\SIMULATIONS OF PANDORA SPECTROMETER SIGNALS VIA RADIATIVE TRANSFER MODELING FOR DETECTION OF AEROSOLS AND TRACE GASES OVER SINGAPORE

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ABSTRACT: Ambient air quality is of utmost importance in Singapore due to its direct impact on public health and environmental sustainability. Airborne pollutants concentration such as Ozone (O_3), Nitrogen Dioxide (NO_2), and Sulfur Dioxide (SO_2) are important indicators to measure ambient air quality in Singapore. These pollutants can originate from various sources, such as vehicular emissions, industrial activities, and transboundary pollution. Accurate measurements are, therefore, key to identifying pollutant sources, understanding the local atmospheric environment, formulating effective mitigation strategies, and evaluating environmental policies and regulations. Our group plans to address the limited availability of in-situ trace gas measurements at ground level by installing a UV-NIR spectroradiometer (Pandora spectrometer) to provide near-real-time air quality information, supporting future measurement campaigns and satellite validations. This instrument will deliver high temporal resolution measurements of total column amounts of key trace gases, including Sulphur Dioxide, Ozone, Nitrogen Dioxide, Formaldehyde, and water vapor. In this work, we perform a range of radiative transfer computations, coupled with Pandora data, aimed to: (a) evaluate upper and lower limits of NO_2 range of observability and (b) the impact that tropical high degree of humidity (water vapor) might have on planned observations. For this purpose, we used the well-validated MODerate resolution atmospheric TRANsmission (MODTRAN) radiative transfer model to perform a range of numerical simulations of scattering and absorption properties of aerosols and trace gases, similar to those that Pandora would observe. Such a study will provide an initial insight into these pollutant characteristics, being used as a reference for our ground instrument installation.

1. Introduction

The monitoring and characterization of atmospheric constituents, particularly aerosols and trace gases, play pivotal roles in the fields of atmospheric science, air quality assessment and climate research. Singapore, located at the southern tip of the Malay Peninsula, is an urban city where ambient air quality holds immense significance due to its direct impact on public health, the environment and key economic sectors such as tourism. Key atmospheric pollutants such as aerosols and trace gases including Ozone (O_3), Nitrogen Dioxide (NO_2), Sulphur Dioxide (SO_2) and Carbon Monoxide (CO) are used to produce the local air quality index as a proxy of overall ambient air quality. It is therefore vital to be able to establish long term measurement of these pollutants with high precision and timeliness. The Pandora spectrometer has emerged as an indispensable tool in atmospheric remote sensing, offering high spectral and temporal resolution measurements of direct solar and hemispherical sky radiance in the ultraviolet-visible (UV-Vis) spectral range. On the other hand, radiative transfer modelling has proven instrumental in simulating the interaction of sunlight with the atmosphere and subsequently deriving valuable information about aerosols and trace gases. This paper presents a study assessing the performance of Pandora instrument in Singapore as well as a range of radiative transfer simulations aimed to evaluate the limits of pollutants observability of the Pandora and the impact of the high degree of humidity found in tropical environments such as Singapore on the Pandora observations.

2. Methods, Instrumentation and Data processing

As part of the Pandonia Global Network (PGN) (Swap et al., 2018), a Pandora 1S spectrometer, Pandora 77, is installed and operated by the Centre for Remote Imaging, Sensing and Processing at the National University of Singapore (1.298◦ N, 103.780◦ E). The Pandora is a UV-NIR spectroradiometer that can provide near real time measurements of important trace gas column concentrations that are vital for satellite data validation and air quality monitoring (Cede, 2017; Herman et al., 2009). Measurements made by Pandora are analyzed using a Differential Optical Absorption Spectroscopy (DOAS) technique capable of producing a data point every 120 seconds. The instrument produces data at 4 levels: raw field spectra (L0), data with instrument characteristics applied (L1), data with spectral fitting applied (L2Fit) and surface concentration, tropospheric column and total columns (L2). The specifications of the Pandora as well as some basic description of the L2 data products used in this study are summarized in Table 1.

	Pandora 77	Nitrogen Dioxide (NO_2)	Ozone (O_3)	Formaldehyde (HCHO)	Sulfur Dioxide (SO_2)
Spectral range	270nm – 530nm	-	-	-	-
Spectral resolution	0.6nm	-	-	-	-
Slit function	Gaussian	-	-	-	-
SNR	350				
Level 2 Data product	-	rnvs3p1-8	rout2p1-8	rfus5p1-8	rsus1p1-8
Observation made	-	Direct sun	Direct sun	Direct sun	Direct sun
Fitting window	-	400nm – 470nm	305nm – 325nm	322.5nm – 359.2nm	306nm – 326.4nm

Table 1: Specification of Pandora and L2 data products.

In this study, L2 data of the Pandora 77 instrument including: NO_2, O_3, HCHO and SO_2 are processed to access the instrument's measurement limit. Data is obtained from 14 June to 11 September 2023. The data processing aims to remove data that is of bad quality through filtering of quality assurance flags. Data with the following quality flags are used in this study: assured high (0), assured mid (1), unassured high (10), and unassured mid (11). Next, the data is filtered based on its measurement solar zenith and azimuth angle to match the simulation parameters of the radiative transfer model described in the next section. Data measured when the sun is near the zenith is obtained by filtering for: solar zenith angle of less than 10 degrees and solar azimuth angle between 340 and 20 degrees. The resulting data is used to determine the measured total column pollutant amount as well as the measurement uncertainty. The total measurement uncertainty is used in this study as this particular instrument (Pandora 77)'s detection limit as it is unable to distinguish any measurement smaller than the total uncertainty between noise and actual presence of pollutants. After the data processing step, only the L2 NO_2 product yielded sufficient data points (61) for further analysis as compared to O_3 (0), HCHO (2) and SO_2 (2). Therefore, this study will focus on the analysis of NO_2 measurements and simulations via MODTRAN.

The MODerate resolution atmospheric TRANsmission (MODTRAN) is a well validated radiative transfer code capable of simulating a wide range of optical properties for various aerosols and trace gases (Berk et al., 1989). Here in the present study, the MODTRAN model was used to assess the impact of the high amount of humidity in the tropical atmosphere such as in Singapore. Table 2 shows the MODTRAN model parameters for Pandora's internal multi-spectral sensor simulation runs. In addition, the sensor or observer view in MODTRAN's path geometry was such that it points directly at the sun which is at the zenith point to simulate a direct sun measurement of the Pandora. This is the reason for the data processing step previously described to filter for measurements as close to the zenith as possible.

MODTRAN run mode	Transmittance/Solar
RT Option	Band model
Band model resolution	0.1 cm^{-1}
Multiple scattering	DISORT MS
DISORT streams	8
Atmosphere model	Tropical
Carbon Dioxide (CO_2)	400 ppm

Cloud model	None
Aerosols model	Urban
Stratospheric model	Background
Phase function	Mie
Observer height	77m
Observer zenith angle	0.0°
Path azimuth angle	0.0°
Solar zenith angle	0.0°
Solar azimuth angle	0.0°
Day of the year	84
Spectral range	270nm – 530nm
Spectral resolution	0.6nm
Slit function	Gaussian
Top-of-atmosphere solar irradiance	Thullier plus 1997 Kurucz

Table 2: MODTRAN simulation parameters

Two run modes are employed in this study: transmittance mode and solar mode. Transmittance mode is able to produce the transmittance due to specific species of interest such as NO_2 and H_2O, and can be used as a preliminary tool to look for water vapor interference in the fitting window of NO_2 (400nm-470nm). The solar run is able to produce the solar irradiance that reaches the sensor, similar to the measurement of the direct sun retrieval of Pandora. By varying only the total NO_2 column amount, contributions due to the presence of NO_2 can be used to identify absorption peaks in the fitting window suitable for NO_2 retrieval. Finally, the impact of humidity or water content on NO_2 retrievals will be investigated by multiple MODTRAN simulations of varying water content at multiple fixed total NO_2 column amounts.

3. RESULTS AND DISCUSSION

3.1 Pandora 77 data and uncertainty

After the data processing step, we obtained 61 valid data measurements from the L2 NO_2 total column amount product. Figure 1 shows the resulting data set of NO_2 total column amount and total uncertainty. The total uncertainty follows a similar trend as the total column amount but is overall much more consistent. Note that the total uncertainty value is expressed on the secondary axis and is a degree of magnitude less than the total column amount. The mean total column amount obtained here is 0.9 Dobson Unit (DU, where 1 DU = 2.69×10^{16} molecules cm^{-2}) while the mean total uncertainty is 0.09 DU. The total uncertainty obtained here differs slightly from the instrument's given accuracy of 0.05 DU (Müller et al., 2017) and is a more accurate representation of the instrument's performance in Singapore's atmospheric environment.

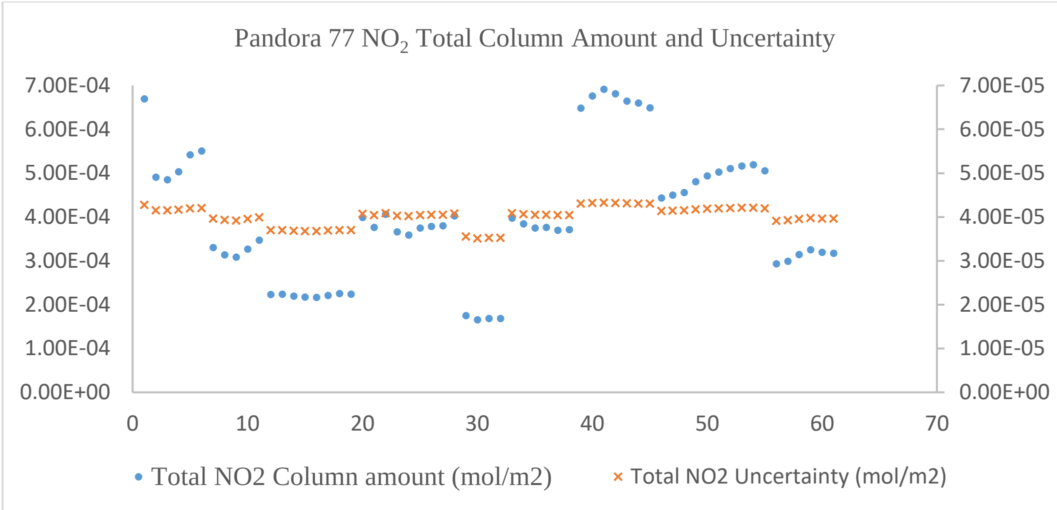

Figure 1: Pandora 77 NO2 Total column amount and uncertainty

Using the mean total column amount and uncertainty obtained above, we developed three cases of NO2 profiles to be simulated with MODTRAN. Case A included the built in MODTRAN tropical atmosphere with the NO2 scaling set to 0, this case enabled simulating hypothetical scenarios without any NO2 presence. Case B profile is simply the MODTRAN tropical model, and has a total column amount of 0.21091 DU of NO_2 (Berk, 2019). Finally, case C represents the mean NO_2 total column amount of 0.9 DU measured by Pandora in NUS and is a more realistic estimation of the amount of NO_2 in Singapore's environment. The three cases are summarized in Table 3 below.

	Case A	Case B	Case C
Description	Tropical model with NO_2 scaling set to 0	Tropical model with default NO_2 scaling	Tropical model with NO_2 scaled to mean Pandora NO_2 total column
NO2 total column amount	0 DU	0.21091 DU	0.9 DU

Table 3: Description of 4 cases of NO_2 profiles used in MODTRAN simulations.

3.2 MODTRAN transmittance only run

A MODTRAN transmittance simulation was done using Case B NO_2 profile to examine the transmittance due to water vapour and NO_2 specifically. Figure 2 and 3 shows the MODTRAN simulated transmittance for H_2O and NO_2 in the full spectral range (270nm – 530nm) and the NO_2 fitting window (400nm – 470nm) respectively. It can be observed in Figure 2 that there are a number of absorption peaks for H_2O, with the largest at 504 nm. While this is outside of the fitting window of Pandora NO_2 retrieval, figure 3 shows another significant H_2O absorption peak at 440 nm. The NO_2 transmittance did not show clear absorption peaks in the fitting window, rather there is likely a continuum contribution at a large number of wavelengths within the window. The impact of the water absorption peak at 440 nm and the behaviour of NO_2 absorption within the fitting window were examined further using solar MODTRAN runs in the following section.

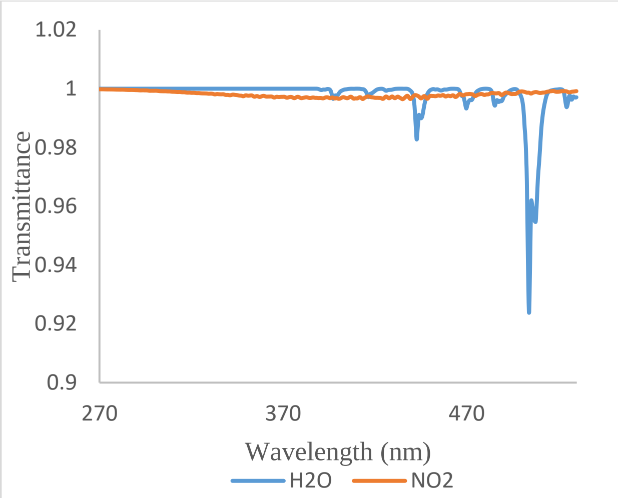

Figure 2: MODTRAN transmittance

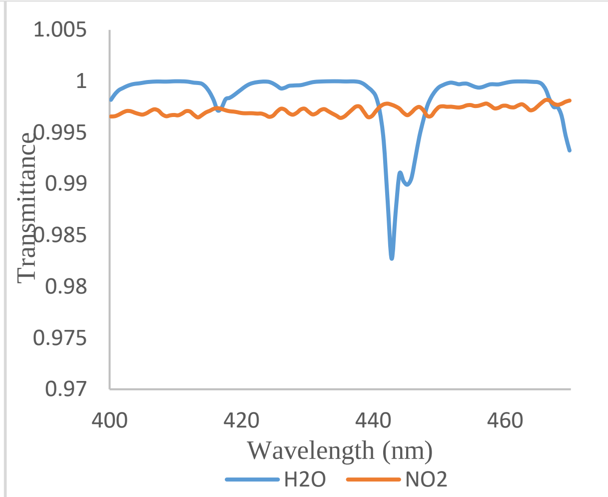

Figure 3: MODTRAN transmittance within NO_2 fitting window

3.3 MODTRAN solar run

A MODTRAN solar run was performed for each of the three cases of NO_2 profiles described in Table 3. Figure 4 shows the solar irradiance of each of the three cases. Case A, with no NO_2 in the atmosphere, is used as the basis of comparison to determine the contribution of only NO_2 to the solar irradiance, I, measured by the sensor. These contributions were obtained by calculating the relative difference between case B and C solar irradiance as compared to case A. The relative difference by case B and C are given by:

$$\frac{|I_B - I_A|}{I_A}, \frac{|I_C - I_A|}{I_A}$$

As the only varying factor between cases B, C and case A is the amount of NO_2 in the atmosphere model, the relative differences measured is directly attributed to NO_2 absorption in the direct sun measurement. The relative difference in the solar irradiance at observer between case B and A and case C and A are shown in figure 5.

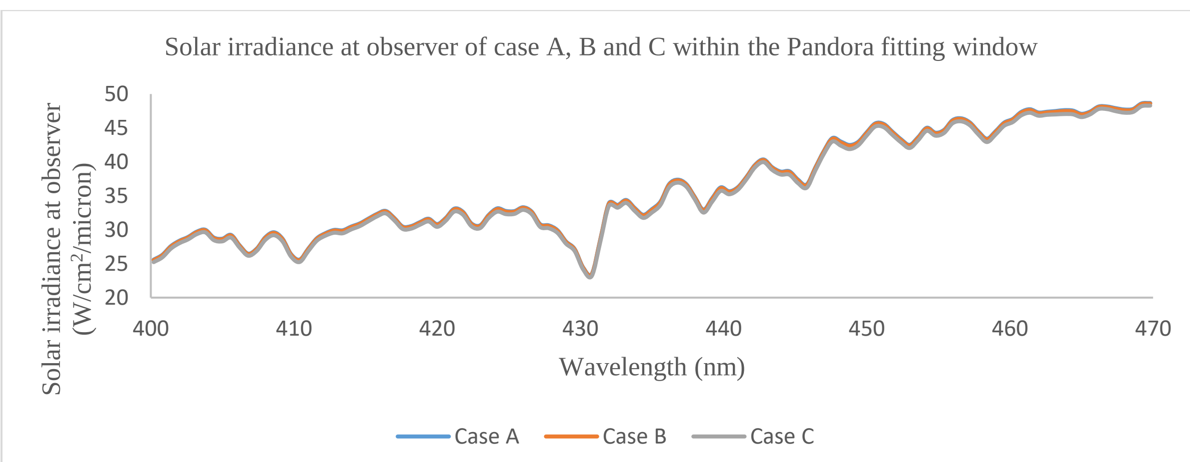

Figure 4: Solar irradiance at observer of case A, B and C within the Pandora fitting window.

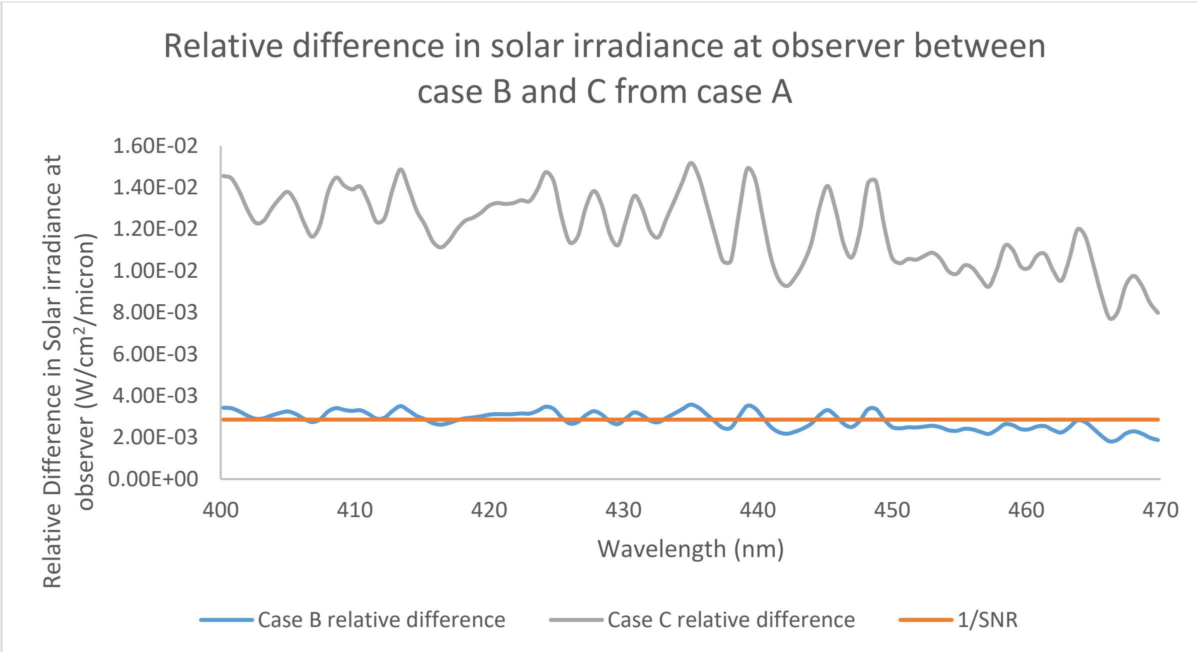

Figure 5: Relative difference in solar irradiance at observer between case B and C from case A within the NO_2 Pandora fitting window.

The largest relative difference in the solar irradiance measured was observed for case C, which corresponded to the mean total NO_2 column amount observed of 0.9 DU. This suggested that the default tropical atmosphere model within MODTRAN is not sufficient alone to represent Singapore's atmospheric environment and therefore would need to be further adjusted for more accurate and realistic simulations of Pandora measurements. Additionally, a horizontal line representing a threshold of the inverse of the signal to noise ratio (SNR) of the Pandora sensor of 350 is overlaid in figure 5. The relative difference found between cases B, C and case A must be above this value for the instrument to clearly resolve the signal. In figure 5, we can clearly observe that the relative difference due to case C is larger than 1/SNR at all wavelengths in the fitting window, however, the relative difference due to case B is only larger than 1/SNR at certain wavelength windows. Finally, Figure 5 can be used to determine the wavelength where absorption due to NO_2 is the most pronounced. The model output suggests that the most absorption due to NO_2 occurred at 435nm and 439.2nm. Note that these wavelengths is relatively close to the water absorption peak at 440nm shown by the transmittance run and the impact of water absorption will be investigated next.

3.4 Impact of water on NO_2 retrieval

Due to the proximity of the largest NO_2 absorption wavelength at 435nm and 439.2nm and the water absorption peak in the NO_2 retrieval fitting window at 440nm, there could be potential degradation in the Pandora direct sun measurement due to the high degree of humidity present in Singapore's atmosphere. Thus, to assess the effect humidity has on NO_2 direct sun measurements, MODTRAN simulations were carried out by varying the column water amount while fixing the NO_2 total column amount. Water content of 0, 2, 4, 6, 8 and 10 g/cm^2 were simulated for each of the 4 cases of NO_2 profiles in Table 3. The resultant solar irradiance at observer produced by MODTRAN are shown in Figure 6 below.

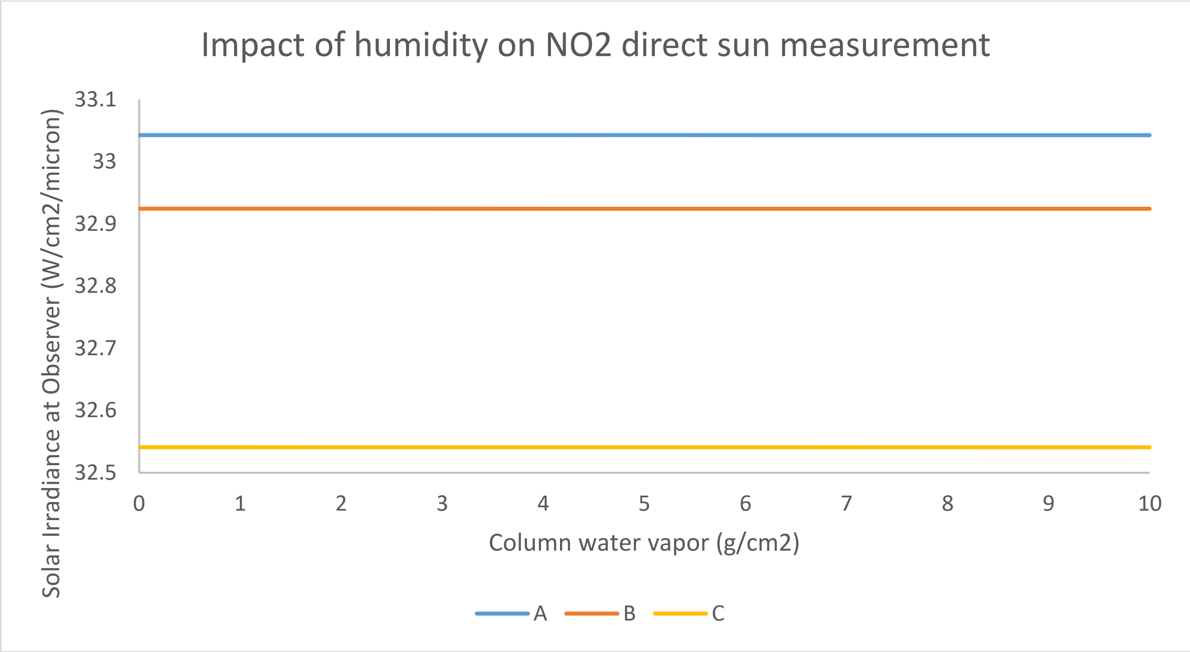

Figure 6: Impact of humidity on NO_2 direct sun measurement at 435nm.

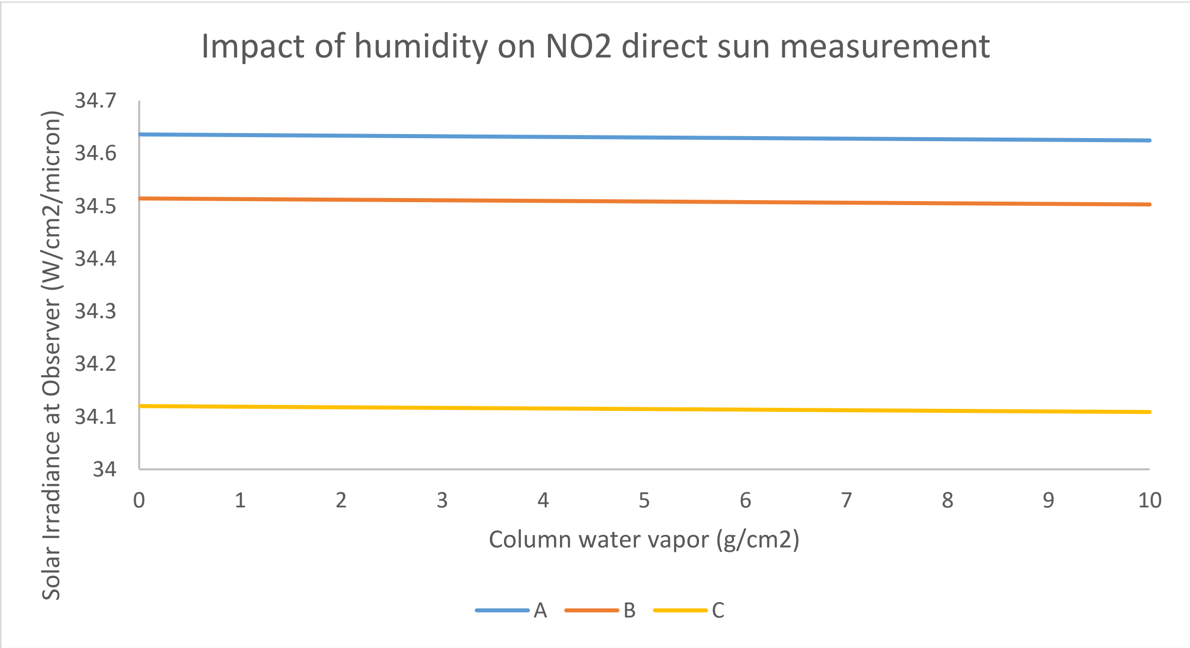

Figure 7: Impact of humidity on NO_2 direct sun measurement at 439.2nm.

A linear degradation of the solar irradiance was observed at all three cases at 439.2nm while there is practically no degradation at 435nm. There is a 3.3% degradation in solar irradiance at 10 g/cm^2 as compared to 0 g/cm^2 for all three cases at 439.2nm. This suggests that the impact humidity has on NO_2 retrieval is relatively consistent for NO_2 column amount up to the environment's background amounts. However, NO_2 retrieval at the wavelength of 439.2 nm should nonetheless take into account any such degradation in the solar irradiance during direct sun measurements. Furthermore, there exists possibility of a larger degradation in the measurement in an elevated NO_2 pollution event which can be further investigated with in-situ data and corresponding simulation of such an event.

4. Conclusion

In this study, we examined Pandora 77 NO_2 total column data since its installation at CRISP, NUS for the period of 14 June to 11 September 2023. We obtained a mean NO_2 total column amount of 0.9 DU with an uncertainty or accuracy of 0.09 DU. Multiple MODTRAN numerical simulations were carried out to study the NO_2 absorption behavior within the Pandora fitting window of 400 – 470 nm. It is found that NO_2 absorption is the most significant at 435 nm and 439.2 nm. The impact of high humidity in Singapore's atmosphere on the retrieval of NO_2 total column amount was investigated using MODTAN and a significant water absorption peak is found at 440 nm, within the NO_2 fitting window. At 439.2 nm, a linear degradation on the simulated signal measured by Pandora is found and the solar irradiance reaching the sensor is found to degrade by 3.3% when water content is increased from 0 to 10 g/cm^2. This degradation appeared to be consistent for NO2 total column amount of up to the ambient mean of 0.9 DU. However, no degradation in data is found at 435 nm, potentially making this wavelength more suitable for NO_2 retrieval in Singapore.

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