



**HAL**  
open science

## Retrieval of tropospheric NO<sub>2</sub> columns from satellite measurements in presence of cirrus: A theoretical sensitivity study using SCIATRAN and prospect application for the A-Train

Jérôme Vidot, Olivier Jourdan, Alexander Kokhanosvky, Frédéric Szczap, Vincent Giraud, Vladimir Rozanov

### ► To cite this version:

Jérôme Vidot, Olivier Jourdan, Alexander Kokhanosvky, Frédéric Szczap, Vincent Giraud, et al.. Retrieval of tropospheric NO<sub>2</sub> columns from satellite measurements in presence of cirrus: A theoretical sensitivity study using SCIATRAN and prospect application for the A-Train. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 2010, 111 (4), pp.586-601. 10.1016/j.jqsrt.2009.10.015 . hal-01971909

**HAL Id: hal-01971909**

**<https://hal.science/hal-01971909v1>**

Submitted on 20 Mar 2022

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1           **Retrieval of tropospheric NO<sub>2</sub> columns from satellite**  
2 **measurements in presence of cirrus: a theoretical sensitivity study**  
3 **using SCIATRAN and prospect application for the A-Train**

4

5

6           Jérôme Vidot<sup>a,\*</sup>, Olivier Jourdan<sup>a</sup>, Alexander A. Kokhanosvky<sup>b</sup>, Frédéric Szczap<sup>a</sup>,

7

Vincent Giraud<sup>a</sup> and Vladimir V. Rozanov<sup>b</sup>

8

9

10 <sup>a</sup>Laboratoire de Météorologie Physique (LaMP), Université Blaise Pascal, OPGC/CNRS,

11

24 avenue des Landais, Aubière, France, F-63177

12

13 <sup>b</sup>Institute of Environmental Physics (IUP), Institute of Remote Sensing (IFE),

14

University of Bremen, Otto-Hahn-Allee 1, Bremen, Germany, D-28359

15

16

17\*Corresponding author. Fax: +33 (0)473405136

18E-mail address: J.Vidot@opgc.univ-bpclermont.fr

19

20

21**Abstract**

22A theoretical sensitivity study of the influence of cirrus cloud properties on tropospheric NO<sub>2</sub>

23columns retrieved from the spaceborne Ozone Monitoring Instrument (OMI) measurements is

24performed. It is conducted within the framework of the synergetic use of A-Train sensors to

1derive more representative trace gas products. We aim to study the potential effects of cirrus  
2clouds on tropospheric NO<sub>2</sub> retrievals using a retrieval algorithm that, unlike the OMI  
3Standard and DOMINO algorithms, does not correct for the effects of clouds. The sensitivity  
4study is based on the radiative transfer code SCIATRAN that performs both simulations of  
5top of atmosphere (TOA) reflectances as measured by an OMI-like band and tropospheric  
6NO<sub>2</sub> column retrievals based on the Differential Optical Absorption Spectroscopy (DOAS)  
7method. The results of the sensitivity study show that if a correction for cirrus clouds is not  
8included in our simple retrieval that does not account for clouds in the first place, the  
9tropospheric column can be underestimated by 55%. This underestimation depends strongly  
10on cirrus parameters as, in order of importance, cloud fraction, cloud optical depth,  
11asymmetry factor of cirrus cloud phase function and cloud top height. The perspective of the  
12synergy between OMI and cloud information obtained from cloud-derived products of the A-  
13Train is evaluated in two parts by applying a simple cloud correction scheme based on the  
14independent pixel approximation (IPA). Firstly, we evaluated the tropospheric NO<sub>2</sub> column  
15retrievals error caused by uncertainties in cirrus cloud properties. Secondly we studied the  
16influence of subpixel cloud optical depth variability on NO<sub>2</sub> retrievals. From our simulations,  
17it is demonstrated that the error will be reduced significantly if the cloud fraction is lower or  
18equal to 0.5. In this case, the cloud fraction and the cloud optical depth must be known within  
19accuracy less than 0.05 and 50 %, respectively. The cloud top height and the asymmetry  
20factor must be known within uncertainty of at least 1 km and less than 0.05, respectively. The  
21latter result shows that the uncertainty of the asymmetry factor is a major source of error in  
22the cloud correction for tropospheric NO<sub>2</sub> retrieval in the presence of cirrus.

23

24**Keywords:** Cirrus properties; OMI; Tropospheric NO<sub>2</sub> vertical column; SCIATRAN; A-  
25Train.

## 11. Introduction

2

3 Nitrogen dioxide (NO<sub>2</sub>) plays an important role in the tropospheric chemistry [1]. NO<sub>2</sub> is  
4 known to be one of the key species in the formation of photochemical smog during pollution  
5 episodes. In the troposphere, the concentration of NO<sub>2</sub> takes part in the chemical budget of  
6 ozone. It also contributes to acid rain and locally, to radiative forcing over industrial regions  
7 and urban areas [2]. To assess accurately our current knowledge of tropospheric chemistry  
8 and its interaction with climate, global information about the amounts and distribution of NO<sub>2</sub>  
9 is required. By their high spatial and temporal coverage, satellite measurements of NO<sub>2</sub> are  
10 essential for air quality monitoring (for health regulation) and regional scale modelling  
11 (improvement of emission estimates).

12 The measurement of tropospheric NO<sub>2</sub> from space began with the precursor satellite  
13 Global Ozone Monitoring Experiment (GOME) [3] followed by the Scanning Imaging  
14 Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) [4] and the Ozone  
15 Monitoring Instrument (OMI) [5]. The retrieval algorithms of the NO<sub>2</sub> vertical columns are  
16 based on the Differential Optical Absorption Spectroscopy (DOAS) approach [6]. This  
17 technique is based on the analysis of differential structure of backscattered signal using non  
18 linear least squares fitting in a specific wavelength window. The spectral fit determines a slant  
19 column density of NO<sub>2</sub> which is converted into a vertical column by application of an air mass  
20 factor (AMF). Retrievals of tropospheric NO<sub>2</sub> columns from GOME, SCIAMACHY and OMI  
21 have demonstrated the weekly cycle of NO<sub>2</sub> [7], its relationship with NO<sub>x</sub> emission, the  
22 annual trend over industrial countries [8], the global cartography of tropospheric NO<sub>2</sub> [9] and  
23 the diurnal evolution of NO<sub>2</sub> driven by emissions and photochemistry [10]. Space-based  
24 measurements of NO<sub>2</sub> have been validated against ground-based measurements [11,12],  
25 atmospheric models [13,14] and aircraft measurements [9,15].

1 Nevertheless, the retrieval of NO<sub>2</sub> from space measurements can be subject to significant  
2 errors. Boersma et al. [16] showed that NO<sub>2</sub> tropospheric vertical column can only be  
3 retrieved with an accuracy of 35-60%. The retrieval errors are dominated by uncertainties in  
4 the tropospheric AMF. The *a priori* NO<sub>2</sub> profile shape, the surface albedo and the cloud  
5 properties (especially cloud fraction) are the leading sources of errors associated with AMF  
6 computation. Moreover, clouds cover approximately 60% of the earth's surface. Hence,  
7 considering the weak spatial resolutions of trace gas monitoring sensors (for GOME: 320×40  
8 km<sup>2</sup>, SCIAMACHY: 60×30 km<sup>2</sup> and OMI: 13×24 km<sup>2</sup> at nadir), more than 90% of their  
9 measurements are contaminated by clouds [17].

10 The effects of clouds on trace gas retrieval from space measurements can be separated  
11 into three parts [18-22]. The first effect, called shielding effect, reduces the interaction  
12 between photons and trace gas underneath the cloud leading to an “apparent” decrease of  
13 depths of absorption lines. The second effect, called albedo effect, increases the depth of  
14 absorption lines of the gas layer above the cloud as compared to the clear sky. This is due to  
15 enhanced single and multiple scattering light paths from Sun to cloud to satellite. The third  
16 effect, called in-cloud absorption, increases the depth of absorption lines of gas layer inside  
17 the cloud. Here, light scattering process due to the cloud is responsible for the light path  
18 enhancement as compared to a nonscattering layer. These three effects are competing together  
19 and the final effect depends on the cloud properties as well as on the profile of the trace gas.  
20 Therefore, accurate space-based retrievals of trace gas column in presence of clouds  
21 necessarily imply a precise assessment of the cloud radiative and macrophysical properties  
22 which are, in turn, determined by their microphysical, optical and geometrical characteristics.

23 The most important point to perform a cloud correction for the retrievals of tropospheric  
24 NO<sub>2</sub> column is to identify the cloud parameters that significantly modify the depth of  
25 absorption lines of NO<sub>2</sub>. Studies of Boersma et al. [16] and Wang et al. [23] showed that the

1cloud fraction, cloud albedo and cloud pressure were important quantities for cloud  
2correction. Errors in these cloud properties will directly end up in errors in NO<sub>2</sub> columns.  
3However the quantitative estimate of the retrieval errors depends on the chosen cloud model.  
4For OMI, the cloud correction scheme of the current algorithm is based on the simplified  
5Lambertian cloud model that assumes a homogeneous cloud with an albedo of 0.8 [24]. This  
6method corrects the cloud contribution of the total reflectance at the top of atmosphere (TOA)  
7measured for an OMI pixel using only two cloud parameters: the effective cloud fraction and  
8cloud pressure. The use of this Lambertian cloud model compared to a scattering cloud model  
9has been estimated to lead to AMF differences between -10% and 10% [23] and mean AMF  
10differences of -12% with a standard deviation of 10% [25], depending on cloud properties,  
11cloud fraction and NO<sub>2</sub> pollution. However, the assumption of a Lambertian cloud model is  
12only valid if the bi-directional properties of light reflectance from clouds can be neglected.

13 In order to overcome these limitations, the cloud correction algorithm can be improved on  
14the basis of collocated data from cloud sensors and gas sensors onboard different satellites.  
15The A-train satellite constellation allows for near simultaneous measurements of cloud and  
16trace gas properties at different spatial scales. For instance, high spatial resolution (typically  
17(1×1 km<sup>2</sup>) cloud properties can be assessed from the Moderate Resolution Imaging  
18Spectroradiometer (MODIS) [26] and from the 5-km resolution cloud product provided by the  
19Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) (Level 2 Algorithm Theoretical  
20Basis Document, available at [http://www-calipso.larc.nasa.gov/resources/pdfs/PC-SCI-](http://www-calipso.larc.nasa.gov/resources/pdfs/PC-SCI-21202_Part4_v1.0.pdf)  
21202\_Part4\_v1.0.pdf). This subpixel cloud information (cloud cover, optical thickness, cloud  
22top height...) can then be used to constrain a cloud correction algorithm applied to OMI  
23measurements. Nevertheless, the use of CALIOP information is limited by the fact that  
24CALIOP only overlaps with the nadir pixel of OMI.

1 This paper lies within the framework of using the synergy of A-Train instruments to  
2 improve trace gas retrievals in presence of cirrus clouds. Due to their low optical thickness,  
3 cirrus clouds are more difficult to detect compared to cumulus, particularly over land and ice  
4 surfaces. Additionally, infrared measurements have shown that thin cirrus clouds (with optical  
5 depths between 0.1 and 1.0) have a global frequency of about 20 to 40% [27]. More recently,  
6 a cirrus occurrence up to 70% near the tropics over the 100°-180°E longitude band has been  
7 derived from active measurements by CALIOP [28]. From these observations, this paper aims  
8 at studying into the potential effects of cirrus clouds on the tropospheric NO<sub>2</sub> column  
9 retrievals from OMI space-based measurements and the impact of uncertainties in cirrus cloud  
10 properties in a cloud correction scheme based on the IPA. It is important to point out that this  
11 study is focusing on the effect of cirrus clouds properties in the NO<sub>2</sub> absorption band and that  
12 the results presented here are not compared with the operational cloud correction scheme of  
13 OMI that is based on the O<sub>2</sub>-O<sub>2</sub> absorption band [24]. Even if we can assume that the O<sub>2</sub>-O<sub>2</sub>  
14 band is sensitive to cirrus, the OMI O<sub>2</sub>-O<sub>2</sub> algorithm will retrieve, in presence of cirrus, a  
15 small value for effective cloud fraction and high cloud height. For small cloud fractions, the  
16 assumption on the surface albedo plays an important role and will impact the retrieval of  
17 tropospheric NO<sub>2</sub> column. This effect is beyond the scope of the paper, but the new surface  
18 albedo database made from OMI itself (Kleipool et al., 2008) will certainly change the  
19 tropospheric NO<sub>2</sub> retrievals, because the previous albedo database was from GOME with a  
20 coarser pixel resolution than OMI.

21 Theoretical sensitivity study of the inversion of the tropospheric NO<sub>2</sub> column to  
22 undetected or retrieved (with errors) cirrus cloud properties is discussed in this paper. The  
23 sensitivity study is based on radiative transfer model (RTM) simulations of TOA reflectances  
24 as measured in an OMI-like band. The first part of the paper will describe the atmospheric  
25 model, the retrieval algorithm and the cirrus cloud model. In the second part of the paper, the

1 results of the sensitivity study that highlight the cirrus parameters that influence tropospheric  
2 NO<sub>2</sub> column retrievals are presented. The last part is devoted to the perspective on application  
3 of A-Train cloud-derived products by estimating tropospheric NO<sub>2</sub> column error caused by  
4 uncertainties in cirrus cloud parameter or caused by subpixel cloud optical depth  
5 inhomogeneity within an OMI pixel at nadir.

6

7

## 82. NO<sub>2</sub> retrieval algorithm and model setups

9

### 102.1 Atmospheric model

11 In our study, the atmosphere is treated as plane-parallel. In the spectral band used by OMI  
12 for NO<sub>2</sub> retrieval (between 405 and 465 nm), the two other trace gases, namely O<sub>3</sub> and H<sub>2</sub>O  
13 are included with fixed standard atmospheric profiles. The collision complex of oxygen  
14 molecules O<sub>2</sub>-O<sub>2</sub> is included as well by assuming a fixed standard atmospheric profile of O<sub>2</sub>.  
15 The influence of cirrus clouds is evaluated in different tropospheric NO<sub>2</sub> situations  
16 characterized by the three atmospheric profiles of NO<sub>2</sub> Volume Mixing Ratio (VMR in ppbv)  
17 shown in Fig. 1. The NO<sub>2</sub> VMR profile represented by the full line corresponds to the lowest  
18 NO<sub>2</sub> contribution in the troposphere whereas the NO<sub>2</sub> VMR profile represented by the dotted-  
19 dashed line corresponds to the largest NO<sub>2</sub> contribution. The tropospheric part of the total  
20 atmospheric NO<sub>2</sub> vertical column is commonly retrieved by assuming that the longitudinal  
21 variation of the stratospheric NO<sub>2</sub> vertical column is small. Therefore, tropospheric NO<sub>2</sub>  
22 vertical column ( $V_r$ ) over a polluted location (e.g., over a city) can be retrieved from the total  
23 NO<sub>2</sub> vertical column by removing the total NO<sub>2</sub> vertical column over a clean site (e.g., Pacific  
24 ocean) at a given location with the same latitude. Here, we simply removed a constant  
25 stratospheric NO<sub>2</sub> vertical column from the total vertical column. The stratospheric NO<sub>2</sub>



1vertical column was calculated as the integral of the NO<sub>2</sub> profile from 12 km to the top of  
2atmosphere (TOA). Accordingly, the three profiles displayed in Fig. 1 correspond to three  
3tropospheric NO<sub>2</sub> conditions defined as: (1) low polluted with  $V_{tr} = 0.43 \cdot 10^{15}$  molec/cm<sup>2</sup> (full  
4line), (2) moderately polluted with  $V_{tr} = 2.81 \cdot 10^{15}$  molec/cm<sup>2</sup> (dotted line) and, (3) heavily  
5polluted with  $V_{tr} = 9.36 \cdot 10^{15}$  molec/cm<sup>2</sup> (dotted-dashed line).

6 The modelling results presented in this paper are performed for single values of the  
7surface albedo  $A = 0.05$ , the solar zenith angle  $\theta_s = 30^\circ$  and the viewing zenith angle  $\theta_v = 0^\circ$   
8(according to OMI nadir viewing geometry). The influence of the solar angle and the surface  
9albedo are discussed in section 3. We note in passing that aerosols were chosen to be not  
10included in the atmospheric model.

11

## 122.2 DOAS retrieval

13 The tropospheric NO<sub>2</sub> vertical column ( $V_{tr}$ ) retrieval algorithm has been developed in the  
14framework of SCIATRAN [29]. SCIATRAN is both a radiative transfer code and a retrieval  
15algorithm for many atmospheric gases (see <http://www.iup.uni-bremen.de/sciatran/> for full  
16description of SCIATRAN). For the retrievals of  $V_{tr}$ , the DOAS technique is applied to  
17simulated TOA reflectance in the OMI-like band with a spectral sampling of 0.21 nm [30] and  
18with a signal to noise ratio (SNR) of 1400 [10]. Retrievals of  $V_{tr}$  are performed by converting  
19the simulated TOA reflectances  $R_{TOA}$  to so-called differential optical densities  $D$ , which are  
20calculated as

21

$$22 \quad D(\lambda) = \ln(R_{TOA}(\lambda)) - P_3(\lambda), \quad (1)$$

23

24where  $P_3(\lambda)$  is a 3<sup>rd</sup> degree least squares polynomial fit of the logarithm of  $R_{TOA}$  with respect to  
25the wavelength  $\lambda$ , that removes slowly varying functions. The conversion of  $R_{TOA}$  into  $D$

1 ensures the better contrast of the NO<sub>2</sub> absorption line depths to improve the accuracy of the  
2 fitting procedure. Then, in the retrieval algorithm the quadratic form

3

$$4 \quad F(\lambda, V_{tr,ret}) = \|D_{true}(\lambda, V_{tr,true}) - D_{ret}(\lambda, V_{tr,ret})\|^2, \quad (2)$$

5

6 is minimized with respect to unknown parameter  $V_{tr,ret}$ .  $D_{true}$  is the differential optical density  
7 spectrum simulated with SCIATRAN for the true tropospheric NO<sub>2</sub> column  $V_{tr,true}$ .  $D_{ret}$  is the  
8 retrieved differential optical density spectrum for the retrieved tropospheric NO<sub>2</sub> column  $V_{tr,ret}$ .

9

### 10 2.3 Cirrus cloud model

11 Cirrus clouds are treated as single layer homogeneous clouds. The most important cirrus  
12 cloud parameters that potentially influence the trace gas retrieval are those which determine  
13 the photon paths in the atmosphere. In the visible spectral range, these parameters are  
14 expected to be the cloud optical depth  $\tau$ , the cloud phase function  $P$ , the cloud top height  $z$  and  
15 the cloud geometrical depth  $h$  [31]. These parameters are used as inputs in SCIATRAN.  
16 Because of the quite large spatial resolution of OMI measurement (13×24 km<sup>2</sup>), one more  
17 important parameter is the geometric cloud fraction  $c$ . In most retrieval,  $c$  is accounted for  
18 considering the independent pixel approximation (IPA). IPA consists in the hypothesis that  
19  $R_{TOA}$  is the sum of the reflectance of a cloudy part ( $R_{cloudy}$ ) and the reflectance of a clear-sky  
20 part ( $R_{clear}$ ) as

21

$$22 \quad R_{TOA} = cR_{cloudy} + (1 - c)R_{clear} \quad (3)$$

23

24 The ice crystals phase function used for SCIATRAN simulations has been retrieved from in  
25 situ aircraft measurements during the Cirrus Cloud Experiment (CIRCLE2) campaign. The

1phase function (Fig. 2) was inferred from the Polar Nephelometer (PN) measurements. The  
2best fit of the PN measurements was achieved using a combination of spherical ice particles  
3with diameters ranging from 1 to 100  $\mu\text{m}$  and rough droxtal shaped particules with maximum  
4dimension between 2 and 200  $\mu\text{m}$ . Accordingly, two particle size distributions were retrieved  
5using the iterative inversion method developed by Oshchepkov et al. [32] and upgraded by  
6Jourdan et al. [33]. Then, the scattering patterns of the retrieved particle size distribution were  
7computed with the Lorenz-Mie theory (for spherical particles) and with an improved  
8geometric-optics model (for droxtal particles) at a wavelength of 420 nm [34]. The retrieved  
9phase function exhibits a featureless behaviour and is flat at side scattering angles which is in  
10accordance with most of the observations [32-33,35-37] or scientific recommendations in ice  
11cloud remote sensing application [38-41]. The corresponding asymmetry factor  $g$  is 0.75.

12 To illustrate the influence of cirrus clouds on the retrieval of  $V_{tr}$ , Fig. 3 shows simulated  
13differential optical densities (calculated with Eqs. 1 and 3) with and without cirrus clouds.  
14 $R_{clear}$  is calculated using SCIATRAN with standard profiles of  $\text{O}_3$ ,  $\text{H}_2\text{O}$  and  $\text{O}_2$ , surface albedo  
15and geometries described in section 2.1. Clouds properties ( $\tau = 1$ , CIRCLE2 phase function,  $z$   
16= 10 km and  $h = 1$  km) are added in the calculation of  $R_{cloudy}$ . The heavily polluted  $\text{NO}_2$  profile  
17is considered. The clear-sky differential optical density ( $c = 0$  in Eq. 3) is represented by the  
18green line. Blue and red lines represent differential optical densities when a cirrus cloud is  
19included and cover the full pixel ( $c = 1$ ) and half of the pixel ( $c = 0.5$ ), respectively. When a  
20cirrus cloud is included, the depths of  $\text{NO}_2$  absorption lines are reduced. This stems from the  
21cloud shielding effect discussed in the introduction, which is greater to both cloud albedo and  
22in-cloud absorption effects. Because  $V_{tr}$  varies with the  $\text{NO}_2$  absorption lines depths, the  
23presence of undetected cirrus would lead to a lower apparent  $V_{tr}$ , that depends on the cloud  
24fraction. Nevertheless, these results are valid for algorithms that do not make any attempt to  
25correct for clouds in the first place. In the current available OMI  $\text{NO}_2$  algorithms such as the

1 OMI standard retrieval (bucsela et al, 2006) and the Dutch OMI NO<sub>2</sub> (DOMINO) retrieval  
2 (Boersma et al. 2008b), the missing tropospheric NO<sub>2</sub> column due to clouds is compensated  
3 by adding a ghost column obtained from chemical transport model. In situations of cirrus, the  
4 OMI O<sub>2</sub>-O<sub>2</sub> algorithm will retrieve a small value for effective cloud fraction and high cloud  
5 height. Because of the small effective cloud fraction, the ghost column effect will also be  
6 small.

7

8

9

10

### 113. **Influence of undetected cirrus clouds on NO<sub>2</sub> retrieval**

12

13 The differential optical densities simulated previously are used to introduce the following  
14 discussion that focuses on the influence of undetected cirrus cloud on the tropospheric NO<sub>2</sub>  
15 column retrieval. The influence of undetected cirrus clouds is evaluated as an error on the  
16 retrieved tropospheric NO<sub>2</sub> column  $V_{tr,ret}$  compared to a true tropospheric NO<sub>2</sub> column  $V_{tr,true}$  :

17

$$18 \quad Error = \frac{V_{tr,ret} - V_{tr,true}}{V_{tr,true}} \times 100, \quad (4)$$

19

20  $V_{tr,ret}$  is retrieved according to the following methodology: A true TOA reflectance spectrum is  
21 calculated following Eq. 3, for an atmosphere characterized by both a cirrus cloud with  
22 prescribed properties and a true tropospheric NO<sub>2</sub> column  $V_{tr,true}$ . Then, this TOA reflectance is  
23 used as the input spectrum for the DOAS-based SCIATRAN retrieval algorithm. The  
24 retrieved quantity is, in that case, called  $V_{tr,ret}$ . It is important to point out that the inversion

1 procedure is performed with the hypothesis of a non cloudy atmosphere. By this mean, errors  
2 caused by a specific cirrus parameter (optical depth, cloud top height, etc...) can be quantified  
3 and used to determine the relative importance of the impact of a given cloud parameter on the  
4 retrieval of tropospheric NO<sub>2</sub> columns for retrievals that do not correct for clouds in the first  
5 place.

6

### 7 3.1 Cloud fraction and cloud optical depth

8 The panel of Fig. 4 displays errors of the retrieved  $V_{tr,ret}$  as a function of cloud optical  
9 depth and cloud fraction for low polluted condition (Fig. 4a), for moderately polluted  
10 condition (Fig. 4b) and for heavily polluted condition (Fig. 4c). For these cases,  $z = 10$  km,  $h$   
11 = 1 km and  $g = 0.75$  (i.e., with the CIRCLE2 ice crystal phase function). The overall negative  
12 error (underestimation) highlights the predominance of the shielding effect. This is explained  
13 by the fact that the tropospheric NO<sub>2</sub> is mainly situated under the cirrus cloud. Even for the  
14 highest value of the cloud optical depth considered in this study ( $\tau = 3$ ), the albedo effect still  
15 remains negligible compared to the shielding effect. The underestimation of  $V_{tr,ret}$  increases  
16 rapidly with both  $\tau$  and  $c$  and can reach, for  $\tau = 3$  and  $c = 1$ , an underestimation of ~35% in  
17 low polluted conditions (Fig. 4a), ~45% in moderately polluted conditions (Fig. 4b) and  
18 ~55% in heavily polluted condition (Fig. 4c). It is interesting to notice that the  
19 underestimation remains less than 5% for optical depth less than 0.2 whatever the cloud  
20 fraction and the polluted conditions are. This implies, from our simulations, that optically thin  
21 cirrus or subvisible cirrus do not significantly influence the NO<sub>2</sub> tropospheric column retrieval  
22 which is not the case for example, in atmospheric CO<sub>2</sub> retrieval [42,43]. This might be  
23 explained by the fact that, in the NO<sub>2</sub> absorption band, the signal is predominantly due to  
24 Rayleigh scattering as compared to CO<sub>2</sub> absorption band that is in the near infrared spectral  
25 band and where the Rayleigh scattering is very low.

1

### 23.2 Cloud top height and cloud geometrical depth

3 The influence of cloud top height ( $z$ ) and cloud geometrical depth ( $h$ ) is evaluated in term  
4of error difference between the maximum and the minimum  $\text{NO}_2$  tropospheric column errors  
5over the different values of  $z$  and  $h$  considered. Fig. 5 displays the error caused by undetected  
6cirrus clouds versus cloud top height  $z$ . In these simulations,  $g = 0.75$  and  $h = 1$  km. Colour  
7lines and symbols differ with pollution conditions, cloud fraction and cloud optical depth. For  
8 $c = 1$  and  $\tau = 3$  in low polluted condition (represented by the green line with triangles), the  
9error is -29% at  $z = 8$  km and -37% at  $z = 15$  km leading to an error difference of 8% between  
10 $z = 8$  km and  $z = 15$  km. As comparison, the error difference is ~6% for moderately polluted  
11condition (represented by the blue line with triangles) and ~4% for heavily polluted condition  
12(represented by the red line with triangles). The underestimation increase with cloud top  
13height mostly between  $z = 8$  to  $z = 12$  km. This is explained by the increase of shielding effect  
14with the altitude of the cirrus cloud. For  $c = 0.5$  and  $\tau = 1$  (represented by lines with  
15diamonds), error differences are almost constant with  $z$ . Table 1 provides a summary of error  
16differences for different values of  $c$  and  $\tau$ . Errors remain less than 1% for low optical depth ( $\tau$   
17= 0.05) or for low cloud fraction ( $c = 0.05$ ). We noticed also that, for fixed  $c$  and  $\tau$ , error  
18differences decrease with polluted conditions, which can be explained by an increase of the  
19in-cloud absorption effect when the cloud is lower due to higher tropospheric  $\text{NO}_2$  (see Fig.  
201). The error caused by undetected cirrus clouds versus cloud geometrical depth  $h$  is  
21represented in Fig. 6 for the same cases as Fig. 5. Underestimation of  $V_r$  decrease slowly with  
22 $h$  due to increase of in-cloud absorption leading to error difference of few percents between  $h$   
23= 0.1 km and  $h = 2$  km. Table 2 provides a summary of error differences for different values  
24of  $c$  and  $\tau$  which, overall, remain less than 3%.

25

### 13.3 Asymmetry factor of ice crystal phase function

2 The influence of ice crystal phase function is evaluated by comparing the retrieval errors  
3 versus the asymmetry factor  $g$ .  $g$  is used to parameterize the general behaviour of the ice  
4 crystal phase function because it is an integrated optical parameter (scalar) taking into account  
5 both ice crystals shape and size variability. The different values of asymmetry factor tested  
6 here are obtained on the basis of nine pre-calculated phase functions for different ice crystal  
7 shape (aggregate, plate, column, bullet and dendrite) and effective diameters comprise  
8 between 4.5 and 150  $\mu\text{m}$  that are used for cirrus clouds modelling [44]. The CIRCLE-2 phase  
9 function is included as well. The asymmetry factor values obtained are between 0.70 and  
10 0.85. Errors caused by undetected cirrus clouds versus asymmetry factors are displayed in  
11 Fig. 7. It appears that the underestimation decreases with the asymmetry factor. This is  
12 explained by the fact that cirrus cloud particles with a large value of  $g$  reflect less solar  
13 radiation than ice crystals with smaller asymmetry factor. Then, for a large value of  $g$ , more  
14 solar radiation exhibited by the cirrus cloud will interact with tropospheric  $\text{NO}_2$  located below  
15 the cloud. From the summary given in Table 3, error differences between  $g = 0.70$  and  $g =$   
16 0.85 are comprise between 0.07 % (for  $c = 0.05$ ,  $\tau = 0.05$  and low polluted condition) and  
17 1.3 % (for  $c = 1$ ,  $\tau = 3$  and heavily polluted condition). These results imply that, for fixed  $c$   
18 and  $\tau$ , the error difference caused by undetected cirrus clouds with  $g$  lying between 0.70 and  
19 0.85 (Table 3) is more important than the one caused by undetected cirrus clouds with  $z$   
20 comprise between 8 and 15 km (Table 1) or  $h$  comprise between 0.1 km and 3 km (Table 2).  
21 The asymmetry factor appears to be a major source of error in tropospheric  $\text{NO}_2$  retrieval in  
22 the presence of cirrus.

23

24 This sensitivity study showed that four cirrus parameters influence the retrieval of  
25 tropospheric  $\text{NO}_2$  columns. These parameters are, in descending order, the cloud fraction  $c$ ,

1the cloud optical depth  $\tau$ , the asymmetry factor  $g$  and the cloud top height  $z$ . Since the error  
2difference caused by cloud geometrical depth variability remains less than 3%, we have  
3considered that cloud geometrical depth does not influence significantly the retrieval of  
4tropospheric  $\text{NO}_2$  columns. The  $V_{tr,ret}$  errors have also been estimated for other solar zenith  
5angles ( $15^\circ$ ,  $45^\circ$  and  $60^\circ$ ) and other surface albedos (0.03 and 0.1) but they do not show large  
6discrepancies with the results obtained for a solar zenith angle of  $30^\circ$  and a surface albedo of  
70.05.

8

9

10

11

12

#### 134. **Error on $\text{NO}_2$ retrieval caused by uncertainties in cirrus clouds** 14**properties**

15

16 The next step of our study is to look at the  $V_{tr,ret}$  error induced by uncertainties of the cirrus  
17parameters that were identified previously as critical. The objective here is to evaluate the  
18precision needed for these parameters in order to constrain the retrievals. To this respect, we  
19simulated a true  $R_{TOA}$  using Eq. (3) with a set of cirrus properties ( $\tau$ ,  $g$ ,  $z$ ,  $h$ ), a cloud fraction  $c$   
20and a true tropospheric  $\text{NO}_2$  vertical column  $V_{tr,true}$ . Then,  $V_{tr,ret}$  is obtained by using as the  
21input reflectance spectrum for the inversion algorithm, the clear-sky part of the TOA  
22reflectance  $R_{clear}$ .  $R_{clear}$  is a function of  $R_{TOA}(p)$  and  $R_{cloud}(p+\delta p)$  where  $p$  is  $\tau$ ,  $g$ ,  $z$ ,  $h$  or  $c$ . The  
23formulation of  $R_{clear}$  is presented for each cloud parameters in the following subsections.



1 Results are presented for moderately polluted condition as results found for the three polluted  
2 conditions did not differ significantly.

3

#### 44.1 Cloud fraction

5 The error on tropospheric NO<sub>2</sub> column retrievals caused by uncertainty in cloud fraction is  
6 calculated as Eq. (4). The input reflectance spectrum of the inversion scheme is defined as:

7

$$8 \quad R_{clear} = \frac{R_{TOA}(\tau, g, z, h, c) - (c + \delta c)R_{cloud}(\tau, g, z, h)}{1 - (c + \delta c)}, \quad (5)$$

9

10 where  $\delta c$  is the uncertainty on cloud fraction. The simulations were performed for a cirrus  
11 cloud characterized by an asymmetry factor of 0.75, a cloud top height of 10 km and a  
12 geometrical thickness of 1 km. The surface albedo, solar zenith angle and the viewing angle  
13 are the same as in Section 3. Fig. 8 displays errors on  $V_{tr,ret}$  versus cloud optical depth for  $\delta c =$   
14 0.01 (Fig. 8a),  $\delta c = 0.05$  (Fig. 8b) and  $\delta c = 0.09$  (Fig. 8c) and for three values of  $c$  (0.1, 0.5  
15 and 0.8) representing low, medium and high cloud fractions. Overall, the overestimation of  
16 cloud fraction results in an overestimation of tropospheric NO<sub>2</sub> columns (i.e., positive errors).  
17 If  $c \leq 0.5$  (represented by lines with triangles or diamonds), overestimations are expected to  
18 be less than 10 % if  $\delta c \leq 0.05$  (Figs. 8a and Fig. 8b) and less than 20 % if  $\delta c = 0.09$  (Fig. 8c).  
19 If  $c \geq 0.8$ , overestimations are expected to be higher, ranging from 10% to more than 100%, if  
20  $\delta c \geq 0.05$  (Figs. 8b and 8c) for large cloud optical depth ( $\tau > 1$ ).

21 Within the framework of combining measurements performed by A-train sensors to  
22 improve OMI NO<sub>2</sub> products, cloud fraction can be derived from independent measurements.  
23 For example, MODIS can provide a subpixel cloud fraction information with a 1×1 km<sup>2</sup>  
24 resolution. Accordingly, the cloud fraction estimation from MODIS within an OMI pixel is  
25 expected to be achieved with lower uncertainty than the value of 0.05 reported by

1 Koelemeijer et al. [45]. However, the estimation of the uncertainty on MODIS cloud fraction  
 2 is a difficult task because it depends on the cloud type. Koren et al. [46] reported that for  
 3 small cumulus cloud, MODIS cloud fraction is almost twice as the cloud fraction obtained  
 4 from a finer spatial resolution (30 m) instrument. Fortunately, cirrus clouds have larger spatial  
 5 extension than small cumulus clouds and would be less influenced by the scale dependence.  
 6 Finally, these results highlight that cloud fraction within an OMI pixel should be determined  
 7 with uncertainty lower than 0.05 in order to reduce the tropospheric NO<sub>2</sub> vertical columns  
 8 errors, especially if the cloud optical depth is greater than 1.

9

#### 10 4.2 Cloud optical depth

11 The error on tropospheric NO<sub>2</sub> columns retrieval caused by uncertainties in the cloud  
 12 optical depth is calculated as Eq. (4) where the retrieval is applied on the clear-sky part of the  
 13 reflectance given by

14

$$15 \quad R_{clear} = \frac{R_{TOA}(\tau, g, z, h, c) - cR_{cloud}(\tau + \delta\tau, g, z, h)}{1 - c}, \quad (6)$$

16

17 where  $\delta\tau$  is the uncertainty in cloud optical depth. The simulations were performed for a cirrus  
 18 cloud characterized by  $g = 0.75$ ,  $z = 10$  km and  $h = 1$  km. The panel of Fig. 9 displays errors  
 19 on  $V_r$  versus cloud optical depth for  $\delta\tau/\tau = 10$  % (Fig. 9a),  $\delta\tau/\tau = 20$  % (Fig. 9b) and  $\delta\tau/\tau = 50$   
 20 % (Fig. 9c) and for three values of  $c$  (0.1, 0.5 and 0.8, differentiated by symbols). Here again,  
 21 the overestimation of cloud optical depth results in an overestimation of tropospheric NO<sub>2</sub>  
 22 columns. This is explained by the fact that an overestimation of  $\tau$  will increase the cloud  
 23 reflectance. The numerator of Eq. (6) will then decrease leading to an overestimation of  
 24 tropospheric NO<sub>2</sub> columns. For  $c \leq 0.5$ , overestimations are expected to be less than 15 % if

1  $\delta\tau/\tau \leq 20\%$  (Figs. 9a and 9b). The same result is seen if  $\delta\tau/\tau = 50\%$  and  $\tau < 1$  (Fig. 9c). If  $c =$   
2 20.8, overestimations are expected to increase quickly, especially if  $\delta\tau/\tau = 50\%$  (Fig. 9c).

3 By comparison, cirrus optical depth from MODIS and ground-based measurement has  
4 shown that MODIS overestimates the optical depth by 30% [47]. However, clouds with low  
5 optical depth (less than 0.4) are not retrieved from MODIS [48]. The use of CALIOP data will  
6 be helpful for a better characterisation of the optical depth since CALIOP is able to detect  
7 clouds with  $\tau < 0.05$  [49]. Nevertheless, the use of CALIOP information is limited by the fact  
8 that CALIOP only overlaps with the nadir pixel of OMI. If no CALIOP data is available and  
9 the cirrus cloud is undetected by MODIS (ie,  $\tau < 0.4$ ), tropospheric NO<sub>2</sub> vertical column  
10 errors are expected to be less than 10% (Fig. 4).

11

12

13

#### 14 4.3 Asymmetry factor

15 The error on tropospheric NO<sub>2</sub> columns retrieval caused by uncertainties in asymmetry factor  
16 is retrieved from the clear-sky part of the reflectance given by Eq. (6) and where the  
17 uncertainty is applied to  $g$  instead of  $\tau$ . Here again,  $g$  was chosen instead of the phase  
18 function because it is a convenient integrated parameter function of the ice particle shape and  
19 effective size. So it is a first guess indicator of the general scattering behaviour of the cirrus  
20 cloud. Fig. 10 displays errors versus cloud optical depth.  $R_{TOA}$  is simulated with  $g = 0.75$ ,  $z =$   
21 110 km and  $h = 1$  km. Here, the overestimation of asymmetry factor results in an  
22 underestimation of tropospheric NO<sub>2</sub> columns (i.e., negative errors). Underestimations up to  
23 40% are expected if  $c = 0.5$  (full lines) and up to 60% if  $c = 0.8$  (dashed lines), but do not  
24 change that much whether  $\delta g = 0.05$  (diamonds) or  $\delta g = 0.1$  (triangles). These results show  
25 that the uncertainty of the asymmetry factor is a major source of error in tropospheric NO<sub>2</sub>

1 retrieval in the presence of cirrus. However, although modelling studies show that for ice  
2 crystals values of  $g$  could vary from 0.7 to 0.9 (see for instance the review paper of Baran  
3 [2009]), measurements do not show such variability in ice clouds. An uncertainty less than  
4 0.05 can be expected since the asymmetry factor ranges more likely from 0.75 to 0.80 for ice  
5 clouds and from 0.80 to 0.85 for mixed phase clouds depending on the liquid water fraction  
6 [50,51]. However, the operational algorithms of MODIS retrieve the ice crystals effective  
7 radius ( $R_{eff}$ ) and not  $g$ . Both information on  $R_{eff}$  and shape are needed to model the scattering  
8 properties of ice crystals. Usually, a combination of particles with different shapes and sizes is  
9 used as an equivalent microphysical model in MODIS retrieval process of the effective radius.  
10 The retrieved  $R_{eff}$  from MODIS corresponding to a specific microphysical shape model could  
11 be used to assess appropriate phase function and asymmetry parameter on the basis of  
12 MODIS Look-Up-Tables (LUT) [52,53]. However, MODIS  $R_{eff}$  seems to be overestimated by  
13 comparison with Lidar retrievals [54].

#### 14 4.4 Cloud top height

15 The error on tropospheric  $\text{NO}_2$  column retrievals caused by uncertainties in the cloud top  
16 height is assumed from the clear-sky part of the reflectance given by Eq. (6) and where the  
17 uncertainty is applied on  $z$  instead of  $\tau$ . The panel of Fig. 11 displays errors on  $V_{tr}$  versus  
18 cloud optical depth for  $\delta z = 0.5$  km (Fig. 11a),  $\delta z = 1$  km (Fig. 11b) and  $\delta z = 3$  km (Fig. 11c)  
19 and for two values of  $c$  (0.5 and 0.8) and three values of  $z$  (8, 10 and 12 km). If  $\delta z \leq 1$  km  
20 (Figs. 11a and 11b), errors are expected to be less than 20 %. If  $\delta z = 3$  km (Fig. 11c), errors  
21 are expected to be less than 20 % if  $c \leq 0.5$ . Uncertainty of  $z$  retrieved from MODIS is  
22 currently under investigation but preliminary results by comparison with Lidar measurements  
23 show difference on the order of 1 km [54]. However, this comparison remains limited due to  
24 the high occurrence of multi-layers ice clouds where MODIS only retrieves an effective cloud

1top height. The use of CALIOP retrieval will allow constraining the cloud top height input in  
2order to retrieve the tropospheric NO<sub>2</sub> column in presence of cirrus cloud.

3

4

## 55. **Contribution of subpixel cloud optical depth**

6

7 The IPA formulation of the TOA reflectance in presence of clouds given by Eq. (3) does  
8not consider the subpixel inhomogeneity of cloud properties. To this respect, we can consider  
9that the optical thickness can be derived more accurately from high spatial resolution  
10measurements (1×1 km<sup>2</sup>) of MODIS. This information will be used to describe the variability  
11of the optical thickness within an OMI pixel (13×24 km<sup>2</sup>). Therefore, two ways of  
12determining the contribution of the cloud to the total reflectance measured by OMI can be  
13considered: (1) by calculating  $R_{cloud}$  from the mean cloud optical depth  $\langle \tau \rangle$  or (2) by  
14calculating the mean cirrus cloud reflectance  $\langle R_{cloud} \rangle$  corresponding to a distribution of  
15optical depth within an OMI pixel. These two ways of calculating the contribution of the  
16cloud would have an important impact on the computing time of the retrieval because in the  
17first way, there is only one calculation for the mean cloud optical depth whereas in the second  
18way there are several calculations for each value of the subpixel cloud optical depth  
19distributions. To compare these two ways, we have simulated different distribution of cloud  
20optical depth considering a gamma distribution. Nine distributions have been selected  
21randomly with three mean cloud optical depths ( $\langle \tau \rangle = 0.5, 1$  or  $2$ ) and three variances ( $\nu_\tau =$   
220.1, 1 or 2). In these distributions, 312 values have been considered representing the  
23maximum total number of MODIS subpixels within an OMI pixel at nadir. Cloud fractions  
24have been included in the study by considering 31 subpixels for  $c = 0.1$ , 62 subpixels for  $c =$   
250.2, etc... to 281 subpixels for  $c = 0.9$ . For the cloud simulations we have used  $z = 10$  km,  $h =$

11 km and  $g = 0.75$ . The influence of the solar zenith angle has been tested by considering two values,  $\theta_s = 30^\circ$  and  $\theta_s = 60^\circ$ . Figure 12 illustrates the tropospheric  $\text{NO}_2$  column error retrieved from the clear-sky part of the reflectance given by

4

$$5 \quad R_{clear} = \frac{\langle R_{TOA} \rangle - c R_{cloud}(\langle \tau \rangle, g, z, h)}{1 - c}, \quad (7)$$

6

7 where  $\langle R_{TOA} \rangle$  is the mean TOA reflectance calculated as the average of TOA reflectances  
 8 calculated using Eq. (3) for the different values of the cloud optical depth distribution. The  
 9 tropospheric  $\text{NO}_2$  column error resulting from the use of a mean cloud optical depth is either  
 10 positive or negative. The error remains negative for  $\langle \tau \rangle = 0.5$  (dotted-dashed lines) and  
 11 remains positive for  $\langle \tau \rangle = 2$  (dashed lines). Interestingly, the error is negative for  $\langle \tau \rangle = 1$   
 12 (full lines) if  $\theta_s = 30^\circ$  and positive if  $\theta_s = 60^\circ$  showing the importance of the solar zenith angle  
 13 in the subpixel inhomogeneity consideration. If  $\theta_s = 30^\circ$  or  $60^\circ$ , errors caused by the use of a  
 14 mean cloud optical depth are expected to be less than  $\pm 15\%$  for  $c \leq 0.5$ . For cloud fractions  
 15 greater than 0.5, errors increase quickly with cloud fraction and strongly with variances and  
 16 cloud optical depth. The increase of errors with cloud fraction is explained by the  
 17 denominator  $1-c$  in Eq. (7). The blow-up of errors at  $\text{SZA}=60^\circ$  is explained by the bias  
 18 between  $R_{cloud}$  calculated with the distribution of subpixels cloud optical depth and calculated  
 19 with the mean cloud optical depth. This bias increases with the variability of cloud optical  
 20 depths and solar zenith angle [55].

21 Finally, when the cloud fraction is lower than 0.5, the subpixel variability of the cloud optical  
 22 depth does not need to be included in the cloud correction scheme. Otherwise, the subpixel  
 23 variability of the cloud optical depth is significant and should be considered in the cloud  
 24 correction scheme. Such information can be deduced from MODIS observations.

25

1

## 26. Conclusion

3

4 In this study, we have theoretically demonstrated the sensitivity of tropospheric NO<sub>2</sub>  
5column ( $V_{tr}$ ) retrieval to the presence of cirrus clouds. In order of importance, we concluded  
6that  $c$  (cloud fraction),  $\tau$  (cloud optical depth),  $g$  (asymmetry factor of ice crystal phase  
7function) and  $z$  (cloud top height) influence the retrieval of  $V_{tr}$ . It appears from our  
8simulations that  $V_{tr}$  is less influenced by  $h$  (cloud geometrical depth). The error on the  
9retrieved tropospheric NO<sub>2</sub> column depends also on the NO<sub>2</sub> profile. Because tropospheric  
10NO<sub>2</sub> is generally situated underneath the cirrus cloud, the shielding effect leads to  
11underestimating the tropospheric NO<sub>2</sub> columns for retrievals that do not correct for clouds in  
12the first place. The underestimation depends strongly on  $c$  and  $\tau$  ranging from few percents  
13for low cloud fraction to 55 % for  $c = 1$  in heavily polluted conditions (i.e. for  $V_{tr} = 9.36 \cdot 10^{15}$   
14molec/cm<sup>2</sup>). In the context of the A-Train constellation, the tropospheric NO<sub>2</sub> column in  
15presence of cirrus clouds can be retrieved by using the independent pixel approximation. The  
16MODIS sensor coupled with CALIOP should be able to describe the optical and physical  
17properties of the clouds in such way to reduce the error on the tropospheric NO<sub>2</sub> column. The  
18use of CALIOP information is however limited by the fact that CALIOP only overlaps with  
19the nadir pixel of OMI. Nevertheless, errors on cloud properties will end up by errors on  
20tropospheric NO<sub>2</sub> column retrieval. From our simulations, we have demonstrated that the error  
21will be reduced significantly if  $c \leq 0.5$  as opposed to a retrieval that does not correct for  
22cirrus. In the case of a cirrus cloud correction based on IPA,  $c$  and  $\tau$  would have to be known  
23within accuracy better than 0.05 and 50 %, respectively.  $z$  would have to be known within  
24accuracy of at least 1 km and  $g$  would have to be known within accuracy better than 0.05.  
25The latter result shows that the uncertainty of the asymmetry factor is a major source of error

1in tropospheric NO<sub>2</sub> retrieval in the presence of cirrus. Using the subpixel information from  
2MODIS cloud flag for  $c$ , from MODIS and CALIOP cloud products for  $\tau$  and  $z$  and from  
3MODIS  $R_{eff}$  for the phase function should be sufficient to reduce the error on tropospheric  
4NO<sub>2</sub> column retrieval. Under this circumstance, the use of a mean value of the cloud optical  
5depth from subpixel information as provided by MODIS should be enough to take into  
6account the cirrus cloud and speed up the retrieval significantly. If the cloud fraction is greater  
7than 0.5, the subpixel variability of the cloud optical depth has to be included in the cloud  
8correction scheme and can be obtained from MODIS. Moreover, differences between  
9radiative transfer models used for cloud retrievals and sensor's calibration issues would  
10introduce some discrepancies in the cloud correction scheme. The results in this study hold  
11for retrievals that do not attempt to correct for cirrus clouds. Current state-of-science  
12retrievals do correct for clouds that include a range of situations including cirrus clouds.  
13Nevertheless, the current NO<sub>2</sub> algorithm have not be evaluated in presence of cirrus. To  
14interpret our results in the context of current retrieval algorithms, the sensitivity for cirrus  
15clouds in the O<sub>2</sub>-O<sub>2</sub> (OMI) and O<sub>2</sub>-A (GOME(-2), SCIAMACHY) band based cloud retrievals  
16needs to be studied. Such studies could point out whether a correction for cirrus based on  
17concurrent A-Train cloud information would be useful

18

19

## 20**Acknowledgments**

21 The authors are grateful to Céline Cornet, Philippe Dubuisson and Nicolas Ferlay from  
22the Laboratoire d'Optique Atmosphérique (LOA) for fruitful discussions. Authors are also  
23grateful to Christophe Gourbeyre, Guillaume Mioche and Jean-François Gayet from the  
24Laboratoire de Météorologie Physique (LaMP) for providing data from the CIRCLE-2



1campaign. This research was supported by the Centre National d'Etudes Spatiales (CNES)  
2and the Institut National des Sciences de l'Univers (INSU/PNTS).

3

4

## 5References

- 6[1] Crutzen PJ. The Role of NO and NO<sub>2</sub> in the Chemistry of the Troposphere and  
7 Stratosphere. *Ann Rev Earth Planet Sci* 1979;7:443-472.
- 8[2] Solomon S, Portmann RW, Sanders RW, Daniel JS, Madsen W, Bartram B, Dutton EG.  
9 On the role of nitrogen dioxide in the absorption of solar radiation. *J Geophys Res*  
10 1999;104: D1012,047.
- 11[3] Burrows JP, Weber M, Buchwitz M, Rozanov V, Ladstätter-Weißenmayer A, Richter A,  
12 DeBeek R, Hoogen R, Bramstedt K, Eichmann KU, Eisinger M, Perner D. The Global  
13 Ozone Monitoring Experiment (GOME): Mission Concept and First Scientific Results. *J*  
14 *Atm Sci* 1999;56:151–175.
- 15[4] Bovensmann H, Burrows JP, Buchwitz M, Frerick J, Noël S, Rozanov VV, Chance K,  
16 Goede PH. SCIAMACHY: Mission Objectives and Measurement Modes *J Atm Sci*  
17 1999;56: 127–150.
- 18[5] Levelt PF, Hilsenrath E, Leppelmeier GW, Van den Oord GHJ, Bhartia PK, Tamminen J,  
19 de Haan JF, Veefkind JP. Science objectives of the ozone monitoring instrument. *IEEE*  
20 *Trans Geosci Remote Sensing* 2006;44:1199- 1208.
- 21[6] Platt U, Stutz J. *Differential Optical Absorption Spectroscopy (DOAS): Principles and*  
22 *Applications*. Heidelberg, Germany: Springer, 2006.
- 23[7] Beirle S, Platt U, Wenig M, Wagner T. Weekly cycle of NO<sub>2</sub> by GOME measurements: a  
24 signature of anthropogenic sources. *Atmos Chem Phys* 2003 ;3 :2225-2232.

- 1[8] Richter A, Burrows JP, Nüß H, Granier C, Niemeier U. Increase in tropospheric nitrogen  
2 dioxide over China observed from space. *Nature* 2005;437:129 – 132.
- 3[9] Martin RV, Sioris CE, Chance K, Ryerson TB, Bertram TH, Wooldridge PJ, Cohen RC,  
4 Neuman JA, Swanson A, Flocke FM. Evaluation of space-based constraints on global  
5 nitrogen oxide emissions with regional aircraft measurements over and downwind of  
6 eastern North America. *J Geophys Res* 2006;111:D15308.
- 7[10] Boersma KF, Jacob DJ, Eskes HJ, Pinder RW, Wang J, van der A RJ. Intercomparison of  
8 SCIAMACHY and OMI tropospheric NO<sub>2</sub> columns: Observing the diurnal evolution of  
9 chemistry and emissions from space. *J Geophys Res* 2008;113:D16S26.
- 10[11] Celarier EA, Brinksma EJ, Gleason JF, Veefkind JP, Cede A, Herman JR, Ionov D,  
11 Goutail F, Pommereau JP, Lambert JC, van Roozendael M, Pinardi G, Wittrock F,  
12 Schönhardt A, Richter A, Ibrahim OW, Wagner T, Bojkov B, Mount G, Spinei E, Chen  
13 CM, Pongetti TJ, Sander SP, Bucsela EJ, Wenig MO, Swart DPJ, Volten H, Kroon M,  
14 Levelt PF. Validation of Ozone Monitoring Instrument nitrogen dioxide columns. *J*  
15 *Geophys Res* 2008;113: D15S15.
- 16[12] Wenig MO, Cede AM, Bucsela EJ, Celarier A, Boersma KF, Veefkind JP, Brinksma EJ,  
17 Gleason JF, Herman JR. Validation of OMI tropospheric NO<sub>2</sub> column densities using  
18 direct-Sun mode Brewer measurements at NASA Goddard Space Flight Center. *J*  
19 *Geophys Res* 2008;113:D16S45.
- 20[13] van Noije TPC, Eskes HJ, Dentener FJ, Stevenson DS, Ellingsen K, Schultz MG,  
21 Wild O, Amann M, Atherton CS, Bergmann DJ, Bey I, Boersma KF, Butler T, Cofala J,  
22 Drevet J, Fiore AM, Gauss M, Hauglustaine DA, Horowitz LW, Isaksen ISA, Krol MC,  
23 Lamarque JF, Lawrence MG, Martin RV, Montanaro V, Müller JF, Pitari G, Prather MJ,  
24 Pyle JA, Richter A, Rodriguez JM, Savage NH, Strahan SE, Sudo K, Szopa S,

1 van Roozendael M. Multi-model ensemble simulations of tropospheric NO<sub>2</sub> compared  
2 with GOME retrievals for the year 2000. *Atmos Chem Phys* 2006;6:2943-2979.

3[14] Blond N, Boersma KF, Eskes HJ, van der A RJ, Van Roozendael M, De Smedt I,  
4 Bergametti G, Vautard R. Intercomparison of SCIAMACHY nitrogen dioxide  
5 observations, in situ measurements and air quality modeling results over Western Europe.  
6 *J Geophys Res* 2007;112:D10311.

7[15] Bucsela EJ, Perring AE, Cohen RC, Boersma KF . Celarier A . Gleason F, Wenig M O,  
8 Bertram TH, Wooldridge PJ, Dirksen R, Veefkind JP. Comparison of tropospheric NO<sub>2</sub>  
9 from in situ aircraft measurements with near-real-time and standard product data from  
10 OMI. *J Geophys Res* 2008;113:D16S31.

11[16] Boersma KF, Eskes HJ, Brinksma EJ. Error analysis for tropospheric NO<sub>2</sub> retrieval from  
12 space. *J Geophys Res* 2004;109:D04311.

13[17] Krijger JM, van Weele M, Aben I, Frey R. Technical Note: The effect of sensor  
14 resolution on the number of cloud-free observations from space. *Atmos Chem Phys*  
15 2007;7:2881-2891.

16[18] Koelemeijer R, Stammes P. Effects of clouds on ozone column retrieval from GOME  
17 UV measurements. *J Geophys Res* 1999;104:8281–8294.

18[19] Liu X, Newchurch MJ, Loughman R, Bhartia PK. Errors resulting from assuming opaque  
19 Lambertian clouds in TOMS ozone retrieval. *JQSRT* 2004;85:337-365.

20[20] Kokhanovsky AA, Mayer B, Rozanov VV, Wapler K, Lamsal LN, Weber M, Burrows J  
21 P, Schumann U. Satellite Ozone Retrieval Under Broken Cloud Conditions: An Error  
22 Analysis Based on Monte Carlo Simulations. *IEEE Trans Geosci Remote Sensing*  
23 2007;45:187-194.

24[21] Kokhanovsky AA, Rozanov VV. The uncertainties of satellite DOAS total ozone  
25 retrieval for a cloudy sky. *Atmos Res* 2008;87: 27-36.

- 1[22] Stammes P, Sneep M, de Haan JF, Veefkind JP, Wang P, Levelt PF. Effective cloud  
2 fractions from the Ozone Monitoring Instrument: Theoretical framework and validation. J  
3 Geophys Res 2008;113:D16S38.
- 4[23] Wang P, Stammes P, Boersma KF. Impact of Effective Cloud Fraction Assumption on  
5 Tropospheric NO<sub>2</sub> Retrievals. Proc of the Atmospheric Science Conference, 8-12 May  
6 2006 at ESRIN, Frascati Italy. Edited by H. Lacoste and L. Ouwehand. European Space  
7 Agency, 2006;SP-628:72.1.
- 8[24] Acarreta JR, De Haan JF, Stammes P. Cloud pressure retrieval using the O<sub>2</sub>-O<sub>2</sub>  
9 absorption band at 477 nm. J Geophys Res 2004;109:D05204.
- 10[25] van Diedenhoven B, Hasekamp OP, Landgraf J. Retrieval of cloud parameters from  
11 satellite-based reflectance measurements in the ultraviolet and the oxygen A-band. J  
12 Geophys Res 2007;112:D15208.
- 13[26] Platnick S, King MD, Ackerman SA, Menzel WP, Baum BA, Riedi JC, Frey RA. The  
14MODIS cloud products: algorithms and examples from Terra. IEEE Trans Geosci Remote  
15Sensing 2003;41:459- 473.
- 16[27] Wylie DP, Menzel WP, Woolf HM, Strabala KI. Four Years of Global Cirrus Cloud  
17 Statistics using HIRS. J Clim1994;7(12):1972-1986,.
- 18[28] Nazaryan H, McCormick MP, Menzel WP. Global characterization of cirrus clouds using  
19 CALIPSO data. J Geophys Res 2008;113:D16211.
- 20[29] Rozanov AA, Rozanov VV, Buchwitz M, Kokhanovsky AA, Burrows JP. SCIATRAN  
21 2.0-a new radiative transfer model for geophysical applications in the 175-2400 nm  
22 spectral range. Adv Space Res 2005;36:1015-1019.
- 23[30] Boersma KF, Eskes HJ, Veefkind JP, Brinksma EJ, van der A RJ, Sneep M,  
24 van den Oord GHJ, Levelt PF, Stammes P, Gleason JF, Bucsela EJ. Near-real time  
25 retrieval of tropospheric NO<sub>2</sub> from OMI. Atmos Chem Phys 2007;7:2103-2118.

- 1[31] Pfeilsticker K, Erle F, Funk O, Marquard L, Wagner T, Platt U. Optical path  
2 modifications due to tropospheric clouds: Implications for zenith sky measurements of  
3 stratospheric gases. *J Geophys Res* 1998;103(D19):25323–25335.
- 4[32] Oshchepkov S, Isaka, H, Gayet JF, Sinyuk A, Auriol F, Havemann S. Microphysical  
5 properties of mixed-phase and ice clouds retrieved from in situ airborne “Polar  
6 Nephelometer” measurements, *Geophys. Res. Lett.*, 27, 209-213, 2000
- 7[33] Jourdan O, Oshchepkov S, Shcherbakov V, Gayet JF, Isaka H. Assessment of cloud  
8 optical parameters in the solar region: Retrievals from airborne measurements of  
9 scattering phase functions. *J Geophys Res* 2003;108:D134572.
- 10[34] Yang P, Liou KN, Geometric-optics-integral-equation method for light scattering by  
11 nonspherical ice crystals. *Appl Opt* 1996;35:6568-6584.
- 12[35] Shcherbakov VN, Gayet JF, Jourdan O, Minikin A, Ström J, Petzold A. Assessment of  
13 Cirrus Cloud Optical and Microphysical Data Reliability by Applying Statistical  
14 Procedures. *J Atmos Ocean Technol* 2005;22:409-420.
- 15[36] Gayet JF, Shcherbakov VN, Mannstein H, Minikin A, Schumann U, Ström J, Petzold A  
16 Ovarlez J, Immler F. Microphysical and optical properties of midlatitude cirrus clouds  
17 observed in the southern hemisphere during INCA. *QJR Meteor Soc* 2006;132;2719-2748.
- 18[37] Francis PN, Foot JS, Baran AJ. Aircraft measurements of the solar and infrared radiative  
19 properties of cirrus and their dependence on ice crystal shape. *J Geophys Res*  
20 1999;104:31685-31696.
- 21[38] Labonnote LC, Brogniez G, Buriez JC, Doutriaux-Boucher M, Gayet JF, Macke A.  
22 Polarized light scattering by inhomogeneous hexagonal monocrystals : Validation with  
23 ADEOS-POLDER measurements. *J Geophys Res* 2001;106:12139-12154.

- 1[39] Baran AJ, Francis, PN. On the radiative properties of cirrus cloud at solar and thermal  
2 wavelengths: A test of model consistency using high-resolution airborne radiance  
3 measurements. QJR Meteorol Soc, 2004;130:763-778.
- 4[40] Baran AJ, Labonnote LC. On the reflection and polarisation properties of ice cloud.  
5 JQSRT, 2006;100:41-54.
- 6[41] Baran AJ, Labonnote LC. A self-consistent scattering model for cirrus. I: The solar  
7 region. QJR Meteorol Soc 2007;133:1899-1912.
- 8[42] Dufour E and Bréon FM. Spaceborne Estimate of Atmospheric CO<sub>2</sub> Column by Use of  
9 the Differential Absorption Method: Error Analysis. Appl Opt 2003;42:3595-3609.
- 10[43] Schneising O, Buchwitz M, Burrows JP, Bovensmann H, Reuter M, Notholt J,  
11 Macatangay R, Warneke T. Three years of greenhouse gas column-averaged dry air mole  
12 fractions retrieved from satellite – Part 1: Carbon dioxide. Atmos Chem Phys  
13 2008;8:3827-3853.
- 14[44] Yang P, Liou KN. Single-scattering properties of complex ice crystals in terrestrial  
15 atmosphere. Contr Atmos Phy 1998;71:223–248.
- 16 [45] Koelemeijer R, Stammes P, Hovenier J, de Haan J. A fast method for retrieval of cloud  
17 parameters using oxygen A band measurements from the Global Ozone Monitoring  
18 Experiment. J Geophys Res 2001;106(D4):3475-3490.
- 19[46] Koren I, Oreopoulos L, Feingold G, Remer LA, Altaratz O. How small is a small cloud?  
20 Atmos Chem Phys 2008;8:3855-3864.
- 21[47] Mace GG, Zhang Y, Platnick S, King MD, Minnis P, Yang P. Evaluation of Cirrus Cloud  
22 Properties Derived from MODIS Data Using Cloud Properties Derived from Ground-  
23 Based Observations Collected at the ARM SGP Site. J Appl Meteor 2005;44:221–240.
- 24[48] Ackerman SA, Holz RE, Frey R, Eloranta EW, Maddux BC, McGill M. Cloud Detection  
25 with MODIS. Part II: Validation. J Atmos Oceanic Technol 2008;25:1073–1086.

1[49] Winker DM, Pelon JR, McCormick MP. The CALIPSO mission: spaceborne lidar for  
2 observation of aerosols and clouds. Proc SPIE 2003;4893:1-11.

3[50] Garrett TJ, Hobbs P., Gerber H. Shortwave, single-scattering properties of arctic ice  
4 clouds. J Geophys Res 2001;106(D14);15155-15172.

5[51] Gayet JF, Asano S, Yamazaki A, Uchiyama A, Sinyuk A, Jourdan O, Auriol F. Two  
6 case studies of winter continental-type water and mixed-phase stratocumuli over the  
7 sea, Part I : Microphysical and optical properties. J Geophys Res  
8 2002;107(D21);doi:10.1029/2001JD001106.

9[52] Baum BA, Heymsfield AJ, Yang P, Bedka ST. Bulk scattering models for the remote  
10 sensing of ice clouds. Part 1: Microphysical data and models. J Appl Meteor  
11 2005;44 :1885-1895.

12[53] Baum BA, Yang P, Heymsfield AJ, Platnick S, King MD, Hu YX, Bedka ST. Bulk  
13 scattering models for the remote sensing of ice clouds. Part 2: Narrowband models. J Appl  
14 Meteor 2005;44:1896-1911.

15[54] Chiriaco M, Chepfer H, Minnis P, Haeffelin M, Platnick S, Baumgardner D,  
16 Dubuisson P, McGill M, Noël V, Pelon J, Spangenberg D, Sun-Mack S, Wind G.  
17 Comparison of CALIPSO-Like, LaRC, and MODIS Retrievals of Ice-Cloud Properties  
18 over SIRTa in France and Florida during CRYSTAL-FACE. J Appl Meteor Clim  
19 2007;46:249-272.

20[55] Cahalan RF, Ridgway W, Wiscombe WJ, Bell TL, Snider JB. The Albedo of Fractal  
21 Stratocumulus Clouds. J Atmos Sci 1994;51;2434–2455.

22Baran AJ. A review of the light scattering properties of cirrus, 2009 JQSRT 2009;110:1239-  
23 1260.

1 Kleipool Q. L., M. R. Dobber, J. F. de Haan, P. F. Levelt (2008), Earth surface reflectance  
2 climatology from 3 years of OMI data, *J. Geophys. Res.*, 113, D18308,  
3 doi:10.1029/2008JD010290.

4 Boersma, K. F., Dirksen, R., Veefkind, J. P., Eskes, H. J., and  
5 van der A, R. J.: Dutch OMI NO<sub>2</sub> (DOMINO) data product  
6 HE5 data file user manual, TEMIS website, [http://www.temis.](http://www.temis.nl/airpollution/no2.html)  
7 [nl/airpollution/no2.html](http://www.temis.nl/airpollution/no2.html), 2008b.



## 1Figure captions

2Figure 1. Atmospheric profiles of NO<sub>2</sub> Volume Mixing Ratio in ppbv used in the study.

3Figure 2. Cirrus cloud phase function inferred from aircraft measurement during the  
4CIRCLE2 campaign.

5Figure 3. Differential optical densities for heavily polluted troposphere without cloud (green  
6line), with  $c = 1$  (blue line) or  $c = 0.5$  (red line).

7Figure 4. NO<sub>2</sub> tropospheric error caused by undetected cirrus cloud as function of cloud  
8optical depth and cloud fraction. Here,  $z = 10$  km,  $h = 1$  km and  $g = 0.75$ .

9Figure 5. NO<sub>2</sub> tropospheric error caused by undetected cirrus cloud versus cloud top height.  
10Green, blue and red lines stand for low, moderately and heavily polluted conditions. Cloud  
11fraction and cloud optical depth were assumed to be 0.5 and 1, respectively for diamonds and  
12to be 1 and 3 for triangles. Here,  $h = 1$  km and  $g = 0.75$ .

13Figure 6. Same as Fig. 5 versus cloud geometrical depth. Here,  $z = 10$  km and  $g = 0.75$ .

14Figure 7. Same as Fig. 5 versus asymmetry factor. Here,  $z = 10$  km and  $h = 1$  km.

15Figure 8. NO<sub>2</sub> tropospheric column error caused by an uncertainty in the cloud fraction  $\delta c =$   
160.01 (a),  $\delta c = 0.05$  (b) and  $\delta c = 0.09$  (c) versus cloud optical depth and for different cloud  
17fraction ( $c = 0.1, 0.5$  and  $0.8$ , differentiated by symbols)., Here,  $z = 10$  km,  $h = 1$  km and  $g =$   
180.75 and moderately polluted condition is assumed.

19Figure 9. Same as Fig. 8 for  $\delta\tau/\tau = 10\%$  (a),  $\delta\tau/\tau = 20\%$  (b) and  $\delta\tau/\tau = 50\%$  (c).

20Figure 10. NO<sub>2</sub> tropospheric column error caused by an uncertainty in the asymmetry factor  
21 $\delta g = 0.05$  (diamonds) and  $\delta g = 0.1$  (triangles) versus cloud optical depth for  $c = 0.5$  (full lines)  
22and  $c = 0.8$  (dashed lines). Here,  $z = 10$  km,  $h = 1$  km and  $g = 0.75$  and moderate polluted  
23condition is assumed.

24Figure 11. NO<sub>2</sub> tropospheric column error caused by an uncertainty in the cloud top height  $\delta z$   
25= 0.5 km (a),  $\delta z = 1$  km (b) and  $\delta z = 3$  km (c) versus cloud optical depth and for  $c = 0.5$  (full

1lines),  $c = 0.8$  (dashed lines),  $z = 8$  km (diamonds) and  $z = 12$  km (triangles). Here,  $h = 1$  km  
2and  $g = 0.75$  and moderate polluted condition is assumed.

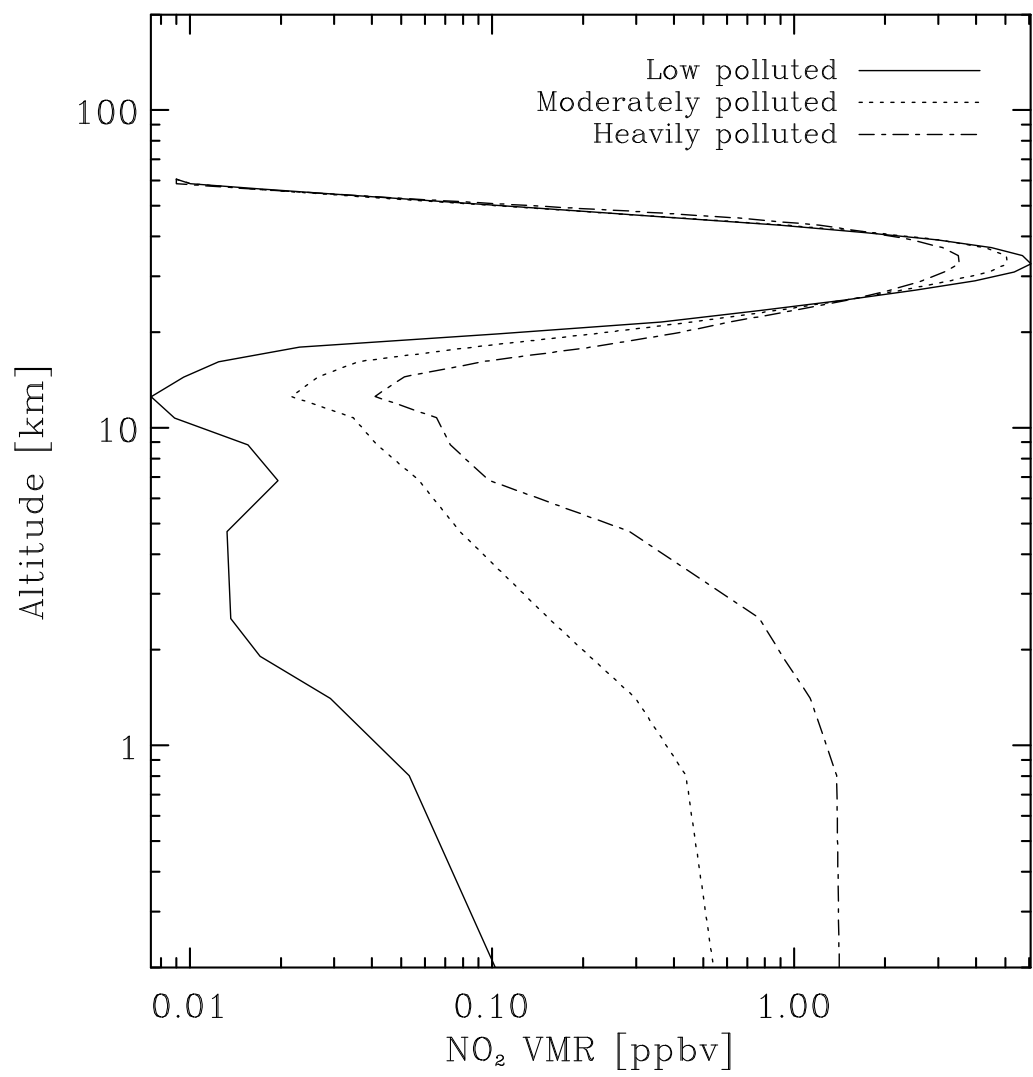
3Figure 12.  $\text{NO}_2$  tropospheric column error caused by the use of a mean cloud optical depth  
4instead of subpixels information versus cloud fraction if  $\theta_s = 30^\circ$  (left) and if  $\theta_s = 60^\circ$  (right).

## 1 Table captions

2 Table 1. NO<sub>2</sub> tropospheric error difference caused by cloud top height  $z$  variability. Values in  
3 each box of fixed  $c$  and  $\tau$  correspond to low, moderately and heavily polluted conditions,  
4 respectively.

5 Table 2. Same as Table 1 for cloud geometrical depth  $h$ .

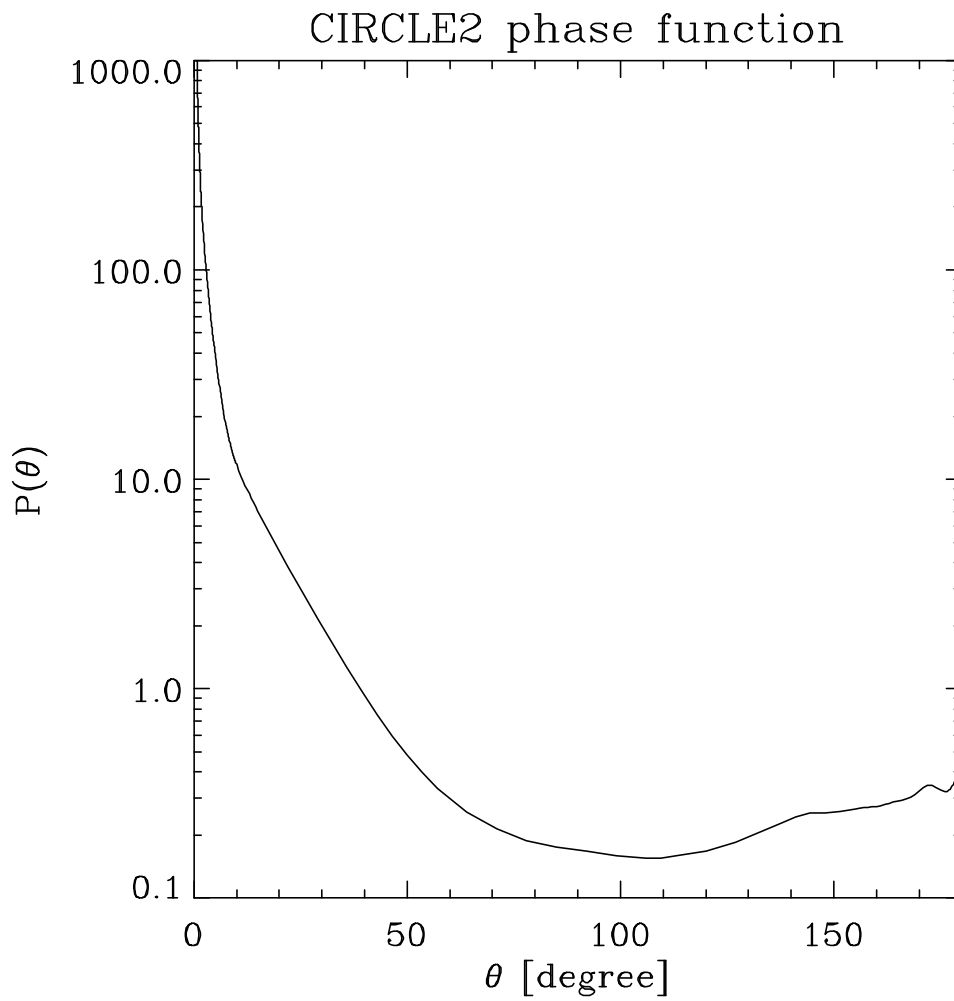
6 Table 3. Same as Table 1 for asymmetry factor  $g$ .



1

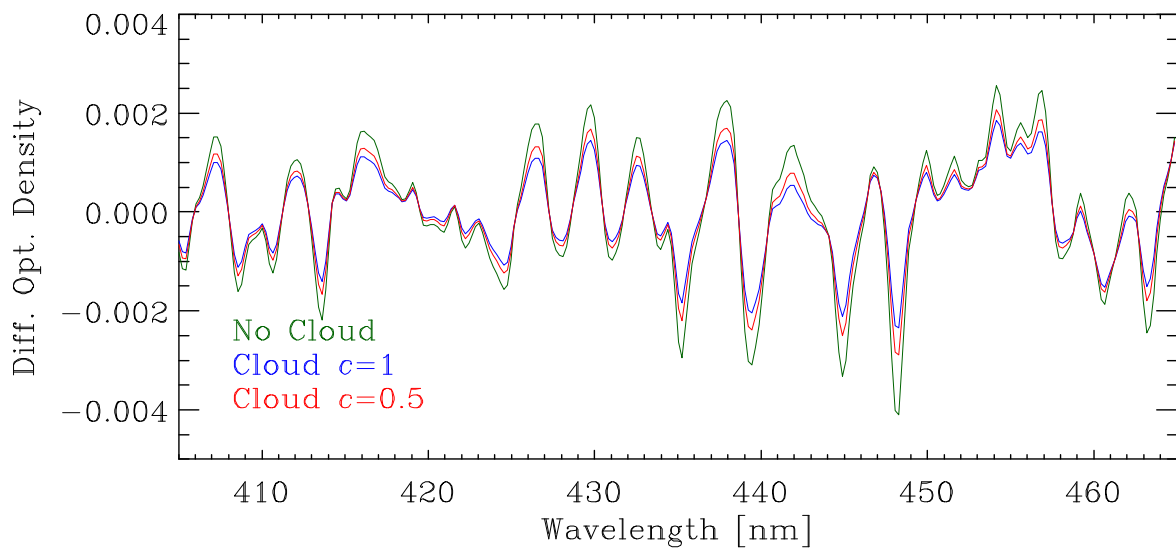
2Figure 1. Atmospheric profiles of NO<sub>2</sub> Volume Mixing Ratio in ppbv used in the study.

3



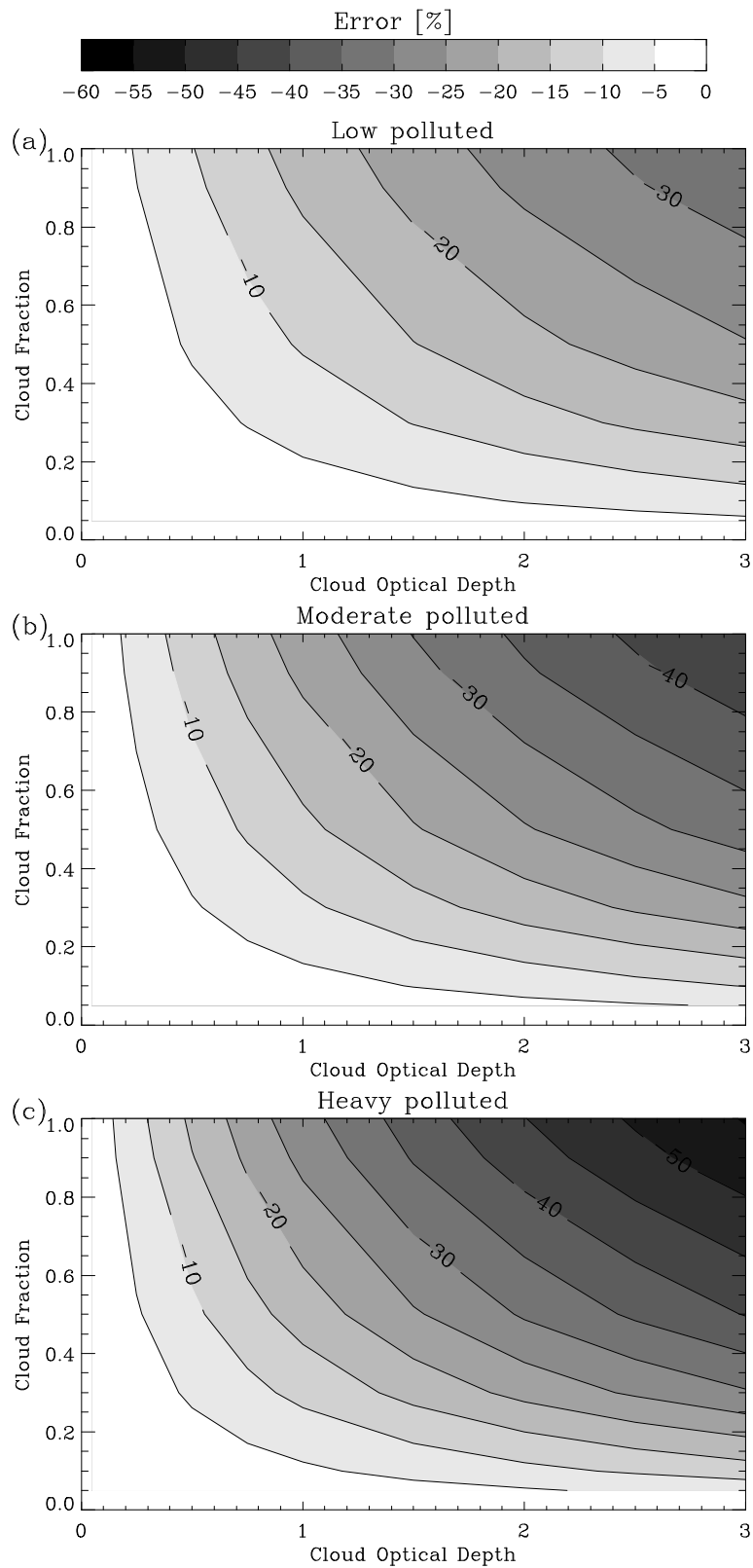
1

2Figure 2. Cirrus cloud phase function inferred from aircraft measurement during the  
3CIRCLE2 campaign.



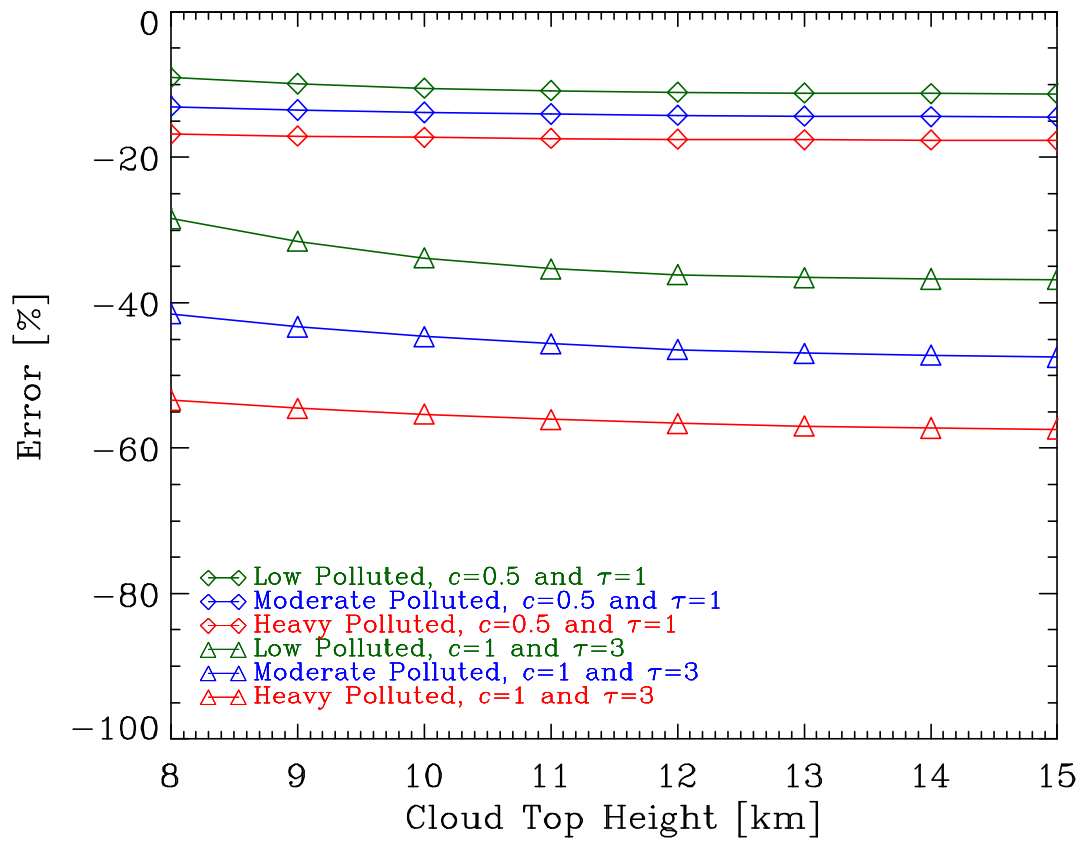
1

2Figure 3. Differential optical densities for heavily polluted troposphere without cloud (green  
3line), with  $c = 1$  (blue line) or  $c = 0.5$  (red line).



1

2Figure 4.  $\text{NO}_2$  tropospheric error caused by undetected cirrus cloud as function of cloud  
3optical depth and cloud fraction. Here,  $z = 10$  km,  $h = 1$  km and  $g = 0.75$ .



1

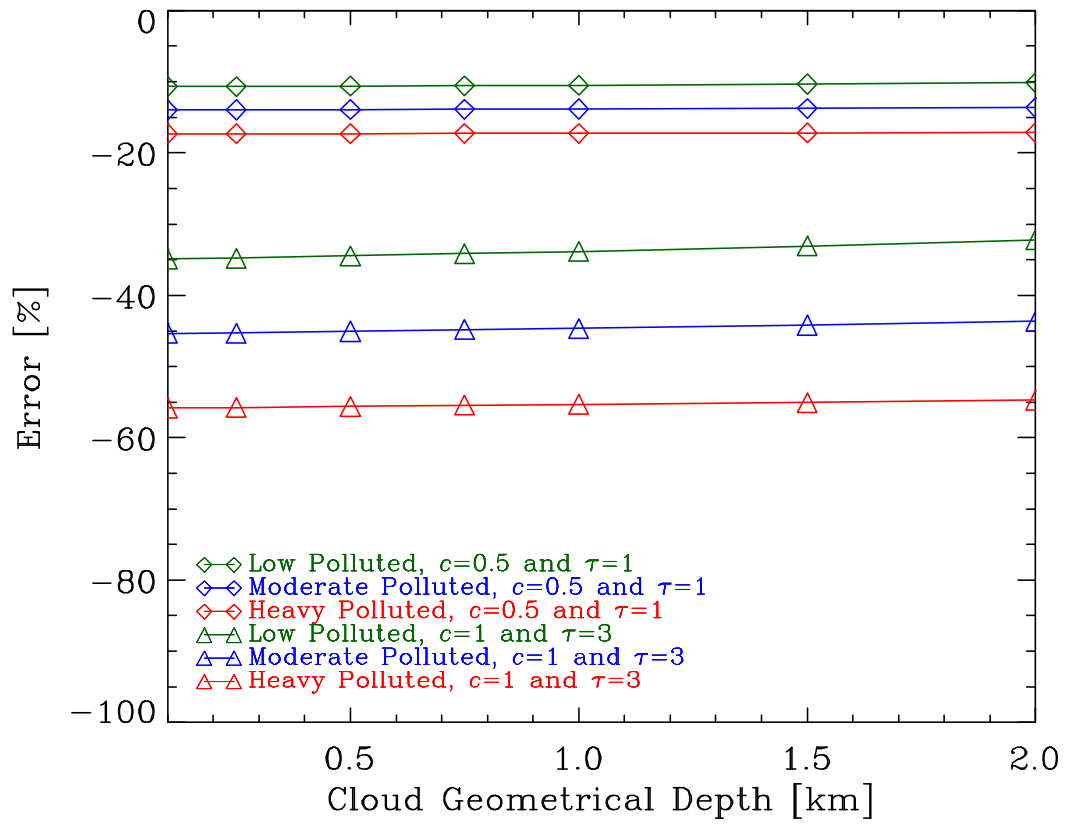
2Figure 5. NO<sub>2</sub> tropospheric error caused by undetected cirrus cloud versus cloud top height.

3Green, bleu and red lines stand for low, moderately and heavily polluted conditions. Cloud

4fraction and cloud optical depth were assumed to be 0.5 and 1, respectively for diamonds and

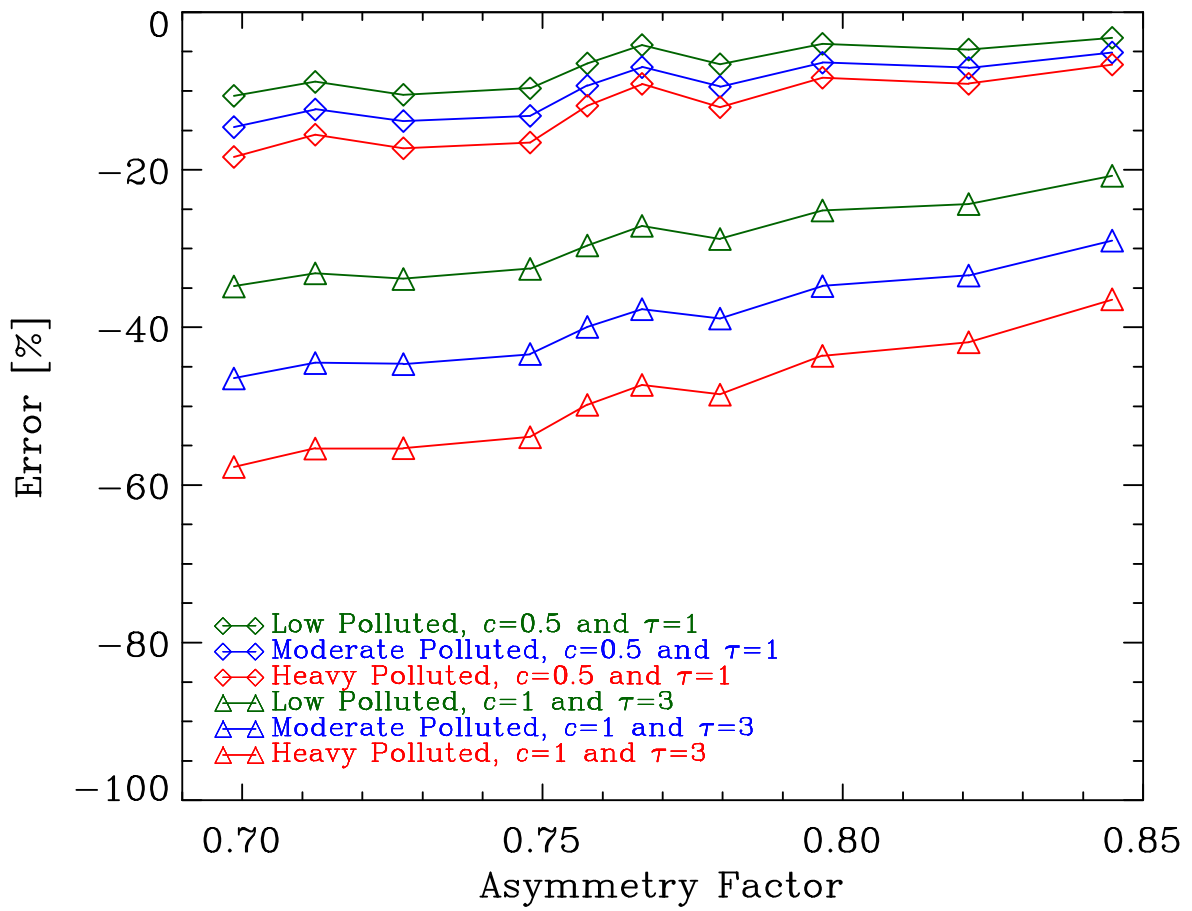
5to be 1 and 3 for triangles. Here,  $h = 1$  km and  $g = 0.75$ .





1

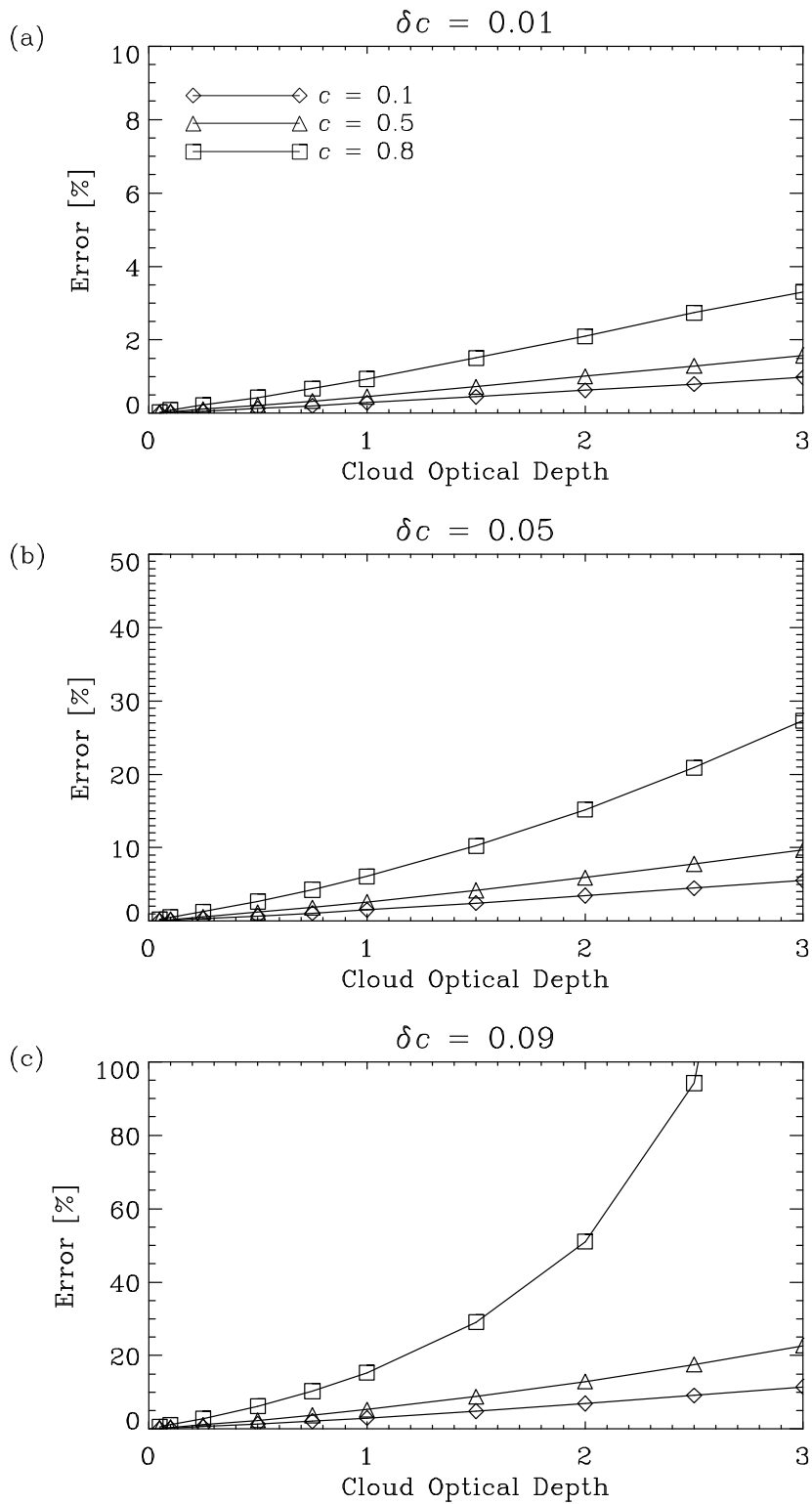
2Figure 6. Same as Fig. 5 versus cloud geometrical depth. Here,  $z = 10$  km and  $g = 0.75$ .



1

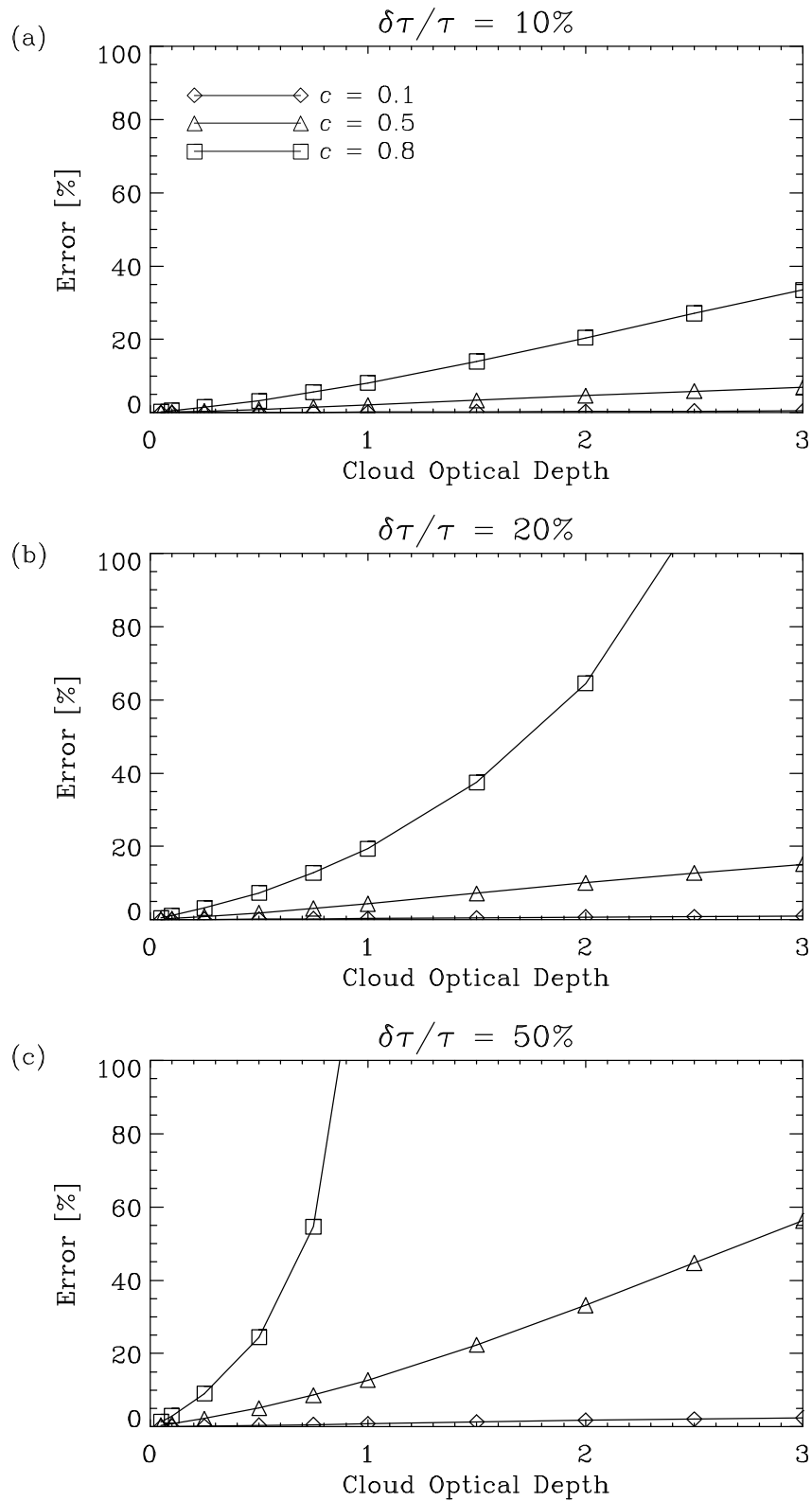
2Figure 7. Same as Fig. 5 versus asymmetry factor. Here,  $z = 10$  km and  $h = 1$  km.

3



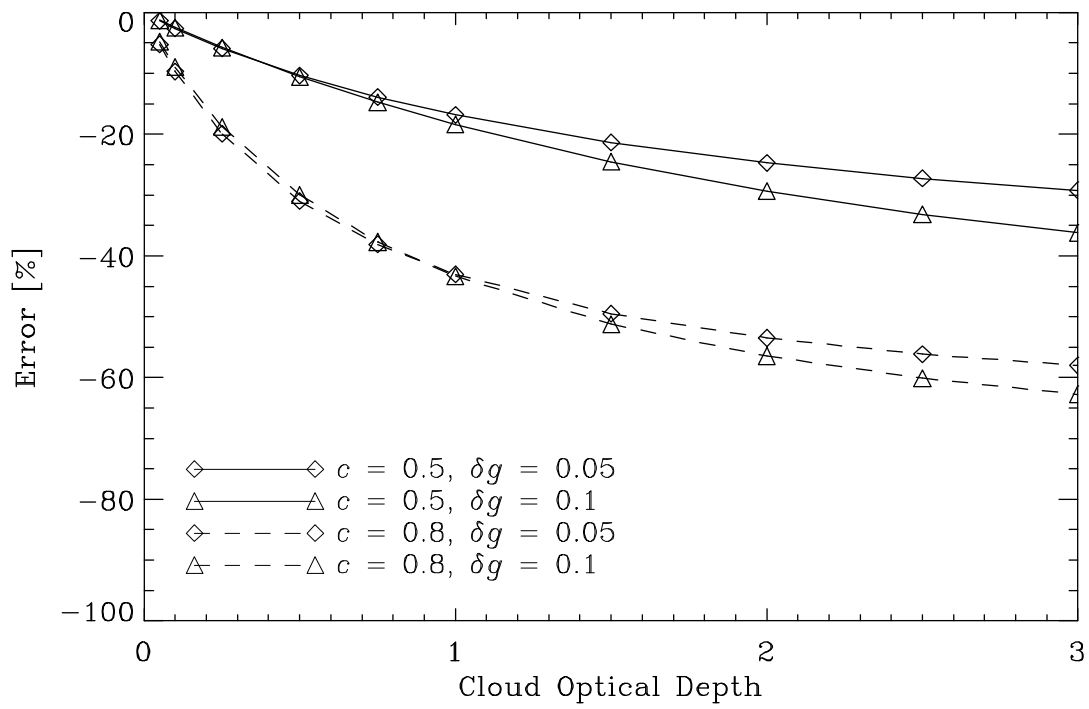
1

2Figure 8. NO<sub>2</sub> tropospheric column error caused by an uncertainty in the cloud fraction  $\delta c =$   
 40.01 (a),  $\delta c = 0.05$  (b) and  $\delta c = 0.09$  (c) versus cloud optical depth and for different cloud  
 4fraction ( $c = 0.1, 0.5$  and  $0.8$ , differentiated by symbols)., Here,  $z = 10$  km,  $h = 1$  km and  $g =$   
 50.75 and moderately polluted condition is assumed.



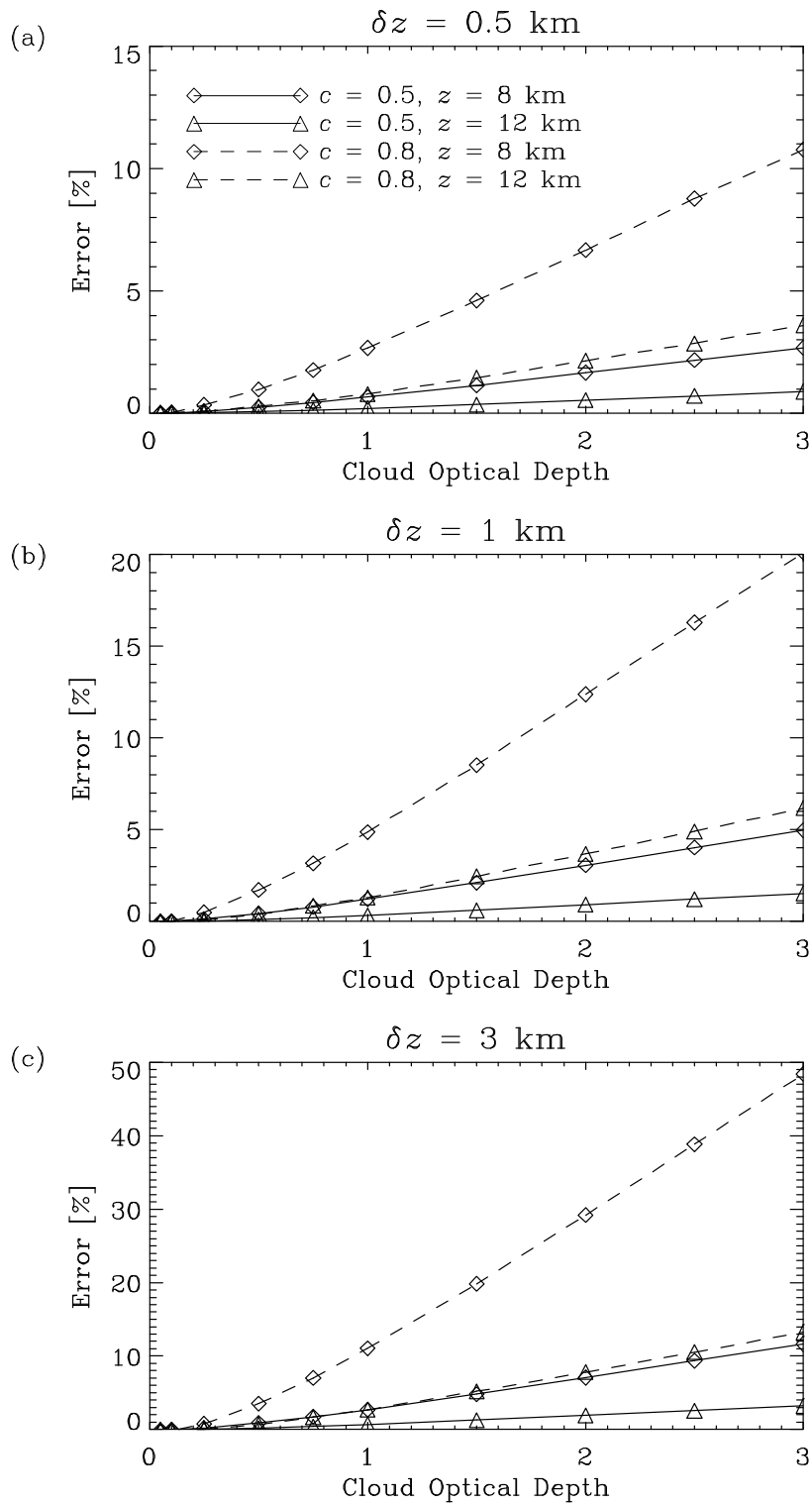
1

2Figure 9. Same as Fig. 8 for  $\delta\tau/\tau = 10\%$  (a),  $\delta\tau/\tau = 20\%$  (b) and  $\delta\tau/\tau = 50\%$  (c).



1

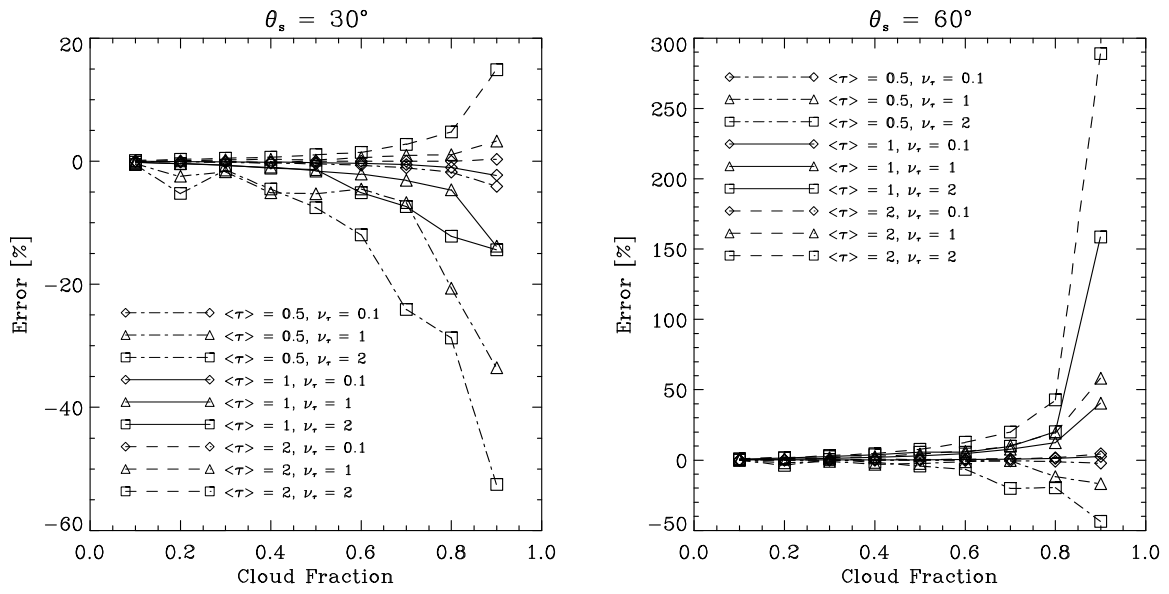
2Figure 10. NO<sub>2</sub> tropospheric column error caused by an uncertainty in the asymmetry factor  
3 $\delta g = 0.05$  (diamonds) and  $\delta g = 0.1$  (triangles) versus cloud optical depth for  $c = 0.5$  (full lines)  
4and  $c = 0.8$  (dashed lines). Here,  $z = 10$  km,  $h = 1$  km and  $g = 0.75$  and moderate polluted  
5condition is assumed.



1

2Figure 11.  $\text{NO}_2$  tropospheric column error caused by an uncertainty in the cloud top height  $\delta z$  3= 0.5 km (a),  $\delta z = 1 \text{ km}$  (b) and  $\delta z = 3 \text{ km}$  (c) versus cloud optical depth and for  $c = 0.5$  (full 4lines),  $c = 0.8$  (dashed lines),  $z = 8 \text{ km}$  (diamonds) and  $z = 12 \text{ km}$  (triangles). Here,  $h = 1 \text{ km}$  5and  $g = 0.75$  and moderate polluted condition is assumed.

1



1

2Figure 12. NO<sub>2</sub> tropospheric column error caused by the use of a mean cloud optical depth  
3instead of subpixels information versus cloud fraction if  $\theta_s = 30^\circ$  (left) and if  $\theta_s = 60^\circ$  (right).

1

Error difference [%] between $z = 8$ km and $z = 15$ km	$\tau = 0.05$	$\tau = 1$	$\tau = 3$
$c = 0.05$	0.01	0.29	1.07
	0.01	0.18	0.78
	0.01	0.12	0.56
$c = 0.5$	0.10	2.26	6.15
	0.06	1.40	4.39
	0.09	0.90	3.03
$c = 1$	0.19	3.63	8.35
	0.12	2.20	5.93
	0.17	1.35	4.02

2Table 1. NO<sub>2</sub> tropospheric error difference caused by cloud top height  $z$  variability. Values in

3each box of fixed  $c$  and  $\tau$  correspond to low, moderately and heavily polluted conditions,

4respectively.



1

Error difference [%] between $h = 0.1$ km and $h = 2$ km	$\tau = 0.05$	$\tau = 1$	$\tau = 3$
$c = 0.05$	0.01	0.08	0.34
	<0.01	0.04	0.21
	<0.01	0.02	0.14
$c = 0.5$	0.03	0.54	1.96
	0.02	0.31	1.23
	0.02	0.17	0.81
$c = 1$	0.05	0.88	2.69
	0.04	0.49	1.69
	0.04	0.27	1.10

2Table 2. Same as Table 1 for cloud geometrical depth  $h$ .

3

1 Error difference [%] between $g = 0.65$ and $g = 0.85$	$\tau = 0.05$	$\tau = 1$	$\tau = 3$
$c = 0.05$	0.07	1.00	2.66
	0.07	1.29	3.40
	0.09	1.61	4.21
$c = 0.5$	0.66	7.37	12.25
	0.77	9.44	15.40
	0.89	11.69	18.85
$c = 1$	1.27	11.30	14.04
	1.52	14.24	17.50
	1.76	17.58	21.27

2Table 3. Same as Table 1 for asymmetry factor  $g$ .

3