Characterisation of trace gases horizontal inhomogeneity in Urban atmosphere: a case study using Pandora sky measurements in the context of the Boundary-layer Air Quality Using Network of INstruments (BAQUNIN) Project

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Method

Use the Sky measurements (Max-DOAS) taken at 6 equispaced azimuth angles to evaluate horizontal inhomogeneity.

Measurement Definitions
- SCAN = set of subsequent pointing zenith angles measurements for a given pointing azimuth angle
- SEQUENCE = set of SCAN measurement for 6 subsequent azimuth angles
- Surface Concentration (SC)= NO2 or H2O concentration estimated from a single SCAN, supposed to be representative for a horizontal conic volume of about 5 km, in $\mu g/m3$ units. The Pandora SC are given in ppb units, conversion to $\mu g/m3$ is based on the species molecular weight, and on 298.15 K and 1 atmosphere conditions
- Tropospheric Concentration = NO2 or H2O concentration estimated from a single SCAN using Multi-Axes DOAS technique (Max-DOAS), relative to a volume approximately covering the troposphere (0-10 km height), in Dobson Units (NO2) and cm (H2O) units
- Vertical Column Density = NO2 or H2O vertical column concentration estimated from Direct Sun measurements using DOAS technique, in Dobson Units (NO2) and cm (H2O) units

Ellipse Definitions
- Eccentricity = how much a conic section (circle, ellipse, parabola or hyperbola) departs from being circular
- Rotation Angle = the angle between the major axis and 0 degrees relative to North clockwise
- Area = square root of ellipse area divided by π, i.e. sqrt(area/π), estimate of the average Surface or Troposphere concentration for a given sequence (the π value comes from the 2D integration of the ellipse and must be removed)

SEQUENCE detection method

Since 1st of November 2018, PAN#117 operation schedule has been modified in order to perform sequences of Max-DOAS (Sky) measurement scans at 6 fixed pointing azimuth angles, i.e. 0, 60, 120, 180, 240 and 300 degrees North.
For each scan, Surface and Troposphere NO2 and H2O concentrations are estimated along with related uncertainties. The time required to complete the six sky measurements and two direct sun measurements is about 30 minutes.
However, being a SEQUENCE the set of 6 subsequent scans, we can impose the first value of the pointing azimuth to be any of the possible values and searching for the subsequent 5. This implies that we can increase the temporal sampling of this type of analysis. To illustrate this, let's indicate the pointing azimuth angles with 0, 1, 2, 3, 4, 5 and the direct sun measurements by D.
In practice, the operation schedule should look like this one:
D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …
If we impose the first azimuth to be 0 (i.e. 0 North), then the SEQUENCE pattern will be 012345; but if we impose the first azimuth to be 1 (i.e. 60 North), then the pattern will be 123450. Similarly, for first azimuth to be 2, the pattern will be 234501, and so on as shown below (first sequence in red, second in dark red).

Sequence type 1: D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …
Sequence type 2: D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …
Sequence type 3: D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …
Sequence type 4: D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …
Sequence type 5: D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …
Sequence type 6: D 0 1 2 3 4 5 D 0 1 2 3 4 5 D 0 1 2 3 4 5 …

For each sequence type, the "sampling time" is the average time of the selected data in a single sequence.
By adopting this strategy and merging the results from all sequence types, we can increase the temporal sampling of the sequences to about 5 minutes.

Ellipse fitting method part 1

For each detected SEQUENCE, the following fitting strategy is adopted:

1) Conversion from troposphere/surface data to pseudo coordinates
Being ρ the density and ϕ the azimuth angle for ith scan we can define the "pseudo coordinates":
$x_i = \rho_i \sin(\phi_i)$
$y_i = \rho_i \cos(\phi_i)$
i = 0,...,5 (in the order of the selected sequence)

2) Exclude in sequence one ϕ value and compute conic curve parameters using the remaining 5 points from the following formula:
$$x^2 + by^2 + cxy + dx + ey + f = 0$$
The fit converges if the pseudo coordinates can be represented by an ellipse, i.e. if the resulting eccentricity "e" value is 0≤e<1 (0=circle, 1=parabola, >1=hyperbola).
The "single ellipse" RMS is estimated from measured-retrieved point distances + original concentration uncertainties (quadratic sum of residuals).

3) Consider the N (1≤N ≤ 6) converged fit results to create the SEQUENCE ellipse. The ellipse parameters are estimated as the weighted average of the "single ellipse" values, where the weights are the inverse of the related RMS (i.e. larger errors = smaller weight)

4) Compute the eccentricity and the rotation angle for the resulting average SEQUENCE ellipse

Ellipse fitting method part 2

The general of a conic section equation is
$$a'x^2 + b'y^2 + c'xy + d'x + e'y + f' = 0$$
If we assume that a'≠0, dividing by a' the equation can be transformed into:
$$x^2 + by^2 + cxy + dx + ey + f = 0$$
The eccentricity of an ellipse can be calculated from the curve parameters as:
$$e = \sqrt{\frac{2(1-b^2)+c^2}{\eta(1+b)+\sqrt{(1-b^2)+c^2}}}$$
Where η = 1 or -1 if the determinant of the following matrix is negative or positive.

$$\begin{vmatrix} 1 & c/2 & d/2 \\ c/2 & b & e/2 \\ d/2 & e/2 & f \end{vmatrix}$$

See https://en.wikipedia.org/wiki/Eccentricity_(mathematics) for details.

The eccentricity of an ellipse is not dependant on the area or the rotation angle of the curve, it only describes the divergence of the curve from a perfect circle. This means that eccentricity values associated to different species and/or units can be directly compared.
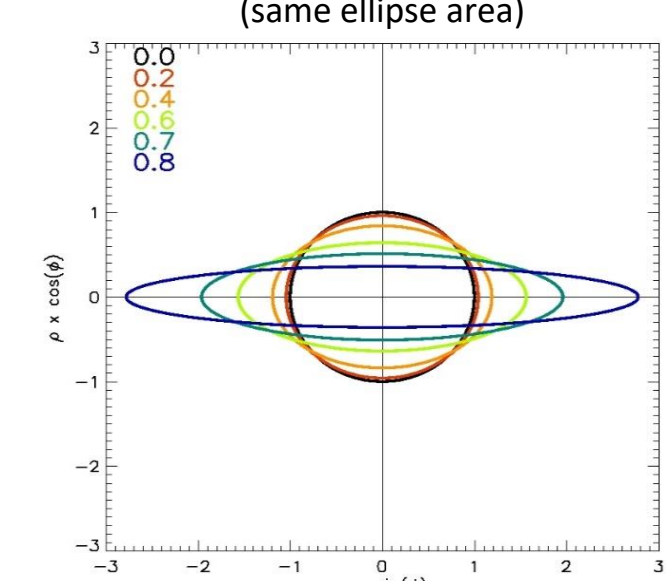
Ellipse fitting method part 3

The counter clockwise rotation angle α of our generic ellipse is defined as follows:
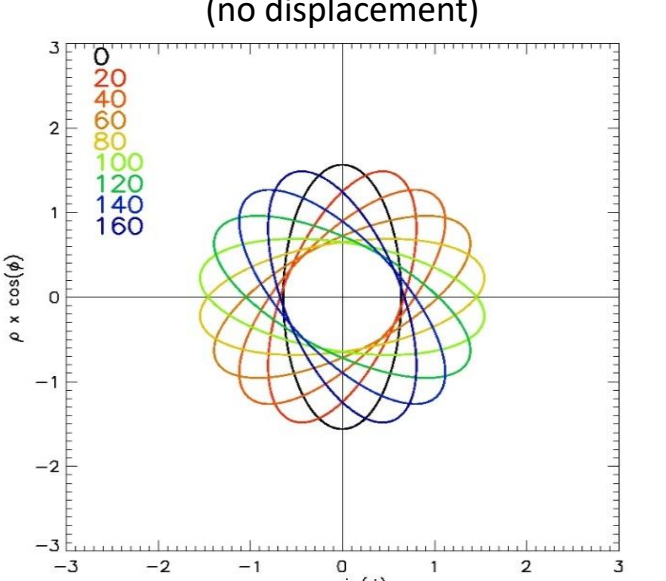$$\cot 2\alpha = \frac{1-b}{c}$$
See: https://math.stackexchange.com/questions/426150/what-is-the-general-equation-of-the-ellipse-that-is-not-in-the-origin-and-rotate
The α values range from 0 to 180 degrees, as shown in the centre panel of Figure 1.
In case the ellipse is not centred in the origin of axes, we can use the coordinates of the barycentre of the ellipse as additional information in order to extend the rotation angle range to 0-360 degrees, as shown in the right panel of Figure 1. In this way we can solve the 180 degrees ambiguity intrinsic in the definition of the cot function.
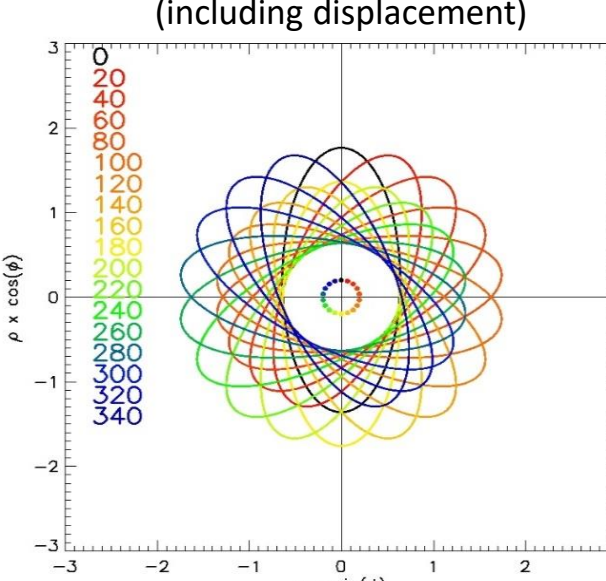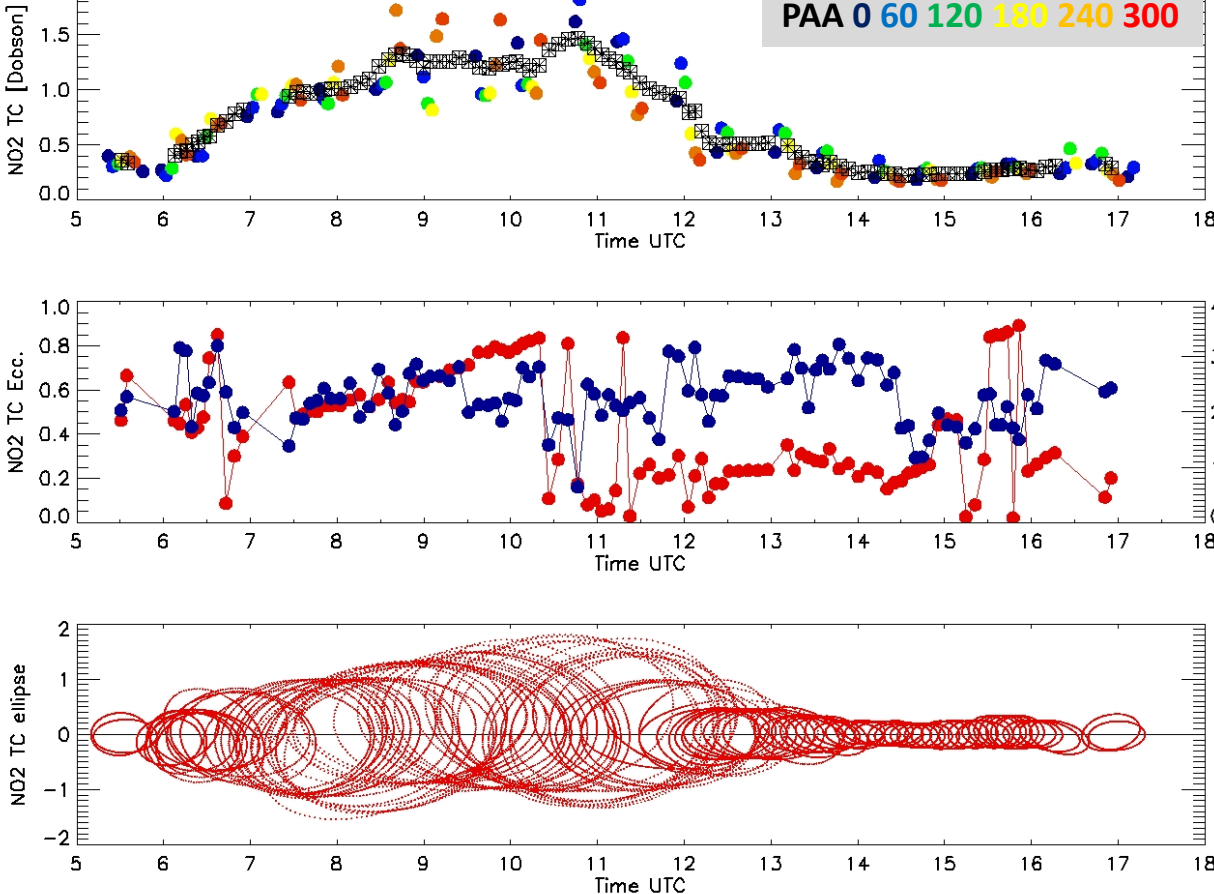

Figure 1 Left panel: ellipse shape for different eccentricity values. Centre panel: possible ellipse rotation angles (no displacement with respect to axis origin). Right panel: possible ellipse rotation angles in presence of a displacement.

Example of daily evolution of NO2 TC (2019/04/01)
- Upper panel: NO2 TC retrieved at 6 PAA (coloured dots) and related ellipse area (squared crosses)
- Centre panel: estimated ellipse eccentricity (blue, left y axis) and rotation angle (red, right y axis)
- Lower panel: retrieved ellipses (pseudo-coordinates)

The ellipses are retrieved only if 6 available MaxDOAS scans per sequence are available.

a) The rotation angle constantly drifts counter-clockwise from about 7:00 UTC
b) the eccentricity increases from 0.4 to 0.8, then drops to 0.3 to rise again
c) The wind intensity (not shown here) was lower than 2 m/s until 11:00, then increased up to 4 m/s at 13:00
d) No clouds were detected during the whole day

Abstract

A simple but effective method for the characterisation of trace gases horizontal inhomogeneity in atmospheric boundary layer is proposed.
It is based on Pandora-2S MaxDOAS measurements taken at 6 equispaced azimuth angles and on the hypothesis that the probed atmospheric 2D field values can be roughly approximated by an ellipse. The aim of this method is to express the horizontal inhomogeneity through a limited number of parameters, such as a) the ellipse eccentricity, which is a measure of departure from perfect homogeneity; b) the ellipse rotation angle, which indicates the direction of maximum species concentration as a function of the azimuth angle; c) the ellipse area, which is directly related to the species average concentration.
The main advantages of this method are:
1) The ellipse "shape" parameters are independent from physical units, allowing for the comparison of results relative to different species or concentration types (e.g. surface, tropospheric, or columnar) and facilitating long term analysis of MaxDOAS data
2) The possibility to infer concentration values for azimuth angles which could not be directly accessible due to obstacles in the observing field of view at low elevation angles, allowing for a more efficient setup of the maxDOAS measurement scheduling resulting in a higher atmospheric sampling frequency
The method has been tested using the Pandora#117 MaxDOAS measurements taken between November 2018 and March 2019 . The Pandora is operated at the Atmospheric Physics Laboratory (APL), located in the centre of Rome, in synergy with a number of atmospheric remote sensing and in situ instruments composing the urban component of the BAQUNIN instrumental suite (see Table 1 below).
In this work, five months of "ellipse" NO2 and H2O preliminary results are presented and discussed in details.
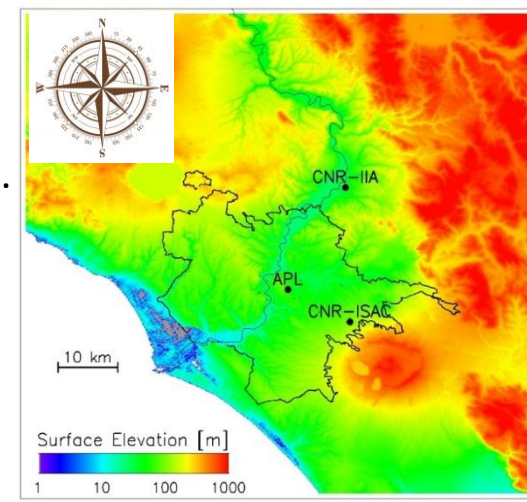
Statistical analysis of 5 months of NO2 and H2O ellipses (Nov 2018 – March 2019)
SC = surface concentration; TC = tropospheric column
Red box: NO2, Green box: H2O

NO2

H2O

SODAR
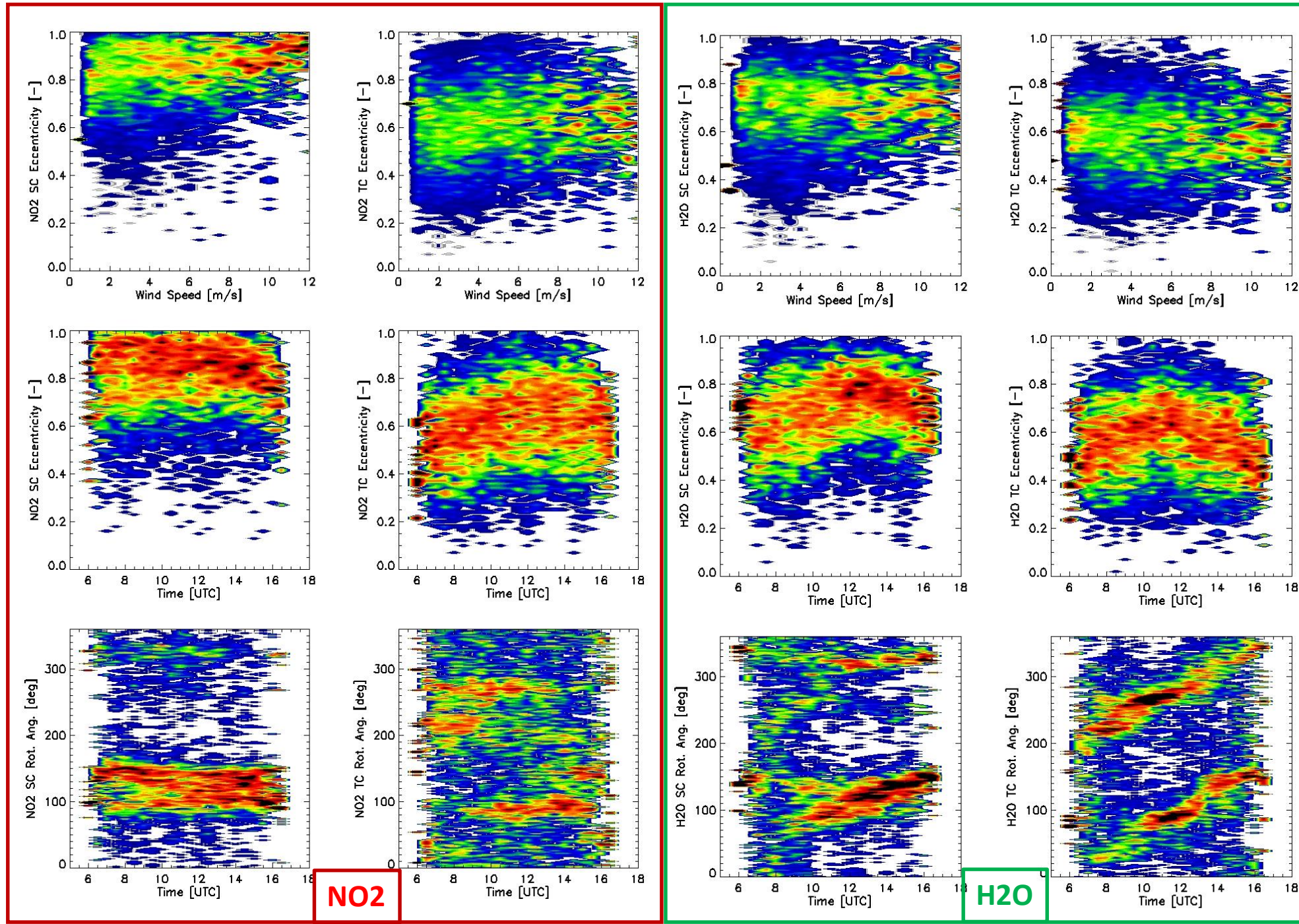
Right panel: orography of the Tiber Valley along with the position of the BAQUNIN sites. The boundaries of the Rome municipality are also shown.

Left panels: Probability Density Functions (histograms) for estimated ellipses eccentricity and rotation angle as function of environmental conditions (time of day and SODAR-PBL wind speed).
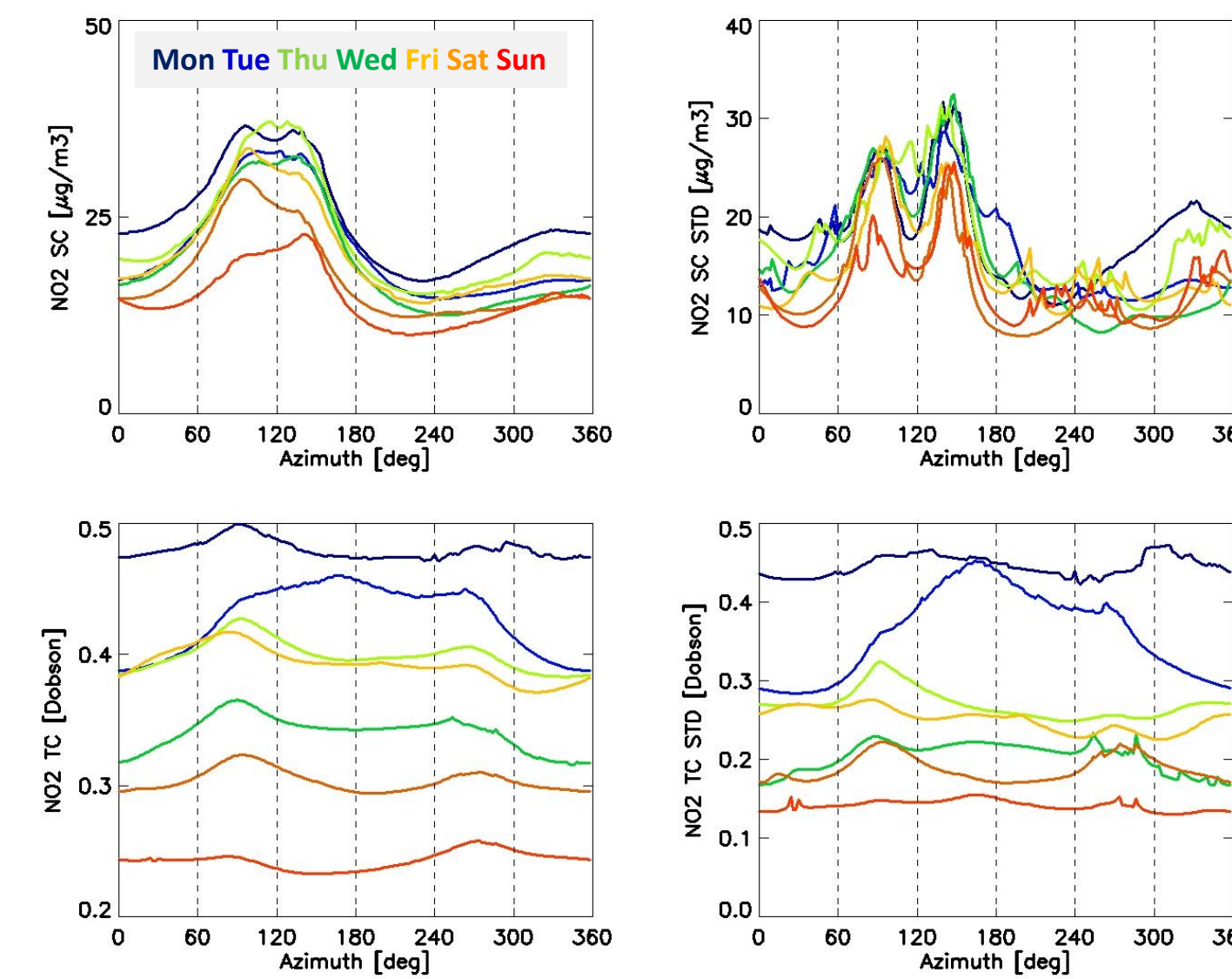The SODAR data have been averaged over the 100-300 m a.m.s.l. range
Upper row: Eccentricity vs. SODAR wind speed (U)
Middle row: Eccentricity vs. hour of day (UTC)
Lower row: Rotation Angle vs. hour of day (UTC)
Colour scale range from low (blue) to high (red) occurrences.

Given that for each sequence an ellipse is fitted, we can use the ellipse polynomial to create "synthetic" concentration values at any azimuth angle from 0 to 360 degrees.
As a test, we used the complete set of ellipses estimated in the period November 2018 to March 2019 to create synthetic NO2 SCs and TCs for 0-360 azimuth angles and 2 degrees resolution. In this way, 180 synthetic values are obtained from the 6 "real" measurements taken during each sequence.
The two upper panels on the right show the results for NO2 SC mean values and related standard deviations vs. azimuth and as function of the day-of-week (from Monday to Sunday). The two lower panels refer to NO2 TC ellipses.
In all plots, the vertical dashed lines indicate the original Pandora observation azimuth angles. As can be seen, the synthetic reconstruction seems to be not influenced by the relatively small number of observing angles.

As can be noticed, the SC and TC values follow a typical weekly evolution for urban areas (lower values and variability during weekend).

Of course, these preliminary results must be considered with caution and should be validated before using them for any purpose.

TROPOMI
PAN#117 Ellipses
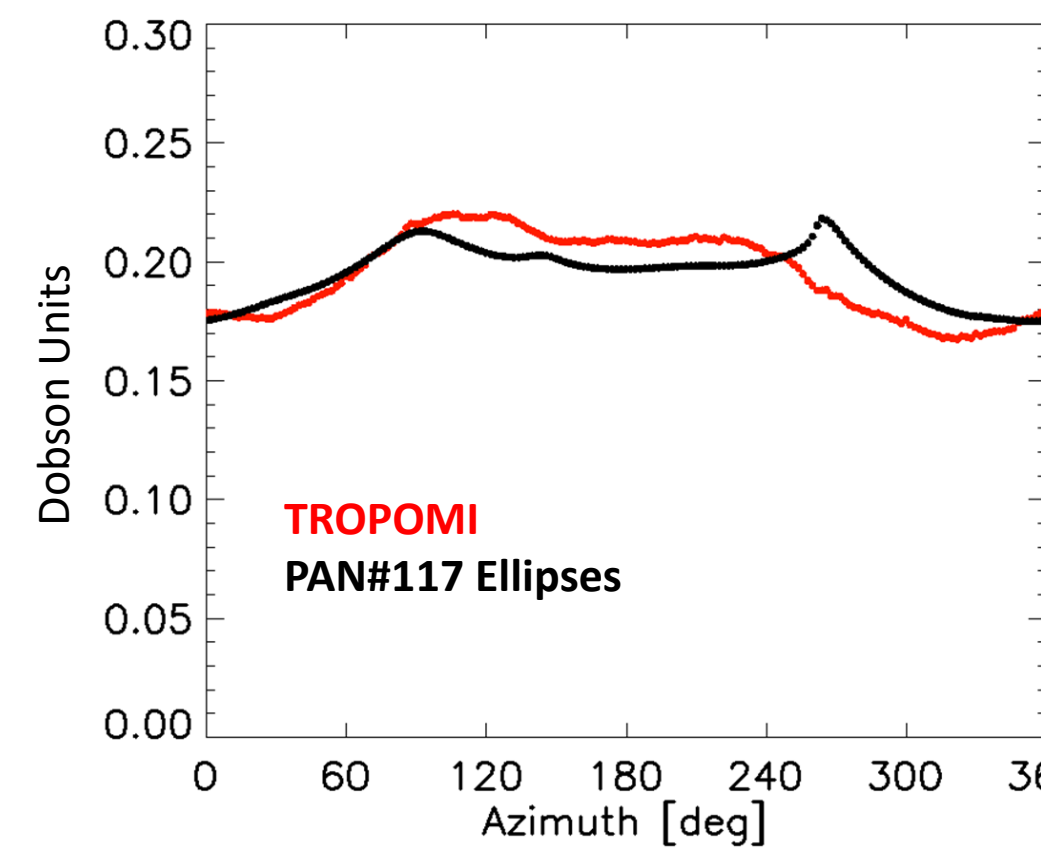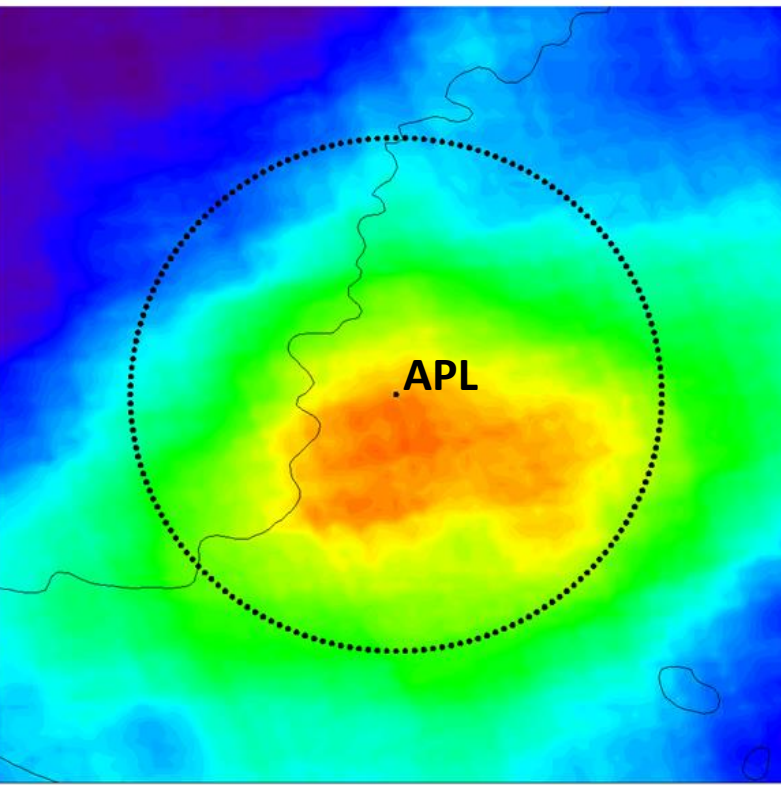
A qualitative test was carried out using the TROPOMI NO2 TC values (v1.2.2) to create a 2D TC field and averaging the TC values along segments anchored at APL (0-360 degrees azimuth, 2 degree step), in order to estimate the horizontal inhomogeneity of the TC field.
The map on the left shows the TROPOMI NO2 TC field (arbitrary colour scale), the position of the APL laboratory (Pandora #117) and the limit distance over which the average is performed (15 km from APL).
The right panel shows the TROPOMI TC results (red dots) and the Pandora ellipse data (black dots, 11:00 – 14:00 UTC sensing time) vs. azimuth. An offset of 0.17 Dobson has been subtracted to the Pandora results for sake of readability.
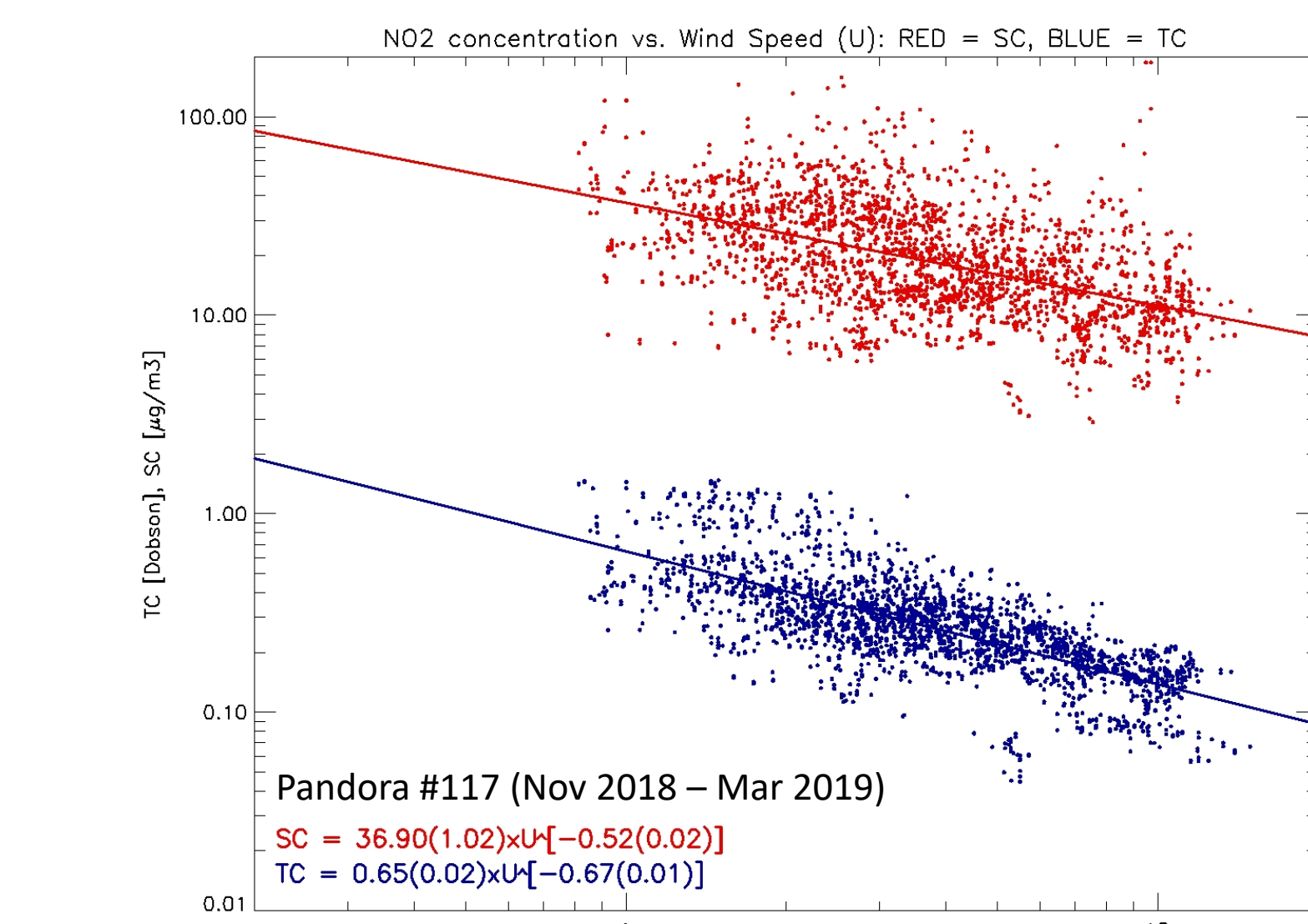
In the Urban environment of Rome, we observed that:
- Pandora NO2 VCD, TC and SC are function of PBL wind speed (not of direction!)
- TROPOMI NO2 VCD and TC are weakly dependant on PBL wind speed
- TROPOMI-Pandora NO2 difference is "predictable" from PBL wind speed measurements!

BAQUNIN sites in the context of Rome Municipality and Tiber Valley
- URBAN: Atmospheric Physics Laboratory (APL) Physics Department of University Sapienza, Lat 41.9017°, Lon 12.5158°, 75m a.s.l.
- SEMI-RURAL: Institute of Atmospheric Sciences and Climate (CNR-ISAC), Lat 41.8403°, Lon 12.6475°, 117m a.s.l.
- RURAL: Institute of Environmental Pollution (CNR-IIA), Lat 42.1057°, Lon 12.6402°, 92m a.s.l.
List of currently available BAQUNIN instruments and related operation sites (* = "maintenance")

Table 1: list of currently available devices composing the BAQUNIN instrumental suite

Instrument	Network	BAQUNIN Site	Type	Coverage
Pandora-2S	PGN	APL, CNR-ISAC, CNR-IIA	Spectrophotometer	Sunrise-Sunset (Moon)
CIMEL	AERONET-EUROPE	APL	Sun-photometer	Sunrise-Sunset
POM-PREDE	EUROSKYRAD	APL, CNR-IIA*	Sun-photometer	Sunrise-Sunset
Brewer MKIV	EUBREWNET	APL	Spectrophotometer	Sunrise-Sunset
LIDAR	N/A	APL	Active Remote Sensing	On demand (day/night)
MFRSR	N/A	APL	Actinometer	Sunrise-Sunset
Pyranometer	N/A	APL	Actinometer	Sunrise-Sunset
All-sky Camera	N/A	APL	Imager	24 hours
Meteo station	N/A	APL	In situ	24 hours
SODAR	N/A	APL	Active Remote Sensing	24 hours
FTIR	N/A	APL*	Fourier Transform Interferometer	On demand
Ceilometer	N/A	APL*	Active Remote Sensing	24 hours
Disdrometer	N/A	APL*	In situ	24 hours
WRF	N/A	ESA-ESRIN	Model	24 hours