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# Retrieval of NO<sub>2</sub> vertical columns under cloudy conditions: A sensitivity study based on SCIATRAN calculations

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#### 1. Introduction

Nitrogen dioxide (NO<sub>2</sub>) plays an important role in the troposphere. It is one of the key players in the formation of photochemical smog during pollution episodes. NO<sub>2</sub> also contributes to acid rain. The vertical columns of the nitrogen dioxide *V* are usually measured using the differential optical absorption spectroscopy technique (Platt, 2007). The technique is based on the fact that the directly transmitted solar light is attenuated in atmosphere and this attenuation is different inside and outside NO<sub>2</sub> absorption bands. As a matter of fact the depth of the nitrogen dioxide absorption bands is influenced by the value of *V* being larger for the atmosphere containing more NO<sub>2</sub>.

Nitrogen dioxide columns can be retrieved not only from the measurements of the direct transmitted solar light but also spectral measurements (e.g., in the interval 420–440 nm) of the diffuse (transmitted or reflected) solar light can be used. The use of the reflected solar light is especially important because then satellite measurements can be utilized for the studies of the global distribution of the nitrogen dioxide total vertical columns (Richter and Burrows, 2002; Platt, 2007; Kokhanovsky et al., 2008; Wagner et al.,

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### ABSTRACT

The accuracy of the retrieved  $NO_2$  vertical columns using satellite observations under cloudy conditions have been studied using the radiative transfer code SCIATRAN. It was found that the tropospheric nitrogen dioxide columns can be retrieved in the case of thin clouds, if their optical properties and the altitude are retrieved from independent observations. The dependence of the retrieval error on the accuracy of the information on cloud top height and cloud optical thickness is studied.

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2008). The tropospheric  $NO_2$  columns are usually retrieved assuming that the variation of the stratospheric part of the NO<sub>2</sub> column with the longitude is small and, therefore, the tropospheric NO<sub>2</sub> columns over a given location (e.g., over a city) can be retrieved subtracting the vertical column over a clean site (e.g., Pacific) from the total vertical column at a given location with the same latitude (Kokhanovsky et al., 2008). Only clear sky pixels are used in the retrieval process because the standard retrieval algorithms are not capable to deal with cloudy pixels. In the case of overcast sky, the information content of measurements with respect to NO<sub>2</sub> tropospheric columns is low and retrievals are not possible. However, for thin clouds, there is a possibility to get information on NO<sub>2</sub> tropospheric columns under them. For this, the NO<sub>2</sub> vertical columns retrievals must be coupled with cloud property retrievals (cloud top height, cloud optical thickness, etc.). The corresponding combined retrieval algorithms are currently under development.

The task of this paper is to perform the sensitivity studies of the tropospheric nitrogen dioxide retrieval algorithm based on satellite measurements for a cloudy sky in the spectral range 420–440 nm with respect to the errors in the cloud property assumptions/retrievals.

#### 2. The retrieval algorithm and results

The NO<sub>2</sub> satellite retrieval algorithm developed in the framework of the radiative transfer software package SCIATRAN

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430

wavelength, nm

435

440

425

(Rozanov et al., 2005) is used in this paper. The algorithm is applied to the simulated spectral measurements of top-of-atmosphere reflectance in the spectral window 420–440 nm. The behavior of the NO<sub>2</sub> vertical optical depth (VOD) in the selected spectral range is shown in Fig. 1. It follows that the VOD is quite small (just 0.002). However, the spectral dependence is significant (about two times change in the selected window). The correspondent spectral variations of the registered back-scattered solar light are used in the retrieval process. The total vertical column *V* for the case demonstrated in Fig. 1 was  $3.2977e + 15 \text{ molec/cm}^2$  with the tropospheric column *V*<sub>t</sub> (below 12 km) of  $2.0128e + 14 \text{ molec/cm}^2$ . The NO<sub>2</sub> volume mixing ratio profile used is shown in Fig. 2 (the clean case).

First of all retrievals have been performed for the clear sky case. SCIATRAN allows calculations both in the forward and inversion modes. The assumed (true) values of V were correctly retrieved running the inverse model for the same conditions as the forward model used for the simulation of the experimental (synthetic) data. The next step was to retrieve the value of  $V_t$  assuming that the stratospheric part of the vertical column is exactly known and a cloud is present in



Fig. 2. The NO<sub>2</sub> vertical profiles used in this work (clean case – circles  $(V_t=2.013e+14 \text{ molec/cm}^2, V=3.2976e+15 \text{ molec/cm}^2))$ , polluted case – squares  $(V_t=3.1430e+15 \text{ molec/cm}^2, V=5.8476e+15 \text{ molec/cm}^2))$ .



**Fig. 3.** The spectral top-of-atmosphere reflectance for the clear (lower line) and cloudy (upper line) cases. The cloud optical thickness is equal to 10.

atmosphere during the measurements. Therefore, the experimental spectrum was modeled for the same atmospheric scenario as used in the retrieval process (see Appendix) except a vertically homogeneous cloud of a geometrical thickness 1 km was put at different levels in atmosphere. Three cloud top heights were assumed: 2, 6, and 12 km. The cloud optical thickness (COT) varied in the range 0.1–10. The ice cloud phase function was calculated using the geometrical optics in the assumption of the Macke's fractal ice grains (Macke et al., 1996).

The retrievals were performed converting the calculated top-of-atmosphere spectral reflectances  $R(\lambda)$  (see Fig. 3) to the so-called differential optical depths (DOD)  $D(\lambda)$ , which are calculated as

$$D(\lambda) = \ln(R(\lambda)) - P(\lambda), \tag{1}$$

where  $P(\lambda)$  is the polynomial of the second order with the respect to the wavelength  $\lambda$ . In the retrieval algorithm the quadratic form

$$F(\lambda, V_{t}) = |D(\lambda) - D_{c}(\lambda, V_{t})|^{2}$$
<sup>(2)</sup>

is minimized with respect to unknown parameter  $V_{t}$ . Here  $D_{c}(\lambda, V_{t})$  is the DOD spectrum calculated with SCIATRAN for various values of  $V_{t}$ .

The DOD is given in Fig. 4. Results shown in Figs. 3 and 4 were obtained for the clean case given in Fig. 2. One concludes that the spectral DODs have much more pronounced dependence on the wavelength as compared to reflectances both for cloudy and a clear sky cases shown in Fig. 3. For instance, absorption peaks are clearly seen. It follows from Fig. 4 that the NO<sub>2</sub> absorption features are less pronounced in the cloudy sky DOD spectrum as compared to a clear sky one.



**Fig. 4.** The differential optical depths for the case shown in Fig. 3. The spectrum for the cloudy case (lower line) is characterized by more shallow absorption bands as seen in the reflected light.

VOD(NO2)

0.0030

0.0028

0.0026

0.0024

0.0022

0.0020

0.0018

0.0014

0.0010

60

50

40

20

height, km 00 420

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