Development of LED-DOAS system for observing aerosol optical properties in the lower troposphere

J Aminuddin1*, S Okude2, I Alimuddin3, I Tursilowati4, N Manago5 and H Kuze2

1Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Jenderal Soedirman, Jl. dr. Suparno 61 Purwokerto, Jawa Tengah, 53123, Indonesia.
2Center for Environmental Remote Sensing (CEReS), Chiba University, 1-33 Yayoi-Cho, Inage-Ku, Chiba, 2638522, Japan.
3Department of Geology, Faculty of Engineering, Universitas Hasanuddin, Jl. Perintis Kemerdekaan km 10, Makassar, Sulawesi Selatan, 90245, Indonesia.
4Space Science Center, Indonesian Institute of Aeronautics and Space (LAPAN), Jl. Dr. Djunjunan No.133, Bandung, Jawa Barat, 40173, Indonesia.
5Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, 3-1-1 Yoshinodai, Chuo-Ku, Sagamihara, Kanagawa, 2525210, Japan.

*Corresponding author: jamrud.aminuddin@unsoed.ac.id

Abstract. Aerosols play an important role in the Earth’s radiation budget through the reflection of incoming solar radiation and formation of cloud droplets working as cloud condensation nuclei. The understanding on aerosol optical properties in troposphere, especially their behavior near the ground level, is still insufficient for precise evaluation of their impact. Although a sunphotometer can provide the aerosol optical thickness, its application is limited to daytime under near cloud free conditions. A visibility meter, on the other hand, can give the value of visibility, but the operation wavelength is limited to a single wavelength, e.g. 875 nm. To attain the multi-wavelength observation of aerosol extinction coefficient near the surface level, here we propose the use of a four-color light emitting diode (LED) source emitting at 455, 530, 590, and 625 nm as a light source for the long-path measurement of aerosol extinction. A near-horizontal light path with a round-trip distance of around 630 m has been established inside the campus of Chiba University. A collimated light beam is produced with a 130 mm diameter telescope, and the reflected beam from a retro-reflector is detected using a 200 mm diameter telescope connected to a photodiode. A sinusoidal wave modulation is applied to the LED source, and the resulting modulated signal amplitude is detected and recorded using a digital oscilloscope (Iwatsu, DS-5614A). The results of the recent observation are discussed in conjunction with the concurrent records of a visibility meter (Vaisala, PWD52) and a nephelometer (TSI3563) that can measure the aerosol scattering coefficient.

1. Introduction

The vital effect of pollutant gases and aerosol both directly and indirectly to the atmosphere has been discussed in the context of the earth radiation budget and public health issues [1,2]. Therefore, the precise observation method of aerosol parameters is important for increasing better understanding their real characteristics especially in the lower troposphere. One of significant aerosols parameter is extinction coefficient which is correlated to the attenuation of light due to aerosol particle size floating in atmosphere and hygroscopicity parameter related to relative humidity [3,4]. The extinction coefficient
is linear summation between absorption and scattering [5]. The leading method in monitoring absorption phenomena, the differential optical absorption spectroscopy (DOAS) is a remote sensing technique which has high sensitivity and time resolution in observing atmospheric particulate concurrently [6,7].

Development of instruments and methods in measuring of extinction coefficient have been provided that the DOAS is quite suitable tool in obtaining of particle characteristics in atmosphere near the source [7,8]. Several groups have represented that the DOAS systems is the potential alternatives instruments to monitor air pollutant and aerosols [9–12]. By means specific DOAS design in our group - Center for Environmental Remote Sensing (CEReS) - Chiba University - Japan, monitoring of pollutants and aerosols is established through measurement extinction coefficient over light path length. On that DOAS approach, the measurement is equipped by white flashlight as visible spectral range to detect atmospheric trace gas such as NO2, SO2, and CO2 [13–16]. The DOAS systems used in previous studies applied near ultraviolet and near infrared rays. Their main targets were gases pollutant.

The dependence of the spectrum regions to the particle size as absorber in atmosphere require an accuracy in exploiting the light source wavelength [17–19]. Since our new target is addressed to aerosol in the lower troposphere, we developed a unique DOAS by means of visible rays. Due to the light source modulated by light emitting diode (LED), the instrument is denominated LED-DOAS as novel active ground-based remote sensing. The LED-DOAS operate by transmitting modulated visible rays through collimate system and receiving reflected signal from retro-reflector. This system is improved from DOAS system which utilized and developed by several researchers. The MAX-DOAS proved capable in measuring tropospheric vertical column with target aerosol [19], NO2 [20], and compound between aerosol and trace gases [21]. The long-path measurement developed in Center for Environmental Remote Sensing - Chiba University - Japan evident not only in retrieving for NO2 [13] but also for both NO2 and aerosols [21][21]. Judging for employing retro-reflector [10,16] and LED [11,22,23], we develop a novel design and experiment using 4 wavelength visible rays.

The main purpose in designing of the LED-DOAS is to measure the temporal change of aerosol extinction coefficient (AEC) near the ground level. In this study we developed and carried out experiment in CEReS area which is completed by some ground-based instrument. Besides, to verify the instrument’s performance, the nephelometer, weather monitor, and visibility meter are employed for comparing several results of the new LED-DOAS. Here, we compare the LED-DOAS data to nephelometer raw data with wavelength 550 nm, relative humidity (RH), and the visibility with wavelength of 870 nm.

2. Instruments and Methods

Components of LED-DOAS system are generally consisted of four main parts: signal modulator, transmitter and reflector, signal detector, oscilloscope and personal computer. The LED-DOAS scheme and specification are showed in figure 1 and listed in table 1. This instrument operates by modulating signal and transmitting light beam in four wavelengths (625, 455, 590, and 530 nm) from LED which is controlled automatically by personal computer. The four lasers beam are collimated using Newtonian telescope in 630 meters of round trip from the source to retro-reflector through ambient and back to the photodetector with internal amplifier.

Stability of the LED-based system has been verified by stabilising of photo detector linearity in long time measurement and sun interference. The alignment for detector linearity is checked by adjusting the voltage of modulation signal (recorded in Channel-1 of oscilloscope and personal computer), then compare to the voltage of both reference signal (Channel-2) and LED-signal from retro-reflector traversing real ambient aerosol (Channel-3). This procedure also confirms the correction factor of LED-signal by fitting and analysing several aspects between reference and ambient LED signals. Furthermore, the signal modulation of functional generator is compared to the signal of both original light from small-mirror and ambient contaminated light from retro-reflector. The comparison is done for instrument data in clear and turbid condition using only several days’ data for each sky condition. All alignment processes show the stability of detector and the signal to noise ratio of LED-DOAS system. Linearity of photo detector used in this system using the square signal, 377 Hz of frequency, and modulation vary from 1 to 4 Volt. The sample of linearity process is showed in figure 2.
Figure 1. The scheme of LED LED-based long-path system

| Table 1. Specification of LED-DOAS |
|-----------------------------------|
| **Signal modulator**             |
| Function generator 10 MHz DDS Model 29 |
| Light Emitting Diode LED4D201; Wavelength 625 nm, 590 nm, 530 nm, 455 nm |
| **Transmitter and Reflector**    |
| Collimator                      |
| Newtonian telescope; D 130 mm; f 650 mm; FOV 0.2 mrad |
| Reflector                        |
| Retro-Reflector (RR) & Small Mirror (SM) |
| **Signal Detector**              |
| Receiver                        |
| Cassegrainian telescope; D 2000 mm; Aperture 8”, f/10 |
| Photo Detector                  |
| Model PDA100A-EC Si Amplified Detector |
| Wavelength range 320-1100 nm |
| Maximum output 10 V             |
| Gain 8 x 10 dB steps            |
| Bandwidth Range DC-2.4 MHz      |
| Noise-Equivalent Power          |
| (NEP) x 10 $9.73^{-11}$ - $2.7 \times 10^{-11}$ W/Hz $^{1/2}$ |
| **Oscilloscope and PC**          |
| Oscilloscope                     |
| Digital Oscilloscope 1000 MHz 2 GS/s |
| Personal Computer                |
| LED control and data recorder    |
Figure 2. Linearity of LED-DOAS system derived from channel-2 as the reference signal without aerosol interaction and channel-3 as the LED signal with ambient aerosol interaction.

Sample data of aerosol measurement shown here is from our observation on May 10 to 12, 2017 with reference and LED signals are showed in figures 3 and 4, respectively. We generated the signal using functional generator with frequency 377 Hz and modulation 4 V. The colour changes every 175 second with average measurement 26 second per data. Indeed, we collected 6-7 data for 1 colour. The temporal change of both signals is retrieved under relative humidity, visibility, and temperature as displayed in figure 5. We can see that there are good correlations between LED-signal and ambient patterns. In the case of lower visibility, higher relative humidity, and lower temperature, LED-signal in the lower condition. Here, we can assume that in the lower visibility and higher relative humidity, aerosol particle reduces light beam from retro reflector based on theoretical concept in refs [4,24]. As a result, the LED-signal from retro reflector recorded in Channel-2 is lower.

Figure 3. The reference signal of LED-system derived from channel-2 as the signal without aerosol interaction.
Furthermore, an algorithm for processing LED-signal to attain aerosol parameter in horizontal direction was develop. By governing Bouguer-Lambert-Beer law and Langley extrapolation methods in refs [25–27] and adapting the results into the LED-DOAS system, we develop procedure for calculating initial signal intensity \( I_{CLN} \) in equation (1), aerosol optical thickness \( \tau_a \) in equation (2), and aerosol extinction coefficient \( \alpha_a \) in equation (3):
\[
\ln I_{CLN}(\lambda) = \ln I_{LED}(\lambda) - \alpha_{SCA}(\lambda),
\]

\[
\tau_a(\lambda, t) = \tau_{NET}(\lambda, t) - \tau_m(\lambda, t) = C(\lambda) \frac{\ln I_{CLN}(\lambda, t)}{\ln I_{LED}(\lambda)} - \frac{P(t)}{P_0} \times 0.000877 \lambda^{-4.05},
\]

\[
\alpha_a(\lambda, t) = \frac{\tau_a(\lambda, t)}{6.36} - \alpha_m(\lambda, t)
\]

Where, the \( I_{LED} \) (LED-signal), \( \tau_{NET} \) (total optical thickness), \( \tau_m \) (molecule optical thickness), \( \alpha_m \) (molecule extinction), \( \alpha_{SCA} \) (scattering of nephelometer), \( C \) (correction factor), \( P \) (pressure), \( P_0 \) (atmospheric standard, 1013.25 hPa) are parameters as function of wavelength (\( \lambda \)) and/or time (\( t \)).

3. Results and Discussion

Figure 6 shows the sample of comparison between LED-signal (590 nm) and scattering nephelometer (550 nm). Here, the temporal change of LED-signal inversely proportional to the scattering nephelometer. The lower the LED-signal, the higher the scattering, and vice versa. The scattering is a part of extinction coefficient which correlated to the attenuation of light due to aerosol particle size floating in atmosphere and hygroscopicity parameter related to relative humidity [4,24]. In the condition of higher scattering, we can assume that aerosol loading in the sample of observation is higher. As a result, the signal recorded by LED-DOAS system will lower. This temporal variation verifies the theoretical concept about attenuation of light in atmosphere. Comparison for the three other wavelengths have been done with the same condition.

![Figure 6. Comparison between LED-signal and scattering coefficient of nephelometer.](image-url)
Figure 7 shows the result of fitting between LED-signal (455, 530, 590, and 625 nm) and scattering nephelometer (550 nm). This procedure is Langley plot like developed from ref [26,28]. The Langley plot is the extrapolation method to determine aerosol loading by fitting reference to measured values. In this case, we used scattering of nephelometer data at wavelength 550 nm as reference values and LED-DOAS signal as measured data. The result of this procedure is fitted logarithmic of LED-DOAS signals, ln $I_{LED}(\lambda)$. Their values are: ln $I_{LED}(455) = -3.9104$, ln $I_{LED}(530) = -3.0117$, ln $I_{LED}(590) = -2.9701$, and ln $I_{LED}(625) = -2.6611$. These values are used in calculation of optical thickness and extinction coefficient in the next step of data processing of LED-DOAS system.

Figure 8 represents the results of aerosol optical thickness (AOT) using equation (2). In computational process, we calculated AOT for four bands of LED-DOAS system. The time series variation of AOT in the day of experiment is relatively stable at around 0.4 to 0.5 for the blue light (455 nm). The green (530 nm) and yellow (590 nm) beams show AOT approximately ~ 0.3 and ~ 0.2, respectively. The red band (625 nm) of LED-DOAS signal also stable at approximately 0.1 to 0.2. The whole temporal change of the AOT derived from data processing of LED-DOAS system on the day of experiment are relatively stable, except for at around 6 am on May 12, 2017. Considering with the green band of the scattering nephelometer (550 nm) in figure 6, we can conclude that this processing method in calculating the AOT show the corresponds with the fluctuation of scattering coefficient.

Figures 9 and 10 show the temporal change of extinction coefficient which is also verified to the nephelometer and visibility-meter data directly as scattering and extinction coefficient, respectively. In figure 9, we show the time series variation of extinction coefficient using LED-DOAS signal which is computed using equation (3). Furthermore, in figure 10, we present another temporal change of extinction coefficient as comparison to the result of LED-DOAS data processing based on visibility-meter observation at the wavelength 550 nm. The method in computing visibility-meter data is the Koschmeider equation developed in refs [28,29]. Here, we found extinction coefficient’s values derived from LED-DOAS system almost similar to the result retrieved from visibility-meter as the certified instrument installed in the CEReS-Chiba University.
Figure 8. Temporal change of aerosol optical thickness.

Figure 9. The temporal change of aerosol extinction coefficient.
Figure 10. Comparison of extinction coefficient between visibility-meter and LED-DOAS system.

4. Conclusion
Considering the time series variation of aerosol optical thickness and extinction coefficient obtained from the present instrument, experiment, and approach will be generally useful to estimate the optical properties of ambient aerosols based on the new LED-DOAS system. The capability of uninterrupted estimation of both aerosols optical thickness and extinction coefficient will provide new insight in the source and sink investigation of aerosols as well as in monitoring local environment. Since the LED-DOAS raw signal directly proportional to visibility and temperature but inversely proportional to scattering and relative humidity, we can conclude that the new instrument is working well.

Acknowledgments
One of the authors (Jamrud Aminuddin), the first author would like to thank the Centre for Environmental Remote Sensing (CEReS) Chiba University for supporting the facility in this study and the University of Jenderal Soedirman-Purwokerto for supporting financial to attend the international conference in science (ICOS) at the University of Hasanuddin-Makassar.

References
[1] Seinfeld H and Pandis S. 1998 Atmospheric Chemistry and Physics, from Air Pollution to Climate Change (New York, USA: John Wiley & Sons)
[2] IPCC 2007 Climate Change 2007: The Physical Science Basis vol 446 (Geneva-Switzerland)
[3] Zieger P, Fierz-Schmidhauser R, Gysel M, Ström J, Henne S, Yttri K E, Baltensperger U and Weingartner E 2010 Effects of relative humidity on aerosol light scattering in the Arctic Atmos. Chem. Phys. Atmos. Chem. Phys. 10 3875–90
[4] Titos G, Lyamani H, Cazorla A, Sorribas M, Foyo-Moreno I, Wiedensohler A and Alados-Arboledas L 2014 Study of the relative humidity dependence of aerosol light-scattering in southern Spain Tellus, Ser. B Chem. Phys. Meteorol. 66 1–15
[5] Chung C E 2012 Aerosol Direct Radiative Forcing: A Review Atmospheric Aerosols – Regional Characteristics – Chemistry and Physics models (In Tech Publication) pp 379–94
[6] Platt U 1994 Differential Optical Absorption Spectroscopy (DOAS): Air monitoring by spectroscopic techniques, Chemical Analysis Series ed M. W. Sigrist (New York, USA: Wiley)
[7] Platt U and Stutz J 2008 Differential Optical Absorption Spectroscopy: principles and applications
[8] Kuze H 2012 Multi-Wavelength and Multi-Direction Remote Sensing of Atmospheric Aerosols and Clouds, Remote Sensing - Applications InTech, Available from: ed D B Escalante (In Tech) pp 279–94

[9] Mathew L, Tai W R and Lo J-G 2001 Measurements of Sulfur Dioxide and Formaldehyde in Taipei Using a Differential Optical Absorption Spectrometer J. Air Waste Manage. Assoc. 51 94–101

[10] Lee J S, Kuk B J and Kim Y J 2002 Development of a Differential Optical Absorption Spectroscopy (DOAS) System for the Detection of Atmospheric Trace Gas Species; J. Korean Phys. Soc. 41 693–8

[11] Kern C, Trick S, Rippel B and Ulrich Platt 2006 Applicability of light-emitting diodes as light sources for active differential optical absorption spectroscopy measurements Appl. Opt. 45 2077–88

[12] Lee H, Kim Y J, Jung J, Lee C, Heue K P, Platt U, Hu M and Zhu T 2009 Spatial and temporal variations in NO2 distributions over Beijing, China measured by imaging differential optical absorption spectroscopy J. Environ. Manage. 90 1814–23

[13] Yoshii Y, Kuze H and Takeuchi N 2003 Long-path measurement of atmospheric NO2 with an obstruction flashlight and a charge-coupled-device spectrometer. Appl. Opt. 42 4362–8

[14] Si F, Kuze H, Yoshii Y, Nemoto M, Takeuchi N, Kimura T, Umekawa T, Yoshida T, Hoki T, Tsutsui T and Kawasaki M 2005 Measurement of regional distribution of atmospheric NO2 and aerosol particles with flashlight long-path optical monitoring Atmos. Environ. 39 4959–68

[15] Kuriyama K, Kaba Y, Saitoh H, Manago N, Harayama Y, Osa K, Yamamoto M and Kuze H 2011 Visible and near-infrared differential optical absorption spectroscopy (DOAS) for the measurement of nitrogen dioxide, carbon dioxide and water vapor Int. J. Technol. 2 94–101

[16] Saito H, Manago N, Kuriyama K and Kuze H 2015 Near-infrared open-path measurement of CO2 concentration in the urban atmosphere. Opt. Lett. 40 2568–71

[17] Mischenko M, Travis L and Lacis A 2002 Scattering, absorption, and emission of light by small particles (New York, USA: NASA Goddard Institute for Space Studies)

[18] Somekawa T, Manago N, Kuze H and Fujita M 2011 Differential Optical Absorption Spectroscopy Measurement of CO2 using a Nanosecond White Light Continuum Opt. Lett. 36 4782–4

[19] Irie H, Kanaya Y, Akimoto H, Iwabuchi H, Shimizu a. and Aoki K 2008 Dual-wavelength aerosol vertical profile measurements by MAX-DOAS at Tsukuba, Japan Atmos. Chem. Phys. Discuss. 8 19357–75

[20] Vlemmix T, Hendrick F, Pinardi G, De Smedt I, Fayt C, Hermans C, Pitera A, Wang P, Levelt P and Van Roozendael M 2015 MAX-DOAS observations of aerosols, formaldehyde and nitrogen dioxide in the Beijing area: Comparison of two profile retrieval approaches Atmos. Meas. Tech. 8 941–63

[21] Saito H, Goto Y, Mabuchi Y, Alimuddin I and Bagtasa G 2014 Simultaneous Monitoring of Nitrogen Dioxide and Aerosol Concentrations with Dual Path Differential Optical Absorption Spectroscopy 20–32

[22] Sihler H, Kern C, Pohler D and Platt U 2009 Applying Light-Emitting Diodes with Narrowband Emission Features in Differential Spectroscopy Opt. Lett. 34 3716–8

[23] Thalman R and Volkamer R 2010 Inherent calibration of a blue LED-CE-DOAS instrument to measure iodine oxide, glyoxal, methyl glyoxal, nitrogen dioxide, water vapour and aerosol extinction in open cavity mode Atmos. Meas. Tech. 3 1797–814

[24] Zieger P, Fierz-Schmidhauser R, Weingartner E and Baltensperger U 2013 Effects of relative humidity on aerosol light scattering: Results from different European sites Atmos. Chem. Phys. 13 10609–31

[25] Qiu J 1998 A Method to Determine Atmospheric Aerosol Optical Depth Using Total Direct Solar Radiation J. Atmos. Sci. 55 744–57

[26] Cerqueira J G, Fernandez J H, Hoelzemann J J, Leme N M P and Sousa C T 2014 Langley method applied in study of aerosol optical depth in the Brazilian semiarid region using 500, 670 and
870 nm bands for sun photometer calibration Adv. Sp. Res. 54 1530–43

[27] Bodhaine B A, Wood N B, Dutton E G and Slusser J R 1999 On Rayleigh optical depth calculations J. Atmos. Ocean. Technol. 16 1854–61

[28] Aminuddin J, Okude S, Lagrosas N, Manago N and Kuze H 2018 Real Time Derivation of Atmospheric Aerosol Optical Properties by Concurrent Measurements of Optical and Sampling Instruments Open J. Air Pollut. 07 140–55

[29] Aminuddin J, Manago N, Lagrosas N, Okude S and Kuze H Simultaneous observation of temporal and spatial distribution of atmospheric aerosol by means of slant-path and plan position indicator lidar Proc. SPIE 10779, Lidar Remote Sensing for Environmental Monitoring XVI, 107790F (28 October 2018) p 15