Design of a Low-cost Differential Optical Absorption Spectroscopy Set-up for Simultaneous Monitoring of Atmospheric NO_2 Concentration and Aerosol Optical Thickness

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ABSTRACT

Air quality monitoring in urban areas is indispensable in understanding the environment and how anthropogenic factors contribute to the increasing volume of pollutants in the atmosphere. Differential optical absorption spectroscopy (DOAS) is a useful technique in identifying and quantifying trace amounts of air pollutants over a wide region. In this paper, a low-cost DOAS set-up was developed and was used to measure nitrogen dioxide (NO_2) concentration and aerosol optical thickness (AOT) in the University of the Philippines Diliman campus. The temporal variation of NO_2 concentration from the DOAS measurement was found to agree with the relative NO_2 integrated absorbance from 430-450 nm. A calibration curve was then constructed with calculated sensitivity of 4.467 per mg•mm^{-3} (8.540 per ppm). The concentration range of the low-cost set-up

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is also able to detect unhealthy NO_2 levels in the Philippines. Aerosol optical thickness was then retrieved and showed similar temporal variation with NO_2 throughout the duration of the experiment. The correlation was attributed to the photochemical reaction of NO_2 to NO_3^-, which then forms into aerosol. Average daily AOT at different wavelengths was then determined and was compared to AERONET's data. The results were in agreement with each other and both displayed decreasing AOT at increasing wavelength, which is an expected behavior for a Mie-scattered light due to aerosol. More importantly, proof-of-concept demonstration of low-cost DOAS set-up, capable of measuring trace amounts of NO_2 and AOT, was successfully performed. Results show that the low-cost design can provide an alternative, cheaper and portable atmospheric NO_2 and aerosol measurement technique with reliable sensitivity for environmental monitoring applications.

Keywords: nitrogen dioxide (NO_2), aerosol, differential optical absorption spectroscopy (DOAS), urban air pollution

INTRODUCTION

Monitoring air quality, particularly in urban areas like Metro Manila, is indispensable in understanding the environment and the issues it poses concerning human health (Wendling et al. 2018; Platt and Stutz 2008). Short- and long-term exposure to air pollutants such as particulate matter (aerosol), nitrogen dioxide (NO_2), sulphur dioxide (SO_2), and ground-based ozone (O_3) have shown the strongest evidence of negative health effects in humans (WHO 2018). Ground-based sampling such as electrochemical, photo-ionization, and metal oxide particle sensors may be employed in quantifying trace amounts of these pollutants; however, they only represent local concentration and do not always represent atmospheric condition in a wider region (Hao et al. 2006; European Commission – JRC 2019). Differential optical absorption spectroscopy (DOAS) offers to be a suitable technique that could measure horizontally averaged concentration of air pollutants over a wider region (Platt and Stutz 2008). The principle of DOAS lies in the absorption and scattering of pollutants when broadband light is transmitted through polluted air. Conventional DOAS set-up consists of a light source that is placed a certain distance away from a detector. The spectrum of the transmitted light is then compared to the spectrum of the light source. The changes in the transmitted light are analyzed using the theoretical absorption and scattering cross sections of air species and is used to calculate the column density of the pollutant (Platt and Stutz 2008).

By utilizing the near-UV region in the visible spectrum, it is possible to measure trace amounts of nitrogen dioxide (NO_2) and aerosol using the DOAS technique (Yoshii et al. 2003; Kuze et al. 2012; Saito et al. 2014). NO_2 is naturally emitted to the atmosphere via forest fires, lightning storms and emissions from soil. However, it is now the anthropogenic sources of NO_2 that contribute greatly to the rapidly increasing NO_2 emissions throughout the world (Platt and Stutz 2008). These anthropogenic sources include fossil fuel combustion from power plants, industries, and automobiles (European Commission – JRC EDGAR 2018), as well as soil emission from agricultural lands (Almaraz et al. 2018).

Compact DOAS systems available in the market are already routinely installed to monitor smog-related chemicals and agro-industrial related emissions, as well as conduct upper atmosphere monitoring of trace gases and pollutants (DENR-EMB 2016; US EPA 2018). However, these compact DOAS systems are not without their high production, installation, and maintenance cost, making air quality management difficult for developing countries like the Philippines. Media reports indicate that a DOAS system could cost PHP 300,000.00 in the Philippines with some even reaching PHP 10,000,000.00 (Alunan 2016; Gamil 2017). A recent report used NO_2 and SO_2 emissions to indicate air quality index per country and revealed that highly industrialized countries tend to curb their emissions while those of developing countries are expected to increase (Bouwman et al. 2002; Wendling et al. 2018). Thus, efforts have been made to develop low-cost DOAS systems (Yoshii et al. 2003; Kuze et al. 2012; Saito et al. 2014) that could potentially be deployed and easily acquired by concerned agencies for their respective air pollution monitoring programs. In this study, the authors developed a proof-of-concept demonstration of a low-cost DOAS set-up shown to be capable of detecting trace amounts of atmospheric NO_2 and aerosol in University of the Philippines Diliman (UP Diliman) campus.

MATERIALS AND METHODS

Theoretical Background

When broadband light of intensity $I_0(\lambda)$ passes through a volume of absorbers such as polluted air, the detected light at the end of the light path experiences extinction due to molecular absorption of trace gases and scattering by air particles present in the atmosphere (see Figure 1).

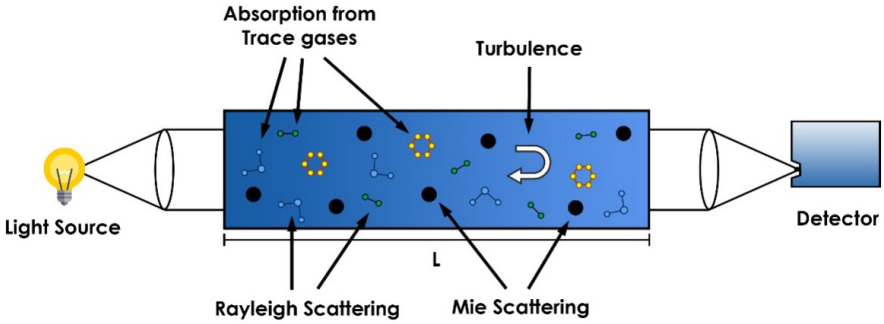

Figure 1. Schematic diagram of light absorption and scattering through a polluted medium.

The absorption and scattering of light follows the Lambert-Beer's law, and thus the detected light $I(\lambda)$ can be expressed as:

$$I(\lambda) = k(\lambda) I_0(\lambda) T_R(\lambda) T_a(\lambda) T_{gas}(\lambda) \qquad (1)$$

Where λ is the wavelength, $k(\lambda)$ is the instrument efficiency, $T_R(\lambda) = \exp[-\tau_R(\lambda)]$ is the transmittance due to Rayleigh scattering with τ_R as the optical thickness due to air molecules, $T_a(\lambda) = \exp[-\tau_a(\lambda)]$ is the transmittance due to Mie scattering with τ_a as the optical thickness due to aerosol particles, and $T_{gas}(\lambda)$ is the transmittance due to the absorbing gas which is also expressed as:

$$T_{gas}(\lambda) = \exp[-N\sigma_{gas}(\lambda)L]. \qquad (2)$$

Here, N and $\sigma(\lambda)$ are the concentration and absorption cross section of the absorbing species, respectively, over the optical distance L. The absorption cross section of NO_2 obtained from the HITRAN2012 database is shown in Figure 2 (Rothman et al. 2013). The cross section is characterized by having a broad feature centered around a 380 nm wavelength superimposed by rapidly varying spikes. NO_2 mostly absorbs in the blue region of the visible spectrum (400-450 nm), and thus an appropriate light source with high emission at this wavelength range is required. There is also no expected significant absorption interference with other gases within the specified range.

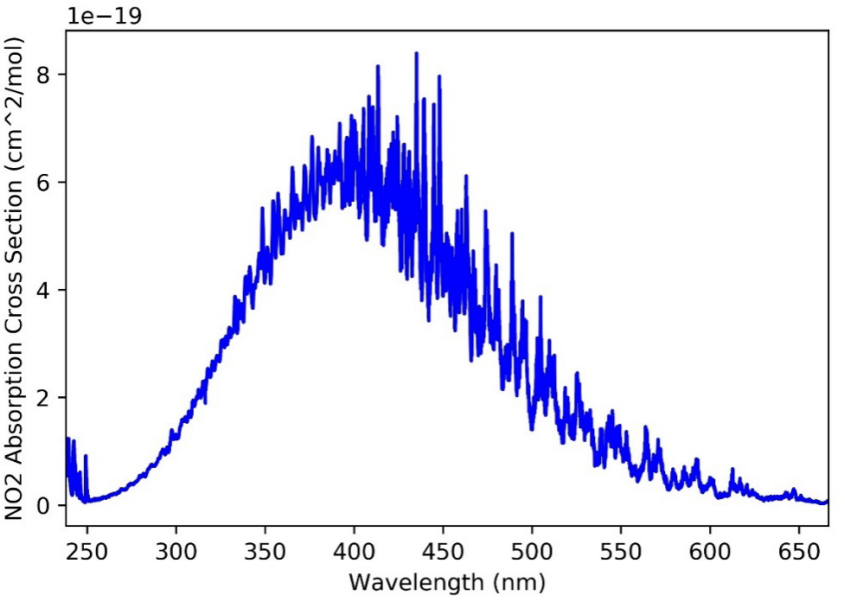

Figure 2. Absorption cross section of NO_2 from 250-650 nm. Data obtained from the HITRAN2012 database (Rothman et al. 2013).

Experimental Set-up

The low-cost DOAS set-up is composed of two astronomical Newtonian telescopes (Celestron Omni XLT150, FL = 750 mm, D = 150 mm) placed at a certain distance away from each other. The first telescope is used to propagate the light coming from a commercially available blue LED while the other is used to focus the transmitted light towards a detector. The schematic diagram of the experimental set-up is shown in Figure 3. The emitter and detector telescopes were placed at the rooftops of the National Institute of Physics building and the Electrical and Electronics Engineering Institute building, respectively, which are 547 m apart. Figure 4 shows the location of the DOAS path while Figure 5 shows the actual DOAS set-up used in this experiment.

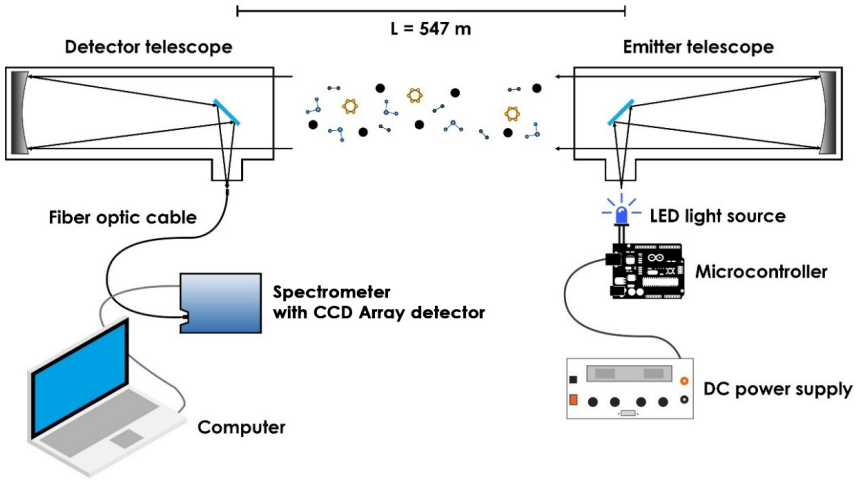

Figure 3. Schematic diagram of the DOAS experimental set-up.

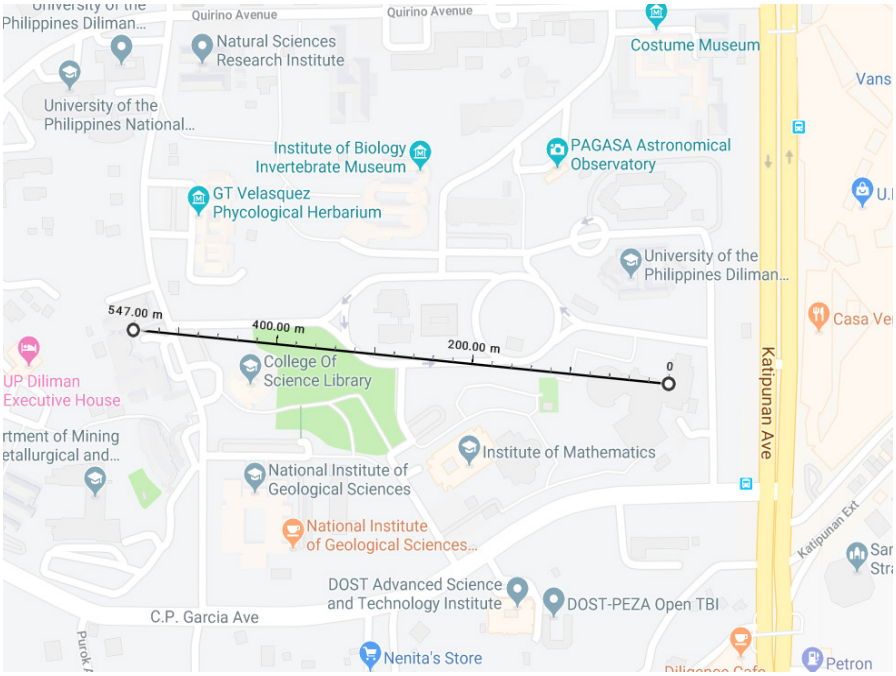

Figure 4. DOAS observation path from the National Institute of Physics (NIP) to Electronics and Electrical Engineering Institute (EEEI), University of the Philippines, Diliman (Google Maps 2019)

Figure 5. The actual DOAS set-up used in the experiment which shows the detector telescope set-up in EEEI (top), and the emitter telescope in NIP as seen through the detector telescope's eyepiece (bottom).

The spectrum of the LED light source used is shown in Figure 6 and shows appreciable emission intensity at around 430-450 nm which makes it suitable for the detection of NO_2 whose absorption cross section is also plotted in the figure. The detector that was used was a fiber-fed CCD spectrometer (Ocean Optics USB2000+) with 2048 elements and is sensitive in the wavelength range of 395-700 nm, thus having a spectral resolution of around 0.127 nm. This enables the differentiation of spectral features within the target spectral region of 430–450 nm. The emission of the light source was set to flash every 2.5 minutes and the spectra with and without the light source (on and off events) were measured. The on and off measurements were used to discriminate background radiation due to sunlight and turbulence.

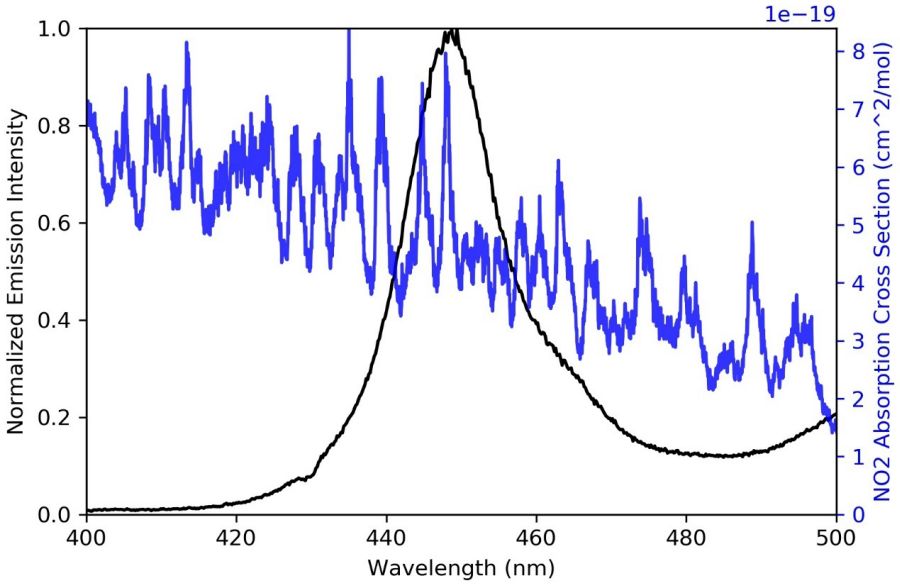

Figure 6. Spectrum of the white LED measured near the light source (left axis) superimposed with NO_2's theoretical absorption cross section obtained from the HITRAN2008 database (right axis).

The measured on and off events were averaged for every 10-minute time interval to represent the on and off events at the particular time interval. The on and off intensities ($I_{on}(\lambda)$ and $I_{off}(\lambda)$) were numerically discriminated by calculating the integrated intensity over the range of 430-450 nm. The background subtracted spectrum was then normalized with respect to the emission spectrum of the LED $I_{ref}(\lambda)$ measured by the same DOAS set-up at a near distance. The absorption optical density D was then calculated as follows:

$$D = \ln\left[\frac{I_{ref}(\lambda)}{I_{on}(\lambda) - I_{off}(\lambda)}\right] = N\sigma_{gas}(\lambda)\, L + \tau_R(\lambda) + \tau_a(\lambda) \qquad (3)$$

The quantity D was used in two ways. First, it was used to determine the relative absorption intensity per time interval by calculating the integrated intensity of the spectrum. The integrated intensity is directly proportional to the amount of absorption due to molecular NO_2 in the atmosphere. Second, the quantity was subjected to standard DOAS analysis procedure discussed below to derive the column density of NO_2.

The spectrum of the optical density *D* is composed of fast- and slowly-varying features that are signatures of NO_2 molecular absorption and aerosol scattering, respectively (Platt and Stutz 2008). The fast-varying features were studied by comparing *D* to the theoretical absorption cross section of NO_2 via linear regression analysis. The slope of the best-fit line determines the concentration in ppm given the optical path distance *L* and under the assumption that 1 ppm of NO_2 in air has a molecular concentration of $2.503 \times mol \bullet cm^{-3}$ and mass concentration of $1.912\ mg \bullet m^{-3}$ (Platt and Stutz 2008). The intercept takes care of the slowly-varying features. After the calculation of NO_2 concentration from the regression analysis, it is then possible to retrieve aerosol optical thickness (AOT) from the same spectra as described below. Thus, simultaneous monitoring of these two pollutants can be performed within the spectral range of interest in this study.

Rearranging equation 3, the wavelength dependence of the optical thickness due to aerosol scattering may be expressed as:

$$\tau_a(\lambda) = D - N\sigma_{gas}(\lambda)\, L - \tau_m . \qquad (4)$$

Here, the concentration *N* calculated previously, $\sigma_{gas}(\lambda)$, and *L*, are substituted in equation 4. The quantity due to Rayleigh scattering was then omitted due to its very low contribution within the chosen wavelength range (Fröhlich and Shaw 1980). Aerosol optical scattering takes the form (Xue et al. 2001; Fuqi et al. 2005):

$$\tau_a(\lambda) = B\left(\frac{\lambda}{\lambda_0}\right)^{-A} \qquad (5)$$

where B (also called turbidity coefficient) is the AOT at λ_0, which is proportional to the column density of aerosol, and A is the Angstrom coefficient, which is related to the wavelength dependence of AOT. The values of A and B are then obtained by performing a least-squares fit to the combined equations 4 and 5 for different center wavelength λ_0. The chosen values were 440 nm, 500 nm, and 675 nm, which are typically used in conventional AOT retrieval instruments.

RESULTS AND DISCUSSION

Figure 7 shows the resulting temporal variation of NO_2 concentration in air observed on November 16, 2018. The left axis shows the concentration in terms of ppm as obtained from the standard DOAS analysis procedure while the right axis shows the relative absorption intensity due to NO_2. The two independent analysis procedures

agree fairly well with each other in terms of the temporal profile of the pollutant with correlation coefficient of 0.998 and slope equal to 4.467 relative units per $mg \bullet m^{-3}$ (or 8.540 relative units per ppm) as shown in Figure 8. Since the integrated intensity is directly proportional to absorption due to NO_2 concentration in the air, therefore the correlation provides a qualitative validation of the measured values via DOAS analysis.

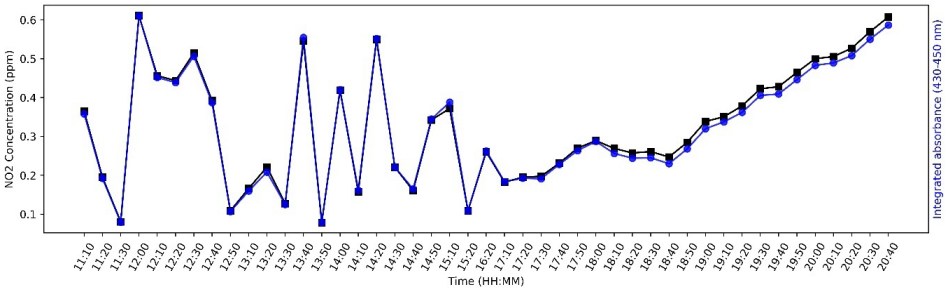

Figure 7. Temporal variation of NO_2 concentration as measured by the DOAS analysis (left axis) and by its relative absorption (right axis) on November 16, 2018.

As shown in Figure 7, NO_2 concentration was found to be fluctuating from 0.1 to 0.6 ppm from 11:10 until 16:20. It then started to increase steadily at 17:10 until the end of the experiment at 20:40. Vehicular NO_2 emissions are attributed to be the primary cause of the monotonic increase starting at 17:10 since the duration also coincided with the evening rush hour. No nearby industries can be found that may emit high concentrations of NO_2. Furthermore, the range of the calculated ppm values in this experiment is within the scale of measurements conducted by other researchers in an urban setting ranging from 0.01 to 0.05 ppm (Yoshii et al. 2003; Kuze et al. 2012; Saito et al. 2014). The average NO_2 concentration throughout the experiment was 0.33 ± 0.15 ppm, which is below NO_2's unhealthy levels (0.65 – 1.24 ppm) as defined in the implementing rules and regulations (IRR) for RA 8749 or the "Philippine Clean Air Act of 1999" (DENR-EMB 2016).

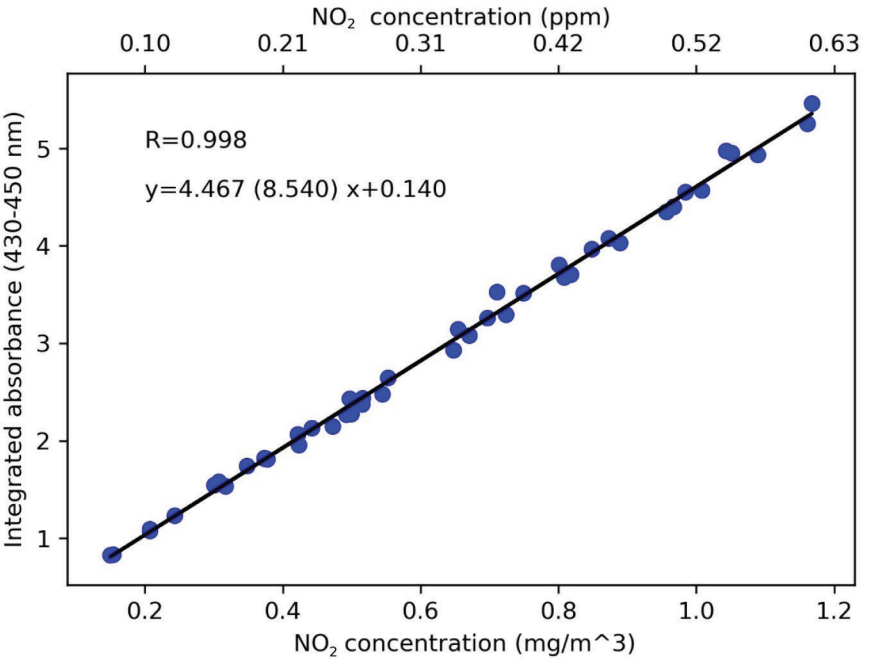

Figure 8. DOAS instrument calibration curve.

Given the obtained NO_2 concentration values, AOT was then retrieved from the measured optical density D following equation 4. The temporal variation of the AOT at 440 nm shown in Figure 9 mimics that of the calculated NO_2 concentration, having a correlation coefficient of 0.969 (shown in Figure 10). The correlation between NO_2 concentration and AOT at 420 nm has also been observed in other studies in the northern hemisphere (Xue et al. 2001; Veefkind et al. 2011) with correlation coefficient of 0.98 and similar spectral shift of AOT for longer wavelengths. It was suggested that the correlation between NO_2 concentration and AOT at 420 is due to the photochemical reaction of NO_2 to NO_3^-, which in turn adds to the aerosol concentration on the ground (Kaneyasu et al. 1994; Xue et al. 2001).

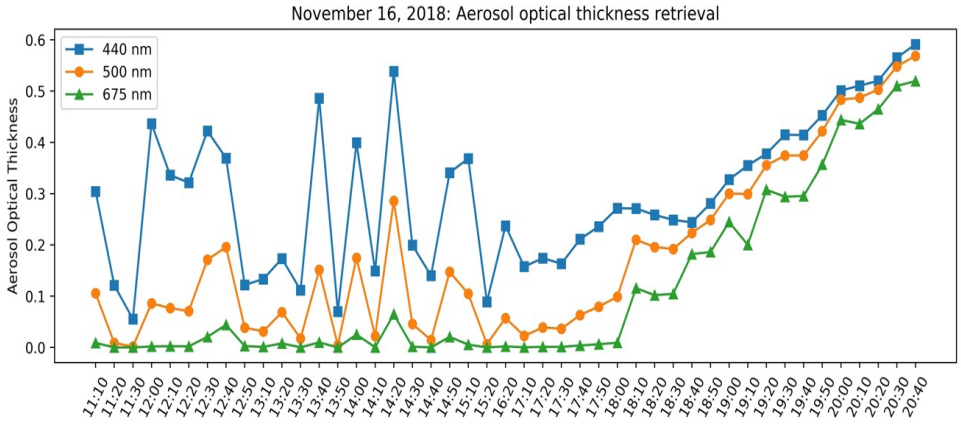

Figure 9. Temporal variation of AOT at 440 nm, 500 nm, and 675 nm on November 16, 2018.

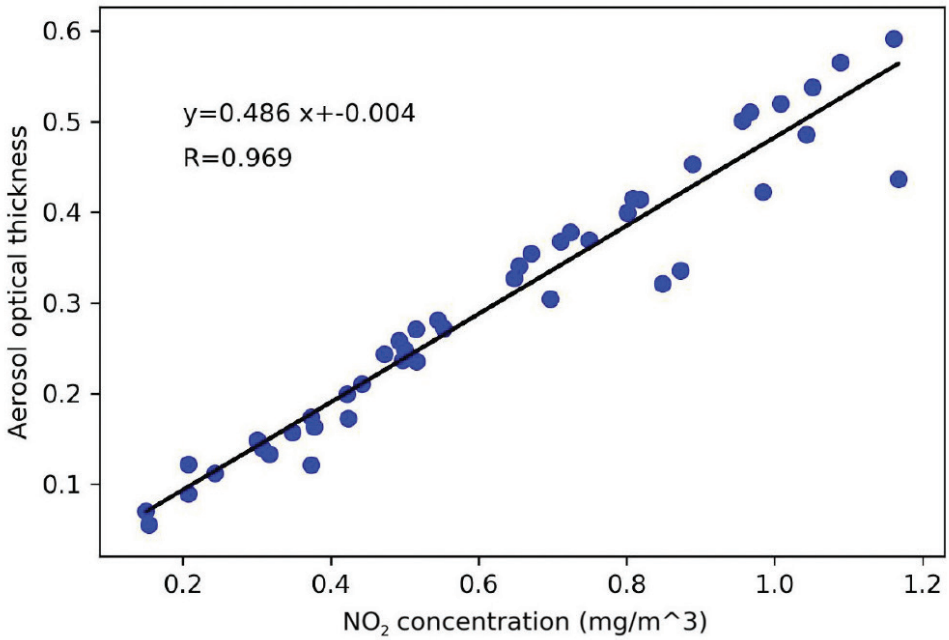

Figure 10. Result of the linear regression between NO_2 concentration and aerosol optical thickness (AOT).

The result from AOT retrieval was then compared to the data obtained by the AERONET (AErosolROboticNETwork) project, a federation of ground-based remote sensing aerosol networks (NASA 2019). The daily average AOT from AERONET ground station at the Manila Observatory was found to be comparable to the average AOT obtained from the DOAS measurement. The comparison is shown in Figure 11.

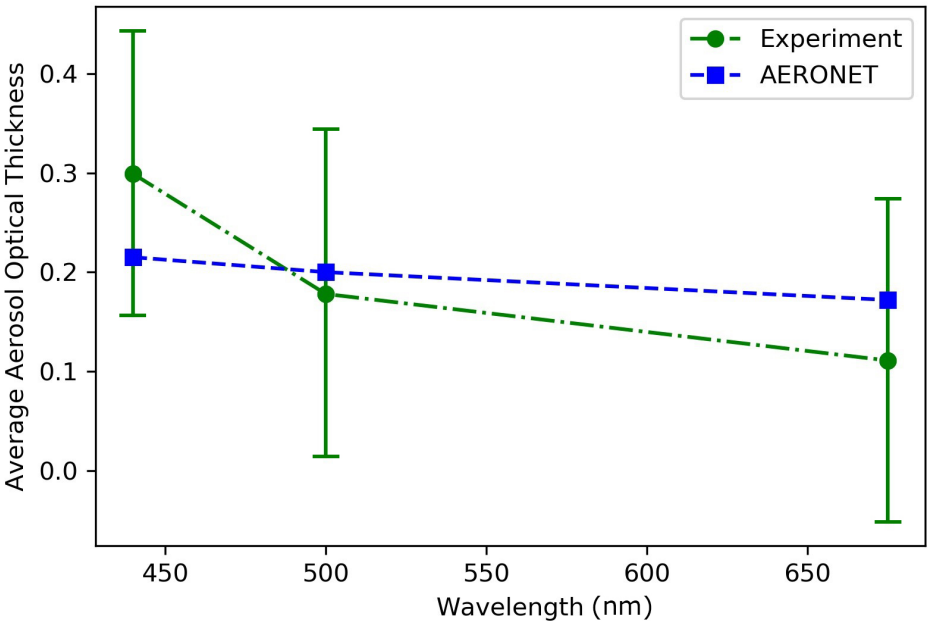

Figure 11. Comparison between the average AOT on November 16, 2018 obtained from the DOAS measurement and AERONET.

The difference between the values may be due to the different times of monitoring conducted by the current DOAS measurement and by AERONET. In this study, the measurement was done from 11:10–20:40, which, as seen in Figure 9, shows great temporal variation thus the high margin of error in Figure 11. Meanwhile, AERONET's measurement was done from 22:00 (of the previous day)–01:00 and 03:00–07:00 (NASA 2019). However, both showed a general decrease in the average AOT for increasing wavelength which is a typical behavior of Mie-scattered light due to aerosol or suspended particulate matter.

The above results show the successful proof-of-concept demonstration of a low-cost DOAS set-up that is capable of measuring trace amounts of atmospheric NO_2 and AOT within the UP Diliman campus. The current design used components whose total cost is estimated to be only around PHP 300,000.00-400,000.00, while compact DOAS systems used by the Department of Environment and Natural Resources are millions of pesos (Alunan 2016; Gamil 2017). The sensitivity of the low-cost DOAS design is also within the scale of NO_2's unhealthy levels in the Philippines. Finally, it must also be noted that the current design can be made portable, allowing for easy deployment in the field unlike most DOAS systems which are fixed.

Summary

A relatively low cost DOAS set-up for the monitoring of atmospheric NO_2 concentration and aerosol optical thickness was successfully designed and demonstrated. The calculated ppm values were found to be in agreement with the integrated NO_2 absorbance from 430-450 nm with correlation coefficient of 0.998. The primary source of NO_2 measured in this study was attributed to vehicular emission. The concentration range (0.1-0.6 ppm) obtained in this experiment was also comparable with the previous studies conducted using similar DOAS setup (Yoshii et al. 2003; Kuze et al. 2012; Saito et al. 2014), and is able to detect NO_2's unhealthy levels in the Philippines.

Aerosol optical thickness (AOT) was then retrieved from the measured optical density using the calculated NO_2 concentration. The temporal variation of NO_2 and AOT exhibited high correlation that was attributed to the photochemical reaction of NO_2 to NO_3^-. The daily average AOT from the DOAS measurement was compared to the data obtained by AERONET, and also appeared to be correlated. The difference was attributed to the different times of measurement conducted by the two experimental techniques.

Results imply that the design can provide a cheaper and portable atmospheric NO_2 and aerosol measurement technique with reliable sensitivity for environmental monitoring applications.

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