	4.4×10^{-24}
CH_4	3310–3330	8.2×10^{-23}
	3200–3450	1.8×10^{-22}

measured over Delhi since 1992. Typical concentration levels are CO_2 ~355–361 ppm, CO ~2–4 ppm, CH_4 ~1.7–2.7 ppm, O_3 ~30–50 ppb, and H_2O ~1.7–2.7 g/cm^2; these values tend to increase in summer months. However, one of the recent studies by Sahay and Ghosh [20] mentions that the average daytime ambient concentration of CO_2 varied from with an average of 541±27 ppm, and CH_4 concentration varied with an average of 3.2±1.0 ppm. Gurjar et al. [21] reported a comprehensive emission inventory for megacity Delhi, India, for the period 1990-2000. It is reported that SO_2 is largely emitted by thermal power plants, while the transport sector contributes most to NO_x, CO, and non-methane volatile organic compound (NMVOC) emissions. Though the power plants largely emitted CO_2 in the past, the contribution by the transport sector is also increasing steadily. Two decades old trucks nearly 80000 in number flit in and out of the city at night. Many of these, run on a mixture of kerosene and diesel, which release a lot of pollutants. An increase in air pollution level will result in more attenuation of laser radiation in the respective wavelength band. Hence, the transmission property in this wavelength band gets altered due to this. Based on the experimentally measured spectral transmission data, one can also detect and deduce the background concentration of the atmospheric trace gases such as methane, water vapor, NO_x, etc. A large number of lines exist, which makes it possible to select one with the ideal cross section for a specific measurement. The large number of water vapor lines leads to interference problems in the measurements of many other pollutants. Hence, one has to choose the selection of wavelengths very carefully to avoid interference from other molecules. We are also analyzing the selection of line pairs and measurement of ambient concentration of atmospheric trace gases mentioned above using a differential absorption method. The results of this work will be discussed in a future work.

Conclusion. A tunable mid infrared differential absorption lidar system has been designed and developed for remote sensing applications. The reflected laser signal collected over the entire spectral band from topographic targets was used to derive the atmospheric transmission property. We have also compared our results with simulated atmospheric transmission spectra in the 3000–3450 nm band using SkyCalc model. The pattern is similar to that of experimentally measured spectra. The absorption regions are more or less similar with a minor shift in the wavelengths. Nevertheless, the general agreement of both traces confirms the usefulness of this system for spectroscopic applications. Weaker transmission lines in the spectra were indentified and molecules are found, which are absorbing these lines using the HITRAN database. We have also indentified molecules such as methane, water vapor, etc. whose background concentration can be measured in ambient conditions. In the future, we are planning to conduct systematic studies to understand the humidity, temperature, and visibility dependence of atmospheric transmittance in the above band during different seasons.

Acknowledgement. The authors wish to thank the Director of LASTEC for his constant support and encouragement.

REFERENCES

1. IPCC, 2001. Climate change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge, University Press, Cambridge, U.K.
2. V. Ramanathan, L. Callis, R. Cess, J. Hansen, I. Isaksen, W. Kuhn, A. Lacis, F. Luther, J. Mahlman, R. Reck, M. Schlesinger, *Rev. Geophys.*, **25**, 1441–1482 (1987).
3. Marilena Kampa, Elias Castanas, *Environ. Poll.*, **151**, 362–367 (2008).
4. Jun Kagawa, *Toxicology*, **181-182**, 349–353 (2002).
5. Richa Singh, Chhemendra Sharma, *J. Sci. Ind. Res.*, **71**, 155–160 (2012).
6. L. D. Hoffland, R. J. Piffath, J. B. Bouck, *Opt. Eng*., **24**, 982–984 (1985).
7. F. M. D’Amico, R. G. Vanderbeck, R. G. Warren, *Proc. SPIE*, **3855**, 128–33 (1999).
8. H. Karimnezhad, P. Pravin, F. Borna, A. Bavali, *Opt. Laser. Eng.*, **48**, No 4, 491–499 (2010).
9. H. B. Walmsley, S. J. O’Connor, *Pure Appl. Opt.*, **7**, 907–925 (1998).
10. V. Vaicikaukas, V. Kabelka, Z. Kuprionias, M. Kaucikas, *Proc. SPIE*, **5988**, 592880A-1 (2005).
11. S. Veerabuthiran, A. K. Razdan, M. K. Jindal, R. K. Sharma, Vikas Sagar, *Opt. Laser Technol.*, **73**, 1–5 (2015).
12. M. Devi, M. Saikia, A. K. Barbara, *Int. J. Remote Sensing*, **37**, No. 21, 6193–6204 (2011).
13. R. L. Byer, M. Endemannt, *AIAA J.*, **20**, No. 3, 395–403 (1982).
14. V. S. Airapetyan, *J. Appl. Spectrosc.*, **76**, No.2, 268–272 (2009).
15. G. C. Bhar, P. Kumbhakar, A. K. Chaudhary, *Indian J. Pure Appl. Phys.*, **39**, 282–285 (2001).
16. L. S. Rothman, I. E. Gordon, A. Barbe, *J. Quant. Spectr. Radiat. Transf.*, **110**, 533–572 (2009).
17. S. Noll, W. Kausch, M. Barden, A. M. Jones, C. Szyszka, S. Kimeswenger, J. Vinther, *Astron. Astrophys.*, **543**, A92 (2012).
18. J.-M. Theriault, P. L. Roney, F. Reid, *Appl. Opt*., **29**, No. 25, 3654–3666 (1990).
19. A. B. Ghosh, S. Bose, M. C. Sharma, B. S. Gera, *Ind. J. Radio Space Phys.*, **29**, 41–46 (2000).
20. Samraj Sahay, Chirashree Ghosh, *Environ. Monit. Asses.*, **185**, 123–142 (2013).
21. B. R. Gurjar, J. A. Van Aardennea, J. Lelievelda, M. Mohan, *Atm. Environ.*, **38**, 5663–5681 (2004).