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Interactive comment on "Retrieval of total column and surface NO_2 from Pandora zenith-sky measurements" *by* Xiaoyi Zhao et al.

Anonymous Referee #2

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General comments

This study presents a new approach to extend direct-sun NO2 measurements from Pandora instruments with a zenith-sky mode also applicable under cloudy conditions. In addition, attempts are made to also derive surface concentration estimates from total column observations. The general methodology is strongly inspired from the empirical zenith-sky mode developed for total ozone measurements by Dobson and Brewer spectrophotometers. Although the adopted approach implies many approximations not always well described or even identified (see detailed comments below), results are surprisingly good and certainly of interest for the ACP readership. I found the manuscript well written, concise and easy to read; also figures are of good quality and adequate in number and the appendices provide useful additional information. With

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one exception, credit to existing literature is appropriate. I therefore recommend publication in ACP, after careful consideration for the comments and suggestions below.

Specific comments

Section 2.1.2, L. 20: what was the temperature used for the NO2 cross-section in the zenith-sky QDOAS retrievals? Is it consistent with the effective temperature assumed for the direct-sun retrieval (254.5°)?

Section 2.1.2, L. 25: how was the NO2 residual amount in the reference spectrum determined here? Generally speaking, this paper lacks a proper analysis of the uncertainties. It would be useful to add a section describing the estimated uncertainties for the zenith-sky column and surface concentrations (which are new data products introduced in this study).

Section 3.1: The approach introduced for the zenith-sky AMF calculation is fully empirical and strongly inspired from the zenith-sky measurement mode used with Dobson and Brewer total ozone spectrophotometers. Basically the idea is to use simultaneous direct-sun and zenith-sky measurements to infer effective AMFs for the zenith-sky geometry. It is then assumed that the established relationship remains valid under moderately cloudy conditions (O4 is used to exclude thick diffusing clouds). I first note that the authors do not refer to the AMT publication by Tack et al. (2015) (https://www.atmos-meas-tech.net/8/2417/2015/) where a more physical approach to derive total and tropospheric NO2 columns from zenith-sky measurements is described. Second, I see a major drawback in the empirical approach used here, which is that the total AMF for zenith-sky measurements is expected to be a strong function of not only the solar zenith angle but also the tropospheric column itself. In first approximation, one can assume that the stratospheric AMF will mostly follow the solar geometry (geometrical AMF) while the PBL AMF is approximately constant and close to one at any solar position. In consequence, for intermediate and low sun conditions, the stratospheric and PBL AMFs differ quite strongly and the total AMF depends on the relative amount of

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NO2 present in the PBL and in the stratosphere. I think that the classification could be improved substantially by taking this dependence into account (probably within an iterative scheme). The dependence on the season accounts somehow for this effect (since it implicitly accounts for the seasonality of the stratospheric NO2 column), but only in a very crude way.

Section 3.1, L. 19: " . . . VCD_Emp shows less SZA dependence than VCD_DS. . . " -> I think that VCD_NDACC is meant here instead of VCD_DS

Section 3.1, last paragraph: note that the zenith-sky AMF could also be affected by aerosols present in PBL (together with NO2), due to their impact on the light path. Although the impact of aerosols is likely to be moderate, it certainly contribute to the uncertainty of the measurements and this should be mentioned.

Section 3.2, L. 5-10: the finding that zenith-sky columns despite their larger uncertainties (in comparison to direct-sun data) show a better agreement with satellite measurements is quite surprising and interesting. I am not convinced by the argument stating that the air mass sampled by zenith-sky measurements is more representative of the air mass sampled by the satellite than the direct-sun. Considering the size of typical OMI pixels (approx. 20x20 km2), one can argue that both direct-sun and zenith-sky measurements are more local in nature, and therefore maybe another explanation can be found. Could it be that direct-sun and zenith-sky measurements sample different meteorological conditions (e.g. different wind patterns), or maybe that zenith-sky are generally more homogeneously distributed around the overpass time of the satellite so that the average value becomes more representative? Please comment on these issues.

Section 4.1: the method used to convert NO2 column measurements into surface concentrations implies a lot of approximations/assumptions. The uncertainties associated to these assumptions should be better described. Equation 3 starts from the total NO2. For the zenith-sky case, this column already contains quite a large uncertainty (cf. pre-

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vious point). Then a correction for the stratospheric column and the free-tropospheric column is made. The stratospheric column is taken from photochemically-corrected stratospheric NO2 OMI measurements without any further verification. Can we exclude any possible systematic bias between OMI and ground-based measurements? Was there any attempt to verify that OMI and Pandora measurements do agree well under clean conditions? Also monthly mean OMI data are used. Can we safely neglect the day-to-day variability in the stratospheric NO2 content (e.g. in Spring it is known that transport patterns can produce short term variations of the stratospheric NO2)? As regards the free-tropospheric NO2 content, it is taken directly from model data. How large and uncertain is this contribution? Finally the last step is based on the assumption that the modelled column to surface concentration ratio is representative of the actual ratio. Is this assumption expected to be correct in all cases? What about the impact of the limited horizontal resolution of the model? In any case, I think that all these uncertainties are responsible for the large scatter of the correlations shown in Figure 6. As such they should be discussed with a little bit more of attention.

This being said, I agree that the average behaviors found (and illustrated in Figs. 9-11) are quite convincing and demonstrate well the potential of the data set.

Finally this study makes use of Pandora direct-sun completed by zenith-sky measurements. At no point in the paper, the potential of extending the data set with multi-axis measurements providing more information on the tropospheric NO2 is mentioned, although this would be a logical evolution for a follow up activity. Please consider adding this in the perspectives.

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