

## Response to Reviewer's comments

Review of: “Assessment of NO<sub>2</sub> uncertainty impact on aerosol optical depth retrievals at a global scale”.  
The paper is an extension of the already published “Evaluating the effects of columnar NO<sub>2</sub> on the accuracy of aerosol optical properties retrievals” Drosoglou et al. 2023, who analyzed the effect for the site of Rome. Extending the results to more sites worldwide is very interesting and can provide very useful information. There are however some points that need a clarification.

(1) The title is misleading since it seems that the authors are evaluating how “the uncertainty in NO<sub>2</sub> estimation” impacts over AOD measurements. I would suggest something like “Assessment of the impact of NO<sub>2</sub> contribution on aerosol optical depth observations in several site worldwide locate”. AOD is not retrieved, because there isn't any inversion analysis to perform. Moreover “global scale” is too much for the number and location of the sites studied in the work.

Response to Comment 1: We thank the reviewer for the suggestion proceeding with which we have updated the title of the manuscript as below by replacing “retrievals” with “measurements” and “global” with “several sites worldwide”

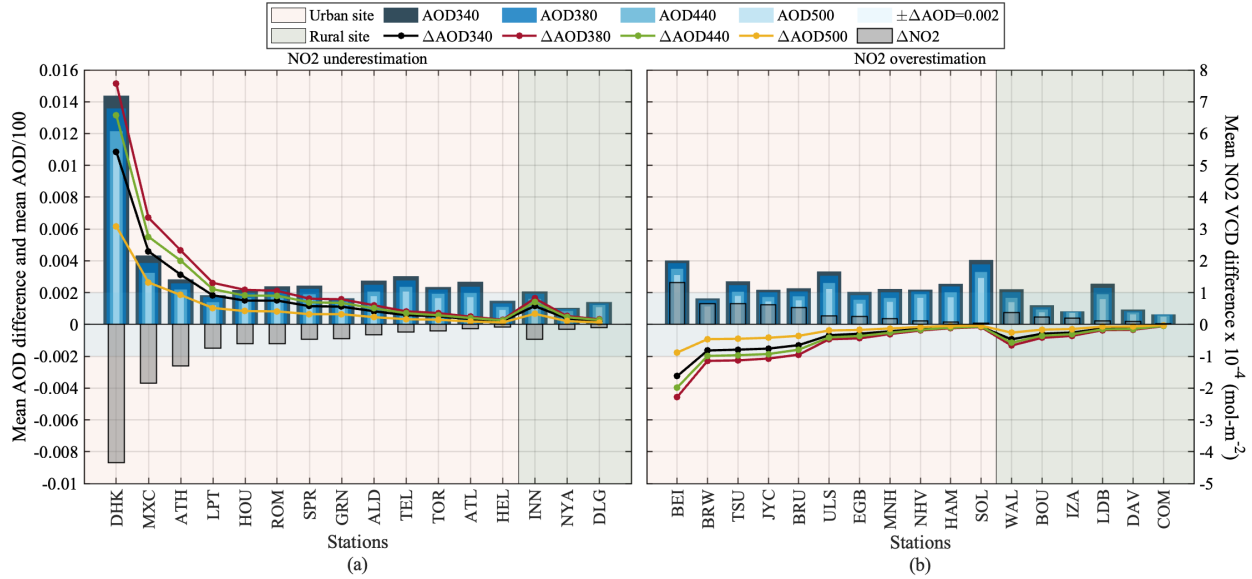
“Assessment of **the impact of NO<sub>2</sub> contribution** on aerosol optical depth **measurements** at **several sites worldwide**”

(2) In the abstract lines 27-28 it is not clear what a “deviation in NO<sub>2</sub>” is. It is understandable reading the text, but it should be clarified also Why the authors preferred “deviation” instead of a simple “difference”?

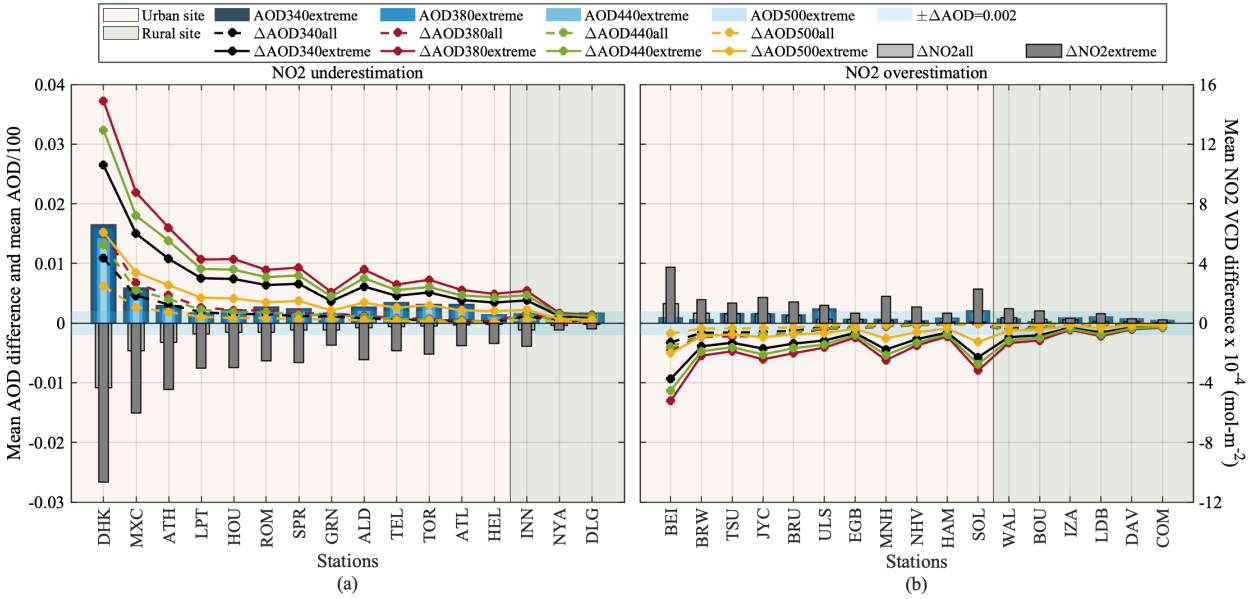
Response to Comment 2: We agree with the reviewer with the use of the word “difference” instead of “deviation” and have corrected this discrepancy throughout the manuscript.

(3) Could you explain the reason why you are looking for the NO<sub>2</sub> effect only at 380 and 440 nm?

Response to Comment 3: We thank the reviewer for this comment. We looked at the NO<sub>2</sub> effect at 380 nm and 440 nm as these wavelengths are the most affected by NO<sub>2</sub> absorption. However, in order to be more consistent with the AERONET methodology and since NO<sub>2</sub> absorption is significant in the UV-VIS spectral range, we have expanded the analysis to 340 nm and 500 nm as 340 nm, 380 nm, 440 nm and 500 nm are the wavelengths that are corrected for NO<sub>2</sub> absorption in AERONET (Reference: Table 1 in Giles et al., 2019). We have provided a table for NO<sub>2</sub> correction based AOD differences at 340 nm and 500 nm in Appendix Table A3 and have updated Table 3 as well as Figure 2 and Figure 5 in the updated manuscript which are also provided below as Figure i and ii. It is evident that AOD bias is the most affected at 380 nm by NO<sub>2</sub> differences followed by 440 nm, 340 nm and 500 nm. Accordingly, we have made changes in the manuscript wherever needed e.g., Line 25-27, Line 287-288, Line 341-342, Line 473, Line 486-487.



**Figure i:**  $\text{NO}_2$  VCD ( $\text{mol}\cdot\text{m}^{-2}$ ) and AOD differences for all station with  $\text{NO}_2$  (a) underestimation and (b) overestimation. The  $\text{NO}_2$  differences are calculated as  $\text{OMIc} - \text{PGN}$  and the corresponding AOD differences as original AERONET AOD – PGN corrected AOD (as described in Section 2.2.2).



**Figure ii:** Comparison of  $\text{NO}_2$  VCD ( $\text{mol}\cdot\text{m}^{-2}$ ) and AOD differences ( $\text{OMIc} - \text{PGN}$ ) in extreme cases with 10% highest  $\text{NO}_2$  (a) underestimation and (b) overestimation by  $\text{OMIc}$  as compared to all datasets.

(4) Line 149: “the nearest matching PGN” to AERONET. Is there any threshold within searching the nearest measurement? The nearest could also be some with some days of difference.

Response to Comment 4: We thank the reviewer for pointing to this. We have performed the comparison between AERONET and PGN time stamps within a day (i.e., on a daily basis) and hence every comparison point is within a day. However, while accepting only points within a maximum of  $\pm 1$  min difference, the coincident comparison points obtained were very few. Hence, to maintain a balance between the accuracy and the number of comparison points, we first found the nearest matching time stamp of Pandora

measurement corresponding to Aeronet time stamp within a day and then time interpolated the Pandora measurement to Aeronet time stamp. In this process, for every Aeronet measurement, we were able to retrieve the corresponding time interpolated Pandora NO<sub>2</sub> measurement. It is to note here that this is for diurnal variation of NO<sub>2</sub> which is anyways not possible with polar orbiting satellites such as OMI/TROPOMI and even with geostationary satellite the exact comparison time stamp will be very few. Hence, we have corrected the sentence in the manuscript as below in Lines 153-155

“Corresponding to every measurement of AERONET (time of measurement) **within a day**, the nearest matching PGN measurement (similar time of measurement) was selected and then the PGN data was time interpolated to the AERONET time stamp **for that day.**”

(5) In the Sections 2.2.2 please describe (or cite a reference) for explaining Eq 3. Moreover in Eq 4: 1) explain the reason of the adding and subtracting each term, and 2) what is TNO<sub>2</sub>(l) in the third term after the first equivalence. Why this term disappears after the second equivalence? The same explanations are necessary for the equivalences in Eq5. How delta\_NO2 is defined. To facilitate the reading, please do a table that summarize the three lines 184-186 (adding also the variables in Eq 4 and 5 that are not defined) and the parameters in Eq. 3.

Response to Comment 5: We thank the reviewer for this comment which we tried to address one by one:

### **Eq. 3**

We have added the reference as well as explanation for Eq. 3 as below in Line 183-187

“..... NO<sub>2</sub> absorption to AOD and the NO<sub>2</sub> optical depth estimations (Eq. 3) (Cuevas et al., 2019) which is calculated as

$$\tau_{\text{NO}_2}(\lambda) = \frac{\sigma_{\text{NO}_2}(\lambda)}{1000} * \frac{m_{\text{NO}_2}}{m_a} * \text{NO}_2 \quad (3)$$

where  $\sigma_{\text{NO}_2}$  is the NO<sub>2</sub> absorption coefficient at wavelength ( $\lambda$ ) **obtained from (Gueymard, 1995) and the expression for  $m_{\text{NO}_2}$  is obtained from (Gueymard, 1995), while  $m_a$  is the optical air mass and NO<sub>2</sub> VCD is in DU.”**

### **Eq. 4**

#### **(1)**

We have added the reason of the adding and subtracting each term in Eq. 4 as below in Line 192-196

“..... **(considering that  $\tau_{\text{aer}}$  is obtained by subtracting  $\tau_{\text{NO}_2}$  from total optical depth, hence  $\tau_{\text{NO}_2}$  is added to  $\tau_{\text{aer}}$  and newly calculated  $\tau_{\text{NO}_2}$  is subtracted to obtain the PGN corrected  $\tau_{\text{aer}}$  in Eq. 4):**

$$\tau_{\text{aer,PGN}}(\lambda) = \tau_{\text{aer,AERONET}}(\lambda) + \tau_{\text{NO}_2,\text{AERONET}}(\lambda) - \left( \tau_{\text{NO}_2,\text{AERONET}}(\lambda) * \frac{\text{NO}_2^{\text{PGN}}}{\text{NO}_2^{\text{OMIc}}} \right) = \tau_{\text{aer,AERONET}}(\lambda) - \tau_{\text{NO}_2,\text{AERONET}}(\lambda) \left( \frac{\text{NO}_2^{\text{PGN}}}{\text{NO}_2^{\text{OMIc}}} - 1 \right) \quad (4)''$$

#### **(2)**

$\tau_{\text{NO}_2}$  is the AERONET calculated NO<sub>2</sub> optical depth which is corrected as  $\tau_{\text{NO}_2,\text{AERONET}}(\lambda)$  and is highlighted in red in the above Eq. 4 and also in the updated manuscript. This correction explains the

comment on disappearance of this term in second equivalence of Eq. 4 (it doesn't disappear but was wrongly written in the previous version of the manuscript).

### **Eq. 5**

Similarly, Eq. 5 is also corrected as

$$\Delta\tau_{\text{aer}}(\lambda) = \tau_{\text{aer,AERONET}}(\lambda) - \tau_{\text{aer,PGN}}(\lambda) = \tau_{\text{NO}_2,\text{AERONET}}(\lambda) \left( \frac{\text{NO}_{2\text{PGN}}}{\text{NO}_{2\text{OMIc}}} - 1 \right) = - \frac{\tau_{\text{NO}_2,\text{AERONET}}(\lambda)}{\text{NO}_{2\text{OMIc}}} (\Delta\text{NO}_2) \quad (5)$$

And the explanation for Eq. 5 is added as below in Line 200-202

**“Eq. 5 represents the difference in the  $\tau_{\text{aer}}(\lambda)$  between AERONET  $\tau_{\text{aer}}$  and PGN corrected  $\tau_{\text{aer}}$  where the expression for  $\tau_{\text{aer,PGN}}(\lambda)$  was obtained from Eq. 4 that led to the second equivalence of Eq. 5 and third equivalence was obtained using Eq. 1.”**

Definition for delta\_NO2 is presented in Eq. 1 in the manuscript and also referred to as mentioned in the above line for explanation of third equivalence of Eq. 5.

In order to facilitate the reading, we have added explanation for Eq. 4 and Eq. 5 as well as a Table 2 in the updated manuscript (also Table i below) summarizing all the variables used in Eq. 4 and Eq. 5. For the parameters of Eq. 3, a reference with brief explanation has been added as mentioned in the response for **Eq. 3** above.

**Table i: Summary and description of the terms used in the methodology**

Symbol	Description	Expression and/or unit
<b>NO<sub>2</sub></b>		
NO <sub>2OMIc</sub>	AERONET OMI climatology (OMIc) based NO <sub>2</sub>	mol-m <sup>-2</sup>
NO <sub>2PGN</sub>	PGN NO <sub>2</sub>	mol-m <sup>-2</sup>
ΔNO <sub>2</sub>	(AERONET – PGN) NO <sub>2</sub> difference	NO <sub>2OMIc</sub> – NO <sub>2PGN</sub> (mol-m <sup>-2</sup> )
<b>τ<sub>aer</sub>: aerosol optical depth (AOD)</b>		
τ <sub>aer,AERONET</sub> (λ)	original AERONET AOD based on OMIc NO <sub>2</sub> at wavelength λ	-
τ <sub>NO<sub>2</sub>,AERONET</sub> (λ)	original AERONET NO <sub>2</sub> optical depth based on OMIc NO <sub>2</sub> at wavelength λ	-
τ <sub>aer,PGN</sub> (λ)	corrected AOD based on PGN NO <sub>2</sub> at wavelength λ	-
Δτ <sub>aer</sub> (λ)	AERONET NO <sub>2</sub> based - PGN NO <sub>2</sub> based AOD difference at wavelength λ	τ <sub>a,AERONET</sub> (λ) – τ <sub>a,PGN</sub> (λ)
<b>α: Ångström exponent (AE)</b>		
α <sub>λ<sub>i</sub>-λ<sub>j</sub>,AERONET</sub>	AERONET retrieved AE between wavelengths λ <sub>i</sub> and λ <sub>j</sub>	-
α <sub>λ<sub>i</sub>-λ<sub>j</sub>,PGN</sub>	AE calculated from the PGN corrected AOD between wavelengths λ <sub>i</sub> and λ <sub>j</sub>	-
Δα <sub>λ<sub>i</sub>-λ<sub>j</sub></sub>	Difference between the AE calculated from original AERONET AOD and PGN corrected AOD	α <sub>λ<sub>i</sub>-λ<sub>j</sub>,AERONET</sub> – α <sub>λ<sub>i</sub>-λ<sub>j</sub>,PGN</sub>

\*AERONET: Aerosol Robotic Network, PGN: Pandonia Global Network, OMI: Ozone Monitoring Instrument

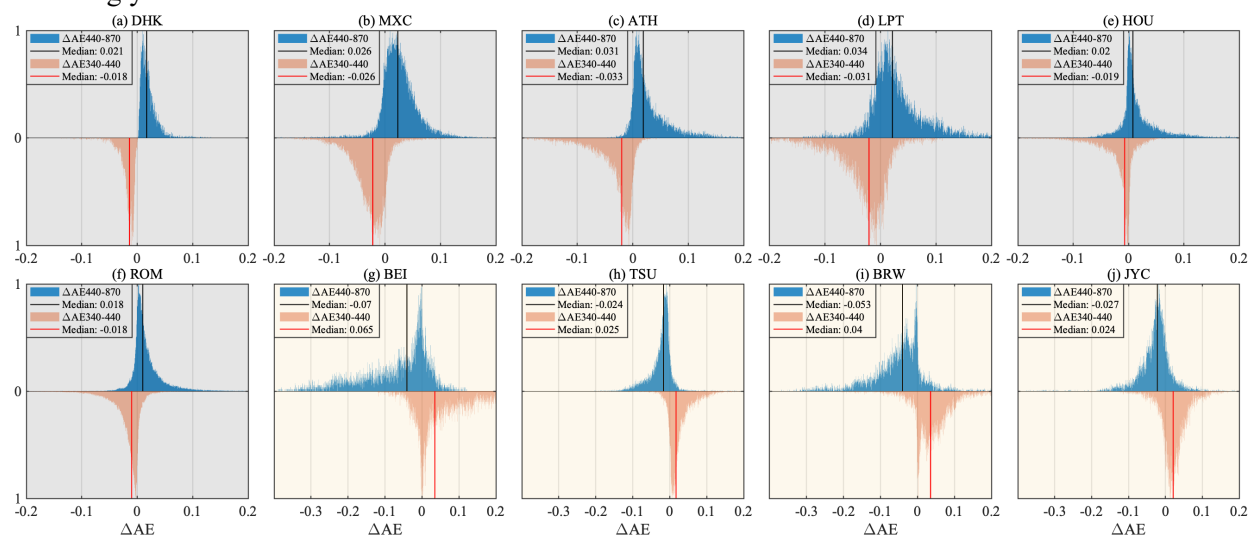
(6) In general, it is better doing an acronyms table.

Response to Comment 6: We thank the reviewer for the suggestion. We have added a table of acronyms at the end in the updated manuscript as follows

<b>AOD</b>	<b>Aerosol optical depth</b>
<b>ÅE</b>	<b>Ångström exponent</b>
<b>AERONET</b>	<b>Aerosol Robotic Network</b>
<b>OMI</b>	<b>Ozone Monitoring Instrument</b>
<b>PGN</b>	<b>Pandonia Global Network</b>
<b>GAWPFR</b>	<b>Global Atmospheric Watch – Precision Filter Radiometers</b>
<b>VCD</b>	<b>Vertical column density</b>
<b>OMIc</b>	<b>OMI climatology</b>
<b>OMId</b>	<b>OMI daily</b>
<b>DU</b>	<b>Dobson Unit</b>
$\tau$	<b>Optical depth</b>
$\alpha$	<b>Ångström exponent</b>
$\lambda$	<b>Wavelength</b>
$\Delta$	<b>Difference</b>

(7) Lines 199-201: Angstrom exponents using AOD corrected for NO<sub>2</sub> from PGN are calculated using two wls. But this method is different from the AERONET one, because the latter uses all the wls inside the intervals 380-675 / 440-870 and not only the range boundaries. Therefore, they can't be compared.

Response to Comment 7: We want to thank the reviewer for this comment. In order to align with the methodology of AERONET, we have updated our methodology as well as the wavelength pairs used for AE calculation. We now use the linear regression in logarithmic coordinates (as used by AERONET) using all 3 and 4 wavelengths for AE<sub>340-440</sub> and AE<sub>440-870</sub>, respectively. Instead of 380-675, we now use 340-440 as we have expanded the analysis of AOD from 380 nm and 440 nm to 340 nm, 380 nm, 440 nm and 500 nm (as described in Response 3). Figure 8 (below Figure iii) in the updated manuscript has been revised accordingly.



**Figure iii: Normalized frequency distributions of (a-j) the difference in AE at 440-870 nm and 340-440 nm retrieved from the AODs based on AERONET OMIc and PGN NO<sub>2</sub>. Shaded background area represents NO<sub>2</sub> underestimation (grey) (a-f), and overestimation (yellow) (g-j) cases.**

We have corrected this in methodology Section 2.2.2 as follows in Line 219-223,

“The negative slope of the least squares regression fit from Equation 7 is used by AERONET to retrieve AE (Eck et al., 1999) with AOD **at all the wavelengths within the considered** spectral ranges (here we use **all three and four wavelengths within 340–440** and 440–870 wavelength pairs, **respectively** for AE estimations) as

$$\alpha_{\lambda_i-\lambda_j} = -\frac{N \sum \ln \tau_{aer,i} \cdot \ln \lambda_i - \sum \tau_{aer,i} \cdot \sum \lambda_i}{N \sum (\ln \lambda_i)^2 - (\sum \ln \lambda_i)^2}. \quad (8)''$$

(8) Section 3.2: “10% highest deviation cases” do you mean that you calculated the differences among NO<sub>2</sub> estimations, then you took the highest and then you increased (or decreased) this difference of 10% for obtaining an extreme scenarios of NO<sub>2</sub> differences ? Please the describe better the meaning of this sentence, also in the conclusion (line 420).

Response to Comment 8: We calculated the differences among NO<sub>2</sub> estimations using Eq. 1 and as presented in Section 3.1 and then looked for the 10% highest differences cases. We did not followed the methodology of taking the highest and then increasing (or decreasing) this difference of 10% for obtaining the extreme scenario. The extreme scenario presented in Section 3.2 is from the actual 10% of highest differences that we obtained from the comparisons.

We have corrected this sentence as below in Line 337-340

“In this section, we present (Table 2) the scenarios with extreme NO<sub>2</sub> situations i.e., 10% highest **difference** cases (**from all the differences as presented in Section 3.1**) taken into account as percentiles of NO<sub>2</sub> differences with 10% and 90% confidence levels for case 1 (NO<sub>2</sub> underestimation by OMIc) and case 2 (NO<sub>2</sub> overestimation by OMIc), respectively (here on referred to as “Extreme” case).”

We also made corrections in the Conclusion section as below in Line 488-491,

“Further assessment of AOD **differences** in extreme NO<sub>2</sub> loading scenarios (i.e., 10% highest **difference** instances taken into account as percentiles of NO<sub>2</sub> differences with 10% and 90% confidence levels for case 1 and case 2) revealed higher AOD **differences** in all cases with much more significant increase in the 10 stations mentioned above along with 3 more stations (ALD, SOL and MNH) as compared to their respective all datasets mean AOD **differences**.”

(9) Some typos errors:

The numbers of sub sessions at lines 118 and 128 are wrong ( 2.2.2=> 2.1.2 etc ).

Line 417: “Among these, 10 stations ..” => “Among these, 6 stations...”.

Response to Comment 9: We thank the reviewer for noticing this error. We have corrected it in the updated manuscript.

## References

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.

Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'Neill, N. T., Slutsker, I., and Kinne, S.: Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols, *J. Geophys. Res.*, 104, 31333–31349, <https://doi.org/10.1029/1999JD900923>, 1999.

Giles, D. M., Sinyuk, A., Sorokin, M. G., Schafer, J. S., Smirnov, A., Slutsker, I., Eck, T. F., Holben, B. N., Lewis, J. R., 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.

We are thankful to the reviewer for providing valuable comments and suggestions that the helped us improve the manuscript manifold.