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Validation of NO_2 Measurements Using a Three-Wavelength OPO Differential Absorption Lidar

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Abstract: Nitrogen dioxide (NO_2) is very important in the tropospheric chemistry. Short-term NO_2 exposure, ranging from 30 minutes to 24 hours, can cause the exacerbation of asthma symptoms, in some cases resulting in hospitalization. Long-term NO_2 exposure is likely to have a causal relationship with respiratory effects, based on evidence for the development of asthma. Hampton University (HU) built a Differential Absorption Lidar (DIAL) based on an Optical Parametric Oscillator (OPO) laser which can obtain more accurate measurements of NO_2 using three-wavelengths elastic signals. NO_2 concentration profiles were obtained using the HU three-wavelength OPO DIAL and these results were compared with simulated data from WRF-Chem model and NO_2 profile from Pandora to assess their accuracy.

1. Introduction

The conventional Differential Absorption Lidar (DIAL) can measure nitrogen dioxide (NO_2) using two wavelengths. However, high concentrations of aerosol within the planetary boundary layer (PBL) can cause significant retrieval errors using only a two-wavelength DIAL technique to measure NO_2. HU developed a three-wavelength DIAL based on an OPO laser to obtain more accurate measurements of NO_2 in PBL [Su, et al]. HU three-wavelength DIAL uses two rules to obtain the optimum choice of the three wavelengths for lidar system to increase NO_2 absorption and reduce aerosol interference. Simulated results show that uncertainties of aerosol extinction using the three-wavelength DIAL technique are reduced to less than 2% of using the two-wavelength DIAL technique. Moreover, the retrieval uncertainty analysis indicates that the three-wavelength DIAL technique can reduce more fluctuation caused by aerosol backscattering than two-wavelength DIAL technique. This study presents NO_2 profiles which were obtained using the HU (Hampton University) three-wavelength OPO DIAL. To assess the accuracy of the HU lidar NO_2 profiles, we compared the NO_2 profiles to WRF-Chem model's and Pandora's. This comparison suggests that the NO_2 profiles retrieved with the three-wavelength DIAL technique agree well with profiles from WRF-Chem model and Pandora.

2. Method

A three-wavelength DIAL technique was proposed with $\lambda_1 < \lambda_2 < \lambda_3$. NO_2 density retrieval equation can be expressed as:

$$N_N(Z) = \frac{\frac{1}{2}\times\frac{d}{dz}\left[ln\frac{X(\lambda_1,Z)X(\lambda_3,Z)}{X(\lambda_2,Z)^2}\right]-AED(z)-MED(z)-OAD(z)-B(z)}{\Delta\sigma_N} \qquad (1)$$

$$\Delta\sigma_N = 2\sigma_N(\lambda_2) - \sigma_N(\lambda_1) - \sigma_N(\lambda_3) \qquad (2)$$

$$B(z) = \frac{1}{2}\frac{d}{dz}\left[ln\frac{\left[\left(\frac{\lambda_3}{\lambda_2}\right)^{-4}\beta_m(Z)+\left(\frac{\lambda_3}{\lambda_2}\right)^{-e}\beta_a(Z)\right]\left[\left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\beta_m(Z)+\left(\frac{\lambda_1}{\lambda_2}\right)^{-e}\beta_a(Z)\right]}{[\beta_m(Z)+\beta_a(Z)]^2}\right] \qquad (3)$$

$$AED(z) = K\alpha_a(Z)$$

$$K = 2 - \left(\frac{\lambda_1}{\lambda_2}\right)^{-e} - \left(\frac{\lambda_3}{\lambda_2}\right)^{-e} \qquad (4)$$

$$MED(z) = \left[2 - \left(\frac{\lambda_1}{\lambda_2}\right)^{-4} - \left(\frac{\lambda_3}{\lambda_2}\right)^{-4}\right]\alpha_m(Z) \qquad (5)$$

$$OAD(z) = 2O_{abs}(\lambda_2,z) - O_{abs}(\lambda_1,z) - O_{abs}(\lambda_3,z) \qquad (6)$$

where X is the lidar signal; the subscripts *a* and *m* represent aerosol, and molecule, respectively; σ_N is the absorption cross section for the gas of interest; N_N is the molecular density of the gas of interest; O_{abs} is absorption of gases other than the gas of interest; *z* is the altitude; β_m and β_a are backscatter from molecules and aerosols for the wavelength of λ_2 ; α_m and α_a are the extinction of molecules and aerosols for the wavelength of λ_2; *e* is the aerosol Ångström exponent and assumed to be equal for the three wavelengths because the three wavelengths are very close; AED, MED, OAD and B are the correction terms of aerosol extinction, molecular extinction, absorption of gases other than the gas of interest and backscattering, respectively. Because the atmospheric molecular density is relatively stable, MED can be corrected using a numerical model or local real-time radiosonde data. OAD can be removed by choosing appropriate wavelengths. However, aerosol is variable especially in PBL. For correction of AED and B, we need accurate aerosol measurements. However, accurate aerosol measurements are not easily to be obtained. From the above NO_2 retrieval relative equation, all of correction terms are related to the three wavelengths, so how to choose the three wavelengths is very critical to reduce correction terms and improve the accuracy of NO_2 retrievals. We designed two rules to obtain the optimum choice for the three wavelengths:

a. The chosen three wavelengths increase differences of the NO_2 absorption cross section ($\Delta\sigma_N$) to improve NO_2 retrieval.

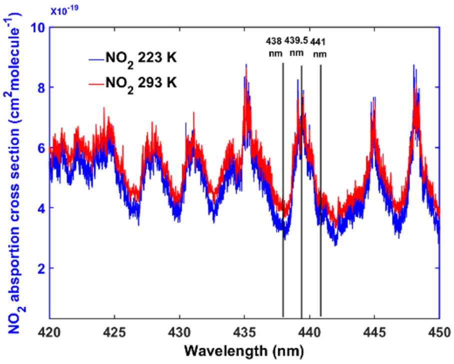


Fig.2 NO_2 strong absorption cross section

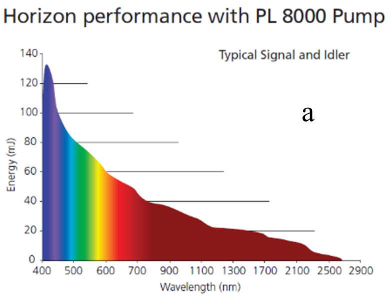
According to Eq. (2), the more $\Delta\sigma_N$ is, the less all of correction terms are. The chosen three wavelengths should help to increase $\Delta\sigma_N$. According to characteristics of the NO_2 absorption spectrum showed in Fig. 2, a bumping absorption method ($\sigma_N(\lambda_1)<\sigma_N(\lambda_2)\&\sigma_N(\lambda_3)<\sigma_N(\lambda_2)$) is designed to choose the three wavelengths which can increase value of $\Delta\sigma_N$ compared to the two-wavelength DIAL technique according to Eq. (2). However, for DIAL systems to measure other atmospheric gases like ozone, it is only practical to use wavelength selection Method B because of the shape of the ozone absorption spectrum (lacking narrow peaks).

b. The chosen three wavelengths can reduce or remove AED.

It means the value of AED is equal or close to 0. Choosing the appropriate three wavelengths to make the value of K in Eq. (4) equal or close to 0, the value of AED will be equal or close to 0. The value of K in Eq. (4) changes with different aerosol Ångström exponents. For example, to remove boundary layer aerosol influence, we can set aerosol Ångström exponents=1 to calculate the value of K to choose the three wavelengths because the size of aerosol in the boundary layer is typically large.

3. HU three-wavelength OPO DIAL system

The HU lidar system currently consists of a Continuum Horizon II tunable OPO laser and a Continuum Powerlite DLS 8000 pump laser as the light source, a 48-inch non-coaxial Cassegrainian-configured telescope receiver, a light separation system that uses beam splitters and interference filters, a detecting system including photomultiplier tubes (PMT) and avalanche photodiodes (APDs) and a Licel optical transient recorder. The wavelength tuning range of the OPO extends from 192 nm to 2750 nm. This range is fully automated with precision scanning for true hands-free operation. Fig. 3(a) and (b) show the Continuum Horizon II output energy and its parameters. The OPO laser energy outputs between 400 nm and 500 nm which overlap with the NO_2 strong absorption spectral zone in Fig. 2 produce near the maximum possible power in the spectrum. Combining the OPO laser energy outputs, NO_2 absorption spectral and two three-wavelength chosen rules, 438 nm, 439.5 nm and 441 nm shown in Fig. 2 result in the wavelengths of HU three-wavelength DIAL system because $\Delta\sigma_N$ of the three-wavelength pair is more than other three-wavelength pairs in NO_2 strong absorption spectral zone and the *K* value of the three-wavelength is 0.000023 (close to 0). A schematic of the lidar system is shown in Fig.3 (c). The system can be configured to measure multi-wavelength aerosols and NO_2 density.



Description b	
Pulsewidth (nsec)	3-7
Pointing Stability (urad)	<±100
Linewidth (cm^{-1})	
Unseeded	3-7
Doubling/Mixing	<10
Energy Stability (%,99% of shots)	<±10
Divergence (mrad,FWHM)	<2 (both axes)
Beam Diameter (mm, near field)	4-7
Beam Roundness (%, near field)	>85
Polarization (%)	
Signal Horizontal	>99
Idle Horizontal	>99

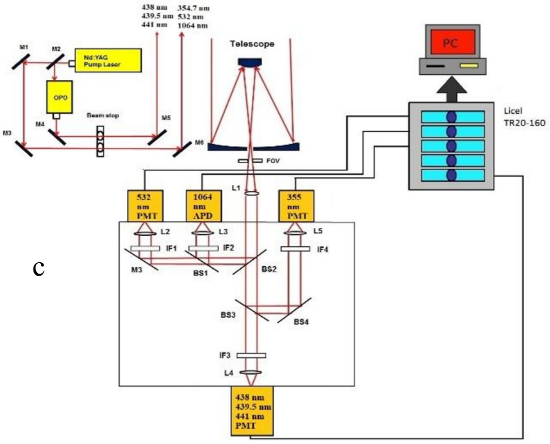
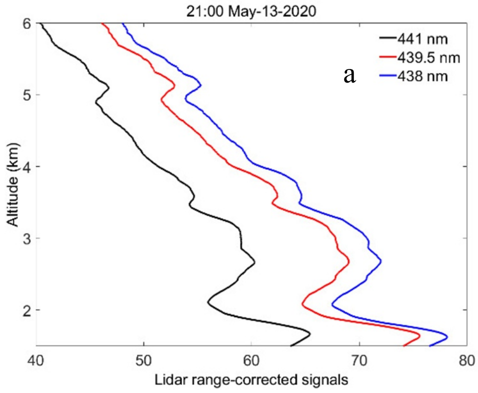


Fig.3 Continuum Horizon II energy outputs (a) and parameters (b) with PL 8000 pump, and the schematic of HU three-wavelength OPO DIAL system (c) (L-lens, M-mirror, BS-beam-splitter, IF-interference filter, FOV-field of view, PMT-Photomultiplier tube, APD-Avalanche Photodetector)

4. Results

Two cases (nighttime and daytime) for vertical profiles of NO_2 obtained from HU three-wavelength OPO DIAL are presented in Fig. 4 and 5. All NO_2 lidar measurements presented here are obtained at times without cloud below 8 km. HU lidar 438 nm (blue line), 439.5 nm (red line) and 441 nm (black line) elastic signals measured at 21:00 (local time) on May 13, 2020 and 11:00 (local time) on December 12, 2023 are shown in Fig. 4 (a) and Fig. 5 (a), respectively. The average integration time for these signals is 2 minutes (night time) in Fig. 4 (a) and 10 minutes (daytime) in Fig. 5 (a). Fig. 4 (b) and Fig. 5 (b) show retrieved NO_2 profiles using the HU three-wavelength OPO DIAL (red line). To assess accuracy of the HU lidar NO_2 profiles, we compare the retrieval results to simulated data from the Weather Research and Forecasting Chemistry (WRF-Chem) model (Grell et al., 2005) at 12 km × 12 km spatial resolution and 200 m vertical resolution. Past studies have demonstrated that WRF-Chem simulated NO_2 results show good agreement between the OMI satellite measurements and aircraft measurements [Amnuaylojaroen et al., 2019] providing a data source to examine the accuracy of the HU retrievals using HU three-wavelength OPO DIAL. A NO_2 profile on May 13 2020 is simulated using WRF-Chem model and shown in Fig. 4 (b). WRF-Chem simulated NO2 magnitudes tend to be lower compared to HU retrieved NO2 profiles using three-wavelength DIAL (typically within ±0.1 ppb), however, the comparison demonstrates a consistent vertical profile shape between observations and the model simulation. In 2022, HU installed a Pandora spectrometer system. The Pandora spectrometer system is an instrument developed to measure vertical column densities of trace gases in the atmosphere using lunar and sun/sky radiation in the UV-visible part of the spectrum. Among currently experimental data products, such as SO_2 and HCHO, validated and near-real-time O_3 and NO_2 total vertical column density data products are included [Herman, et al., 2009; Cede, et al., 2021]. With development of Pandora system, it can obtain vertical profile of NO_2 now. A NO_2 profile on December 12 2023 is obtained using pandora system and shown in Fig. 5 (b). The NO_2 profile retrieved with the HU three-wavelength OPO DIAL agrees well with the profile from Pandora system. However, the NO_2 profile from Pandora system has a low vertical resolution and cannot show more vertical details like HU lidar's.



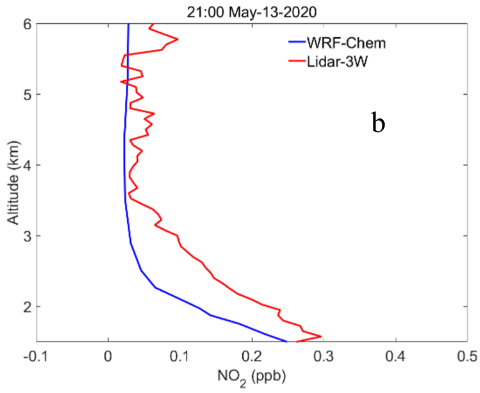


Fig.4 HU lidar 438 nm, 439.5 nm and 441 nm elastic signals measured (a); NO2 profiles obtained using HU three-wavelength OPO DIAL and WRF-Chem model (b) at 21:00 (local time) on May 13, 2020.

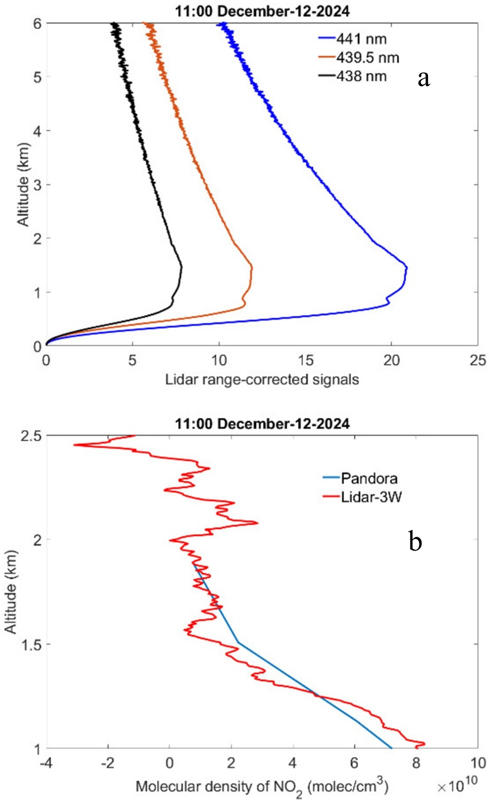


Fig.5 HU lidar 438 nm, 439.5 nm and 441 nm elastic signals measured (a); NO$_2$ profiles obtained using HU three-wavelength OPO DIAL and Pandora (b) at 11:00 (local time) on December 12, 2023.

5. Conclusion

This study describes the measurements of tropospheric NO$_2$ profiles using three wavelengths simultaneously emitted from an OPO laser. The three-wavelength OPO DIAL can decrease errors caused by aerosol interference. Comparing the NO$_2$ profile from HU three-wavelength OPO DIAL to WRF-Chem model output demonstrates that the NO$_2$ magnitudes and vertical structure are in much better agreement with simulated data. Compared to Pandora, the NO$_2$ profile retrieved with HU three-wavelength OPO DIAL agrees well with the profile from Pandora system. However, the NO$_2$ profile from Pandora system has a low vertical resolution and cannot show more vertical details like HU lidar’s. In the future, we plan to purchase NO$_2$ radiosonde for acquiring more validation data to evaluate HU lidar NO$_2$ results.

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