Modeling quality objectives in the framework of the FAIRMODE project: working document.

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Summary of previous papers and scope of this work

Thunis et al. (2012) (here referred as **T2012**) proposed to use as a Model Quality Objective (**MQO**) an indicator based on the Root Mean Square Error (RMSE) between measured and modeled concentrations divided by twice the Root Mean Square of the measurement Uncertainty (RMSU). This methodology is based on a simplified formulation of the measurement uncertainty as a function of the measured concentration. The best fit coefficients for the NO2 and PM10 uncertainties were estimated in Pernigotti et al. (2013, here referred as **P2013**).

In the first section we review the robustness of the formulation for NO2. An errata found in the calculations for NO2 uncertainty in P2013 (calculation based on year 2009 data) is first detailed. Corrected fitting coefficients are calculated for 2009 and kept as reference. We then assess the robustness of the corrected coefficients by repeating the same calculation on two independent datasets (i.e. other years).

In the derivation of the yearly NO2 and PM10 MQO, the formulation is simplified by assuming a linear relationship between the averaged concentration and the standard deviation of the measurements. In the second section we discuss the validity of this assumption (see P2013 for more details).
In the third section we extend the methodology to PM2.5 while in the fourth section a first and preliminary attempt to extend the methodology to wind-speed and temperature is presented. Section 5 describes first-guess uncertainty estimates for PM components based on expert-judgment while Section 6 provides an overview of the currently available MQOs.

1. Review of the MQO formulation for NO2

a. Errata

Unfortunately an error was found in the scripts used for the statistical calculations of the NO2 model quality objective. A consequence of this error is that some uncertainty terms have been neglected, e.g. the PAN (only for non-traffic stations) and HNO2 related uncertainty terms. Neglecting these terms results in an underestimation of the non-proportional terms by about 6 ug/m3 (important at the lowest range of concentrations) but this is for the worst case considered, i.e. non-traffic stations. The overall NO2 uncertainty is however underestimated only by about 3 ug/m3 as part of the error was mitigated by the fact that the rpan term was set to the lowest value of 0.41ppb for more than 50% of the stations (traffic stations) and by the combination of NO and NOx uncertainty in the NO2 uncertainty calculation (see P2013 annex).

A comparison of the fitting parameters before and after correction is presented in the table below.

	P2013C	P2013
m	0.01	0.01
q	22	13
Ur^{RV}	0.12	0.12
alpha	0.04	0.02
(yearly data) Np	5.2	4.7
(yearly data) Nnp	5.5	6.7

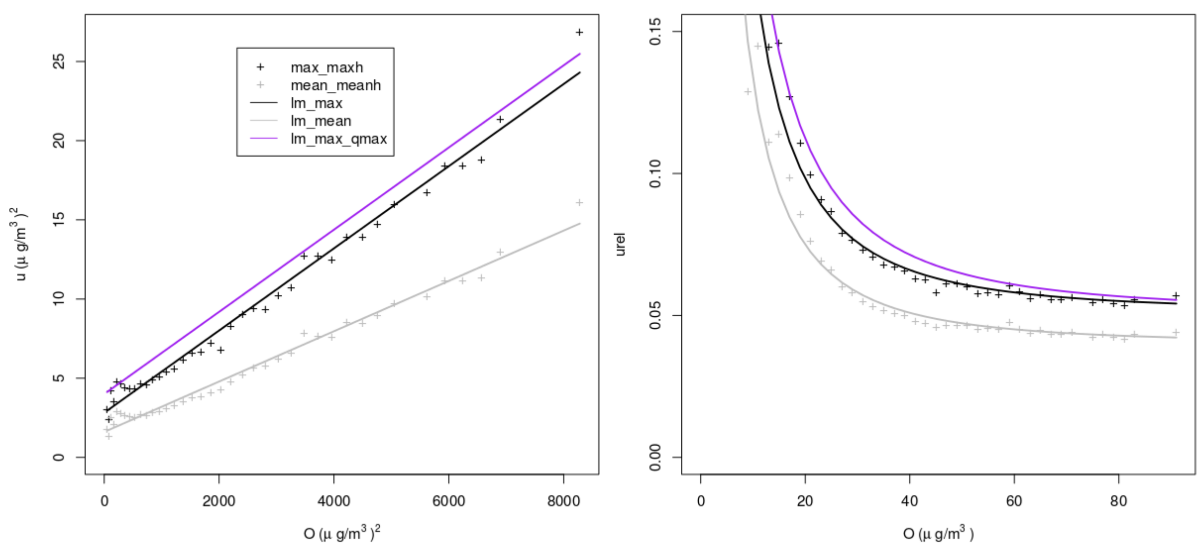
Table 1: Corrected (P2013) and old fitting parameters for hourly and yearly NO2 uncertainties

After correction, the relative uncertainty generally increases (Fig 2), by about 20% for concentrations around 5ug/m^3, 10% around 5ug/m^3, 4% at 20ug/m^3, 2% at 40ug/m^{3,} , and 1% around 75ug/m^3.

For the yearly uncertainty the correction leads to an increase of the non-proportional component. In P2013 we arbitrarily chose 2(ug/m^3)^2 as a minimum fixed level of uncertainty to make sure that our approximate simplified uncertainty function always overestimates the measured uncertainties in the low range of NO2 concentrations (conservative approach). Following the same reasoning with the corrected values we now increased this value to 4(ug/m^3)^2, see following figure (purple line in Fig1).However the two lines (see Fig 3) obtained with the corrected and P2013 formulations do not differ significantly.

In the following the corrected numbers (referred to as P2013C) will be kept as reference, i.e. the left column in the above table.

Fig 1: Corrected fitting curves for yearly NO2 uncertainty. Scatter plot (left panel) between NO2 combined uncertainty u^2 and observation classes $NO2^2$, with the draw of the best fit for the maximum (black) and mean (gray) yearly values. The purple line corresponds to the best fit for the only growing maximum uncertainty. The same data are reported (right panel) but squared and in terms of the NO2 relative uncertainty (urel).



b. **Robustness of the formulation**

The P2013 and P2013C uncertainty estimates have been obtained with the Airbase data for the year 2009 (994 stations for a total of 7421864 hourly data). We apply here the same methodology for years 2008 (849 stations, 6388781 data) and 2006 (615 stations, 4433322 data) to test the robustness of the fitting parameters.

The following table compares the P2013C 2009 results with the results obtained with the 2006 and 2008 data.

	2009 (P2013C)	2006	2008
m	0.01	0.01	0.01
q	22	31	19
Ur^{RV}	0.12	0.12	0.12
alpha	0.04	0.05	0.03
(yearly data) Np	5.2	5.5	5.2
(yearly data) Nnp	5.5	7.8	4.5

Table 2: comparison of fitting parameters obtained for 3 independant years datasets

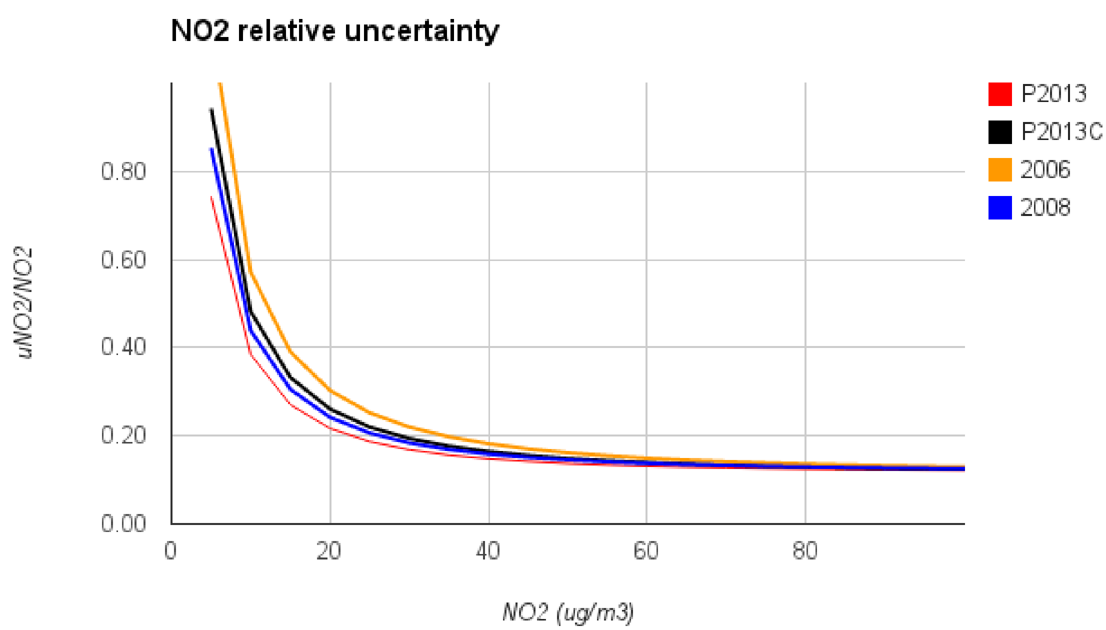


Fig 2: Hourly relative uncertainties for NO2 obtained after correction and with two other independent datasets, compared with the reference values (P2013 - in red).

As seen from the Table above differences in the uncertainty estimation arise especially for the non-proportional part (i.e. Nnp) and mostly for year 2006. When the relative uncertainty is plotted against concentration (figure 2) these differences are around 10% up to 10□g/m3, over 4% up to 35□g/m3 and 1% up to 60□g/m3 with negligible values for higher hourly concentrations.

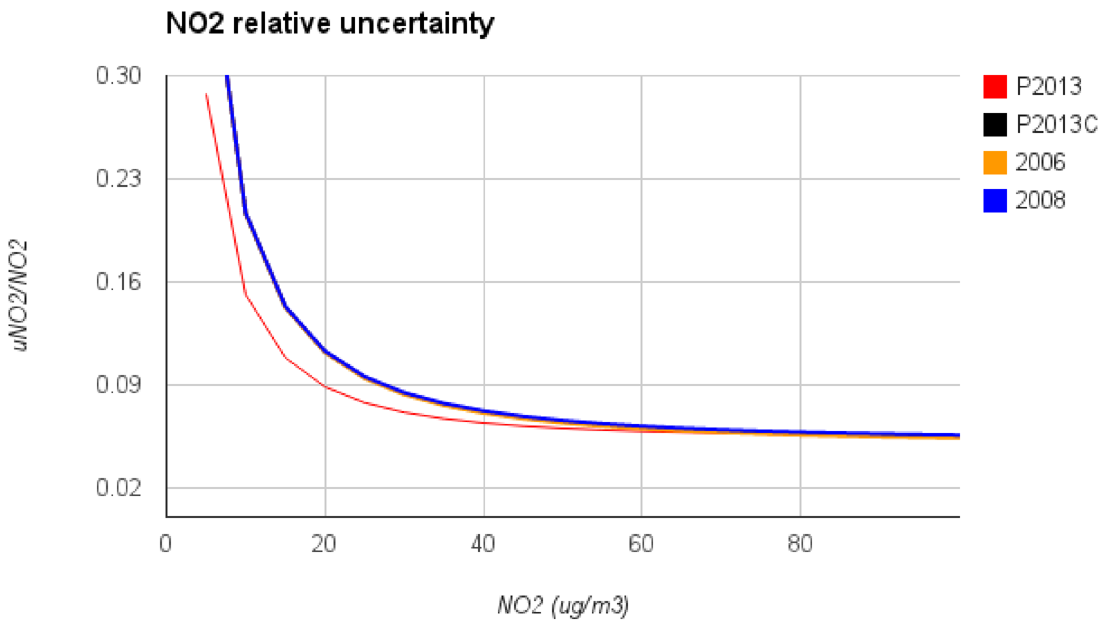


Fig 3: Yearly relative uncertainties for NO2 obtained after correction and with two other independent datasets, compared with the reference values (P2013 - in red).

For yearly data the 3 curves of fitted relative uncertainty do overlay almost perfectly.

Given the little changes in terms of parameters across years the parameters obtained in P2013C discussed above have been inserted in the current version of the Delta tool.

c. Linearity of NO2 automatic analysers - check of the randomness of the linearity deviations

a) In the NO2 uncertainty budget (see table A1, under "Lack of Fit, linearity", P2013), it is assumed that during a full year the linearity deviations can be either positive or negative over the full NO/NOx ranges with overestimations compensating for underestimations. This assumption was checked by observing a set of twenty 6-point linearity checks of analysers calibrated in 2000 (courtesy of Ricardo AEA).

Certified thermal mass flow controllers (MFC) were used to produce dynamic dilution of a high concentration gas cylinder. One MFC was used to supply a constant flow of dilution gas while another MFC controlling the high concentration gas cylinder was varied. Unfortunately the effect of possible non-linearity of the MFCs in this process cannot be excluded.

Fig 4 shows the relative linearity deviations for the 20 checks. At first glance, both positive and negative linearity deviations. Therefore, there is no reason to reject the assumption that overestimations compensates for underestimations.

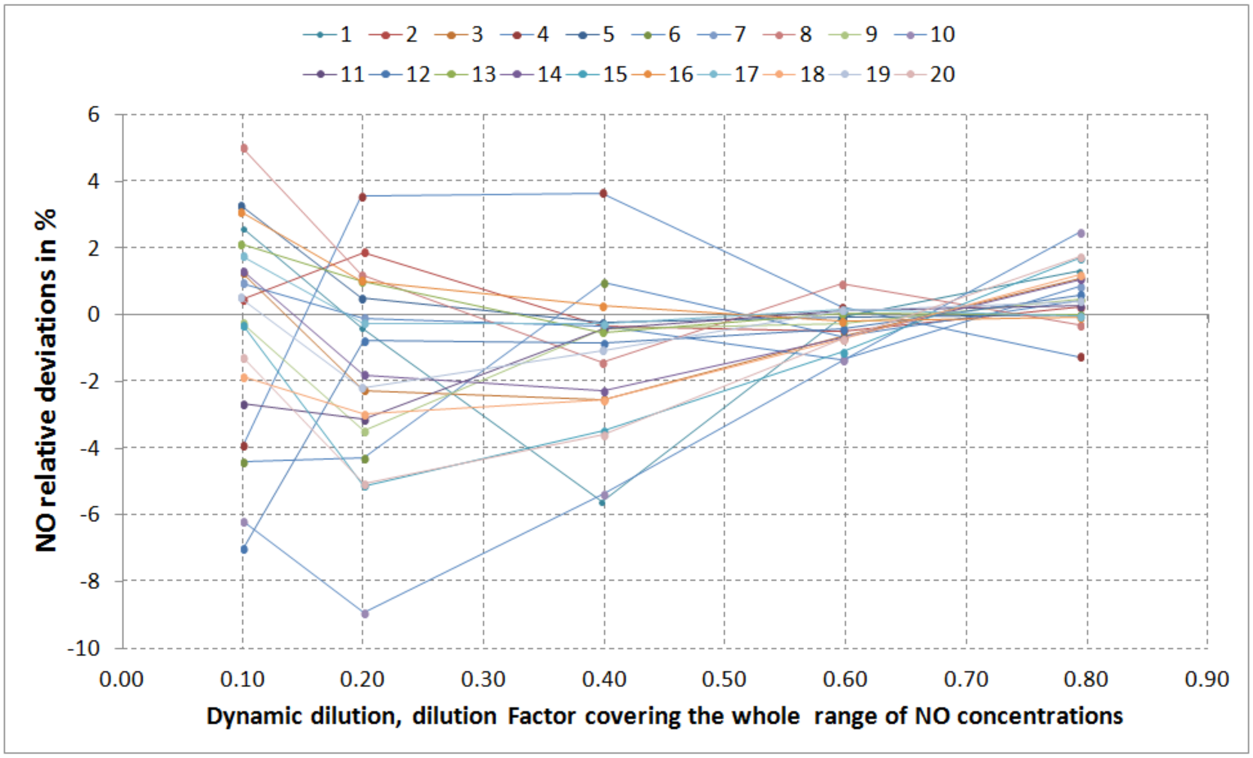


Fig 4: Relative linearity deviations of NOx analysers for 20 analysers checked by multipoint calibrations in 2000

b) In the NO2 uncertainty budget, it was assumed that the maximum linearity deviation is 5.4 % with a rectangular distribution (see table A1, under "Lack of Fit, linearity", P2013). Fig 4 shows that the maximum deviation of 5.4 % is quite in line with the reported results, with only 3 linearity deviations at low concentrations exceeding this limit. These 3 linearity deviations exceeding the 5.4-% limit at low concentrations result in low deviations in NO concentration unit.

c) The decreasing trend of the relative deviations in Fig 4 suggests that the linearity deviations in given concentration unit may show more stable value than relative ones. Linearity deviations in mV are given in Fig 5. One can observe the absence of decreasing trend of linearity deviations in Fig 5 as in Fig. 4. Therefore, it would be wise to change the estimation of relative linearity deviations to absolute deviations in the NO2 uncertainty budget.

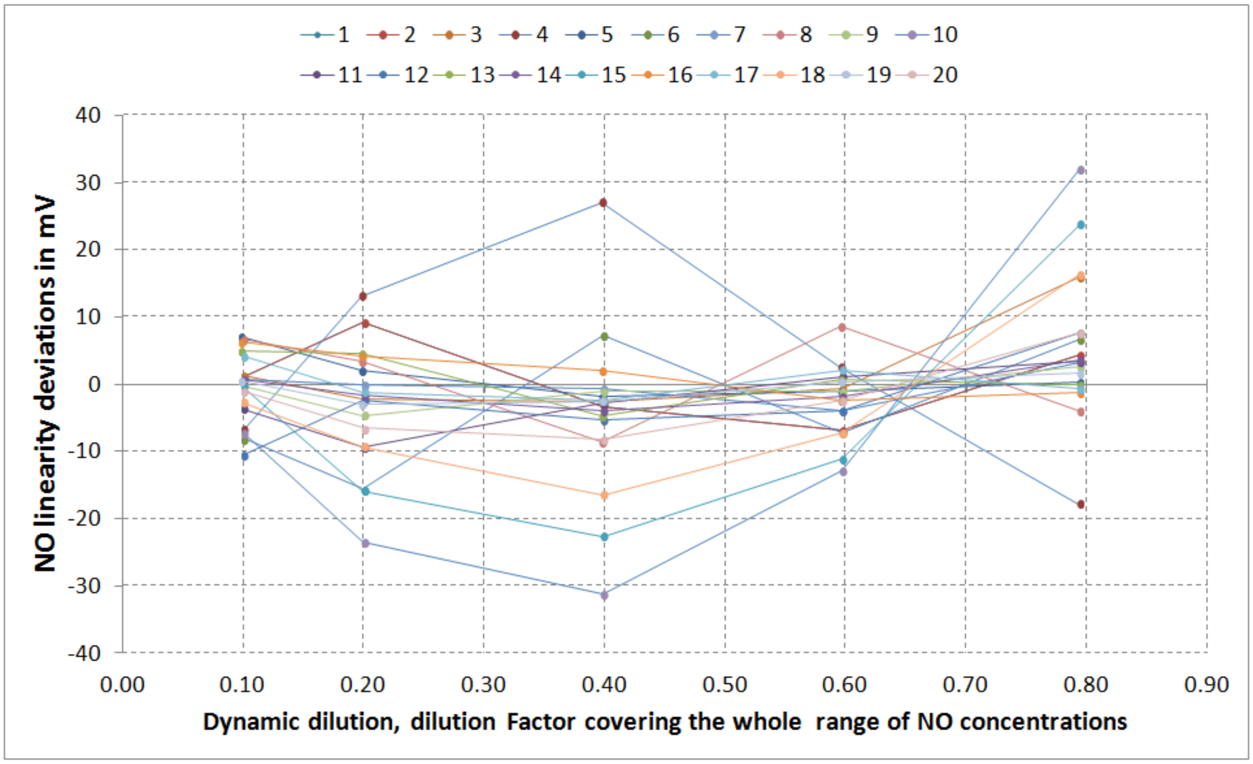


Fig 5: Linearity deviations of NOx analysers in mV for 20 analysers checked by multipoint calibrations in 2000. The linearity deviations are given in mV as registered by the data aquisition systems connected to the analysers

2. Assumption on the standard deviation term in the MQO formulation for yearly uncertainties

In P2013 the assumption is made that the standard deviation (σ) term in the yearly average uncertainty formulation can be expressed as a linear function of the mean observed value, concentration, i.e. $\sigma = l\underline{x}$ in which x is the averaged concentration value and l the proportionality coefficient, or in other words:

$$u = u_r\sqrt{\frac{(1-\alpha)\left(\underline{x}^2+\sigma^2\ \right)}{N_p} + \frac{\alpha RV^2}{N_{np}}} \approx u_r\sqrt{\frac{(1-\alpha)\underline{x}^2}{N_p^*} + \frac{\alpha RV^2}{N_{np}}} \text{ with } N_p^* = \frac{N_p}{(1+l^2)}$$

where $u_r$is a reference uncertainty, α is the non-proportional component of the uncertainty, RV is a reference value (generally equivalent to the limit value), $\underline{x}$ and σ are the observation mean and standard deviation, respectively and N_{np} and N_p are fitting coefficients introduced to account for the reduction of the uncertainty when yearly averages are considered (see P2013 for more details). In this section we evaluate this assumption between the concentration mean and the standard deviation. To do so we will check the quality of the linear fit between x^2 and σ^2+x^2.

a. NO2

Out of the 994 stations considered for 2009, 600 had at least 90% of valid data over the whole year. The linear fit of σ^2+x^2 as a function of x^2 for these stations' average data gave a slope of 1.28 (the intercept was forced through zero) and a correlation coefficient of 0.98. Note that in this section x is equivalent to NO2.

The graph below reports u^2 (u_NO2^2) as a function of x^2 ($NO2^2$) which are the data (intercept and slope) that have been used in P2013 to retrieve the formulation of the yearly uncertainty.

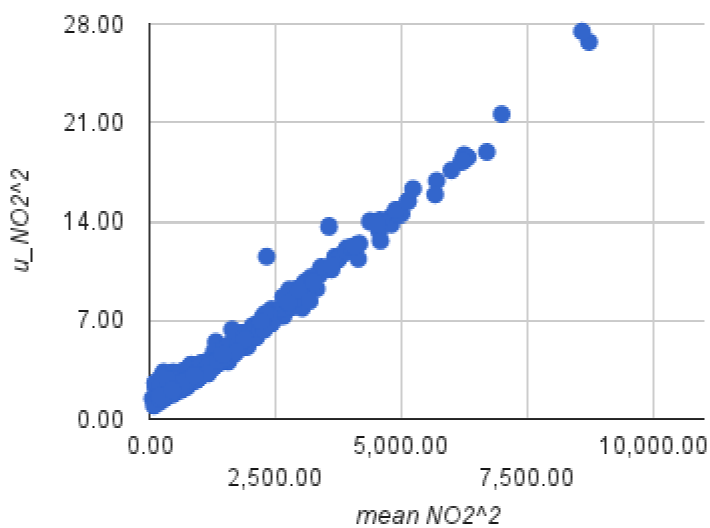


Fig6: Scatter plot of squared uncertainties against squared concentrations

We can see that this graph is equivalent to the next (within the factor of scale 1.28), in which we substituted the abscissa by the σ^2+x^2 values.

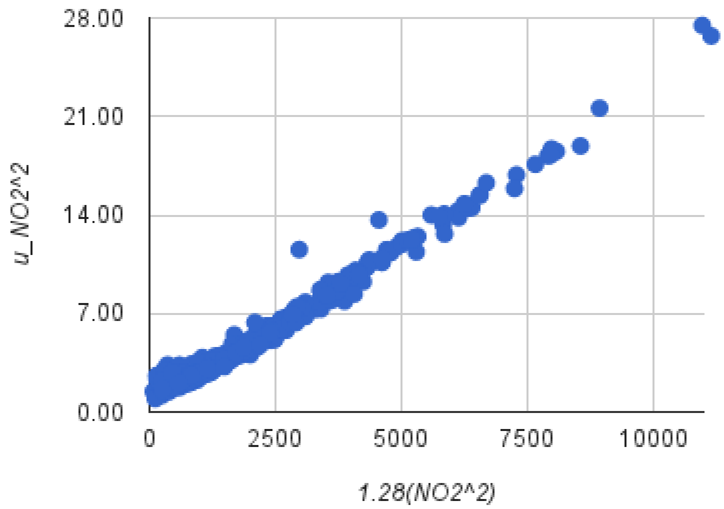


Fig7:*Scatter plot of squared uncertainties against scaled (x 1.28) squared concentrations*

While the data which should be used in theory ("true data") for the fitting are given in the next graph, with no approximation for σ^2+x^2.

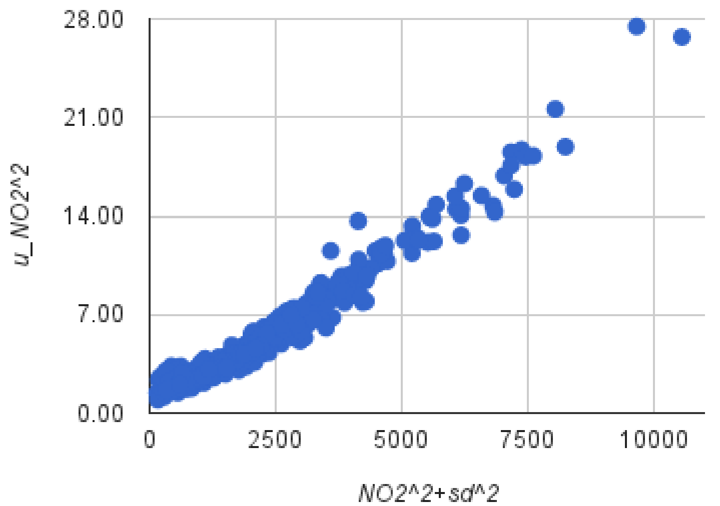


Fig8:*Scatter plot of squared uncertainties against squared concentrations plus squared standard deviation*

In the next plot we compare the uncertainty fitting obtained with the last two dataset ("original" and "fitted").

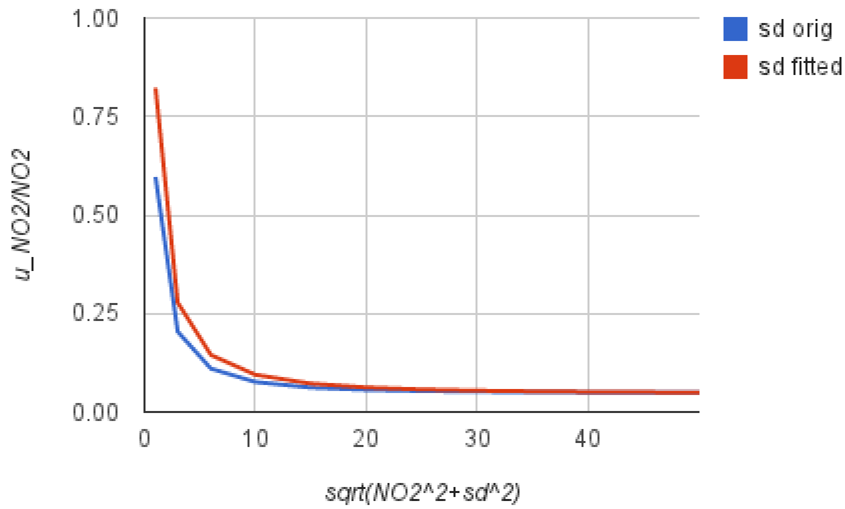


Fig9:Comparison of the NO2 yearly uncertainty functions with and without the standard deviation linearisation assumption

From the above graph it can be seen that using a fitting approximation for σ^2+x^2 in terms of x^2 leads to some slight differences in terms of relative uncertainty for small values while differences become negligible at the higher range of concentrations. We also note that the approximated curve always over-estimate the uncertainty values (conservative approach).

Given that the fitting of u^2 versus x^2 is equivalent to the fitting of u^2 versus $1.28x^2$ we therefore demonstrated here that the approximation made in P2013 can be considered as valid.

b. PM10

In P2013 a holistic approach was used to determine the uncertainty related to PM10 daily measurements and the uncertainty coefficients were estimated via an inter-comparison campaign data where parallel measurements using gravimetric technique were available.

In order to estimate yearly uncertainty the coefficients found for the gravimetric method were used in stations where gravimetric and beta-ray techniques were both available (AirBase database, 5 stations for 2009 having data coverage over 80% for both gravimetric and beta, located in Austria). The assumption was made here that Np and Nnp were approximately equal for gravimetric and beta-ray. An iterative process led to the final result of Np=40 whereas Nnp was difficult to estimate and was assumed equal to 1 (conservative estimate).

The linear fit of σ^2+x^2 as a function of x^2 on all available gravimetric daily data averaged over the whole year (3009 stations) gives a slope of 1.39 and a correlation of 0.98. We make the assumption that this fitting coefficient is also valid for beta ray stations. Indeed almost the same fitting coefficients were found with both the beta and gravimetric data for the limited dataset of 5 stations.

In all three approaches, u^2 vs. x^2, u^2 vs. $(1.39x)^2$ and the “true” fitting line, the uncertainty-concentration slope is approaching zero (of the order 10^{-3}) meaning that the proportional part of the absolute uncertainty disappears and the absolute uncertainty is practically constant for the whole range of concentrations with Np ranging between 20 to 40. The absolute uncertainty is then fixed by the non-proportional term. As the assumption on the standard deviation is made on the proportional term and given that this part of the uncertainty disappears for yearly data the three different methodologies lead to the same result. In the next picture an arbitrary value of 0.8ug/m^3 was set for the fixed uncertainty.

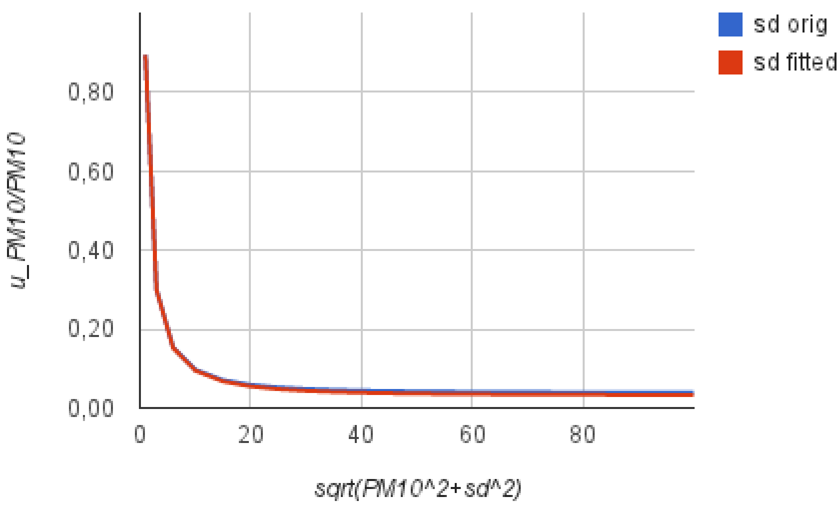


Fig10: *Comparison of the PM10 yearly uncertainty functions with and without the standard deviation linearisation assumption*

3. Extension of the MQO formulation to PM2.5

For PM2.5 a similar to the PM10 approach (see P2013) is used, namely the holistic or direct approach based on the guide for demonstration of equivalence (ECWG, 2010, hereon GDE). With this methodology parallel measurements are used to estimate the uncertainties related to the various PM2.5 measurements technique. Field gravimetric, TEOM and BETA instrument uncertainties are estimated based on measurements collected in the frame of the European PM QA/QC Programme (Lagler et al., 2011) in which a JRC (Joint Research Centre - European Commission) gravimetric reference sampler was co-located with monitoring network instruments in 18 European countries between 2006 and 2009 (15 days campaigns).
The first step was to apply the methodology on the gravimetric data from the European PM2.5 QA/QC Programme. The concentrations have been monitored with 19 other gravimetric

instruments but the requirement of 10 days of data reduced the dataset to 140 days for 14 stations (actually 139 since 1% of extreme values are excluded from the analysis). Two measurements obtained with the same method (gravimetric) are compared. According to the results discussed in Lagler et al. (2011), a value of 5% has been used for the uncertainty of the JRC reference data (see P2013 for details on the methodology). The fitting coefficients obtained from the analysis described above are listed in the table below and compared to those obtained for PM10.

	RV	urRV	α	m	q
PM10	50	0.14	0.018	0.02	0.9
PM2.5	25	0.18	0.018	0.03	0.4

Table 3: Fitting parameters for PM2.5. PM10 values are recorded for convenience

For TEOM (30 day data for 3 stations) and Beta-ray (50 day data for 5 stations) the results give extremely high uncertainties (urRV respectively 0.524 and 0.400) with a relative uncertainty increasing with the measured concentration (negative values for α). The following plots depict the analysis results. In conclusion more data are certainly needed to assess the robustness of the formulation for other techniques than gravimetric.

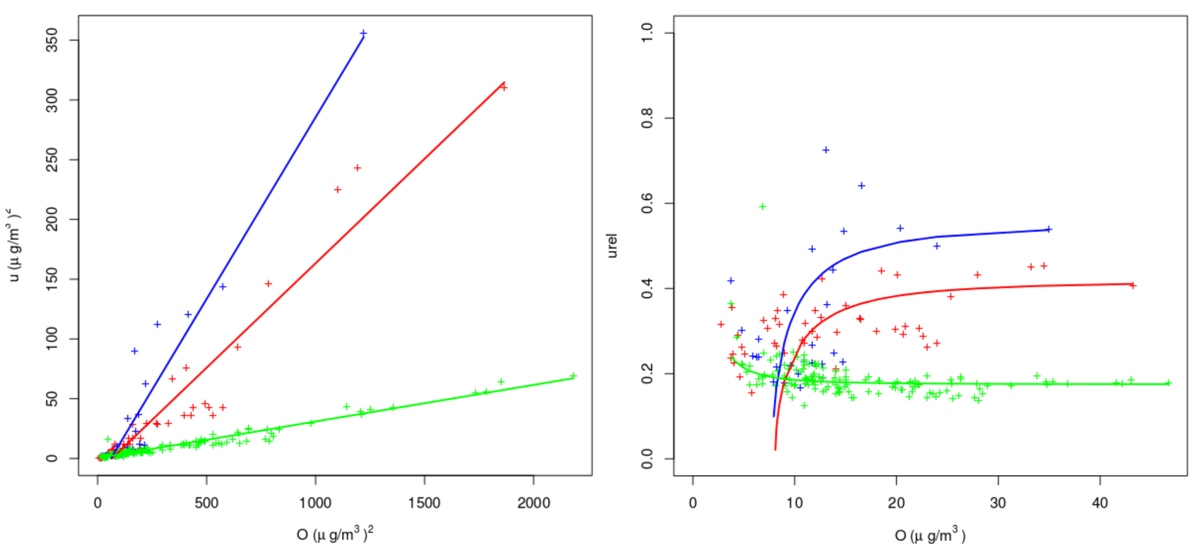


Fig11: Scatter plot (left panel) between the PM2.5 uncertainty u^2 and observations PM2.5^2 , with the draw of the best fit for daily values. The same data are reported (right panel) but squared and with the uncertainty relative (urel) to PM2.5 concentration as y-axis. For the uncertainty calculation data from an inter-comparison measuring campaign have been used (green for gravimetric, blue for TEOM and red for beta).

4. Extension of the MQO formulation to temperature and wind-speed

NOTE: THE CONTENT OF THIS SECTION IS STILL VERY PRELIMINARY!!!!!

a. Temperature

The first dataset considered here was provided by VITO and consist of two meteorological stations located in the same site from 2012-05-29 16:15 to 2012-06-13 17:30 for a total of 1447 data with a time resolution of 15'. In particular it consists of two PT1000 air temperature measurement inside actively ventilated radiation shield.

An attempt is made to extend the technique adopted for PM10 and PM2.5 to these data.

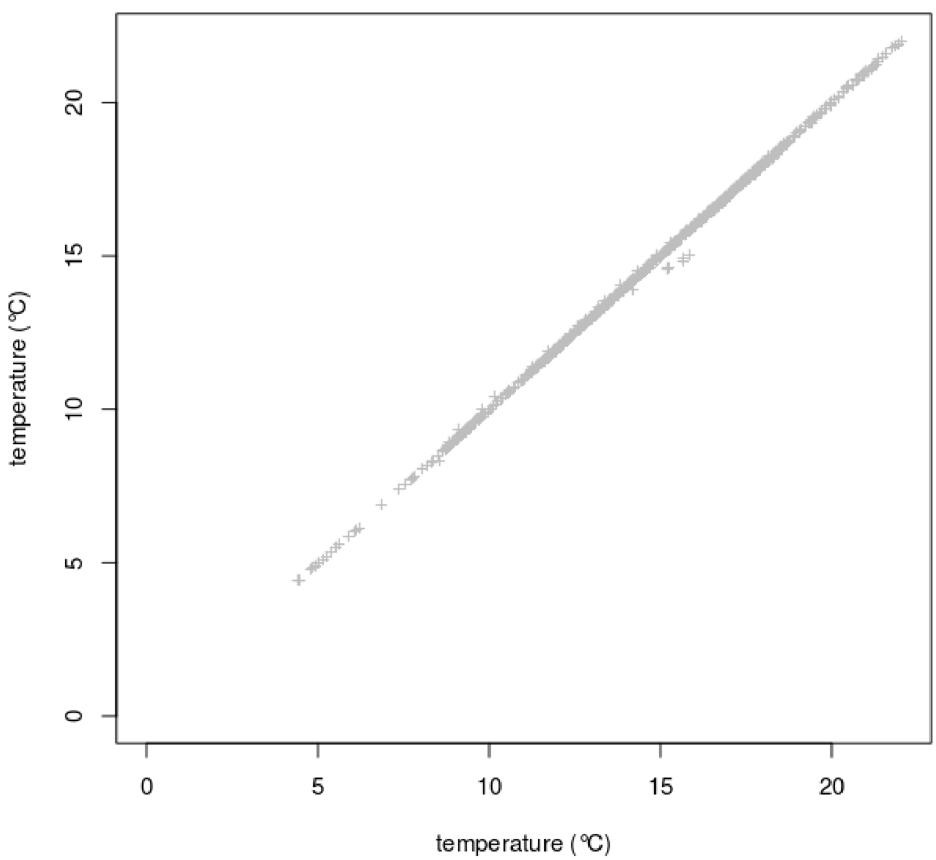


Fig 12: Scatter plot of measured (1) vs. measured (2) temperature measurements

The scatter plot this dataset (Fig. 10) shows that the two sources of data are very well correlated, with data practically indistinguishable (RMSE ~ 0.02 °C). Similar results are found with the different instruments located in ventilated radiation shields (RMSE 0.03 – 0.06 °C).

The methodology used for PM10 was applied to temperature using a trial and error procedure in order to give the same uncertainty to both the stations. As expected the result of the fitting of u^2 as a function of the t^2 shows practically null coefficients m=1.4 10^{-5} q=3.9 10^{-5}.

In this case it is probably necessary to use a different approach and/or to test different dataset. Given the lack of data for deriving robust estimates, we temporarily opted for a simplified approach based on the conclusions contained in Leroy (2002). The source of uncertainty is not so much the temperature measurement in itself but rather the quality of the shielding structure and the location where the measurement is made. In a regular temperature shield, many effects can influence the precision of temperature measurement: ventilation within the shield, wind speed, radiative effects...As a result of all these uncertainty sources Leroy states that the error on temperature measurement might be around 1 degree (in case of high radiation and low wind). In the case of badly designed shields this error might be doubled or tripled. In the current MQO version, we opted for a constant and equiprobable (i.e. rectangular distribution) uncertainty (i.e. u) over the whole range of temperature values. This results in a constant value for the combined uncertainty (u) of 0.575 (=$1/\sqrt{3}$). This choice results in the following fitting parameters. Note that this choice of u=0.575 K leads to U=1.15K and an MQO tolerance margin about 2.3 K

	k	α	u_r^{RV}	RV
TEMP	2.00	1.00	0.023	25

Table 4: list of parameters used to define the simplified uncertainty formulation for temperature

b. Wind speed

The first dataset considered here was again provided by VITO and consist of two meteorological stations located in the same site from 2012-05-29 16:15 to 2012-06-13 17:30 for a total of 1447 data with a time resolution of 15’. In particular two average wind speed (measured by sonic anemometer : WindSonic).

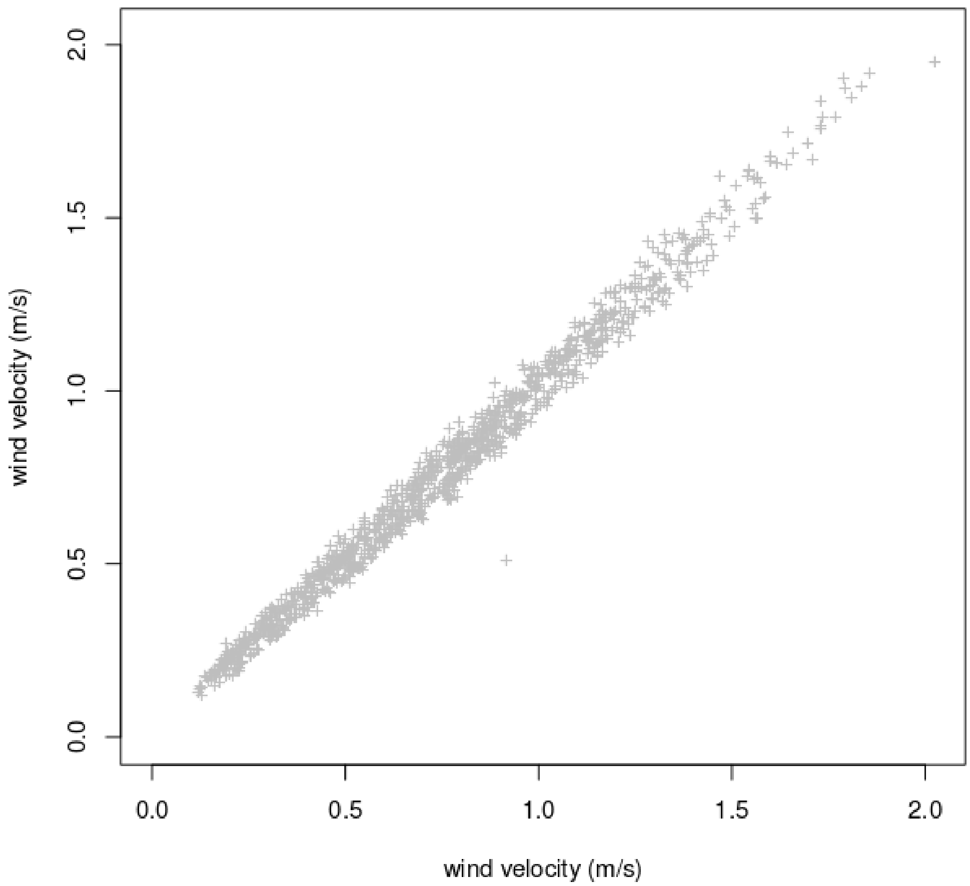


Fig 13: Scatter plot of measured (1) vs. measured (2) wind measurements

When the PM10 methodology is applied to wind speed more reasonable but still small values of uncertainty are found, i.e. m=2.4e-03 q=8.1e-05. Above 0.5m/s the relative uncertainty is around 5%.

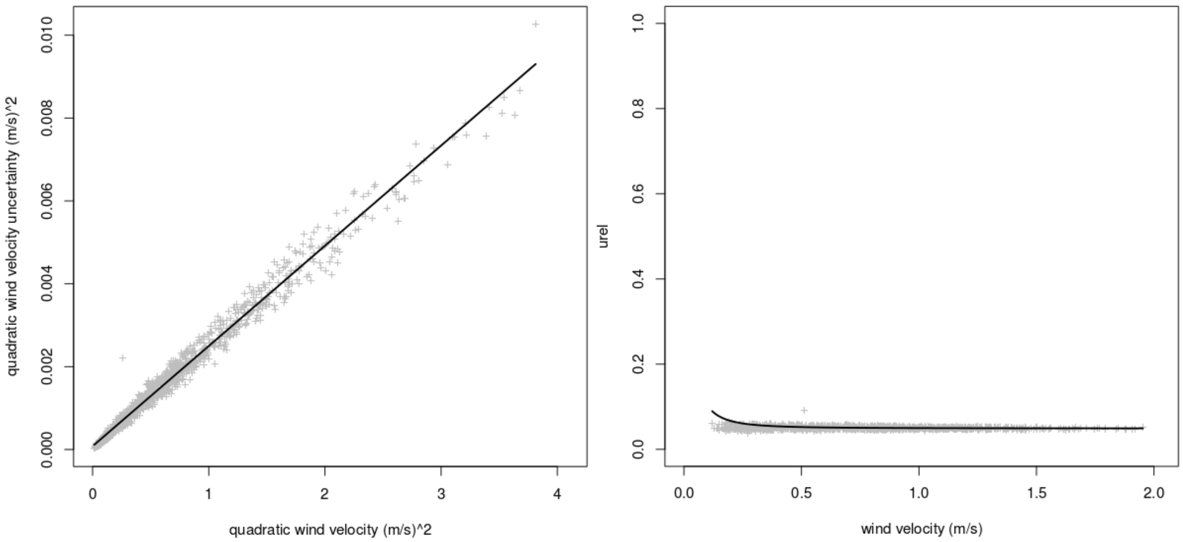


Fig 14: Scatter plot (left panel) between the wind speed uncertainty u^2 and observations^2 , with the draw of the best fit for 15' values. The same data are reported (right panel) but squared and with the uncertainty relative (urel) against the wind speed. For the uncertainty calculation VITO data from an inter-comparison measuring campaign with sonic anemometers is used.

Also in this case it seems that these values of uncertainty are very low and may refer to sonic anemometers only but not to routine anemometers normally used for meteorological applications.

Another dataset was provided by the Croatian Meteorological institute consisting of a monitoring campaign for wind speed and direction on the island of Vis at 600m altitude. In this case 46716 data every 10' from 2009-01-05 09:10 to 2009-12-27 14:40.
In the scatter plot of the two stations it is evident that while the comparison is coherent for most data, different wind regimes are however measured under certain circumstances, .

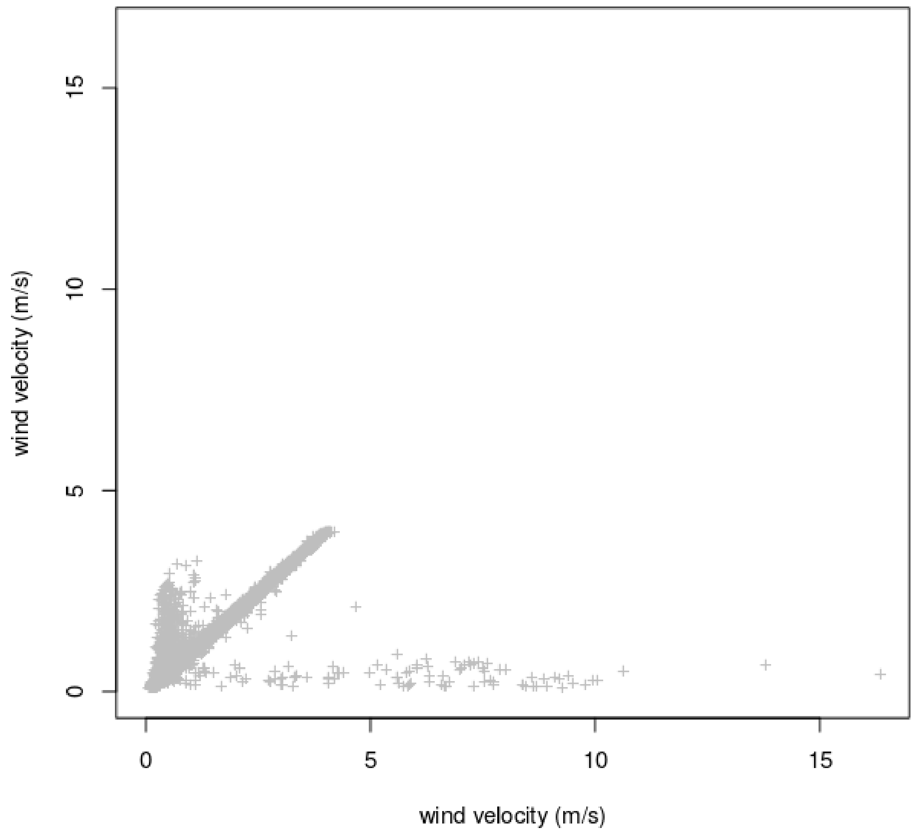


Fig15: Scatter plot of measured (1) vs. measured (2) wind speed measurements for the site of Vis.

When the methodology used for PM10 is applied we find uncertainty coefficient of q=0.0006 and m=0.026. Above 0.5m/s the relative uncertainty remain stable around 16%.

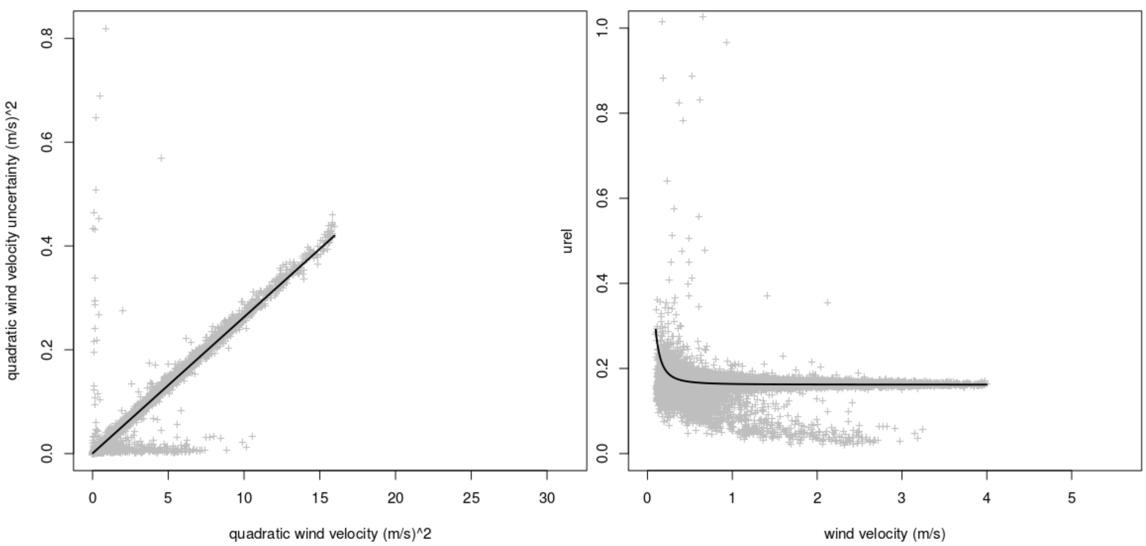


Fig 16: Scatter plot (left panel) between the wind speed uncertainty u^2 and observations^2 , with the draw of the best fit for 10' values. The same data are reported (right panel) but squared and with the uncertainty relative (urel) against the wind speed. For the uncertainty calculation Croatian Meteorological institute data from an inter-comparison measuring campaign with anemometers is used.

Similarly to temperature, given the lack of sufficient data, we also opted here for a simplified approach based on the WMO technical guidance document which report a fixed uncertainty of 0.5 m/s below 5 m/s and a proportional uncertainty equal to 10% of the wind speed value above 5 m/s. On top of this we also assumed an equi-probable uncertainty (+/- 0.5 m/s error) resulting from the frequent rounding of wind measurements to an integer value. These assumptions lead to the following fitting parameters and uncertainty function.

	k	α	u_r^{RV}	RV	N_p	N_{np}
WS	2.00	0.8	0.13	5	N/A	N/A

Table 5: *list of parameters used to define the simplified uncertainty formulation for temperature*

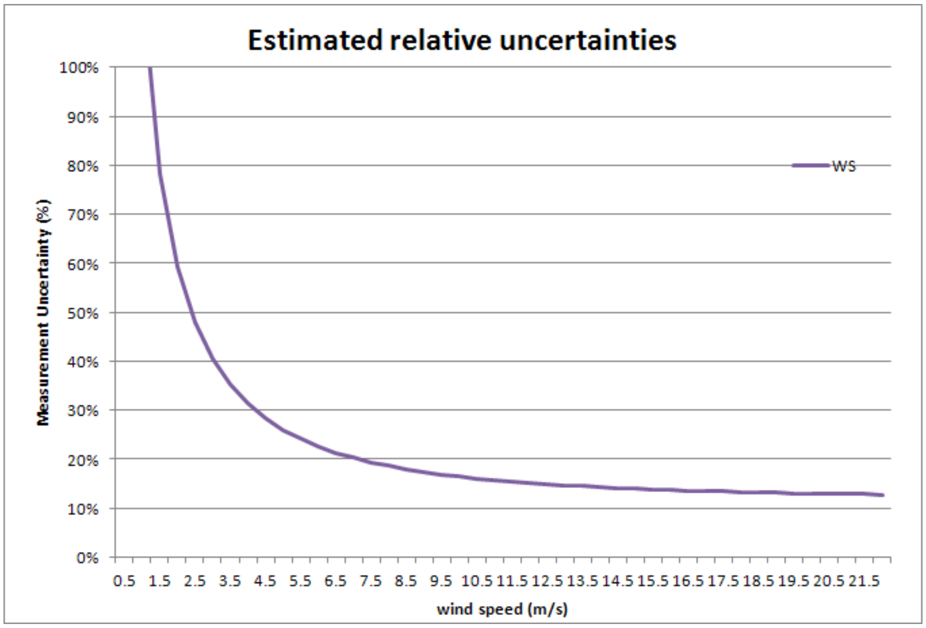


Fig 17: Overview of the combined relative hourly uncertainties (u) for wind speed

5. Extension of the MQO formulation to PM components

Estimation of the uncertainties for these PM components rely on expert judgment. They are here provided only for testing purposes and as a starting point for further discussion and before refinement. Values presented in the table below are valid for both their fine and coarse (if relevant) fractions. The same repartition between the proportional and non-proportional fractions of the uncertainty has been assumed similar for all PM components.

	k	α	u_r^{RV}	RV	N_p	N_{np}
PM10	2.00	0.018	0.140	50	40	1
PM25	2.00	0.018	0.180	25	40	1
SO4	2.00	0.018	0.150	7	40	1
NO3	2.00	0.018	0.150	8	40	1
NH4	2.00	0.018	0.225	4	40	1
EC	2.00	0.018	0.375	5	40	1
TOM	2.00	0.018	0.375	10	40	1

Table 6: *list of parameters used to define the simplified uncertainty formulation for PM components*

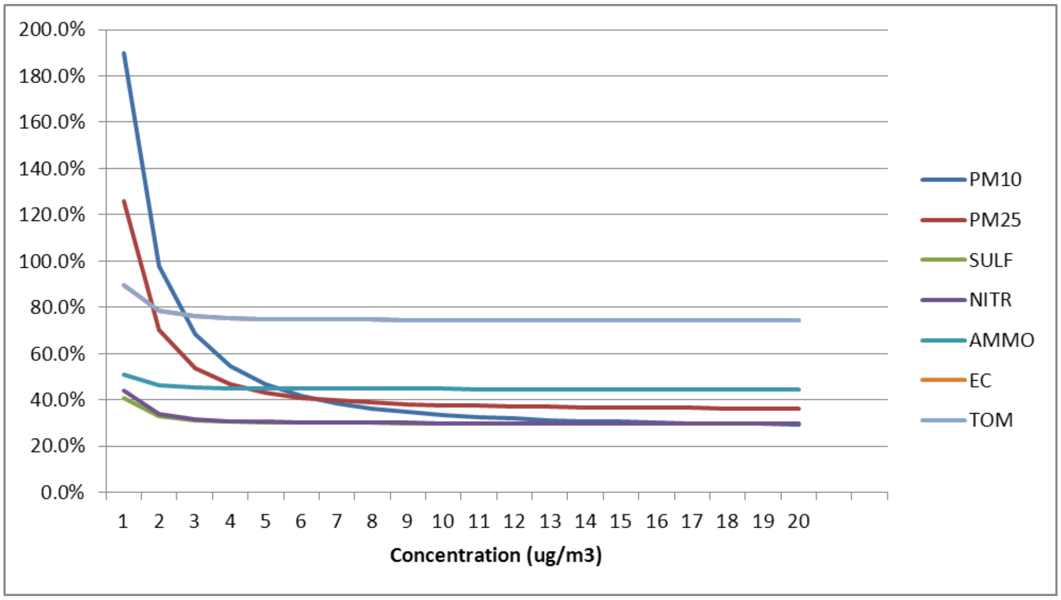


Fig 18: Overview of the combined relative uncertainties (u) for PM components

6. Overview of currently available Air Quality MQO

In this section we summarize the MQO available for air quality variables. The table summarizes the current parameter fitting values while the two figures below provide the uncertainty curves for daily/hourly and yearly data respectively.

Pollutant	k	α	u_r^{RV}	RV	N_p	N_{np}
NO2	2.00	0.040	0.12	200	5	12
PM10	2.00	0.018	0.14	50	40	1
PM25	2.00	0.018	0.18	25	40	1
O3	1.40	0.62	0.09	120	N/A	N/A

Table 7:Fitting parameters for the simplified uncertainty formulations

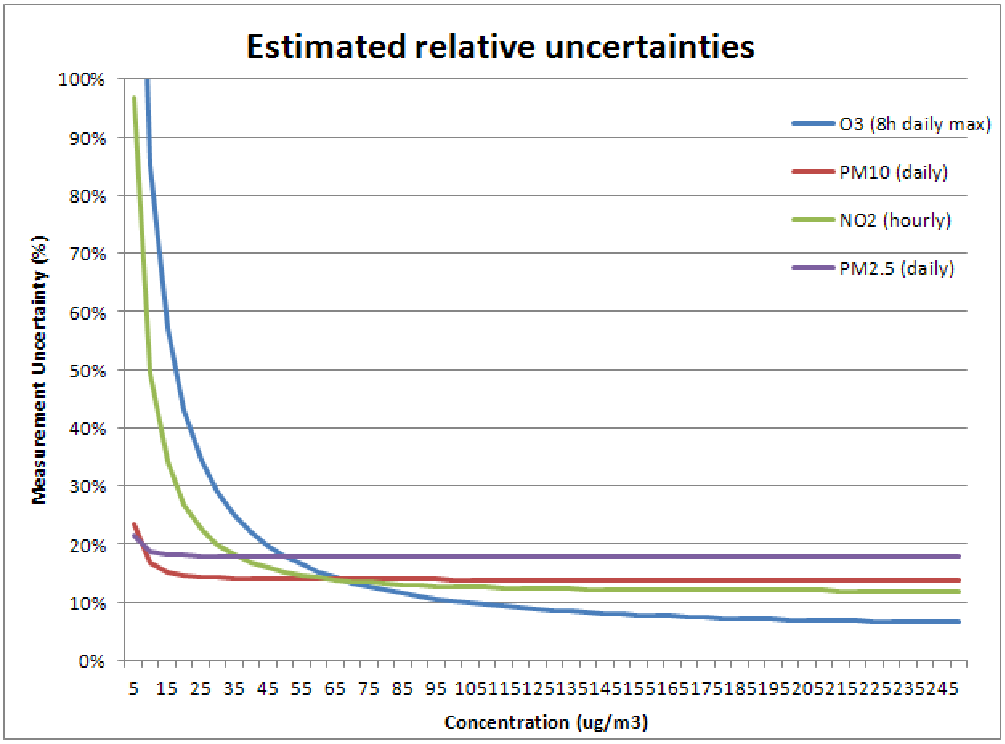


Fig 19: Overview of the combined relative hourly/daily uncertainties (u) for different pollutants

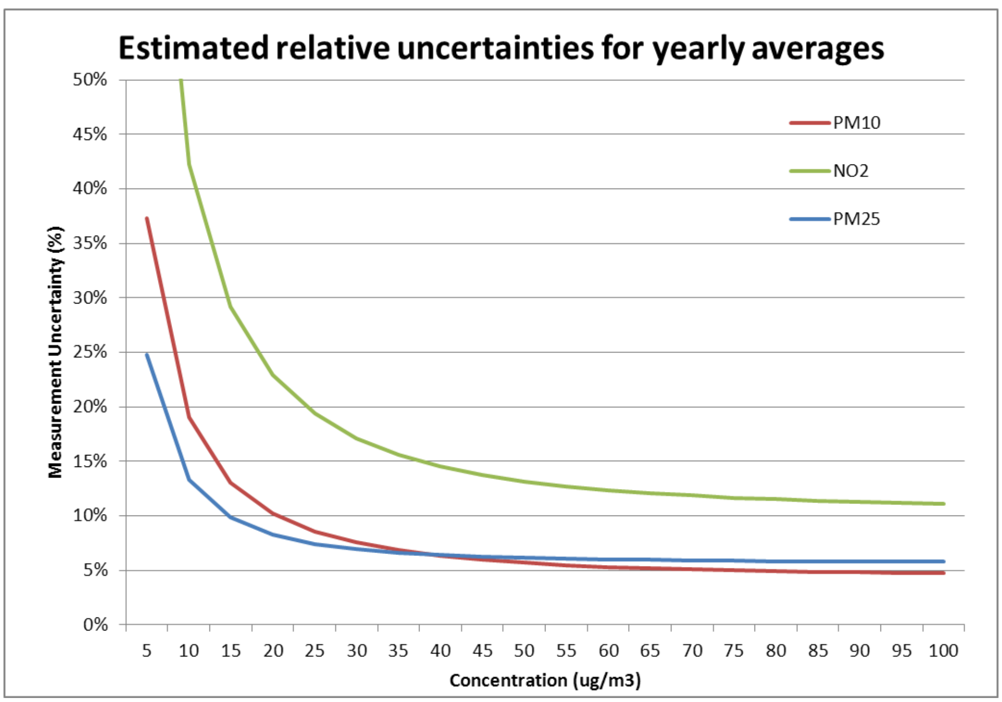


Fig 20: Overview of the combined relative yearly uncertainties (u) for different pollutants

7. Bibliography

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