ARM2 METHOD TO ESTIMATE NO_2 AIR CONCENTRATIONS BY USING NO_X AIR CONCENTRATIONS OBTAINED BY AIR POLLUTION MODELS: VERIFICATION AND ADAPTATION BY USING AIR QUALITY NETWORK OF TUSCANY DATA

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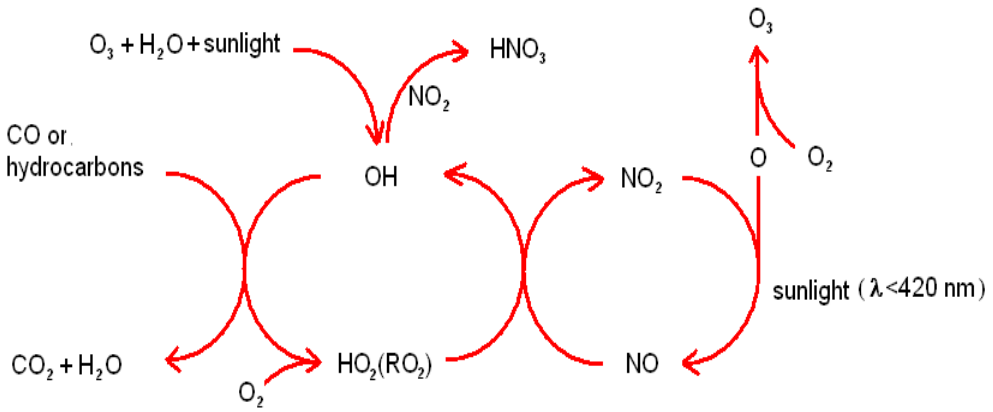
Air Quality Standards for Nitrogen dioxide:

- 40 µg/m^3 for the annual average concentration
- 200 µg/m^3 for the 99.8° annual percentile of the hourly concentrations.

(European Air quality Directive 2008/50/EC)

Nitrogen oxides are emitted as a mixture of NO and NO_2 by the majority of antropic sources (traffic, industrial processes, energy production, etc).

Both gases are also highly reactive through the oxidation of NO with ozone and the photo-dissociation of NO_2 to NO.



Therefore, it is critical to be able to assess as precisely as possible the actual NO_2 increase in ambient concentrations generated by industrial and traffic sources, whose emission ratios are usually expressed as total Nitrogen oxides ($NOx \approx NO+NO_2$).

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US-EPA has approved[1] a three-tiered estimation approach to calculate NO_2 concentrations based on NOx air concentrations obtained by models, that includes the ”Tier 2” method Ambient Ratio Method (ARM). ARM applies an empirically derived conversion factor, based on observed NO_2/NO_x ratios of monitoring data, to the modeled NO_x concentrations

Data indicates that the NO_2/NO_x ratio is variable and decreases with the proximity to emission sources, characterized by low NO_2/NO_x in-stack ratio. (usually traffic and combustion-based industrial processes).

This suggests that fixed ratios overestimate NO_2 air concentrations in the near field of the emission, where usually the most relevant impacts are expected.

ARM version 2 (ARM2) has been developed[2] using 1-h air monitoring data to take into account the variability of the conversion factor as a function of NO_x concentration. Currently US-EPA is proposing[3], as "Tier 2" approach, to replace the old ARM method with the more refined ARM2

[1] *US-EPA 2005: Revision to the guideline on Air quality Model: Adoption of a Preferred General Purpose (Flat and Complex Terrain) Dispersion Model and Other revisions; Final Rule*

[2] *RTP Environmental Associates, Inc. 2013: Ambient Ratio Method Version 2 (ARM2) for use with AERMOD for 1-hr NO_2 Modeling, Development and Evaluation Report.*

[3] *US-EPA 2017: Technical support Document (TSD) for NO_2-related AERMOD modifications*

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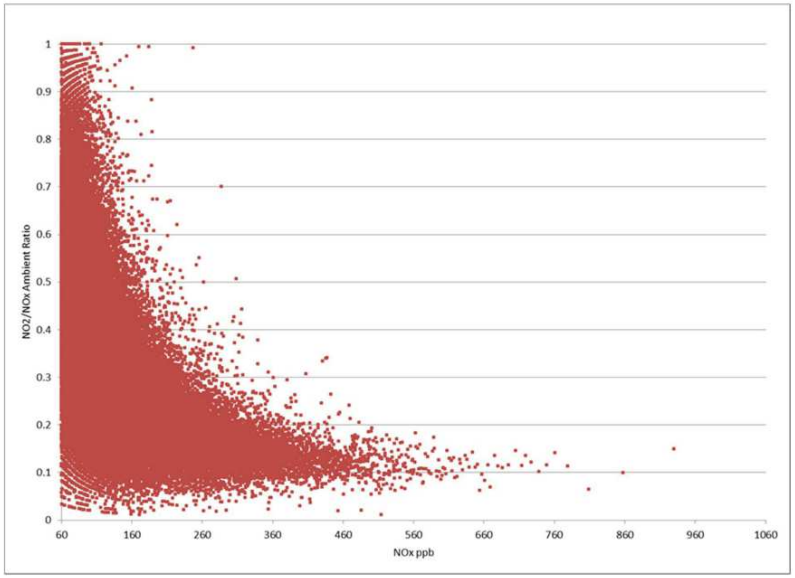
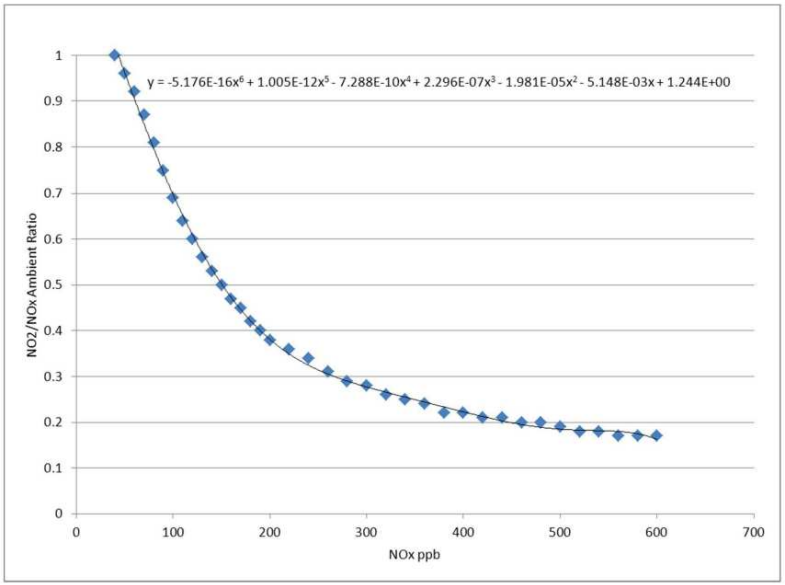
The ARM2 empirical relationship (a 6th grade polynomial function) between NO_x hourly average concentrations and the corresponding NO_2/NO_x ratios has been obtained analyzing a 10-year data set extracted from the US air quality monitoring network.





RTP Environmental Associates, Inc. 2013: Ambient Ratio Method Version 2 (ARM2) for use with AERMOD for 1-hr NO_2 Modeling, Development and Evaluation Report.

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We present an adaptation of the US-EPA ARM2 method to the local environmental conditions of Tuscany, using the data from the Tuscany Air Quality Monitoring Network database

The utilized data-set includes:

- all the NO_x and NO_2 hourly concentrations from all the stations of the network active from 2007 to 2016
- all the NO_x and NO_2 hourly concentrations from the stations active from 1999 to 2006 where the NOx averages are greater than 300 $\mu g/m^3$.

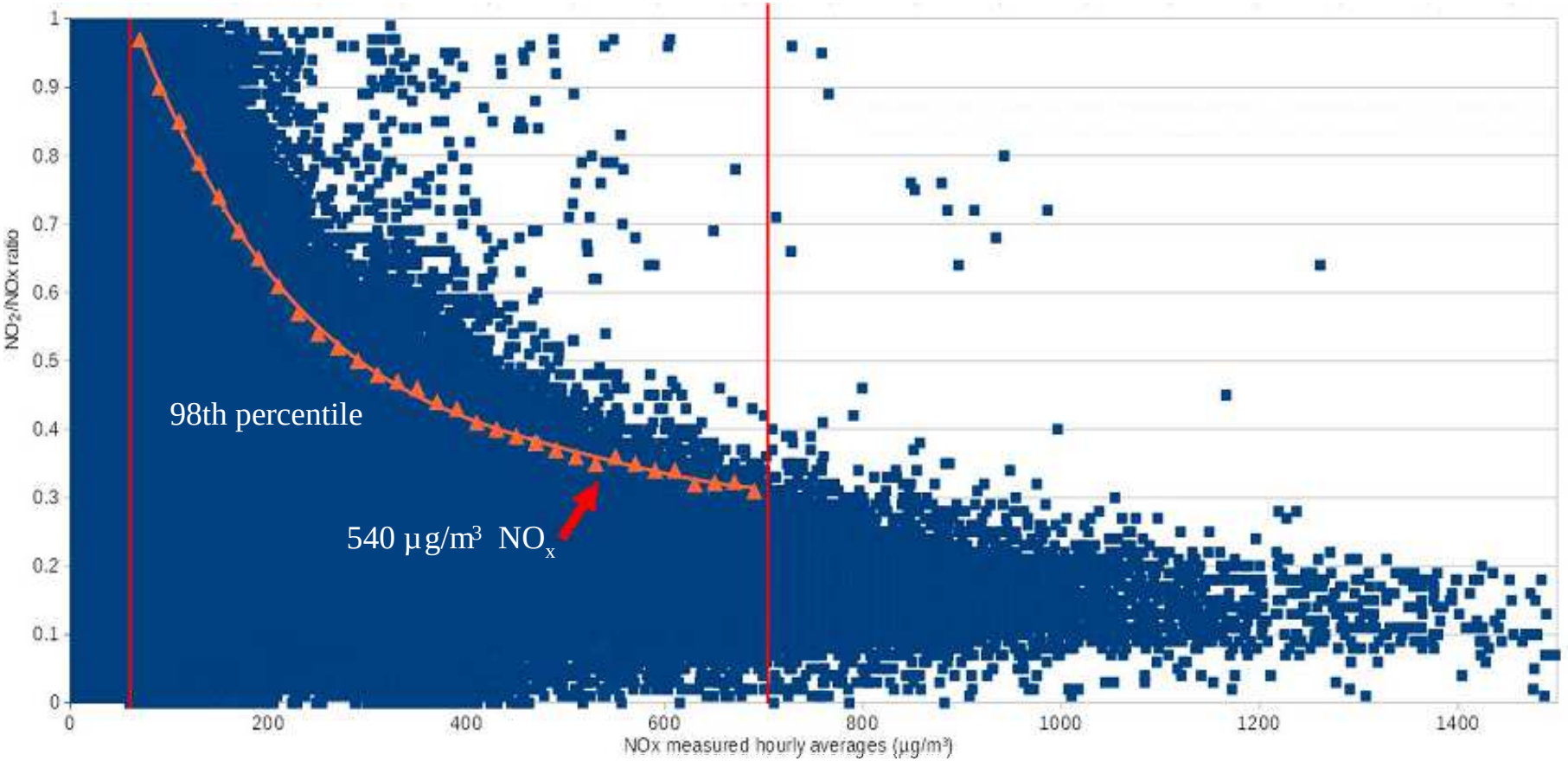
The final data-set contains more than 2.300.000 hourly averages in total.

Lower cut-off value

60 µg/m³ NO_x

Upper cut-off value

700 µg/m³ NO_x



Scatterplot of Tuscany NO_2/NO_x ambient ratios as a function of NO_x hourly concentration

The hourly NO_2/NO_x ambient ratios have been calculated by using the Tuscany Air Quality Monitoring Network database and subsequently sorted into NO_x (as NO_2) concentration ”bins”, 20 µg/m3 wide.

For each “bin”, the median of NO_x hourly concentrations, has been plotted as x-coordinate to the corresponding 98th percentile of the NO_2/NO_x ratios.

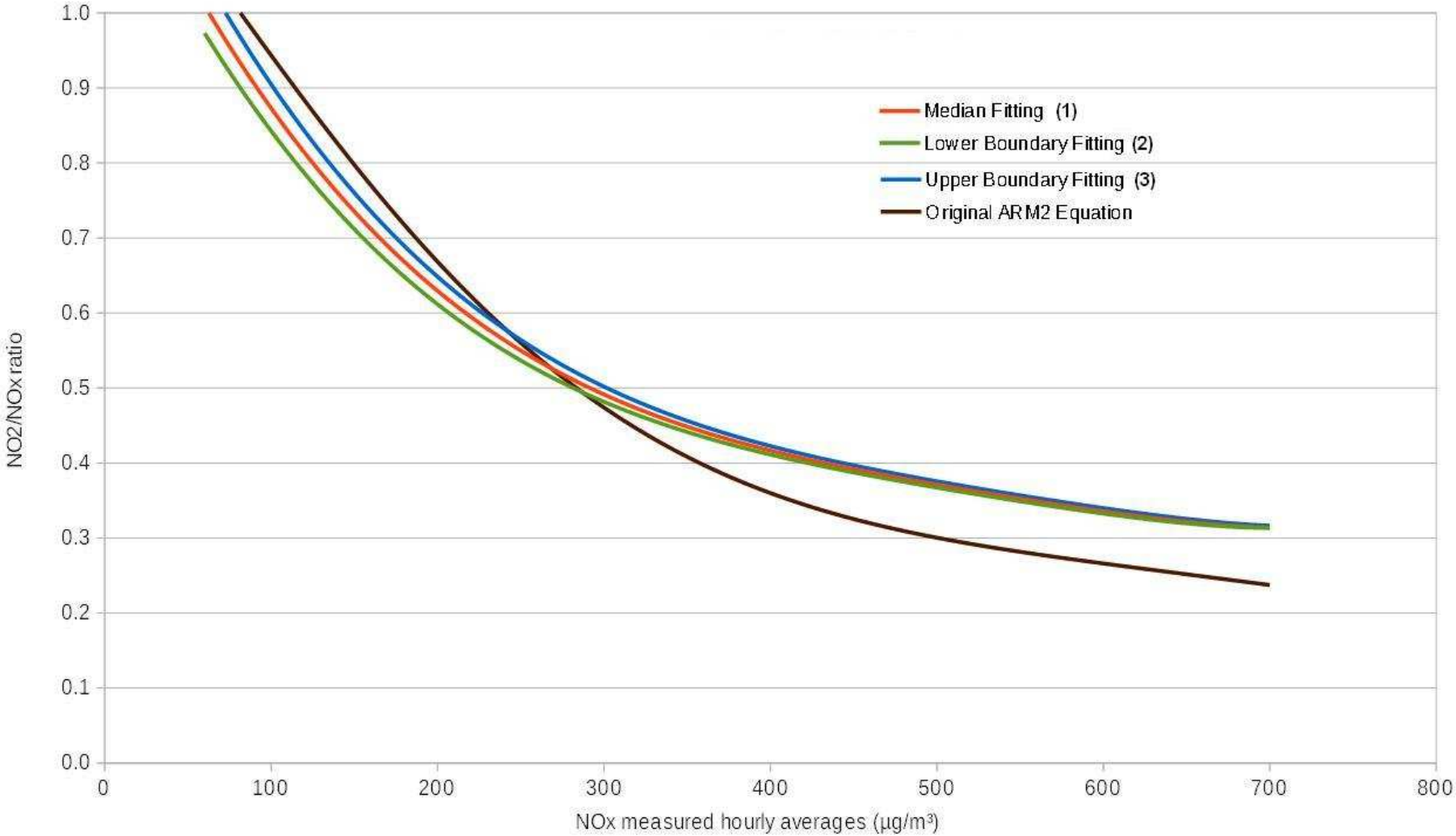
In order to assess the magnitude of the variability in the above relationship, the lower and upper boundary values of each ”bin” have also been used as x-coordinates for the corresponding 98th percentile of NO2/NOx ratios.

The following 5th grade polynomials are the functions that best fit the aforementioned plots, and can be used to represent the empirical relationship between the upper limits of the NO_2/NO_x ambient ratio (R) and the NO_x hourly concentration in µg/m^3 (x) in Tuscany:

$$\mathbf{Rmedian = 6.0635E\text{-}15x^5 - 5.8028E\text{-}12x^4 - 5.1576E\text{-}9x^3 + 9.2741E\text{-}6x^2 - 4.7886E\text{-}3x + 1.2647} \quad \mathbf{(1)}$$

$$\mathbf{Rlower = 6.0635E\text{-}15x^5 - 5.4996E\text{-}12x^4 - 5.3837E\text{-}9x^3 + 9.1159E\text{-}6x^2 - 4.6047E\text{-}3x + 1.2177} \quad \mathbf{(2)}$$

$$\mathbf{Rupper = 6.0635E\text{-}15x^5 - 6.1060E\text{-}12x^4 - 4.9194E\text{-}9x^3 + 9.4253E\text{-}6x^2 - 4.9756E\text{-}3x + 1.3135} \quad \mathbf{(3)}$$



plots of the polynomial equations relating NO_2/NOx ambient ratio and NOx hourly concentration: Tuscany data derived (green, blue, red lines) and ARM2 original equation (brown line)

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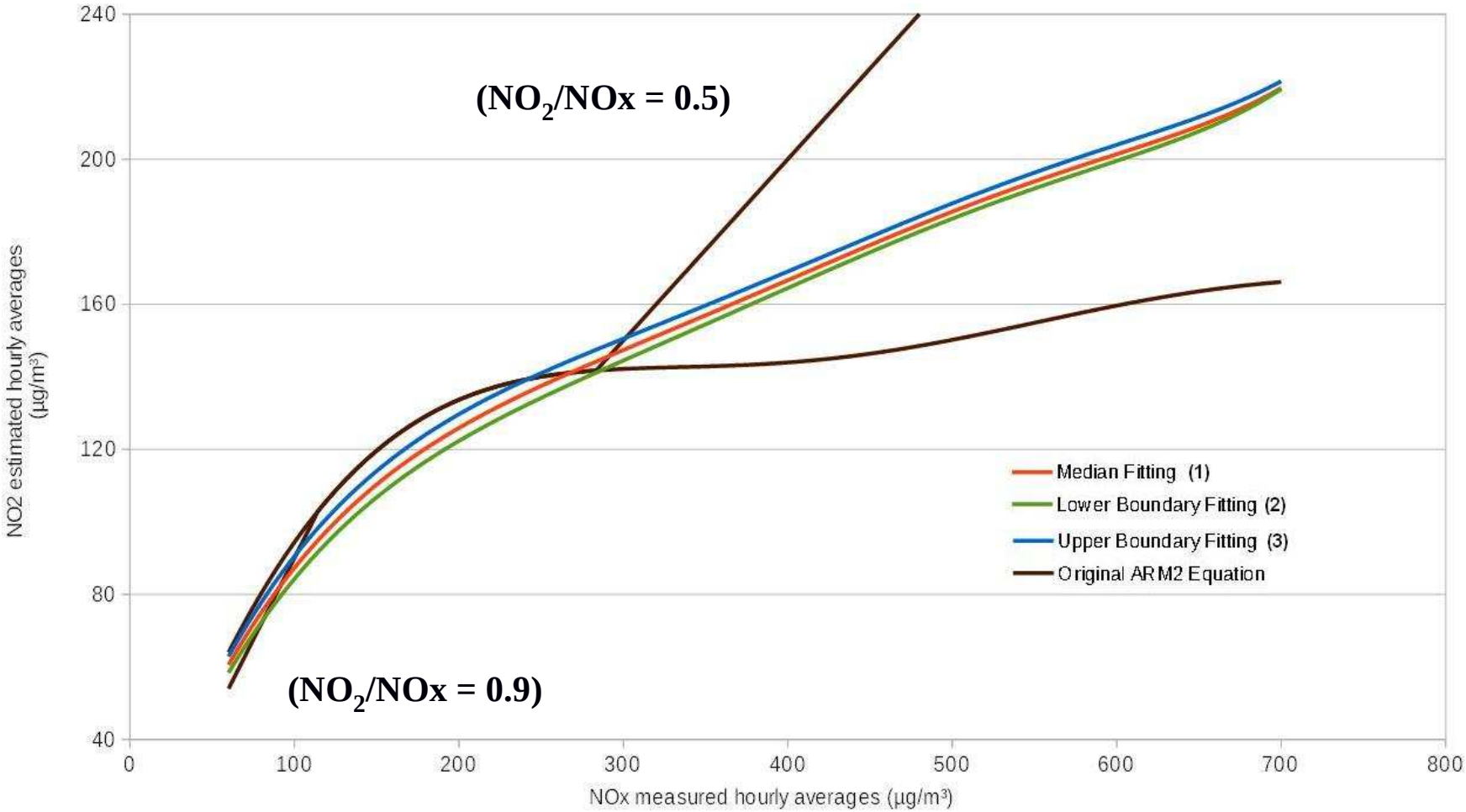

estimated NO_2 ambient concentrations as a function of the NO_x measured ambient concentrations using ARM2 original equation and the Tuscany ambient data adapted ones (equations (1), (2), (3))

Experience in Nitrogen oxides modeling and evaluation for regulatory purposes in Tuscany region suggests that:

1) a simple screening evaluation is usually enough to proceed when modeled concentrations of NO_x are lower than 100 µg/m^3, since they usually do not imply the risk of exceeding the European Air quality Directive 2008/50/EC limit values for Nitrogen dioxide ambient concentrations, when added to the typical background levels in Tuscany;

2) the availability of a good ”Tier 2” method is especially crucial to estimate NO_2 concentrations when the modeled NO_x hourly concentrations due to antropic sources are expected to be in the 100-300 µg/m^3 range without considering the background levels. Since they are obtained from locally measured data, the adapted equations are probably more coherent with the environmental conditions of Tuscany than the original ARM2 polynomial.

Experience in Nitrogen oxides modeling and evaluation for regulatory purposes in Tuscany region suggests that:

3) modeled NO_x hourly concentrations higher than 300 µg/m^3 are usually associated with NO_2 hourly concentrations higher than 150 µg/m^3. Such concentrations increase the likelihood of exceeding the air quality limits when added to background levels. If possible, it is advisable to further refine the ”Tier 2” evaluation, implementing measured NO_2/NO_x in-stack emission ratios of local sources into models that take into account NO_x chemistry in the atmosphere.

Finally, it has to be taken into account that the ARM2 method is probably less conservative when the to the emission source’s NO_2/NO_x in-stack ratios are very high (more than 0.5). In Tuscany this is an occurrence rare enough to warrant a case-by-case approach.

Thank You

for your attention