Dear Anonymous Referee #2,

The authors are very grateful to the reviewer for their insightful comments and suggestions, which has contributed substantially to the content and presentation of our new manuscript.

General comments

There are not many papers about aerosol vertical profile retrievals from oxygen absorption bands, so the topic of this paper is welcome and fits in AMT. But the paper has serious shortcomings, which will be described below.

The first author is cognizant of the lack of structure in the previous manuscript and apologizes sincerely to the reviewer for the difficulty it must have caused in reading the paper. I have posted an author comment entitled "General modifications" which presents a summary of the main changes, both structural and content-related, made to the new manuscript.

In the following we answer all reviewer comments on a point-by-point basis:

Although the paper has the relevant Section titles, the paper is not written in a structured way. Essential information that belongs to one section appears in other sections.

This holds for example for the algorithm description and its uncertainties.

The Abstract, Introduction, and Conclusions had many parts that belonged in other sections. These sections have been shuffled accordingly. An introductory paragraph outlining the structure of the paper has been added in the Introduction.

Referencing is too limited. There are more relevant papers about cloud height retrieval from the oxygen A-band of SCIAMACHY.

We have added the following references on the subject of O2 A-band retrievals from SCIAMACHY:

Kokhanovsky et al. 2005, van Diedenhoven et al. 2005, De Graf et al, 2007, Wang et al, 2008 and de Vries e al, 2009.

Also we have added the following references on the general subject of O2 absorption retrievals not focused on SCIAMACHY:

Koelemeijer et al. 2002, Daniel, Solomon et al. 2003, Guanter et al. 2010.

The introduction of the topic of the paper in Section 1 is rather weak. No mention is made of lidars.

We have added a brief description of lidar data, contrasting their excellent vertical information with their limited spatial sampling.

There is no proper introduction of the SCIAMACHY data; the limited information given about SCIAMACHY is scattered throughout the paper.

A new section has been added to introduce the reader to the SCIAMACHY instrument.

On the positive side: the discussion of the behaviour of the Jacobian matrix in Sect. 3 and the sensitivity studies in Sect. 4 are interesting and may be useful for others, when extended with the relevant additions, mentioned below.

At various places in the paper it is said that cloud filtering is not applied. So the retrieved aerosol properties (optical thickness, vertical profile) could very well belong to clouds. This means a drastic change of the scope of the paper; the reader should be informed about this in the title, abstract, introduction, and conclusions.

The title of the paper has been changed to "Retrieval of the optical depth and vertical distribution of particulate scatterers in the atmosphere using O2 A- and B-band

<u>SCIAMACHY observations over Kanpur: a case study."</u>
Corresponding changes have been made throughout the paper.

The main problem of the paper is that the real retrievals using SCIAMACHY data are not convincing at all. The retrieved AOD does not agree well with AERONET data.

The main problem of the comparison of SCIAMACHY with AERONET data is that they do not occur at the same time in most instances. Also, cloud contamination affects SCIAMACHY to much greater extent due to the large pixel size of the instrument. This makes comparison of data impossible for monsoon months (June-September), during which most days are cloudy. To work around these problems, we produces monthly means of both AERONET and SCIAMACHY measurements of AOT, which show a correlation coefficient of r=0.92 for non-monsoon months (Jan-May, Oct-Dec).

This correlation was not explicitly pointed out in our previous manuscript. The corrected manuscript has been accordingly updated.

Furthermore, there is no validation of aerosol height, so it can not be concluded that the retrieved aerosol vertical profile has any merit. What has to be added is a good error analysis for the retrievals of real SCIAMACHY data. It is not clear which algorithm assumptions are most contributing to the poor end result. Is it the missing cloud detection, the surface albedo or the aerosol microphysical parameters? Please discuss in Section 6 whether the problem is due to the retrieval method or due to the SCIAMACHY data, or due to another reason. We have added a new section entitled "Information content and error analysis" that addresses all the points raised by the reviewer.

The paper needs to be rewritten according to the specific comments before it can be accepted.

Specific comments Abstract:

- More specific and quantitative information about the results of the paper is needed in the abstract.
- The abstract has been modified in accordance with the reviewer's suggestion.
- Now lines 1 10 of the abstract are largely an introductory text which belongs to Section 1.
- These lines have been moved to the Introduction.
- Line 18 mentions "good agreement" but this is not a true summary of the results shown in the paper, especially those of Figure 11. It should be mentioned that the SCIAMACHY retrievals of AOT do not agree well with AERONET data; there is a poor agreement in Figure 11. There is no validation of the aerosol height. The problems in the real retrievals should be mentioned.
- In view of the fact that we cannot show any direct comparison with AERONET measurements, we have modified "good agreement" to "reasonable agreement". We have drawn attention to the correlation coefficient of 0.92 of 8 non-monsoon monthly mean AOTs that we obtain with AERONET data, so that it is clear that our agreement is not poor.

Section 1, Introduction:

Section 1 has several shortcomings and should be rewritten: - A discussion of lidars to measure aerosol height is missing here.

A short note describing Lidars has been added.

The last paragraph of Section 6 in fact belongs in the Introduction. - The difficulty of aerosol

height retrievals from passive satellite measurements should also be discussed.

Our comments comparing our method to LIDAR have been removed for lack of a rigorous comparison.

- This section is a mixture of previous work and own work. Please separate the two, and do not summarize your own results already in the introduction (e.g. on p. 6781, l. 21-22). Several parts of the Introduction have been moved to the Conclusions.
- This section should introduce the topic by properly referencing to earlier work, and by mentioning the goal of the paper and its unique contribution.

 The following line has been added to the Abstract and (with references) to the Conclusions: "Aerosol retrievals from SCIAMACHY have so far been limited to simple indices such as the Aerosol Absorbing Index (AAI) or the {SCattering} Index (SCI). Physics-based retrievals of optical thickness have been applied to thick clouds but not yet extended to aerosols. This paper shows the feasibility of the physics-based retrievals of aerosol optical thickness coupled with vertical information from satellites like SCIAMACHY. Nonetheless, large-scale application to SCIAMACHY data is also severely challenged by radiometric uncertainties which make a simultaneous retrieval of aerosol microphysical parameters practically impossible. Addressing these issues in future instruments similar to SCIAMACHY, coupled with computational resources and speed-up of the current line-by-line radiative transfer calculations, can allow our approach to be extended to the global scale, especially because it is not restricted to dark surface types."
- What is new in the current paper as compared to Sanghavi et al., Appl. Opt., 2010? Please briefly summarize this paper, since it is an essential part of the algorithm in Sect. 2 and later sections.
- The 2010 paper has been summarized in the section 3 (Retrieval Algorithm) and section 5 (Retrievals over Kanpur, India, using SCIAMACHY data).
- Some sections of the paper (4 and 5) are mentioned without introducing the structure of the paper. Please add a last paragraph to the introduction with the structure of the paper. A paragraph describing the structure of the paper has been added at the end of Section 1 (Introduction).

Last paragraph: Apparently, clouds are not discriminated from aerosols. This is not so much of a problem for cloud retrievals, since clouds have much larger optical thick- nesses than aerosols, but it is a severe problem for aerosol retrievals. What is the value of the retrieved AOT? It seems that non-discriminating between clouds and aerosols undermines the scope of the paper, which is aerosols.

Our choice of Kanpur for our case-study partly alleviates this problem, mainly during non-monsoon months due to the generally low likelihood of cloudy skies in the region. Application of our algorithm to other parts of the globe may require very reliable cloud-screening if only aerosols are targeted, but by changing our title to focus on atmospheric particulate matter and not only aerosol, we have shifted the scope of the paper.

- p. 6780, l. 22: large eruptions of the Eyja volcano took place in April and May 2010. Corrected April 2010 -> April-May 2010.
- p. 6781, I. 7ff: there are more relevant references on the retrieval of cloud height from the O2 A-band from SCIAMACHY, like the FRESCO method papers: Koelemeijer et al., JGR, 2011; Wang et al., ACP, 2008.

These references have been included.

p. 6781, l. 10ff: please refer to the paper by Solomon et al. (JGR, 2003) on the information content of the different O2 absorption bands.

<u>Done.</u> (We could not find such a paper by Solomon et al., but Daniel, Solomon et al, 2003, seemed to fit the description perfectly.)

p. 6781, l. 18-19: which parametrized form?

This part has been moved from the Introduction to the Conclusions, after being modified to "retrieve a lognormal approximation of the vertical profile of the aerosol column", instead of "parametrized vertical profile".

Section 2:

p. 6782, I. 14: The Aerosols and UV part of the 2002 OMI ATBD, referenced here as Stammes and Noordhoek (2002) is published as a refereed paper, namely Torres, Tanskanen, Veihelmann et al., JGR, 2007. This is the preferred reference (also later in the paper).

Reference added.

p. 6783, l. 21ff: which aerosol model was assumed?

The aerosol type assumed is a coarse urban absorbing type (IA3). This has been mentioned in section 3 (Retrieval algorithm). It has also been pointed out that for some of the retrievals, this assumption clearly does not work, due to which we have to make use of a finer, less absorbing urban aerosol type, IS1, to achieve a better fit with the observed measurements.

What is the Angstrom parameter of this aerosol type? Does 10 % allow sufficient flexibility? Please include these points in the error analysis to be discussed in Sect. 6. Instead of considering Angstrom exponents directly, we use the quantity dtau/dlambda, related to the Angstrom exponent alpha as dtau/dlambda = -alpha*tau/lambda (obtained by considering the reference wavelength to be very close to the wavelength of interest and under the (valid at visible wavelengths) assumption that alpha remains spectrally constant). The assumption of +/-10% variation holds true for all aerosol types other than water cloud and desert dust, so that an error would be caused by our constraints when clouds or desert dust particles are present. A detailed error analysis has been provided in section 3.2.1., which shows that while a more relaxed constraint on dtau/dlambda it would make our retrieval better and faster, the effect of this parameter is not strong enough to affect the stability of our retrieval.

- p. 6784, I. 3: what are these measurement data? The SCIAMACHY data (mainly its spectral and spatial properties) are not properly introduced in the paper. This should be done in Sect. 1 or anyway before the retrieval method is discussed.

 Added Section 2 (The SCIAMACHY instrument).
- p. 6784, I. 20-21: I do not see how Eq. (6) is testing that the difference between successive iterations is an order of magnitude less than the estimated error.

 Equation modified to agree with the corresponding statement.
- p. 6784, Eq. (7) and I. 23: is this state vector length n the same as the N in Section 3, I. 3 on p. 6785? Please use one symbol for one variable. <u>Changed n to N</u>

Section 3: p. 6785, l. 6: please number this equation for later reference Done

p. 6785, l. 12: . . ., respectively \rho_A, and \rho_B, Added "respectively"

p. 6785, l. 13-14: continental surface > land surface (this also occurs later on)

Done throughout the paper

p. 6785, l. 26ff: please explain the 'depth of the radiance' in terms of the quantity dlnR/dx which is shown in Figure 1 and successive figures. Define this R. Necessary definitions added with equations explaining 'depth of radiance'.

p. 6786, l. 6: optical thickness > aerosol optical thickness Done

p. 6786, l. 6: . . . is a direct consequence . . .: this should also #depend on the height of the aerosol layer

Yes, this is true. In view of the fact that our results could be over-generalized, we have added the following lines in Section 3.2.1 before describing the Figures illustrating the Jacobians: "Before moving on to describe the Jacobians illustrated in Figures \ref{fig:K black}, \ref{fig:K black}, \ref{fig:K hc} and \ref{fig:K dm}, it must be pointed out that the behavior of the Jacobians is specific to the state vector at which it is calculated. This is a consequence of the non-linearity of our problem. For example, the ``depth", \$\Delta R\$, is seen to increase with increasing aerosol optical thickness in both bands for the black surface. While we see in the following that this behavior varies with underlying surface albedo, it is not immediately apparent (but true) that it can also change if a different value of \$\tau {700}\$ and/or \$z p\$ is chosen."

p. 6786, l. 14: 'Since the bulk of the Rayleigh scatterers resides close to the surface': this is too simplistic, because the Rayleigh scale height is about 8 km; is that close to the surface? This is true. We have changed our wording to "Since more Rayleigh scatterers reside closer to the surface"

I. 15: net > mean changed

I. 20: the log normal tails: of the vertical distribution function changed

I. 22: sensitivity > vertical sensitivity changed

I. 23: depths > geometric depths <u>changed</u>

I. 27-28: optical thickness > aerosol optical thickness (also in the remainder of this section) changed

p. 6787, l. 15: also evident > are also evident changed p. 6787, l. 15: What is new in Fig. 4 as compared to the convolved curves in Figs. 1-3? Is there a change of slit function, since the curves in Fig. 4 look less smooth than the convolved curves in Figs. 1-3?

There is no change (even though the thickness of the lines might make them look less smooth), the convoluted curves have only been plotted together to facilitate comparison between them at a glance.

Section 4:

p. 6788, l. 3: 0.5-1.0 > 0.5-1.0 km

The peak widths \sigma p are dimensionless for the form of the lognormal distribution shown in Eq.1. Hence no unit has been used.

p. 6788, I. 3: is the assumed error of 1 % in SCIAMACHY data not much too small, given the fact that SCIAMACHY has quite some radiometric calibration uncertainty? Please give a reference. Is this the error in the radiance or in the reflectance? Is it a random error or a systematic error?

The measurement error covariance matrix typically consists of the purely statistical noise errors while the radiometric calibration uncertainty is a systematic bias, which has to be considered differently. In terms of pure noise, a 1% error across the band is even somewhat conservative. We have discussed the radiometric calibration as a potential source of bias in the new Section 3.2.3.

p. 6788, l. 6-7: remove 'between the x- and y-axes'. Done

I. 7: what are vertical parameters?

Changed to: vertical parameters, z p and sigma p

I. 9: 'truth lines'> true values Done

I. 10: negligible > small Done

I. 14: vertical profile > the vertical profile Done

I. 17: 'the difference in surface albedos adds to the information content of the latter measurement': can you please explain this? What is the physical reason?

This has been illustrated quantitatively in an in-depth information content analysis (Section 3.2)

p. 6789, l. 3-4: truth line > true value Done

p. 6789, I. 5: 'we can assume very robust retrievability. . .': Please remove 'very'. Done

I. 6: 'moderately good': this is too optimistic, please replace by: 'only limited information on \sigma_p'.

Done

I. 12-15: An important question rises here: is in this synthetic retrieval the surface albedo assumed to be known perfectly? But in reality this is never the case. How do you deal with the unknown surface albedo in retrieval of real data? Wouldn't it be better to simultaneously retrieve the surface albedo together with the aerosol parameters?

We have added a new error analysis for this problem, addressing exactly the impact of this assumption. This point has been addressed in detail in the error analysis of Section 3.2.2

In the sensitivity studies of Section 4 no mention is made of the dependence of the results on the following parameters: 1. assumed error in the satellite data 2. neglect of error in surface albedo (see point above). 3. assumed aerosol microphysical parameters, like phase function and single scattering albedo 4. geometry of satellite and sun. These parameters seem important and belong to the sensitivity studies of the retrieval algorithm.

The first 3 points have now been discussed in depth in the error analyses of Sections 3.2.3, 3.2.2 and 3.2.1/3.2.3 respectively. The 4th point has not been addressed directly, but in a private analysis, the first author has plotted retrieved surface albedos for different near-clear sky days from SCIAMACHY data over Kanpur against the solar and line-of-sight zenith angles as well as the solar azimuth, and these do not exhibit much variation with geometry. For application of our approach to a pixel showing strongly asymmetric BRDFs we would need to take geometric effects into consideration, but for Kanpur such a discussion would go beyond the scope of the paper.

Section 5:

p. 6790, l. 5: There could be a real seasonal cycle in the relative contrast of the surface, so the value to be chosen may depend on the season.

We explain in Section 3.2.2 why we do not consider a seasonal cycle in the surface albedos (or their relative contrast) to be substantial over Kanpur.

Also, the relative contrast hypothesis is based on radiative transfer theoretical arguments and not on empirically observed phenomena, so that a coincidental agreement of the observed data would be highly unlikely. Since we see a good anti-correlation between retrieved AOT and relative contrast without any competing trends due to seasonality, we feel more confident about the lack of seasonality in surface reflectance.

I. 10-16: This paragraph seems to suddenly change the scope of the paper. If the paper is aiming to determining atmospheric scatterers, whether they are clouds or aerosols, this should be said clearly in the title, abstract, introduction and conclusion of the paper.

We have changed the title to reflect this and also made it clearer throughout the manuscript now.

In fact, the part of Section 5 from line 10 on p. 6790 to line 19 on p. 6791, discussing fundamental issues of the retrieval method, does not belong here. It belongs to Sect. 4, or even earlier in the paper. After the SCIAMACHY results are presented this earlier fundamental discussion can be included in the discussion of the results.

Done

I. 19: 'such oscillations': probably more well-separated wavelengths are needed to prevent such oscillations (cf. the MODIS aerosol algorithm).

This is true, and we had initially conceived an algorithm additionally using several SCIAMACHY aerosol "windows" together with the still weaker O2 gamma band and the O2-O2 absorption feature around 550 nm. However, we had to abandon most of these additional

spectral regions due to radiometric uncertainties and also due to the fact that SCIAMACHY has different ground pixels corresponding to different spectral channels due to differences in the integration times of the various channels – which implied that measurements at different wavelengths would be made for different ground scenes. This is not an issue for trace gas retrievals, but would have thrown off most aerosol retrievals. This resulted us in settling for the O2 A- and B-bands, which had the same ground pixels and were not too erratic in their radiometric uncertainties.

p. 6792, l. 3: 'scene inhomogeneities': this is not a probable reason of the bad agreement, since aerosol fields are much more homogeneous than clouds. More probable is that the assumed aerosol microphysical properties are not correct in reality.

This is true, though inhomogeneities due to the large pixel size of SCIAMACHY also lead to uncertainties. This case has been stated more clearly in the manuscript on several occasions including the following in the Section 5 on Retrievals over Kanpur:

"Radiometric calibration issues around the O₂ A- and B-bands are not very severe and are more easily alleviated by co-retrieving the correction factors under tight constraints, so as not to undermine the retrieval of aerosol parameters. However, we have to rely almost entirely on climatology for information on aerosol type, since we do not have the sensitivity to track changes in the microphysical properties of the aerosol. In order to ameliorate resulting errors in our case-study retrievals, we have to make occasional use of other aerosol types, typically IS1, when our principal climatological assumption of the IA3 aerosol type fails to achieve an acceptable convergence.

Also, little can be done to eliminate pixel inhomogeneities, especially those in the form of partially clouded scenes. This results in fits that frequently lie outside of the limits set by measurement error, since the uncertainty in the retrieval predominantly stems from the lack of accurate representation by the forward model. This is a problem endemic to aerosol retrievals in general, which is exacerbated in the case of the SCIAMACHY instrument due to 1. Its inter-channel radiometric uncertainties, making it practically impossible to use additional wavelengths to develop a sensitivity to aerosol microphysical parameters, and 2. Its large pixel size, which introduces uncertainties not only due to clouds but also surface reflectance, making a homogeneous pixel representation unrealistic."

I. 27: what do you mean with 'later CALIPSO measurements'? Did you make a climatology for later years and compare this to your SCIAMACHY retrievals? Please quantity this statement. Please perform the retro-active comparison to show that there is merit in the vertical profile information from SCIAMACHY.

We have not made any detailed intercomparison, primarily because of the fact that Calipso was not in orbit in 2003. Hence, we have withdrawn our CALIPSO-related statements.

p. 6793, l. 11: see > show

The word see has been used here as in "witness" or "experience" and is not used to mean show.

I. 14ff: This is an honest discussion of the problems of the retrieval algorithm. This should also be reflected in the Abstract and Conclusions.

Modifications have been made to this effect. All statements describing our results as "very good" have been removed, and have been replaced by arguments taking the uncertainties associated with SCIAMACHY measurements and other sources of error into consideration. The large pixel-size and the inter-channel radiometric irregularities have been identified as the main drawbacks of using the SCIAMACHY instrument for aerosol retrievals.

Section 6, Conclusions: p. 6794, l. 3: 1 nm? but SCIAMACHY has 0.3 nm spectral resolution. The instrumental FWHM is 0.43nm for the B-band and 0.44nm for the A-band. We have summarized this as a spectral resolution of approx. 0.4 nm. The 1nm must have been a typo, our apologies for the confusion.

I. 6-7: 'this is true for most ...': but this is not true for ocean, which is the largest terrestrial surface

'Terrestrial' has been changed to 'land'

- I. 14: 'good agreement': this is not correct. Please indicate the problems of the retrieval algorithm, and give an honest report of the results found in Section 5.

 Modifications have been made to this effect as detailed previously
- I. 15: no comparison of the vertical distribution was shown with CALIPSO, also not retroactively. Please remove sentence.

 <u>Done</u>

Please indicate what the uniqueness is of this work. Apparently AOT from real SCIA-MACHY data shows large discrepancies. Perhaps the aerosol altitude is a unique contribution?

Aerosol retrievals from SCIAMACHY have so far been limited to simple indices such as the Aerosol Absorbing Index (AAI) (de Graaf et al., 2007) or the SCattering Index (SCI) (de Vries et al., 2009). Physics-based retrievals of optical thickness have, to the best of our knowledge, been applied to thick clouds (Kokhanovsky et al., 2005) but not yet extended to aerosols. In this paper, we show that the retrieval of aerosol optical thickness from satellites like SCIAMACHY is feasible and could be, given computational resources and speed-up of the current line-by-line radiative transfer calculations, extended to the global scale since it can be applied to dark as well as bright surfaces.

Currently, large-scale application to SCIAMACHY data is severely challenged by radiometric uncertainties which make a simultaneous retrieval of key aerosol microphysical parameters by using more, spectrally farther-spaced wavelengths practically impossible. However, addressing these issues in future instruments similar to SCIAMACHY, coupled with computational resources and speed-up of the current line-by-line radiative transfer calculations, can allow our approach to be extended to the global scale, especially because it is not restricted to dark surface types.

References: - Martonchik: place of dissertation? - Rodgers: publisher? References completed

Figures and captions:

Figures 1 - 3: Please give the fixed aerosol parameters of each subplot in the legend or caption. Please specify the sun-satellite geometry. Please specify the aerosol type. A coarse biomass aerosol (BL3) has been used. This was an arbitrary choice, motivated only by the wish that the aerosol have a slightly higher peak height, which the BL3 aerosol does as a standard setting in our code. The viewing geometry is statistically representative for SCIAMACHY observations over Kanpur with SZA=43 deg, LZA\$=11deg, Azimuth=149 deg.

Figures 5-7: Please specify the geometry. Is \sigma also given in km? Please state that the black line is the 1:1 line.

<u>Done.</u> As mentioned before, our \sigma_p is dimensionless and hence no units have been used.

Caption Fig. 9: which measurement data were used?

The relative contrast was calculated as explained in the paper: "Our method calculates the apparent lam- bertian equivalent albedo (ALEA), ρ , obtained by calculating the lambertian surface albedo that would produce the same reflectance at a given wavelength for an aerosol/cloud-free atmosphere as that measured by the satellite instrument. By calculating the value of ALEA at two different wavelengths, one near the A-band, ρ A, and the other near the B-band, ρ B, we compute the "relative contrast" of a given scene as $(\rho$ A – ρ B)/(ρ A + ρ B). For a given geometry, the relative contrast is found to decrease monotonically with increasing aerosol loading."

The AOTs are the ones retrieved from SCIAMACHY.

Caption Fig. 11: what is the value of the correlation coefficient of the bottom panel? R=0.92 for non-monsoon months (Jan-May, Oct-Dec)

Some textual comments:

Abstract: trace-gases > trace gases

Done

Introduction: Ejya . . . > Eyja. . .

Corrected spelling

p. 6780, l. 21: punctuated by? please find another word, like: stressed, emphasized, underlined

punctuated -> underlined

p. 6782, l. 1 (and in many other places): scenarios > scenes (scenario is a model situation; a scene is a real situation)

Changed

p. 6792: time progression > time

<u>Done</u>