Author response to Referee Comments and Editor Comment

Many thanks to Dr. David Topping and the anonymous referees for the constructive comments. The manuscript is definitely improved. Below is our response to the RC1, EC1, and RC2, respectively. The original comment is in black, while our response is in blue. The added/modified text is shown in italic. The reference list is at the end of the document. Note that we've added one new figure as Fig. 1, so the figure number in the revised manuscript equals to the figure number in this response plus 1.

(RC1)

General comments

This preprint presents high-resolution air quality modelling at local scale/street level across Europe, performed with the uEMEP_v6 model, which downscales emission input and combines regional calculations with the EMEP model with Gaussian plume modelling of receptor points to obtain annual mean concentrations of NO_2, $PM_{2.5}$, PM_{10} and O_3 in a very high resolution subgrid (down to 100 m resolution). Results presented are comparisons of EMEP and uEMEP model results country by country for Airbase monitoring stations across Europe, as well as sensitivity studies with respect to resolution, weighting of relative traffic distribution, proxies for residential combustion emissions, use of national emission and proxy data with higher detail than the EMEP data, and NO_2 chemistry schemes for the NO_2-NO_x-O_3 reactions. Model results are seen to significantly improve in comparison with measurements for NO_2 and O_3 when the uEMEP model is applied whereas little improvement for $PM_{2.5}$ and PM_{10} is gained using the downscaling approach.

The manuscript addresses the highly relevant scientific question concerning how to obtain high resolution air quality estimates at the urban scale in an operational way, using publicly available data. The different parts of the methodology are known, but the combination is novel and the uEMEP model tool is potentially extremely useful for air quality and population exposure studies across Europe. The manuscript is well-written and the structure and argumentation of the study

is easy to follow with a few minor details that could be clarified (see Specific comments). All model code for the uEMEP model and the visualization tools is publicly available for this study.

Specific comments:

When investigating annual mean values, a rotation symmetric approach for the Gaussian dispersion is used. This implies the assumption that the wind is equally distributed from all directions over a year. In reality these wind distributions will probably be quite different. Could the authors elaborate on if this rotation symmetric approach is more or less accurate in different locations? And what potential comparisons of the approach to applying actual annual met-data would show?

The assumption is usually not met in reality, but the result of this simplified method agrees very well with the hourly calculation. In Sect. S5.1 of Denby et al. (2020), we compared the annual and hourly calculations for the same 100 x 100 km^2 region surrounding Oslo with a resolution of 100 m. Figure S2 and S3 show very similar results for the two model calculations. We have not assessed the error on a European wide basis and so cannot quantify this. From the tests carried out in Norway there is no indication that this approximation leads to errors larger than other uncertainties in the modelling.

When the OSM data are applied across Europe, a weighting based on the Norwegian traffic data is used – can the authors elaborate on what this means for the distribution of local emissions in the rest of Europe?

In Sect. 5.2, we tested other weighting scenarios by changing the power law index which is applied to the initial weights. Results show that the spatial correlation is highest for the initial weights based on Norwegian traffic, and bias is also reasonable. It indicates that weightings derived from Norwegian traffic can reflect a general distribution of traffic in Europe. Moreover, the results also indicate that higher weightings for the minor roads can improve the bias, as the urban background level is increased. This provides insights for the rest of Europe where local traffic data is available.

The NO-NO$_2$-O$_3$ chemistry for annual mean values is based on a calculation including a frequency distribution of the concentrations of NO, NO$_2$ and O$_3$. It is a little unclear from the manuscript, if these frequency distributions are acquired from Norwegian stations only, or if all available measurements from Europe have been taken into account? The dependency on solar input in the photochemical reactions must mean that the frequency distribution will differ across Europe?

The frequency distributions of NO$_X$ and O$_X$ are log-normally distributed. The standard deviation and mean values are derived from earlier modeling results at 72 Norwegian stations. Since the variability of NO$_X$ reflects the variability of the traffic emissions, this should be generally applicable throughout Europe for stations within the influence of traffic. The magnitude of the O$_X$ variability will likely be different across Europe, depending on O$_3$ levels but this distribution has less impact on the results than NO$_X$. Ideally this analysis should have been done for all European stations but this requires a sizable effort to assess hourly data from paired stations.

As Eq. (3) shows, the distribution of J follows the distribution of the solar zenith angle (ZA). This distribution is based on the latitude and longitude of each European grid point so is valid everywhere.

The results of the uEMEP calculations correspond to street-level, but the building configuration is not included, and common situations with development of street-canyon circulation vortices are therefore also not taken into account. Can the authors elaborate on what this means for the results at street-level in the large cities with tall and dense building mass?

There will likely be an underestimate at the street-canyon sites, since the Gaussian model does not consider obstacles and does not include any form of street canyon parameterisation. In Europe there are a large number of traffic sites in fairly open environments that do not require street canyon modelling. Exactly how many traffic sites are not known. Reporting to the EU Commission (e-reporting) on siting of traffic stations is generally quite incomplete. Metadata provided in e-reporting was analysed for the EU commission in 2018 (Tarrasón et al., 2021). This indicated that the majority of stations are not street canyon sites, though the exact number was unclear.

In a study by Lefebvre et al. (2013) modelling of NO_2 was carried out in Antwerp using both Gaussian and street canyon modelling for 49 stations, a mix of both open road and street canyon sites. On average over all sites the increase in concentration was around 4% with the use of the street canyon model. For individual street canyon sites (15 sites) this increase was 11%. If Antwerp can be interpreted as a representative European city then we can expect a similar underestimation for all sites in Europe.

The uEMEP model is intended for applications over country scales, and the level of detail required for street canyon modelling is not easily achievable. Without including obstacles, the increased model resolution (up to 25 m) allows the concentration gradients at roadside to be better described.

At the end of the first paragraph of Sect. 4, we've added: "...traffic sites in street canyons or built up areas may be *underestimated. A study by Lefebvre et al. (2013) in Antwerp, where both Gaussian and street canyon models were applied at 15 street canyon modelling sites, showed an average street canyon modelling increment of just 11% for NO_2. We include all available sites in this study because in Europe the majority of traffic sites appear not to be street canyons, though information on this is unclear (Tarrasón et al., 2021). Also, even without including obstacles, the increased model resolution (up to 25 m) allows the concentration gradients at roadside to be better described.*"

Regarding PM2.5: All annual mean concentrations for (NO2 and) PM2.5 increases when the uEMEP downscaling is applied compared to the EMEP model results. In two countries (Austria and Finland), the EMEP model is already overestimating the PM2.5 concentration, and applying the uEMEP model only increases the overestimation. Wouldn't the authors expect that downscaling using proxies would give a more precise result for the distribution, and thereby a more accurate replica of what is observed? Or is there an underlying risk, that uEMEP increases the concentrations in general?

The downscaling proxies only redistribute the original EMEP emissions, so that the total emissions in EMEP and uEMEP are the same. Therefore, uEMEP does not always increase the concentrations in general, but can resolve more variability, i.e. higher concentrations in heavy

traffic and populated areas, and lower concentrations vice versa, as shown in the example maps Fig. 3-5. The concentrations at validation stations are in general higher for uEMEP, because emissions around those stations are higher after redistribution, and validation results show that uEMEP has done a better job than EMEP in most of the countries and EU as a whole. However, as the reviewer points out, uEMEP further overestimates $PM_{2.5}$ concentrations in two countries (Austria and Finland). This could be due to the uncertainty of the proxies in those countries or that the EMEP emissions themselves are overestimated in these countries. Recent analysis of the CAMS residential combustion emissions by TNO themselves have indicated, personal communication, that these emissions are indeed overestimated in Austria. We discuss more about the uncertainty of residential emission proxy in Sect. 5.3.

Figure 10: is this result for the whole of Europe, i.e. a mean of all countries?

The result is for the whole of Europe, which is averaged over all Airbase stations with >75% coverage. We've added an explanation into the caption of Fig. 10: "*The results are based on the European calculations and all available Airbase stations are included.*"

Figure 11 and 12: It would be good with a little more explanation in the Figure captions, e.g. a note whether this is all of Europe, or only Norway.

In the caption of Fig. 11, we've added that "The scenarios are tested in the European calculations." In the caption of Fig. 12, we've added that "*The results are based on the European calculations and all available Airbase stations are included.*"

Figure 12: the conclusion that the correlation is clearly highest for power law index 1 is putting much trust in the decimals of the correlations. As the numbers are 0.567 (~0.57), 0.574 (~0.57) and 0.557 (~0.56), one could wonder how much the third decimal of the correlation estimate is worth in terms of accuracy?

The reviewer is right that the correlations don't differ much, but we can still say the power law index 1 can reflect a good (though not clearly the best) general distribution of traffic in Europe. We rephrase the sentence like this: "The spatial correlation is *among* the highest for the current weighting with a power index of 1. This confirms that the initial estimate, based on Norwegian traffic, reflects a *good* general distribution of traffic in Europe."

In the discussion: "Though the problem remains that uEMEP does not take into account dispersion in street canyons, where a number of traffic site measurements are made, it is generally the case that the spatial representativeness of the uEMEP calculations is suitable for comparison with these measurements." How do the authors know that this is the case?

In a study by Lefebvre et al. (2013) modelling of NO_2 was carried out in Antwerp using both Gaussian and street canyon modelling for 49 stations, a mix of both open road and street canyon sites. On average over all sites the increase in concentration was around 4% with the use of the street canyon model. For individual street canyon sites (15 sites) this increase was 11%. If Antwerp can be interpreted as a representative European city then we can expect a similar underestimation for all sites in Europe. We've added this reference to the text "... suitable for comparison with these measurements *(Lefebvre et al, 2013)*."

Technical comments:

Figure 14: the order of the components is NO_2, $PM_{2.5}$ PM_{10}, but in the text the order of discussion is NO_2, PM_{10} and $PM_{2.5}$. Would be easier to read if the order is the same in both places.

Thanks for pointing it out. We've changed the order of the text to go along with the order of the figure, i.e., NO_2, $PM_{2.5}$, PM_{10}.

Line 315: all five methods are described in suppl,. but in the figure, there are results for 6 methods? Not easy to follow the names of the 6 methods in the text in the manuscript as they are not consistently defined (nor in the supplementary material).

There are 6 methods mentioned in the Supplement as well, 5 described in detail and 1 cited from another paper. To avoid confusion, we've changed the text into: "*All methods are described in Sect. S1.*" Besides, we've added corresponding method names in the figure into the text when introduced the first time.

Line 319: "Since the Romberg scheme is specifically designed to reflect measurements, providing the correct NO_2/NO_x ratio, this means that the chemistry schemes are overestimating the NO_2 contribution when applied to annual mean concentrations." It is somewhat difficult to understand what is meant here?

There is indeed a logic jump which is unclear. We've rephrased it like this: "Since the Romberg scheme is specifically designed to reflect measurements, providing the correct NO_2/NO_x ratio, *it can be regarded as the closest to the measurements. The bias differences between chemistry schemes and the Romberg scheme indicate that chemistry schemes have higher concentrations of NO_2 than the Romberg scheme, thus* overestimate the NO_2 contribution when applied to annual mean concentrations."

Table 2: within an region – should be: *within* a region

We've corrected "within an region" into "within a region".

Line 374: verses – should be *versus*

We've corrected "verses" into "versus".

Figure S5: verses – *versus* + include should be – *included*

We've corrected accordingly. We've also corrected the same typo in Figure S1.

(EC1)

This paper presents work around downscaling of the EMEP model. Such tools are extremely valuable for personal exposure estimates. The authors present a concise narrative of the background of the work conducted and discuss wider needs. I recommend the paper is published after the following discussion points are acted on.

Section 1, page 2 line 31 'where access to good quality emission data is available'. Can you please add more details and/or a reference? This is obviously important to understand. How is the quality and, presumably, density better in Norway? I can see this is covered in section 5.4 so please summarise and reference that section. You comment on accessibility in the proceeding sentence so is this an issue of data access?

We've added references to explain how the quality is: "where access to good quality emission related data is available. *The quality of the Norwegian emissions is summarized in Sect. 5.4 and details can be found in Sect. S4.2 of Denby et al. (2020)*".

Obtaining high resolution, 250 m, emission data from individual countries is a process in itself and requires significant time and resources. This is why we have used openly available emission proxy data that cover all of Europe and redistributed existing EMEP gridded emissions. This also allows for consistency between EMEP and uEMEP calculations.

Lines 35-40. I wonder how you could add an estimation of relative error on the downscaling predictions. Have you looked at mapping error as a function of land-use? I ask because another way to arrive at annual means is through land use regression. Can you clarify the differences?

We are unsure about what the reviewer actually wanted to see, but estimation of relative error on the downscaling predictions is validated against observations in Sect. 4. We don't know what "mapping error as a function of land-use" is. As for the land use regression (LUR) models, We haven't tested it, but we are aware of relevant studies, e.g. a European-wide LUR model study from Vizcainoand and Lavalle (2018). Though LUR is

commonly used to project the spatial concentration of atmospheric pollutants, it's a statistical method based on regression analysis. The downscaling model used in this manuscript is an atmospheric Gaussian dispersion model, which is based on physics and chemistry formulations.

Section 2 Methodology.

- Why focus on annual means and not seasonal differences? Presumably across most of Europe this changes significantly?

High-resolution simulation of uEMEP at such a large area is quite computing heavy, though parallelled in tiles. The manuscript is intended as a method description primarily, so we demonstrate the method focusing on annual means. More scientific analysis, e.g. seasonal differences across Europe, is beyond the scope of this current manuscript.

- Please can you clarify 'EMEP local fractions' – I think some of this information is provided in section 2.2 so you may wish to consider re-ordering some of the information.

Following your suggestion in the next question, we've added a new schematic figure in Sect. 2 to illustrate all steps (Fig. 1 in the revised manuscript). At the end of Sect. 2.1, we've added a new paragraph to explain 'EMEP local fractions': "*The EMEP model calculates and outputs the 'local fraction', used by the uEMEP downscaling to remove double counting of emissions (Denby et al., 2020; Wind et al., 2020). The local fraction is the contribution of emissions in one EMEP grid to itself and to its neighbouring grids. For this application only a 3 x 3 grid contribution region is calculated, though for other applications this can be much larger. By tagging the grid emissions in this way the local contribution from EMEP can be removed and replaced by the high-resolution uEMEP sub-grid calculation.*" We've also added explanations accordingly in several places of Sect. 2.2.

- may I suggest a schematic illustrating all of the steps might help the reader?

Thanks for the suggestion. We've added a new schematic figure in Sect. 2 to illustrate all of the steps.
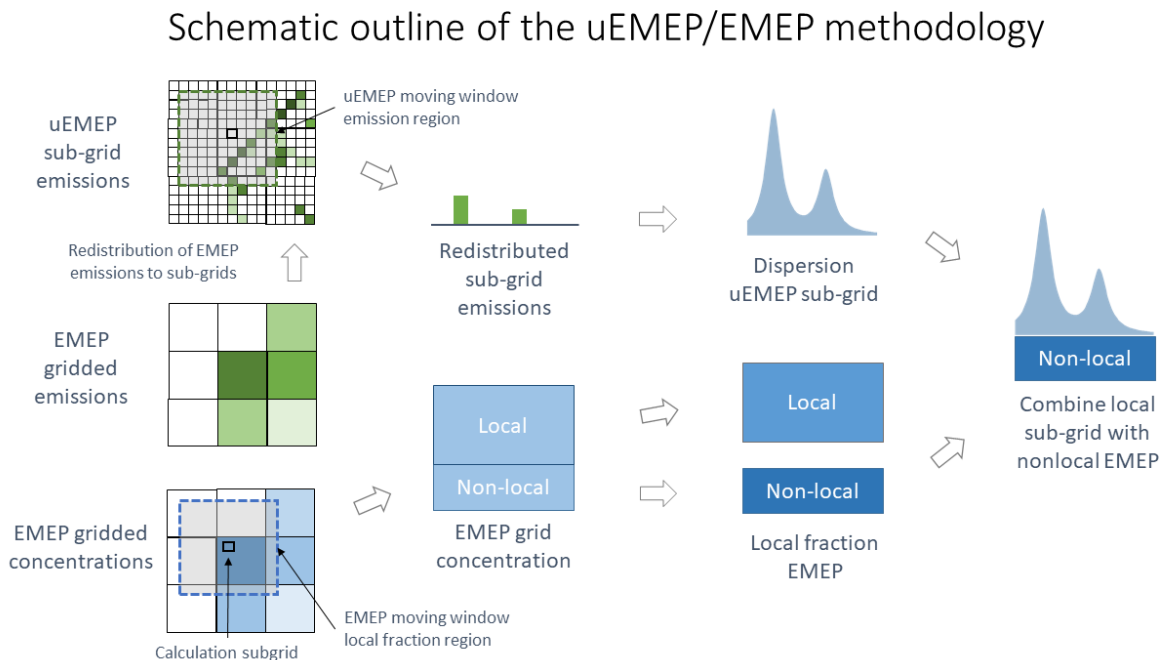

Figure 1. Schematic illustration of the EMEP/uEMEP coupled modelling system.

Line 49, page 2 – '2 x 2 EMEP grids' to clarify, is this independent of the resolution of the EMEP simulations?

It's independent of the resolution of the EMEP simulations.

Table 1, page 3 – can you please clarify, are these parameters for all primary pollutants? If so, why? Please also clarify this with respect to the weight factors displayed in figure 2. It might be useful to reference section 5 at this point.

These parameters in Table 1, as well as the road weightings in Fig. 2 are for all primary pollutants. This is because the initial dispersion, emission height, and road weighting parameters are very much source specific, not species specific. We've added in the caption of Table 1: "...*for all primary pollutants.*" and caption of Fig. 2: "*Applied to all primary pollutants.*"

Section 5.1, line 216 'when calculating at station positions..'- calculating concentrations?

Yes thanks, we've rephrased it into "When calculating *concentrations* at station positions...".

Figure 10 please use black font for all axes, I found the text a little hard to read.

We've used black font for all axes and increased the font size in Fig. 10. It looks clearer now.

Page 21, line 340 – 'the largest contributor to PM is residential heating'. Please clarify whether you are referring to model outcomes or compiled data across Europe. I would like to see more references to support wider known trends in measured PM properties.

The reviewer is correct to ask for a clarification. In fact we mean 'the largest contributor to PM in the downscaled sources is residential heating'. It was not intended as a general overall statement for all concentrations of PM. We haven't analysed all sources and all precursor species to be able to make a broad statement like this. As for trends in measured PM properties, we've added a reference "Screening for High Emission Reduction Potentials for Air quality (SHERPA)" $PM_{2.5}$ urban atlas, as this provides an assessment of urban background source contributions for 150 European cities. Here they find an average contribution of 13% from primary PM from residential combustion (p. 13).

We've corrected this in the text around line 340: "The largest contributor to PM *in the downscaled sources* is residential heating*, with contributions 19% and 16% for $PM_{2.5}$ and*

PM$_{10}$, respectively (Table 2). This is inline with other estimates of residential combustion in Europe. Thunis et al. (2017) calculated a contribution of 13% from residential combustion from primary PM$_{2.5}$ averaged over 150 European cities, without downscaling. Population is used as a downscaling proxy for the residential source, but...".

Code and data availability – please can you also add links to the EMEP and IFS version used, with relevant configuration files.

We've added the link to the EMEP model version and the configuration file used in this study. We run the IFS model version Cycle 40r1 (ECMWF-IFS cy40r1) to generate the meteorological files, as input for the EMEP model. As far as we know, the IFS model is not open-source, so that users need to request a licence from ECMWF. Therefore we can't provide a link to the IFS model used with relevant configuration file. However, all input files can be shared upon request, including the meteorological input files that are generated by the IFS model.

In "Code and data availability", we've added the following text: "*The EMEP model version rv4.36 used in this study can be found at https://github.com/metno/emep-ctm. The configuration file of the EMEP model is archived at https://doi.org/10.5281/zenodo.5648144. Other model input files can be shared upon request, including the meteorological input files that are generated by the IFS model version Cycle 40r1 (ECMWF-IFS cy40r1)*." The EMEP model version has also been corrected from rv4.35 to rv4.36 in Sect. 2.1.

Title – please change uEMEP_v6 to uEMEP v6. I would also consider adding the year, since there will be variation during periods of large scale change?

The uEMEP description paper (Denby et al., 2020) named the model as "uEMEP_v5", so "uEMEP_v6" just keeps the consistency. When we submitted that article to GMD the editor requested this model naming. We can change to "uEMEP v6", if very necessary, but we think this is a question for the editor and the journal's requirements. So far, the uEMEP model only

uses numbers to denote versions after large scale changes. The exact code version used for these calculations has been given a doi, as requested by the journal.

(RC2)

This publication presents a downscaled version of the EMEP model and its application to Europe for air quality pollutants. The topic is very interesting and certainly relevant with respect to the scope of the journal. The methodological approach is sound and the work is well structured and well presented. I list below a few comments.

Comments:

1. The introduction dives directly into the proposed uEMEP downscaling. I would suggest starting with a paragraph to provide some context and explain why we actually need such a downscaling.

   We've now added why we need such downscaling following on from L28: "...so that we can have air quality modelling at street-level all over Europe. *Modelling at high resolution provides a better assessment of air quality mitigation strategies in Europe, as well as improved population exposure estimates for use in health impact studies.*"

   While the Authors state (L107) that "uEMEP downscales only primary pollutants", downscaled results for O3 are discussed and shown. A few lines to explain how secondary pollutants like O3 can be downscaled would clarify a possible confusion.

   How this is done is explained in the rest of section 2.4 but we have added a little bit more to the sentence on L107 that may help: "uEMEP downscales only primary pollutants. It is thus necessary to apply chemistry parameterisations to the NO_X and *EMEP* O_3 concentrations to derive *downscaled* NO_2 *and* O_3 *concentrations*."

2. Orography variations can be important over two 0.1deg grid cells. Can the authors comment on the applicability of their Gaussian approximation to hilly cities?

   Gaussian approximations have of course their limitations and the quality of the results will reflect, amongst other things, the complexity of the terrain. In essence we use winds from 0.1° meteorological fields so the accuracy of these in complex terrain can always be questioned. We have not assessed this, nor have we assessed the impact of buildings that are also not included in the modelling. We cannot provide a quantitative answer to the reviewers question.

3. A few remarks regarding Table 2:

1. Caption: an region à a region

   Corrected.

2. Please add that the region of +/- 0.1 deg also represents 2x2 grid cells.

   We've modified it into "... within a region of 2 x 2 EMEP grids (± 0.1°) …"

3. If my understanding is correct, the source column should precise that only primary contributions are considered (e.g. primary traffic…) and that non-local EMEP includes all secondary contributions as well.

   We've made it clear in the text that "uEMEP local contributions are from *primary* emissions…", also "Non-local EMEP model contributions are all emissions from outside this region, for the downscaled sources, as well as all other primary and precursor emission sources from within this region that are not downscaled."

   It is unclear how the non-local EMEP is obtained from the 2x2 EMEP local fractions. If summed up, how is double-counting avoided? Indeed, the non-local fraction of one given cell reaching the neighbouring cell will be counted as non-local while it is actually local within the 2x2 area, right?

   At every calculation point in uEMEP the local contribution from the surrounding 2 x 2 region in EMEP is removed to provide the non-local contribution. The EMEP local fraction calculation tracks over several EMEP grids, so it is not just the grid itself that the local contribution is removed from, but also neighbouring grids as required. We've added a schematic figure in Sect. 2 to illustrate it.

4. Can the Authors detail the 2x2 grid cell kernel calculation (L48-49)? For an emission located within a given EMEP cell, say near its SW corner, how are these 2x2 cells defined?

   We've added a new schematic figure to illustrate all steps in Sect. 2. In Sect 2.2, we have changed text to: "*For these simulations this region corresponds to 2 x 2 EMEP*

grids, i.e. within an area that is ± 0.1° in both latitude and longitude. This ensures that no matter where the uEMEP calculation sub-grid is placed that the moving window region will always be covered by the 3 x 3 local fraction region".

Technical remarks

1. L45: even though detailed in other references (listed), some explanations on how the EMEP local fraction is calculated would be helpful in this work.

    We've added a new schematic figure in Sect. 2 to illustrate all steps (Fig. 1 in the revised manuscript). At the end of Sect. 2.1, we've added a new paragraph to explain 'EMEP local fractions': "*The EMEP model calculates and outputs the 'local fraction', used by the uEMEP downscaling to remove double counting of emissions (Denby et al., 2020; Wind et al., 2020). The local fraction is the contribution of emissions in one EMEP grid to itself and to its neighbouring grids. For this application only a 3 x 3 grid contribution region is calculated, though for other applications this can be much larger. By tagging the grid emissions in this way the local contribution from EMEP can be removed and replaced by the high-resolution uEMEP sub-grid calculation.*" We've also added explanations accordingly in several places of Sect. 2.2.

2. L56: Can the authors comment on the height of the first vertical layer (50m). Does this impact the downscaled results?

    The height of the lowest layer in EMEP has some, but little, impact on the results since the lowest layer concentrations calculated by EMEP are removed and replaced by uEMEP in the 2 x 2 grid region surrounding each sub-grid. There are many details such as this that we could have provided sensitivity studies for, but we have limited these to the studies provided.

3. L85: Is the split in tiles motivated by CPU gains only or does it improve the efficiency for other aspects?

We can run multiple simulations in parallel, one simulation for each tile. So it's mostly motivated by making full use of the computing power.

4. L93: "Weights are based on Norwegian average road situations". Can the authors comment on the validity of this assumption when applying it to other countries

At the end of the first paragraph of Sect 2.3, we commented that: "Sensitivity tests with alternative weighting, see Sect. 5.2, show the choice of weighting does impact on the results but that the current choice provides close to optimal spatial correlation when compared to measurements."

5. L99: The choice of population as proxy to redistribute residential heating emissions is known to lead to important issues for some cities (as shown later in the document). Is this initial choice related to data availability?

It should be noted that we redistribute each grid's emission by population within that grid. This does not give the same problems as distributing a country's emissions using population. If some cities are known to have reduced residential combustion in the EMEP emissions inventory then they will be reduced there already. As shown in Sect. 5.3, we tested a range of other proxies and variants of the population density as proxy. Population data is indeed very available data for this proxy and was the first choice because 1) it is available everywhere and 2) it does spatially delineate where the emissions come from, even though the emissions may not be directly related to the actual population. That is why we have also tried building density and building density masked with population and a number of population power factors that reduce or enhance the impact of population density for scaling emissions. The results in Section 5.3 address this but do not provide a convincing 'best proxy'.

6. L171 and L334: The spatial representativeness of some traffic stations, especially those located in street canyons is lower than 25 meters. Can the authors comment on their choice to keep those stations anyway for the comparison?

We don't exactly know how many traffic sites are street canyon sites. Reporting to the EU Commission (e-reporting) on siting of traffic stations is generally quite incomplete. Metadata provided in e-reporting was analysed for the EU commission in 2018 (Tarrasón et al., 2021). It indicated that the majority of stations are not street

canyon sites, though the exact number was unclear and most countries do not report this at all.

7. L169 and others: Note that the correlation is actually "r" not "r2". The latter is referred to as the coefficient of determination. Please adapt the text or figures accordingly.

In the first paragraph of Sect. 4, we've corrected the expression as "Results focus on the spatial correlation, *expressed in terms of the coefficient of determination* (r^2), and …". We've also made sure that it is "spatial correlation" instead of "correlation" in the text, and put in "coefficient of determination" in the related figure captions.

8. Figures 10 and 12: please explain which station or station averages are shown.

We've added an explanation in the captions of Fig. 10 and Fig. 12: "The results are based on the European calculations and all available Airbase stations are included."

9. L229: "the" is doubled twice!

Corrected.

10. L311: Can the Authors comment on the robustness of this fitting for other years?

The fitting parameters have only been assessed for European stations for the year 2017, though several years have been fitted for Norwegian stations. These Norwegian fits show a robust, though slightly different to the Europe, relationship since O_3 levels are built into the fitting parameters. The fitting parameters are expected to be slightly different in different years and for different datasets due to changes in NO_2/NO_x emission ratios for traffic and also possibly reduced NO_x emissions and varying ozone levels. If such fitted data were to be used for later years, e.g. 2021, then these fits would need to be updated. We do not intend to use the Romberg scheme for further work but find it instructive for this study.

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Downscaling of air pollutants in Europe using uEMEP_v6

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Abstract. The air quality downscaling model uEMEP and its combination with the EMEP MSC-W chemical transport model are used here to achieve high-resolution air quality modeling at street level in Europe. By using publicly available proxy data, this uEMEP/EMEP modelling system is applied to calculate annual mean NO_2, $PM_{2.5}$, PM_{10} and O_3 concentrations for all of Europe down to 100 m resolution and is validated against all available Airbase monitoring stations in Europe at 25 m resolution. Downscaling is carried out on annual mean concentrations, requiring special attention to non-linear processes, such as NO_2 chemistry, where frequency distributions are applied to better represent the non-linear NO_2 chemistry. The downscaling shows significant improvement in NO_2 concentrations where spatial correlation has been doubled for most countries and bias reduced from -46% to -18% for all stations in Europe. The downscaling of $PM_{2.5}$ and PM_{10} does not show improvement in spatial correlation but does reduce the overall bias in the European calculations from -21% to -11% and from -39% to -30% for $PM_{2.5}$ and PM_{10} respectively. There is improved spatial correlation in most countries after downscaling of O_3, and a reduced positive bias of O_3 concentrations from +16% to +11%. Sensitivity tests in Norway show that improvements in the emission and emission proxy data used for the downscaling can significantly improve both the NO_2 and PM results. The downscaling development opens the way for improved exposure estimates, improved assessment of emissions as well as detailed calculations of source contributions to exceedances in a consistent way for all of Europe at high resolution.

1 Introduction

The EMEP Meteorological Synthesizing Centre - West (EMEP MSC-W) at the Norwegian Meteorological Institute has been developing and implementing a downscaling methodology to enhance the capabilities of the EMEP MSC-W chemical transport model (Simpson et al., 2012, 2020) (hereafter the EMEP model). This downscaling model is known as uEMEP (urban EMEP) and can achieve high-resolution air quality modelling down to 100 m for entire countries (Denby et al., 2020). Even though the methodology is referred to as 'downscaling', uEMEP is actually an independent Gaussian plume modelling system which is added as post-processing to the EMEP model. This makes the modelling similar to other local-scale air quality models and allows for a good physical representation of air quality concentrations.

uEMEP was first reported in the 2016 EMEP status report (Denby and Wind, 2016). Since then uEMEP has been further developed and operationally implemented in the Norwegian Air Quality Forecasting System (Miljodirektoratet, 2020b) as well as providing air quality data, maps and information to Norwegian municipalities through the Air Quality Expert Service (Miljodirektoratet, 2020a). These model applications and validations are described in detail in Denby et al. (2020).

The long-term aim of the uEMEP/EMEP modelling system is to extend uEMEP to cover all the EMEP model domain, so that we can have air quality modelling at street-level all over Europe. Modelling at high resolution provides a better assessment of air quality mitigation strategies in Europe, as well as improved population exposure estimates for use in health impact studies.

30 Previous air quality modeling across Europe could not reach such a high resolution of 100 m (Sofiev et al., 2015; Menut et al., 2013), or the street-level air quality modeling studies were limited to individual cities (Stocker et al., 2012; Kim et al., 2018). The uEMEP/EMEP modelling system is now established in Norway, where access to good quality emission related data is available. The Norwegian emissions are summarized in Sect. 5.4 and details can be found in Sect. S4.2 of Denby et al. (2020). Unfortunately, the same quality of high-resolution emission data that is available in Norway is not directly available

35 for all of Europe. Many countries have suitable high-resolution data but these are not readily accessible for use. In order to implement uEMEP for all of Europe then proxy data that can be used to redistribute emissions to fine scales are required. Three datasets are available for all of Europe, also globally, and have been used to enable the high-resolution modelling in Europe: OpenStreetMap (OSM) (OpenStreetMap contributors, 2020) for redistributing road traffic emissions, population data from Global Human Settlement (GHS) (Schiavina et al., 2019) gridded to 0.0025 degrees for redistributing residential

40 heating emissions, and Automatic Identification System (AIS) (Kystverket, 2020) data for shipping emissions gridded to 0.0025 degrees. These datasets allow downscaling of the traffic, residential heating and shipping emission sources. All other sources are not included in the downscaling.

Results of the European modelling for NO_2, $PM_{2.5}$, PM_{10} and O_3 are presented as example maps in Sect. 3, validation against Airbase stations is in Sect. 4 and results of a number of sensitivity studies are reported in Sect. 5.

2 Methodology

45

Downscaling with uEMEP applies the following methodology where the steps are illustrated in Fig. 1 and additional details are in Sections 2.1 to 2.4:

- Calculations are made using the EMEP model for all of Europe in a similar way to the official EMEP model calculations but with the additional output of the EMEP local fractions (EMEP Status Report 1/2017, 2017; Wind et al., 2020).

50 - uEMEP is implemented as a post-processing routine to the annual mean output from the EMEP model. EMEP emission grids per sector and per compound are redistributed onto high-resolution sub-grids using the emission proxies.

- uEMEP then calculates the local dispersion from these sub-grid emissions using a dispersion kernel within a moving window region defined to be the size of 2 x 2 EMEP grids.

- uEMEP removes the local fraction contribution from the EMEP grid, within the same moving window region, and
55 replaces these with the uEMEP sub-grid results.

- A frequency distribution based chemistry scheme is applied to calculate downscaled NO_2 and O_3 concentrations from annual mean NO_X (NO + NO_2) and O_X (O_3 + NO_2) concentrations.

– Resolution of the sub-grids varies according to the application but maps are made at 100 m and calculations at monitoring sites are made at 25 m.
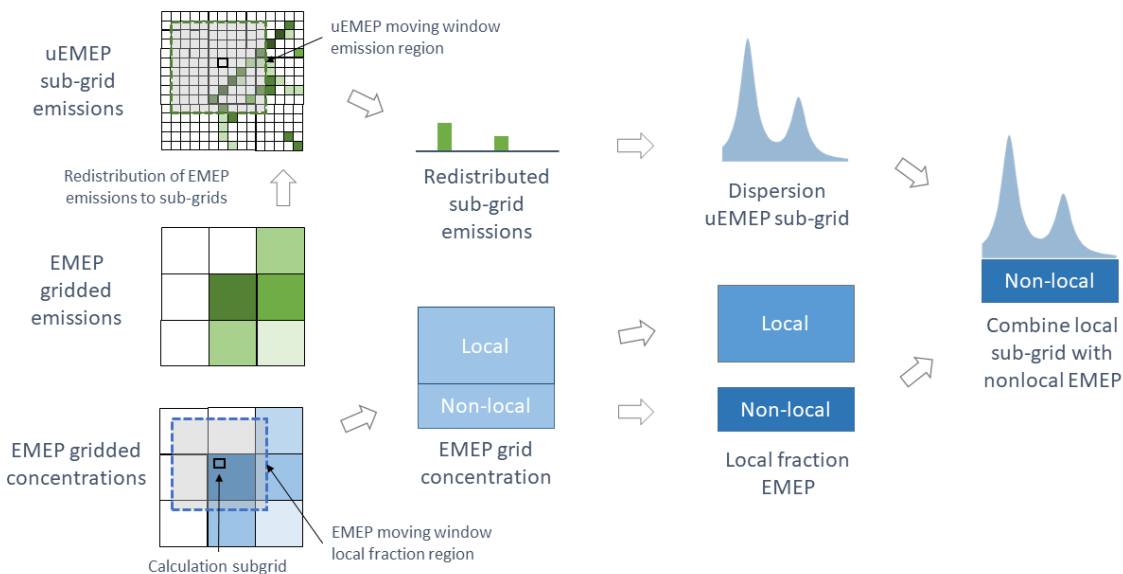
Schematic outline of the uEMEP/EMEP methodology

Figure 1. Schematic illustration of the EMEP/uEMEP coupled modelling system.

2.1 EMEP model implementation

The EMEP model setup follows that in EMEP/MSC-W et al. (2020). Model version rv4.36 is used in this study, with a horizontal resolution of 0.1°x 0.1°and 20 vertical layers (the lowest layer height of approximately 50 m). The model domain covers the geographic area between 30°N-82°N latitude and 30°W-90°E longitude. The simulation year is 2018.

The meteorological data is taken from the Integrated Forecast System (IFS) of the European Centre for Medium-Range Weather Forecasts (ECMWF), with the version IFS Cycle 40r1 (ECMWF-IFS cy40r1). The emission inventory for 2018 is based on the official data submissions to EMEP Centre on Emission Inventories and Projections (Pinterits et al., 2020) in 2020, in which the PM emissions from the residential combustion sector (GNFR C) are replaced by a bottom-up estimate of TNO (Denier van der Gon et al., 2020; Fagerli et al., 2020) for 2017. This TNO dataset should represent an improved estimate of residential combustion emissions of PM, accounting for condensable organics in a consistent way.

The EMEP model calculates and outputs the 'local fraction', used by the uEMEP downscaling to remove double counting of emissions (Denby et al., 2020; Wind et al., 2020). The local fraction is the contribution of emissions in one EMEP grid to itself and to its neighbouring grids. For this application only a 3 x 3 grid contribution region is calculated, though for other

applications this can be much larger. By tagging the grid emissions in this way the local contribution from EMEP can be removed and replaced by the high-resolution uEMEP sub-grid calculation.

2.2 uEMEP model implementation

The uEMEP model is described in a recent publication (Denby et al., 2020). In that article the Norwegian forecast application of uEMEP is described where hourly downscaling using bottom-up emission inventories is carried out. For the European application calculations are made on annual mean data, creating air quality maps for Europe down to 100 m resolution and calculating concentrations at Airbase stations positions down to 25 m.

Downscaling is carried out in the following way. EMEP grid emissions per sector and per source are redistributed to uEMEP sub-grids using the proxy emission data described in Sect. 2.3. Each sub-grid dispersion calculation uses only the emission sub-grids within the moving window emission region, see Fig. 1. For annual mean calculations dispersion is carried out using a rotationally symmetric Gaussian dispersion kernel (Denby et al., 2020), given an initial plume size and height. These parameters are provided in Table 1. The initial horizontal plume size is determined by the size of the sub-grid. The Gaussian dispersion parameters used are based on the K_z dispersion methodology described in Denby et al. (2020) but adapted to the rotationally symmetric dispersion kernel. The EMEP model local fraction contribution originating from within the moving window region is removed and replaced with the sub-grid dispersion calculation from uEMEP, thus avoiding double counting of emissions (Fig. 1). For these simulations this region corresponds to 2 x 2 EMEP grids, i.e. within an area that is $\pm 0.1^o$ in both latitude and longitude. This ensures that no matter where the uEMEP calculation sub-grid is placed that the moving window region will always be covered by the 3 x 3 local fraction region.

Downscaling with uEMEP occurs only for primary emissions. NO_2 is calculated from NO_X and O_X using the same travel time parameterisation described in Denby et al. (2020) but applied to annual mean wind speeds, photo-dissociation rates and concentrations. To account for non-linearity in the NO_2 chemistry, when calculating with annual means, an additional frequency distribution correction factor is implemented, see Sect. 2.4. Annual mean downscaled O_3 is also determined using this same parameterisation.

To improve efficiency of the calculations, Europe is split into a number of tiles that cover the European land domain. For the 100 m resolution mapping calculations there are 1097 tiles, each of which is 100 km x 100 km. These tiling regions are shown in Fig. 2.

Table 1. Initial dispersion (σ_{z0}) and emission height (h_{emis}) for the three downscaled sources for all primary pollutants.

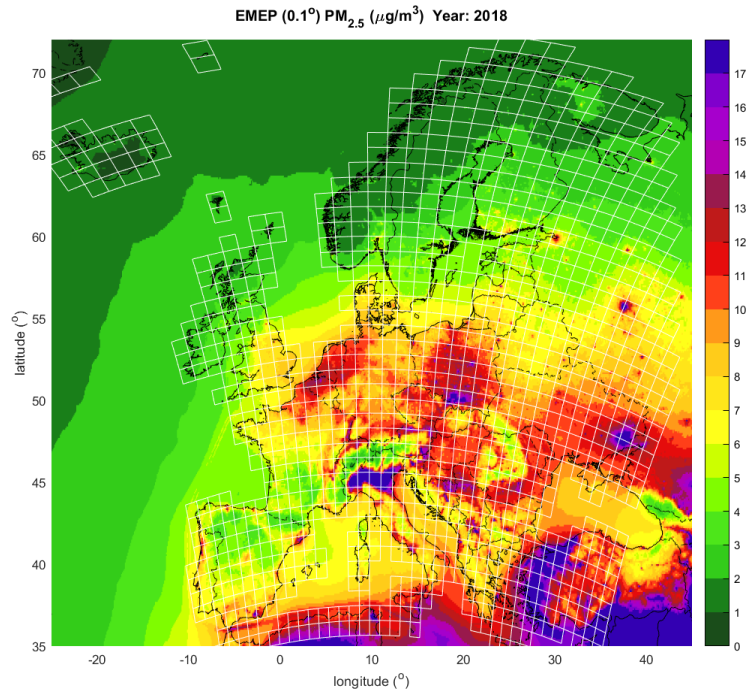| Source	Initial dispersion (σ_{z0})	emission height (h_{emis})
Traffic (GNFR6)	2 m	1 m
Residential heating (GNFR3)	10 m	15 m
Shipping (GNFR7)	15 m	70 m

4

Figure 2. Annual mean PM$_{2.5}$ concentrations calculated with the EMEP model at 0.1^o for 2018. Shown are the uEMEP tiling regions used in the calculations. In total 1097 100 km x100 km tiles with 100 m resolution are used to model Europe.

2.3 uEMEP proxy data

100 We use road data from OSM to redistribute the traffic emissions. Though the spatial coverage of OSM is very good, it does not contain actual traffic data. Redistribution of the emission data is achieved by weighting the different road categories provided in OSM. The following road categories are considered: motorway, trunk, primary, secondary, tertiary, unclassified and residential. Each is weighted relative to the other so that emissions can be redistributed and attributed to the road links. Estimates of the weights are based on the representative average daily traffic (ADT) for different road categories for Norwegian average road

105 situations. The weighting currently employed for the redistribution is shown in Fig. 3. It is also worth noting that for major roads, such as motorways, OSM often represents these as dual carriageways, i.e. as two separate road links. In these cases the weighting of a motorway will be twice that indicated here. Sensitivity tests with alternative weighting, see Sect. 5.2, show the choice of weighting does impact on the results but that the current choice provides close to optimal spatial correlation when compared to measurements.

110 A global population dataset from the GHS is used as the proxy for redistributing residential heating emissions. We choose the highest available resolution of 9 arcsec (0.0025^o) from the year 2015. The coordinate system is WGS84. This dataset indicates

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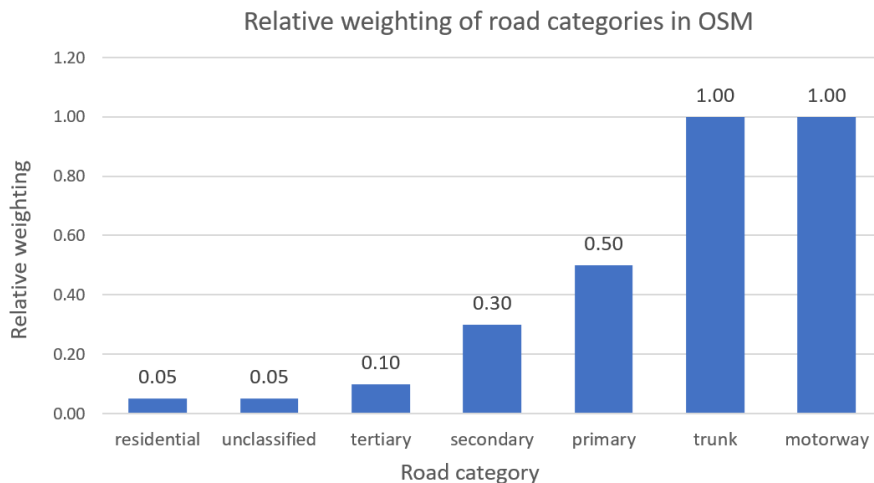
Figure 3. Weighting of the OpenStreetMap road categories used to redistribute EMEP traffic emissions for downscaling. Applied to all primary pollutants.

the distribution of population as the number of people per cell. A number of alternative formulations of the population proxy, as well as an alternative proxy based on building density, are assessed in Sect. 5.3

AIS data for shipping emissions are provided by the Norwegian Coastal Administration. The raw data, which contains a list of instantaneous point emissions, is averaged over the year 2017 and gridded to 0.0025^o. Though these data are actual emissions we still use them as proxy data to redistribute EMEP gridded emissions, to be consistent in the methodology.

2.4 uEMEP chemistry parameterisation for annual mean NO$_2$

uEMEP downscales only primary pollutants. It is thus necessary to apply chemistry parameterisations to the NO$_X$ and EMEP O$_3$ concentrations to derive downscaled NO$_2$ and O$_3$ concentrations. Two methods for doing this are described in Denby et al. (2020). One for hourly concentrations using a weighted travel time parcel method and the other for annual means, which is based on a simple empirical relationship between observed NO$_2$ and NO$_X$. It is desirable to apply the model based chemistry scheme rather than an empirical scheme, however due to the non-linearity of the NO$_2$ chemistry the chemical scheme cannot be directly applied to annual mean concentrations.

To solve this, chemistry is not calculated on just a single annual mean value for NO$_X$ and O$_3$ but on a frequency distribution for these parameters that represent the variability over a year. This can be illustrated for the photostationary case where the NO$_2$ concentrations can be derived from NO$_X$ and O$_X$ using

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

6

where the concentrations are annual mean values in molecules/cm^3, k_1 is the production reaction rate and J the photo-dissociation rate for NO$_2$. If the frequency distribution for the three annual mean variables [NO$_X$], [O$_X$] and J/k_1 is known then we can integrate over Eq. (1) using these distributions as weighting functions. An appropriate probability distribution function for the concentrations is the log-normal distribution, which can be written as

$$PDF_x = \frac{1}{x\sigma\sqrt{2\pi}}exp\left(-\frac{(log(x)-\mu)^2}{2\sigma^2}\right) \tag{2}$$

where the log-normal parameters μ and σ are determined from the mean (m) and the standard deviation (s) by

$$\mu = log\left(\frac{m^2}{\sqrt{m^2+s^2}}\right) \text{ and } \sigma^2 = log\left(1+\frac{s^2}{m^2}\right)$$

The frequency distribution of J is not log-normally distributed, since it is dependent on the solar zenith angle (ZA) and various other meteorological parameters, such as cloud cover and water vapour content. The EMEP model uses lookup tables based on precalculated J values from the Phodis model (Jonson et al., 2000). In order to implement the frequency distribution of J in uEMEP a power law fit is made to the tabulated values. This can be written as:

$$J = C_j cos(ZA)^{-p_j} \tag{3}$$

where C_j is a constant that is normalised out when producing the normalised frequency distribution and $p_j = 0.28$. The standard deviation of k_1, dependent on air temperature, is significantly smaller than for J so it is treated as a constant.

A new value [NO$_2$]$_{pdf}$ can then be determined using these frequency distributions

$$[NO_2]_{pdf} = \int\!\!\!\int\!\!\!\int_0^\infty [NO_2]PDF_{ox}PDF_{nox}PDF_j\,d[NO_X]\,d[O_X]\,dJ \tag{4}$$

and a correction term showing its difference from the mean is defined as

$$f_{no2,pdf} = \frac{[NO_2]_{pdf}}{[NO_2]_{mean}} - 1 \tag{5}$$

When calculating in three orthogonal dimensions then it is assumed there is no correlation between the variables.

To implement this procedure the standard deviation s must be known for NO_X and O_X. Values for s_{nox} and s_{ox} have been

155   derived from earlier model calculations for Norwegian stations. Linear regression provides robust values for s_{ox}/m_{ox} and s_{nox}/m_{nox} of 0.21 and 1.14 respectively (see Fig. S5). The variability of NO_X reflects the variability of the traffic emissions, for stations within the influence of traffic, and this should be generally applicable throughout Europe. 72 sites are used for the calculation. The calculation of the distribution correction is carried out numerically after calculation of the NO_2 concentrations.

Implementation of the frequency distribution for concentrations has a significant impact, with a general reduction in NO_2

160   concentrations compared to the annual mean calculation using Eq. 1. This reduction leads to correction terms ($f_{no2,pdf}$) of between 0 to -25%. The highest corrections occur around $NO_X=O_X$, where Eq. (1) shows the most non-linear behaviour. On average for all station sites in Europe, around a -16% reduction on the initially calculated $[NO_2]_{mean}$ has been determined. In contrast to the distribution correction for concentrations, the distribution correction for J leads to an increase in NO_2 of around 6%. This is because roughly half of the frequency distribution for J is 0, i.e night time, when there is no photo-dissociation.

165   In Sect. 5.5 the impact of this and other chemistry schemes is further discussed. More information concerning this scheme is contained in the supplementary material S1.4.

3   Example maps
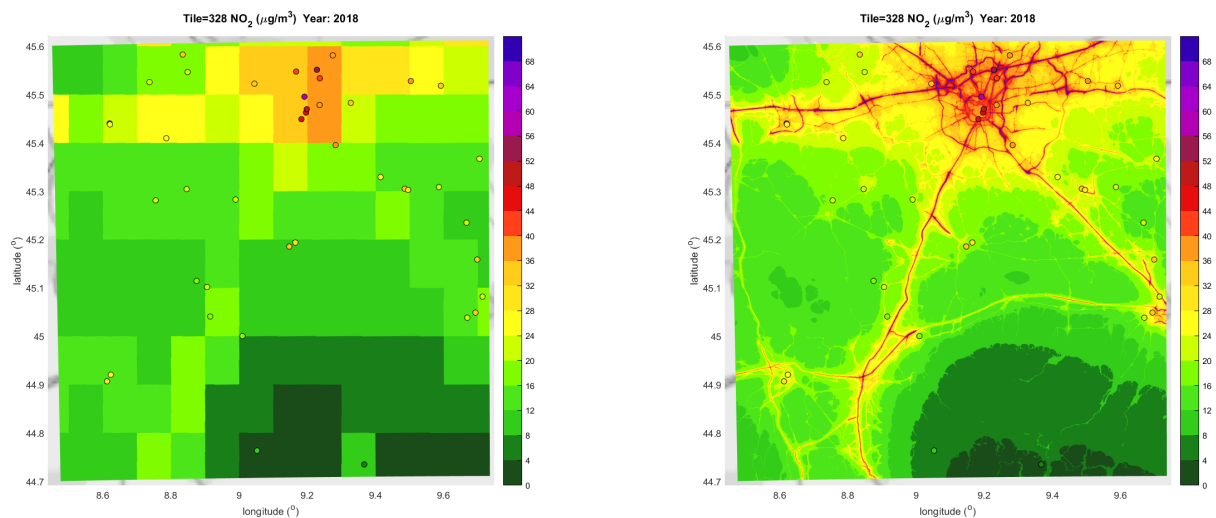
Figure 4. Calculated NO_2 concentrations in the 100 km tile (nr. 328) for 2018, part of the all European calculation at 100 m resolution. Left the EMEP model calculation at 0.1^o and right the uEMEP calculation at 100 m resolution. The city in this tile is Milan. Airbase stations are shown as circles.

In this section we present example maps that are generated from the EMEP model and uEMEP simulations. 100 km example tiles are shown in Fig. 4 - 6, demonstrating the original EMEP model calculations and the downscaled maps using uEMEP for
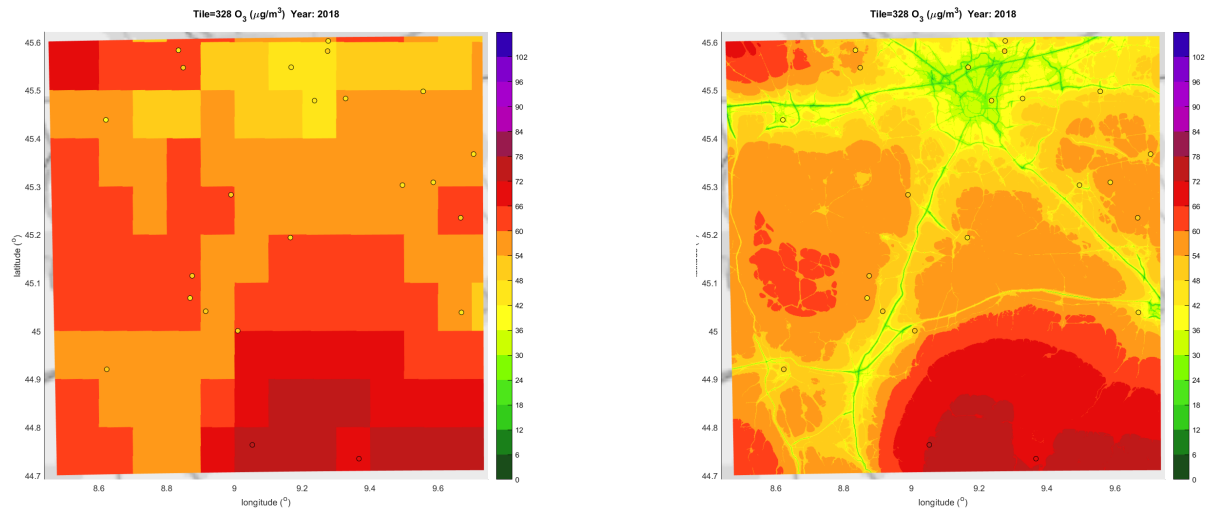
Figure 5. Calculated O$_3$ concentrations in the 100 km tile (nr. 328) for 2018, part of the all European calculation at 100 m resolution. Left the EMEP model calculation at 0.1^o and right the uEMEP calculation at 100 m resolution. The city in this tile is Milan. Airbase stations are shown as circles. This is the same tile as is shown in Fig. 4.
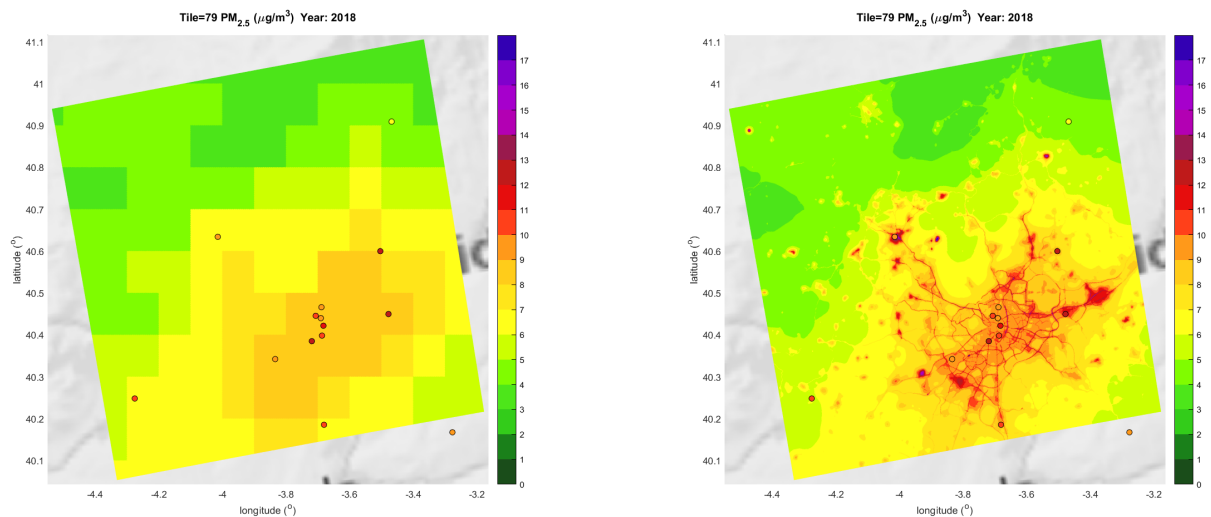

Figure 6. Calculated PM$_{2.5}$ concentrations in the 100 km tile (nr. 79) for 2018, part of the all European calculation at 100 m resolution. Left the EMEP model calculation at 0.1^o and right the uEMEP calculation at 100 m resolution. The city in this tile is Madrid. Airbase stations are shown as circles.

170   NO$_2$, O$_3$, and PM$_{2.5}$. The uEMEP calculations are made on an x-y projected map commonly used for European mapping. The projection used is the European ETRS89-LAEA projection (EPSG: 3035). Maps presented are shown on latitude and longitude which means that the projected uEMEP tiled maps do not necessarily follow the North-South direction.

The downscaled maps resolve more variability between stations. Compared with the EMEP model maps, uEMEP maps have higher concentrations of NO$_2$ and PM$_{2.5}$, and lower concentration of O$_3$, in heavy traffic and populated areas due to the
175   high-resolution proxy dataset.

4   Validation

Observed annual mean concentrations of NO$_2$, PM$_{2.5}$, PM$_{10}$, and O$_3$ from Airbase (European Environment Agency, 2018) are used for comparison with both the EMEP model and uEMEP calculations. All valid Airbase stations with more than 75% coverage are used in the validation and are assumed to be sited at 3 m above the surface. Results for the year 2018
180   are presented. Results focus on the spatial correlation, expressed in terms of the coefficient of determination (r^2), and on the relative bias (Bias). For station sites the downscaling with uEMEP is performed on 25 m sub-grids, which is of sufficient resolution to spatially represent traffic sites. However, since the Gaussian model used does not take into account buildings or obstacles, then traffic sites in street canyons or built up areas may be underestimated. A study by Lefebvre et al. (2013) in Antwerp, where both Gaussian and street canyon models were applied at 15 street canyon modelling sites, showed an average
185   street canyon modelling increment of just 11% for NO$_2$. We include all available sites in this study because in Europe the majority of traffic sites appear not to be street canyons, though information on this is unclear (Tarrasón et al., 2021). Also, even without including obstacles, the increased model resolution (up to 25 m) allows the concentration gradients at roadside to be better described.

4.1   NO$_2$

190   In Fig. S6 and Fig. S7 scatter plots for NO$_2$ are shown for each country and Europe as a whole. These results are summarised in Fig. 7 where the annual mean concentration and spatial correlation are shown.

In a majority of countries the spatial correlation for NO$_2$ is more than doubled when implementing uEMEP. The two exceptions are Ireland (IE), where the spatial correlation hardly changes with the downscaling, and Bosnia and Herzegovina (BA), where the spatial correlation is significantly reduced. Both these countries have very few stations. The highest spatial
195   correlation is for Poland (PL) with $r^2 = 0.85$.

It is worth noting that the average spatial correlation per country is $r^2 = 0.62$ which is higher than the spatial correlation when assessed for all stations in Europe ($r^2 = 0.57$). This indicates that a part of the variability occurs between countries and can be interpreted to reflect differences related to emission reporting from each country. If the NO$_X$ emissions from individual countries have uncorrelated bias then this will reduce the overall spatial correlation.

200   The relative bias is improved for all countries with the exception of Greece (EL), which is the only country with a significant positive bias. Overall for Europe bias is improved from -46% for the EMEP model to -18% when using uEMEP. Of the 28

countries with 10 or more monitoring sites, 18 of these have an absolute bias less than 25% after downscaling. Turkey (TR) has the largest negative bias, after downscaling, of -45%.

4.2 PM$_{2.5}$

205   In Fig. S8 and Fig. S9 scatter plots for PM$_{2.5}$ are shown for each country and Europe as a whole. These results are summarised in Fig. 8 where the annual mean concentration and spatial correlation are presented.

Unlike the NO$_2$ downscaling, there is generally no improvement in the spatial correlation when applying uEMEP for PM$_{2.5}$. Only 6 out of 17 countries show improved spatial correlation and overall for Europe there is a slight decrease, from $r^2 = 0.49$ for the EMEP model to 0.46 for uEMEP. This result is further discussed in Sect. 6.
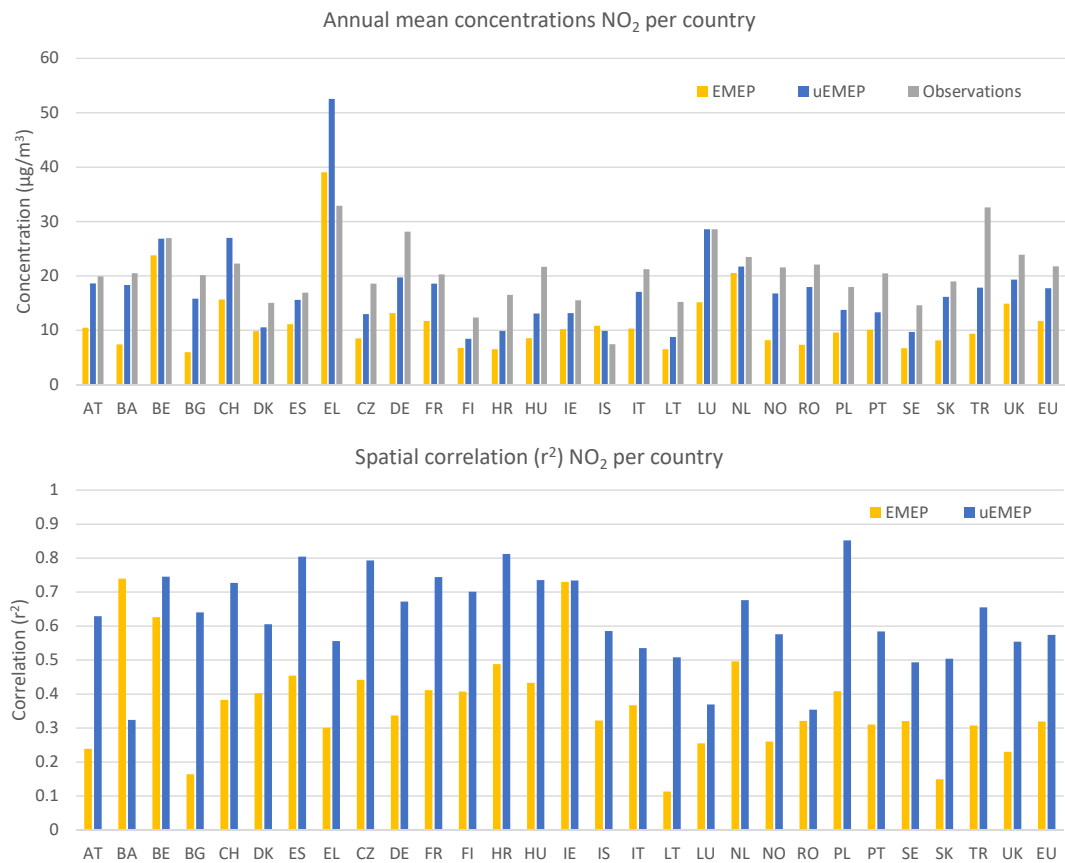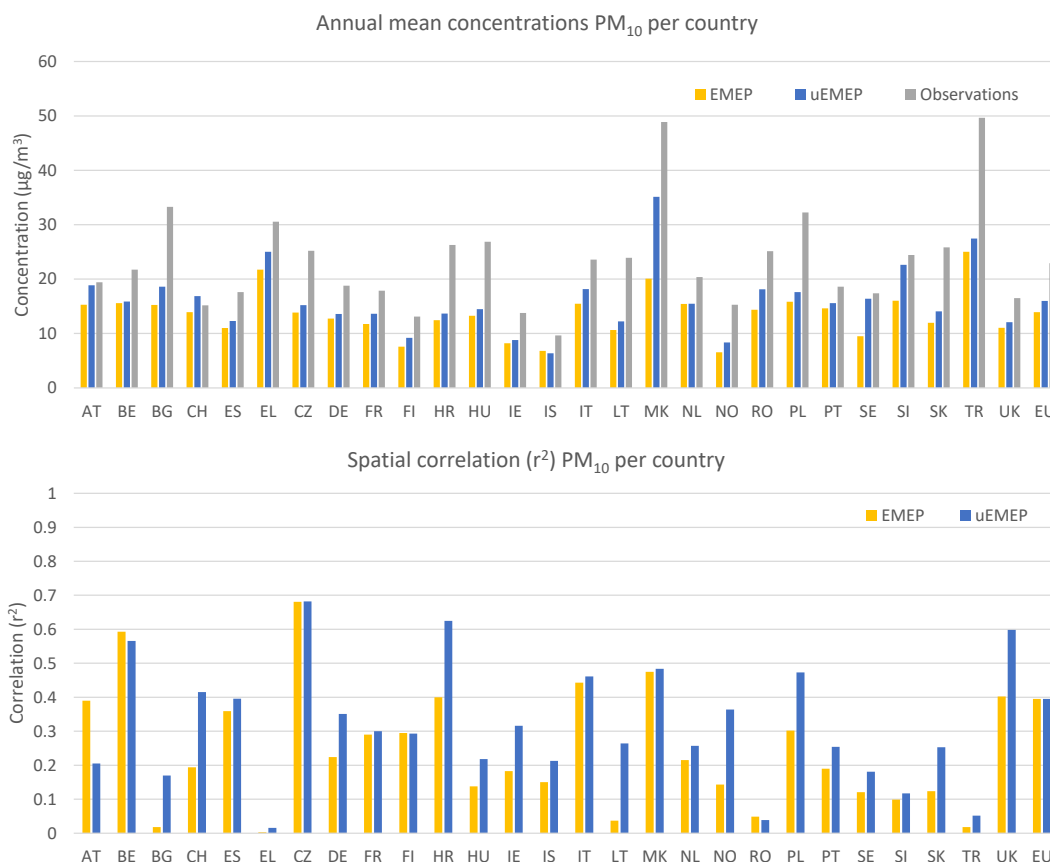

Figure 7. Annual mean NO₂ concentrations and spatial correlation (coefficient of determination r^2) per country for 2018 calculated with the EMEP model and uEMEP compared to Airbase observations. Only countries with 10 or more stations are shown but all stations are included in the final EU result. 3313 stations are included in the comparison.
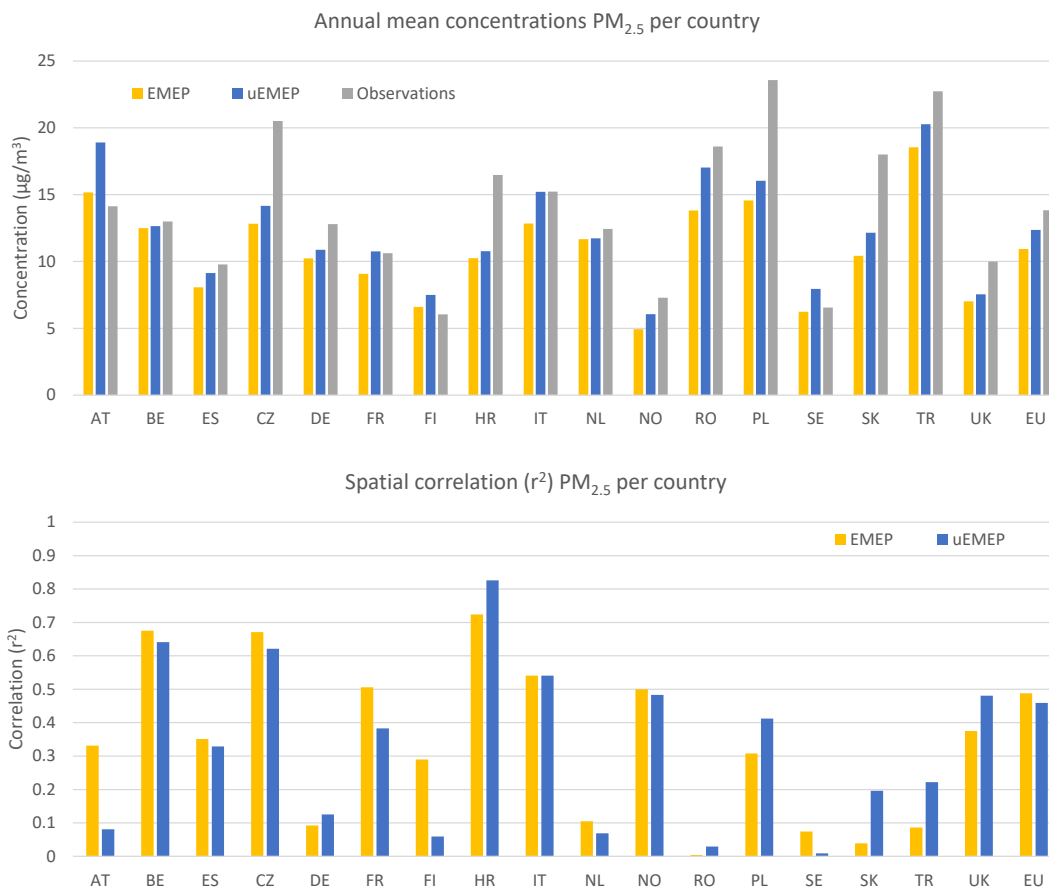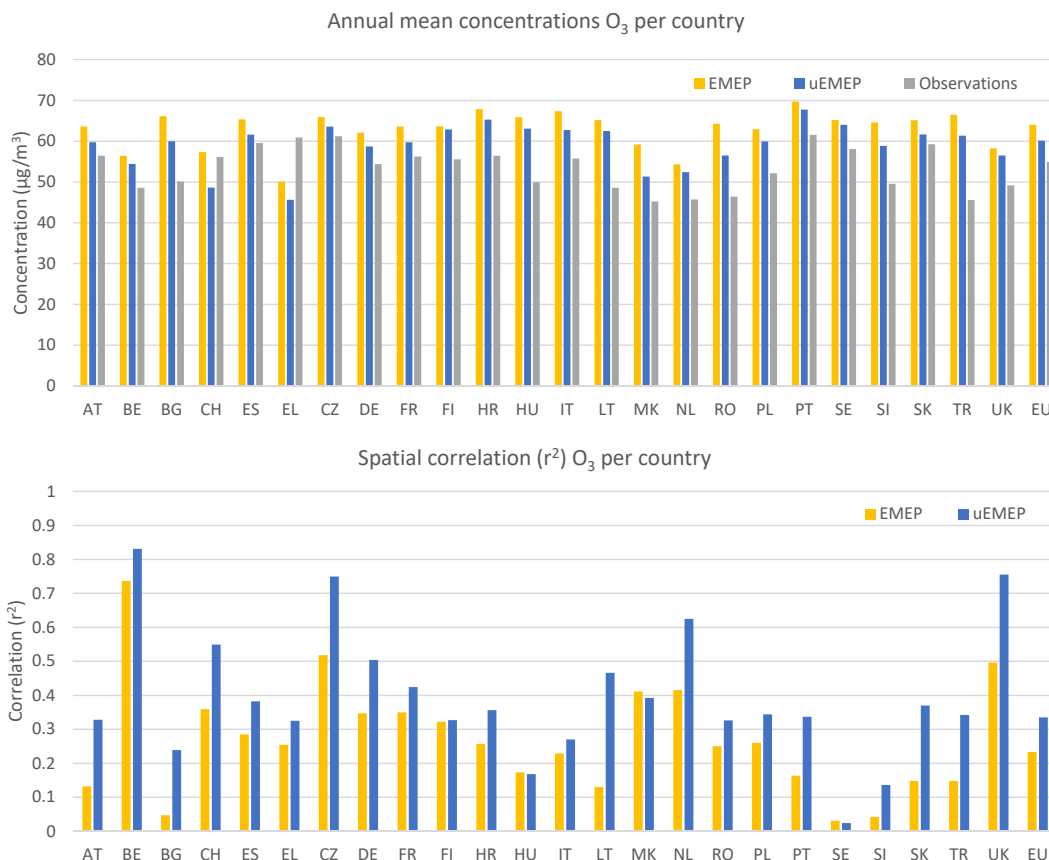
Figure 8. Annual mean PM$_{2.5}$ concentrations and spatial correlation (coefficient of determination r^2) per country for 2018 calculated with the EMEP model and uEMEP compared to Airbase observations including all types of stations. Only countries with 10 or more stations are shown but all stations are included in the final EU result. 1376 stations are included in the comparison.

210   The relative bias is however reduced for almost all countries. For Europe as a whole the relative bias went from -21% for the EMEP model to -11% for uEMEP. Only the three countries Austria (AT), Sweden (SE) and Finland (FI), that had almost no bias with the the EMEP model calculation, achieve a positive bias with uEMEP.

4.3 PM$_{10}$

In Fig. S10 and Fig. S11 scatter plots for PM$_{10}$ are shown for each country and Europe as a whole. These are summarised in
215 Fig. 9.

     The results for PM$_{10}$ are similar to those for PM$_{2.5}$. In this case though the majority of countries, 21 out of 27, have improved spatial correlation with the application of uEMEP. The spatial correlation for all of Europe using uEMEP is unaltered compared to the EMEP model calculation, with r^2=0.34. This is lower than the spatial correlation found for PM$_{2.5}$ by around 0.12.

Figure 9. Annual mean PM$_{10}$ concentrations and spatial correlation (coefficient of determination r^2) per country for 2018 calculated with the EMEP model and uEMEP compared to Airbase observations including all types of stations. Only countries with 10 or more stations are shown but all stations are included in the final EU result. 2891 stations are included in the comparison.

As with PM$_{2.5}$ the relative bias is reduced with the uEMEP downscaling. For Europe we see the relative bias went from -39% for the EMEP model to -30% for uEMEP.

4.4 O$_3$

In Fig. S12 and Fig. S13 scatter plots for O$_3$ are shown for each country and Europe as a whole. These are summarised in Fig. 10.

Ozone is generally reduced with the downscaling due to an increase in NO$_X$ concentrations. In general for Europe we see a reduced positive bias from +16% for the EMEP model to +11% for uEMEP. Spatial correlation is also improved in 21 of the 24 countries. The only countries to show significant degradation in the downscaling results are Switzerland (CH) and Greece (EL). This is likely due to the overestimated NO$_X$ concentrations there (Fig. 7).

Figure 10. Annual mean O_3 concentrations and spatial correlation (coefficient of determination r^2) per country for 2018 calculated with the EMEP model and uEMEP compared to Airbase observations including all types of stations. Only countries with 10 or more stations are shown but all stations are included in the final EU result. 1974 stations are included in the comparison.

5 Sensitivity studies

In this Section we present the results of several sensitivity calculations using uEMEP. These include sensitivity to sub-grid resolution, traffic emission proxies, residential combustion emission proxies, sensitivity to alternative bottom up emissions in Norway and sensitivity to the NO_2 chemistry scheme.

5.1 Sensitivity to resolution

When calculating concentrations at station positions a grid resolution of 25 m is used. However, when mapping all of Europe a lower resolution of 100 m is employed. In Fig. 11 we show the results of a change in resolution on the annual mean NO_2 and $PM_{2.5}$ concentrations for resolutions from 25 to 500 m.

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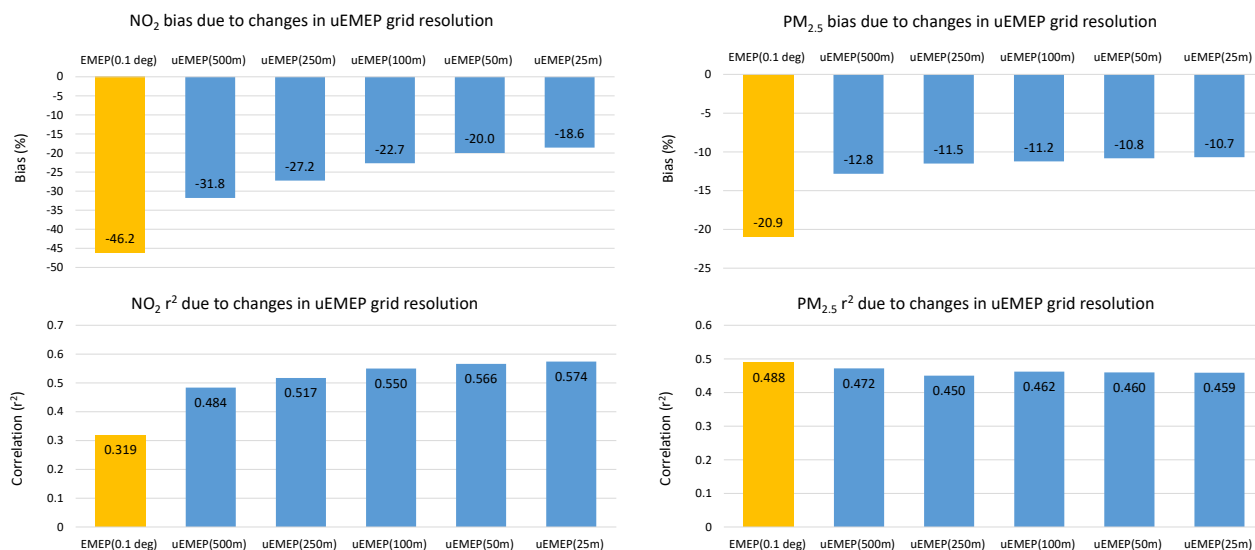
Figure 11. Change in bias and spatial correlation (coefficient of determination r^2) as a result of changes in uEMEP resolution. Shown are the results for NO_2 and $PM_{2.5}$. Also included is the EMEP model 0.1^o calculation in yellow. The results are based on the European calculations and all available Airbase stations are included.

For NO_2 both bias and spatial correlation improve with increasing resolution. 100 m calculations are on average 4% lower than the 25 m calculations. For $PM_{2.5}$ there is little change in bias between the different resolutions. Both shipping and residential combustion sources are only provided at 250 m so any further change in model results at lower resolutions will be due to the traffic contribution only. Spatial correlation is basically unchanged for $PM_{2.5}$ at all resolutions.

5.2 Sensitivity to OSM weighting

In Fig. 3 the weighting imposed on the OSM road categories is shown. This weighting specifies the relative contribution of the different road categories to the redistribution of the gridded traffic emissions in uEMEP. This weighting is based on an analysis of Norwegian traffic data but it is worthwhile assessing the sensitivity of the calculated NO_2 concentrations using different weights. To assess this sensitivity a power law is applied to the weighting. For power indices greater than 1 then more weighting is applied to the major roads, for power law indices less than 1 then more weight is applied to the the minor roads. The different weights for the different power law indices are shown in Fig. 12. The results of this sensitivity test, presented in terms of relative bias and spatial correlation (r^2), are shown in Fig. 13.

Bias is quite strongly affected by the change in weighting. Higher concentrations are calculated when more weight is given to the minor roads. This is likely because most measurement sites are not on major roads. Increasing the weighting to minor roads will generally increase the urban background levels. The spatial correlation is among the highest for the current weighting
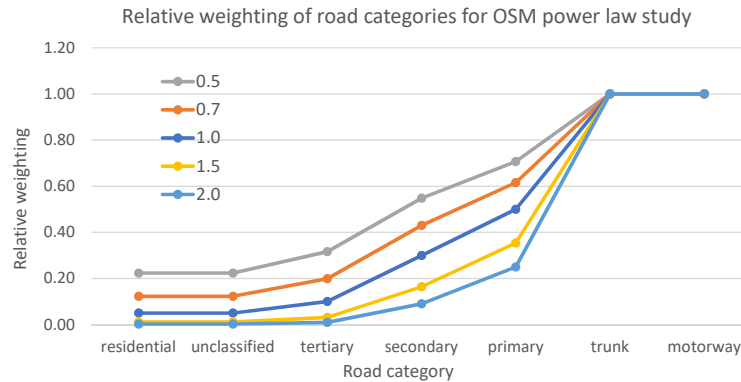
Figure 12. OSM weighting, relative to Motorways, that results with a change in the power law index when applied to the initial weights. The scenarios are tested in the European calculations.
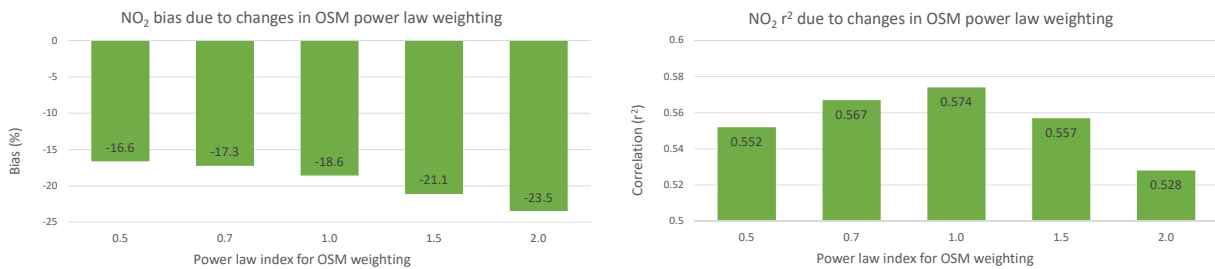

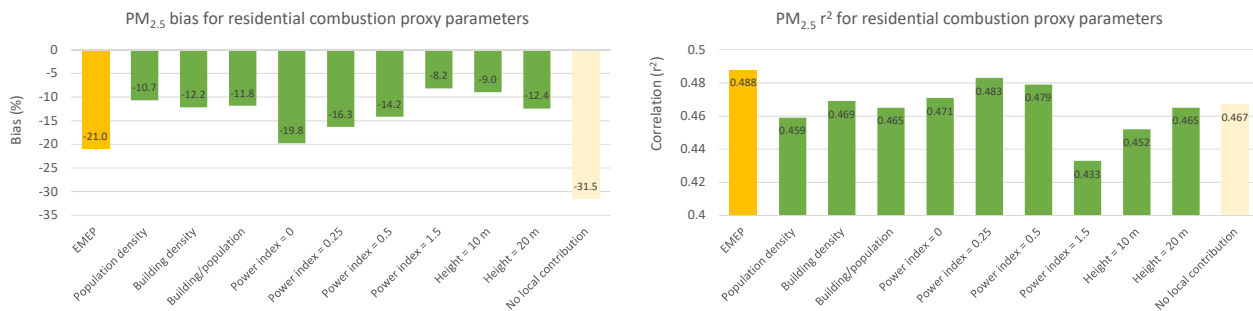**Figure 13.** Change in bias and spatial correlation (coefficient of determination r^2) for NO_2 as a result of changes in the power law index applied to the OSM road traffic weighting. Lower power law indices give more weight to minor roads, higher power law indices give more weight to major roads. The results are based on the European calculations and all available Airbase stations are included.

with a power index of 1. This confirms that the initial estimate, based on Norwegian traffic, reflects a good general distribution of traffic in Europe. If real traffic volume were available then the weighting would be more precise. Tests on Norwegian data, Sect. 5.4, confirm that spatial correlation is significantly improved when using real traffic data for the redistribution weighting.

5.3 Sensitivity to the residential combustion emission proxy

255   For the $PM_{2.5}$ calculations presented in Sect. 4.2 population density data at 0.0025^o has been used to redistribute the residential combustion emissions in uEMEP. The results indicate a slightly reduced spatial correlation but also with an improved negative bias. In this section we assess the sensitivity of the redistribution proxy to a number of alternative proxies. Firstly a power law is applied to the population density data. A lower power law index will reduce the weighting towards highly populated regions.

Figure 14. Change in bias and spatial correlation (coefficient of determination r^2) for PM$_{2.5}$ as a result of changes in the residential combustion proxy. A lower power law index gives less weight to the population redistribution, a higher power law indices give more weight. 'Building/population' is the building density masked by population data. See text for details.

A power law index of 0 will work as a mask, redistributing the EMEP emissions evenly to any 250 m sub-grid that contains
260 population. As an alternative to the population data, building density data has also been extracted from the OpenStreetMap dataset. This has also been placed on a 0.0025^o grid for all of Europe. Two alternatives with this proxy are tested. The first using building density as the weighting proxy and the second using building density masked with population, so that only areas with both buildings and population are used for redistribution. In addition to the alternative proxy data the sensitivity of the calculations to emission height, currently set to 15 m, is also assessed.

265 The results are shown in Fig. 14. Here we see that a power law of 0.25 gives slightly improved spatial correlation and that the use of building density also slightly improves spatial correlation compared to population. However, none of the alternative proxies significantly improves the spatial distribution of PM$_{2.5}$ and none attain the spatial correlation of the EMEP model calculations at 0.1^o. There is a general trend for reduced negative bias to lead to reduced spatial correlation in all calculations, so when the contribution from the downscaled residential combustion increases then spatial correlation decreases. This infers
270 that the redistribution is not improving the results.

In addition to the proxy sensitivity the result of the EMEP model calculation where all local EMEP model grid contributions ($\pm 1^o$) have been removed is shown in Fig. 14. This shows firstly that around 10% of the PM$_{2.5}$ in the EMEP model comes from within this local region and that the inclusion of these emissions does add to improved spatial correlation at the EMEP model 0.1^o scale, from $r^2 = 0.467$ to 0.488. Here we see more clearly that while the bias is improved by downscaling the spatial
275 correlation is not and is similar to the spatial correlation obtained from the non-local contributions. However, it is possible to achieve improved spatial correlation when more appropriate downscaling proxies are used. This is presented in Sect. 5.4.

5.4 Results of improved emission data in Norway

Throughout the uEMEP downscaling simulations we used the 0.1^o country reported emission data and redistributed it using population, OpenStreetMap data and AIS shipping data as redistribution proxies. However, many countries have more detailed

emission data sets, including Norway, that could be used to improve the downscaling calculations. To test the impact of more realistic spatial distributions of emissions, the emission and emission proxy data used in Norway are replaced in the EMEP model and uEMEP calculations with the emission data currently used in the national air quality forecasting in Norway. Details surrounding these emissions can be found in Denby et al. (2020) and Grythe et al. (2019). The most important differences between the Norwegian and European emissions and emission proxy data are: (1) Traffic volume data from the Norwegian national road database is used instead of OSM weighting. Exhaust emissions are based on emission factors using a bottom up methodology and NO_X emissions are additionally corrected for temperature. (2) The non-exhaust road dust emissions are calculated with the NORTRIP model (Denby et al., 2013a; Denby et al., 2013b) which are significantly larger than the current national estimates reported for Norway. (3) The total Norwegian residential heating emissions of PM are the same for both the Norwegian and the European emissions but the Norwegian emissions have been redistributed using the MetVed model (Grythe et al., 2019), which uses much more detailed information than just population to distribute the residential heating emissions at 250 m. (4) The Norwegian emissions and the uEMEP proxy data are entirely consistent with each other since the Norwegian emissions are aggregated grid emissions based on the fine scale emission data.

We make four separate downscaling calculations for Norway using the two emissions, 'European emissions' and 'Norwegian emissions', and the two high-resolution proxy datasets, 'European proxy downscaling' and 'Norwegian proxy downscaling', respectively. Shipping is not changed in these simulations and in this case the calculation year is 2017. Though the resolution of the EMEP model in the Norwegian forecasting system is nominally 2.5 km, for these simulations we use the same 0.1^o EMEP model grid resolution. The results are shown in Fig. 15 for NO_2, $PM_{2.5}$ and PM_{10} where the relative bias (%) and spatial correlation (r^2) are presented.

For NO_2 in Norway the large negative bias seen in the EMEP model is almost completely removed by the use of the traffic downscaling, using either the Norwegian or European emission data. On a national level the local Norwegian (bottom up) traffic NO_X emissions are roughly 25% higher than the EMEP (top down) emissions. NO_2 concentrations are slightly overestimated when using the Norwegian proxy data for traffic. Spatial correlation is improved with the use of the Norwegian proxy data for traffic, compared to European emissions that use OSM data, from $r^2 = 0.6$ to 0.72. It is worth noting that in the complete Norwegian calculation reported in Denby et al. (2020) using hourly calculations that the spatial correlation is even higher at r^2 = 0.78, but the bias is less at -5%.

For $PM_{2.5}$ biases are very similar for both the European and Norwegian proxy data sets when using either the European or Norwegian emissions. The spatial correlation however is significantly higher when using the Norwegian emissions, both at grid level and after downscaling. There is significant improvement, r^2 increases from 0.37 to 0.55, when both changing European emission to Norwegian emission and changing the residential heating proxy from population (European proxy) to the MetVed model (Norwegian proxy). This indicates that improved spatial representation can be attained when both the gridded and the proxy data are consistent and more representative. However, little can be improved with downscaling when the initial gridded emissions are not well distributed, even with improved proxy data. Interestingly we see the same result as reported in Sect. 4.2, that the spatial correlation is reduced when applying the European proxy data to the European emissions. These results
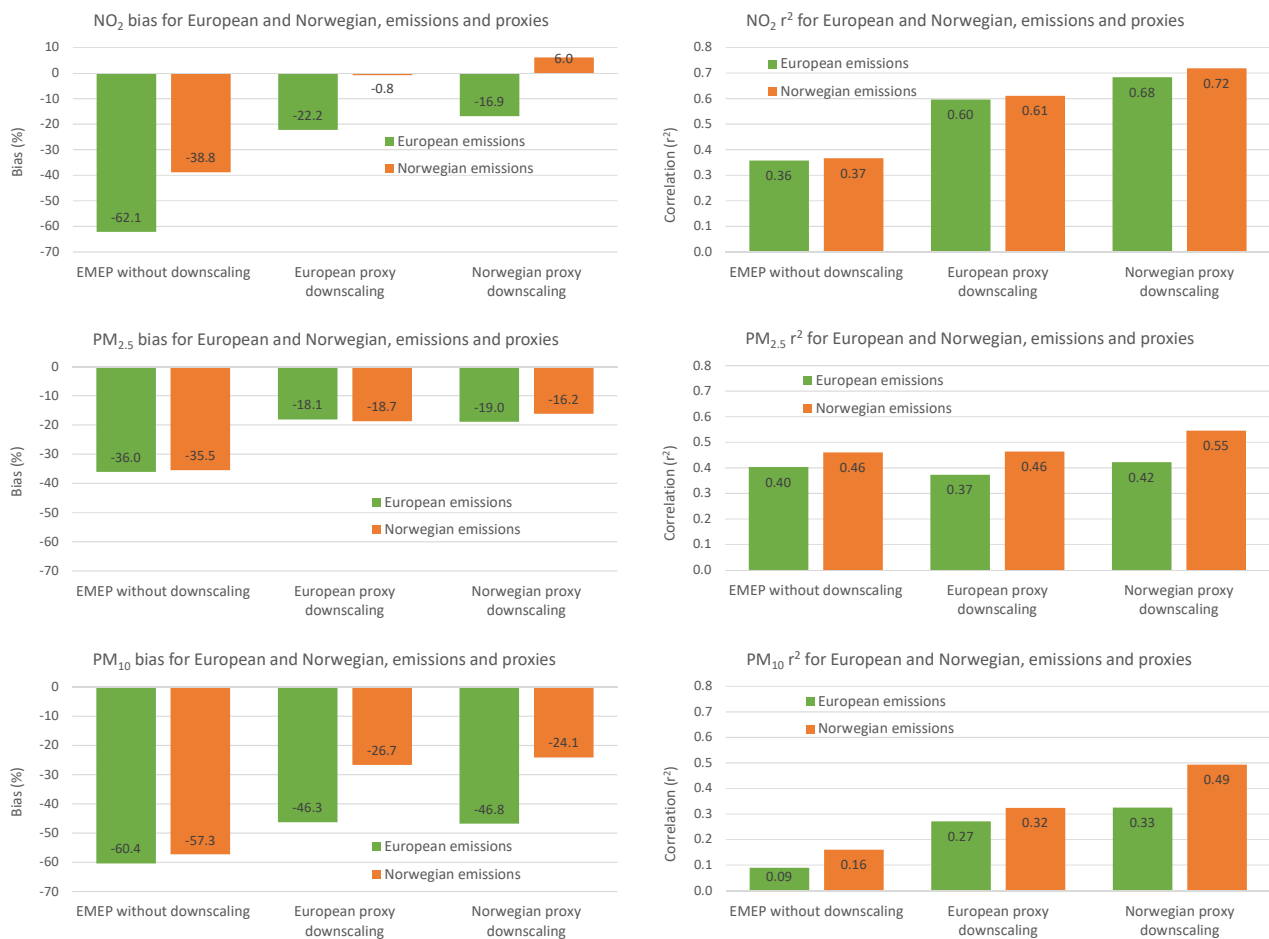
Figure 15. Change in bias and spatial correlation (coefficient of determination r^2) as a result of changes in Norwegian emission and emission proxy data for NO_2, $PM_{2.5}$ and PM_{10} calculations. 'European emissions' are the emissions used for all of Europe and 'Norwegian emissions' replaces these emissions for traffic and residential heating with alternative emissions used in the Norwegian air quality forecasting system. 'European' and 'Norwegian' proxy downscaling are explained in the text. Calculation year is 2017. The number of available stations is 41, 36 and 44 respectively for NO_2, $PM_{2.5}$ and PM_{10}.

indicate that significant improvements can still be obtained in the downscaling if improved emissions and emission proxies are implemented.

For PM_{10} both bias and spatial correlation are significantly improved with the implementation of the local emissions and proxies. This is to a large extent due to the improvement in the road dust emission contribution but also due to an improvement in the residential heating distribution. Spatial correlation is also significantly increased, from $r^2 = 0.27$ to 0.49.

5.5 Sensitivity to the NO$_2$ chemistry scheme

320 Included in uEMEP are a number of simplified NO$_2$ chemistry schemes, used to derive downscaled NO$_2$ concentrations from NO$_X$ and O$_3$ concentrations. In the results presented so far we have used the weighted travel time parcel method, as applied and described in Denby et al. (2020), with the additional use of the frequency distribution correction described in Sect. 2.4 ("Travel time" in Fig. 16). Two additional chemistry-based schemes and two empirically-based schemes are also available. The two alternative chemistry schemes are the photo-stationary formulation ("Photostationary" in Fig. 16) and an alternative

325 stationary formulation ("Stationary" in Fig. 16) that also allows for deviation from the photo-stationary state (Maiheu et al., 2017). The first empirical scheme is the Romberg scheme (Romberg et al., 1996) ("Romberg" in Fig. 16), also described in Denby et al. (2020), that directly converts NO$_X$ to NO$_2$ concentrations. The parameters for this equation have been updated by fitting to all available Airbase data for the year 2017. The other empirical formulation is the SRM scheme (Wesseling and van Velze, 2014) ("SRM" in Fig. 16) that is also based on a fit to measurement data but includes background O$_3$ as one of the input

330 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 directly, without any correction for non-linearity. All methods are described in Sect. S1.

In Fig. 16 we provide the results of the sensitivity tests, showing bias and spatial correlation for both NO$_2$ and O$_3$. The three chemistry based schemes give similar results indicating that in all three cases the calculations are close to photo-stationary.

335 The two empirical fits also give similar results, with the largest negative bias in NO$_2$ given by the Romberg scheme with -25%. Since the Romberg scheme is specifically designed to reflect measurements, providing the correct NO$_2$/NO$_X$ ratio, it can be regarded as the closest to the measurements. The bias differences between chemistry schemes and the Romberg scheme indicate that chemistry schemes have higher concentrations of NO$_2$ than the Romberg scheme, thus overestimate the NO$_2$ contribution when applied to annual mean concentrations. This is partially due to the positive bias in the EMEP model O$_3$ concentrations of

340 16%, but this only accounts for around 4% of the additional NO$_2$. Included in Fig. 16 is the annual mean calculation without the frequency distribution correction ("Travel time (annual)"), showing a 10% difference in bias when compared to calculations that use this correction. Spatial correlation is also improved by using the frequency distribution methodology.

6 Discussion

Downscaling only applies to emissions within a limited region of $\pm 0.1^o$ surrounding each receptor sub-grid. Based on the

345 uEMEP calculation, the local contributions to NO$_X$ are significantly larger than for PM. The different source contributions at measurement sites are given in Table 2 and this shows that, on average in Europe, 58% of the NO$_X$ contributions come from traffic within this limited region. In contrast only 19% of the PM$_{2.5}$ is attributable to residential heating, the largest downscaled contribution, from inside this region.

NO$_2$ is well modelled with high spatial correlation for many countries, but still with a significant negative bias of -18%.

350 There is significant variation in bias between countries even though the methodology is consistently applied to all countries. This may be attributable to the various methods used for generation of the national emissions. Though the problem remains
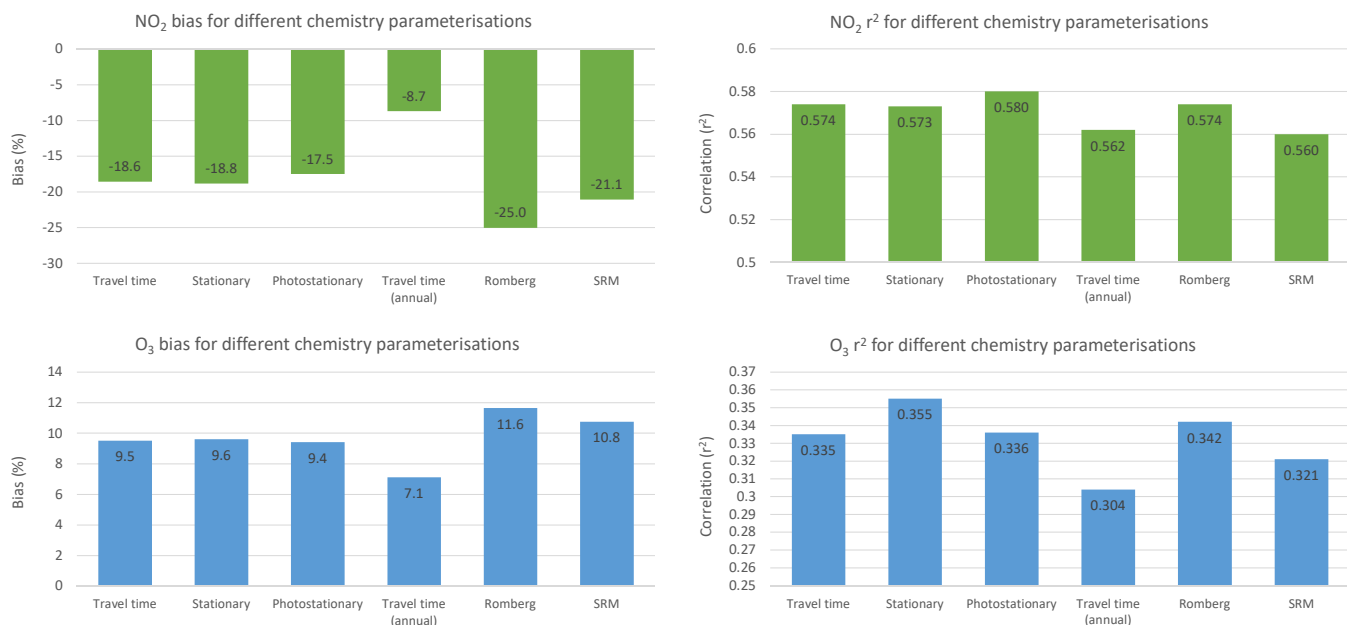
Figure 16. Change in bias and spatial correlation (coefficient of determination r^2) for NO_2 and O_3 with implementation of 6 different versions of the chemistry schemes. See text for details.

Table 2. Source contribution to all air quality stations in Europe calculated with uEMEP. uEMEP local contributions are from primary emissions within a region of 2 x 2 EMEP grids ($\pm\ 0.1^o$) in both latitude and longitude. Non-local EMEP model contributions are all emissions from outside this region, for the downscaled sources, as well as all other primary and precursor emission sources from within this region that are not downscaled.

Source	NO_X ($\mu g/m^3$)	$PM_{2.5}$ ($\mu g/m^3$)	PM_{10} ($\mu g/m^3$)
Traffic (GNFR6)	13.9 (58%)	0.71 (6%)	1.1 (7%)
Residential heating (GNFR3)	1.8 (8%)	2.2 (19%)	2.6 (16%)
Shipping (GNFR7)	0.30 (1%)	0.01 (0.1%)	0.01 (0.1%)
Non-local EMEP	7.9 (33%)	8.4 (75%)	12.3 (77%)
Total	23.9 (100%)	11.3 (100%)	16.0 (100%)

that uEMEP does not take into account dispersion in street canyons, where a number of traffic site measurements are made, it is generally the case that the spatial representativeness of the uEMEP calculations is suitable for comparison with these measurements (Lefebvre et al., 2013). Variation in bias between countries is then no longer a case of a mismatch in resolution

355 but most likely reflects bias in the national emissions. uEMEP may be used to investigate this variability between countries further and to help harmonise future emission inventories across Europe.

There is a significant difference between the results achieved for the downscaling of PM compared to NO_2. NO_2 is dominated by traffic emissions and this is spatially well defined using OSM as a proxy. The largest contributor to PM in the downscaled sources is residential heating, with contributions of 19% and 16% for $PM_{2.5}$ and PM_{10}, respectively (Table 2). This is inline

360 with other estimates of residential combustion in Europe. Thunis et al. (2017) calculated a contribution of 13% from residential combustion from primary $PM_{2.5}$ averaged over 150 European cities, without downscaling. Population is used as a downscaling proxy for the residential source, but it appears that this is not a good proxy for high-resolution emission redistribution. Though clearly residential heating emissions occur where people live there can be large variation from city to city and from urban to suburban and to rural areas as heating practices vary significantly depending on housing type and on availability of alternative

365 heating sources. To some extent this has been taken into account in the emission inventory at 0.1^o, but the emission proxy used in uEMEP is likely not consistent with the EMEP emission inventory.

The Norwegian sensitivity tests show that when consistent emissions and emission proxies are used then spatial correlation can be significantly improved. For the application of uEMEP in Europe this was not the case since each country has their own methodology for calculating gridded EMEP emissions that may or may not make use of the downscaling proxies applied in

370 uEMEP. A more consistent approach, as applied in Norway, would be to use the same spatial redistribution proxies in both the gridded EMEP emissions and the downscaling proxies. This would require additional interaction and cooperation between emission inventory developers and air quality modellers.

It is worth noting that no selection of the Airbase monitoring data was carried out. All available stations with more than 75% coverage were used. This includes mountain stations, all traffic stations as well as industrial sited stations. In comparisons with

375 the EMEP model these types of sites are often removed. All stations were also assumed to be sited at 3 m above the surface. It is quite possible that different results would be obtained if a selection of stations was carried out. This will be assessed at a later time.

7 Conclusions

Downscaling of annual mean concentrations from the EMEP model have been carried out for NO_2, $PM_{2.5}$, PM_{10}, and O_3 using

380 the uEMEP model. Downscaling redistributes EMEP gridded emission data, using suitable proxy data, to high-resolution sub-grids and then calculates the sub-grid concentrations using a Gaussian dispersion model. These are then recombined with the EMEP model concentrations in a consistent way that avoids double counting of the emissions. Maps for all of Europe have been produced at a resolution of 100 m and concentrations at all Airbase measurement sites have been calculated at 25 m.

The results for NO_2 show significant improvement with a doubling of spatial correlation for most countries and a significant

385 reduction in negative bias. For NO_2 the downscaling works very well, which is due to the fact that NO_X emissions are mainly attributable to traffic and these emissions are well defined spatially with the proxy data used. O_3 concentrations are decreased due to higher NO_X concentrations. Both concentrations and spatial correlations of O_3 are better simulated with uEMEP.

Neither $PM_{2.5}$ nor PM_{10} shows any improvement in spatial correlation with the downscaling, though the negative bias in PM concentrations is improved. The spatial distribution of PM emissions can be improved, as demonstrated for Norway, with more accurate proxy data, but emissions of PM remain difficult to quantify properly at high resolutions and will require further effort. One way forward is to harmonise the proxies used for both the EMEP gridded emissions and the uEMEP downscaling. This has been shown to improve results in Norway.

Downscaling can provide additional information concerning the contributions of local sources. This may be combined with the EMEP model source-receptor calculations to provide a more complete picture of local and long-transported contributions. The method can lead to a better assessment of local versus regional mitigation strategies to improve air quality in Europe at high resolution. It also shows good potential to be used to improve exposure estimates.

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Supplementary material for "Downscaling of air pollutants in Europe using uEMEP_v6"

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S1   NO$_2$ chemistry schemes

Included in uEMEP are a number of simplified NO$_2$ chemistry schemes, used to derive downscaled NO$_2$ concentrations from NO$_X$ and O$_3$ 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$_X$ to NO$_2$ 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$_2$ that only considers the NO$_X$ titration of O$_3$ (reaction rate k_1) and the photo-disassociation of NO$_2$ (photo-disassociation rate J) is already presented in Section 2.4. This steady state solution implies

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

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 λ such that

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

For $\lambda < 1$ this implies a sink of NO_2 and for $\lambda > 1$ a source. The steady state solution can then be written, as in Eq. (1), as

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

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_2 concentrations. At the station sites used in this study we find on average $\lambda = 0.95$ with a standard deviation of 0.35. Around 60% of the sites have $\lambda < 1$. This means there is significant divergence from the photo-stationary assumption in the EMEP annual mean calculations. λ is also strongly correlated with NO_2, increasing with increasing NO_2, indicating that emissions are perhaps the dominating factor leading to non photo-stationary equilibrium. It should be noted that the term $[NO_X] + [O_X]$ in Eq. (S3) is usually significantly larger than J/k_1 so even with a significant deviation of λ from unity then the NO_2 concentrations will not be largely affected.

This parameterisation is particularly useful if NO_2 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 λ from unity are equally applicable for the downscaling chemistry.

S1.2 Updated Romberg scheme

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

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

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_2/NO_X emissions, i.e. the asymptotic limit for large NO_X. 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_2 levels provided by EMEP are not altered by this calculation. Background NO_2 and O_3 concentrations are calculated in uEMEP after the removal of the local fraction contribution from NO_X. In Denby et al. (2020) this is referred to as the non-local contribution. To achieve this we rewrite Eq. (S4) as follows

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

where $\Delta[NO_2]_{bg}$ is the difference between the EMEP calculated NO_2 non-local contribution and the Romberg calculation of the non-local contribution given by

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

This ensures that the calculated NO_2 concentrations are unchanged when the local contribution to NO_X is negligible.
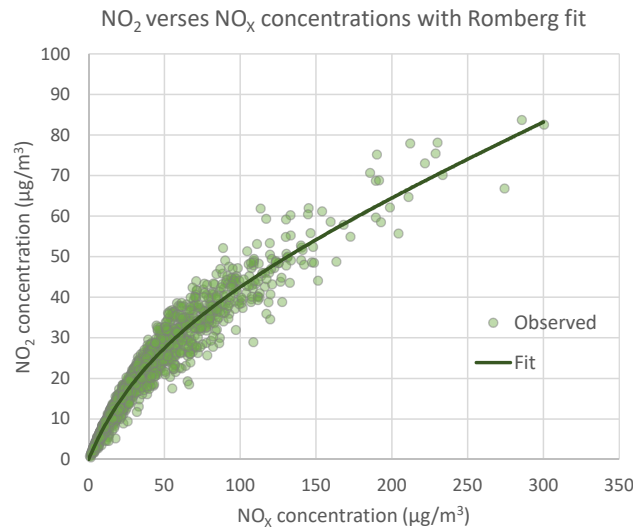
Figure S1. Annual mean NO_2 versus NO_X concentrations for 2017 showing 1774 available Airbase stations. The fit using the Romberg equation is also shown, using the parameters provided in the text.

S1.3 SRM scheme

The SRM scheme for NO_2 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:

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

where

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

The value for c_{srm} applied here is the traffic exhaust NO_2/NO_X emission ratio used in uEMEP, and the value for b_{srm} $(\mu g/m^3)$ comes directly from Wesseling and van Velze (2014). The parameters c_{srm} and b_{srm} are refered to as F and K respectively in Wesseling and van Velze (2014).

This equation clearly resembles the Romberg scheme, Eq. (S4), with the exception that a background NO_2 and a local NO_X 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

A frequency distribution correction scheme for annual mean calculations of NO_2 is described in Section 2.4. This is implemented to deal with the non-linear nature of the NO_2 chemistry when calculating annual means. Here we present some of the background information used to derive this scheme.

An example of the NO_X, O_X 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_X and high O_3.

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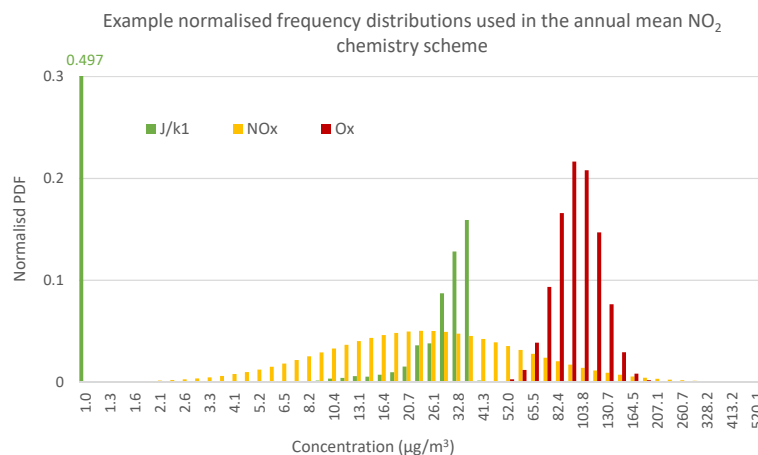
Figure S2. Example of the frequency distribution for NO_X, O_X and J concentrations used in the model. This is taken from the calculation of an actual site in Southern Europe. X axis is logarithmic.

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.
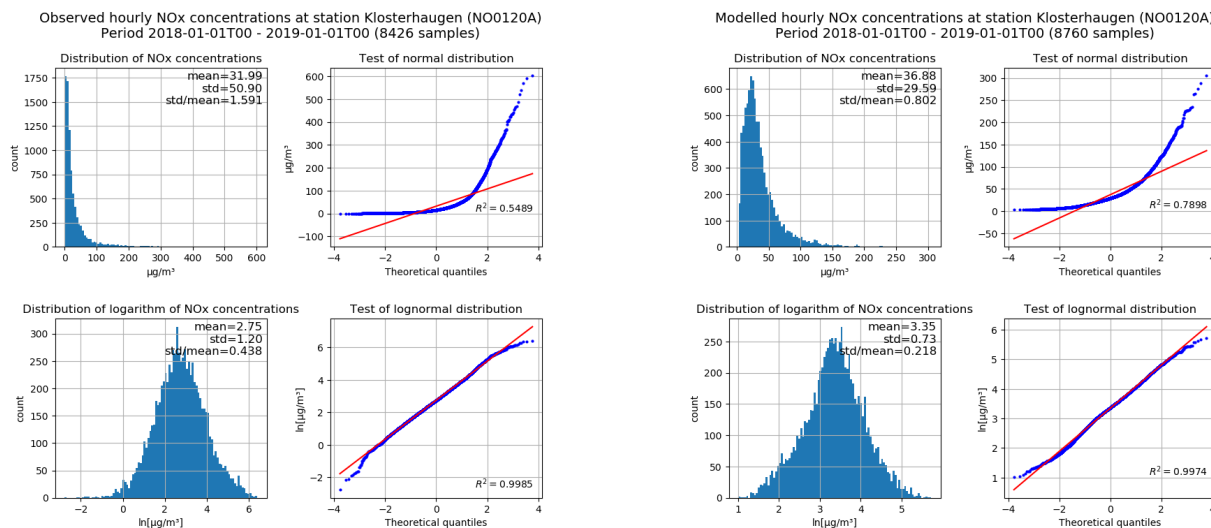

Figure S3. Measured and modelled NO_X distributions for year 2018 at the station Klosterhaugen, Bergen, shown on both a linear and a logarithmic scale.
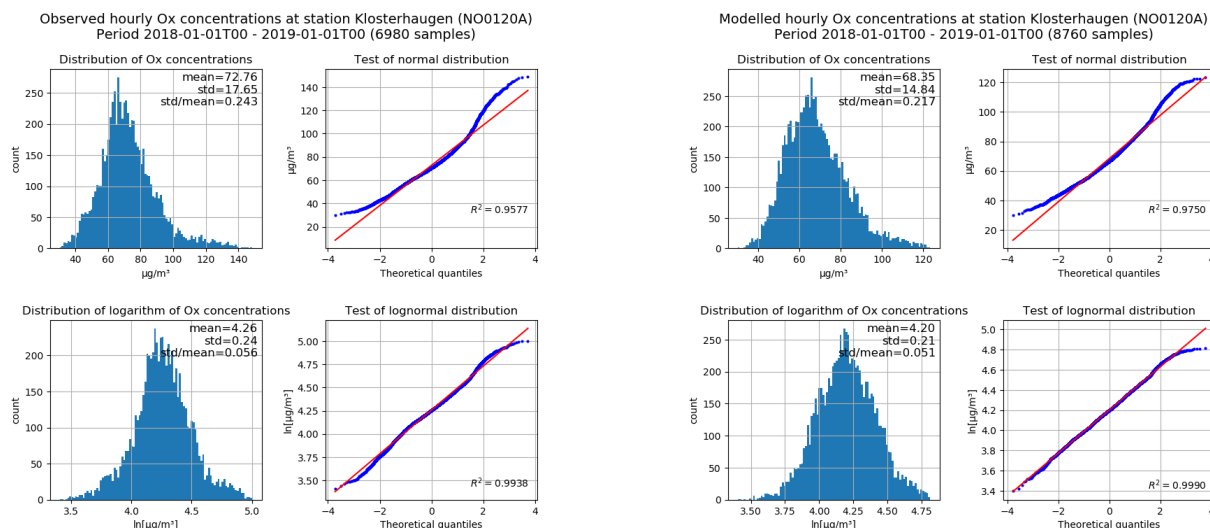
Figure S4. Measured and modelled O_X distributions for year 2018 at the station Klosterhaugen, Bergen, shown on both a linear and a logarithmic scale.

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 relationship for NO_X is very robust and will reflect the temporal variation of the NO_X 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_X.

Finally the correlation between NO_X 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_X and O_X but this would be difficult to take account of in the frequency distribution correction currently implemented in the model.
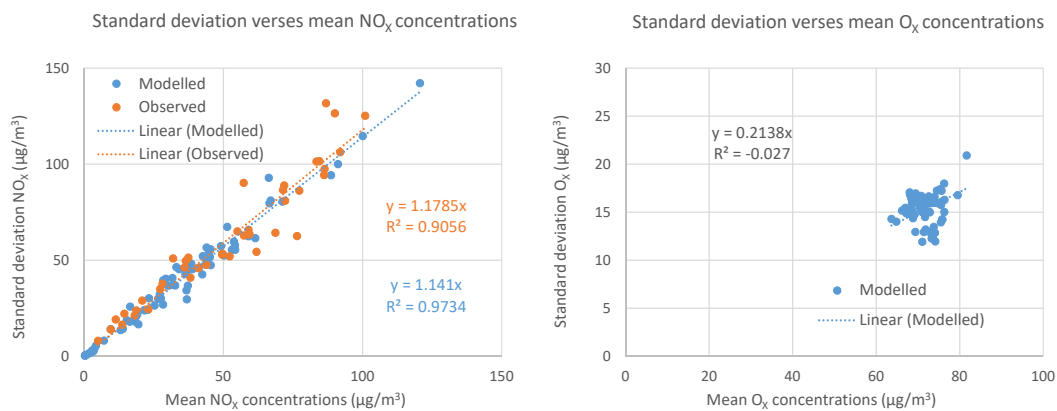
Figure S5. Scatter plot of hourly standard deviation versus annual mean NO_X and O_X for 2018 for all the 72 modelled sites. Also included are measurement data from 41 of the sites that measured NO_X with data coverage >75%. The regression slope, intercept set to 0, and the correlation are also shown in the plots.
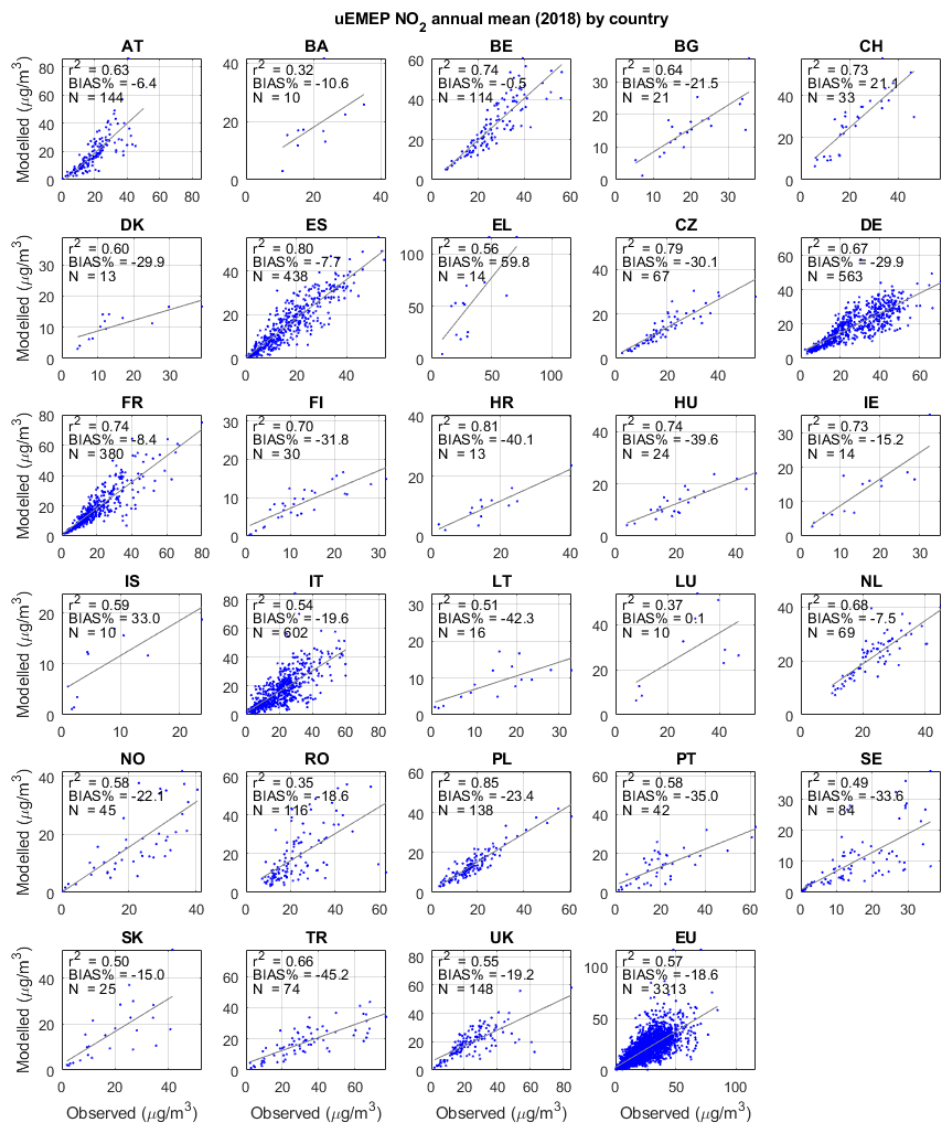
S2   Scatter plots per country

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.
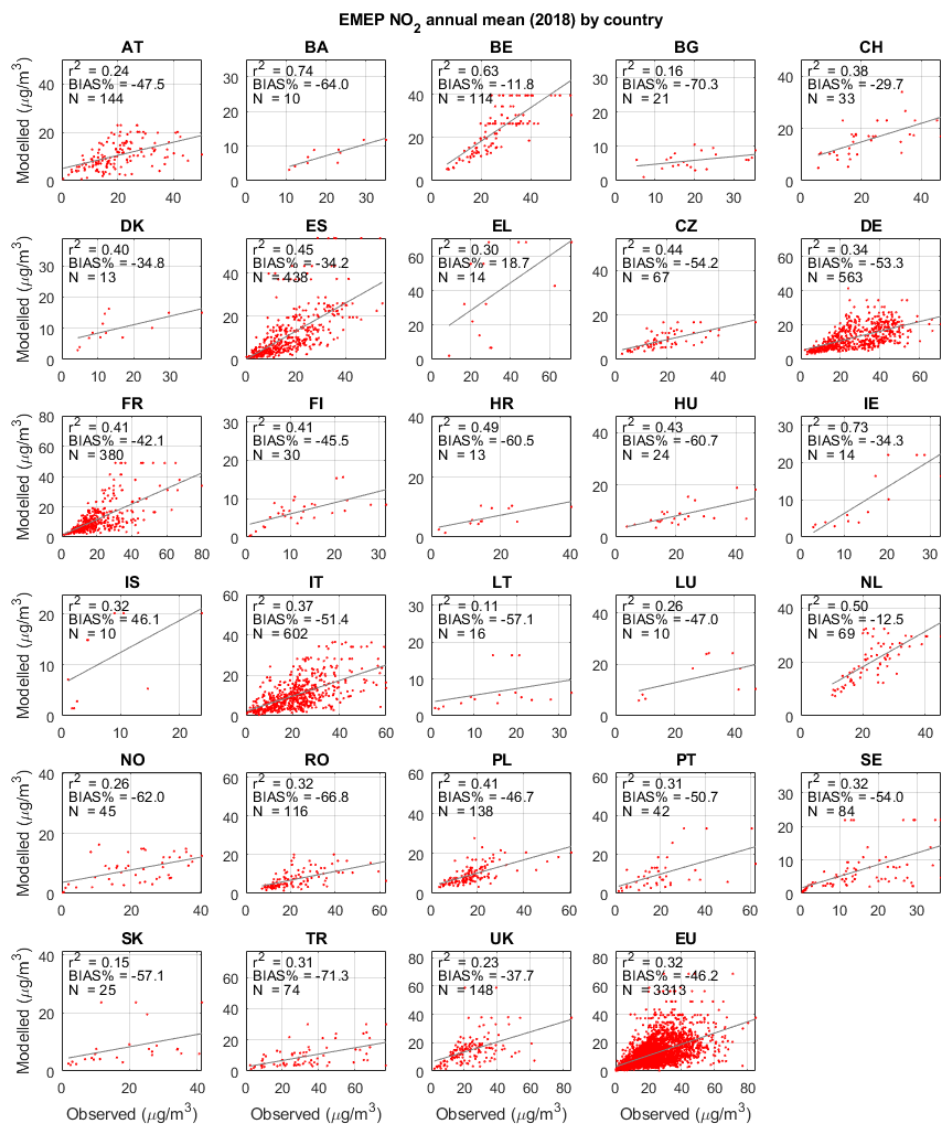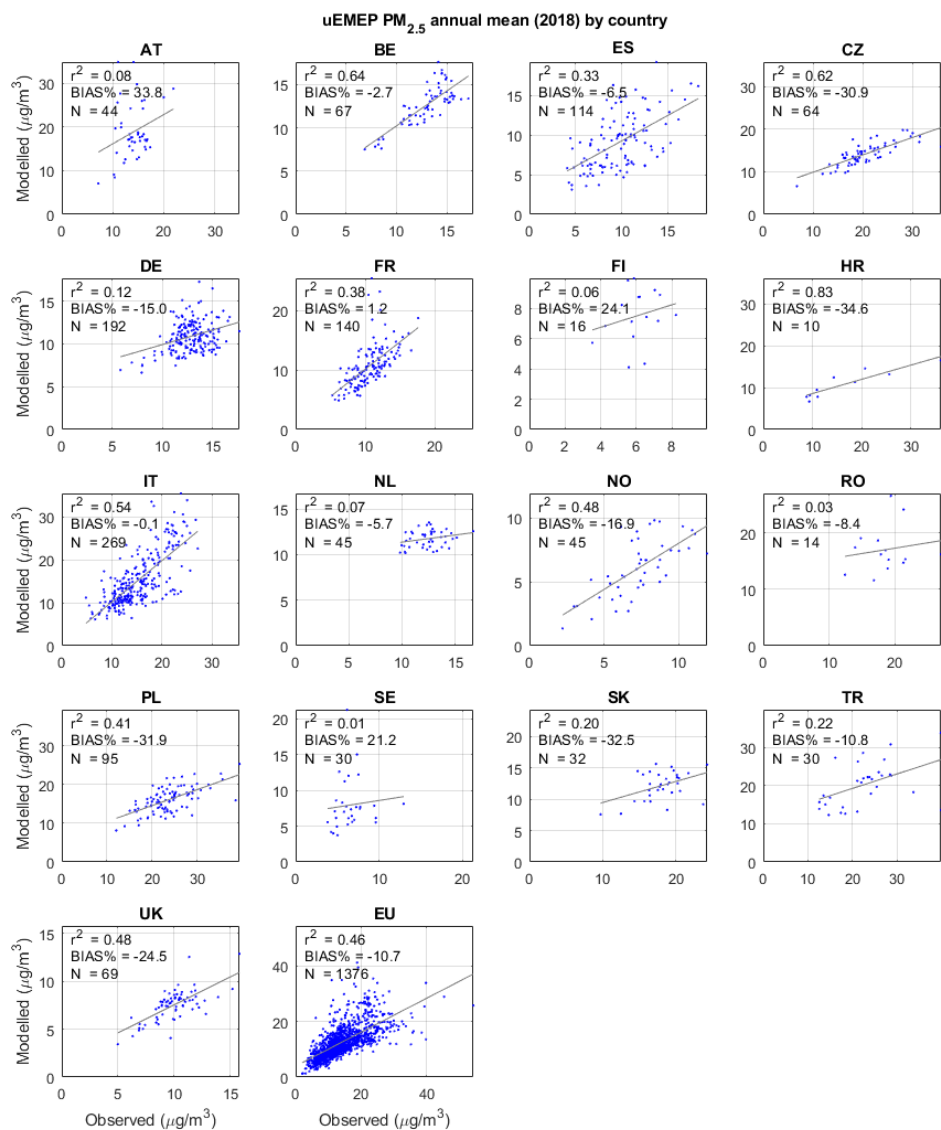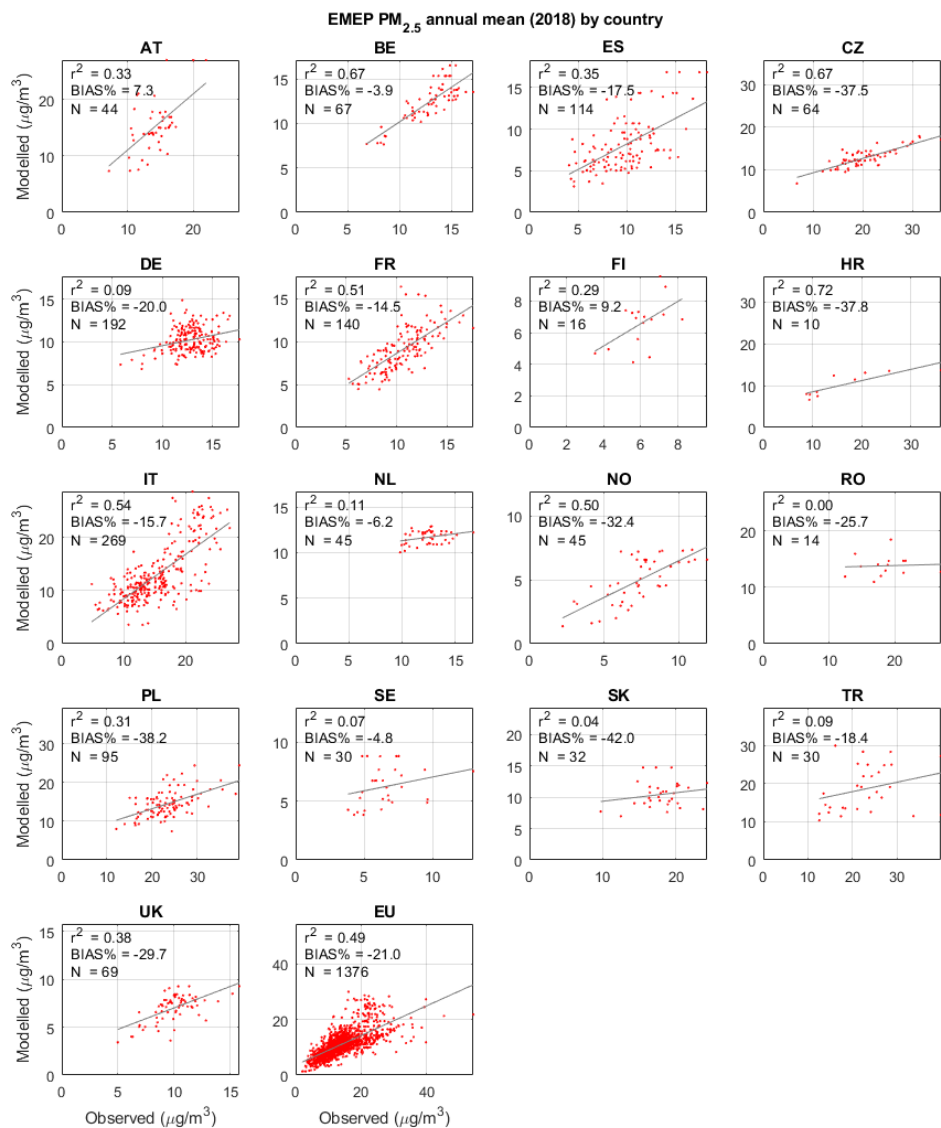
Figure S7. Scatter plots of annual mean NO$_2$ concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.

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.
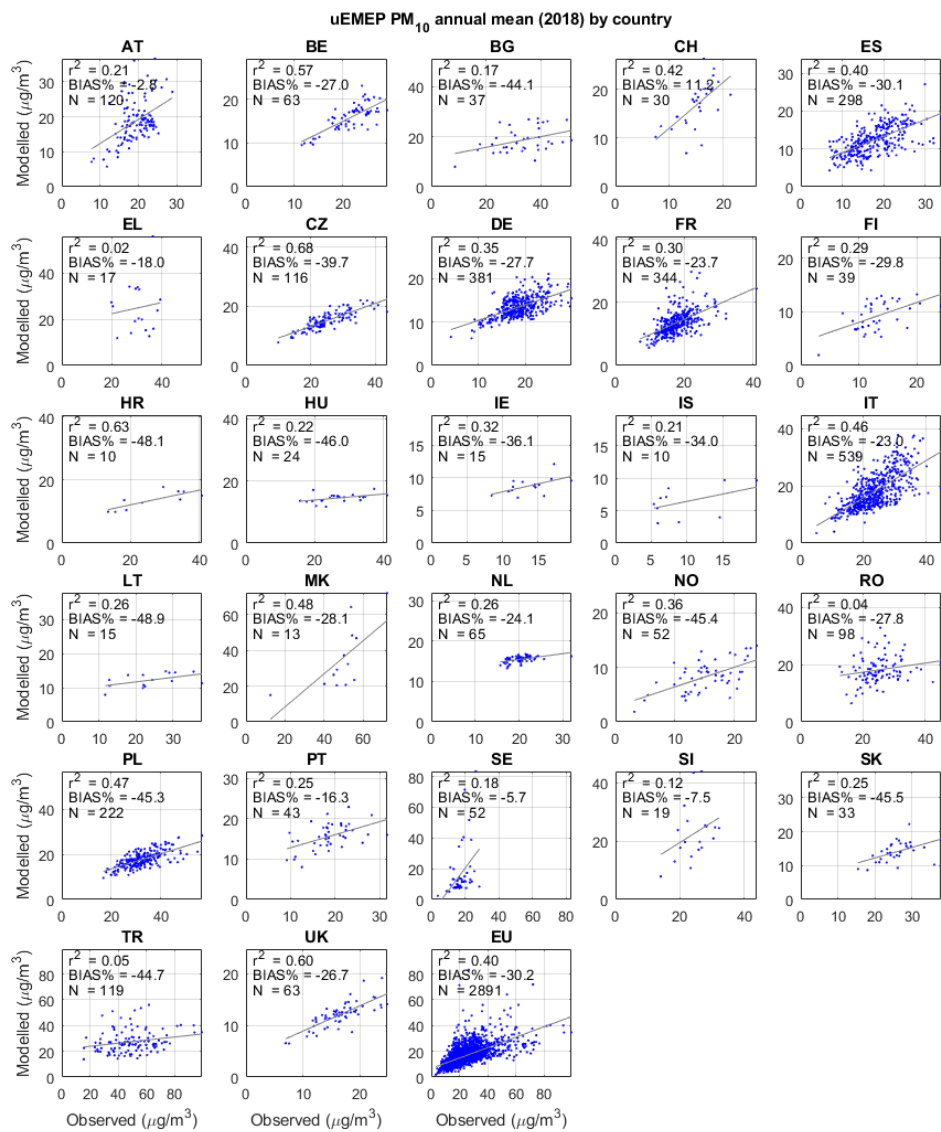
Figure S9. Scatter plots of annual mean PM$_{2.5}$ concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.
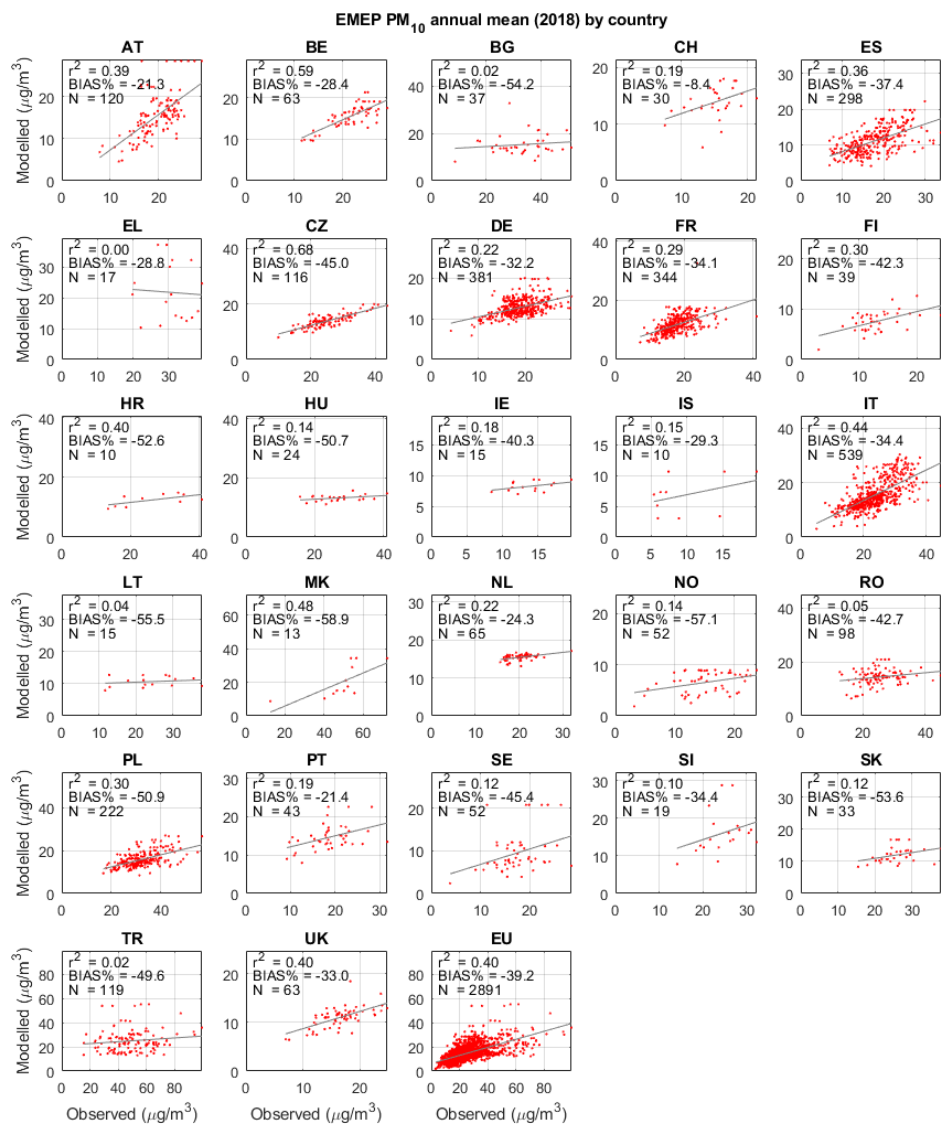
Figure S10. Scatter plots of annual mean PM$_{10}$ concentrations per country for 2018 calculated with uEMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.

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.
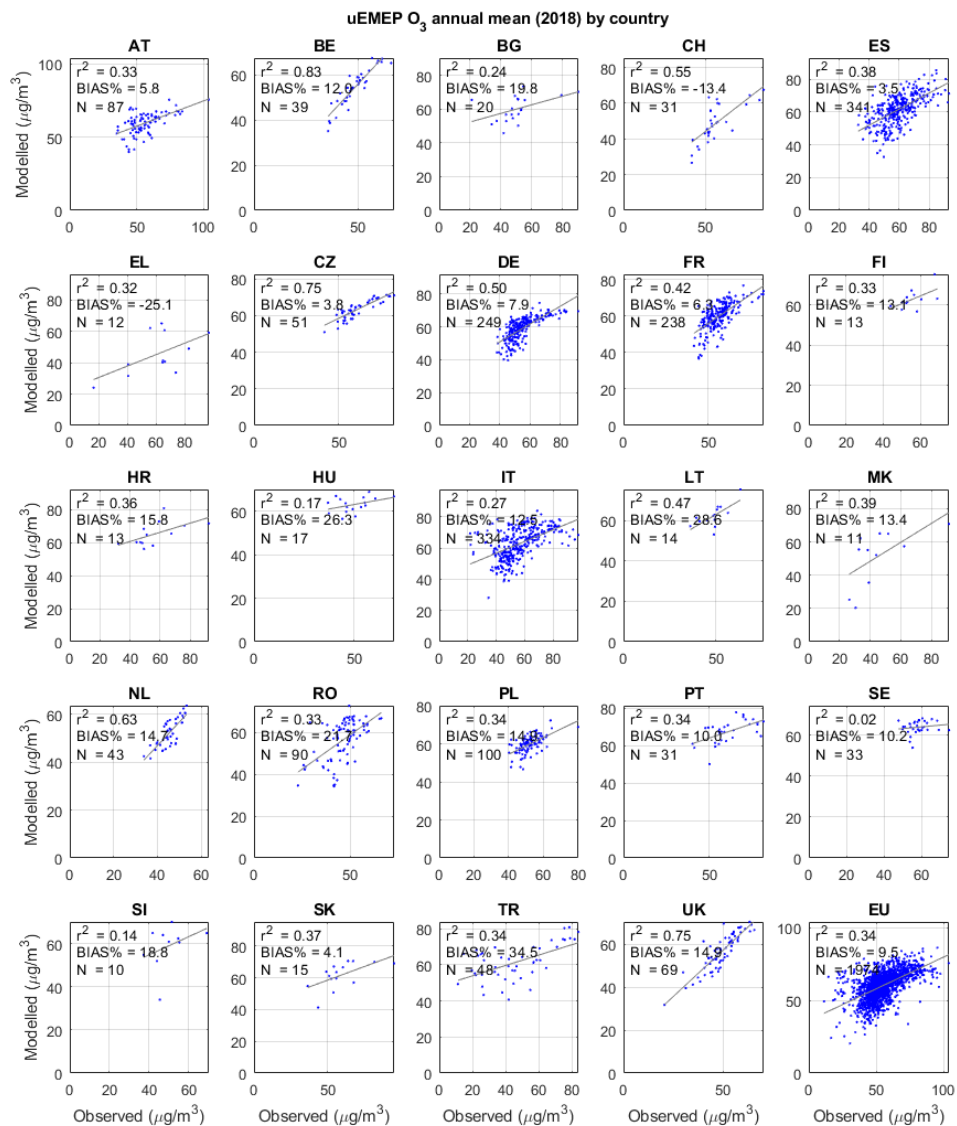
Figure S12. Scatter plots of annual mean O₃ concentrations per country for 2018 calculated with uEMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.
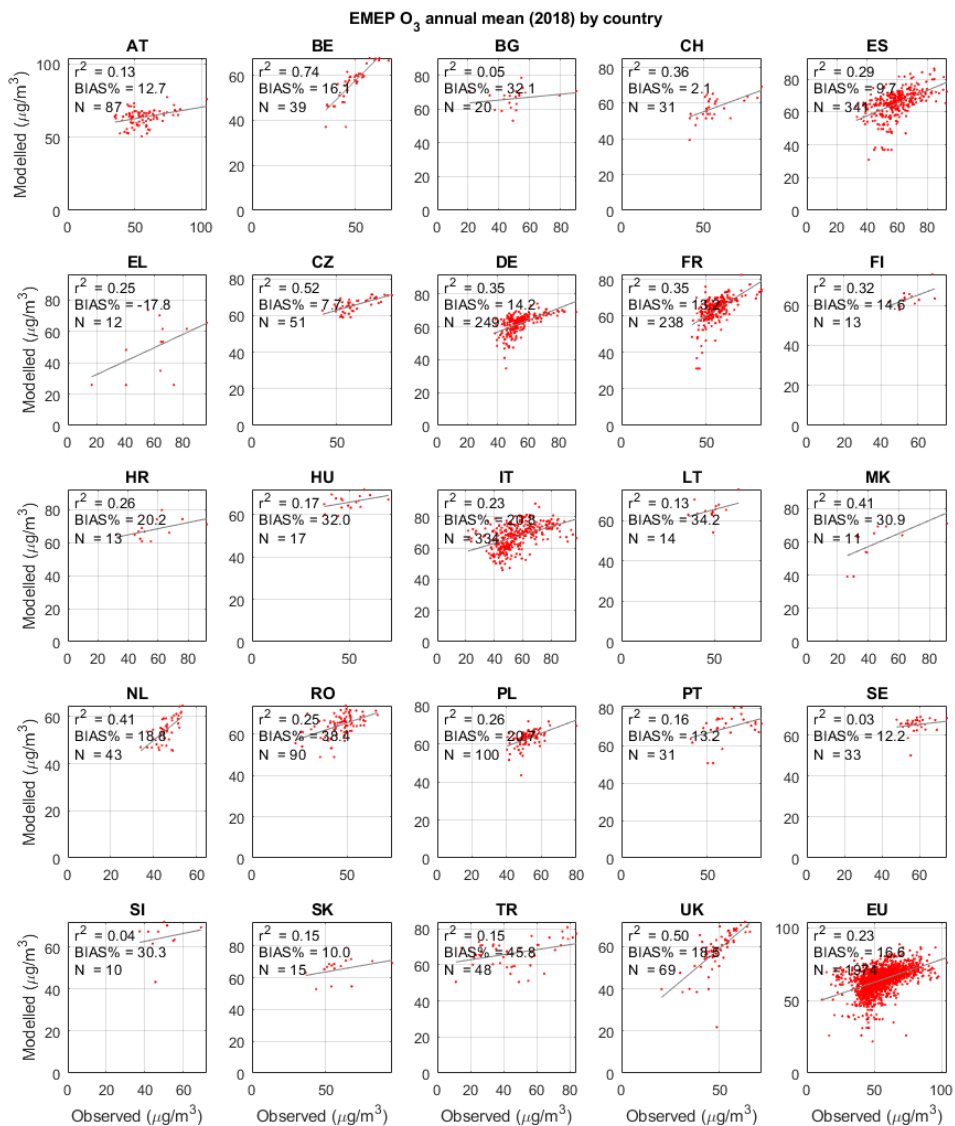
Figure S13. Scatter plots of annual mean O₃ concentrations per country for 2018 calculated with EMEP. Only countries with 10 or more stations are shown individually but all stations are included in the final EU plot.

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