Response to reviewer #1

We thank the reviewer for his/her evaluation of our paper and useful comments that helped improve the manuscript. We appreciate reviewer's time and effort in reviewing the manuscript. Below are responses to each comment. All reviewer's comments are in the standard font while the responses are in the italic font.

On behalf of the authors, Alexander Vasilkov

General comments:

1, The explicit aerosol corrections are only applied to the clear-sky part of a pixel, which causes inconsistency between the cloud and NO2 retrieval.

We do not think that there is inconsistency between the cloud and NO_2 retrieval. Both cloud and NO_2 algorithms make use of the MLER model based on the independent pixel approximation with the same treatment of surface BRDF and aerosol in the clear-sky part of a pixel. The cloudy-sky part of a pixel is treated in the same manner as an opaque Lambertian surface with the same reflectivity of a commonly-accepted value of 80%. The basic assumption here is that aerosol is affecting the clear-sky part of the pixel only and is negligible in the cloudy part of the pixel. This assumption would fail for absorbing aerosol present above a cloud layer as discussed in the response to the next comment.

The studies of Lin et al., (2014) and Liu et al., (2019) have already shown the impacts of aerosols on cloud retrievals. Jethva et al., (2016) also showed absorbing aerosols were observed over clouds in spring and winter in Eastern China. So at least, the authors should discuss the impacts of explicit aerosol treatments on cloudy-sky retrievals.

It should be noted that NO_2 retrievals for cloudy sky conditions are highly uncertain and uncertainty can reach up to 100% (Boersma et al., 2011, Bucsela et al., 2013). Therefore, we recommend using NO_2 retrievals for clear and partially cloudy conditions only. Our focus here is on retrievals for low effective cloud fractions only, typically less than 0.25. We think that non-absorbing aerosol above the cloud with high reflectivity can be neglected. However, we agree that the impact of absorbing aerosol above the cloud is important and should be discussed.

We added the following in Sect. 2.5.1:
"It should be noted that a contribution of non-absorbing aerosol above a cloud with high reflectivity, as we assume within the MLER concept, to the cloud radiance is negligible. However, absorbing aerosol above the cloud can affect the cloud radiance. Analysis of frequency of occurrence of absorbing aerosol above the cloud derived from the 12-year record (2005–2016) of OMI led to the identification of regions with frequent aerosol–cloud overlap (Jethva et al., 2018). Figure 5 of that work showed that the most frequent aerosol–cloud overlap occurs over the oceans where the long-range transport of aerosols plays an important role and low-level marine stratocumulus clouds are observed. This fact is also confirmed in a recent paper by Zhang et al. (2019). Those oceanic regions are of less interest for tropospheric NO2

retrievals because of the small contribution of anthropogenic NO_2 pollution. Additionally, tropospheric NO_2 retrievals over the oceanic regions are prone to errors from other aspects of retrievals (e.g., separation of stratospheric and tropospheric components), which are more important than aerosol effects. The springtime biomass burning activities such as burning of forest, grassland and crop residue over Southeast Asia release significant amounts of smoke particles observed over the widespread cloud deck over southern China on about 20%–40% of the cloudy days. NO_2 retrievals are typically not performed for those events owing to high cloud fractions. It is possible to flag and discard such retrievals if they were to occur in partial or thin cloud conditions using the absorbing aerosol index (Jethva et al., 2018). The treatment of absorbing aerosol over the cloud for NO_2 retrieval in such scenarios is beyond the scope of this work."

Jethva, H., Torres, O., Ahn, C. A 12-year long global record of optical depth of absorbing aerosols above the clouds derived from the OMI/OMACA algorithm. Atmos. Meas. Tech., 11, 5837–5864, 2018.
Zhang, W., Deng, S., Luo, T., Wu, Y., Liu, N., Li, X., Huang, Y. and Zhu, W.: New global view of above-cloud absorbing aerosol distribution based on CALIPSO measurements, Remote Sens., 11, 2396; doi:10.3390/rs11202396, 2019.

2, The authors have listed plenty of comparisons in the introduction to illustrate the difference between satellite retrievals and other measurements. However, only one case study without any comparisons with ground- or aircraft-based data are discussed in the paper. The reviewer is doubt about the applicability of this method.

There is no doubt that explicit aerosol correction will reduce OMI low bias compared with ground-based data documented in numerous previous publications. Figure 9 compares current operational (b) and future (c) OMI tropospheric NO_2 retrievals. It shows that explicit aerosol correction enhances tropospheric NO_2 VCDs for all OMI pixels, which goes in right direction to mitigate general low bias in satellite NO_2 retrievals.

The authors should collect some measurements and make further comparisons since it seems that the method can be applied to anywhere globally. At least, more cases should be collected to reach comprehensive/valid analysis

Extensive OMI NO_2 comparisons with ground-based and aircraft measurements have been recently documented by our group (e.g. Choi et al., AMT, 2019) as well as other groups. Consensus has been reached that all current satellite tropospheric NO_2 measurements (OMI/GOME-2/TROPOMI) have low bias for highly polluted environments (Herman et al., AMT, 2019). Additional new comparisons are beyond the scope of this paper and are subject of a separate paper by our group (Lamsal et al., 2020, in preparation).

What this paper demonstrates, is that aerosol related uncertainties of current OMI (and for that matter TROPOMI) operational cloud (version 2.0) and tropospheric NO_2 (version 4.0) products can be unacceptably large under certain polluted conditions (e.g., Figs. 8-10), which certainly justifies consideration for implementation in the next version. Before operational *implementation, more rigorous comparison with a large collection of available ground-based data will be conducted.*

Indeed, since we have already implemented GLER surface reflectance (which, by itself is explicitly aerosol corrected) in the current OMI operational algorithms, the next necessary step will be implementing explicit aerosol treatment in both cloud and NO_2 retrievals to make them consistent. Thus, the explicit aerosol correction is not only physically based, but indeed required for self-consistency of both cloud and NO_2 retrievals, which is necessary logical step prior to comparisons with ground-based and aircraft data.

Specific comments:

1, Line 3: “over norththeast Asia” should be “over northeastern Asia”. Based on the context, the authors only discussed a specific case over northeastern China.

Corrected.

2, Line 17: “top down” should be “top-down”. “assimilation” should be “assimilations”

Corrected.

3, Line 26: “optical properties” would be more accurate than “scattering properties”

Agree and done.

4, Line 27: Please cite (Castellanos et al., 2014; Liu et al., 2019)

Added.

5, Line 31: What do you mean by “detailed”?

We mean “modeling that includes treatment of clouds, the surface, and aerosols” as it is stated in the sentence.

6, Line 33: I do not think the definition of the Jacobians (AK) is the same as AMF.

Agree. We clarified this by replacing “or” by “needed for calculation of”.

7, Line 47: “southeast Asia” should be “Eastern China”

Corrected.

8, Line 48: “using data from the GEOS-Chem model with further adjustment through MODIS monthly AOD dataset.”

Added.

9, Line 51: Not exactly. Lin et al., (2014, 2015) and Liu et al., (2019) claimed that they used parallel RTM to ensure the efficiency of the calculation.

We mean that even with parallel RTM computations the cited studies were not carried out on a global scale as needed in operational processing of satellite instrument data. That is why we state that "these studies were carried out on a regional scale" in this sentence.

10, Line 175-180: See the general comment 1. Please add some specific case to help valid the argument here. "part of pixel only" should be "part of a pixel only"

We added some discussion here (see the answer to general comment #1). The missed article is added.

11, Line 244: What causes such an enhancement? It happens in this specific case or it related to the way that GEOS-5 uses to separate the troposphere and stratosphere?

This relatively small aerosol enhancement at altitudes of 11 km is thought to happen in this specific case. An exact cause of the enhancement is not clear.

12, Based on my understanding, the procedure for deriving GLER does not include aerosol optical properties, either. GLER is an important concept in this paper. Please add the definition or give a brief introduction to it.

You are right, GLER does not include aerosol contribution. It is calculated using atmospherically corrected BRDF data. We stated this in Sect. 2.4. We agree that adding the definition of GLER will be helpful. We added the definition and corresponding equation in Sect. 2.4. Lines 144-146 read now:

"The GLER is derived from TOA radiance computed for Rayleigh scattering and full surface BRDF for the particular geometry of a satellite instrument pixel. The TOA radiance computed by VLIDORT is then inverted to derive GLER using the following exact equation:

$I_{TOA} = I_0 + GLER*T / (1 - GLER*S_b)$,

where I_0 is the TOA radiance calculated for a black surface, T is the total (direct+diffuse) solar irradiance reaching the surface converted to the ideal Lambertian-reflected radiance (by dividing by π) and then multiplied by the transmittance of the reflected radiation between the surface and TOA in the direction of a satellite instrument, and S_b is the diffuse flux reflectivity of the atmosphere for the case of its isotropic illumination from below (Vasilkov et al., 2017). All quantities, I_0, T, and S_b are calculated using a known surface pressure for a given OMI pixel. The GLER concept has been evaluated with OMI over both land (Qin et al., 2019) and ocean (Fasnacht et al., 2019)."