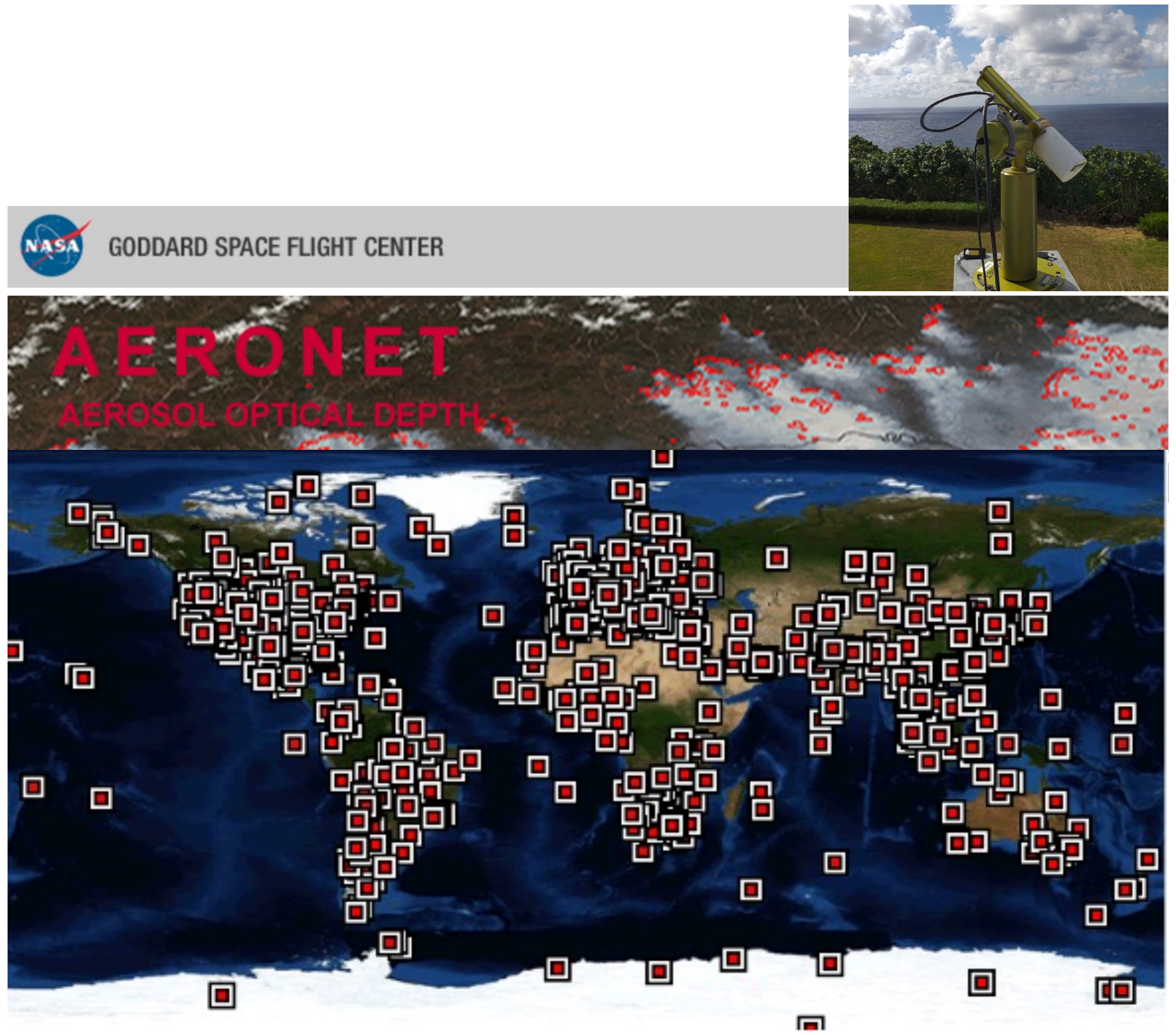
Synergy of ground-based remote sensing instrumentations to explore the impact of NO_2 absorption on aerosol optical depth measurements

Akriti Masoom[*1], Theano Drosoglou[2], Stelios Kazadzis[1], Ioannis-Panagiotis Raptis[3,4]

[1]Physikalisch-Meteorologisches Observatorium Davos, World Radiation Center (PMOD/WRC), Davos, 7260, Switzerland
[2]Institute Information Technologies Institute, Harilaou-Thermi, Thessaloniki, 57001, Greece
[3]Institute for Environmental Research and Sustainable Development, National Observatory of Athens (IERSD/NOA), Athens, 15236, Greece
[4]Laboratory of Climatology and Atmospheric Environment, Sector of Geography and Climatology, Department of Geology and Environment, National and Kapodistrian University of Athens, Athens, 15784, Greece

Introduction and data utilization: Sun photometer, spectroradiometer and global networks



Sun photometer: Aerosol optical depth (AOD) measurement

- Using Lambert–Beer law from direct sun measurements
- Considering contribution from Rayleigh scattering by atmospheric molecules and absorption by atmospheric constituents other than aerosols like ozone, nitrogen dioxide, water vapour, etc.

Spectroradiometers: trace gas measurements

- NO_2, O_3, HCHO total and tropospheric column, profile, surface concentration

Why we looked for NO_2 absorption impact on AOD measurements?

- Tropospheric NO_2 has high spatiotemporal variation and regional confinement near its source
- Likelihood for deviation from climatology in regions with high NO_2 emissions or trend reversal
- Also, there can be significant diurnal variation in NO_2 concentration

Objective

Assessment of the impact of NO_2 contribution on AOD measurements at several sites worldwide

Instrumentation

- AERONET (Aerosol Robotic Network) CIMEL sun photometers
- PGN (Pandonia Global Network) Pandora spectroradiometers

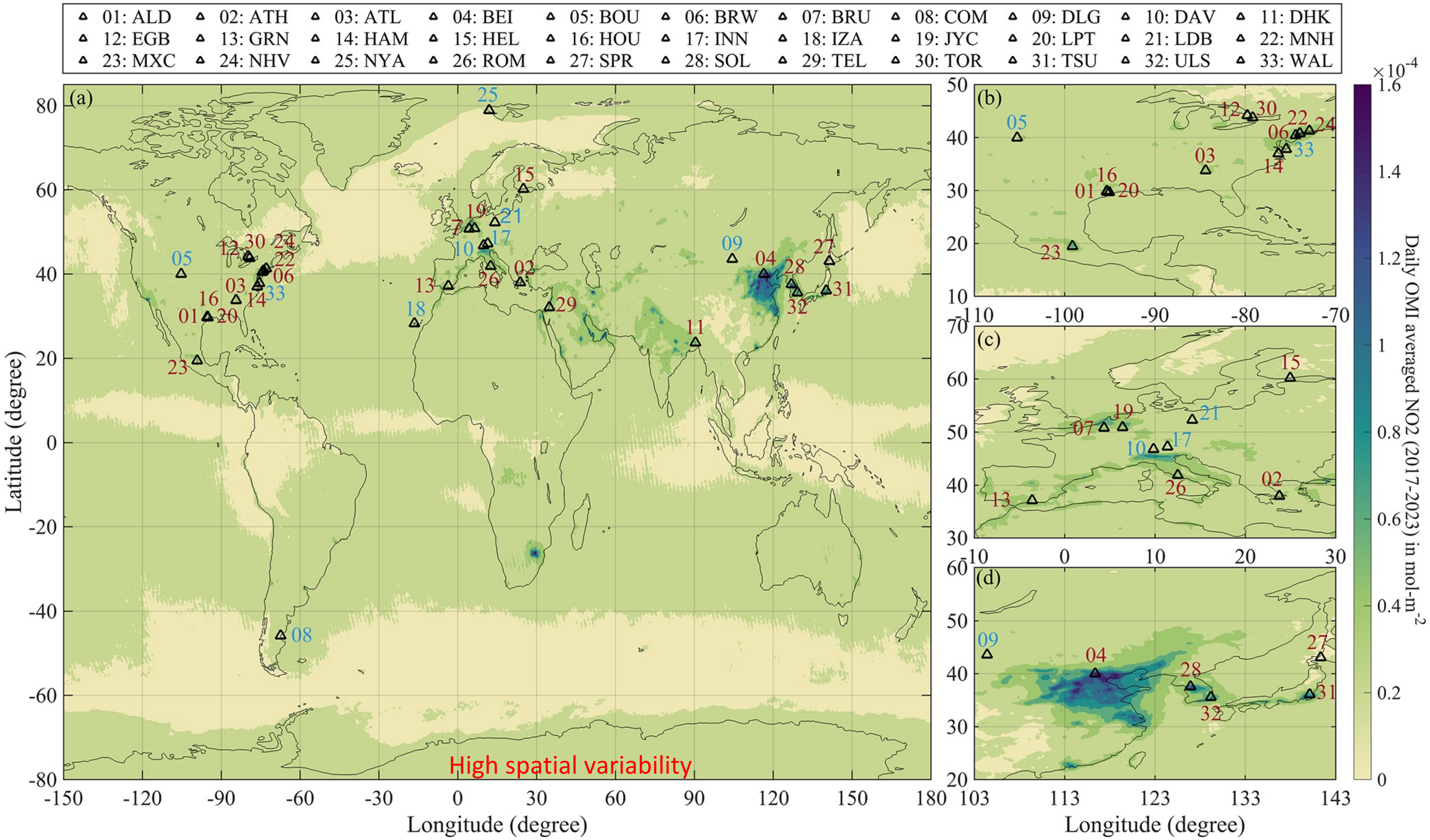
Two global networks → a number of co-locations useful for such analysis

Analysis

- PGN NO_2 vertical column density (VCD) → high (0, 10) and medium (1, 11) quality flags
- NO_2 correction on AERONET AOD at λ (nm): 340, 380, 440 and 500 and Ångström Exponent λ (nm): 440-870 and 340-440

* Pictures: https://aeronet.gsfc.nasa.gov, https://pandora.gsfc.nasa.gov, https://www.pandonia-global-network.org

Methodology: Co-located AERONET and PGN sites worldwide



Urban sites	
ALD	Aldine
ATH	Athens
ATL	Atlanta
BEI	Beijing
BRW	Brunswick
BRU	Brussels
DHK	Dhaka
EGB	Egbert
GRN	Granada
HAM	Hampton
HEL	Helsinki
HOU	Houston
JYC	Jülich/Joyce
LPT	La Porte
MNH	Manhattan
MXC	Mexico City
NHV	New Haven
ROM	Rome
SPR	Sapporo
SOL	Seoul
TEL	Tel Aviv
TOR	Toronto
TSU	Tsukuba
ULS	Ulsan

Rural Sites	
BOU	Boulder
COM	Comodoro
DLG	Dalanzadgad
DAV	Davos
INN	Innsbruck
IZA	Izaña
LDB	Lindenberg
NYA	Ny-Ålesund
WAL	Wallops

- PGN stations coordinates → AERONET latitude ± 0.09° and longitude ± 0.07° (in most of the cases with the exact same)
- Corresponding to daily AERONET time → selection of nearest matching PGN time → time interpolation of PGN data to AERONET time stamp
- Categorized all the stations as urban/rural site ('rural' as small cities that are in the countryside or adjacent to ocean and other sites as 'urban')

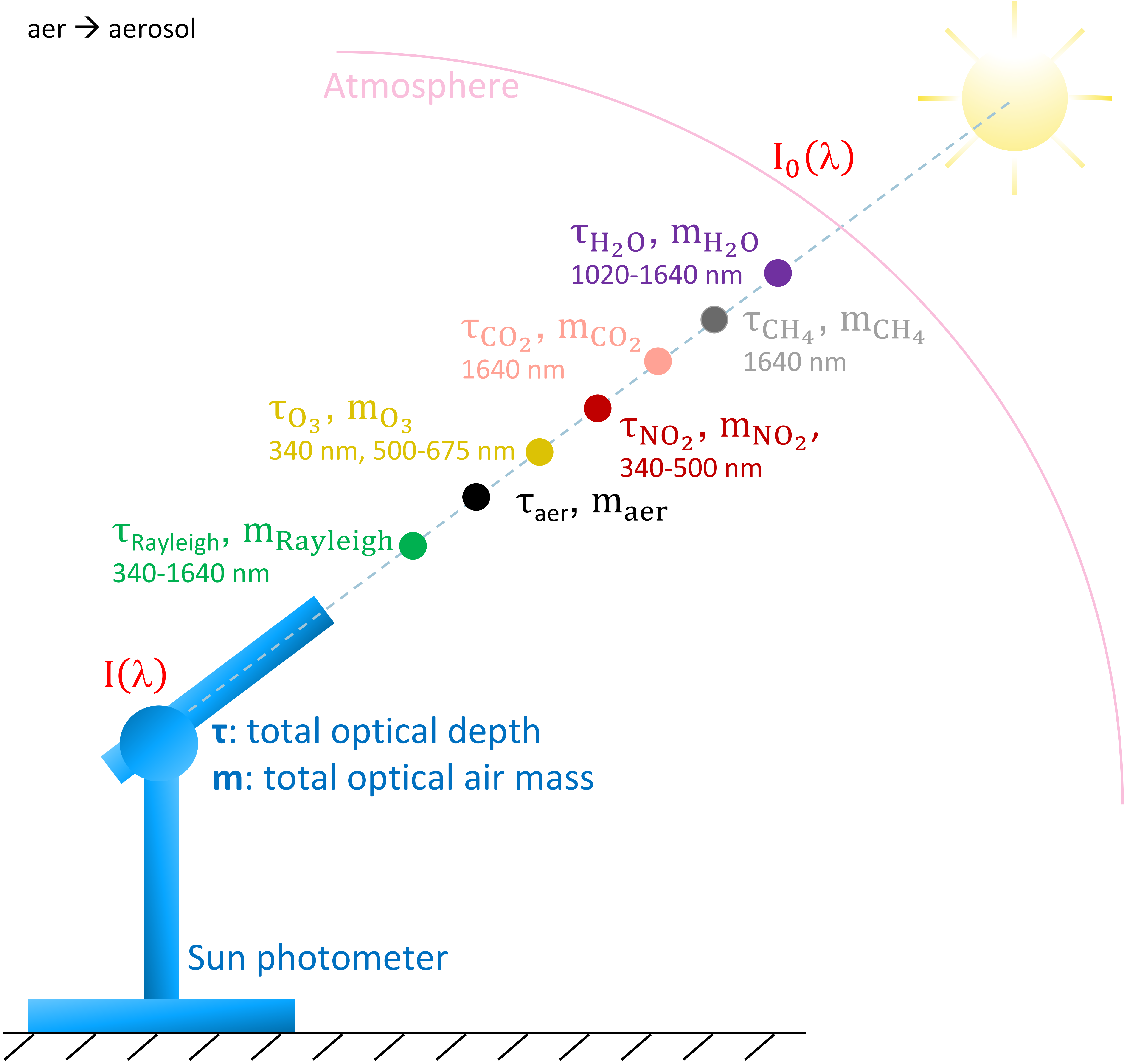
PGN: Pandonia Global Network; OMI: Ozone Monitoring Instrument

AOD calculation from direct sun measurements of sun photometers: NO_2 correction

$$I(\lambda) = I_0(\lambda) * e^{-m\tau} = I_0(\lambda) * e^{-(m_{Rayleigh}\tau_{Rayleigh} + m_{aer}\tau_{aer} + m_{O_3}\tau_{O_3} + m_{NO_2}\tau_{NO_2} + m_{CO_2}\tau_{CO_2} + m_{CH_4}\tau_{CH_4} + m_{H_2O}\tau_{H_2O})} \quad (1)$$

Lambert Beer law

aer → aerosol



$$\tau_{NO_2}(\lambda) = \frac{\sigma_{NO_2}(\lambda)}{1000} * \frac{m_{NO_2}}{m_a} * NO_2(DU) \quad (2)$$

Cuevas et al., 2019

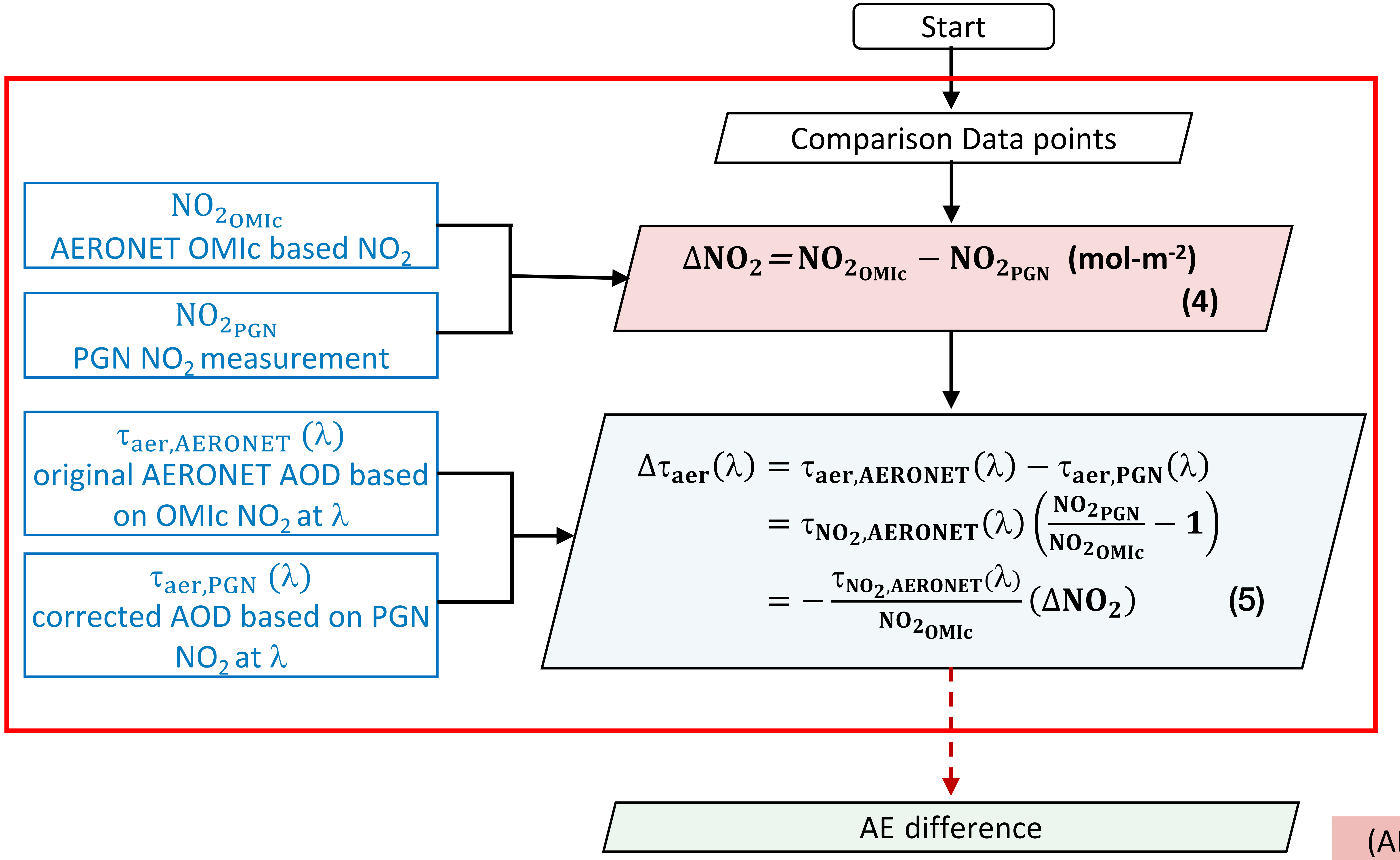
$$\tau_{aer,PGN}(\lambda) = \tau_{aer,AERONET}(\lambda) + \tau_{NO_2,AERONET}(\lambda) - \left(\tau_{NO_2,AERONET}(\lambda) * \frac{NO_{2_{PGN}}}{NO_{2_{OMIc}}}\right) = \tau_{aer,AERONET}(\lambda) - \tau_{NO_2,AERONET}(\lambda)\left(\frac{NO_{2_{PGN}}}{NO_{2_{OMIc}}} - 1\right) \quad (3)$$

- NO_2 optical depth is directly proportional to NO_2 vertical column density at a specific wavelength and sun elevation
- σ_{NO_2}, m_{NO_2}: NO_2 absorption coefficient at wavelength (λ) and NO_2 optical air mass obtained from (Gueymard, 1995)

* Cuevas, E., et al.: 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, 2019.
* Gueymard, C.: SMARTS2: a simple model of the atmospheric radiative transfer of sunshine: algorithms and performance assessment, Florida Solar Energy Center Cocoa, 1995.

DU: Dobson unit; PGN: Pandonia Global Network; OMIc: OMI Climatology

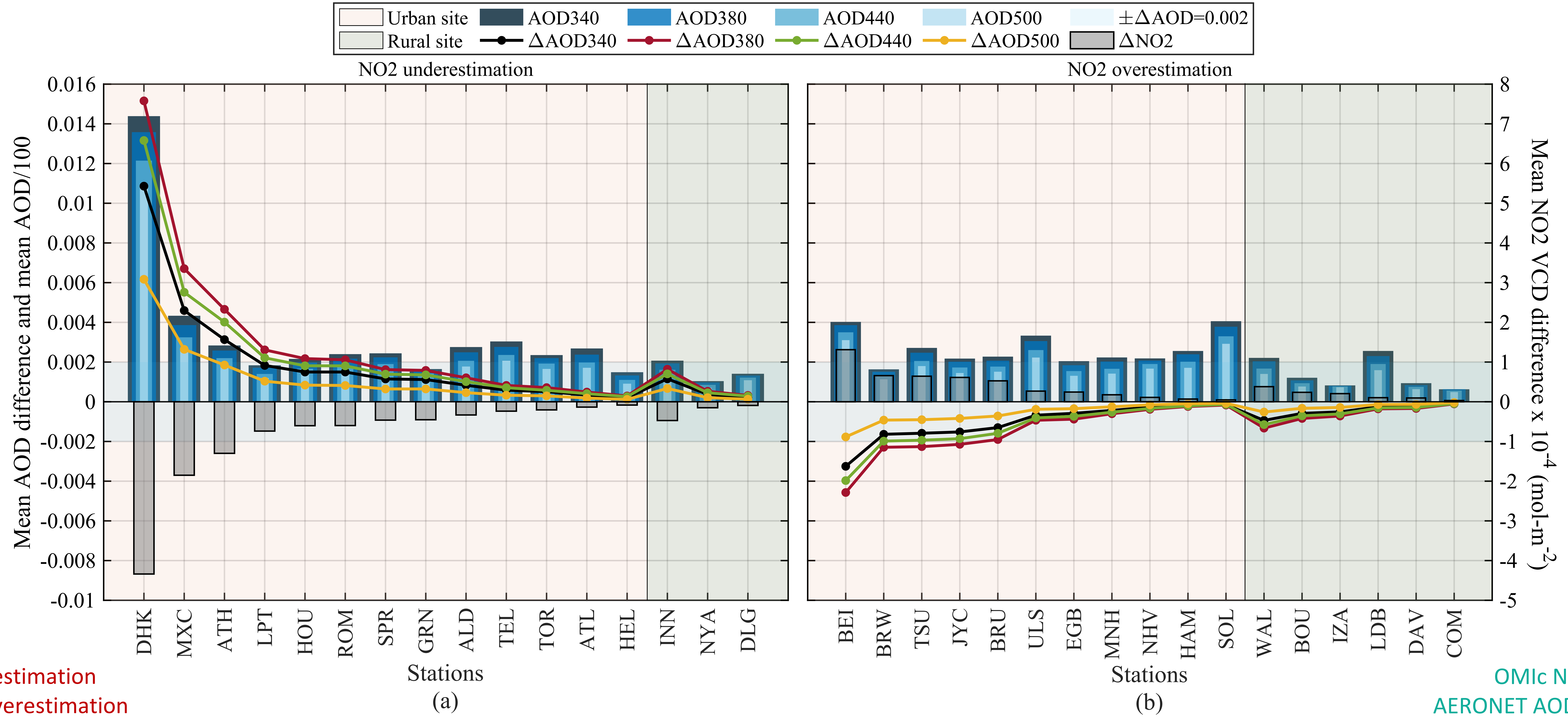
NO_2 absorption induced differences in AOD calculations



	Case 1	Case 2
(AERONET – PGN) NO_2 difference **ΔNO_2**	< 0 OMIc NO_2 underestimation	> 0 OMIc NO_2 overestimation
(AERONET – PGN) AOD difference **ΔAOD ($\Delta\tau_{aer}(\lambda)$)**	> 0 AERONET AOD overestimation	< 0 AERONET AOD underestimation

AOD: aerosol optical depth; PGN: Pandonia Global Network; AE: Ångström Exponent; OMIc: OMI Climatology

Differences between AERONET OMI NO_2 climatology and PGN NO_2 measurements: Impact on AOD measurements



Case 1
OMIc NO_2 underestimation
AERONET AOD overestimation

Case 2
OMIc NO_2 overestimation
AERONET AOD underestimation

- 16 stations in case 1 (13 are urban sites and 3 are rural sites)
- 6 urban stations (DHK, MXC, ATH, LPT, HOU and ROM) → mean NO_2 underestimation > 0.5 x 10^{-4} mol-m^{-2} and AOD overestimation ≥ 0.002
- AERONET OMIc NO_2 underestimation → higher pollution levels, which averaged OMIc climatological interpretation of NO_2 fails to depict

- 17 stations in case 2 (11 are urban sites and 6 are rural sites)
- 4 urban stations (BEI, TSU, BRW and JYC) → mean NO_2 overestimation > 0.5 x 10^{-4} mol-m^{-2} and AOD underestimation ≥ 0.002
- AERONET OMIc NO_2 overestimation → trend reversal of tropospheric NO_2 during the last decade due to reduction in pollution levels (OMIc is based on average values during 2004–2013)

AOD: aerosol optical depth; PGN: Pandonia Global Network; OMIc: OMI Climatology

Differences between AERONET OMI NO_2 climatology and PGN NO_2 measurements

Case 1: AERONET AOD overestimation

DHK: Mean PGN NO_2 is ~4 times higher than OMIc, OMIc NO_2 remains mostly constant and below 5 x 10^{-4} mol-m^{-2}

MXC: Mean PGN NO_2 is ~2 times above OMIc

- PGN NO_2 variation is mostly above OMIc values

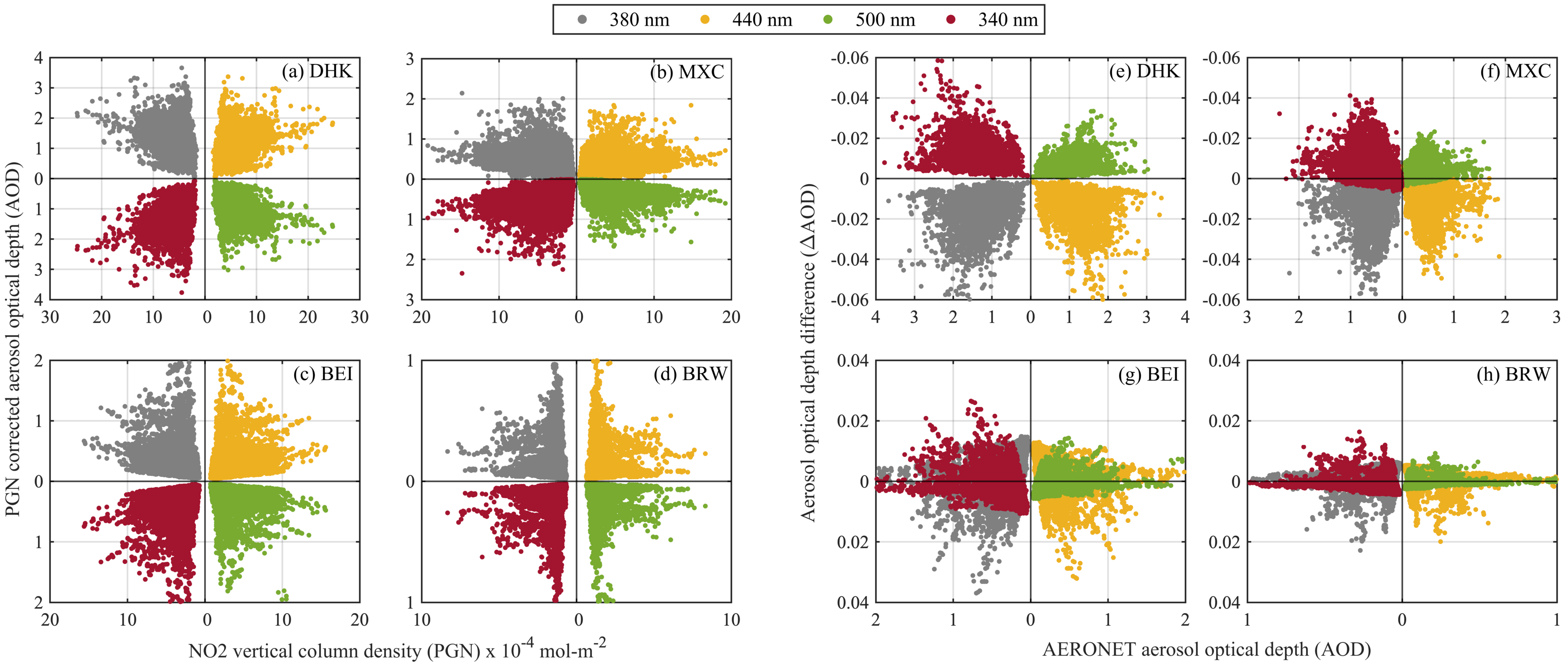
Case 2: AERONET AOD underestimation

BEI, BRW: Mean PGN NO_2 is ~1.5 times lower than OMIc, PGN NO_2 levels reaching ~20 x 10^{-4} mol-m^{-2} for BEI and to 10 x 10^{-4} mol-m^{-2} for BRW

- PGN NO_2 variation is on both side of the OMIc values

AOD: aerosol optical depth; PGN: Pandonia Global Network; OMIc: OMI Climatology

Are AOD, NO_2 values or AOD differences correlated?

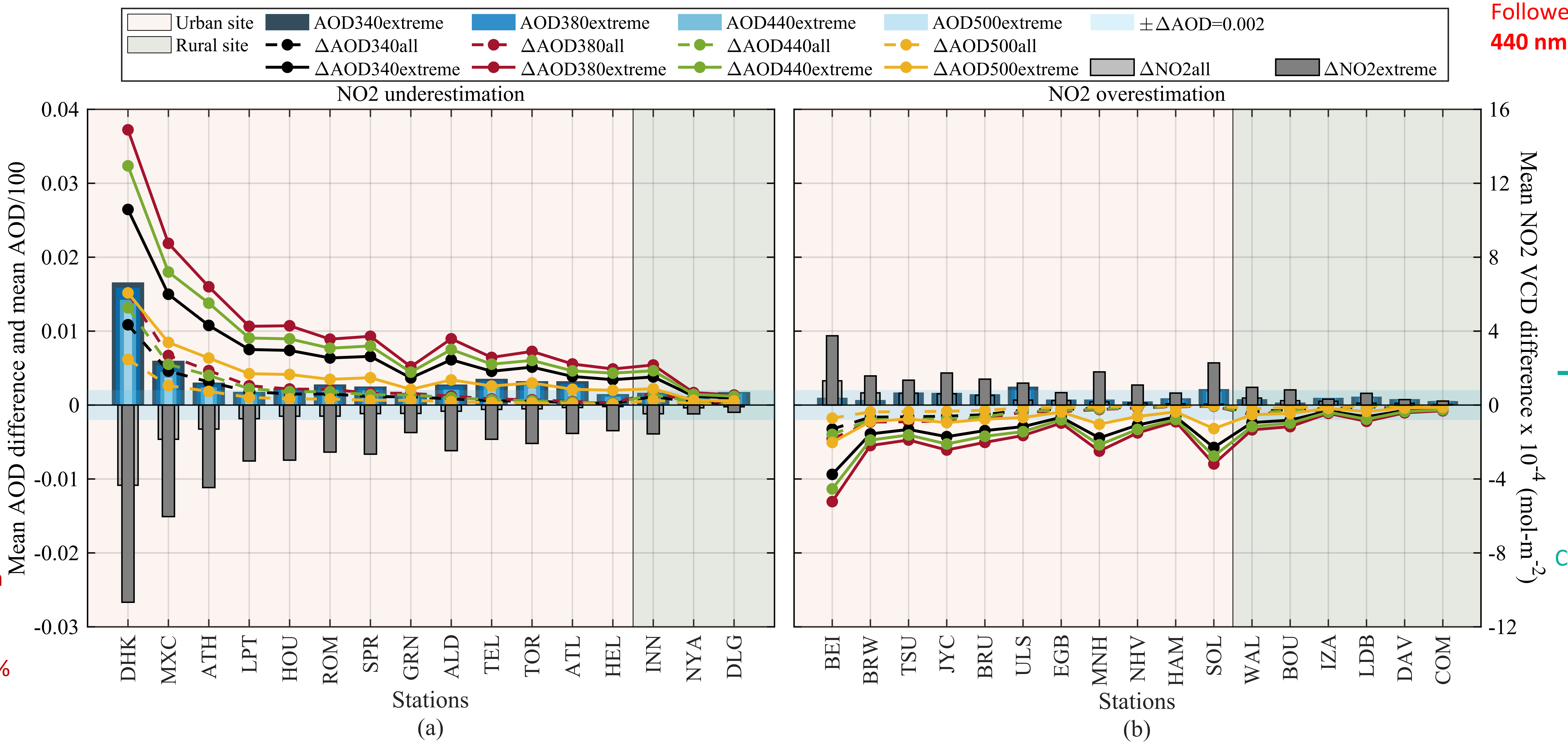


- AOD variation as a function of NO_2 VCD and AOD differences as a function of AOD values
- AOD is not correlated with NO_2 VCD values and AOD differences are also not correlated with AOD values
- NO_2 differences are related to AOD differences, and vice versa

AOD: aerosol optical depth; PGN: Pandonia Global Network; VCD: vertical column density

Extreme NO_2 scenario: 10% of highest differences

Highest affected wavelength
380 nm
Followed by
440 nm, 340 nm, 500 nm



Case 1: AERONET AOD overestimation

percentiles of NO_2 differences with 10% confidence levels

Case 2: AERONET AOD underestimation

percentiles of NO_2 differences with 90% confidence levels

- Increase in AOD differences for the 6 stations was found to be above 0.007 in "Extreme" case from the "All" dataset
- Even reaching up to 0.023 and 0.015 for DHK and MXC, respectively
- Similarly, ALD showed ~7 times and ~8 times increase in the differences in NO_2 and AOD, respectively in "Extreme" scenario as compared to "All" datasets

Stations BEI, JYC, MNH and SOL

- mean NO_2 difference is 1 x 10^{-4} mol-m^{-2} higher in "Extreme" than "All" dataset
- AOD differences increased by 0.004 (380 nm) and 0.003 (440 nm)
- For BEI, mean AOD underestimation reached to 0.013 and 0.011 at 380 nm and 440 nm, respectively

AOD: aerosol optical depth

Ångström Exponent calculation from spectral AOD measurement

$$\tau_{aer}(\lambda) = \beta \,.\, \lambda^{-\alpha} \quad (4)$$

Ångström power law

$$\ln\tau_{aer}(\lambda) = \ln\beta - \alpha \,.\, \ln\lambda \quad (5)$$

least squares regression fit

$$\alpha_{\lambda_i-\lambda_j} = -\frac{N\sum \ln\tau_{aer,i} \,.\, \ln\lambda_i - \sum \tau_{aer,i} \,.\sum \lambda_i}{N\sum(\ln\lambda_i)^2 - (\sum \ln\lambda_i)^2} \quad (6)$$

AE calculation

AE340-440 nm

$\tau_{aer,PGN}$

λ (nm): 340, 380 and 440

AE440-870 nm

$\tau_{aer,PGN}$

λ (nm): 440 and 500

$\tau_{aer,AERONET}$

λ (nm): 675 and 870

$\alpha_{\lambda_i-\lambda_j,AERONET}$

AERONET retrieved AE between λ_i and λ_j

$\alpha_{\lambda_i-\lambda_j,PGN}$

AE calculated from PGN corrected AOD between λ_i and λ_j

$$\Delta\alpha_{\lambda_i-\lambda_j} = \alpha_{\lambda_i-\lambda_j,AERONET} - \alpha_{\lambda_i-\lambda_j,PGN}$$

End

PGN: Pandonia Global Network; AE, **α**: Ångström Exponent; OMIc: OMI Climatology

Effect of climatological vs real NO_2 values on Ångström Exponent

- AE440–870 nm difference median was found to be −0.07 and −0.05 for BEI and BRW, respectively, and within ±0.03 for other stations
- AE340–440 nm difference median was 0.07 for BEI, 0.04 for BRW, and within ±0.03 for the remaining stations
- Narrower frequency distribution for stations like DHK can be attributed to broader AOD distribution (Wagner and Silva, 2008) and a broader AE distribution can be attributed to the narrower AOD distributions

Case 1: AERONET AOD overestimation

ΔAE440-870 Shift in peak of AE difference distribution towards a positive value → Higher relative positive error in AOD at shorter wavelength (440 and 500 nm)

ΔAE340-440 Shift in Peak of distribution is towards the other direction → Higher error at higher wavelength (440 nm) than at lower wavelength (340 nm)

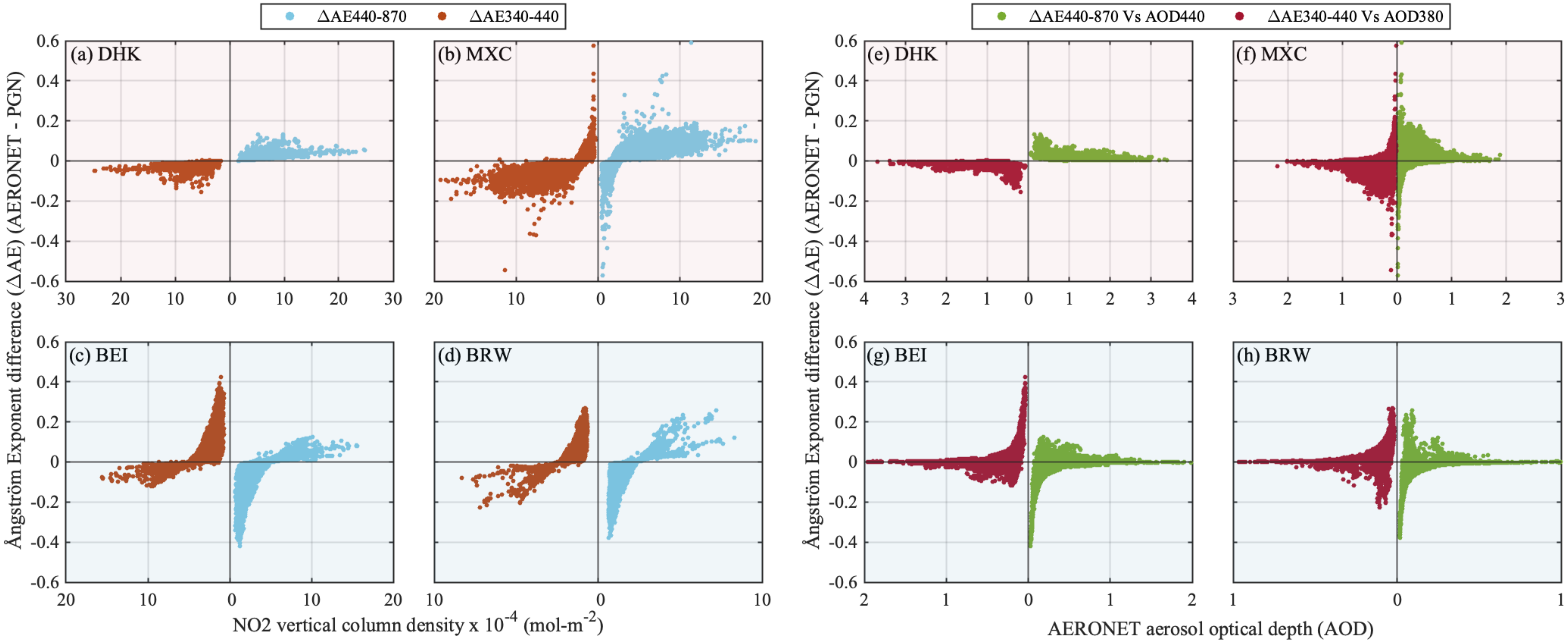
Case 2: AERONET AOD underestimation

ΔAE440-870 and ΔAE340-440 Similar but opposite (in sign) to case 1

* Wagner, et al.: Some considerations about Ångström exponent distributions, Atmos. Chem. Phys., 8, 481–489, https://doi.org/10.5194/acp-8-481-2008, 2008

Ångström Exponent differences variation with NO_2 and AOD magnitude

Case 1:
AERONET AOD
overestimation



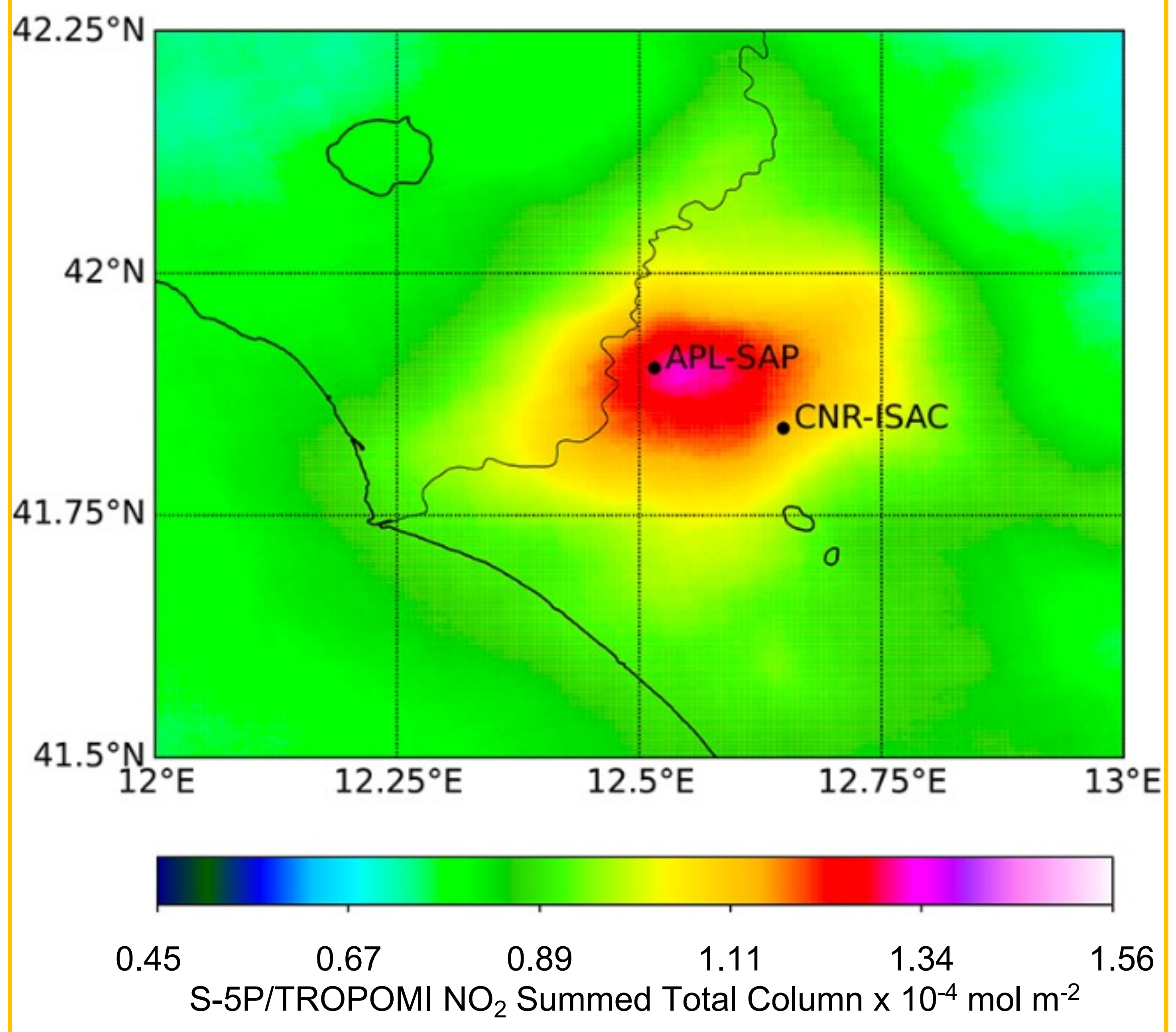
Case 2:
AERONET AOD
underestimation

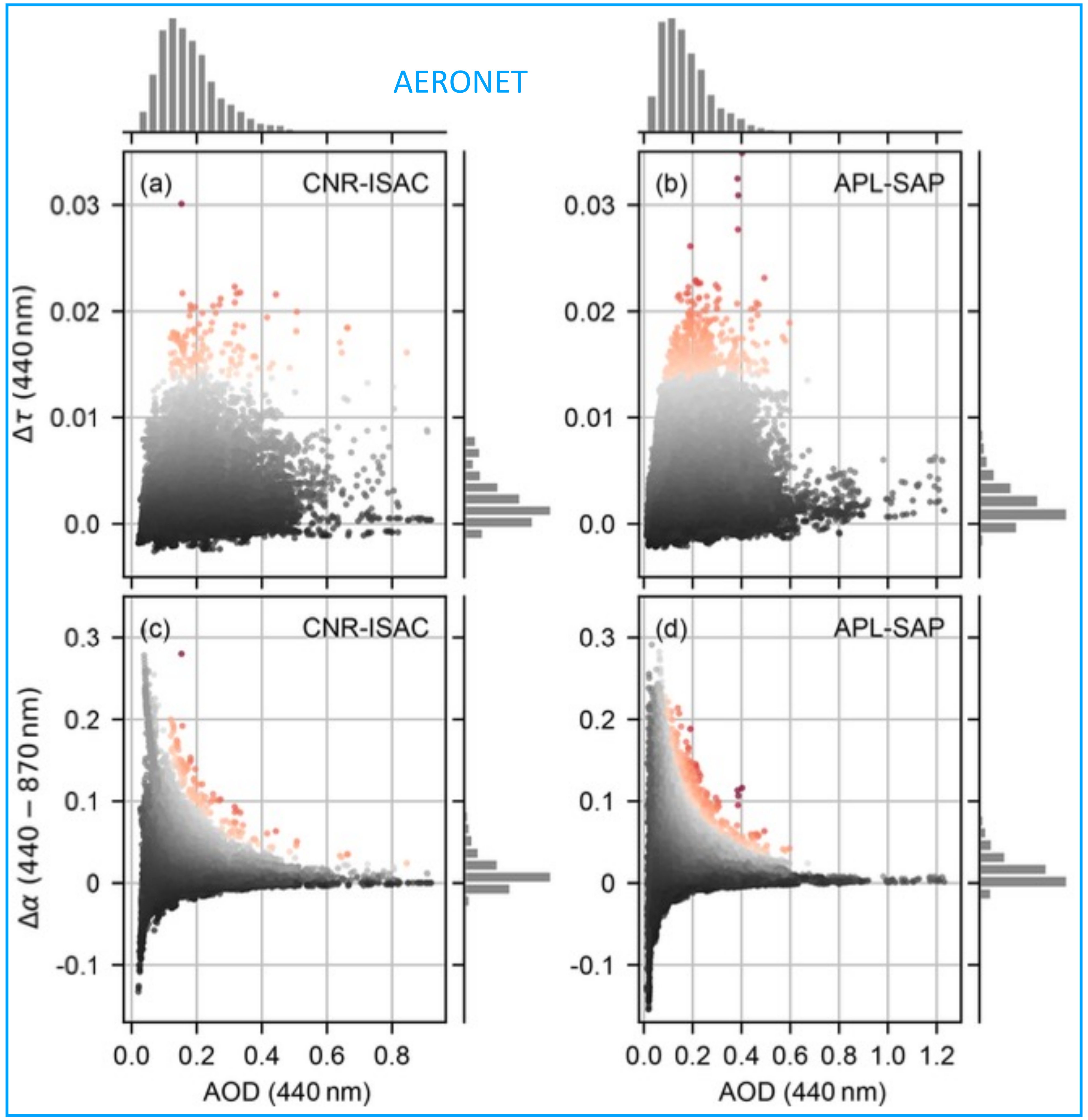
- AE uncertainty is not very simple to interpret as
  - it is a derivative quantity, sensitivity depends on AOD and depends on any spectral correlations in AOD uncertainty
- ΔAE variation with NO_2
  - Case 1: there is a strong positive and negative bias in AE440–870 and AE340–440, respectively
  - Case 2: the positive and negative biases are not that strongly present
- ΔAE variations with AOD showed high AE differences associated with low AOD instances

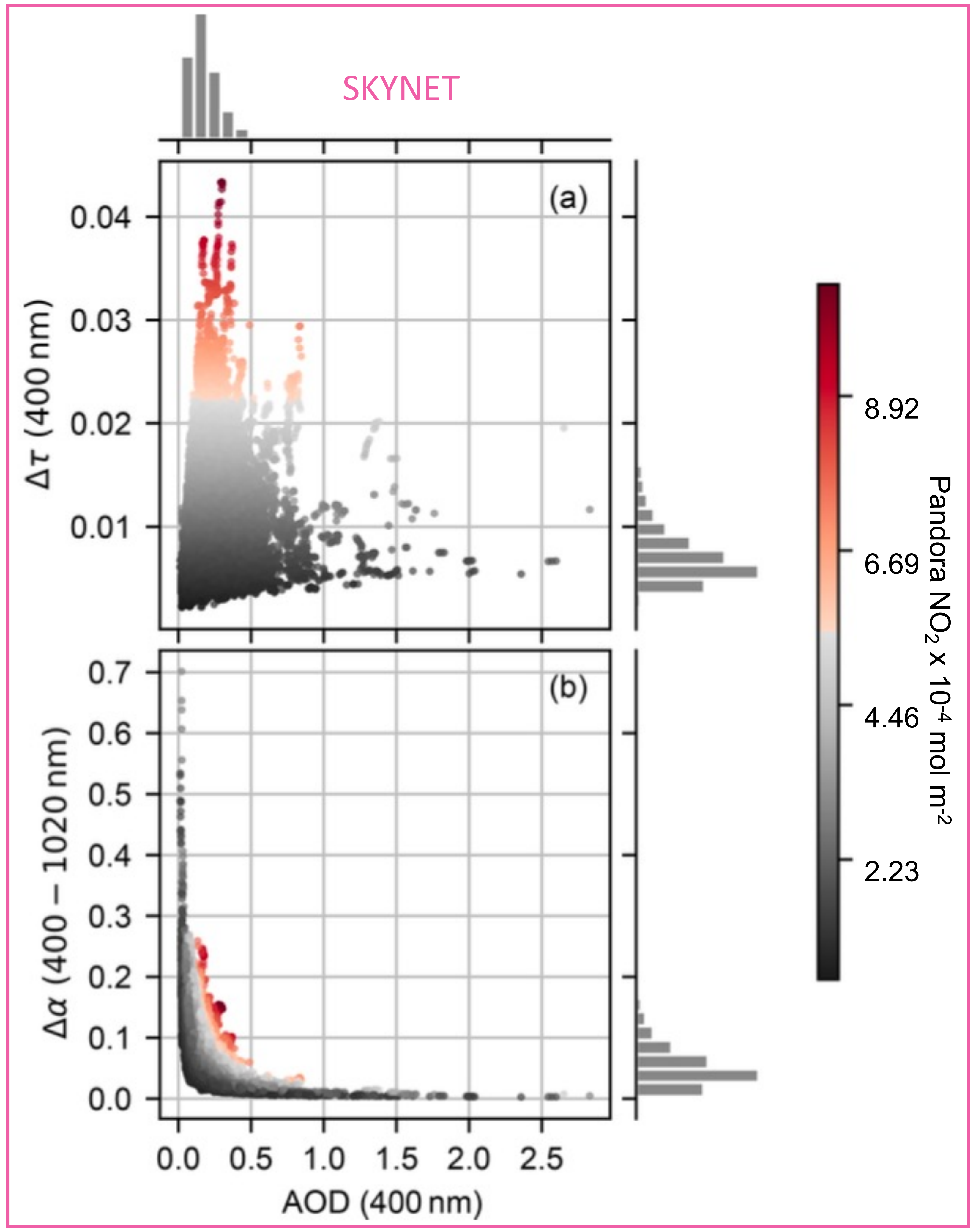
AOD: aerosol optical depth; PGN: Pandonia Global Network

Site specific analysis for Rome - AERONET and SKYNET

APL-SAP: Sapienza University, Rome
CNR-ISAC: Tor Vergata, Rome





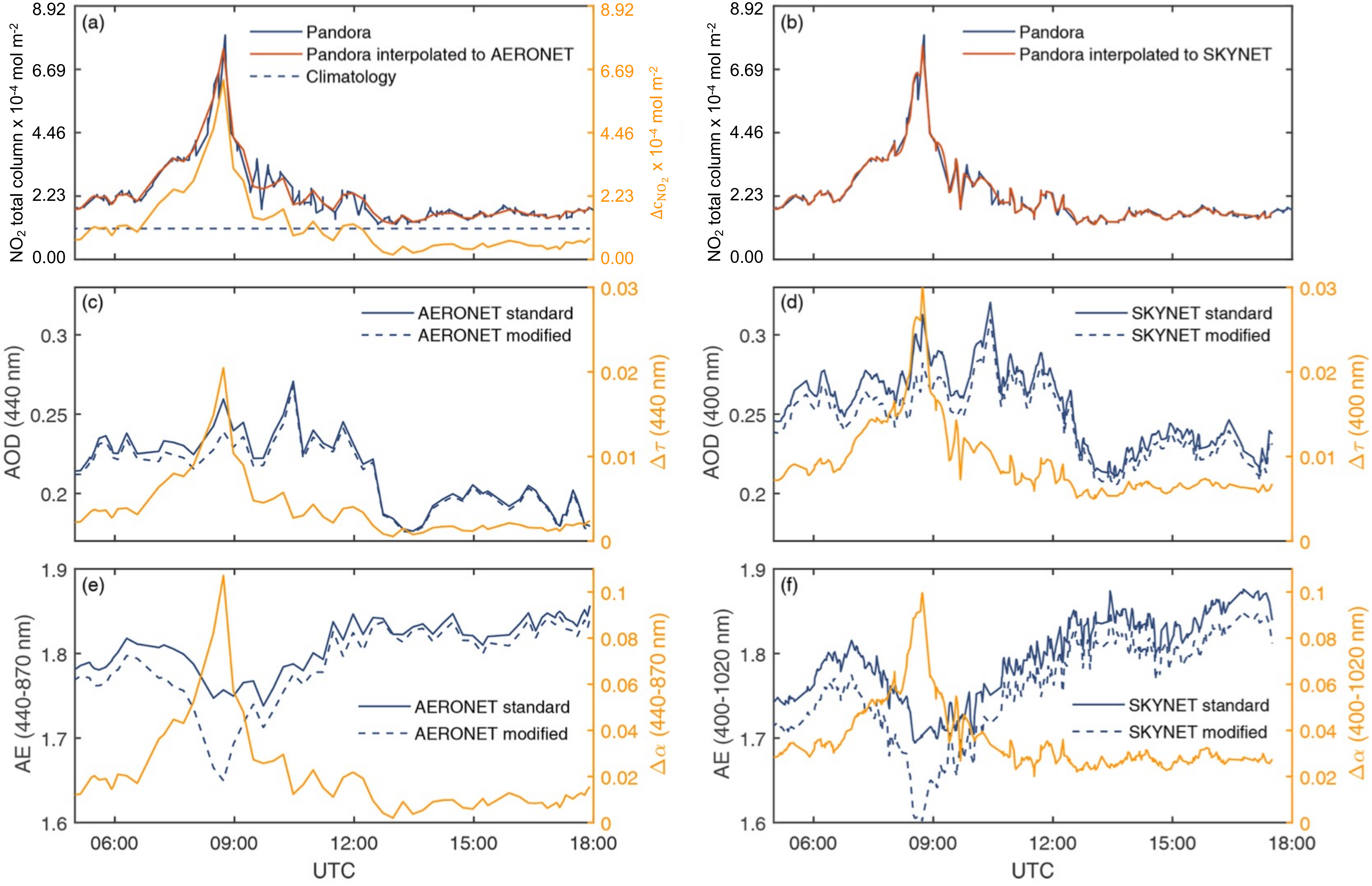


- Average AOD bias for Rome was ~0.002 and ~0.003 at 440 and 380 nm, respectively for AERONET but was a bit higher for SKYNET (~0.007)
- AE bias was ~ 0.02 and ~ 0.05 for AERONET and SKYNET, respectively
- Higher ΔAOD are obtained for higher NO_2 concentrations, regardless of the initial measured AOD (i.e., higher ΔAOD are also observed for lower AOD values)

AOD: aerosol optical depth; PGN: Pandonia Global Network; AE, **α**: Ångström Exponent; OMIc: OMI Climatology

*Drosoglou, T., et al.: Evaluating the effects of columnar NO_2 on the accuracy of aerosol optical properties retrievals, Atmos. Meas. Tech., 16, 2989–3014, 2023

Site specific analysis for Rome: High Pandora NO_2 and low AOD - AERONET and SKYNET

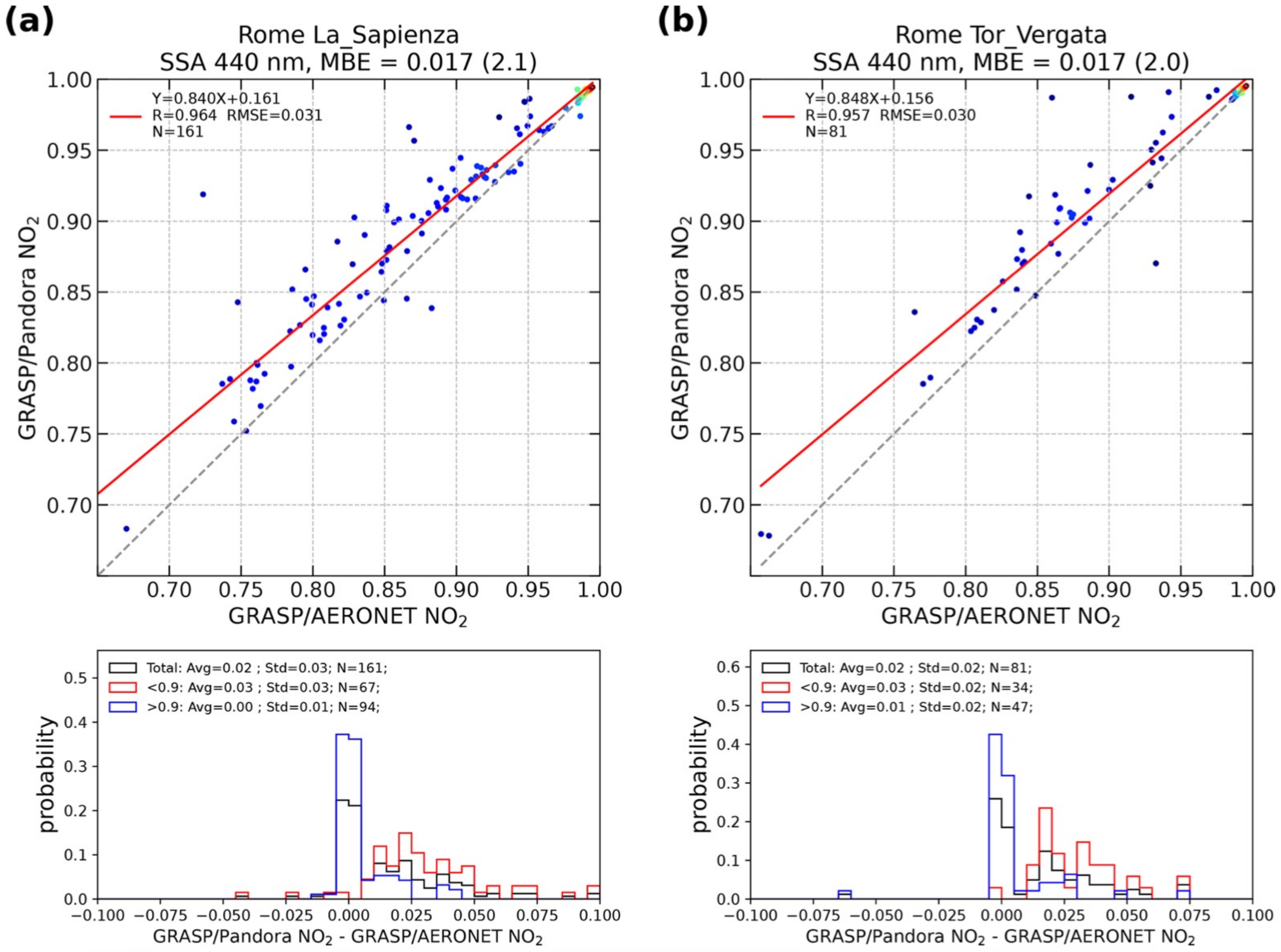


- A high NO_2 event in Rome on 25 June 2020
- For AERONET,
  - median and maximum AOD bias was ~0.003 and ~0.02, respectively
  - median and maximum AE biases are 0.014 and 0.11, respectively
- For SKYNET,
  - AOD bias median and maximum was ~0.008 and ~0.03, respectively
  - AE bias median and maximum was ~0.03 and ~0.10, respectively
  - both AOD and AE deviations are higher compared to AERONET
  - can be due to the fact that SKYNET AOD calculations do not account for NO_2 absorption

AOD: aerosol optical depth; AE: Ångström Exponent

*Drosoglou, T., et al.: Evaluating the effects of columnar NO_2 on the accuracy of aerosol optical properties retrievals, Atmos. Meas. Tech., 16, 2989–3014, 2023

Site specific analysis for Rome: Impact on single scattering albedo (SSA)



- Comparisons of SSA at 440 nm obtained from GRASP with AERONET and Pandora NO_2 for NO_2 values above 3.12 x 10^{-4} mol m^{-2} ($\mu + 2\sigma$)
- Consistent positive bias of ~ 0.02 (~ 2 %) in high NO_2 conditions
- Previous studies found SSA retrieval uncertainties in the range of 0.02–0.03 (Eck et al., 2003), whereas the correction, when high NO_2 is recorded, is usually higher

*Drosoglou, T., et al.: Evaluating the effects of columnar NO_2 on the accuracy of aerosol optical properties retrievals, Atmos. Meas. Tech., 16, 2989–3014, 2023
* Eck, T. F., et al.: Variability of biomass burning aerosol optical characteristics in southern Africa during the SAFARI 2000 dry season campaign and a comparison of single scattering albedo estimates from radiometric measurements, J. Geophys. Res.-Atmos., 108, 2156–2202, https://doi.org/10.1029/2002JD002321, 2003

Key findings and way ahead

- NO_2 climatological input was found to underestimate NO_2 values in a number of cases as compared to PGN (ground-based) measurements which led to an overestimation of AOD calculations in spectral range with prominent NO_2 absorption and vice versa for other cases

- Satellite based NO_2 climatology used for AOD calculations can be updated with focus on urban areas that can also have high diurnal variability in NO_2 concentrations

- Instrument co-location of different instrument/networks can potentially be used to improve stand-alone algorithms and retrievals using different outputs from co-located instrumentations (e.g., in this analysis PGN NO_2 output was used as an input in AERONET AOD calculations)

- This analysis highlights the importance of accurate NO_2 representation with the best possible scenario, however, concerning implementation into global AOD networks (such as AERONET, GAW-PFR or SKYNET), synergistic use of satellite data is required to account for all stations and also concerning the times series of data availability from Pandora instruments that start from 2016

Questions !!!!

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Preprint. Discussion started: 19 March 2024

EGUsphere
Preprint repository

Assessment of NO_2 uncertainty impact on aerosol optical depth retrievals at a global scale

Akriti Masoom[1], Stelios Kazadzis[1], Masimo Valeri[2], Ioannis-Panagiotis Raptis[3,4], Gabrielle Brizzi[2], Kyriakoula Papachristopoulou[5], Francesca Barnaba[6], Stefano Casadio[2], Axel Kreuter[7,8], Fabrizio Niro[9]

[1]Physical-Meteorological Observatory in Davos, World Radiation Center (PMOD/WRC), Davos, 7260, Switzerland
[2]Serco Italia S.p.A., Frascati, Rome, 00044, Italy
[3]Institute for Environmental Research and Sustainable Development, National Observatory of Athens (IERSD/NOA), Athens, 15236, Greece
[4]Laboratory of Climatology and Atmospheric Environment, Sector of Geography and Climatology, Department of Geology and Environment, National and Kapodistrian University of Athens, Athens, 15784, Greece
[5]Institute for Astronomy, Astrophysics, Space Applications and Remote Sensing, National Observatory of Athens (IAASARS/NOA), Athens, 15236, Greece
[6]National Research Council, Institute of Atmospheric Sciences and Climate, CNR-ISAC, Rome, 00133, Italy
[7]Institute for Biomedical Physics, Medical University Innsbruck, Innsbruck, 6020, Austria
[8]LuftBlick OG, Innsbruck, 6020, Austria
[9]ESA-ESRIN, Frascati, Rome, 00044, Italy

Correspondence to: Akriti Masoom (akriti.masoom@pmodwrc.ch)

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Atmospheric Measurement Techniques

Evaluating the effects of columnar NO_2 on the accuracy of aerosol optical properties retrievals

Theano Drosoglou[1], Ioannis-Panagiotis Raptis[1,2], Massimo Valeri[3], Stefano Casadio[3], Francesca Barnaba[4], Marcos Herreras-Giralda[5], Anton Lopatin[5], Oleg Dubovik[6], Gabriele Brizzi[3], Fabrizio Niro[7], Monica Campanelli[4], and Stelios Kazadzis[8]

[1]Institute for Environmental Research and Sustainable Development, National Observatory of Athens (IERSD/NOA), 15236 Athens, Greece
[2]Laboratory of Climatology and Atmospheric Environment, Sector of Geography and Climatology, Department of Geology and Environment, National and Kapodistrian University of Athens, 15784 Athens, Greece
[3]Serco Italia S.p.A., 00044 Frascati, Rome, Italy
[4]National Research Council, Institute of Atmospheric Sciences and Climate, CNR-ISAC, 00133 Rome, Italy
[5]GRASP SAS, Remote Sensing Developments, 59260 Lezennes, France
[6]Laboratoire d'Optique Atmosphérique (LOA) – UMR 8518, CNRS/Université de Lille, Villeneuve-d'Ascq, 59650, France
[7]ESA-ESRIN, 00044 Frascati, Rome, Italy
[8]Physicalisch-Meteorologisches Observatorium Davos, World Radiation Center, 7260 Davos, Switzerland

Correspondence: Theano Drosoglou (tdroso@noa.gr)

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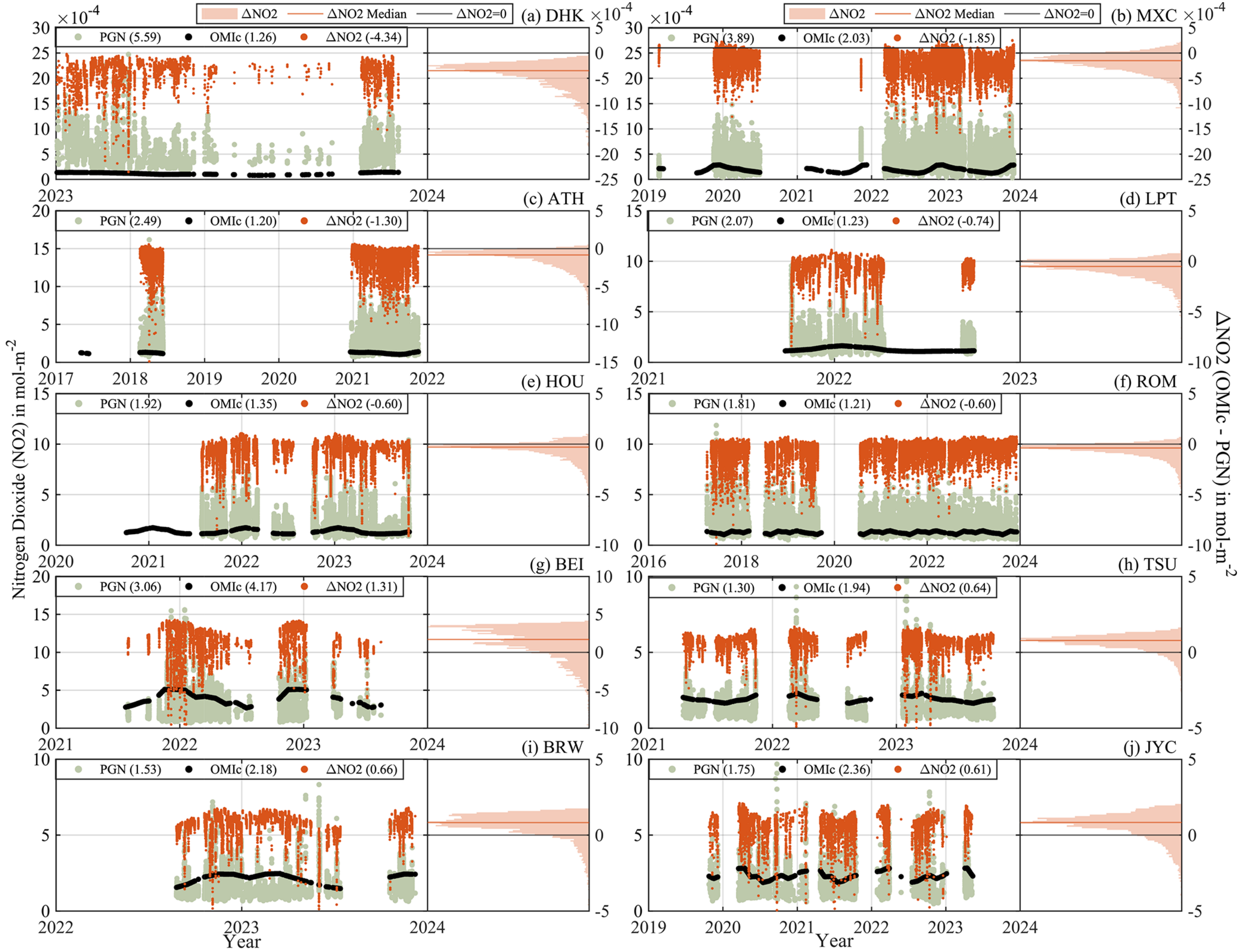
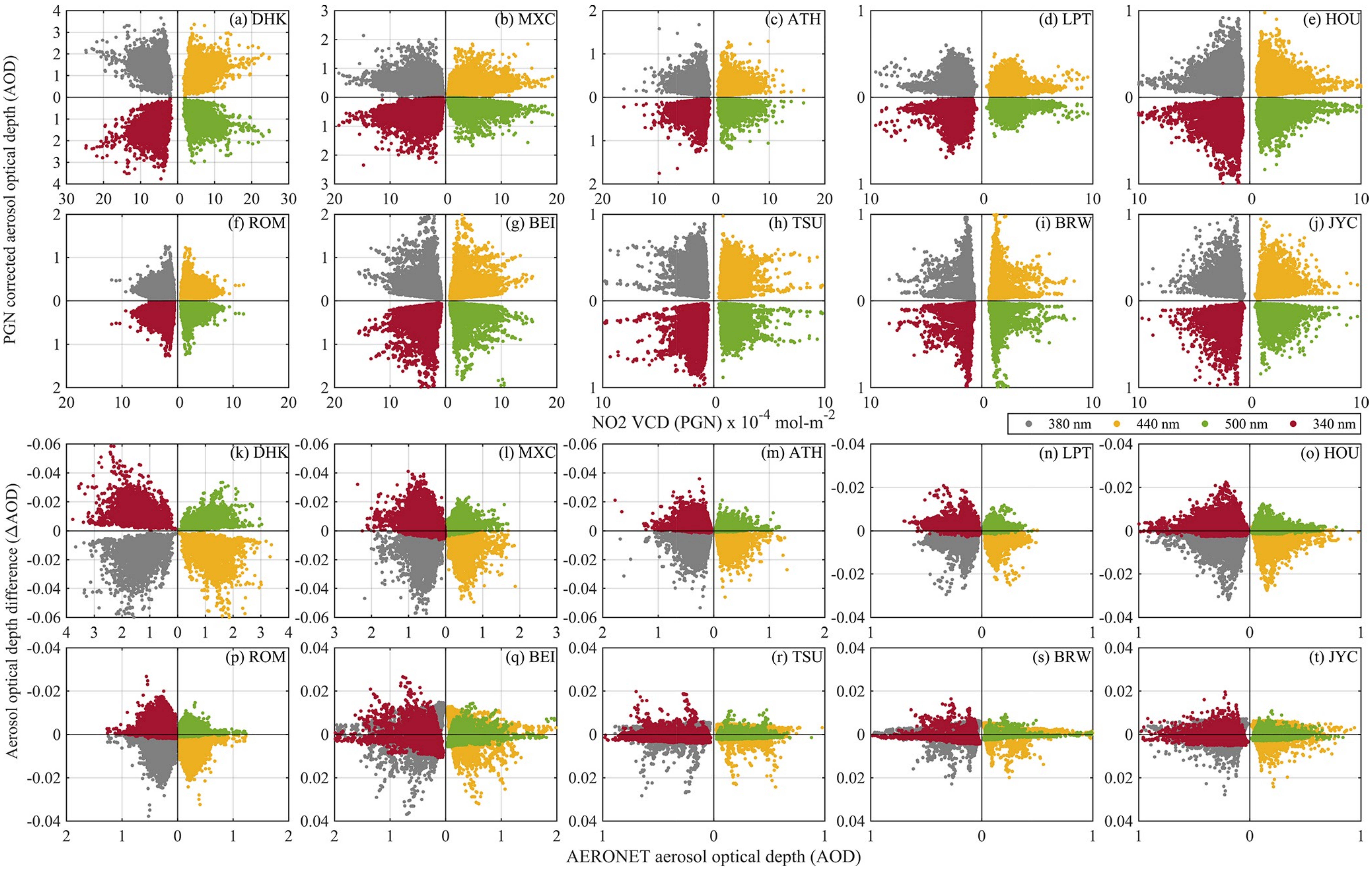
Extra Slides

Differences between AERONET OMI NO_2 climatology and PGN NO_2 measurements: 10 stations



Are AOD, NO_2 values or AOD differences correlated?: 10 stations



Effect of climatological vs real NO_2 values on Ångström Exponent: 10 stations

Ångström Exponent differences variation with NO_2 and AOD magnitude: 10 stations