Response to anonymous referee #3

The reviewer comments are given in black, followed by the authors' response in blue. Text copied from the revised manuscript is in blue italic.

Based on a comment from referee #1, Fig. 8 and 9 have been merged and, as a result, the numbering of the following figures has been changed. In addition, the structure of Sect. 2 has been modified in order to include a new subsection with the description and implementation of the GRASP algorithm (Sect. 2.6).

General Comments

This study aims to evaluate the impact of the NO_2 correction for the aerosol retrievals based on ground-based instruments (i.e., AERONET and SKYNET). They utilized multiannual data collected at urban and suburban sites in Rome, Italy. For the NO_2 correction, they used ground-based Pandora instruments as well as the TROPOMI data. The NO_2-corrected aerosol retrievals are compared with the operational methods to assess their effects. This manuscript analyzed valuable collocated data from the AERONET, Pandora, and SKYNET, and presented various results using the data. However, I do not fully agree with their main conclusions, which insist significance of the NO_2 corrections for the AERONET and SKYNET products. In the major part of the results, the effects of NO_2 correction seem to be negligible to me, which is the reason why the previous algorithms neglected NO_2 effects or utilized climatology. I believe the authors need to demonstrate their conclusions based on the statistical test to assess the impacts of the NO_2 corrections on aerosol retrievals. Therefore, I would recommend considering the publication of this manuscript after clarifying the below comments.

We would like to acknowledge the referee for their helpful and thorough review. We believe that their comments improved the quality of this work.

In general, we agree that, in the major part of the results, the effects of NO_2 correction seem not to be so significant, which is the reason why the previous algorithms neglected NO_2 effects or utilized climatology. However, we think that the average systematic underestimation of AOD found for SKYNET (0.007) cannot be considered negligible. Moreover, according to the findings of this study, significant errors may be introduced over polluted areas for cases with high NO_2. Those cases are quite a few for Rome, but the error introduced is comparable to the AOD uncertainties. In addition, there are areas with higher NO_2 levels and more frequent events of high NO_2 compared to the Rome stations used in this study. For studies that do not deal with averages and use individual days in the analysis, the NO_2 correction could be important when intraday NO_2 variability is high especially in cities or in episodic NO_2 cases.

Statistics and references have been included in the text to support the importance of NO_2 correction in the above cases. Also, revisions have been made in parts of the manuscript where the significance of the results may has been excessively or inappropriately overstated.

We answer to each point in detail below.

Major comments

Lines 316-317: This overestimation should be quantified by suggesting statistical values in the main script although the values are listed in the tables. The values should be compared with the reported uncertainties of the AERONET (i.e., 0.01 in the visible and NIR and 0.02 in the UV) and SKYNET.

The resulted values of mean deviations from our analysis and reported uncertainties for the two networks have been included in the text as follows:

"The estimated AOD and AE deviations are below 0.01 and 0.1, respectively, for the majority of observations, i.e., about 96 - 98% of occurrences for both CNR-ISAC and APL-SAP (see also distributions in Fig. 6). The average AOD bias is between 0.002 ± 0.003 and 0.003 ± 0.003 (with the higher values observed at 380nm), while the average AE bias is ~0.02 ± 0.03. Overall, the mean AOD bias is low compared to the estimated uncertainties for the standard AERONET product, i.e., 0.01 - 0.02 (with the higher errors observed in the UV) (Sinyuk et al., 2020). However, the mean AOD bias for the cases of high NO_2 levels (> ~0.7 DU) is ~0.011 ± 0.003 at 440 nm and ~0.012 ± 0.003 at 380 nm for APL-SAP and ~0.009 ± 0.003 at 440 nm and ~0.010 ± 0.003 at 380 nm for CNR-ISAC, which is comparable to the AERONET reported uncertainties. The estimated mean bias of AE retrievals for the cases with high NO_2 (> ~0.7 DU) is ~0.08 ± 0.04 for both Rome sites. The threshold for NO_2 has been selected as the average Pandora NO_2 (~0.4) calculated from the whole data set plus two times the standard deviation.....

… Similarly to AERONET, the derived AOD and AE biases for SKYNET are below 0.01 and 0.1, respectively, for the majority of observations, i.e., about 85% of occurrences for AOD and about 90% for AE (see also distributions in Fig. 7). The overall average AOD bias is ~0.007 ± 0.003, which can be assumed low considering that Nakajima et al. (2020) have estimated a root-mean-square difference (RMSD) of about 0.03 for wavelengths < 500 nm in city areas in AOD comparisons with other networks. However, the mean AOD bias for the cases with high NO_2 levels (> ~0.7 DU) is found to be about 0.018 ± 0.003, which is comparable to the RMSD value reported by Nakajima et al. (2020). The overall average AE bias calculated in this study is ~0.05 ± 0.04, whereas the AE bias averaged over the high NO_2 cases is about 0.10 ± 0.05."

Section 3.4: The authors summarized the trend analysis in the abstract and conclusion sections. However, this section suggests that the impact of NO_2 absorption on the aerosol retrievals is insignificant for their measurements, but suggested "possible importance". I think this can mislead the readers unless they show other cases showing the significance of the NO_2 absorption on the aerosol trend analysis.

Section 3.4 has been revised as follows:

"In this section, a first attempt is conducted to investigate the effect of the modified AOD and AE retrievals based on the Pandora total NO_2 observations on the annual trends of those aerosol properties. The annual trends of AERONET/SKYNET AOD and AE over both APL-SAP and CNR-ISAC sites, calculated by applying the approach described in paragraph 2.5, as well as their uncertainties (standard errors of the regression slope) are presented in Table 3.

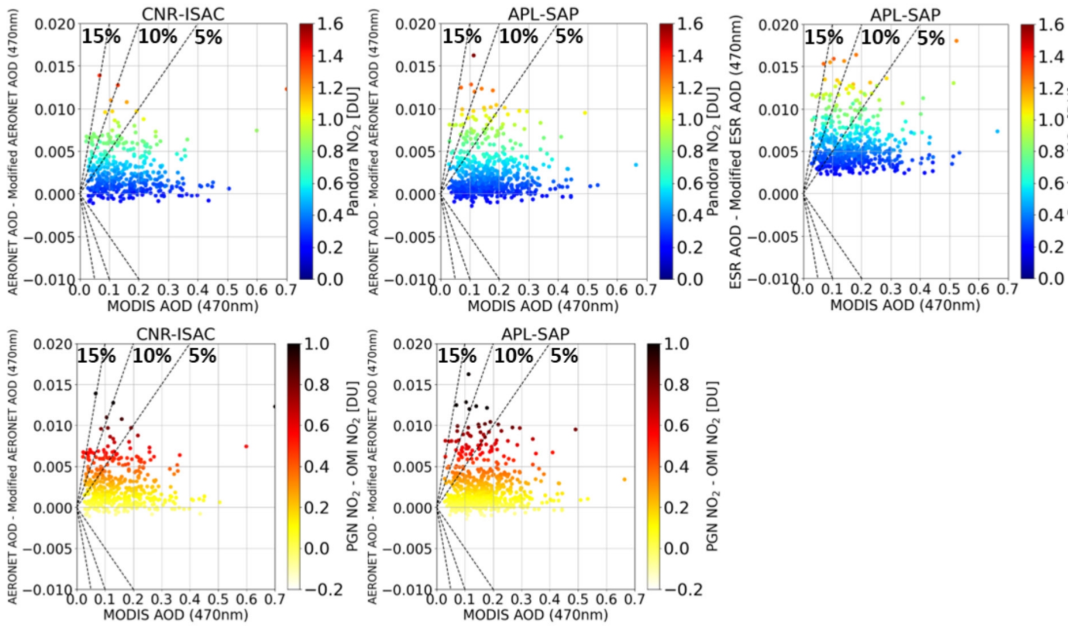
It should be noted here that the aerosol data sets from the two networks correspond to slightly different time periods. In addition, there are significant gaps in the time series from CNR-ISAC due to instrumental problems and the COVID-19 lockdown period (February – May 2020) has been excluded from the data analysis. Therefore, the results in Table 3 are mainly intended to highlight how a different NO_2 correction may affect the aerosol trends and should be interpreted separately for each individual site. Interpretation of the trend significance for the Rome area is not possible using this short period of time (~5.5 years), considering that the estimated trends are quite small and the uncertainties introduced by linear regression are relatively high.

One aspect shown here is that the difference in the AOD and AE trends for the two data sets (original and NO_2-modified) is comparable with the calculated trends. As expected, AE trends with and without NO_2 correction show relatively higher differences, as AE is much more sensitive to spectral AOD changes. However, the linear fitting uncertainty on AE is also high. NO_2 effects on AOD trends would be more obvious in the case of a significant NO_2 trend during a certain period. A thorough long-term trend analysis is out of the scope of this work and could be the topic for a future study."

Lines 415-418: As this result is one of the main conclusions, the authors should report the statistical significance of differences between original and modified data.

We agree with the reviewer that the average effects of NO_2 correction are relatively small, which is why the previous algorithms neglected NO_2 effects or utilized NO_2 climatology. As already highlighted in the paper, the proposed correction and the consequent improvement are, on average, not statistically significant. This result is explainable considering that the suggested correction depends on the amount of NO_2 and that the relative frequency distributions of absolute Pandora-OMI deviation decrease for high NO_2 values (lower panel of Fig. 4). Basically, we are focusing on those situations in which the NO_2 climatology is not able to represent the real scenario. The present work highlights that when significant discrepancies between climatology and PGN NO_2 values are observed, the improvement due to the proposed correction is also statistically significant, i.e., larger than combined instantaneous uncertainties.

To better highlight this result, we decided to update Fig. 10, adding in the lower panels the absolute correction as a function of the corresponding MODIS DB AOD data and the absolute difference between PGN and climatology NO_2 data for CNR-ISAC (left panel) and APL-SAP (central panel) sites. This type of plot is not included for ESR data, since it would be identical to the upper right panel of the figure, as NO_2 absorption is not accounted in the official SKYNET retrieval chain.



The last part of section 3.5 has been revised as follows:

"This inter-comparison exercise demonstrated that the proposed correction slightly improves the agreement between MODIS DB AOD data and AERONET and SKYNET AOD products, even if, on average, it is not statistically significant. Nevertheless, as shown in Fig. 10, the improvement becomes significant when the differences between the NO_2 values observed by Pandora and the OMI NO_2 climatology are also significant (lower panels of Fig. 10). Furthermore, since the proposed correction depends on the amount of NO_2, the improvement is more evident in the correspondence of high values of NO_2 (upper panels of Fig. 10), typical of highly polluted areas such as the urban area of Rome (APL-SAP). Also, a slight improvement is also achieved in the suburban area of Rome (CNR-ISAC). Finally, in the case of SKYNET AOD products, the systematic overestimation, due to neglected NO_2 extinction in the official retrieval chain, is eliminated."

The caption of Fig. 10 was also changed as follows:

"Upper row: Absolute correction as a function of the corresponding MODIS DB AOD data and PGN NO_2 data for CNR-ISAC (left panel) and APL-SAP (middle and right panels) sites. In the left and middle panels, the inter-comparison was performed using AERONET AOD products, in the right panel SKYNET AOD was used. The color scale represents the PGN NO_2 retrieved in correspondence with the AERONET/SKYNET AOD products. The analysis was performed considering a maximum distance between the center of the MODIS DB pixel and the site location of 5 km and Δt_max of ±30 minutes. Lower row: As in the upper row, but the color scale represents the absolute difference between PGN and OMI climatological NO_2 data in correspondence with the AERONET AOD products."

Section 3.6: I believe this section is one of the most meaningful results to me. If the impact of the NO_2 corrections on the AOD and trend analysis is not statistically significant, I recommend elaborating on this section (e.g., adding more cases or locations, etc.).

We tried to focus on Rome datasets, as the setup of having two sites in such a close distance, two NO_2-retrieving photometers and three AOD-retrieving ones is unique.

In general, we think that we demonstrated that not accounting for NO_2 or using NO_2 climatologies, which are systematically lower than the actual NO_2 measured in real time, introduces a systematic error on AOD retrievals. This error is low and within the AOD reported uncertainties on an average level, but it becomes more significant for a number of cases with relatively high NO_2.

This is also the case for SSA. We aimed to demonstrate that different than near real-time measured NO_2 could affect SSA retrievals in certain wavelengths. So, inversion algorithms for retrieving properties like SSA need to account for NO_2 for "high" NO_2 cases, where "high" is defined by the NO_2 climatology used.

For all sites globally, the effect would be directly proportional to the difference of the climatological NO_2 from the actual NO_2 for each specific case/measurement. So another study could probably shed light on how accurate are satellite-based climatologies compared with existing ground-based data. Such a study, which is beyond the scope of our analysis, could probably be then used in order to revise the NO_2 inputs in the aerosol retrieval algorithm. Of course, in the case of co-located NO_2-retrieving instruments at the same site, the AOD retrieval algorithms could be fed with real-time measured NO_2.

Lines 463-464: I don't agree that the difference (i.e., lower than 0.003 in table 1) is "quite significant errors" as the errors are typically smaller than the reported uncertainties of the AERONET and/or SKYNET.

The statement has been revised as follows:

"However, significant errors could be introduced in the AOD retrievals, especially over urban areas, where NO_2 variability can be high and also the occurance of high NO_2 events can be more frequent. Such errors may occur only in the cases where NO_2 is not taken into account or the used NO_2 climatology underestimates such high-NO_2 events."

This statement refers to the significant errors that may be introduced over polluted areas for cases with high NO_2. Those cases are quite a few, but the error introduced is comparable to the AOD uncertainties. In addition, there are areas with higher NO_2 levels and more frequent events of high NO_2 compared to the Rome stations used in this study.

The mean bias derived for the high NO_2 cases (> ~0.7 DU) in our study is ~0.011 ± 0.003 at 440 nm and ~0.012 ± 0.003 at 380 nm for APL-SAP and ~0.009 ± 0.003 at 440 nm and ~0.010 ± 0.003 at 380 nm for CNR-ISAC for AERONET AOD and ~0.08 for both Rome sites for AERONET AE. In the case of SKYNET, the mean bias for the cases with high NO_2 levels (>~0.7 DU) is ~0.018 and ~0.10 for AOD and AE, respectively. These numbers have been included in the manuscript (in Section 3.1 as well as in the abstract and conclusions).

Lines 477-479: Again, according to table 2, it is lower than 0.0011 for AERONET, and 0.0051 for SKYNET, which is much lower than 0.01. I don't believe it is significant given that the AEORNET uncertainty is higher than 0.01.

The corrections in Table 2 are based on space-borne NO_2 data. The purpose for including them is to show the possibility for corrections on a global scale. The underestimation of TROPOMI NO_2 compared to Pandora leads to lower and less accurate AOD corrections. However, in the case of high NO_2 (> ~0.7 DU) the corrections are not negligible. More specifically, a mean AOD bias of ~0.004 ± 0.001 at 440 nm and ~0.005 ± 0.002 at 380 nm for AERONET APL-SAP and ~0.003 ± 0.001 at both 440 nm and 380 nm for AERONET CNR-ISAC was estimated. The mean bias of AE retrievals is ~0.05 ± 0.04 and ~0.02 ± 0.01 for APL-SAP and CNR-ISAC, respectively. In the case of SKYNET, the average bias is about 0.011 ± 0.002 and ~0.07 ± 0.04 for AOD and AE, respectively. These numbers have been included in the manuscript in Section 3.2.

Table 1, which is based on less uncertain ground-based NO_2 measurements, shows a 0.002-0.003 (depending on wavelength) and 0.007 difference on the average for AOD for AERONET and SKYNET, respectively. Especially for SKYNET, we think that an average systematic underestimation of AOD of 0.007 cannot be considered negligible, having also in mind that there are parts of the world with much higher average NO_2.

WMO (2005) states that 95% of AOD differences compared with a reference standard should lie within ± (0.005 + 0.01/m) of AOD, where m is the optical air mass. The first term of equation (0.005) represents the maximum tolerance for the uncertainty due to the atmospheric parameters used for the AOD calculation (additional atmospheric trace gas corrections, i.e., Ozone and NO_2, and Rayleigh scattering). The second term (0.01/m) describes the calibration-related relative uncertainties (WMO recommends an upper limit for the calibration uncertainty of 1 % (e.g., Cuevas et al., 2019, Kazadzis et al., 2018a)).

Based on the above, we consider the average systematic AOD underestimation found in our study, mainly the 0.007 (Table 1 / using Pandora NO_2) and 0.005 (Table 2 / using TROPOMI NO_2) for SKYNET, important to be reported here.

The above discussion and references have been added in Section 3.1.

WMO: WMO/GAW Experts Workshop on a Global Surface-Based Network for Long Term Observations of Column Aerosol Optical Properties, GAW Report No. 162, WMO TD No. 1287, available at: https://library.wmo.int/index.php?lvl=notice_display&id=11094, 2005.

Cuevas, E., Romero-Campos, P. M., Kouremeti, N., Kazadzis, S., Räisänen, P., García, R. D., Barreto, A., Guirado-Fuentes, C., Ramos, R., Toledano, C., Almansa, F., and Gröbner, J.: Aerosol optical depth comparison between GAW-PFR and AERONET-Cimel radiometers from long-term (2005–2015) 1 min synchronous measurements, Atmos. Meas. Tech., 12, 4309–4337, https://doi.org/10.5194/amt-12-4309-2019, 2019.

Kazadzis, S., Kouremeti, N., Nyeki, S., Gröbner, J., and Wehrli, C.: The World Optical Depth Research and Calibration Center (WORCC) quality assurance and quality control of GAW-PFR AOD measurements, Geosci. Instrum. Method. Data Syst., 7, 39–53, https://doi.org/10.5194/gi-7-39-2018, 2018a.

Lines 505-506: I don't agree that NO_2 absorption is very important for the AE, AOD, and SSA retrievals.

We agree with the referee that this is a very strong statement based on the results presented here.

The text has been revised as follows:

"In general, the effect of NO_2 absorption can be relatively important in the retrievals of aerosol properties, especially AE, AOD and SSA at 440 nm and 380nm, when NO_2 is not included in the retrieval algorithms or in cases where NO_2 absorption is significantly higher than the NO_2 climatology used."

Minor comments

Lines 61-62: Please add references of the SKYNET, GAW-PFR, AERONET regarding the NO_2 corrections for the aerosol retrievals.

The following references have been added in the text:

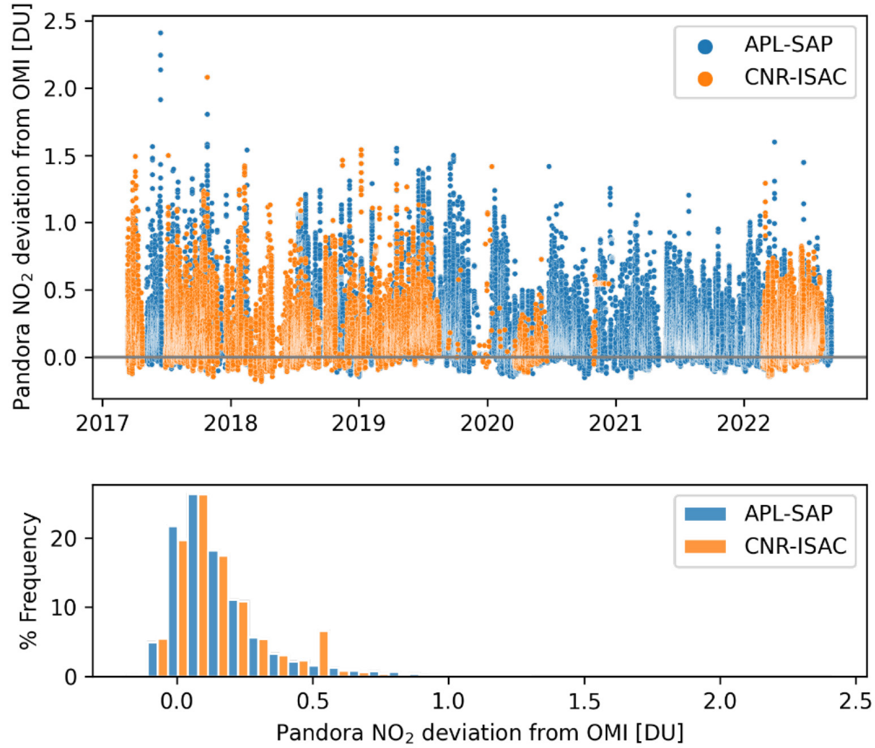
AERONET - Giles, D. M., Sinyuk, A., Sorokin, M. G., Schafer, J. S., Smirnov, A., Slutsker, I., Eck, T. F., Holben, B. N., Lewis, J. R., 645 Campbell, J. R., Welton, E. J., Korkin, S. V., and Lyapustin, A. I.: Advancements in the Aerosol Robotic Network (AERONET) Version 3 database – automated near-real-time quality control algorithm with improved cloud screening for Sun photometer aerosol optical depth (AOD) measurements, Atmos. Meas. Tech., 12, 169–209, https://doi.org/10.5194/amt-12-169-2019, 2019.

GAW-PFR - Kazadzis, S., Kouremeti, N., Nyeki, S., Gröbner, J., and Wehrli, C.: The World Optical Depth Research and Calibration Center (WORCC) quality assurance and quality control of GAW-PFR AOD measurements, Geosci. Instrum. Method. Data Syst., 7, 39–53, https://doi.org/10.5194/gi-7-39-2018, 2018a.

SKYNET - Nakajima T., Campanelli, M., Che, H., Estellés, V., Irie, H., Kim, S.-W., Kim, J., Liu, D., Nishizawa, T., Pandithurai, G., Soni, 740 V. K., Thana, B., Tugjsurn, N.-U., Aoki, K., Go, S., Hashimoto, M., Higurashi, A., Kazadzis, S., Khatri, P., Kouremeti, N., Kudo, R., Marenco, F., Momoi, M., Ningombam, S. S., Ryder, C. L., Uchiyama, A., and Yamazaki, A.: An overview of and issues with sky radiometer technology and SKYNET, AMT, 13, 4195-4218, 2020.

Figure 4: I'm not quite sure if the upper panels of figure 4 are meaningful. I would recommend adding temporal plots of the biases (Pandora - OMI) vs. time over whole measurement periods. I believe that chart can show how the simple assumption of the AERONET can affect the temporal analysis of the AOD over a few years.

The upper panels of Fig. 4 have been replaced with the time series of Pandora – OMI deviations (see the following figure).



Lines 194-197: Underestimation of satellite NO_2 retrievals (e.g., OMI, TROPOMI) compared to ground-based retrievals (e.g., MAX-DOAS, Pandora, etc) is quite a well- known phenomenon and it is attributable to the different field of view (FOV). I think it is worth noting that NO_2 correction using the Pandora is more accurate than the satellite retrievals since the FOV of the Pandora is similar to that of the AERONET in the main script.

Discussion on the underestimation of satellite NO_2 retrievals due to their limited spatial resolution has been added in the manuscript.

In Section 2.3.1:

"This underestimation of the NO_2 levels over urban locations, characterized by strong spatial gradients, can be attributed to the fact that OMI climatology cannot capture the temporal and spatial NO_2 variability within an urban context (e.g., Drosoglou et al., 2017; Herman et al., 2019)."

In Section 3.2:

"Satellite sensors perform measurements globally and provide information on the air quality even over regions that lack ground-based observations. However, as already mentioned for OMI in Sect. 2.3.1, the spatial resolution of the satellite retrievals is limited by the pixel size… Despite the improved spatial resolution of TROPOMI, the NO_2 corrections using TROPOMI data are expected to be less accurate than those performed with the Pandora product. For example, Lambert et al. (2021) showed a bias between TROPOMI and Pandora total NO_2 column ranging from -23% over polluted stations to +4.1% over clean areas with a median bias of -7.1%, in the frame of the standard validation process of TROPOMI Level 2

NO_2 products. Other studies have concluded similar results. For example, Zhao et al. (2020) showed negative bias for the standard TROPOMI total NO_2 product in the range 23 - 28% over urban and suburban environments and a positive bias of 8 - 11% at a rural site, while Park et al. (2022) showed 26 - 29% negative bias and R^2 within 0.73-0.76 over the Seoul Metropolitan Area in Korea."

Lines 305-306: Is there any reason for the opposite definition between $\Delta\tau$ and $\Delta\alpha$?

Both $\Delta\tau$ and $\Delta\alpha$ are defined in the calculations as the difference of the standard minus the modified value. The equation in the text was wrong. The manuscript has been revised accordingly.

Lines 311-312: This sentence is not clear to me. Typical "pollution events" do not always accompany high loadings of NO_2, which depends on emissions sources and environmental conditions. Also, Figure 4-6 does not directly demonstrate the relationship between the AOD and NO_2. Scatter plots between AOD and NO_2 might be helpful for this statement.

This is a finding from Fig. 6. Reddish colors (indicating high NO_2 values) do not correspond to high AOD loadings. The text has been revised so that it is clearer that we refer to high NO_2 episodes and a reference to the figure has been added:

"Interestingly, based on Fig. 6, the highest Pandora NO_2 retrievals (reddish colors) are not associated with the highest AOD values, indicating that in Rome the high AOD loadings are not strictly associated with high NO_2 pollution events. In fact, high AODs are frequently related to long-range transport of elevated layers of desert dust, fires plumes or a combination of both (e.g., Barnaba et al., 2011; Gobbi et al., 2019; Campanelli et al., 2021; Andrés Hernandez et al., 2022). Hence, it might be worth to modify aerosol retrievals for high NO_2 in those pollution-related events with low to medium AOD levels. More about AOD and aerosol type climatology for the Rome area can be found in Di Ianni et al., (2018) and in Campanelli et al. (2022)."

Andrés Hernández, M. D. et al.: Overview: On the transport and transformation of pollutants in the outflow of major population centres – observational data from the EMeRGe European intensive operational period in summer 2017, Atmos. Chem. Phys., 22, 5877–5924, https://doi.org/10.5194/acp-22-5877-2022, 2022.

Barnaba, F., Angelini, F., Curci, G., and Gobbi, G. P.: An important fingerprint of wildfires on the European aerosol load, Atmos. Chem. Phys., 11, 10487–10501, 10.5194/acp-11-10487-2011, 2011.

Campanelli, M., Iannarelli, A.M., Mevi, G., Casadio, S., Diémoz, H., Finardi, S., Dinoi, A., Castelli, E., di Sarra, A., Di Bernardino, A., Casasanta, G., Bassani, C., Siani, A.M., Cacciani, M., Barnaba, F., Di Liberto, L., Argentini, S.: A wide-ranging investigation of the COVID-19 lockdown effects on the atmospheric composition in various Italian urban sites (AER – LOCUS), Urban Climate, Volume 39, 100954, ISSN 2212-0955, https://doi.org/10.1016/j.uclim.2021.100954, 2021.

Campanelli, M., Diémoz, H., Siani, A. M., di Sarra, A., Iannarelli, A. M., Kudo, R., Fasano, G., Casasanta, G., Tofful, L., Cacciani, M., Sanò, P., and Dietrich, S.: Aerosol optical characteristics in the urban area of

Rome, Italy, and their impact on the UV index, Atmos. Meas. Tech., 15, 1171–1183, https://doi.org/10.5194/amt-15-1171-2022, 2022.

Di Ianni A, Costabile F, Barnaba F, Di Liberto L, Weinhold K, Wiedensohler A, Struckmeier C, Drewnick F, Gobbi GP.: Black Carbon Aerosol in Rome (Italy): Inference of a Long-Term (2001–2017) Record and Related Trends from AERONET Sun-Photometry Data. Atmosphere. 9(3), 81, https://doi.org/10.3390/atmos9030081, 2018.

Gobbi, G.P., Barnaba, F., Di Liberto, L., Bolignano, A., Lucarelli, F., Nava, S., Perrino, C., Pietrodangelo, A., Basart, S., Costabile, F., Dionisi, D., Rizza, U., Canepari, S., Sozzi, R., Morelli, M., Manigrasso, M., Drewnick, F., Struckmeier, C., Poenitz, K., Wille, H.: An inclusive view of Saharan dust advections to Italy and the Central Mediterranean, Atmospheric Environment, 201, 242-256, 10.1016/j.atmosenv.2019.01.002, 2019.

Lines 342-343: As spatiotemporal variabilities of the NO_2 are significantly high, the authors should state the spatial and temporal window of this collocation.

This information has been included in the text as follows:

"Based on the current satellite footprint (5.5 km × 3.5 km), a radius of 5 km around each ground-based station was selected for the spatial co-location. The TROPOMI NO_2 data were time-interpolated to AERONET and SKYNET measurements."

Line 403: font of "Wei et al., 2019" needs to be "times new roman"?

The font type has been corrected.

Lines 407-408: Which data were used to calculate the NO_2-modified AERONET? (Pandora or TROPOMI?)

The Pandora data were used. This is now mentioned in the text as follows:

"The NO_2-modified AERONET and SKYNET AOD at 470 nm were also computed with the same approach and the AOD and AE retrievals that have been modified using the Pandora NO_2 data."