Comparing Emission Inventories and Model-Ready Emission Datasets between Europe and North America for the AQMEII Project

George Pouliot[1*], Thomas Pierce[1], Hugo Denier van der Gon[2], Martijn Schaap[2], Michael Moran[3], and Uarporn Nopmongcol[4]

[1] Atmospheric Modeling and Analysis Division, USEPA, RTP, North Carolina, USA

[2] TNO Environment and Geosciences, Utrecht, The Netherlands

[3] Air Quality Research Division, Environment Canada, Toronto, Ontario, Canada

[4] ENVIRON Corporation, Novato, California, USA

For consideration in the AQMEII Special Issue of *Atmospheric Environment*

25 May 2011

* Corresponding author. Email: pouliot.george@epa.gov; Phone: +1-919-541-5475; Fax: +1-919-541-1379; Address: USEPA (MD-E243-04), RTP, NC 27711, USA.

Comparing Emission Inventories and Model-Ready Emission Datasets between Europe and North America for the AQMEII Project

George Pouliot[1*], Thomas Pierce[1], Hugo Denier van der Gon[2], Martijn Schaap[2], Michael Moran[3], and Uarporn Nopmongcol[4]

[1] Atmospheric Modeling and Analysis Division, USEPA, RTP, North Carolina, USA

[2] TNO Environment and Geosciences, Utrecht, The Netherlands

[3] Air Quality Research Division, Environment Canada, Toronto, Ontario, Canada

[4] ENVIRON Corporation, Novato, California, USA

Abstract

This paper highlights the similarities and differences in how emission inventories and datasets were developed and processed across North America and Europe for the Air Quality Model Evaluation International Initiative (AQMEII) project and then characterizes the emissions for the two domains. We focus specifically on the creation of "model-ready" gridded emission datasets for 2006 across the two continental study domains. The practice of creating and processing the two inventories is discussed with a focus on emission factors, spatial allocation, temporal variability, speciation of PM and VOCs, and the mechanics of distributing the data and supporting emission algorithms to the modeling community. The spatial and temporal distribution on common scales is compared for the pollutants of primary concern: NO_x, VOCs, SO_2, $PM_{2.5}$, CO, and NH_3. Because of differences of population distribution, emissions across North America tend to be more heterogeneous in spatial coverage than in Europe. The temporal

patterns in the estimated emissions are largely the result of assumptions used to characterize human activity, with the exception of "natural" emissions, which are modulated by meteorological variability, and emissions from large electric generating units in the U.S., which have the benefit of continuous emission monitors that provide hourly resolved profiles. Emission estimates in both study domains are challenged by several important but poorly characterized emission source sectors, notably road dust, agricultural operations, biomass burning, and road transport. Finally, this paper provides insight on the strengths and weaknesses of emission inventory preparation practices on both continents. One important outcome of this comparison of 2006 emissions between Europe and North America is the greater understanding provided into how the emission estimates developed for the AQMEII project impact regional air quality model performance.

Key words: AQMEII, regional air quality models, emission inventories

1. Introduction

To support the goals of the Air Quality Model Evaluation International Initiative (AQMEII), emission datasets have been created for the European and North American study domains. Guiding principles for constructing these datasets were as follows: (1) that they be based on model inventories used by the air quality management community; (2) that they be distributed to the AQMEII participants in a timely fashion using the (limited) resources available for the project; and, (3) that the datasets be constructed such that their size and format could be conveniently distributed to the international community for use in regional air quality model

simulations with a minimum amount of preprocessing. While the construction and delivery of these datasets are the obvious motivators for this effort, having emission datasets for the same period for two different continental study domains offers the opportunity to investigate and compare how emission inventories and datasets are developed and processed across North America and Europe for regional air quality model application.

In this paper, we focus on the creation of "model-ready" gridded emission datasets for 2006 across the two continental study domains. The practice of creating and processing the two inventories will be discussed with a focus on spatial and temporal allocation. The spatial and temporal distribution on common scales will be compared for the pollutants of primary concern: nitrogen oxides (NO_x), volatile organic compounds excluding methane (VOCs), sulfur dioxide (SO_2), particulate matter 2.5 microns or less ($PM_{2.5}$), carbon monoxide (CO), and ammonia (NH_3). It is hoped that this paper will provide insight on the strengths and weaknesses of emission inventory preparation practices on both continents. One important outcome of this comparison of 2006 emissions between Europe and North America will be the greater understanding on how the emission estimates developed for the AQMEII project might impact regional air quality model performance.

2. **How the emissions were assembled**

In this section, we describe the basis of model-ready inventory datasets created for the European and North American domains.

2.1 European domain

The AQMEII inventory is based on the TNO (Nederlandse Organisatie voor Toegepast Natuurwetenschappelijk Onderzoek or Netherlands Organization for Applied Scientific Research) inventory (Denier van der Gon et al., 2010) that consists of a gridded emission database for the year 2005 across the European region. The dataset is a follow-on to the widely used "Global and regional Earth-system Monitoring using Satellite and in-situ data" (GEMS) emission database by Visschedijk et al. (2007) constructed by TNO in the framework of the GEMS project. The dataset consists of European anthropogenic emissions by country for the ten Source Nomenclature for Air Pollution (SNAP) sectors: energy transformation, small combustion sources, industrial combustion, industrial processes, extraction of fossil fuels, solvent and product use, road transport, non road transport, waste handling, and agriculture. The TNO 2005 emission inventory is not a bottom-up emission inventory combining activity data and emission factors but is set up using official reported emissions at the source sector level, to the extent possible without reducing the overall quality of the inventory. Emissions were downloaded from the European Environment Agency (http://www.eea.europa.eu/data-and-maps/data). However, the reported emissions by an individual country may contain gaps and errors, therefore various consistency checks were made as described in detail by Denier van der Gon et al. (2010). If necessary, gaps and unreliable data were replaced by emissions estimated from the IIASA-GAINS (International Institute for Applied Systems Analysis - Greenhouse Gas and Air Pollution Interactions and Synergies) model (http://gains.iiasa.ac.at/) or TNO's own default emission database. The advantage of this approach is that the inventory made optimal use of national emission experts (who are responsible for reporting emissions to EMEP). The

country emissions and international shipping emissions were spatially distributed on a 0.125 by 0.0625 degree longitude-latitude resolution. Source sector total emissions were broken down into contributions from approximately 200 source categories. Each source category was linked, when possible, to spatial distribution proxies such as population density, power plant capacity and location, road network and traffic intensity (Denier van der Gon et al., 2010).

In the framework of the European Integrated Project MACC (http://www.gmes-atmosphere.eu) the base year 2005 emission inventory was extended to the years 2003-2007. This was done by developing a set of scaling factors as described in detail by Kuenen et al. (2011). Similar consistency checks and gap filling as done for the year 2005 database was performed for each year. For countries where no emission data for 2005 were available, linear interpolation was used between available years. The result was a set of scaling factors relative to 2005 emission by individual source category by country by year. The international shipping emission totals for the individual years in the European domain were taken from the EMEP Centre on Emission Inventories and Projections (CEIP; http://www.ceip.at/). The AQMEII emission inventory is the year 2006 out of this so-called European TNO-MACC inventory.

The dataset distributed to the AQMEII community includes CH_4, CO, NO_x, SO_2, NMVOC, NH_3, PM_{10}, and $PM_{2.5}$. The black carbon (BC) and organic carbon (OC) contributions to primary PM were supplied as well. While the data consist of annual total emissions, temporal factors from the Eurodelta Modelling Intercomparison Exercise (EURODELTA - Stuttgart University/TNO GENEMIS), were made available for calculating hourly inputs for the regional air quality models. The database distinguishes area and point sources. The dataset included

example VOC splits for the CBM-IV and CBM-V mechanisms, but it was assumed (as is common practice in Europe) that chemical speciation would be performed internally by each modeling system. While biomass burning emissions have been developed by researchers in Finland, they were not available for distribution with this dataset. An important limitation in the emission inventories is that semi-natural sources like wind-blown dust and re-suspension emissions were not included in either the anthropogenic and natural emissions data set.

2.2 North American domain

The 2006 AQMEII modeling inventory for the North American domain is comprised of data from the United States, Canada, and Mexico. The primary basis for the inventory is U.S. EPA's 2005 National Emission Inventory, version 3 (NEIv3), which has been translated onto the 2005 modeling platform described in EPA's Technology Transfer Network Clearinghouse for Inventories and Emissions Factors (TTN/CHIEF) (www.epa.gov/ttn/chief/eiinformation.html). Pollutants in the inventory include CO, NH_3, VOC, NO_x, $PM_{2.5}$, and SO_2. For AQMEII, the 2005 dataset was updated for the United States to include 2006 Continuous Emission Monitoring (CEM) data of SO_2 and NO_x from major point sources; 2006 episodic wildland fire emissions; integration of criteria and hazardous air pollutant inventories for benzene, acetaldehyde, formaldehyde, methanol, chlorine, and HCL; and, the allocation of large marine vessel emissions as near-surface, grid cell area-averaged emissions. Fire emissions were based on 2006 daily fire estimates using the Hazard Mapping System fire detections from the National Oceanic and Atmospheric Administration (NOAA) and Sonoma Technology's SMARTFIRE emission processing system (Raffuse et. al, 2009). Criteria emissions were integrated with EPA's

Hazardous Air Pollutant (HAP) inventory for benzene, acetaldehyde, formaldehyde, methanol while chlorine and HCL emissions were obtained directly from the HAP inventory. Version 2 of the 2005 point source inventory was used, which included aircraft and airport emissions. Ethanol-related updates were not included because they were not yet available. Oil and gas emissions in the nonpoint sector were updated for states in the western U.S. Continuous Emission Monitoring (CEM) data from 2006 were used for the Electric Generating Units (EGU) sector. Environment Canada supplied a 2006 inventory with updated surrogates for the domain but no biomass burning. No additional information was available for Mexico, which comes from a 1999 emission inventory (http://www.epa.gov/ttn/chief/net/mexico.html), in which population was the only spatial surrogate used to allocate emissions across the model domain.

The Sparse Matrix Operational Kernel for Emissions (SMOKE) processor (http://www.smoke-model.org/version2.6/html/) was used to process the emissions into a "model ready" format. Use of SMOKE allowed the emissions to be tabulated by source categories, allocated into grid cells, temporally resolved, and output into two different chemical mechanisms. Emissions were separated into the 15 source categories shown in Table 1. These source categories were chosen because of their use in regulatory modeling

To reduce space, the data were distributed in a two-dimensional format rather than in a three-dimensional format. This was accomplished by providing, for major point sources and location-specific wildland fires, the plume top and bottom (in units of meters, above ground level) on an hourly basis. These sources were denoted by latitude/longtitude rather than grid cell id. Emissions for all other sources were given as a grid-cell average value.

As noted above, all the major point sources and wildland fire emission sources were output on an hourly basis. For the remaining grid-cell average emission sources (except for biogenics), to save space and to enable distribution among the AQMEII participants, monthly emissions were provided for a single week, as emissions for most sectors vary little on a week-to-week basis (within a month). Sectors allocated in this manner included afdust, ag, alm_no_c3, nonpt, nonroad, on_nmim2moves, on_nmim2moves, othar, othon, and othpt. In addition, holidays were not accounted for in this modeling inventory, since typically the first full week in the month is assumed. The weekly emissions did account for weekends. Within the SMOKE processing system, hourly time resolution is achieved by using the system's source-category specific temporal allocation factors.

Day-specific emissions were developed for the following sectors: ptnonipm, ptipm, othpt, beis, ptfire. Data in these files include (1) hourly continuous emissions monitoring (CEM) information from electrical generating units (EGU); (2) biogenic emissions based on hourly meteorology; (3) hourly point source emissions and plume rise calculations (based on hourly meteorology and stack operating characteristics); and (4) daily fire emissions for the ptfire sector. For the point source sectors (ptnonipm, ptipm, othpt), three files are provided for each day: (1) layer 1 emissions that are considered "minor" sources with little/no plume rise; (2) a locations file that includes the grid cell locations including the latitude/longitude, row and column, and the location in the grid relative to the origin of the point sources; and, (3) emissions per source "matched" with the location file and including the estimated plume rise based on the

hourly meteorology. To reduce data size and ease transferability, the data were stored in the IOAPI/NETCDF format (http://www.baronams.com/products/ioapi/AA.html).

Gridded biogenic VOC and NO emissions were computed on an hourly resolution for the same chemical species as the anthropogenic emissions. Emissions were based on version 3.14 of the Biogenic Emissions Inventory System (BEISv3.14). Environmental correction factors were derived from the meteorological data generated by the WRF model (see Gilliam et al, this issue). Like the European inventory, it was assumed that sea salt would be calculated internal to most chemical transport models. Currently, lightning, wind blown dust, and geogenic sources are not included in the North American inventory. As indicated above, biomass burning emissions are included in the "anthropogenic" emissions although in most cases, these emissions should be considered natural in origin.

The manner in which emissions are processed for modeling in North America involves an emissions processing system, in our case the SMOKE system, which includes a speciation module to take total VOC and PM emissions and to allocate those emissions to specific chemical classes. For the North American domain, emission data were developed for the CB05 (Yarwood et al., 2005) and SAPRC07 (Carter, 2010) chemical mechanisms. These mechanisms are commonly used in regional air quality model simulations in the United States, and the lumping assumed for VOCs with SAPRC07 enables its emissions to be translated conveniently to other chemical mechanisms in use by the international community.

A few possible limitations of the North American dataset include the following:

(1) Some area source estimates were based on an older 2002 inventory.

(2) Mobile source emissions were based on a 2005 estimate using EPA's NMIM model, which included the MOBILE6 system for on-road emissions. Monthly state-level temperatures were used to create monthly county-level mobile source emission estimates.

(3) It did not include emissions from wind blown dust or NO generated from lightning.

(4) Aircraft emissions were limited to takeoffs/landings/ground operations.

(5) Information from Mexico is limited and all spatial allocations used a single population surrogate.

3. Comparison of emission estimates

To compare emissions between the two modeling domains requires a harmonization of the two source categorization systems: the Source Nomenclature for Air Pollution (SNAP) in Europe and the Source Classification Codes (SCC) in North America. Given that the two systems differ significantly at the fully coded classification level, comparisons are made at a high level using the 11 SNAP sectors and one or two leading digits of the North American Source Classification Code (SCC) system. Table 2 provides a "high level" cross-walk between the two systems that is used for comparing emissions between the two study domains.

Tables 3 and 4 summarize the annual emission estimates for the European and North American domains by pollutant and broad emission categories. For the European and North American domains, we have chosen not to include international shipping estimates in the overview tables but they are present in the emission databases and have included them in the

spatial analysis. Since the European reported totals did not include biogenic or biomass burning emissions, we have not included these in the North American totals to allow for a consistent comparison. Therefore the comparisons in tables 3 and 4 include only land-based anthropogenic sources. A first remarkable and coincidental feature is that the overall total emissions are the same order of magnitude across both inventories within less than 15%; only CO and NH3 differ more (Table 3 and 4). For example, SO_2 is about 15,000 Gg per year for both domains. Carbon monoxide and NO_x are dominated by mobile and combustion sources as expected. VOCs are dominated by non-combustion and mobile sources (also as expected). $PM_{2.5}$ is dominated by combustion (because biomass burning has been excluded from this comparison). PM10 is not included in the comparison because total emissions for the North American domain are dominated by fugitive (road) dust which is not included in the European domain inventory because it is not seen as a primary emission source in Europe and falls outside of any reporting or inventory obligation.

To better understand the differences and similarities between the two inventories, we have calculated the relative percentage of each pollutant by sector for each domain. These percentages are shown in Tables 5 and 6. We have further highlighted the pollutant/sector combinations where the significant differences were found to exist between the two inventories.

Similarities between the two inventories include the following: The relative contribution of source categories to the total emissions is highly similar for all pollutants except for CO. The ratio of pollutant emissions differs between source categories but for each source category is rather similar in both domains (e.g., the VOC/NOx ratio or PM2.5/NOx ratio are similar for the

five categories). Of the five categories, combustion sources have the smallest VOC/NO_x ratio for both domains. Ammonia (NH_3) is dominated by the miscellaneous category (agriculture) (94% for Europe and 86% for North America).

The following differences are noteworthy: While the overall total of CO is similar, the relative differences in combustion and mobile are quite large; CO from combustion is a factor of 4 higher in the North American domain, while CO from mobile is a factor of 3 higher in the European domain. The VOC/ NO_x ratio is much lower in the NA domain than the EU domain (0.13 vs 0.26) for combustion sources, suggesting different assumptions about combustion between the two domains. In Tables 5 and 6, we see that 85% of the CO inventory comes from mobile sources for North America, but for Europe it is 43%. This suggests that either the inventory methods for CO are significantly different between the two domains or the emission standards and emission limit values are different, and we might expect different model performance for CO in the two domains. An explanation, but only partial, is the higher fraction of gasoline passenger cars in the NA domain. Gasoline fuelled cars emit substantially more CO per vehicle km than diesel fueled cars. Tables 5 and 6 suggest two additional pollutant/source combinations of large relative differences: SO_2 from non-combustion sources and $PM_{2.5}$ from combustion sources. The discrepancy for SO_2 from non-combustion sources is not too surprising as this category covers a wide range of activities (Table 2) that may be quite different between the two domains. More interesting is the difference for the category combustion sources. NOx and SO_2 emissions are quite similar in both domains but PM2.5, VOC and CO emissions are relatively enhanced in the European domain (Table 3 and 4). In a future study, it would be

296 worthwhile to explore if this is due to different emission factors or reflects a real difference in
297 type of combustion devices, fuels and fuel qualities.
298
299 Figures 1 through 6 show the spatial patterns of the annual emission estimates for each
300 modeling domain normalized by the grid resolutions. In these figures, we have included
301 international commercial shipping since this information was available for both domains.
302 However, we have excluded biogenic and biomass burning (wildfire) emissions. Emission
303 estimates are displayed on the same logarithmic coloring scale to capture the spatial ranges of
304 variability. In Figure 1, NO_x emission density is largest in urban areas and smallest in rural
305 areas in North America and in Europe. However, in Europe the distribution is less variable
306 compared to North America. For SO_2 emissions the expected spatial patterns in both domains
307 (Figure 2) are only partially visible in the maps. Highest densities in coal-fired electric power
308 burning regions of the Ohio Valley are visible, but in Europe, land based SO_2 emissions are
309 completely dominated by point sources (66% power plants and another 16% industrial
310 combustion). These point sources are not as visible in the emission grids depicted in Figure 2
311 because of the grid spacing used to visualize the emission inventory. The contribution of solid
312 fuels (coal) and fuel oil in residential combustion is clearly visible in figure 2 as a more diffuse
313 pattern and important in Germany and Poland but as pointed out earlier, for SO_2, this is of minor
314 importance compared to point source emissions. $PM_{2.5}$ has its highest densities in the mid-
315 western and south-central regions in North American compared to Europe where the densities
316 are distributed fairly evenly across the domain (Figure 3). CO patterns are similar to NO_x
317 patterns (Figure 4). In contrast, ammonia emissions are dominated by agricultural operations and
318 animal husbandry in rural areas of both continents and lack an urban signal. Ammonia emissions

appear widespread in areas east of the Rocky Mountains, in the Central Valley of California, in portions of France, Germany, Italy, and the Netherlands (Figure 5). Figure 6 shows that VOC emissions are largest in urban areas corresponding to concentrations with population, road transport and industrial solvent use.

Another comparison of the two emission inventories is an analysis of the temporal allocation of emissions by pollutant for each domain. This analysis was performed both for the hourly emission changes (diurnal variation) averaged over the year and for the daily variation during the year including both seasonal changes and weekday/weekend changes. Figures 7-12 show the normalized emission temporal profiles by pollutant for both the North American and European domains. To harmonize the comparison, emissions from biogenic sources, wildfires, and international shipping have been excluded. However, we were unable to harmonize all differences across time zones. For the North American domain, hour of the day emissions are summarized by Universal Time (UT) rather than by Local Time (LT). This causes the hourly emissions, which span four time zones, to appear to have a less pronounced diurnal pattern than the European emissions. For the European domain, the temporal profiles were applied to each SNAP sector to compute the daily emissions and the daily values were divided by the mean. No adjustments were made for local times, so the European domain results represent the normalized temporal profile at LT compared to the North American which are all computed at UT. A first observation from Figures 7-12 is that the European profiles work with a first break down by month causing an artificial jump going from one month to the next (see PM2.5, CO and NH3 for clear examples). For annual analysis, this step function in the emissions will not cause problems but when focusing on an episode that includes days from different months it should be taken into

342 account. Figure 7 shows the normalized temporal profile for NO_x. We see consistent temporal
343 allocation for both domains both seasonally and for weekday/weekends trends. Since mobile
344 sources dominate the NO_x emission variations and this source has consistent activity patterns in
345 both domains, this consistency in the NO_x profiles is not surprising. Figure 8 shows the
346 normalized SO_2 temporal profile for both the European and North American domains. Since the
347 Electric Generating Units (EGUs) dominate the SO_2 emission variations, the temporal pattern in
348 the SO_2 for North America is consistent with the Continuous Emission Monitor (CEM) emission
349 measurements. Lower summer demand for electricity probably accounts for the minimum in
350 SO_2 emissions for the summer in the European domain, compared to the local maximum in SO_2
351 emission in the summer for North America. The European temporal profile for electricity
352 demand has not been updated recently and, although we do not expect a summer peak, we do
353 expect a flatter profile today than is provided here based on data for over 10 years ago. Since
354 fugitive dust dominates the $PM_{2.5}$ inventory in North America, and the temporal allocation is
355 assumed to be relatively flat throughout the year, we see a relatively flat $PM_{2.5}$ profile compared
356 to Europe (Figure 9). The European profile mirrors the SO_2 profile indicating that $PM_{2.5}$ and SO_2
357 are derived from the same source sector in the European inventory. Figure 10 shows the
358 normalized CO temporal profile for both the European and North American domains.
359 Surprisingly, the weekday/weekend trend is opposite phase between North America and Europe.
360 This is because in the North American domain, higher weekend emissions are assumed from the
361 off-road sector (which dominate the CO emission variations) likely due to recreational weekend
362 activities in North America. Lower weekend emissions are assumed in the European domain.
363 An important general observation from the above discussion is that the NA domain appears to
364 lack the summer dip that is characterizing the European combustion related temporal emission

365 profiles (Figures 7-10). Possibly the more extensive use of air-conditioning in the NA domain
366 plays a role, but the discrepancy is quite remarkable and deserves more detailed analysis in the
367 future. Figure 11 shows the normalized temporal profile for ammonia. We see significant
368 differences in the seasonal assumptions from ammonia. For the North American domain, the
369 seasonal variations were derived from inverse modeling techniques (Gilliland et al., 2006). For
370 the European domain, it is a simple combination of agricultural management activities (timing of
371 fertilizing and manuring) combined with assumptions about animal densities and timing of
372 moving cattle from stables into the fields. However, from air quality modeling studies, it is
373 known that the timing of ammonia emissions in Europe is not correct and needs improvement.
374 Figure 12 shows the normalized temporal profile for VOC. Similar to CO, the EU emissions and
375 the NA emissions have opposite phases with respect to weekday/weekend trends. We note that
376 CO and VOC emission trends in weekday/weekend differences are dominant in the North
377 American inventory because of the assumptions made for the off-road sector, which includes
378 recreational activities.
379
380 We now consider the hourly profiles used in each inventory and compute a normalized
381 temporal profile for both North American and Europe. For the European inventory, we take the
382 annualized emission estimates per SNAP sector and calculate the fraction of emissions that are
383 allocated to each hour of the day in local time for all parts of the domain. Then we divide the
384 hourly emissions by the mean to get a normalized profile for each pollutant. Figure 13 shows the
385 mean diurnal profile by pollutant using the annualized emissions excluding international
386 shipping, biogenic, and wildfire emissions. For the North American domain, we applied a
387 similar approach except that we calculated the hourly emissions in UT across the whole domain.

In addition, the North American emission inventory involves a large number of profiles that are applied per pollutant and sometimes on a per county basis. The impact of calculating the mean diurnal profile using so many profiles and at a fixed time reference is to smooth the emission patterns. Figure 14 shows the normalized profile at UT for the North American domain, excluding international shipping, biogenic and wildfire emissions. Although there is a minor difference in methods for the two inventories, we can still see the similarities and differences in emission inventories. First, SO_2 has the least variability or flattest profile for both domains. This is understandable as it is dominated by continuous operating power plants and industrial combustion units. Second, the European inventory has a bimodal pattern of a morning and afternoon peak, whereas the smoother North American inventory has a single peak later in the day. The bimodal pattern in the European domain is due to the morning and evening rush hour in road transport, further enhanced by a similar bimodal peak in residential combustion for heating which starts slightly earlier in the morning and somewhat later, lasting longer in the evening. In North America, most of the diurnal profiles have only one maximum and one minimum per day, with some of the mobile source profiles having two peaks similar to the European inventory. However, these get smoothed out when combined with all emission sources. The ammonia peak is determined by the daily temperature profile with only one peak just after noon in the European domain. In North America, the ammonia emission profile is more a smooth function.

4. Discussion

We now focus on three particular emission sources or sectors because these typically are difficult to estimate regionally. These are fugitive dust (which includes road dust), agricultural operations (including animal husbandry), and mobile sources (on-road and off-road).

Fugitive dust emissions in the North American domain are known to be poorly characterized (Reff et. al, 2009). The current assumptions used to estimate this sector are not robust and need improvement. Therefore, we will note major deficiencies in the North American inventory for fugitive dust. First, the temporal allocation is assumed to flat for the entire year without accounting for weekday/weekend effects, meteorological effects of snow cover and rainfall, and an assumed "transportable fraction" that is applied to the emission estimate to account for capture by near source vegetation. This likely creates an over estimate of this source during the winter and an underestimate during the summer. Nevertheless, fugitive dust is an important contributor to total PM emissions. It is important to note that this category is not properly covered in the European inventories, which do not cover road dust resuspension and include only a very limited contribution – if at all - due to agricultural land management. The main argument to exclude these sources in Europe is that they are considered semi-natural and therefore less affected by policies. Moreover, it is beyond a country's influence whether it has a more arid climate and consequently having larger dust emissions. Inclusion of such sources in official reporting would severely influence the level playing field with respect to PM emission reduction targets in Europe. However, this does not reduce the importance of such dust sources for air

433 quality modeling and a solution to consider the semi-natural dust sources in air quality modeling
434 studies, without disrupting the policy process towards emission reductions, should be pursued.
435
436 Ammonia emissions are dominated by agricultural operations. For North America, extensive
437 work has been done to estimate seasonal variation in ammonia sources through inverse modeling
438 techniques. However, these techniques do not capture the daily or diurnal variations and focus
439 on the larger time scales and seasonal changes. Figure 11 captures the state of the science with
440 respect to ammonia estimates for both European and North American domains, but it is clear
441 more work needs to be done to improve this part of the emission inventory for both continents.
442 A logical step, based on process understanding, is that part of the timing of ammonia emissions
443 e.g., from fertilization, is related to actual climate conditions in a particular year. Thus, the
444 timing of release may have to be (partly) determined inside the air quality models driven by the
445 meteorology of the year of study rather than by generic emission profiles that are the same for
446 each year.
447
448 Mobile sources for the North American domain were estimated using EPA's National Mobile
449 Inventory Model (NMIM) model (EPA, 2005). This modeling system estimates mobile source
450 emissions at the county level for all on-road sources and off-road sources (except for aircraft,
451 locomotive, and marine sources) and uses a county level database of inputs to drive the
452 MOBILE6 (EPA, 2003) and NONROAD models. Future mobile inventories for the US will be
453 use the Motor Vehicle Emission Simulator (MOVES) model (EPA, 2010). Clearly there are
454 some substantial differences between European and North-American emission profiles, most
455 notably for CO, VOC and PM10. The latter refers explicitly to the earlier road dust discussion.

This analysis of the European inventory and the North American Inventory has been limited to those sectors that are common to both inventories. Therefore, since the European inventory did not include biomass burning from wildfires, we have not included the North American estimates in our comparisons. In addition, biogenic emissions are dependent on the meteorological model and are computed internally in most models. Thus, biogenic emission estimates are not discussed here.

5. Conclusions and Recommendations

This summary of the spatial and temporal differences in the European and North American emission inventories used for AQMEII highlights the strengths and weaknesses of the two inventories. The strengths in the two inventories is that they have overall similar magnitudes for most of the pollutants and there is broad consistency between the two. The weaknesses are in the details of the inventories themselves: emission factors, assumptions, and limitations. These comparisons will hopefully provide additional insight into the performance of the different air quality models applied to each domain. We have also highlighted some rather robust discrepancies that warrant further study. These include particular pollutants like CO and PM10 but also important features for air quality modeling, like the timing of emission release throughout the year as well as the diurnal pattern, and the proportional release of pollutants, which may influence atmospheric chemistry. A clear benefit from the AQMEII project is that such differences are now documented.

Acknowledgements and disclaimer

This work was conducted under the auspices of the AQMEII project (http://aqmeii.jrc.ec.europa.eu/aqmeii2.htm) under the leadership of S.T. Rao (USEPA) and Stefano Galmarini (Joint Research Center, European Union). Contributions to the North American emission inventory were made by the Computer Sciences Corporation in Research Triangle Park, North Carolina. The authors also wish to thank K. Foley in helping to create plots using the R software package. Research leading to the European emission inventory was partly funded by the MACC European Union's Seventh Framework Programme (FP7/2007-2013) under Grant Agreement no. 218793. A. Visschedijk, J. Kuenen and R. van Gijlswijk (TNO) are thanked for their part in preparing the European emission inventory. Although this manuscript has been reviewed and approved for publication, it does not necessarily reflect the policy or views of the U.S. Environmental Protection Agency, TNO, Environment Canada, or the Environ Corporation.

References

Carter, W. P. L. 2010. Development of a condensed SAPRC-07 chemical mechanism. *Atmospheric Environment* **44**(2010): pp. 5336-5345.

Denier van der Gon, H.A.C.., Visschedijk, A., Van der Brugh, H., Dröge R., 2010. A high resolution European emission data base for the year 2005, A contribution to UBA- Projekt PAREST: Particle Reduction Strategies, TNO report TNO-034-UT-2010-01895_RPT-ML, Utrecht.

Gilliam et. al., *Atmospheric Environment* this issue

Gilliland, A. B., K. Wyat Appel, R. W. Pinder and R. L. Dennis 2006. Seasonal NH3 emissions for the continental united states: Inverse model estimation and evaluation. *Atmospheric Environment* **40**(2006): pp. 4986-4998.

Kuenen J., Denier van der Gon H.A.C., Visschedijk A., Van der Brugh H.,Gijlswijk R., 2011 MACC European emission inventory for the years 2003-2007, TNO-report TNO-060-UT-2011-00588 Utrecht.

Reff, A., P. V. Bhave, H. Simon, T. G. Pace, G. A. Pouliot, J. D. Mobley and M. Houyoux 2009. Emissions Inventory of PM2.5 Trace Elements across the United States *Environmental Science & Technology* **43**(2009): pp. 5790-5796.

Raffuse S.M., Pryden D.A., Sullivan D.C., Larkin N.K., Strand T., and Solomon R. 2009. SMARTFIRE algorithm description. Paper prepared for the U.S. Environmental Protection Agency, Research Triangle Park, NC, by Sonoma Technology, Inc., Petaluma, CA, and the U.S. Forest Service, AirFire Team, Pacific Northwest Research Laboratory, Seattle, WA STI-905517-3719.

USEPA, 2003 User's Guide to MOBILE6.1 and MOBILE6.2: Mobile Source Emission Factor Model, U.S. Environmental Protection Agency, Ann Arbor, MI.

USEPA, 2005 EPA's National Inventory Model (NMIM), A Consolidated Emissions Modeling System for MOBILE6 and NONROAD, Office of Transportation and Air Quality.

USEPA, 2010 Motor Vehicle Emission Simulator, Office of Transportation and Air Quality. Availble at http://www.epa.gov/otaq/models/moves/.

Visschedijk, A. J. H., Zandveld, P. Y. J., & Denier van der Gon, H. A. C., 2007. High resolution gridded European emission database for the EU integrate project GEMS. TNO-report 2007-A-R0233/B.

Yarwood, G., S. Rao, M. Yocke, and G. Whitten, 2005. Updates to the Carbon Bond chemical mechanism: CB05. Final report to the U.S. EPA, RT-0400675 (http://www.camx.com/publ/pdfs/CB05_ Final_ Report_120805.pdf).

Table 1. Source categories used to distribute emissions across the North American domain

Source type	Description
afdust	Area source fugitive dust from anthropogenic sources, PM10 & $PM_{2.5}$ only
ag	Area source emissions from agricultural operations, NH_3 only
alm_no_c3	Area source emissions from aircraft, locomotive, and marine (except class 3 vessels) (Aircraft includes only takeoff, landing, and ground operations.)
beis	Biogenic VOC and soil NO emissions estimated with BEIS3.14 and meteorology from WRF
nonpt	Area source emissions not included in other sectors (e.g., residential wood combustion)
nonroad	Off-road mobile source emissions from EPA's National Mobile Inventory Model (NMIM)
on_nmim2moves	On-road mobile source emissions (part 1)
on_nmim_plus_ca	On-road mobile source emissions (part 2, includes California)
othar	Area source emissions from Canada and Mexico (no fires from Canada are included)
othon	Mobile source emissions from Canada and Mexico
othpt	Point source emissions from Canada and Mexico
ptipm	Electric generating unit (EGU) point source emissions, includes Continuous Emission Monitoring (CEM) hourly data
ptnonipm	Non-EGU point source emissions (industrial source and no power generation)
ptfire	Wildland fires and prescribed burning fire emissions identified as point sources
seca_c3	Commercial marine port and inter-port Class 3 (C3) vessels defined as having displacement greater than 30 liters per cylinder.

544 Table 2. Categories used to compare European emissions organized by Selected Nomenclature
545 for Air Pollution (SNAP) codes and North American emissions organized by Source
546 Classification Codes (SCC)

Combined category for comparison	SNAP Code	SNAP Description	SCC	SCC Description
Combustion	1	Combustion in energy and transformation industries	1	External combustion boilers
Combustion	2	Non-industrial combustion plants	2	Internal combustion engines
Combustion	3	Combustion in manufacturing industry	21	Stationary source fuel combustion
Non-Combustion	4	Production processes	3	Industrial processes
Non-Combustion	4	Production processes	23	Industrial processes
Non-Combustion	4	Production processes	6	MACT source categories
Non-Combustion	5	Extraction and distribution of fossil fuels and geothermal energy	4	Petroleum and solvent evaporation
Non-Combustion	5	Extraction and distribution of fossil fuels and geothermal energy	25	Storage and transport
Non-Combustion	5	Extraction and distribution of fossil fuels and geothermal energy	33	LPG distribution
Non-Combustion	6	Solvent and other product use	4	Petroleum and solvent evaporation
Non-Combustion	6	Solvent and other product use	24	Solvent Utilization
Non-Combustion	6	Solvent and other product use	6	MACT Source Categories
Mobile	7	Road transport	22	Mobile sources
Mobile	8	Other mobile sources and machinery	22	Mobile sources
Waste Disposal	9	Waste treatment and disposal	5	Waste disposal
Waste Disposal	9	Waste treatment and disposal	26	Waste disposal, treatment, and recovery
Miscellaneous[a)]	10	Agriculture	28	Miscellaneous area sources

547 [a)] semi-natural sources (EU; SNAP 11 and NA; SSC 27) are not included.

548

549 Table 3. Annual 2006 emission estimates for the European study domain (Gg/Year)

Category	CO	NH_3	VOC	NO_x	$PM_{2.5}$	SO_2
Combustion	22979	33	1831	7175	1619	13868
Non-Combustion	5254	167	7954	721	660	980
Mobile	22421	86	4511	8642	598	501
Waste Disposal	1726	121	148	36	114	14
Miscellaneous	224	5885	657	197	211	8
Total	52604	6292	15101	16771	3202	15371

550

551 Table 4. Annual 2006 emission estimates for the North American study domain (Gg/Year)

Category	CO	NH_3	VOC	NO_x	$PM_{2.5}$	SO_2
Combustion	5619	63	843	6206	1150	12026
Non-Combustion	3009	205	7960	1724	758	2708
Mobile	69735	291	6913	10810	962	499
Waste Disposal	1416	25	375	131	241	27
Miscellaneous	1949	3679	495	61	561	20
Total	81728	4263	16586	18932	3672	15280

552

553

554 Table 5. Relative distribution of annual 2006 emission estimates for the European study domain

555 by pollutant with entries of interest highlighted.

Category	CO	NH_3	VOC	NO_x	$PM_{2.5}$	SO_2
Combustion	44%	1%	12%	43%	51%	90%
Non-Combustion	10%	3%	53%	4%	21%	6%
Mobile	43%	1%	30%	52%	19%	3%
Waste Disposal	3%	2%	1%	0%	4%	0%
Miscellaneous	0%	94%	4%	1%	7%	0%

556

557 Table 6. Relative distribution of annual 2006 emission estimates for the North American study

558 domain by pollutant with entries of interest highlighted.

Category	CO	NH_3	VOC	NO_x	$PM_{2.5}$	SO_2
Combustion	7%	1%	5%	33%	31%	79%
Non-Combustion	4%	5%	48%	9%	21%	18%
Mobile	85%	7%	42%	57%	26%	3%
Waste Disposal	2%	1%	2%	1%	7%	0%
Miscellaneous	2%	86%	3%	0%	15%	0%

559

560

561

562

563 List of Figures

579

580

Figure 1

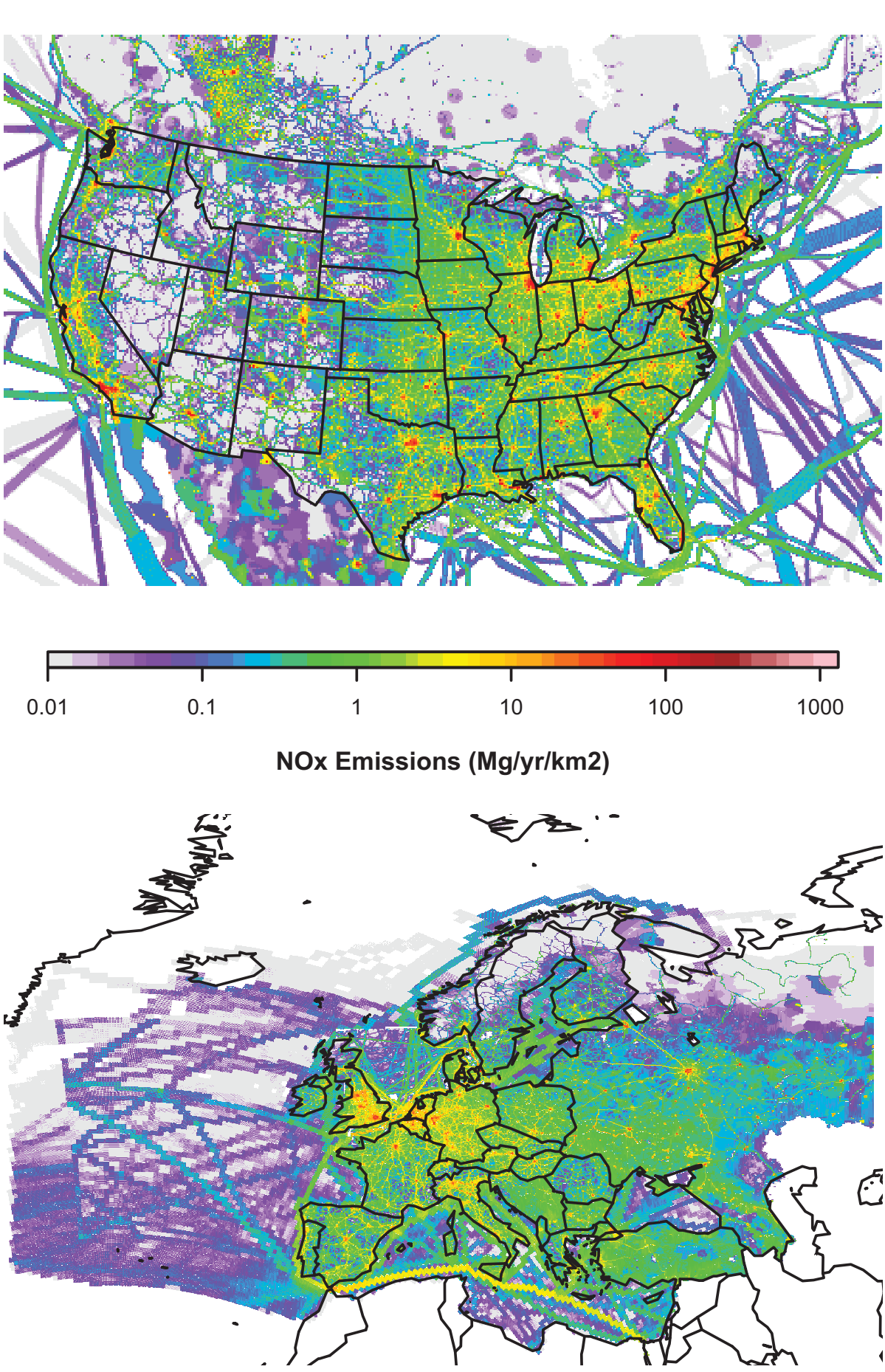

NOx Emissions (Mg/yr/km2)
0.01
0.1
1
10
100
1000
NOx Emissions (Mg/yr/km2)

Figure 2

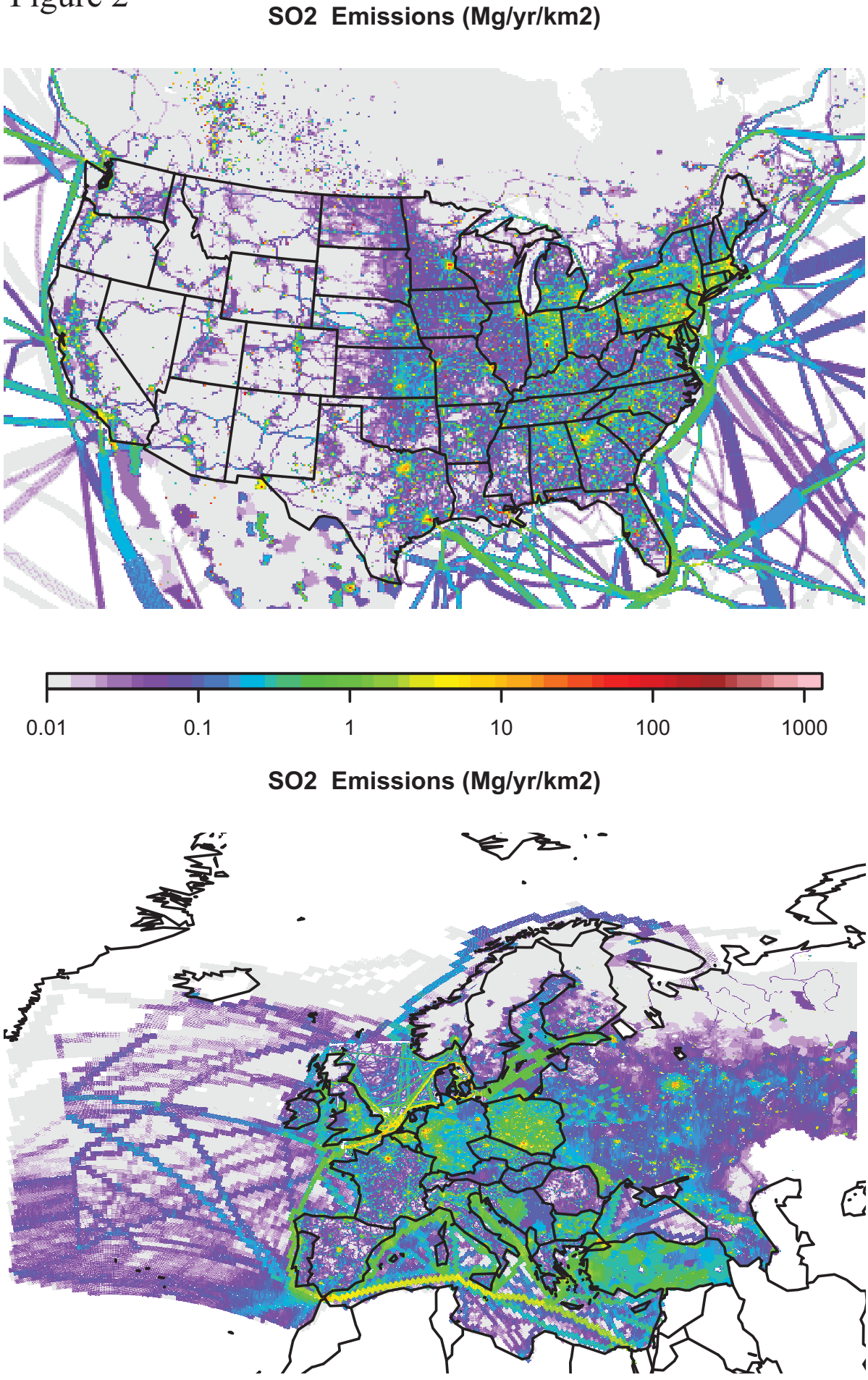

SO2 Emissions (Mg/yr/km2)
0.01
0.1
1
10
100
1000
SO2 Emissions (Mg/yr/km2)

Figure 3

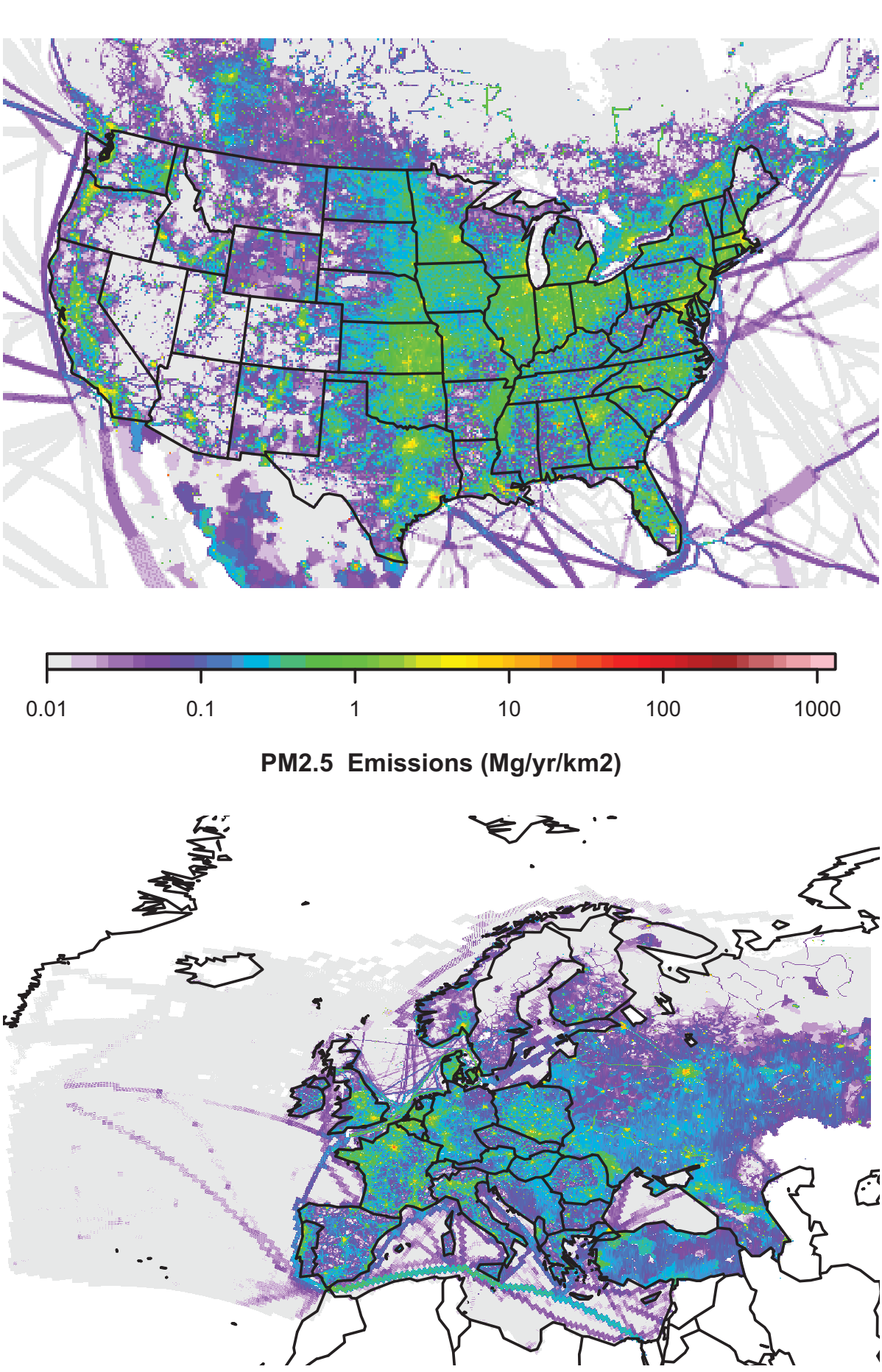

PM2.5 Emissions (Mg/yr/km2)
0.01
0.1
1
10
100
1000
PM2.5 Emissions (Mg/yr/km2)

Figure 4

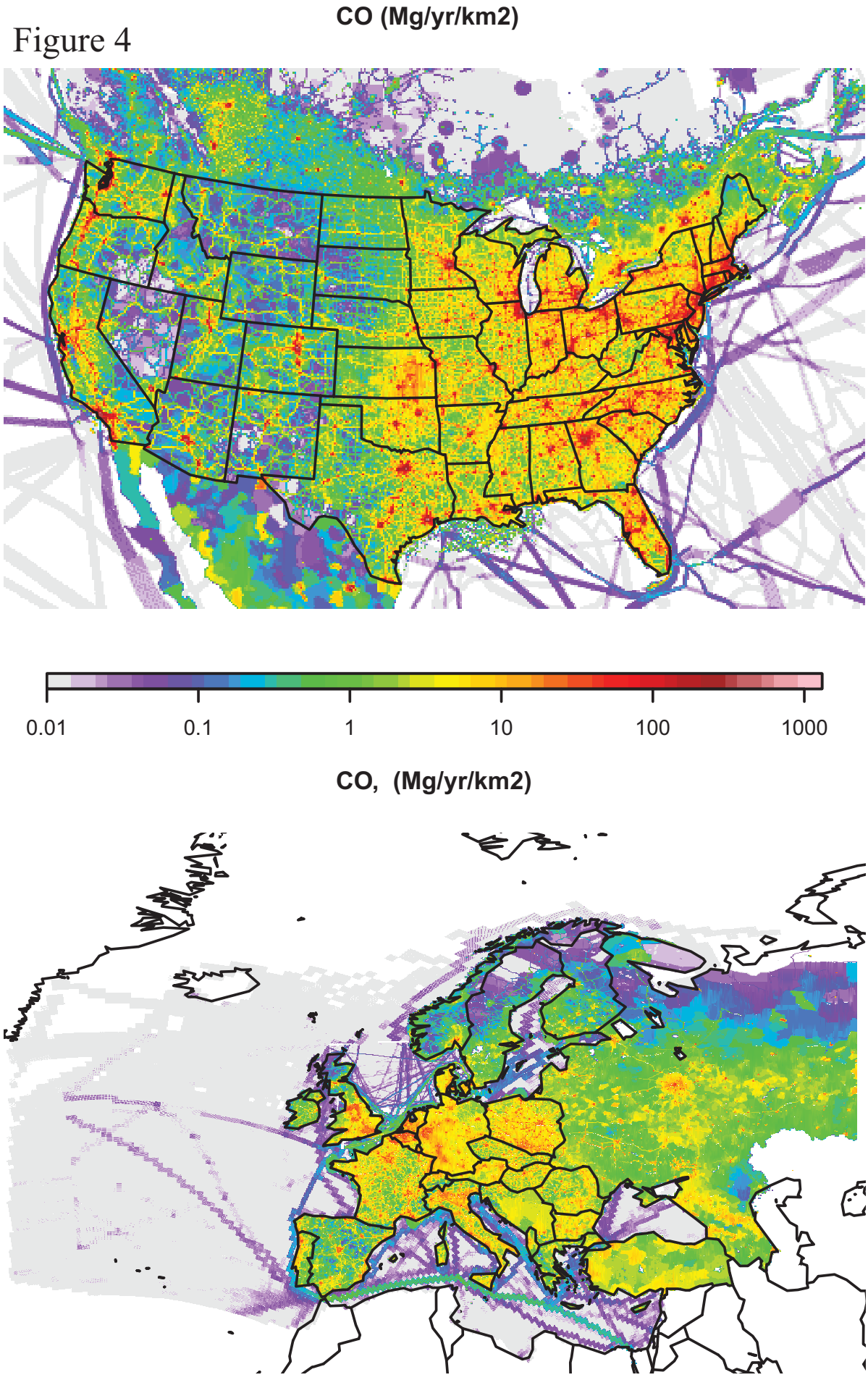


Figure 5

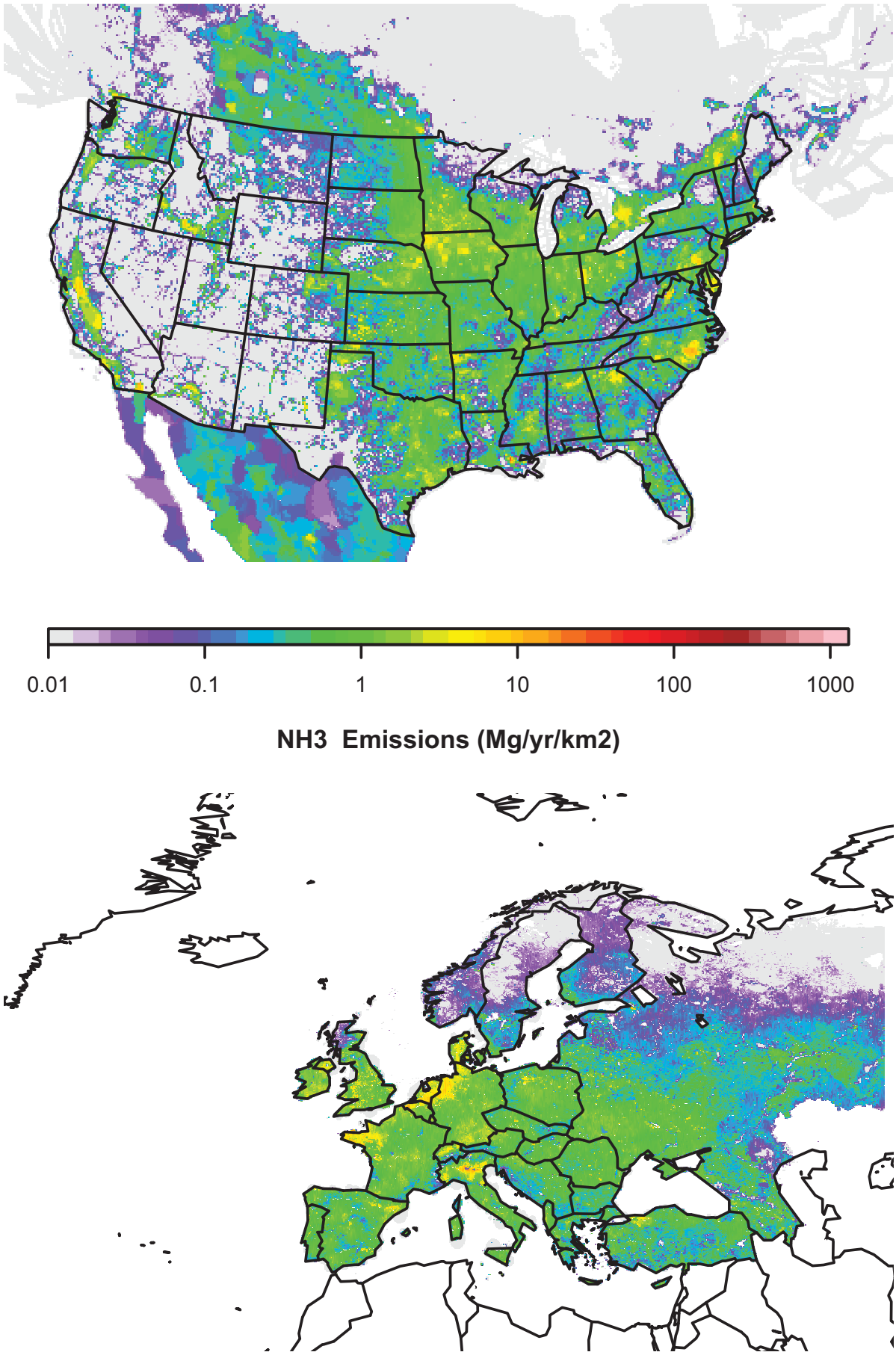
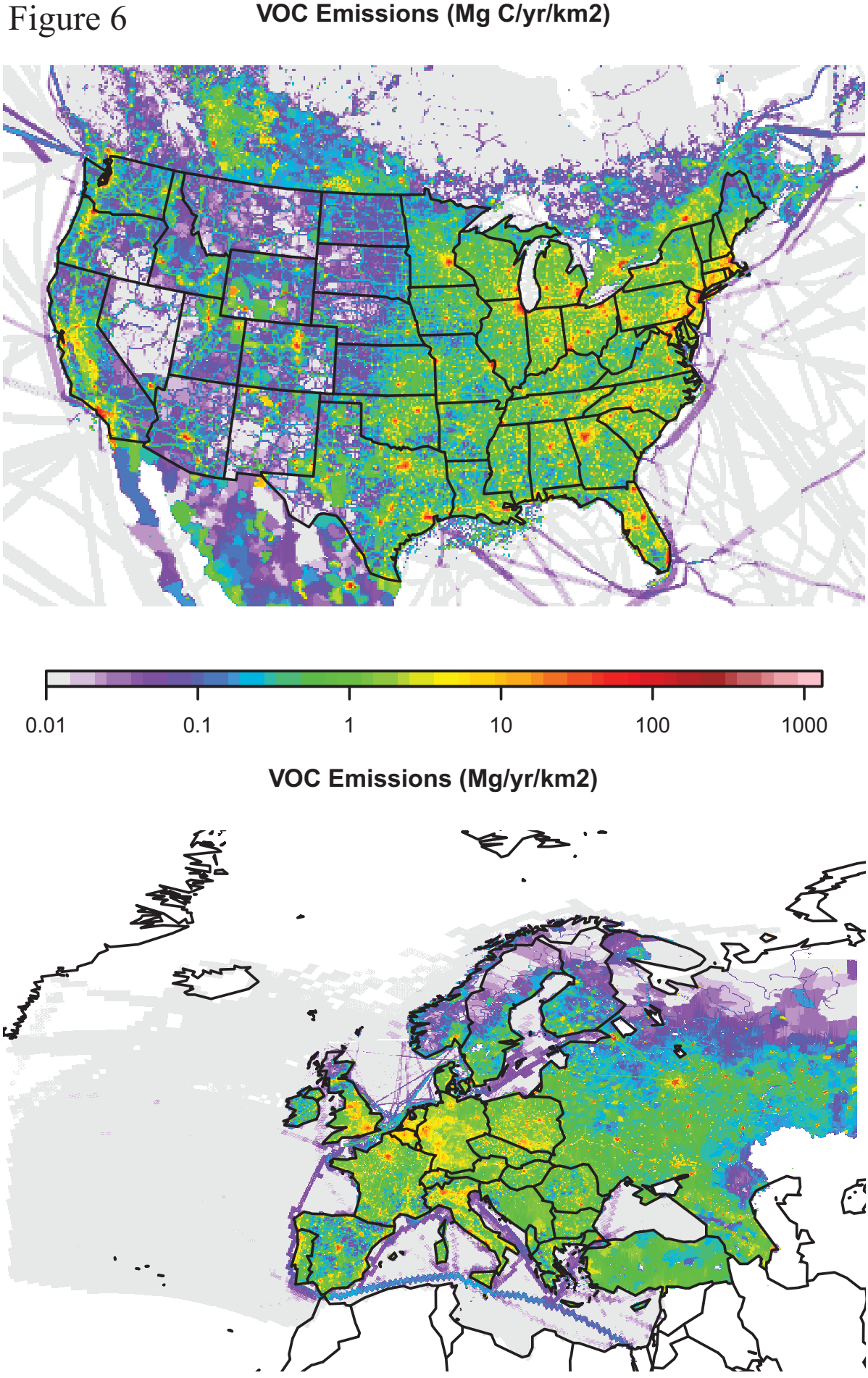
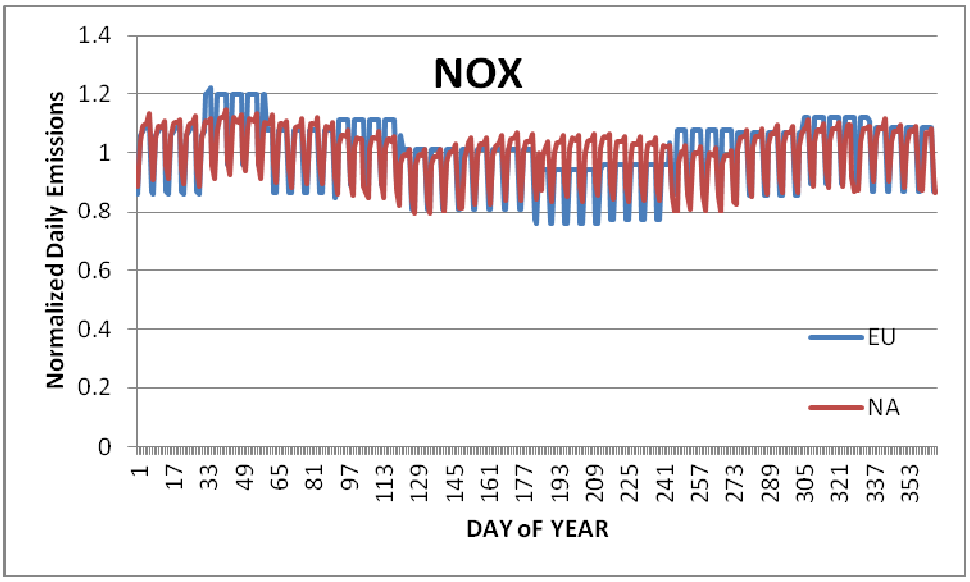

NH3 Emissions (Mg/yr/km2)
0.01
0.1
1
10
100
1000
NH3 Emissions (Mg/yr/km2)

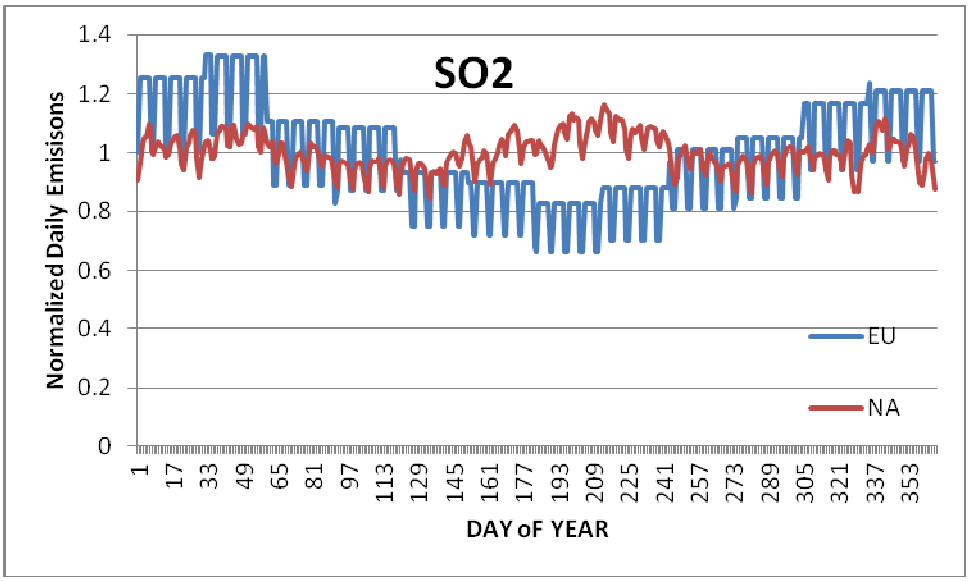
Figure 6


VOC Emissions (Mg C/yr/km2)
0.01
0.1
1
10
100
1000
VOC Emissions (Mg/yr/km2)

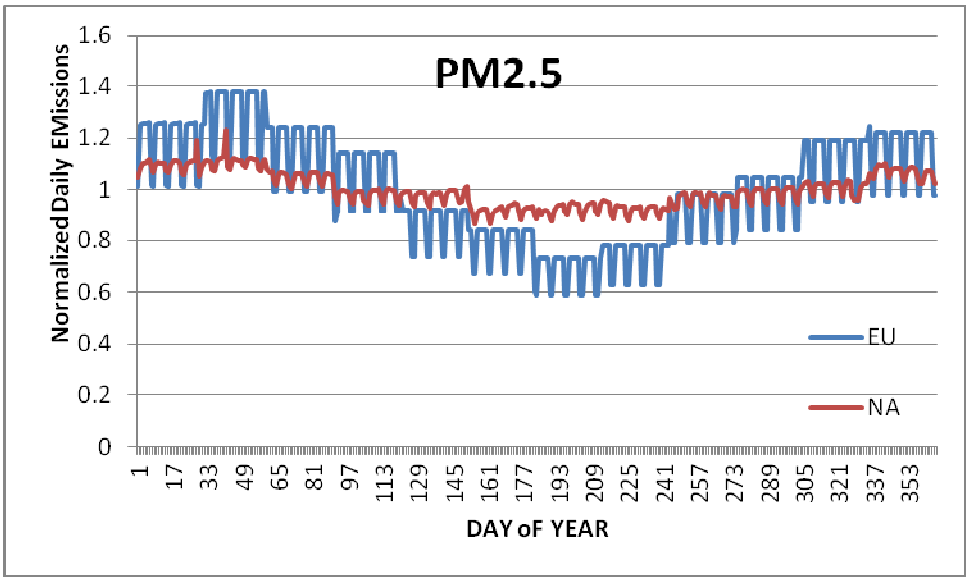
581


NOX
Normalized Daily Emissions
1.4
1.2
1
0.8
0.6
0.4
0.2
0
1
17
33
49
65
81
97
113
129
145
161
177
193
209
225
241
257
273
289
305
321
337
353
DAY oF YEAR
EU
NA

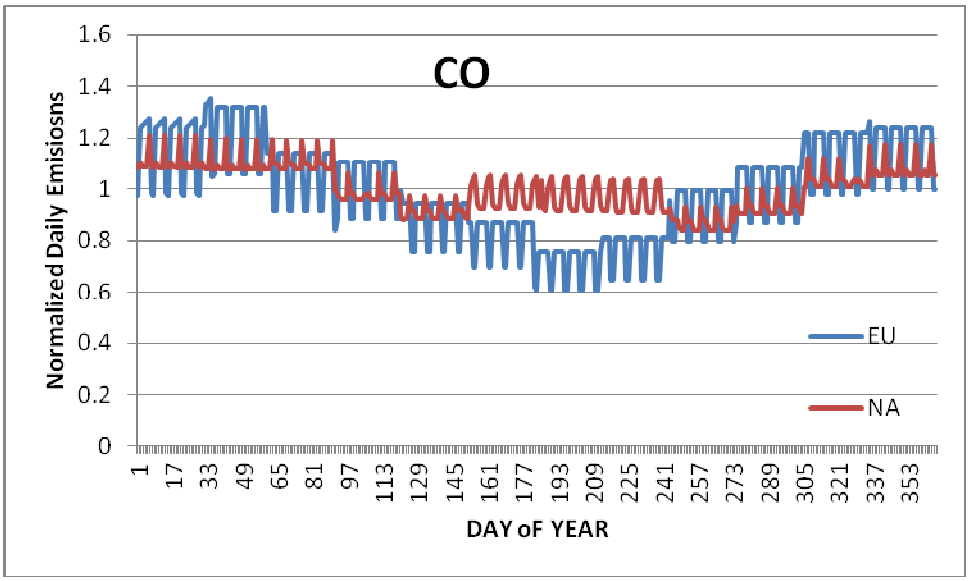
582

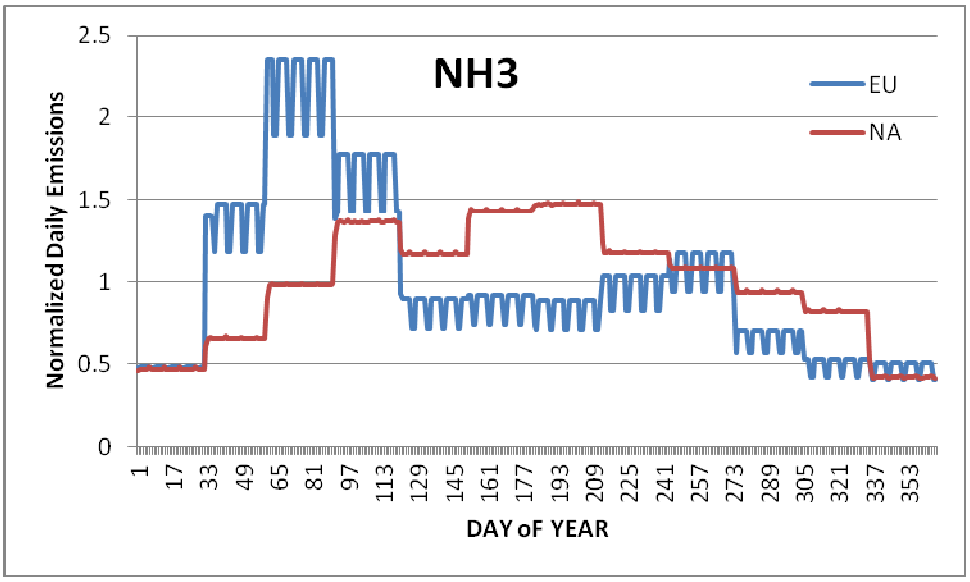

SO2
Normalized Daily Emisisons
1.4
1.2
1
0.8
0.6
0.4
0.2
0
1
17
33
49
65
81
97
113
129
145
161
177
193
209
225
241
257
273
289
305
321
337
353
DAY oF YEAR
EU
NA

583


PM2.5
Normalized Daily EMissions
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
1
17
33
49
65
81
97
113
129
145
161
177
193
209
225
241
257
273
289
305
321
337
353
DAY oF YEAR
EU
NA

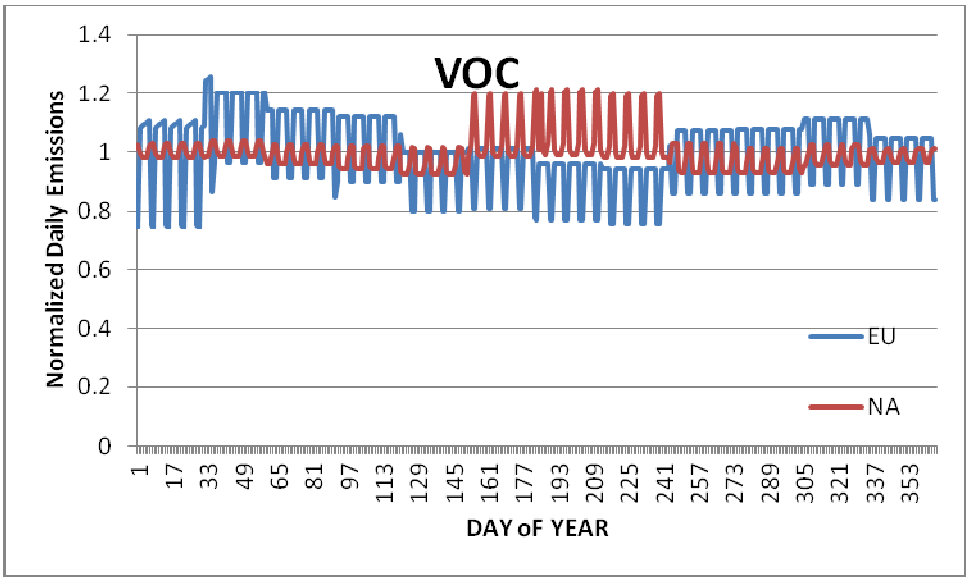
584


CO
Normalized Daily Emisiosns
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
1
17
33
49
65
81
97
113
129
145
161
177
193
209
225
241
257
273
289
305
321
337
353
DAY oF YEAR
EU
NA



585

586

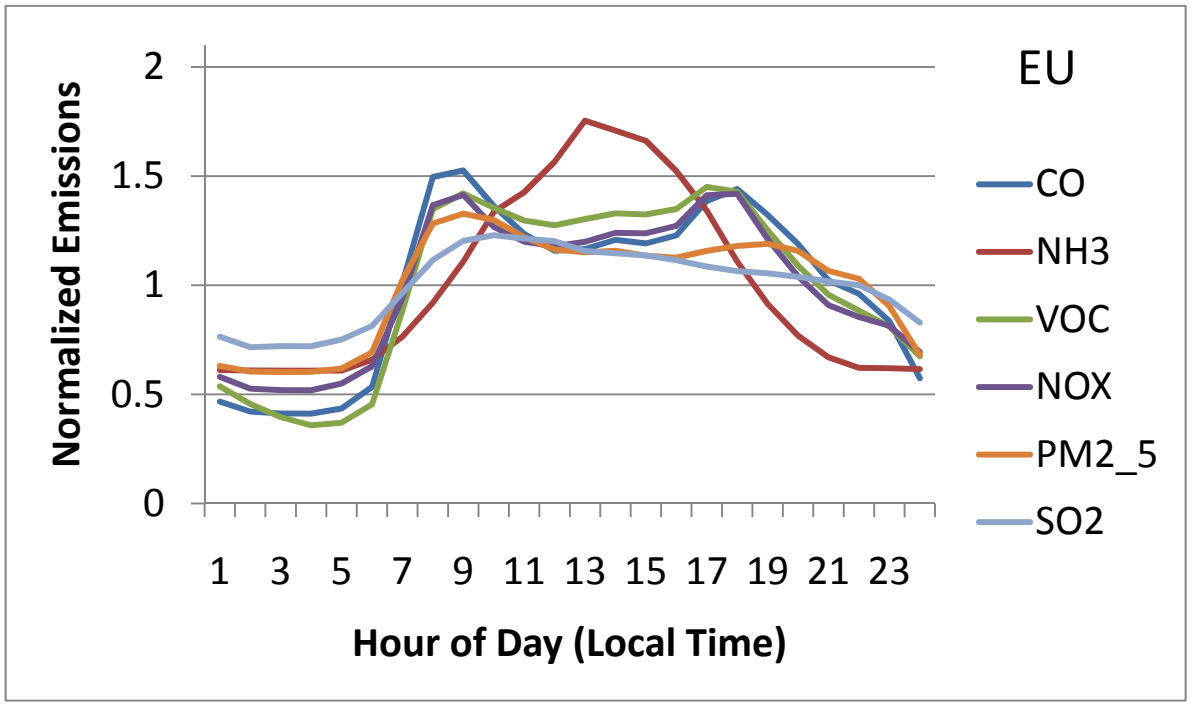


587

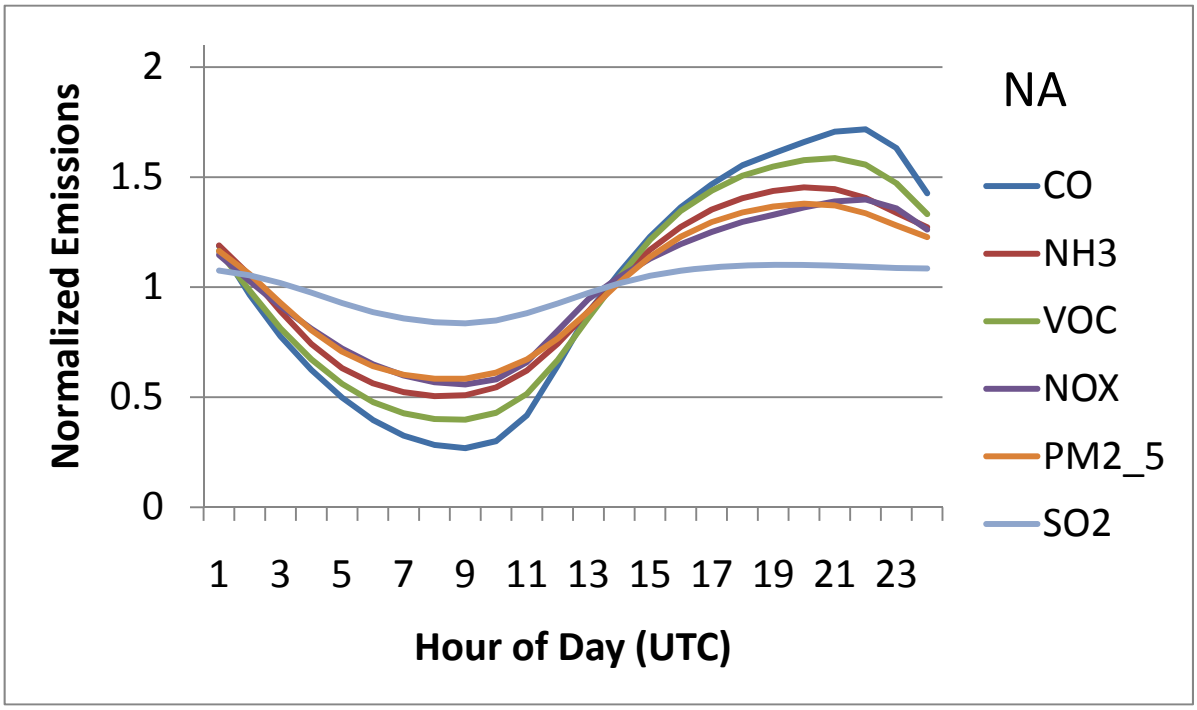
588

589

590



591



592

593

594