Updated Authors' response to the comments by Anonymous Referees #1 and #2

We would first like to thank both referees for their useful and constructive comments. We have combined the responses to both Anonymous Referees and the responses have been grouped by topic to improve readability. Page and line numbers refer to the manuscript version with Tracked Changes (included at the end of this document).

Approach used

Ref#1: Referring to this technique as 'modeling the sub grid scale variability' is misleading because there is nothing in the formulation of the CTM that has been changed here to actually model the unresolved variability. In my view, what they develop is a post-processing, downscaling technique to map regional scale simulations on finer resolution emission data using some parametrization to account for dispersion of these emissions.

Ref#1: The parametrization of emission dispersion does not include any sub grid process such as fast chemistry or deposition. What is more, the time scale of their application is much coarser than the CTM's (annual vs. several minutes though the time step of the EMEP model is not explicitly mentioned). Those choices make it clear, in my opinion, that this effort is not meant to solve the subgrid variability. To do so, the effort would rather focus on high resolution meteorology and emissions on line with the regional CTM to capture the unresolved features of atmospheric chemistry and dispersion.

Authors' response: *We appreciate the referee's opinion on this matter. We think that it is a question of definitions. The referee's interpretation of the term "sub-grid model" appears to be that of a parameterisation within a coarse resolution model (e.g. a CTM) that explicitly takes into account sub-grid variability during the simulation. We use the term "sub-grid model" to mean a parameterisation that can refine the outputs of a coarse resolution model, by providing an estimate of the spatial distribution of the model output (e.g. atmospheric concentrations) within each grid cell of the coarser model. In this respect we a using the term "sub-grid model" in the same sense as that used by various publications cited in the manuscript (Ching et al., 2006; Denby et al., 2011; Dragosits et al., 2002 and Isakov et al., 2007). The manuscript has been revised to clarify what we mean by sub-grid modelling (see e.g. p 4 lines 13-14).*

Ref#2: I have difficulties understanding why NO2 photochemistry is not needed here. By using the NOx fine-scale emission pattern and distribution kernel for redistributing NO2 concentrations this approach assumes a constant NO2/NOx concentration ratio in the whole EMEP 50km grid cell, which is hard for me to believe. Typically this ratio should show a dip close to NOx sources due to the high NO ratio in primary emissions. Also this fraction of primary NO2 in NOx emissions varies strongly between different sources and this should play a role at the local scale. I thought that at least ADMS includes a NOx photochemistry formulation, why is this not used? Ideally such a chemistry scheme should be used, or at least the potential errors discussed. Along similar lines, also the formation of NH4 can influence local NH3 concentrations – it would be interesting to know what the potential errors are when these processes are ignored.

Authors' response: *The assumption of a constant NO_2/NO_x concentration ratio is made to provide a simple parameterisation that is universally applicable. It would be difficult to include a variable ratio in a simple model since it*

would depend on the local pollution climate (e.g. ozone concentrations), photochemical reactions of NO and NO_2 emitted from the different source types and complex variations in diurnal emission patterns and meteorological drivers. Both AERMOD and ADMS include optional simple photochemistry schemes but they depend on background ozone concentrations, which are not homogeneous across the domains, and so these model options cannot be used for developing a simple generic model, which was the aim of the study.

Of course this simplification will introduce uncertainty into the model. This uncertainty can be estimated by analysing the variability of the NO_2/NO_x ratio in the measured data. For the Scottish domain, the coefficient of variation of the ratio is 20% whereas for the Dutch domain it is only 7.5%. Estimating the NO_2 concentrations from the measured NO_x concentrations, assuming a constant ratio, gives mean errors (NMGE) of 18% and 6.3%, for the Scottish and Dutch domains, respectively. Extending this analysis to the annual mean concentrations for the sites in the European air pollution database "airbase" that simultaneously measure NO and NO_2 (1478 sites), gives a coefficient of variation of 17.5% and a mean error of 14.6%. Based on the European dataset, we estimate that the mean error introduced by assuming a constant NO_2/NO_x ratio is about 16%. A discussion of these uncertainties has been included in the Discussion section (p 12 lines 23-28).

Similarly the formation of NH_4^+ from NH_3 depends on the concentrations of other pollutants, as well as the complex variations in diurnal emission patterns and meteorological drivers, the inclusion of which is beyond the scope of a simple sub-grid model. However, a discussion of the uncertainty due to this omission has been included in the Discussion section (p 12 line 28 to p 13 line 1).

Ref#1: How would results look like if no dispersion was taken into account and the same process was done only by using the 1km (or 7km or both) emission proxy?

Authors' response: *We have now included these analyses using the 1 x 1 km^2 emission data for both NO_2 and NH_3 (p 10 lines 6-15 and Figure 6). As now mentioned in the discussion, these results show that short-range dispersion estimates are necessary for improving on the EMEP model predictions.*

Meteorological data

Ref#2: I find it hard to understand why meteorological data from just one station was used here, instead of grid-specific meteorology consistent with the fields driving the CTM. Even the two "domain-specific" meteorological data sets come only from one station each. I think this needs to be better justified and compared to the effects a grid specific met data set would have.

Ref#2: Since the authors mention this as a possibility I would suggest that they add a sensitivity analysis using grid-specific meteorology consistent with the CTM

Ref#1: Wouldn't it make more sense to use the same meteorology as in the EMEP model at least for this sensitivity test?

Authors' response: *The model was developed using a single meteorological dataset to provide a simple parameterisation that could be of benefit to the wider air quality modelling community. The model was tested with meteorology specific to each modelling domain to show that the model results were not that sensitive to the meteorological data set used. However, we agree that it makes more sense to use the same meteorology as the EMEP model and we have done this for the revised manuscript. The original approach using the modified meteorological data from just one station is now included*

as a sensitivity test and is now referred to as the simulation using "synthetic meteorological data". As can be seen in Figure 5, the use of the EMEP model meteorology does not make a large difference to the concentration predictions and model performance is similar, strengthening the conclusion that the approach is fairly insensitive to the meteorological dataset used. The manuscript has been revised to reflect this change in methodology (see e.g. p5 lines 5-12; p 7 lines 7-8 and lines 17-19).

Ref#2: Why is it justified to assume rotationally symmetric dispersion kernels, should not the local predominant wind direction have a significant influence? How big is the error introduced by this rotationally symmetric formulation?

Authors' response: *In order to keep the model parameterisation simple and universally applicable, we decided to remove the influence of local wind direction distributions (which are not valid for the whole domain) by assuming rotationally symmetric dispersion kernels. It can anyway be noted that in the real world local wind-directions will differ from that of a larger scale model. For example, cities are frequently located in locations subject to topographic (e.g. valley) winds, or sea-breezes.*

Although this rotation symmetry obviously does introduce some error, the results shown in Figs. 4 and 5 demonstrate that even with this assumption the sub-grid model provides a valuable improvement over that of the larger scale CTM. The use of EMEP model meteorology, however, now relegates this approach to that of a sensitivity test.

Emissions

Ref#1: I am wondering what do emission sources as large as 1km2 could possibly represent. In my understanding, dispersion models are conceived to represent emission from point sources such as industrial stacks. Is this the right model to represent large area sources such as crops or residential emissions? Is this type of modeling adequate to represent dispersion around busy roads? Don't dispersion patterns depend on the emission sector?

Authors' response: *The use of 1 km^2 emission sources is a simplification due to the fact that we do not have emission inventories for the study domains at a higher spatial resolution. So in reality we may have a large point source within a specific 1 x 1 km^2 grid cell but since we do not know exactly where, we distribute this emission over the whole grid cell. Of course this is a simplification which is going to introduce uncertainty in the model but it is the best we can do with the available emission data. The model, therefore, would not be expected to be suitable for estimating concentrations close to busy roads but the model performs well for the Dutch traffic sites. This has been made clearer in the revised manuscript (p 13 lines 5-11).*

Averaging period

Ref#1: It would be interesting to look at the effect of the meteorological dataset at a finer time-scale. Especially since the authors claim in their conclusions that this method is easily applicable at finer time scales.

Authors' response: *As stated in the manuscript, the annual mean limit values for NO_2 are generally more stringent than the hourly ones and impacts of NH_3 are only assessed using annual mean concentrations. We agree that it would be interesting to develop a model with a higher temporal resolution, but such a study would raise other issues (e.g. as discussed above, or*

with the availability and accuracy of time-resolved emissions). Thus, we did not consider this to be 'easily applicable' at finer time-scales, but rather, as we state in the discussion, this is a potential future improvement and is out of the scope of the presented work.

Model evaluation

Ref#1: I don't think it is appropriate to say that "the sub grid model performed better than the EMEP model". It would be more fair to say that the downscaled version of the
EMEP model compares better with observations.

Authors' response: *This is a fair point and the manuscript has been modified accordingly.*

Ref#2: Does this mean the downscaling decreased agreement to NO2 non-traffic stations, which are the ones we would actually expect it to improve? As mentioned above, I would not expect the 1x1km model to be representative of traffic stations.

Authors' response: *You would expect the sub-grid model to perform best for the non-traffic stations but for the Dutch domain it had the lowest bias and error for the traffic sites. This suggests that the model may be suitable for simulating concentrations at traffic stations, providing they are located several metres from the roadside (as is the case for most of the Dutch sites), and the good agreement is likely the result of the fact that the majority of roadside sites are not situated along isolated motorways, but rather embedded in urban areas with dense road networks. This is now highlighted in the Discussion (p 13 lines 8-10).*

Manuscript structure

Ref#2: I would find it helpful if the downscaling equations were written out as equations and not only described verbally

Ref#2: Why moving window? Is the final subgrid distribution pattern at a given 1km grid cell not simply made up from the sum of all emissions times kernel within the range of the kernel? Perhaps a better formulation could be to say that the kernels are not cut off at the EMEP grid boundaries (I hope). Again, formulating this with equations would help here.

Authors' response: *This is a good suggestion and the downscaling equation is now included in the Model Development section. The dispersion kernels are not cut-off at the EMEP grid boundaries. This is now clarified in the description of the downscaling equation (p 7 lines 26-27).*

Miscellaneous comments

Ref#1: The inevitable question arises of whether a direct EMEP run at 7km resolution with its corresponding meteorology would bring about the same improvement as the downscaling developed in the present study. And in this case the data would be directly at hourly resolution.

Authors' response: *Although it is currently possible to run the EMEP model at a 7 km resolution for specific projects, they are far too CPU intensive for routine use, and especially where CTMs need to be run 10s-100s of time for emission control assessments. However even at a spatial resolution of 7 km, the EMEP model would not be able to resolve the large*

horizontal concentration gradients found close to sources and a sub-grid treatment may still be necessary. This is now clarified in the Introduction (p 3 lines 15-16).

Ref#2: One issue that is not addressed here nor anywhere else in the manuscript is that (to my understanding) the lowest vertical layer of the EMEP model extends from the surface to about 90 meters. In this context, it would be important to explain what is meant by 'the mean atmospheric concentration in each grid square': Is this the estimated surface concentration calculated by applying some standard vertical distribution? But if so, is it then justified to assume that the modelled mean concentration on the 50km grid is 'correct' and just needs to be re-distributed spatially within the grid cell?

Authors' response: *The EMEP model estimates the near surface concentrations by extrapolating the concentration at the mid height of the first vertical layer (45 m) assuming an approximately constant vertical deposition flux. Of course, there are uncertainties in this procedure, especially for NH_3 where deposition velocities can be relatively high, but where bi-directional exchange complicates even the sign of atmosphere-biosphere exchange in real situations. However, we have to work with the EMEP values as provided by the model, and our working assumption is that the errors in the vertical distribution are less important than the errors we seek to minimise – namely those of sub-grid horizontal variability. This is now clarified in the Introduction (p 2 lines 20-22).*

Ref#1: If the mean value is correct it would be surprising that the urban background concentration is underestimated. It would make sense to say that near-sources concentration levels are under predicted but if the background value is off as well, I don't see how we could get the mean value right.

Authors' response: *Although this comment refers to the conclusions stated by Denby et al. (2011), we will attempt to respond to the referee´s comment. Both rural and urban areas can be present within a 50 km grid square of the EMEP model and so it is possible that the model predicts the correct mean value for the entire grid square but underestimates concentrations within the urban area (at both traffic and background locations). This would be the case, for example, if the rural area occupies the majority of the grid square.*

Ref#1: If I think that the comparison with Denby et al., 2011 study is off mainly because they worked on hourly data and not annual.

Authors' response: *We disagree with this comment because the study of Denby et al. (2011) is based on annual mean concentrations.*

Ref#1: If I think that the correlation in Schaap et al., 2015 is on time and not in space as in the present study.

Authors' response: *We are referring to Figure 7 of Schaap et al. (2015), which is included in the section on spatial analysis (although it appears to be erroneously referred to as "temporal analysis" in the figure caption).*

Ref#2: In case of PM10 to my knowledge the limit on daily means (35 exceedances of 50ug/m3) is the more stringent one.

Authors' response: *Yes the referee is correct, for PM10 the limit for daily means is the more stringent one. The reference to PM10 has been removed from the revised manuscript (p 12 line 17)*

Ref#2: I wonder whether the quotes are really necessary here.

Authors' response: *Maybe not for the "moving window" but we think that the quotes necessarily highlight the new term "sub-grid distributions" in order to distinguish it from the term "sub-grid concentrations".*

Ref#2: SO4-2 should be SO-24?

Authors' response: *Yes, that is correct and the change has been made.*

Ref#2: Figure 4: I would suggest to split up the two regions into separate figures, since the model performance is so different.

Authors' response: *We agree and the figures for the individual domains have now been included in the supplementary material.*

Additional changes to the manuscript

In addition to the changes mentioned above, the following additional modification has been made to the manuscript:

- *The model name and version has been included in the manuscript title, as per journal guidelines and the model name and acronym used throughout the manuscript*

~~A sub-grid model for improving~~Improving the spatial resolution of air quality modelling at a European scale – Development and evaluation of the Air Quality Re-gridder Model (AQR v1.1)

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Abstract. Currently, atmospheric chemistry and transport models (~~CTMs~~ACTMs) used to assess impacts of air quality applied at a European scale lack the spatial resolution necessary to simulate fine-scale spatial variability. This spatial variability is especially important for assessing the impacts to human health or ecosystems of short-lived pollutants, such as nitrogen dioxide (NO_2) or ammonia (NH_3). In order to simulate this spatial variability, ~~a sub-grid~~the Air Quality Re-gridder (AQR) model has been developed to estimate the spatial distributions (at a spatial resolution of 1 × 1 km^2) of annual mean atmospheric concentrations within the grid squares of ~~a CTM~~an ACTM (in this case with a spatial resolution of 50 × 50 km^2). This is done as a post-processing step by combining the coarse resolution ACTM concentrations with high spatial resolution emission data ~~with~~and simple parameterisations of atmospheric dispersion. The ~~sub-grid~~AQR model was tested for two European sub-domains (the Netherlands and central Scotland) and evaluated using NO_2 and NH_3 concentration data from monitoring networks within each domain. A statistical comparison of the performance of the two models shows that ~~the sub-grid model represents~~AQR gives a substantial improvement on the predictions of the ~~CTM~~ACTM, reducing both mean model error (from ~~60~~61% to ~~40~~41% for NO_2 and from 42% to ~~26~~27% for NH_3 and increasing the spatial correlation (r) with the measured concentrations (from 0.0 to 0.~~42~~39 for NO_2 and from 0.74 to 0.~~85~~84 for NH_3). This improvement was greatest for monitoring locations close to pollutant sources. Although the model ideally requires high spatial resolution emission data, which is not available for the whole of Europe, the use of a Europe-wide emission dataset with a lower spatial resolution also gives an improvement on the ~~CTM~~ACTM predictions for the two test domains. The ~~sub-grid~~AQR model provides ~~a simple~~an easy to use and robust method to estimate sub-grid variability that can potentially be extended to different time scales and pollutants.

Keywords: Air quality; Impacts; Atmospheric chemistry and transport models; Sub-grid variability

1 Introduction

The impacts of air pollution on human health and natural ecosystems are often evaluated using data from atmospheric dispersion models or atmospheric chemistry and transport models (~~CTMs~~ACTMs). The scale of these evaluations ranges from local assessments with domains of several kilometres (e.g. Dragosits et al., 2002, Aggarwal and Jain, 2015, Galvis et al., 2015) to global assessments using grid cells of 1–10 degrees (see for example Dentener et al., 2006). The spatial resolution used in these assessments depends on many factors, including availability of input data, model assumptions, receptor type (e.g. people, forests, etc~~).~~.) and computation time. Many of the impact assessments at a European scale are carried out using atmospheric concentration or deposition predictions of the model developed by the Meteorological Synthesizing Centre-West (MSC-W) of the European Monitoring and Evaluation Programme (EMEP). The EMEP MSC-W model (Simpson et al., 2012), called the EMEP model hereafter, has commonly been applied for policy purposes at a spatial resolution of ca. 50 × 50 km^2 (e,g. Fagerli and Aas, 2008, Simpson et al., 2006). Although the model is increasingly used at even finer resolution (e.g. 0.1 × 0.1 degrees) even for official MSC-W purposes (EMEP, 2015), such runs are extremely CPU intensive for European-scale modelling, and cannot be used for the 100s–1000s of simulations required by the source-receptor matrices, which are an important output of MSC-W (EMEP, 2015). EMEP model results also underpin the Greenhouse gas - Air pollution Interactions and Synergies (GAINS) model, which is a key tool in developing European policy within both UN-ECE and the European Union (Amann et al., 2011). However, the resolution of the EMEP model (or any other European scale ~~CTM~~ACTM), at least when run in typical policy mode, is not currently high enough to resolve the large horizontal concentration gradients found close to sources of relatively short-lived pollutants, such as ammonia (NH_3), nitrogen dioxide (NO_2) or sulphur dioxide (SO_2) (CLRTAP, 2014; Denby et al., 2011).

The EMEP model predicts the mean near-surface atmospheric ~~concentration~~concentrations within each grid square~~.~~ assuming a constant deposition flux between the centre of the first vertical layer (ca. 45 m) and a height of 3 m (Simpson et al., 2012). However, within a grid square there may be concentrations an order of magnitude (or more) above and below this mean value, even if the mean prediction is correct. Neglecting this sub-grid variability (SGV) can strongly bias assessments of air pollution impacts. For example, Denby et al. (2011) estimated that urban background exposure to NO_2 is underestimated by an average of 44% when the 50 × 50 km^2 grid concentrations of the EMEP model are used. This problem is not restricted to the low grid resolution used by the EMEP model, it also occurs in assessments with higher resolutions. For example, Hallsworth et al. (2010) used ~~a CTM~~an ACTM to estimate NH_3 concentrations in the UK at spatial resolutions of 5 × 5 km^2 and 1 × 1 km^2. They found that the 5 km model estimated that the NH_3 critical level of 1 ug m^{-3} was exceeded for 40% of the total area of UK Special Areas of Conservation (SAC), whereas the 1 km model estimated an exceedance for only 21%. This reduction in the area of exceedance when the model resolution was increased was due to the ammonia sources (agricultural areas) and the SAC being separated spatially. Modelling at a higher resolution resolved the large horizontal concentration gradients better, thus predicting higher concentrations in the agricultural areas and lower concentrations within the SAC. By contrast, Oxley and ApSimon (2007) found that increasing the model spatial resolution

from 50 km to 5 km and from 5 km to 1 km increased the estimates of exposure to primary particles with a diameter of 10 µm or less (PM_{10}) in urban areas. This is because, in this case, the urban areas are also some of the largest sources of primary PM_{10}. A multi-model study involving five ~~CTMs~~ACTMs to simulate pollutant concentrations across Europe found a large increase in annual mean concentration predictions of PM_{10} and NO_2 in urban locations when increasing the spatial resolution through the range 56, 28, 14 and 7 km (Cuvelier et al., 2013; Schaap et al., 2015). For most of the models, about 70% of the model response to the change of resolution was due to the change in the spatial distribution of emissions. By comparing the concentration predictions in urban areas with measured values, model performance (slope, bias and correlation) was generally found to improve for all models as the resolution was increased. In order to resolve the large horizontal concentration gradients found in urban areas, Cuvelier et al. (2013) suggested that a resolution of a few km down to one km would be needed, but added that this is not currently feasible for application across Europe. However, even this might not be sufficient for resolving the large horizontal concentration gradients of NO_2, for example.

Several potential methods could be used to estimate the SGV of the concentration predictions of short-lived air pollutants across Europe. Firstly, the EMEP model could be applied at a higher resolution. This has been done in the UK for a resolution of 5 × 5 km^2 (EMEP4UK) (Vieno et al., 2010; Vieno et al., 2014), and for Europe at ca. 7 × 7 km^2 (Schaap et al., 2015, EMEP, 2015), but such runs are extremely CPU demanding~~.~~ and are not suitable for routine use, especially where ACTMs need to be run 10s-100s of times for emission control assessments, for example. A European application at 1 × 1 km^2 resolution or higher is currently not feasible~~. Such~~, even for research purposes. As well as being too demanding on computation time, such runs would also require a consistent and accurate high resolution emission dataset, which is not currently available. A second solution is the 'stitching together' of national modelling simulations at a high resolution (see, for example, de Smet et al., ~~(~~2013~~);~~ Janssen and Thunis, 2016). This approach has the advantage of making use of national expertise and emission and meteorological datasets. However, the disadvantages are that it is likely to lead to 'border effects' as a result of differing methodologies and/or input datasets used by neighbouring countries and results may not be available for all countries, making it difficult to carry out a consistent assessment for the whole of Europe. The third solution is to apply geo-statistical techniques to the low resolution concentration data (e.g. from the EMEP model) that makes use of other relevant spatial datasets. These techniques can be used to either estimate the probability distribution of the concentration (or a related quantity) within each grid square or to explicitly estimate the spatial distribution of the concentration within the grid square. An example of the former approach is that of Denby et al. (2011) who estimated the population-weighted concentrations of NO_2, PM_{10} and O_3 within each EMEP 50 × 50 km^2 grid square using information on measured concentrations and their covariance with population density, that was then parameterised using emission and altitude data. Another example is the SGV parameterisation of Ching et al. (2006) for the CMAQ model based on sub-grid concentration distributions of benzene and formaldehyde calculated using the ISCST3 short-range dispersion model. The same CMAQ simulations were used by Isakov et al. (2007) to develop a method to explicitly model the sub-grid spatial distributions of concentrations at a resolution of 200 × 200 m^2. Their method used relationships between the sub-grid concentrations and sub-grid emission strengths derived from short-range dispersion modelling results, although it was only

applied to a small area (Philadelphia County). A different geo-statistical approach was used by Janssen et al. (2012), in which they estimated sub-grid concentrations for Belgium by using empirical relationships between long-term atmospheric concentrations and land use characteristics. A Europe-wide approach was developed for NO_2 and particulate matter by Kiesewetter et al. (2013 and 2014), although only at a resolution of 7 × 7 km^2. In their work, concentrations simulated by the EMEP model at a resolution of 28 × 28 km^2 were disaggregated using an 'urban increment'. This increment was calculated from the concentration predictions of the CHIMERE model (Bessagnet et al., 2004) at a resolution of 7 × 7 km^2. The relationship between the differences in the concentration predictions of the two models and the emission rate (from near-ground-level sources only) used for each 7 km grid square was used to calculate the urban increment. Model evaluation using annual mean concentrations from more than 1500 urban background monitoring stations showed that the model can predict concentrations within a factor of two of the measured value for most locations. The authors also developed a parameterisation to estimate the additional concentration increment at the locations of roadside air quality stations, although this approach relies heavily on measurement data.

In this paper we present the development, testing and evaluation of a simple ~~sub-grid model~~geo-statistical post-processing methodology (the Air Quality Re-gridder (AQR) model) that combines high-spatial-resolution emission data and a simple parameterisation of short-range dispersion to estimate the spatial distribution of concentrations of short-lived pollutants within the EMEP model grid squares. ~~The~~This sub-grid model is used to calculate the annual mean concentrations of NO_2 and NH_3 for 2008 at a resolution of 1 × 1 km^2 for two test domains (central Scotland and the Netherlands) and evaluated using monitoring network data from within the two domains. Section 2 provides information on the methods and datasets used and Section 3 describes the model development process. Section 4 presents the results of the sub-grid modelling, model evaluation and an analysis of the sensitivity of the model to some of the parameters and datasets used whilst Section 5 discusses model performance, its applicability, uncertainties and potential improvements and extensions.

2 Materials and Methods

The two domains used in this study are central Scotland and the Netherlands (Fig. 1). These domains were chosen because they provide a contrast between a built-up, industrialised and agricultural region (the Netherlands) and a region with both large cities, intensive industrial and agricultural areas as well as more extensively used or semi-natural areas (central Scotland). Both domains also have NH_3 and NO_x emission inventory data at a ca. 1 × 1 km^2 resolution. Spatially distributed annual NH_3 and NO_x emission data for the study year (2008) were obtained from the National Atmospheric Emissions Inventory (http://naei.defra.gov.uk/) for the Scottish domain and from the National Institute for Public Health and the Environment (RIVM), for the Netherlands (Fig. 1). In order to evaluate ~~the sub-grid model~~AQR for an emission dataset with a lower spatial resolution that could be used for a Europe-wide application of the ~~sub-grid~~ model, the 2008 'EC4MACS' emissions with a spatial resolution of ca. 7 × 7 km^2 (EC4MACS, 2012, also used in Schaap et al., 2015) were also used for the two domains.

~~Meteorological data used to develop the sub-grid model were taken~~

In order to parametrise the pollutant dispersion from source areas, three different atmospheric dispersion models were used. These were ADMS (v4.1) (Carruthers et al., 1994), AERMOD (v12345) (Cimorelli et al., 2002) and LADD (Dragosits et al., 2002). These three models were chosen because they have been extensively evaluated for the atmospheric dispersion of NO_2 and NH_3, with the exception of LADD, which has only been evaluated for NH_3 (Theobald et al., 2012). The meteorological data used for the atmospheric dispersion simulations were derived from the meteorological data used in the EMEP model simulation (generated by the Weather Research Forecast (WRF) model version 3.6.1; http://www.wrf-model.org). Surface and vertical profile data at the centre of each EMEP model grid square were extracted in AERMOD format using the Mesoscale Model Interface Program (MMIF; https://www3.epa.gov/ttn/scram/dispersion_related.htm#mmif) and subsequently converted into the input formats for ADMS and LADD. In order to test the sensitivity of AQR to the meteorological data used, additional simulations were carried out using two domain-specific real meteorological datasets and a synthetic meteorological dataset derived from data from an arbitrary location. The two domain-specific datasets used were from Easter Bush, for Scotland (von Bobrutzki et al., 2010), and Cabauw, for the Netherlands (obtained from the Cesar Database: http://www.cesar-database.nl/). The synthetic dataset was derived from data from the Lyneham meteorological station in the UK for 1995 (LYNE95) (Spanton et al., 2004), which was a fairly typical year with regards to mean air temperature and wind speed. Although an arbitrary choice, this dataset was chosen because it has been used in various model evaluation studies and has been made freely available to the dispersion modelling community (e.g. Hall et al., 2000; Theobald et al., 2012). ~~In order to make the~~This dataset ~~less location specific~~was modified (LYNE95mod) by randomising the wind direction data ~~were randomised~~ and scaling the wind speed ~~was scaled~~ so that the annual mean value was equal to the annual domain mean value used in the EMEP model for the 2008 study year (5.1 m s^{-1}). ~~The use of a single UK meteorological dataset from a different year to the study year for the development of a model applied at the European scale may introduce a large amount of error or uncertainty in the predictions. In order to assess this uncertainty, two domain-specific meteorological datasets for the study year were also tested. These datasets were from Easter Bush, for Scotland (von Bobrutzki et al., 2010), and Cabauw, for the Netherlands (obtained from the Cesar Database: http://www.cesar-database.nl/).~~The wind directions were randomised for two reasons: 1) to make the meteorological data less location specific so that they can be used within different modelling domains and 2) to provide a generic dataset that could be of use to the air quality modelling community.

Evaluation of the AQR model

~~In order to parametrise the pollutant dispersion from source areas, three different atmospheric dispersion models were used. These were ADMS (v4) (Carruthers et al., 1994), AERMOD (v12345) (Cimorelli et al., 2002) and LADD (Dragosits et al., 2002). These three models were chosen because they have been extensively evaluated for the atmospheric dispersion of NO_2 and NH_3, with the exception of LADD, which has only been evaluated for NH_3 (Theobald et al., 2012).~~

~~Model evaluation~~ was carried out using 2008 annual mean concentration data from local and national monitoring networks in the two study domains. For Scotland, NO_2, data were obtained from the Air Quality in Scotland website

(http://www.scottishairquality.co.uk/) (48 stations: 37 traffic and 11 non-traffic sites) and from RIVM for the Netherlands (43 stations: 13 traffic and 30 non-traffic). The evaluation was done for all sites~~,~~ and for the traffic and non-traffic sites separately since the traffic sites are strongly influenced by the exact site location and are unlikely to be representative of a 1 × 1 km^2 grid square. For NH_3 concentrations in the Scottish domain, monitoring data were obtained from the UK National Ammonia Monitoring Network (NAMN) (Sutton et al., 2001) (http://uk-air.defra.gov.uk/networks/network-info?view=nh3), which has 14 sites within the domain. In addition, NH_3 monitoring data from 21 sites in a local network covering 36 km^2 (Vogt et al., 2013) were also used. For the Netherlands, NH_3 concentration data from the Measuring Ammonia in Nature (MAN) network (Lolkema et al., 2015) were provided by RIVM (108 stations). Model performance was assessed using the evaluation statistics of the R package "OpenAir" (Carslaw and Ropkins, 2012). Four performance metrics were used to compare the modelled concentrations with the observed values: fraction of model predictions within a factor of two of the observations (FAC2), normalised mean bias (NMB), normalised mean gross error (NMGE) and the Pearson correlation coefficient (r) (see Appendix A for definitions).

3 Model development

The sub-grid 1 × 1 km^2 concentration estimates were calculated from three components: the EMEP 50 × 50 km^2 concentration predictions, the 1 × 1 km^2 emission data and an estimate of short-range (< 50 km) pollutant dispersion. Figure 2 shows a schematic of the process. Short-range pollutant dispersion was parameterised using a simple scenario of a single 1 × 1 km^2 source with an emission rate of 1 Mg km^{-2} yr^{-1} in the centre of a square domain (of dimensions 101 × 101 km^2). Although individual sources are generally smaller than this, this value was used to match the spatial resolution of the emission data. For NO_2, the assumption was made that annual mean NO_2 concentrations are linearly correlated with those of NO_x. This allowed us to use the NO_x emissions for the calculation of NO_2 concentrations without considering photochemical reactions. An analysis of the 2008 mean annual concentrations for the 1478 sites in the Air Quality e-Reporting database (formerly AirBase) of the European Environment Agency shows that measured NO_2 and NO_x concentrations are approximately linearly correlated with a linear correlation coefficient, r^2 of 0.93. For the dispersion of NH_3, the source was assumed to be at ground level (a suitable approximation for most agricultural sources, which account for more than 90% of emissions in Europe). ~~For NO_2, the assumption was made that NO_2 concentrations were linearly correlated with NO_x concentrations (valid for annual mean concentrations).~~Emissions of NO_x, on the other hand, ~~This allowed us to use the NO_x emissions for the calculation of NO_2 concentrations without considering photochemical reactions.~~ ~~Emissions of NO_x~~ can occur over a range of emission heights, depending on the source type. Since the emission height will affect the resulting NO_2 concentrations at ground level, it needs to be taken into account. This was done by assigning a representative emission height for each emission sector (Selected Nomenclature for Air Pollution (SNAP) code) that contributed more than 1% of the total domain emissions (Table 1). These emission heights correspond ~~loosely~~approximately to the mean effective emission heights used in the EMEP model for the sector emissions. In order to test the sensitivity of the ~~sub-grid~~AQR model to the

emission heights used, additional simulations were carried out using emission heights half and double these values. For the ground level source, all three dispersion models (ADMS, AERMOD and LADD) were used to simulate the annual mean near-ground-level concentrations of NH_3 and NO_2 on a 1 km grid (for the 101 × 101 km^2 domain~~) using the LYNE95mod meteorological dataset.~~). For the elevated source scenarios, only ADMS and AERMOD were used to simulate the annual mean concentrations because the LADD model is not suitable for simulating dispersion from elevated sources (Theobald et al., 2012). A height of 1.5 m was used for the near-ground-level concentrations, because this height is commonly used for concentration monitoring and impact assessments (Cape et al., 2009). These short-range dispersion simulations were carried out using the meteorological data extracted from the WRF simulations at the centre of each EMEP model grid square. No removal processes (chemical reactions, dry or wet deposition etc.) were simulated because these processes depend strongly on local conditions (concentrations of other chemical species, meteorological conditions, surface characteristics, etc.).

The result of these simulations was nine concentration fields~~,~~ (kernels), three for ground level sources (three models × one source height) and six for elevated sources (two models × three source heights) ~~centred on the source location. For each source height, a rotationally symmetric concentration field (or kernel) was obtained by fitting regression curves to the modelled concentrations (natural log of concentrations vs. natural log of distance from source centre), which was then averaged over all models (more details are provided in the supplementary material).~~for each meteorological dataset (corresponding to each of the EMEP model grid squares). A model-average dispersion kernel (D) for each source height was obtained by taking the mean value of the dispersion model concentration estimates for each kernel grid cell.

These model-average kernels were then ~~multiplied by~~combined with the emission data ~~(for each SNAP sector separately in the case of NO_x)~~ using a ~~"~~moving window~~"~~ approach to obtain the sub-grid concentration estimate (C):

$$C(i,j) = \sum_{s}^{SNAP} \sum_{i'}^{n} \sum_{j'}^{m} E(i',j')\, D'(i-i', j-j')$$

where i and j are the ~~results summed over~~sub-grid-cell coordinates, s is the ~~entire~~emission sector, i' and j' are the emission grid cell coordinates, E is the emission rate of the emission grid cell (Mg km^{-2} yr^{-1}) and D' an interpolated dispersion kernel (inverse distance squared weighted interpolation of the kernels for the source EMEP grid square and the eight adjacent grid squares). Since the dispersion kernel has a size of 101 × 101 grid cells, the values of i' and j' range from i-50 and j-50 to i+50 and j+50, respectively, with the constraint that they lie within the modelling domain~~ (central Scotland or the Netherlands).~~ .

The resulting "sub-grid distributions" provide an estimate of the spatial variability of the concentrations at a 1 × 1 km^2 resolution, which were then used to "redistribute" the EMEP predictions within each 50 × 50 km^2 grid square. This step is necessary since ~~the sub-grid model~~AQR does not take into account large scale processes such as long-range transport or

chemical transformations of pollutants, processes that are included in the large scale model (the EMEP model, in this case). The simplest way to do this redistribution would be to multiply the sub-grid distributions by the EMEP predictions and then divide by the mean value of the sub-grid distribution for each 50 × 50 km^2 grid square. This approach conserves the sub-grid distribution for each 50 × 50 km^2 square and also has the same mean concentration as the EMEP prediction. However, it also could lead to large discontinuities at the edges of the EMEP grid squares if the ratio between the mean of the sub-grid distribution and the EMEP prediction differ greatly from that of adjacent squares. To avoid this problem, the ratio of the EMEP predictions to the mean value of the sub-grid distribution for each 50 × 50 km^2 square was interpolated on a 1 × 1 km^2 grid (using a spline interpolation of the values at the centre of each grid square in ArcGIS 10.2 (Environmental Systems Research Institute, Redlands, CA, USA)). The interpolated field was then multiplied by the sub-grid distribution and then the process was repeated over ten iterations. In fact only four-five iterations were necessary to give concentration fields that differed by a maximum of 1%. A more detailed description of the process is provided in the supplementary material.

In order to test the sensitivity of the model to the meteorological data, the above process was repeated with the kernels obtained from the dispersion simulations using the domain-specific meteorological data and with kernels derived from the dispersion simulations using the synthetic meteorological data (more details provided in the supplementary material).

4 Results

4.1 Sub-grid concentration predictions and model evaluation

Figure 3 shows the sub-grid concentration predictions for NO_2 and NH_3 for the two domains~~.~~ (data for the individual domains are provided in Figure S3.1 in the supplementary material). The EMEP concentration fields are also shown for comparison. Table 2 shows the evaluation statistics of the EMEP and ~~sub-grid~~AQR models for annual mean NO_2 concentrations for the Dutch and Scottish monitoring data. In general~~ the sub-grid model performed notably better than~~, AQR is an improvement on the EMEP model ~~as a result of a consistent underestimation by~~alone because the latter generally underestimates concentrations (negative NMB). The mean error of the EMEP model is largest for the Scottish dataset with a NMGE of 82% and 70% for the datasets with and without traffic stations, respectively. The model performs worst for the Scottish traffic stations with a mean underestimation of 84%. The EMEP model performs considerably better for the Dutch dataset, with 91% of predictions within a factor of two of the observed values, although this drops to 69% when considering the traffic stations only. The ~~sub-grid~~AQR model (using 1 x 1 km^2 emissions) also performed best for the Dutch dataset, with a smaller mean bias and error and a better correlation than the EMEP model alone. However, the EMEP model had a lower mean bias and error for the non-traffic stations. The ~~sub-grid~~AQR model is also ~~performed better than~~an improvement on the EMEP model alone for the Scottish dataset (both with and without traffic stations), as well as for the combined dataset (Netherlands plus Scotland). Similarly to the EMEP model, ~~the sub-grid model~~AQR performed worst for the Scottish traffic stations, although ~~notably better than~~was a notable improvement over the EMEP model alone. The use of the lower

resolution emissions actually improved the performance of ~~the sub-grid model~~AQR for some of the statistics (most notably for the non-traffic stations in the Netherlands domain).

Table 3 shows the evaluation statistics of the EMEP and ~~sub-grid~~AQR models for annual mean NH_3 concentrations for the Dutch and Scottish monitoring data. In general ~~the sub-grid model performed notably better than~~AQR was an improvement on the EMEP model~~. The EMEP model~~ alone, which performed worse for the local monitoring network, as all monitoring locations were within a single EMEP 50 × 50 km^2 square. The ~~sub-grid~~AQR model (using 1 x 1 km^2 emissions) also performed worst for this dataset, although its performance was ~~better than~~still an improvement on that of the EMEP model alone, as it was for all the datasets except for the National Ammonia Monitoring Network sites in Scotland. The use of the 7 x 7 km^2 emissions worsened the performance of ~~the sub-grid model~~AQR for all datasets except for the National Ammonia Monitoring Network sites, for which it had a similar performance to the model using the higher resolution emissions. Figure 4 shows the scatterplots of NO_2 and NH_3 concentration predictions of the EMEP and ~~sub-grid model~~AQR models vs. the observed values for all sites in both domains.

4.2 Sensitivity of sub-grid model predictions to model parameters

The use of ~~domain-specific~~alternative meteorological datasets only had a small effect on the concentration estimates of the ~~sub-grid~~AQR model (Fig. 5). ~~Mean differences~~The use of domain-specific data from a single location affected the ~~estimates using the generic meteorological dataset (LYNE95mod) were 7~~concentration predictions by an average of 6% for ~~both~~ NO_2 and 5% for NH_3~~,~~ although differences of up to ~~29~~23% were found for individual measurement sites. ~~Model performance was barely affected (not shown).~~Similarly, the use of the synthetic meteorological data affected concentrations, on average, by 6% and 5% for NO_2 and NH_3, respectively, with a maximum difference of 28%. Randomising the wind direction data of the domain-specific datasets gave very similar results to those using the ~~generic~~synthetic meteorology dataset, with maximum differences of only 1% (not shown). This suggests that the meteorological factor that most influences the ~~sub-grid model~~ estimates of the AQR model is the wind direction distribution.

The ~~sub-grid~~AQR model estimates are also not very sensitive to the NO_x emission height. On average, the effect on the concentration predictions of halving or doubling the emission heights is less than 2%, with a maximum difference of 6~~%.~~% (not shown). This lack of sensitivity to the exact source height ~~used~~ reflects the fact that ground-level sources contribute significantly more to near-source concentrations than elevated sources. Since the concentrations predicted by AQR were not greatly affected by the meteorological data or the emission heights, model performance was very similar (not shown).

5 Discussion

5.1 An improvement, but is it enough?

These results show that a simple and robust geostatistical approach can be used to improve the EMEP model predictions of NO_2 and NH_3 annual concentrations. This improvement is not surprising considering the large difference in spatial resolutions (50 km vs. 1 km) and the strong link between short-lived pollutants and the spatial distribution of emissions. ~~However, is this improvement~~In fact, it is worth looking at whether this improvement is mainly a result of the high resolution emissions and has very little to do with the use of short range dispersion estimates. This can be done by repeating the analyses with the 1 × 1 km^2 grid cell emissions as the initial sub-grid distribution. Figure 6 shows that doing this for NO_2 substantially overestimates concentrations for the mid-range of measured values, whereas for NH_3 concentrations are substantially underestimated at many sites. The model performance statistics for these simulations show that using just the emissions gives lower values of FAC2 (0.60 vs 0.70 for NO_2 and 0.28 vs 0.84 for NH_3) and larger bias and error (NMB: 0.36 vs -0.27 for NO_2 and -0.36 vs 0.09 for NH_3; NMGE: 0.72 vs. 0.41 for NO_2 and 0.79 vs 0.27 for NH_3). Model error is even larger than that for the EMEP model alone (0.72 vs 0.61 for NO_2 and 0.79 vs 0.42 for NH_3), which demonstrates that short-range dispersion estimates are necessary for improving on the EMEP model predictions. However, is the improvement of AQR over the EMEP model alone large enough to warrant the inclusion of such a sub-grid model into the output processing options of a chemical transport model? In order to answer this question, we can use the concept of model acceptability suggested by Chang and Hanna (2004). This concept can be used to evaluate whether the EMEP model and/or the ~~sub-grid~~AQR model perform acceptably and, therefore, whether the ~~sub-grid~~AQR model represents an improvement on the EMEP model alone, in terms of model acceptability. Hanna and Chang (2012) suggested that an 'acceptable' model is one that meets the criteria for more than half of a series of statistical tests. The performance metrics used are: fractional bias, geometric mean bias, normalised mean square error, geometric variance and FAC2 (see Appendix A for definitions and acceptability criteria). In the current study, we define an acceptable model as one that meets at least three of these five criteria (for each dataset). Although the concept of model acceptability of Chang and Hanna (2004) was defined for research-grade experimental data, the fact that we are considering annual mean concentrations (instead of high temporal resolution measurements), should make the approach suitable for use with operational models and monitoring data, such as those used here. For the two combined datasets (NO_2-All and NH_3-All) shown in Fig. 4, the EMEP model meets none and five of the five criteria for NO_2 and NH_3 respectively, whereas ~~the sub-grid model~~AQR meets three and five criteria, respectively (Table 4). This suggests that ~~the sub-grid model~~AQR is a significant improvement (in terms of model acceptability) for NO_2 (even when the lower resolution emission dataset is used), but not for NH_3., since the EMEP model alone already performs acceptably for this dataset. This can be explained by looking at the number of criteria met for the individual datasets (Table 4). For NO_2, The EMEP model performed acceptably for the Netherlands (All) but not for Scotland (All). This is partly due to the Dutch network having a larger proportion of non-traffic sites (70% vs. 23%), which would be more representative of the 50 × 50 km^2 grid cells. However, the EMEP model also performed acceptably for the Dutch traffic stations but neither

the EMEP model nor the ~~sub-grid~~AQR model performed acceptably for the Scottish traffic stations. Looking more carefully at the traffic stations used in the domains reveals that station siting may have an influence on model performance. According to the information available regarding the Scottish traffic sites, monitoring stations are located between 0.5 and 16 m from the road edge. Although no information is available regarding the exact locations of the Dutch monitoring stations, Nguyen et al. (2012) point out that one station in the Amsterdam Municipal Health Service (GGD) network (not used in this study) "is very close to the road (< 2.5 m)". This suggests that, in general, sites in the Dutch network are > 2.5 m from the road, whereas in the Scottish network 17 of the 37 traffic sites are closer than this. This difference in station siting could be the reason why neither the EMEP nor ~~sub-grid~~the AQR model ~~performs~~performed acceptably for the Scottish dataset. For NH_3, the EMEP and ~~sub-grid model perform~~AQR models performed acceptably for the two national networks but only ~~the sub-grid model performs~~AQR performed acceptably for the local network. This is probably because the national networks site their monitoring stations far from the influence of individual emission sources in order to be representative of a large area, whereas the local network was located in an area with intensive poultry farming and was designed to assess the influence of individual sources. Since the majority (86%) of the sites used in the analysis belonged to the national networks, overall model performance was similar to model performance for those networks. The sub-grid approach, therefore, is most useful where there are large horizontal concentration gradients, such as within large cities (for NO_2) or areas with intensive agriculture (for NH_3), which is where the largest impacts are most likely to occur.

It is also worth briefly comparing the improvements in model performance with those reported by other studies. Denby et al. (2011) showed that the population weighted concentration for NO_2 was, on average, 44% higher with their sub-grid parameterisation than that calculated using the original concentrations from the EMEP model. Although not directly comparable (since we do not calculate population weighted concentrations), NO_2 concentrations estimated using ~~our sub-grid~~the AQR model ~~were~~are, on average 77% higher than those of the EMEP model at the monitoring station locations. Despite this increase, the ~~sub-grid model~~AQR estimates ~~were~~are still, on average, 27% lower than the measured concentrations. Janssen et al. (2012) showed that their approach of downscaling modelled concentrations from 15 × 15 km^2 to 3 × 3 km^2 reduced model error by about 20%. ~~Our sub-grid~~The AQR model for NO_2 reduced model error by 30–40%, although for a larger change in resolution (50 × 50 km^2 to 1 × 1 km^2). In the study by Schaap et al. (2015), increasing the spatial resolution from approx. 56 × 56 km^2 to 7 × 7 km^2 increased the correlation (r) between the models' predictions and hourly urban background NO_2 concentrations from approx. 0.1–0.4 to 0.6–0.7 and reduced model bias by approx. 60–90% for most of the models. For a similar change in spatial resolution (50 × 50 km^2 to 7 × 7 km^2), ~~our sub-grid~~the AQR model for annual mean NO_2 concentrations using the low resolution emissions increased r from 0.16–0.54 to 0.51–0.~~79~~85 and reduced model bias by approx. 20–70%.

5.2 How can the sub-grid approach be applied?

Two potential uses of the sub-grid approach can be envisaged: a Europe-wide application to provide a spatial assessment of exceedance of NO_2 and NH_3 annual limit values or critical levels and the assessment of individual emission hot-spots in areas where detailed modelling assessments are not available but high resolution emission data are. In the latter case, if the hot-spot domain is located within a single EMEP 50 × 50 km^2 grid square, the smoothing step would not be necessary. The Europe-wide application would require high spatial resolution emission data for the whole domain. There is, as far as we are aware, currently no European emission inventory with a spatial resolution close to 1 × 1 km^2. The highest resolutions available are the 7 × 7 km^2 emission inventories produced for various EU projects (Kuenen et al., 2014, EC4MACS, 2012). As shown above, the use of emission data at this resolution still gives an improvement on the concentration predictions and even performs better than the sub-grid model using the higher resolution emissions, in some cases.

5.3 Advantage, disadvantages, uncertainties and potential improvements

The ~~sub-grid~~AQR model can provide more accurate concentration predictions than the EMEP model alone, especially close to emission sources. However, this approach has only been tested for annual mean NO_2 and NH_3 concentrations, although could potentially be extended to other short-lived pollutants and shorter time scales (daily or hourly). This means that the model cannot currently be used to assess exceedance of short-term limit values (e.g. for Europe, an hourly mean concentration of 200 µg NO_2 m^{-3} more than 18 times in one year) although, as shown by Kiesewetter et al. (2013), the annual mean limit values for NO_2 ~~and PM_{10} are~~is the more stringent ~~targets~~target. Critical levels for ammonia are expressed as annual mean concentrations and so a sub-grid model with a higher temporal resolution is not necessary. The other limitation of the approach is the need for high resolution emission data although, as shown above, the use of emission data with a resolution of 7 × 7 km^2 already produces improvements in model performance compared with the original ~~CTM~~ACTM concentration estimates.

The various assumptions and simplifications made in the development of AQR introduce uncertainty in the model estimates. The omission of NO_x photochemistry and the assumption that annual mean NO_2 concentrations are linearly correlated with those of NO_x was justified above by the fact that measured concentrations across Europe are approximately linearly correlated (r^2 = 0.93). However, a more in-depth analysis of the European measurements shows that if a constant factor is used to estimate NO_2 concentrations from the measurements of NO_x, the estimated NO_2 concentrations differ from the measure values by an average of 16%, which is a small uncertainty compared with the uncertainty in emissions, meteorological conditions, etc. The uncertainty as a result of not modelling the chemical transformation of NH_3 (e.g. to particulate ammonium) is more difficult to quantify since the reactions depend on many factors such as the meteorological conditions and the concentrations of other pollutants. However, the fact that the errors (NMGE) in the AQR estimates of NH_3 concentrations are of a similar order of magnitude to the errors in the NO_2 estimates suggests that the benefits of AQR in handling sub-grid distributions outweigh any chemical impacts. In addition, such errors would be largest far from the

sources, once NH_3 concentrations are diluted more to levels comparable to incoming sulphate or HNO_3 concentrations. Another source of uncertainty is the omission of deposition processes in the short-range dispersion parameterisations, but wet-deposition has been implicitly included in ACTM predictions, and time-scales for dry deposition are usually far larger than those for sub-grid mixing. Again, given the AQR model has a mean error of 41% and 27% for NO_2 and NH_3, respectively, the benefits of AQR seem greater than any uncertainty as a result of omitting these processes. Finally, another simplification is the use of a 1 × 1 km^2 source for parameterising short-range dispersion. In reality sources are generally smaller than this and so this simplification may result in incorrect concentration gradients close to small or linear NO_x sources (e.g. chimney stacks or motorways). However, on average, transport emissions contribute more than 90% of the estimated concentrations, most of which are in urban areas where a 1 × 1 km^2 source is probably an adequate representation of a dense urban road network. In addition, we rarely know the location of stacks in emission inventories to better than 1 km resolution, and usually with no or very limited information on plume rise and height.

With regards to potential improvements, in addition to the extension to shorter time periods, it ~~would also be possible to include spatially varying wind data since this has the potential to better represent local conditions. Such data could be obtained directly from the meteorological fields of the CTM. It also would~~also should be possible to incorporate stack parameters (effective emission heights and the contribution of stack emissions to the emissions of a particular grid square) from officially reported data and/or other data sources~~.~~, if these become more readily available. This would potentially improve concentration estimates close to large stack sources. As shown above, model performance is poorer for sites very close to roads and so the inclusion of a roadside increment model could also improve the model estimates. However, by increasing the complexity of the model, we have to careful not to lose sight of the objective of the ~~sub-grid~~AQR model, which is to provide a robust and simple method of post-processing concentrations estimated by a chemical transport model.

The sub-grid approach also has the potential to be applied to other pollutants for which there is a strong relationship between emissions and concentrations. Zhang and Wu (2013) analysed air quality simulations of the CMAQ model to quantify the influence of a range of processes on the atmospheric concentrations of several pollutants. The species that were most strongly influenced by emission processes were: NH_3, NO, NO_2, SO_2, $PM_{2.5}$, ~~SO_2~~ SO_4^{2-}, elemental carbon, and primary organic aerosol and are, therefore, potential candidates for an extension of the model. The spatial distribution of ozone, a secondary pollutant, cannot be estimated based on emissions but its inverse relationship with NO_x could be exploited to model the sub-grid variability. Apart from concentrations, it may also be possible to develop a sub-grid model for processes such as wet deposition of nitrogen or sulphur, for which high resolution rainfall maps could be used to estimate the sub-grid distributions. Dry deposition of reduced nitrogen could also be modelled using the NH_3 concentration distribution and land cover parameters, assuming that most of the deposition is in the form of NH_3. Dry deposition of oxidised nitrogen would be more difficult since there is no one dominant species that contributes.

Conclusions

The sub-grid spatial variability of the annual mean NO_2 and NH_3 concentrations ~~predictions~~ predicted ~~of NO_2 and NH_3 of~~by an atmospheric chemistry and transport model can be estimated by combining the predictions with high spatial resolution emission datasets and short-range dispersion fields. This paper describes the development of ~~this technique~~ the Air Quality Re-gridder (AQR) model and its application to two test domains in Europe. Comparison of ~~the sub-grid model predictions with~~ annual mean concentrations estimated by AQR with measured values within both domains shows that the ~~sub-grid~~AQR model represents an improvement on the predictions of the atmospheric chemistry and ~~chemical~~ transport model, reducing both model error and bias and increasing the spatial correlation with the measured concentrations.

Code availability

The ~~sub-grid~~AQR model code (in R) plus example input and output files for the simulations using synthetic meteorological data are provided in the supplementary material.

Data availability

The data shown in Fig. 4, Fig. 5 and Fig. ~~5~~6 are provided in the supplementary material.

Appendix A: Descriptions of the performance metrics used

Table A1: The four metrics relating modelled concentrations (M_i) with the observed values (O_i), used for evaluating model performance.

Performance measure	Definition
Fraction of model predictions within a factor of two of the observations (FAC2):	$0.5 \le \frac{M_i}{O_i} \le 2.0$
Normalised mean bias:	$NMB = \frac{\sum_{i=1}^{n} M_i - O_i}{\sum_{i=1}^{n} O_i}$
Normalised mean gross error:	$NMGE = \frac{\sum_{i=1}^{n} \lvert M_i - O_i \rvert}{\sum_{i=1}^{n} O_i}$
Pearson correlation coefficient:	$r = \frac{1}{(n-1)} \sum_{i=1}^{n} \left(\frac{M_i - \overline{M}}{\sigma_M} \right) \left(\frac{O_i - \overline{O}}{\sigma_O} \right)$

Table A2: The five performance measures relating modelled concentrations (M_i) with the observed values (O_i) used to assess model acceptability.

Performance measure	Definition	Optimum value	Acceptability Criterion
Fractional bias (FB)	$FB = \frac{2(\overline{O} - \overline{M})}{(\overline{O} + \overline{M})}$	0	|FB| < 0.3
Geometric Mean Bias (MG)	$MG = \exp(\overline{\ln O} - \overline{\ln M})$	1	0.7 < MG < 1.3
Normalised mean square error (NMSE)	$NMSE = \frac{\overline{(O - M)^2}}{\overline{O}\ \overline{M}}$	0	NMSE < 1.5
Geometric variance (VG)	$VG = \exp\left[\overline{(\ln O - \ln M)^2}\right]$	1	VG < 4
Fraction of model predictions within a factor of two of the observations (FAC2)	$0.5 \le \frac{M_i}{O_i} \le 2.0$	1	FAC2 > 0.5

Author contribution

Model development and evaluation was principally carried out by M. R. Theobald with contributions on the design of the sub-grid modelling methodology from D. Simpson and M. Vieno. Emission datasets were prepared by M. Vieno and the manuscript was prepared by M. Theobald with contributions from both co-authors.

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Table 1: Emission heights used for each main emission sector

SNAP Code	Emission sector	Effective emission height used (m)
1	Combustion in energy and transformation industries	400
2	Non-industrial combustion plants	0
3	Combustion in manufacturing industry	400
4	Production processes	50
7	Road transport	0
8	Other mobile sources and machinery	0
9	Waste treatment and disposal	200

Table 2: Performance evaluation of the EMEP and sub-grid models for annual mean NO_2 concentrations. The best performing model for each statistic is highlighted in bold. FAC2 is the fraction of model predictions within a factor of two of the observations, NMB is the normalised mean bias, NMGE is the normalised mean gross error and r is the Pearson correlation coefficient. Shaded cells highlight the model performance for the sub-grid model using the lower resolution emission data.

Dataset	n	EMEP model				Sub-grid model				
		FAC2	NMB	NMGE	r	Emission data	FAC2	NMB	NMGE	r
Netherlands (All)	43	0.91	-0.24	0.31	0.54	1 × 1 km^2	**1.**0.98	**0.0405**	0.2224	**0.8483**
						7 × 7 km^2	**1.0**	-0.08	**0.21**	0.79
Netherlands (No traffic stations)	30	**1.00**	-0.06	**0.18**	0.73	1 × 1 km^2	**1.**0.97	0.07	0.2729	**0.86**
						7 × 7 km^2	**1.0**	**0.01**	0.21	0.81
Netherlands (Traffic stations only)	13	0.69	-0.45	0.45	0.17	1 × 1 km^2	**1.0**	**0.0002**	**0.1716**	**0.5848**
						7 × 7 km^2	**1.0**	-0.18	0.21	0.32
Scotland (All)	48	0.06	-0.82	0.82	0.16	1 × 1 km^2	**0.4846**	**-0.4850**	**0.5254**	0.4643
						7 × 7 km^2	0.23	-0.63	0.63	**0.51**
Scotland (No traffic stations)	11	0.27	-0.70	0.70	0.40	1 × 1 km^2	**0.91**	**-0.0708**	**0.30**	0.80
						7 × 7 km^2	0.64	-0.38	0.39	**0.85**
Scotland (Traffic stations only)	37	0.00	-0.84	0.84	0.05	1 × 1 km^2	**0.3532**	**-0.5456**	**0.5557**	0.5048

						7 × 7 km^2	0.11	-0.67	0.67	**0.51**
All	91	0.46	-0.58	0.~~60~~61	--	1 × 1 km^2	**0.~~73~~7 0**	**-0.27**	**0.~~40~~41**	**0.~~42~~ 39**
						7 × 7 km^2	0.59	-0.40	0.46	0.27

Table 3: Performance evaluation of the EMEP and sub-grid models for annual mean NH_3 concentrations. The best performing model for each statistic is highlighted in bold. FAC2 is the fraction of model predictions within a factor of two of the observations, NMB is the normalised mean bias, NMGE is the normalised mean gross error and r is the Pearson correlation coefficient. Shaded cells highlight the model performance for the sub-grid model using the lower resolution emission data.

Dataset	n	EMEP				Sub-grid model				
		FAC2	NMB	NMGE	r	Emission data	FAC2	NMB	NMGE	r
Netherlands	108	0.85	0.23	0.39	0.69	1 × 1 km^2	**0.~~93~~9 2**	**0.10**	**0.~~22~~24**	**0.~~85~~ 84**
						7 × 7 km^2	0.90	0.28	0.40	0.71
Scotland – Local network	21	0.52	-0.47	0.65	--	1 × 1 km^2	**0.62**	**-0.~~08~~0 4**	**0.~~54~~52**	**0.~~48~~ 55**
						7 × 7 km^2	0.52	-0.48	0.66	--
Scotland (National Ammonia Monitoring Network)	14	**0.71**	**0.07**	0.46	0.73	1 × 1 km^2	0.~~50~~5 7	0.~~24~~2 6	**0.~~42~~45**	0.~~80~~ 77
						7 × 7 km^2	0.57	**0.07**	0.43	**0.81**
All	143	0.79	0.17	0.42	0.74	1 × 1 km^2	**0.84**	**0.09**	**0.~~26~~27**	**0.~~85~~ 84**
						7 × 7 km^2	0.81	0.20	0.42	0.75

Table 4: Number of model acceptability criteria met for each model and evaluation dataset. Shaded cells represent acceptable model performance (≥ 3 criteria met).

Pollutant	Dataset		No. of criteria met		
			EMEP	Sub-Grid (1 × 1 km^2 emissions)	Sub-Grid (7 × 7 km^2 emissions)
NO_2	Netherlands	All	5	5	5
	Netherlands	No traffic stations	5	5	5
	Netherlands	Traffic stations only	3	5	5

	Scotland	All	0	2	1
	Scotland	No traffic stations	0	5	3
	Scotland	Traffic stations only	0	2	0
	All		0	3	3
NH_3	Netherlands		5	5	5
	Scotland	Local network	2	5	2
	Scotland	National Network	5	5	5
	All		5	5	5

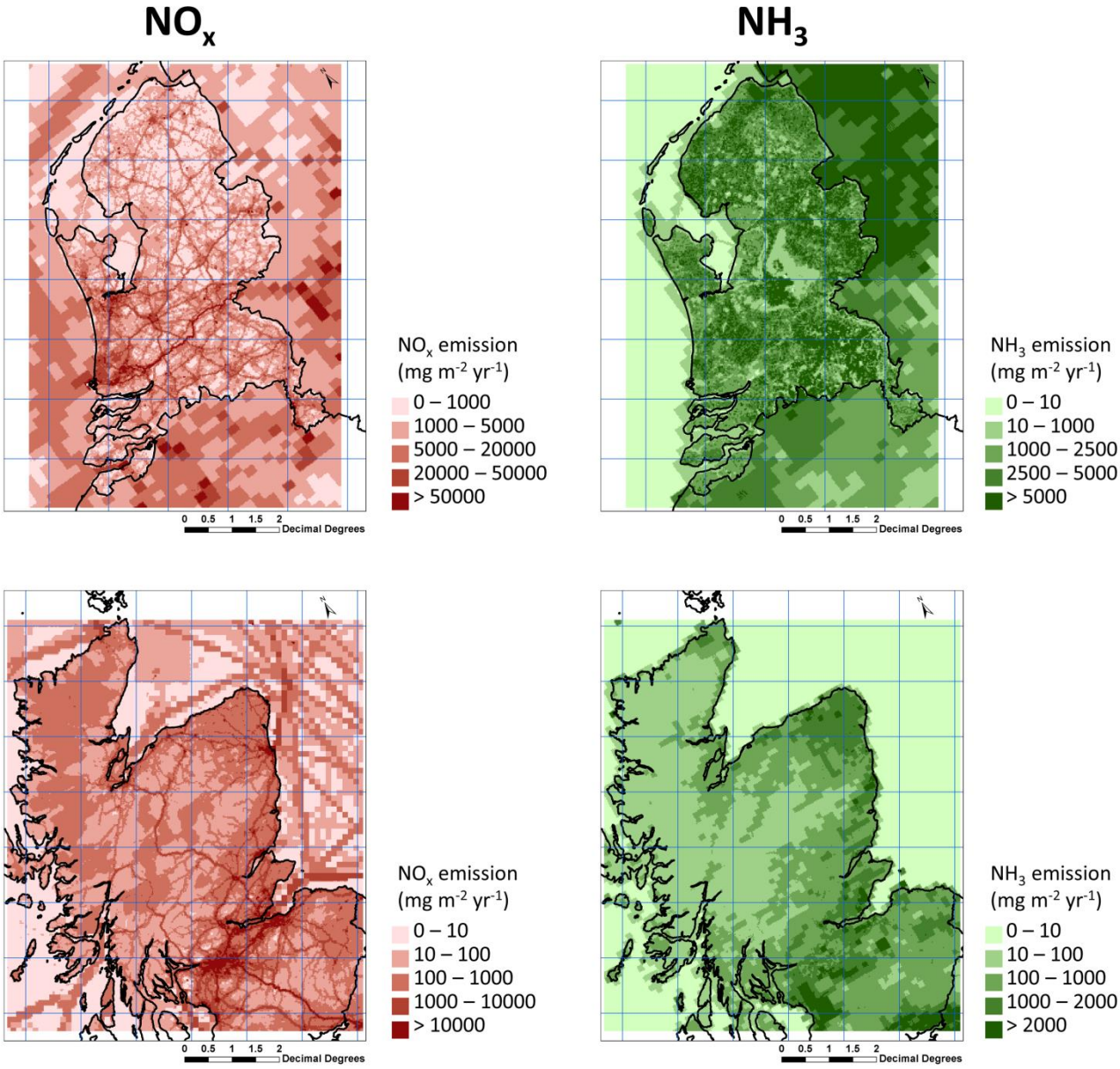


Figure 1: Spatial distributions of annual emissions of NO_x (left) and NH_3 (right), for the Dutch (top) and Scottish (bottom) domains. The EMEP 50 × 50 km^2 grid is also shown (in blue).

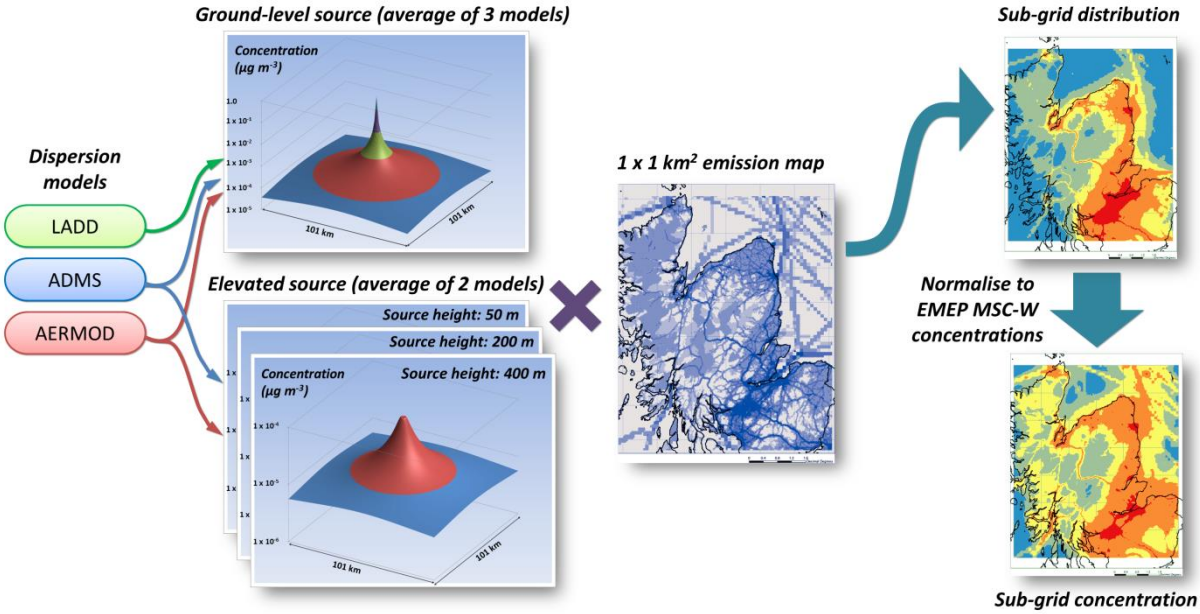
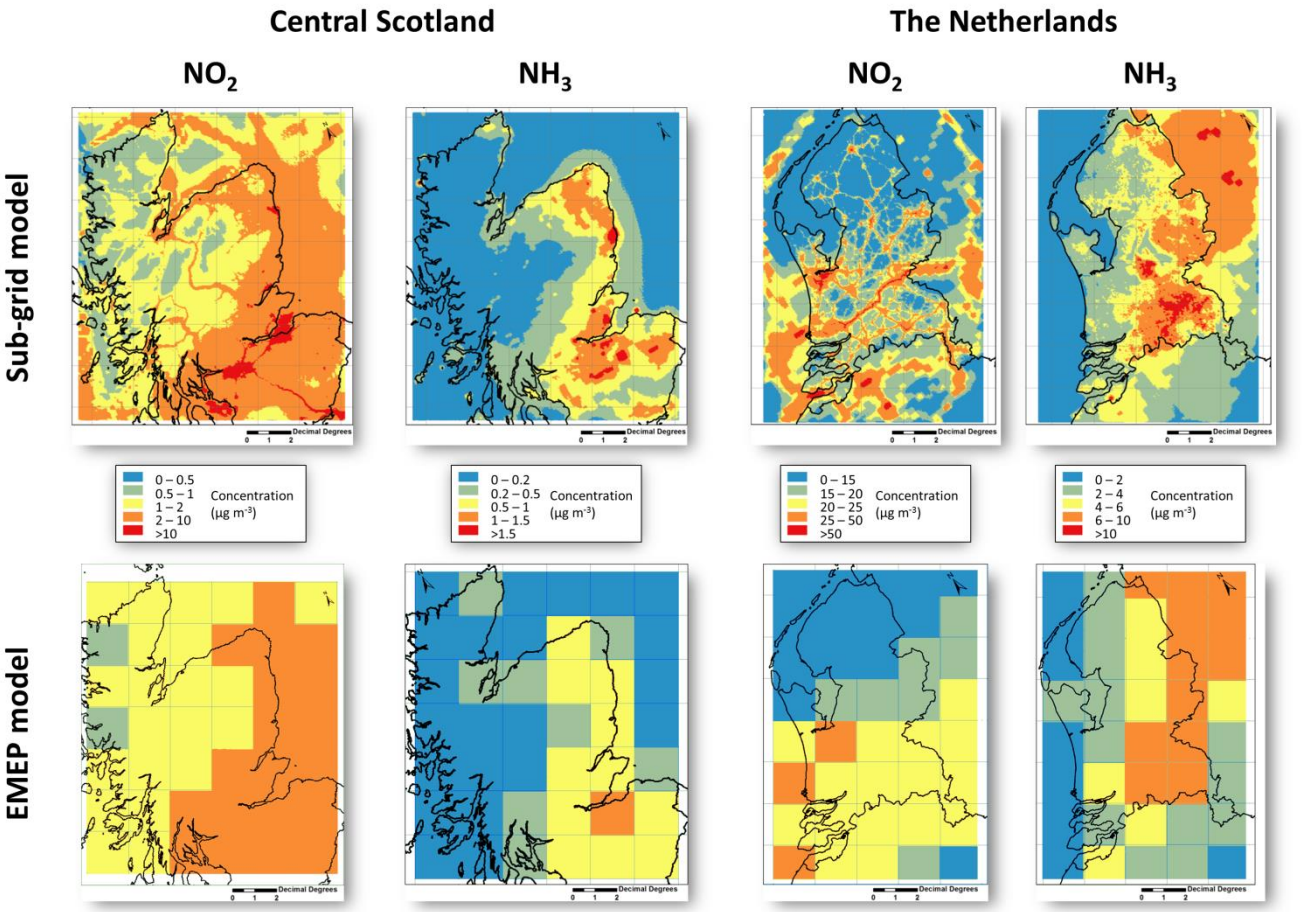


Figure 2: Schematic showing the process of producing the sub-grid concentration predictions from short-range dispersion model simulations and high spatial resolution emission data.


Central Scotland
The Netherlands
NO2
NH3
NO2
NH3
Sub-grid model
EMEP model
0 – 0.5
0.5 – 1
1 – 2
2 – 10
>10
Concentration (µg m-3)
0 – 0.2
0.2 – 0.5
0.5 – 1
1 – 1.5
>1.5
Concentration (µg m-3)
0 – 15
15 – 20
20 – 25
25 – 50
>50
Concentration (µg m-3)
0 – 2
2 – 4
4 – 6
6 – 10
>10
Concentration (µg m-3)
Decimal Degrees

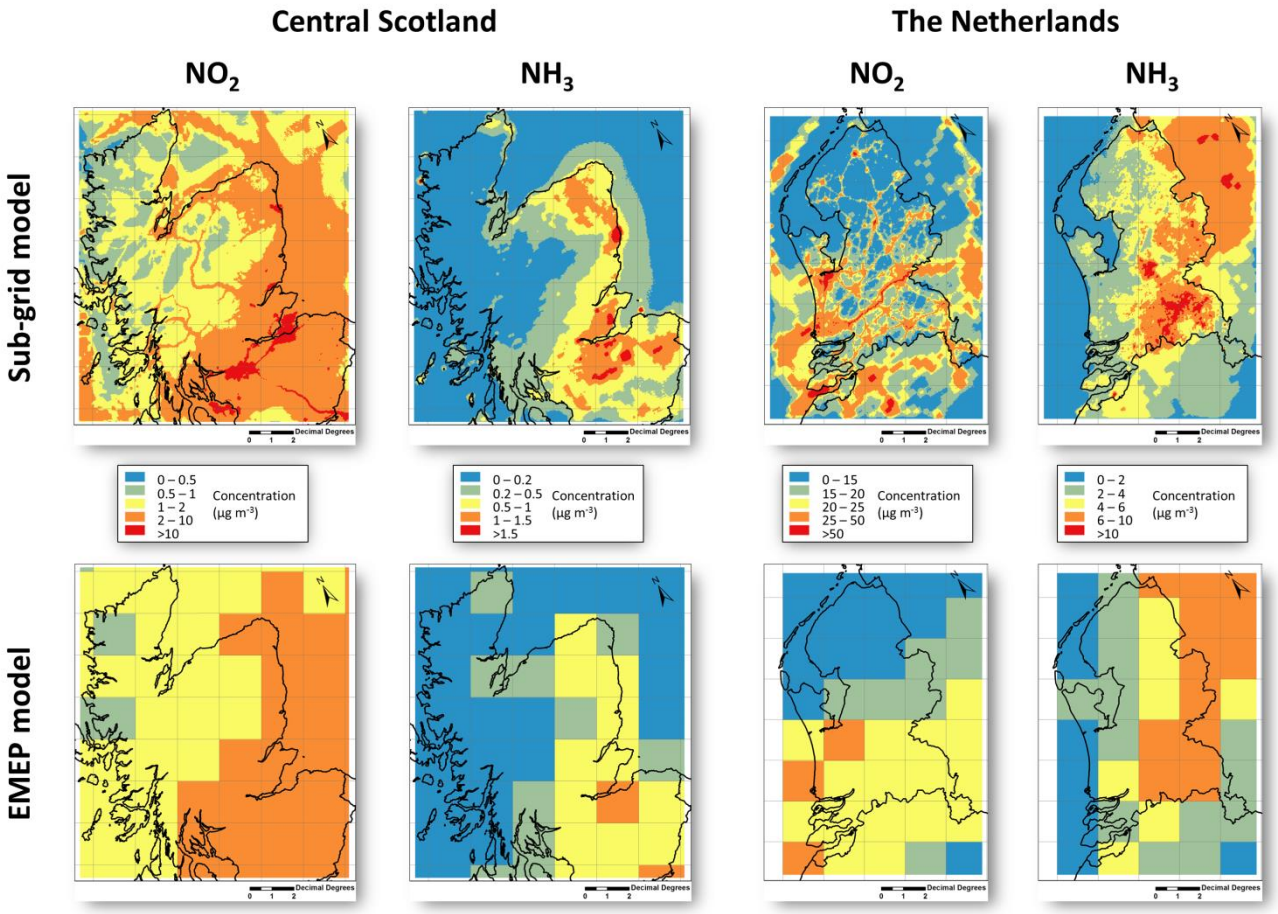


Figure 3: Sub-grid model predictions (top row) of annual mean concentrations of NO_2 and NH_3 for the two domains. EMEP model predictions at a resolution of 50 × 50 km^2 are shown for comparison (bottom row).

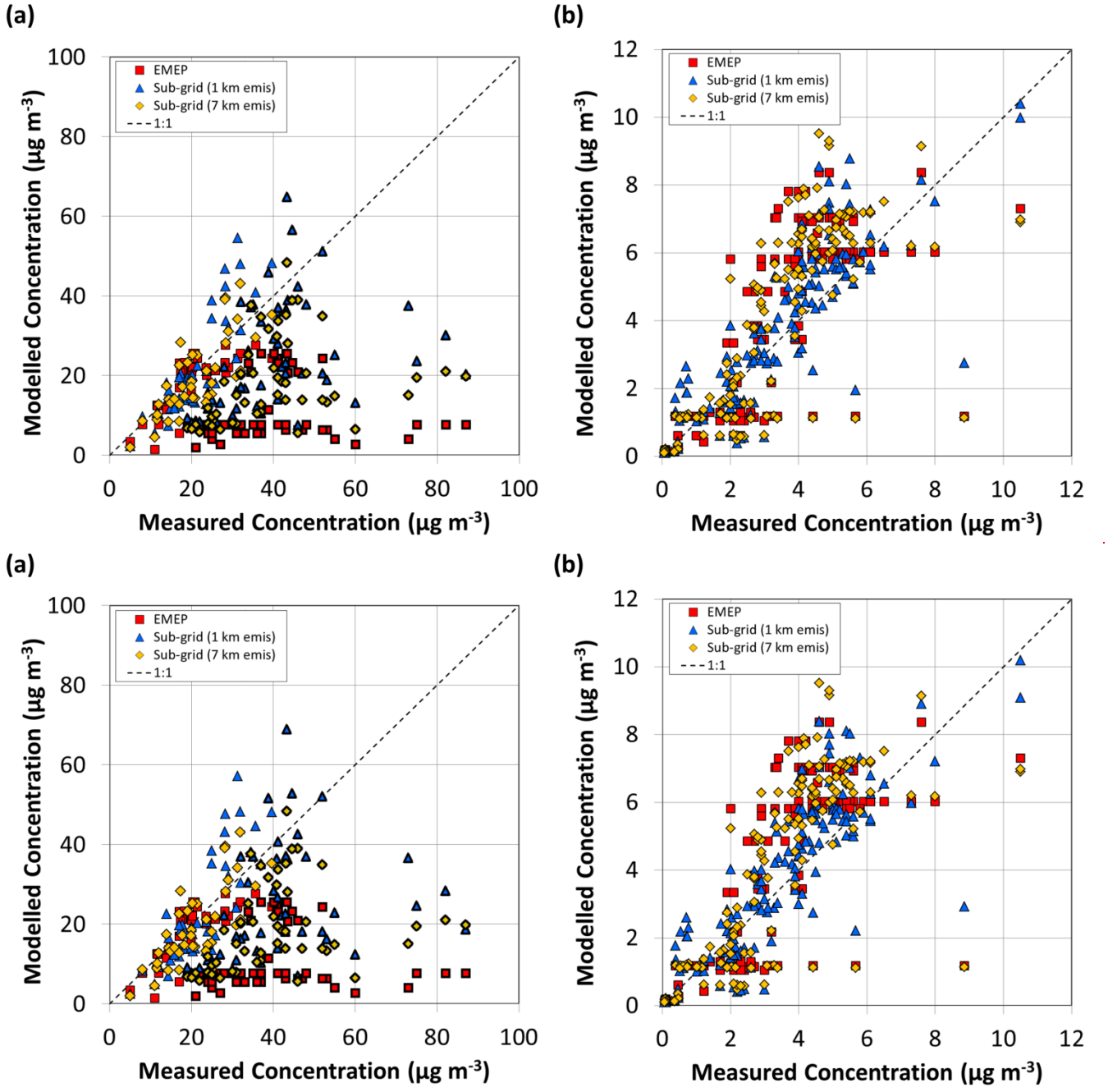


Figure 4: Modelled concentrations plotted against measured values for all sites for (a) NO_2 and (b) NH_3. NO_2 traffic stations are indicated by bold symbol outlines. Plot data provided in the supplementary material.

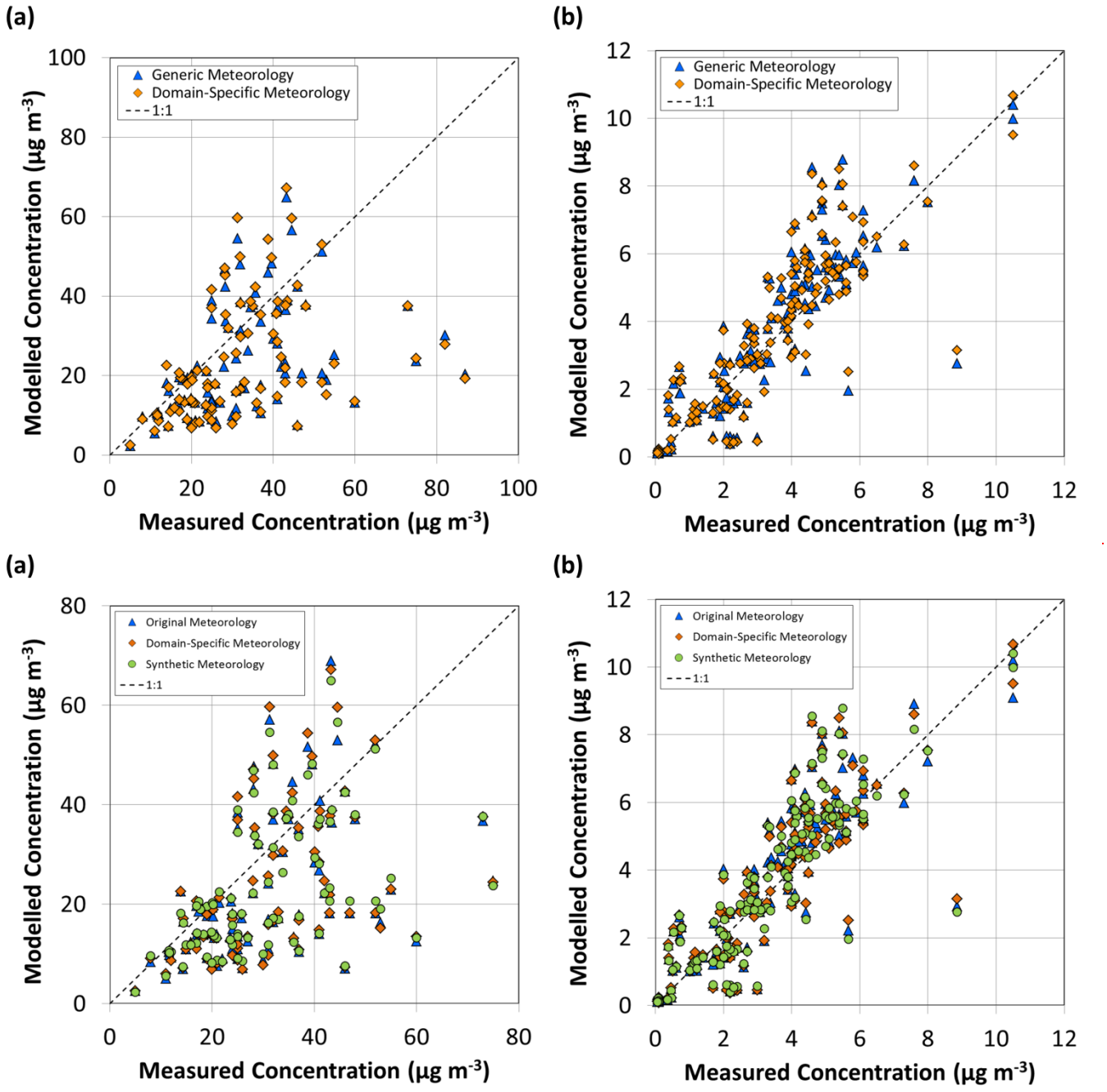


Figure 5: Modelled concentrations plotted against measured values for all sites for (a) NO_2 and (b) NH_3 using ~~both~~ the ~~generic meteorological dataset (LYNE95mod~~original meteorology (as in Figure 4) and using the domain-specific and synthetic meteorological ~~data. Plot data provided in the supplementary material~~datasets.

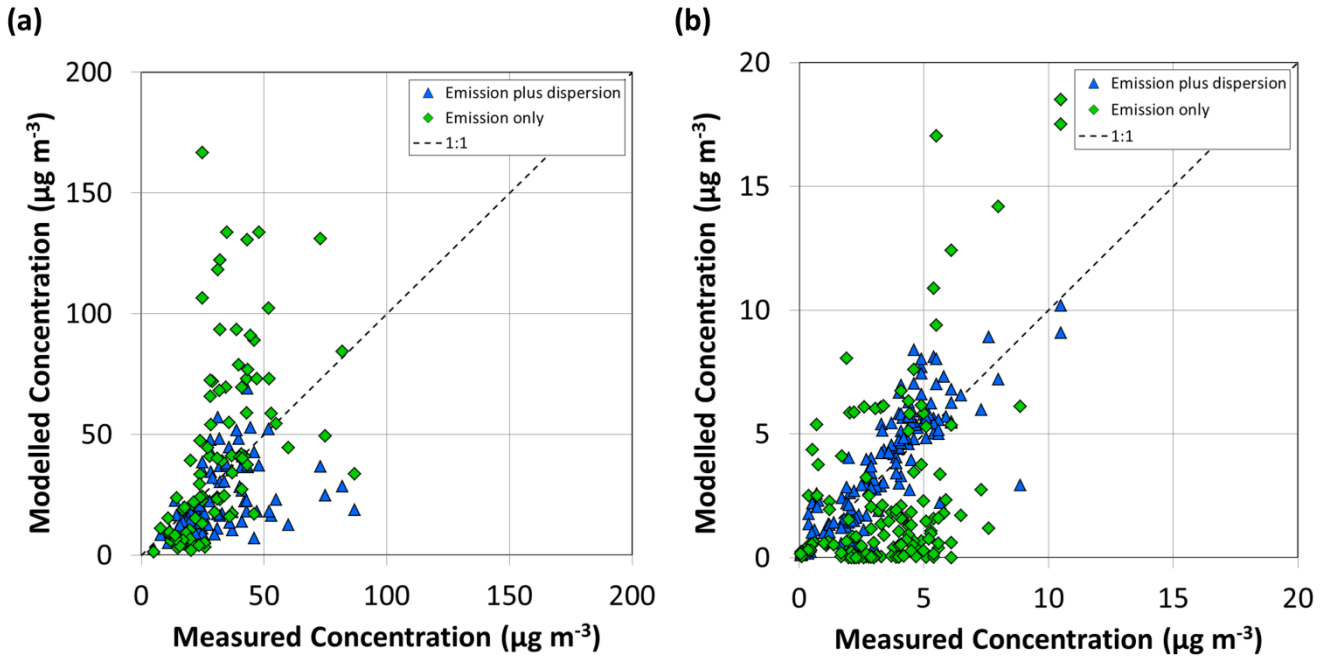


Figure 6: Modelled concentrations plotted against measured values for all sites for (a) NO_2 and (b) NH_3 using the original sub-grid parameterisation (Emission plus dispersion) and using just the spatial distribution of emissions as the sub-grid distribution (Emission only).