



## Supplement of

# Downscaling of air pollutants in Europe using uEMEP\_v6

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#### S1 NO<sub>2</sub> chemistry schemes

Included in uEMEP are a number of simplified NO<sub>2</sub> chemistry schemes, used to derive downscaled NO<sub>2</sub> concentrations from NO<sub>X</sub> and O<sub>3</sub> concentrations. The results presented in this paper have used the weighted travel time parcel method, as applied and described in Denby et al. (2020), together with the frequency distribution correction scheme (Section 2.4). In addition to

- 5 this parameterisation two other chemistry based formulas are available. The first being the photo-stationary formulation, also described in Denby et al. (2020), and the second a stationary formulation that allows for deviation from the photo-stationary state that may be caused by emissions, advection gradients or additional chemistry (Maiheu et al., 2017). Two empirical formulations are also included that are based on a fit to measurement data. The first, the Romberg scheme (Romberg et al., 1996), is already described in Denby et al. (2020) and directly converts NO<sub>X</sub> to NO<sub>2</sub> concentrations. The parameters for this
- 10 equation have been updated by fitting to all available Airbase data for the year 2017. The second empirical scheme, the SRM scheme (Wesseling and van Velze, 2014), includes background  $O_3$  and  $NO_2$  combined with local  $NO_X$  as input parameters. The advantage of the two empirical fits is that they should convert  $NO_X$  to  $NO_2$  in a manner that is consistent with the observations, and as such can be applied to annual mean concentrations without correcting for non-linear chemistry. The first method is already described in (Denby et al., 2020), the remaining methods are presented in the following sections.

#### 15 S1.1 Stationary scheme based on EMEP equilibrium

The solution to the photo-stationary equilibrium of NO<sub>2</sub> that only considers the NO<sub>X</sub> titration of O<sub>3</sub> (reaction rate  $k_1$ ) and the photo-disassociation of NO<sub>2</sub> (photo-disassociation rate J) is already presented in Section 2.4. This steady state solution implies

$$\frac{k_1[NO][O_3]}{J[NO_2]} = 1$$
(S1)

For the case where there is equilibrium but other terms such as emissions, deposition or additional chemistry are involved then we can define a constant  $\lambda$  such that

$$\frac{k_1[NO][O_3]}{J[NO_2]} = \lambda \tag{S2}$$

25 For  $\lambda < 1$  this implies a sink of NO<sub>2</sub> and for  $\lambda > 1$  a source. The steady state solution can then be written, as in Eq. (1), as

$$[NO_2] = \frac{1}{2} \left( ([NO_X] + [O_X] + \lambda J/k_1) - \sqrt{([NO_X] + [O_X] + \lambda J/k_1)^2 - 4[NO_X][O_X]} \right)$$
(S3)

By simply extracting the parameters listed in Eq. (S1) from the EMEP model we can determine λ and by using Eq. (S3) we can calculate the downscaled NO<sub>2</sub> concentrations. At the station sites used in this study we find on average λ = 0.95 with a standard
deviation of 0.35. Around 60% of the sites have λ < 1. This means there is significant divergence from the photo-stationary assumption in the EMEP annual mean calculations. λ is also strongly correlated with NO<sub>2</sub>, increasing with increasing NO<sub>2</sub>, indicating that emissions are perhaps the dominating factor leading to non photo-stationary equilibrium. It should be noted that the term [NO<sub>X</sub>] + [O<sub>X</sub>] in Eq. (S3) is usually significantly larger than J/k<sub>1</sub> so even with a significant deviation of λ from unity then the NO<sub>2</sub> concentrations will not be largely affected.

This parameterisation is particularly useful if NO<sub>2</sub> concentrations from EMEP are to be recalculated, for example when using the local fraction to assess the impact on concentrations with changes in emissions in a post-processing step. It is not necessarily the case that the conditions leading to a deviation of  $\lambda$  from unity are equally applicable for the downscaling chemistry.

#### S1.2 Updated Romberg scheme

40 The Romberg scheme (Romberg et al., 1996) is the simplest method for converting  $NO_X$  to  $NO_2$ . It is based on an empirical fit to the formula

$$[NO_2] = a \frac{[NO_X]}{[NO_X] + b} + c[NO_X]$$
(S4)

The parameters *a*, *b* ( $\mu g/m^3$ ) and *c* are derived by fitting to observed annual mean concentrations. The term *c* should in some way reflect the ratio of NO<sub>2</sub>/NO<sub>X</sub> emissions, i.e. the asymptotic limit for large NO<sub>X</sub>. Values for these parameters were given in Denby et al. (2020), based on a fit to Norwegian measurement data. For the European application a new fit is made to annual mean concentrations from all available European observations for 2017. This is shown in Fig. S1. The fitted parameters are: a = 41.1,  $b = 56.4 \ \mu g/m^3$  and c = 0.162. The root mean square error of this fit is 2.9  $\mu g/m^3$ , or a normalised error of around 14%.

When implemented in uEMEP a small adjustment is made to Eq. (S4). It is desirable that background NO<sub>2</sub> levels provided by EMEP are not altered by this calculation. Background NO<sub>2</sub> and O<sub>3</sub> concentrations are calculated in uEMEP after the removal of the local fraction contribution from NO<sub>X</sub>. In Denby et al. (2020) this is referred to as the non-local contribution. To achieve this we rewrite Eq. (S4) as follows

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$$[NO_2] = a \frac{[NO_X]}{[NO_X] + b} + c[NO_X] + \Delta [NO_2]_{bg}$$
 (S5)

where  $\Delta [NO_2]_{bg}$  is the difference between the EMEP calculated NO<sub>2</sub> non-local contribution and the Romberg calculation of the non-local contribution given by

$$\Delta[NO_2]_{bg} = [NO_2]_{nonlocal} - a \frac{[NO_X]_{nonlocal}}{[NO_X]_{nonlocal} + b} + c[NO_X]_{nonlocal}$$
(S6)

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This ensures that the calculated  $NO_2$  concentrations are unchanged when the local contribution to  $NO_X$  is negligible.

#### S1.3 SRM scheme

The SRM scheme for NO<sub>2</sub> calculation is implemented in The Netherlands as part of the Standard Calculation Method for air quality (Wesseling and van Velze, 2014). This scheme requires information on background levels of  $O_3$  and  $NO_2$ . Whilst this information is directly available from the model calculations this is more difficult to extract from measurement data. For

this reason no fitting or changes to the parameters given in Wesseling and van Velze (2014) have been made. The equation is slightly re-written to be comparable with the Romberg scheme and the uEMEP formulation as:

$$[NO_2] = [NO_2]_{bg} + [O_3]_{nonlocal} \frac{[NO_X]_{local}}{[NO_X]_{local} + b_{srm}} + c_{srm} [NO_X]_{local}$$
(S7)

#### 70 where

$$b_{srm} = \frac{100}{1 - c_{srm}}$$
 and  $c_{srm} = 0.15$ 

The value for  $c_{srm}$  applied here is the traffic exhaust NO<sub>2</sub>/NO<sub>X</sub> emission ratio used in uEMEP, and the value for  $b_{srm}$  ( $\mu g/m^3$ )



Figure S1. Annual mean  $NO_2$  versus  $NO_X$  concentrations for 2017 showing 1774 available Airbase stations. The fit using the Romberg equation is also shown, using the parameters provided in the text.

comes directly from Wesseling and van Velze (2014). The parameters  $c_{srm}$  and  $b_{srm}$  are referred to as F and K respectively in 75 Wesseling and van Velze (2014).

This equation clearly resembles the Romberg scheme, Eq. (S4), with the exception that a background NO<sub>2</sub> and a local NO<sub>X</sub> are used. Here the background ozone would then be equivalent to the parameter  $a_{srm} = [O_3]_{nonlocal}$ .

#### S1.4 Additional information on the frequency distribution correction scheme

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A frequency distribution correction scheme for annual mean calculations of NO<sub>2</sub> is described in Section 2.4. This is imple-80 mented to deal with the non-linear nature of the NO<sub>2</sub> chemistry when calculating annual means. Here we present some of the background information used to derive this scheme.

An example of the NO<sub>X</sub>, O<sub>X</sub> and J frequency distributions is shown in Fig. S2. Here we see the log-normal distribution of the concentrations and the quite different distribution of the photo-dissociation rate J. J has a frequency of close to 0.5 at J = 0, since this, over the course of a year, is the amount of time the sun is below the horizon. The example shown here is taken from a low latitude European urban background station, with moderate NO<sub>X</sub> and high O<sub>3</sub>.

An assessment of measurement and model data for 2018 was carried out for Norwegian stations in order to derive the standard deviations of the  $NO_X$  and  $O_X$  concentrations. This assessment was limited to available data and a more substantial assessment could be carried out on a larger set of data. Focus is on the modelled distributions since this is what needs to be reproduced in order to correct the annual mean calculation. Firstly the concentration distributions were tested for a log-normal

90 distribution. An example for both  $NO_X$  and  $O_X$ , both modelled and observed is shown for the urban background station Klosterhaugen in Bergen, Norway, in Fig. S3 and Fig. S4. This is one of the few stations measuring both  $NO_X$  and  $O_3$  in Norway. Further assessment of the log-normal distribution was carried out for all modelled data at the 72 sites modelled in Norway for that year. This assessment showed that the concentrations were very close to log-normally distributed at all sites.

The standard deviations were then derived and compared to mean values. The results for both  $NO_X$  and  $O_X$  at modelled stations are shown in Fig. S5. For  $NO_X$  the observed values are also included since these are more readily available. The



Figure S2. Example of the frequency distribution for  $NO_X$ ,  $O_X$  and J concentrations used in the model. This is taken from the calculation of an actual site in Southern Europe. X axis is logarithmic.



Figure S3. Measured and modelled  $NO_X$  distributions for year 2018 at the station Klosterhaugen, Bergen, shown on both a linear and a logarithmic scale.

relationship for NO<sub>X</sub> is very robust and will reflect the temporal variation of the NO<sub>X</sub> sources, mostly traffic, and of the meteorology. It is worth noting that the normalised standard deviation of the traffic time profile applied in the Norwegian calculations is 0.71. Additional variability will be added due to meteorology. For  $O_X$  the standard deviation is less dependent on the mean concentration. Even so, the normalised standard deviation of  $O_X$  is significantly smaller than that of NO<sub>X</sub>.



Figure S4. Measured and modelled  $O_X$  distributions for year 2018 at the station Klosterhaugen, Bergen, shown on both a linear and a logarithmic scale.



Figure S5. Scatter plot of hourly standard deviation versus annual mean  $NO_X$  and  $O_X$  for 2018 for all the 72 modelled sites. Also included are measurement data from 41 of the sites that measured  $NO_X$  with data coverage >75%. The regression slope, intercept set to 0, and the correlation are also shown in the plots.

- 100 Finally the correlation between NO<sub>X</sub> and  $O_X$  was addressed since the frequency distribution correction assumes these two concentrations are not correlated. This assessment showed both negative and positive correlations at the modelling sites. Sites with high  $NO_X$  concentrations showed positive correlation and background sites showed mostly negative correlation. Since  $NO_2$  is part of both  $NO_X$  and  $O_X$  then this positive correlation is not surprising. Values for the correlation (r) ranged from -0.4 to +0.6. This result shows there is correlation between NO<sub>X</sub> and O<sub>X</sub> but this would be difficult to take account of in the frequency distribution correction currently implemented in the model. 105



Figure S6. Scatter plots of annual mean  $NO_2$  concentrations per country for 2018 calculated with uEMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S7. Scatter plots of annual mean  $NO_2$  concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S8. Scatter plots of annual mean  $PM_{2.5}$  concentrations per country for 2018 calculated with uEMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S9. Scatter plots of annual mean  $PM_{2.5}$  concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S10. Scatter plots of annual mean  $PM_{10}$  concentrations per country for 2018 calculated with uEMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S11. Scatter plots of annual mean  $PM_{10}$  concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S12. Scatter plots of annual mean  $O_3$  concentrations per country for 2018 calculated with uEMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.



Figure S13. Scatter plots of annual mean  $O_3$  concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.

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