Analysis of the Emission Inventories and Model-Ready Emission Datasets of Europe and North America for Phase 2 of the AQMEII Project

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Abstract

This paper highlights the development of the emission inventories and emission processing for Europe (EU) and North America (NA) in the second phase of the Air Quality Model Evaluation International Initiative (AQMEII) project. The main purpose of the second phase of the AQMEII project is to understand the importance of coupled meteorological-chemical models in our understanding of the feedback of chemistry on the meteorology. A second purpose of the second phase of the AQMEII project is to explore the differences between EU and NA in a dynamic evaluation of two modeling years (2006 and 2010). For the EU domain, there was substantial decrease in CO (-19%), NH_3 (-11%), and SO_2 (-12%) emissions between phase 2 and phase 1 emissions for 2006. For the NA domain, there were decreases in CO (-10%), non-methane hydrocarbons (-5%), $PM_{2.5}$ (-8%), PM10 (-18%), SO_2 (-12%), with an increase of 4% in NOx. Between 2006 and 2009, considerable emission reductions were achieved for 17 EU

countries, Norway and Switzerland as well as EU-Non-Member States, for all emissions classes aside from NH_3, which increased. Non-EU countries showed little change in emissions levels, though this may be a result of poor data quality. Shipping emissions decreased for PM and SO_2 due to Sulfur Emission Control Areas on the North Sea and the Baltic Sea, while increasing for other species. Between 2006 and 2010, estimated US NOx emissions decreased by 17%, SO_2 by 29%, CO by 21%, $PM_{2.5}$ by 12%, PM10 by 7%, NMHC by 4% and NH_3 by 2%. Between 2006 and 2010, biogenic emissions in North America increased in the east and decreased in the west, due to regional temperature differences between the years. The European emissions of isoprene peaked earlier in 2006, but achieved higher levels in 2010.

Highlights

We compared the 2006 anthropogenic emission estimates between phase 1 and phase 2.

We compared the 2006 and 2010 anthropogenic emission estimates.

We compared the biogenic emission estimates from BEIS and LOTOS_EUROS for 2006 and 2010

Key words: AQMEII; regional air quality models; emission inventories, emission preparation for modeling; biogenic emissions

1. Introduction

A key input to all regional air quality models is the emission estimates that provide the chemical forcing for chemical transport models. To support one of the protocols of the second phase of the Air Quality Model Evaluation International Initiative (AQMEII), emission datasets were created for the European and North American study domains, to be used by the participating modeling groups. The domains were not changed from those used in the first phase of AQMEII, and descriptions can be found in Pouliot et al. (2012). For this second phase of the project,

guiding principles for the construction of the emission datasets included: (1) consistent base inventories be applied for the EU and NA domains; (2) emission datasets be made available to all modeling groups for both 2006 and 2010 study years; (3) consistent methodologies be used in the two study years so that any differences in emission estimates arise from actual trends and not differing methodologies.

Here, we focus on relative differences in the emission inventories in order to provide insight into the evaluation of the chemical transport modeling results. This necessitated collecting emissions data and generating new model input emissions relative to phase 1 of AQMEII – comparisons with the earlier emissions highlight changes in methodologies between the two phases of the experiment, while ensuring that the emissions for the current phase are internally consistent. This paper focuses on two aspects of the emission inventory: (1) how the emission inventories in phase 2 compare to the emission inventories used in phase 1; (2) the emission trends between 2006 and 2010 that provide a "signal" to the chemical transport modeling for the two years. Numerous additional inputs and assumptions are used in the development of model-ready emission datasets, including annual emission estimates, and their temporal allocation, spatial allocation, and chemical speciation. The purpose of this paper is to focus on relative differences in the emission inventories in order to provide insight into the evaluation of the chemical transport modeling results. We summarize the biogenic emission estimates two of the biogenic models shown in Table 1 of Im et al. (2014).

2. How the emissions were assembled

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

2.1 European domain

A major difference between the EU and NA AQMEII domains is that, despite being of about equal size, the European domain consists of many individual countries. European countries are party to the Convention for Long-Range Transboundary Air Pollution (CLRTAP; http://www.unece.org/env/lrtap/), which requires parties to the convention to report their emissions to European Monitoring and Evaluation Programme (EMEP), a co-operative program for monitoring and evaluation of long-range transmission of air pollutants in Europe. The reporting follows well-defined Guidelines and asks countries to complete a pre-defined template with emissions by year, pollutant and sector (defined by the Nomenclature for Reporting; NFR). These emissions data can be accessed through the EMEP Centre on Emission Inventories and Projections (CEIP; http://www.ceip.at). Despite this organizational framework, the available emissions data are not directly suitable or optimal for air quality modeling. In many cases gaps and errors exist in the reported emission data. Consistency in emissions reporting for consecutive years is a particular problem. Moreover the spatial resolution of gridded data is rather coarse (currently still ~50 x 50 km) and not consistent with the requirements of current air quality models. The procedure to process, gap fill, and redistribute the European emissions data on a high- resolution (0.125° x 0.0625° longitude-latitude) grid is described by Kuenen et al., 2014. The time series of the most recent data set covers the years 2003-2009: an update to the years 2003-2011 is foreseen for summer 2014.

For AQMEII phase 2 the years 2006 and 2010 were selected as the years of study. Unfortunately, for the European domain no emissions data for the year 2010 were available in fully processed form at the start of AQMEII phase 2. Therefore, it was decided to use year 2009

data as the best approximation of year 2010 for the anthropogenic emissions data. Biogenic VOC emissions, driven by year-specific meteorology (see section 5), were calculated for the years 2006 and 2010.

2.1.1. European fire emission data for 2006 and 2010

The European wildfire emissions for 2006 and 2010 were provided by the Finnish Meteorological Institute using the IS4FIRES model (Sofiev et al., 2009). The current fire emission dataset has been obtained by re-analysis of fire radiative power data obtained by MODIS instrument onboard of Aqua and Terra satellites. The algorithm of the emission evaluation and comparison of the results for Europe with the GFEDv2 (Giglio et al., 2006) is presented in the IS4FIRES description (Sofiev et al., 2009). The IS4FIRES output data for Europe consist of daily emissions data at a 0.1 x 0.1 degree spatial resolution), were provided in a netcdf format and described in detail by Soares et al. (2014, this issue). We refer to Soares et al. (2014, this issue) for further details including an uncertainty analysis of the European fire emissions data.

2.2 North American domain

The emissions for the North American domain used for the 2006 and 2010 study years were based on selected national emission inventories from the USA, Canada, and Mexico.

2.2.1 US emissions

For the US, the 2008 National Emission Inventory (NEI) (version 2, released April 10, 2012) was used as the basis for both the 2006 and 2010 model-ready emission datasets (http://www.epa.gov/ttn/chief/net/2008inventory.html). The 2008-based modeling platform

dated 12/14/2012 at http://www.epa.gov/ttn/chief/emch/index.html#2008 provided all the inputs and datasets for processing with SMOKE (http://cmascenter.org/smoke). These files contain the chemical speciation files, the temporal allocation, and spatial allocation used for emission processing with SMOKE. A technical support document (Mason et al. 2012) for this modeling platform containing the full details of the inventory preparation and processing is available. We will focus on year-specific updates made to particular sectors in this paper. Table 1 provides a list of sectors and their descriptions for the US inventory. We note that the US inventory has been divided somewhat differently compared to phase 1. Notes indicating differences between sector names in phase 2 versus phase 1 are included in Table 1.

Year-specific updates were made for the following US sectors: Electric Generating Unit (EGU) point sources, wildfires and prescribed burns, on-road mobile sources, residential wood combustion (temporal allocation only), biogenic sources, and area source fugitive dust, each of which will be discussed later. All other sectors (nonpoint sources, off-road (or nonroad) sources, non-EGU point sources, agricultural ammonia sources, agricultural fires, commercial marine vessels classes 1 and 2, and commercial marine vessels class 3) were assumed to be constant for the two study years.

Emission estimates for SO_2 and NO_x for the EGU sector were based on the Continuous Emission Monitors (CEMs) database from EPA's Clean Air Markets Division (CAMD) (http://ampd.epa.gov/ampd). For all other EGU sector pollutants, the emissions were allocated using the hourly heat inputs divided by the annual heat input as a temporal allocation factor multiplied by annual emission estimates. The annual emission estimates for pollutants other than

164 NO_x and SO_2 were based on the 2008 NEI and did not change for the two study years of 2006
165 and 2010 for the EGU sector. For wildfires and prescribed burns, emission estimates were
166 based on year-specific satellite data and SMARTFIREv2 for 2010 and SMARTFIREv1 for 2006
167 (Raffuse et al. 2009). The SMOKE/MOVES modeling system (a combination of the MOVES
168 model with the SMOKE emissions processing system) was used to estimate mobile source
169 emissions. This is in contrast to MOBILE6 modeling system used in phase 1 of AQMEII. The
170 MOVES model creates emission factors (not emission totals) by county, pollutant, vehicle mode,
171 vehicle type, temperature bin, and road type for the SMOKE system. In addition to the MOVES-
172 generated emission factor inputs, SMOKE uses year-specific hourly meteorological data
173 (temperature and relative humidity) and vehicle activity data, to create model-ready emissions. In
174 phase 1 of the AQMEII project, MOBILE6 emission estimates were created at the county level
175 using monthly average temperatures. The processing required by MOVES can be very time
176 consuming when applied to the entire continental US, due to the large number of combinations
177 of the inputs, and the high level of detail in the inputs themselves on a county basis. MOVES-
178 generated emission factors were already available for 2005 and 2009 at the inception of phase 2
179 of AQMEII – these were used for this project rather than reprocessing 2006 and 2010 separately,
180 in order to meet AQMEII-2 emissions delivery requirements. Vehicle activity data from 2006
181 and 2010 along with these emission factors were used to create the model-ready mobile source
182 emissions. To further reduce processing requirements, representative counties by state were used
183 rather than county-specific factors.
184 For residential wood combustion and anthropogenic fugitive dust sectors, the annual estimate
185 for both 2006 and 2010 remained the same as for 2008 but the temporal allocation was a function
186 of the meteorological conditions. For residential wood combustion, the annual estimates are

temporally allocated as a function of 2 meter temperature derived from a previously created annual WRF simulation. For anthropogenic fugitive dust, the emissions are adjusted downward based on the soil moisture and snow fields from a WRF simulation of that particular year (Appel et al. 2013). This adjustment was applied after the emissions were processed with SMOKE but before the chemical transport model simulation. Some modeling groups used the model-ready emissions input files from SMOKE; here it has been applied to both US and Canadian fugitive dust emissions. However, if the pre-processed annual emission estimates were used, then the adjustment would not carried out by all models making use of the combined inventory – the Environment Canada simulations were based on separate processing of the same inventory. Without this correction, a subsequent degradation of $PM_{2.5}$ and PM_{10} performance would be expected. Biogenic Emissions based on BEIS3.14 for the Model labelled "M14" from Group "US6" for the NA domain are discussed in later in this paper in section 5.2. For a complete summary of the biogenic models used in this phase 2 of AQMEII, see Table 1 of Im et al, (2014).

2.2.2 Canadian and Mexican emissions

Canadian emissions were based on the national 2006 Criteria Air Contaminants Emissions Inventory (Environment Canada, 2008; Sassi et al., 2010). This inventory was used to represent Canadian emissions for both 2006 and 2010. The version of the Canadian inventory used for AQMEII-2 contains more recent updates compared to the 2006 Canadian inventory used in AQMEII phase 1 (Pouliot et al., 2012). These updates included revised residential wood combustion (RWC) emissions, which were generally revised downward (Moran et al., 2012), and a more detailed description of agricultural ammonia emissions (from 10 to 53 subsectors and from provincial-level to agricultural-census subdivision level: c.f. Lillyman and Buset, 2009),

which were 3% lower overall than in the older inventory version. Canadian wildfire and prescribed burning emissions were not used in either AQMEII phase because they were only available as annual values without any information on fire timing or location that could be used for temporal and spatial allocation. Lack of Canadian fire emissions may negatively affect model performance in the northern part of the NA domain. Note that the same downward adjustment applied to U.S. anthropogenic fugitive dust emissions based on day-specific soil moisture and snow fields was also applied to Canadian anthropogenic fugitive dust emissions in the processed emissions discussed here.

Mexican emissions were based on a projected national 2008 Mexican inventory (Wolf et al., 2009). This inventory was used to represent both 2006 and 2010 Mexican emissions. This projected 2008 inventory was a change from AQMEII phase 1, which had used a regional 1999 inventory. Table 2 contains a list of the source sectors and their descriptions for the Canadian and Mexican part of the 2008-based modeling platform inventory for North America. Canadian and Mexican emissions were separated into their own sectors in phase 2 whereas they had been combined in phase 1.

3. Emission Differences between phase 1 and phase 2 for the year 2006

We begin with a detailed comparison of the emissions for the study year of 2006 from phase 1 and phase 2 of the AMQEII project.

3.1 European Domain

The emissions data prepared and used in the AQMEII project phase 1 and 2 were respectively partly developed in the framework of the EUFP 7 projects: Monitoring Atmospheric Composition and Climate (MACC) and the successor to MACC (MACC-II) (https://www.gmes-atmosphere.eu). The AQMEII phase 1 year 2006 emissions were a subset of a time series of a dataset constructed for the years 2003-2007. These data are described in more detail by Pouliot et al., 2012 and Kuenen et al., 2011. For AQMEII phase 2 the new TNO-MACC-II emissions data set was used, an emissions data time series for the years 2003-2009 (Kuenen et al, 2014). To get consistent time series within the new TNO-MACC-II dataset, the years 2003 to 2007 were re-estimated and re-processed for AQMEII phase 2. The new, re-estimated European emissions for 2006 are lower than in the previous AQMEII phase I dataset (Table 3). The changes differ by country (not shown) and by country group and by pollutant. Overall NO_x and PM10 emissions were quite similar but CO, NH_3 and SO_2 emissions were substantially lower in the more recent data set (Table 3). There are several reasons for these differences. For the EU15 including Norway and Switzerland as well as the group of EU new member states (EU-NMS), the differences are due to recent revisions of the reported historic emissions data by countries. For the other non-EU countries in general no reported emissions were available but emissions were taken from the International Institute for Applied Systems Analysis (IIASA) Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model (IIASA, 2012). For the countries Armenia, Azerbaijan and Georgia data from Emissions Database for Global Atmospheric Research (EDGAR) (JRC, 2011) were used at SNAP (Selected Nomenclature for Air Pollutants) level 1 for all pollutants and all years. See Table S2 for a mapping of SNAP codes to NFR codes. The revision of these bottom-up emissions data in recent years caused substantial changes. In particular, the revision of NH_3 and SO_2 emissions in the IIASA GAINS model had an impact on

255 the year 2006 emissions data (Table 3). The large discrepancy for CO is a special case. In the
256 AQMEII phase 1 dataset no CO data were available for the non-EU countries to make a time
257 series. Therefore a TNO estimate for the year 2000 from Visschedijk and Denier van der Gon
258 (2005) was used and kept constant over time. In the new TNO-MACC-II data set used in
259 AQMEII phase 2 this estimate was replaced by a new TNO bottom-up estimate for the year
260 2006. Although the uncertainty in CO emission estimation is high, simply because it receives
261 little attention, the new estimate is considered better than continuing the use of the previous year
262 2000 estimates because 2006 estimates are closer to the modeling year of 2010. Here, the
263 objective is merely to document changes between the data used in AQMEII phase 1 and
264 AQMEII phase 2.
265
266 **3.1.1. Suitability of 2009 European anthropogenic emission data as approximation for 2010**
267 At the start of AQMEII phase 2 no high resolution gridded data for Europe for the year 2010 was
268 available. Therefore, European anthropogenic emissions for the year 2009 were used as the best
269 approximation for the year 2010. The effects of this assumption can be analyzed through the use
270 of official reported emissions data for the EU27 taken from the Centre on Emission Inventories
271 and Projections (CEIP) (Table 4); estimated total emissions changes between 2009 and 2010
272 were calculated. It can be seen that the decreasing trend in SO_2 emissions continues, CO was
273 estimated to be 6% higher in 2010 compared to 2009, while other pollutants show rather small
274 changes. The data in Table 3 cannot be directly compared to the data in Table 4 since the latter
275 does not include the further corrections for the European gridded data finally used (see also
276 Kuenen et al., 2014) – In Table 4 we directly compare reported 2009 and 2010 emissions data
277 only to derive the trend. This trend however, is indicative for the trend that would apply to

corrected gridded data as well. Given uncertainties in emission inventories and the importance of actual meteorology we conclude that differences between 2009 and 2010 emissions will not critically influence the modeling results. Table 4 also provides information on the extent to which potential biases observed in the models may be due to the inventory year. For example, SO_2 emissions in 2009 were slightly higher than 2010. Hence if the models (slightly) overestimate SO_2 at measurement locations that might be partly related to using 2009 data, but if negative model SO_2 biases were found, these are less likely to be attributable to the emissions year.

3.1 North American Domain

This year 2006 was used in both phase 1 and phase 2 of the AQMEII project for North America. We will highlight the most significant sector-specific differences. Different grouping of a few sectors between the two phases of the project requires that some sectors be combined for consistent analysis. Table 5 shows the 2006 emission totals by pollutant and sector for phase 1 and Table 6 shows the 2006 emission totals by pollutant for phase 2. Significant percent changes (> 5%) are highlighted in Table **7**. Differences that are greater than 5% will be discussed in this section and divided into 3 groups. Changes that are most attributable to methodological differences are highlighted in red in Table **7**. Changes that primarily resulting from changes in growth and/or controls between base inventory years of 2005 and 2008 are colored in green. If a combination of emissions estimation method changes, growth, and controls is the reason for the difference, the percent differences are highlighted in blue. One important difference between phase 1 and phase 2 arose from the estimation of on-road mobile source emissions. In phase 1, EPA's MOBILE6 model was used to estimate on-road emissions

whereas in phase 2, EPA's MOVES model was used to estimate on-road emissions. This method change has a substantial impact on on-network mobile source emissions. NO_x on-road emissions are increased by 35% and $PM_{2.5}$ on-road emissions increased by a factor of 1.54. Revised NO_x emission factors in MOVES account for the increase in NO_x whereas a newly incorporated temperature dependence of $PM_{2.5}$ emissions in MOVES resulted in the change in $PM_{2.5}$. In MOVES, mobile source $PM_{2.5}$ emissions are highest at low temperatures whereas in MOBILE6, there is no temperature dependence. More information on the differences between MOVES and MOBILE6 can be found in (CRC, 2010). More information on the temperature adjustments in MOVES can be found in (EPA, 2010). The introduction of a meteorological adjustment to fugitive dust emissions accounts for the reduction in PM for this sector between the two project phases. As noted above, this meteorological adjustment was not applied to all North American models using the inventory – while its use resulted in greater magnitude PM_{10} negative biases compared to observations, the PM_{10} spatial correlations were improved (Im et al., 2014). The "c3marine" or 'seca_c3" sector was simply grown using the assumption of 3% growth per year from 2005 to 2008 and this accounts for the 9% increase for all the pollutants in this sector. The "ptipm" sector which consists of the Electricity Generation Units (EGU) reflects changes in growth and controls between the two base years. Continuous Emission Monitors (CEM) data for NO_x and SO_2 were used for both phases of the project; emissions differences associated with this sector are due to changes in the non-CEM sources and non-CEM pollutants. For the remaining US sectors shown in blue ("nonpt", "ptnonipm", "c1c2rail","rwc"), we note that method changes in the inventory development process account for decreases in emission estimates which are offset by growth assumptions for these sectors.

Together, these changes resulted in some moderate differences in some species as shown in the last line of Table 7. US total NH_3 increased by 1%, and NO_x increased by 4%, while SO_2 estimates decreased by 12%, $PM_{2.5}$ decreased by 8%, PM_{10} decreased by 18%,CO decreased by 10%, and NMHC decreased by 5%.

The changes to the Canadian and Mexican emission inventories between phase 1 and phase 2 are described in Sect. 2.2.2. These changes explain the net differences seen in rows 5 to 7 of Table 7. In addition, subsequent to AQMEII phase 1, Environment Canada reviewed and implemented a number of updates to the spatial surrogates, temporal profiles, and related source-sector cross-references that are used during emissions processing for spatial and temporal allocation. These updates included an expansion in the number of spatial surrogates used to allocate on-road emissions from one to six and a large increase in the number of spatial surrogates used to allocate agricultural ammonia emissions (e.g., Makar et al., 2009; Zhang et al., 2012). While they did not affect inventory totals, these additional updates changed the spatial and temporal allocation of Canadian emissions.

3.1.1 U.S. Residential Wood Combustion

As an example, we highlight one U.S. emission sector that was revised significantly between phase 1 and phase 2 of the AQMEII project, the residential wood combustion (RWC) sector. The changes made to this sector are expected to have a significant impact on winter time model performance for primary $PM_{2.5}$ because of changes the spatial and temporal distribution of the RWC emissions. Residential wood combustion represents the use of wood in a fireplace, woodstove, or hydronic heater as a fuel for residential heating. This sector is one of the more

346 difficult sectors to estimate both spatially and temporally. As indicated in Pouliot et al. (2012),
347 the base emission inventory used in phase 1 was the 2005 NEIv3 and the 2005 modeling
348 platform was used for temporal and spatial allocation. In the 2005 modeling platform, emissions
349 from residential wood combustion were annual county based estimates based on the
350 methodology carried over from the 2002 NEI (EPA, 2006, Appendix Page A-142). This
351 approach is a top-down approach based on national surveys with no adjustments for high-density
352 urban areas. Temporal allocation was based on monthly, state level factors with a default
353 diurnal profile shown in Figure 1(dashed line) In the 2008 modeling platform used here, the
354 emissions were again estimated at the county level on an annual basis (EPA, 2013). However,
355 data supplied from the Midwest and Southeast planning organizations replaced the EPA-derived
356 data because it was expected to be more accurate. A new daily temporal allocation method was
357 introduced to account for the temperature dependence (Adelman et al, 2010). A new diurnal
358 profile shown in Figure 1 (solid line) was also implemented based on a survey from the state of
359 Delaware (Roe and Lindquist, 2004). This new profile allocated the majority of the emissions in
360 the evening hours and was based on residential wood survey data rather than the old default
361 diurnal profile which has no relationship to activity profiles for this sector. Where EPA data was
362 used, there was again no adjustment for high density urban areas. The impact of the changes in
363 this sector can be summarized as follows: substantial increase in nighttime emissions in high
364 density urban areas and regional changes in emissions due to variation in data sources at the
365 regional level. Figure 2 (top) shows the spatial distribution of RWC emissions from phase 1 on
366 the top. Figure 2 (bottom) also shows the spatial distribution of the RWC emission from phase 2.
367 Figure 3 shows the absolute difference in $PM_{2.5}$ emissions between phase 1 and phase 2. We note
368 that RWC emissions increased in the Midwest and decreased in the southeast in phase 2

compared to phase 1. Increases and decreases were noted in the northeast states. These changes reflect the use of multiple data sources within the sector. The higher emission in urban areas coupled with the change in the diurnal profile would be expected to increase $PM_{2.5}$ concentrations substantially during meteorological conditions conducive to $PM_{2.5}$ stagnation.

4. Anthropogenic Emission Differences between the two years

4.1 European Domain

For the European domain the year 2009 was used as approximation for 2010, as explained in the previous section. We will therefore compare changes in emissions going from 2006 to 2009, as this is what was used as input for the models in AQMEII phase 2. This does not apply to the biogenic emissions which are discussed separately in section 5. To keep an overview, emissions are presented by country group (Table 8). The rationale behind the country groups is that they roughly combine countries with similar policy implementation levels. The emission levels for individual countries in both years can be found in the supplementary material of Kuenen et al. (2014).

In the EU-15+NOR+CHE, considerable emission reductions are achieved for all pollutants going from 2006 to 2009 (Table 3), with the remarkable exception of NH_3. This illustrates that the dominating source of NH_3 emissions (agriculture, SNAP10) is not effectively addressed with controls. PM10 emission reductions of 10% are encouraging given known human health effects of PM10 and PM10 reduction goals, provided that this trend can be continued. NO_x and CO emissions were reduced at a faster rate. Similar trends are observed for the EU-NMS, probably related to the implementation of similar EU policies. The non-EU countries behave different with

392 little emission reductions achieved except for NO_x and NMVOC (-5%). However, it should be
393 noted that our knowledge base for these countries is poor. Reliable data for these countries are
394 difficult to obtain, and there is little interest in supporting and funding research to improve the
395 data quality substantially. So, it is fair to say that the trends in the non-EU countries are highly
396 uncertain. Shipping emission totals by sea were taken directly from the EMEP Centre on
397 Emission Inventories and Projections (CEIP, 2012;http://www.ceip.at/). The emission estimates
398 from international shipping by EMEP increased with the exception of PM and SO_2 due to the
399 SECA (Sulfur Emission Control Areas) policies being implemented for the North Sea and the
400 Baltic Sea. The shipping emission methodology underlying the CEIP emission estimates is
401 transparent but rather simple consisting of scaling emissions from a base year to following years
402 based on growth estimates. This may lead to an overestimation of the emissions because
403 autonomous development like increasing ship size and/or slow steaming may not be fully
404 accounted for. In general these developments will lead to fuel savings and thereby emission
405 reductions. The emission data by sea were distributed using the shipping tracks as available from
406 Wang et al. (2008). An example of the spatial distribution of 2009 emissions is given for SO_2
407 emissions in Figure 4. Total anthropogenic European domain emissions declined with about 10%
408 from 2006 to 2009, with the exception of NH_3 (Table 8). Spatial maps for both the absolute
409 difference and percent change in NO_x emissions are shown in Figure 5. It can be seen that the
410 emission from road transport decreased. The road networks are clearly recognizable as lines
411 where the emissions were reduced, especially in the EU15 but NOx emissions from shipping
412 increased. The relative change is sometimes misleading as some small diffuse sources that may
413 have changed will note effect the absolute emissions much but will show up as a relative large

414 change. Therefore it is recommended to look at both the absolute concentrations difference as
415 well as the relative change.
416 The most relevant trend is the ongoing reduction in SO_2 emissions by almost 20% (Table 8); a
417 more detailed analysis of the changes in SO_2 emissions is provided in Figure 6 and Figure 7. The
418 emission reduction is achieved mainly in the EU-27 and more specifically in the energy
419 production sector (SNAP01: Combustion in energy and transformation industries). Furthermore
420 the implementation of the SECA causes SO2 emission reduction on the North Sea and the Baltic
421 Sea (Figure 6). Similar graphs of the spatial component of the emission changes for PM2.5 are
422 presented in Figure 8, showing the decrease in the North Sea and Baltic Sea due to
423 implementation of the SECA. However, PM2.5 emission decreases between 2006 and 2009 over
424 land are small in the EU15 (decreasing road transport emissions) and the EU-NMS (decreasing
425 emission in industrial combustion) or virtually zero (Non-EU countries).
426
427 **4.2 North American Domain: US emissions changes between 2006 and 2010**
428 In this section, we will examine the differences in emission between the two study years of 2006
429 and 2010. Canadian and Mexican emissions did not change between the years for the inventory
430 constructed for AQMEII-2, hence these findings relate only to the US portion of the North
431 America domain. As stated previously, year-specific updates were made for the following
432 sectors: EGU point sources (NO_x and SO_2 only), wildfires and prescribed burns, on-road mobile
433 sources, residential wood combustion, biogenic sources, and area source fugitive dust. All other
434 sectors (nonpoint sources, off-road (or nonroad) sources, non-EGU point sources, agricultural
435 ammonia sources, agriculture fires, commercial marine vessels classes 1 and 2, commercial
436 marine vessels class 3 were constant for the two study years. The reason that these sectors were

437 not changed is two-fold: 1) We do not have reliable year specific information for these sectors 2)
438 We assume that year to year changes in these sectors are small compared to the other sectors
439 which we have year specific information. However, because we have held these sectors
440 constant, the "signal" in the modeling results between the two years may be reduced because of
441 this missing information. We provide emission estimates here in terms of the more detailed
442 sectors used in phase 2 of this project. Table 9 shows the 2006 emission estimates used in phase
443 2 of this project. Table 10 shows the 2010 emission estimates. Table 11 shows the percent
444 change in emissions from 2006 to 2010 for those sectors/pollutants for which there was a change
445 of more than a half percent.
446
447 Spatial maps for both the absolute difference and percent change in NO_x emissions are shown in
448 Figure 9. We see an overall decrease of 10% to 30% for many regions with a total net domain-
449 wide decrease of 17% (Table 11). We note that the changes in emissions in Table 11 are due to
450 (1) implementation of additional emission controls between 2006 and 2010 (EGU, mobile,
451 nonroad sources) and (2) natural variability (dust and fire sources). There are a few isolated
452 areas with increases. We would expect regional decreases in ozone throughout the Contiguous
453 United States as a result of this decrease in NO_x. In Canada and Mexico, on the other hand,
454 since the same base inventories were used for both 2006 and 2010 (see Sect. 2.2.2), only
455 differences due to numerical noise can be seen. Spatial maps showing both the absolute
456 difference and percent change in SO_2 are shown in Figure 10. We see a larger decrease on the
457 order of 40 to 80% for various regions and a domain-wide total decrease of 29%. Spatial maps
458 showing both the absolute difference and percent change in $PM_{2.5}$ are shown in Figure 11.
459 Differences can be seen in Canada due to the downward adjustment to fugitive dust emissions

from soil moisture and snow cover (Appel et al. 2013). In addition, US differences in $PM_{2.5}$ are also due to the natural variability in dust and fire sources as well as changes in emission controls from on/off road sources and EGU point sources. We see a mix of increases and decreases and in this case, a well-defined "signal" change in emission between the two years may be difficult to interpret. Emission controls of NO_x were changed between 2006 and 2010 for the EGU sector. To show the difference for the annual cycle, we show the annual domain-wide daily time series of 2006 compared to 2010 for NO_x (Figure 12) and SO_2 (Figure 13). Note that NO_x changes are most prominent during the winter months because summer controls were extended for the whole year between 2006 and 2010. Thus we would expect a smaller decrease in summer ozone since most of the NO_x reductions occur outside the ozone season. For SO_2, the reductions occurred for all months, so we expect model differences to be similar during all seasons. Stoekenius et al. 2014 contains a summary of the regional and seasonal differences in the anthropogenic emissions between 2010 and 2006 in the NA domain.

5. Biogenic Emission Differences between 2006 and 2010

In this section we will examine the differences in the biogenic emission estimates that were used in some (but not all) of the models for both the North American and European domains.

5.1 European Domain

Biogenic VOC emissions (BVOC), mainly in the form of isoprene, can have a substantial impact on ozone production and ozone concentration variability (e.g., Curci et al., 2009). The rates of isoprene emission are largely dependent on plant species, and increase strongly with temperature and with photosynthetic active radiation intensity (Guenther et al., 1993; Guenther, 1995). BVOC emissions are often calculated on-line in chemistry and transport models (CTM) using

meteorological fields for the climate variables in combination with detailed information on spatial distribution of tree types in Europe. Here we present the calculated BVOC emissions for the years 2006 and 2010 using the LOTOS-EUROS model (Schaap et al., 2008; 2009), which is a three-dimensional CTM that simulates air pollution in the lower troposphere. These emissions were used for model "M10", Group "NL2" for the EU domain as shown in Table 1 of Ulas et.al. (2014). The implementation of biogenic VOC emissions in LOTOS-EUROS is similar to the approach by Steinbrecher et al. (2009) and further documented in Schaap et al., 2009 and Beltman et al., 2013.The land use dataset (Steinbrecher et al., 2009) was combined with the distributions of 115 tree species over Europe (Koeble and Seufert, 2001). The role of the local temperature and photo-synthetically active radiation are taken into account in the biogenic emissions by following the empirically designed algorithms proposed by Guenther et al. (1993) and Tingey et al. (1980). The projection used in the BVOC emission calculations is longitude–latitude at a grid resolution of 0.50° longitude × 0.25° latitude (in Europe approximately 25 × 25 km). The simulated domain is a rectangular grid from 15°W to 35°E and 35°N to 70°N, which roughly encompasses Europe but some models may use a larger extent. Various CTMs may have slightly different solutions or parameterizations but in general the emission source strength and the emission timing will be comparable with what was calculated by LOTOS-EUROS. In AQMEII phase 2 these calculated emissions were used by at least one other CTM.

BVOC emissions vary considerably between years (Figure 14). Over the total European domain isoprene emissions were about 20% higher in 2010 compared to 2006 and the releases were slightly later in the year (Figure 14). Since the emissions are driven by climate variables, the timing throughout the year is important. About 90% of the isoprene emissions occur in the period

May-September (Figure 14; Figure 15). Terpene emissions also occur in winter months but still about 75% of the emissions occur in May-September (Figure 14). Since the absolute emissions calculated by the various CTMs participating in AQMEII may vary, the fractional release per month gives more insight in the important patterns (Figure 15). Figure 15 illustrates that from year to year the fraction of total emission in a particular month can vary substantially due to climatic variability. Also individual locations (in this example East France and Southern Italy) may deviate from the average European pattern. For example, the emissions in East of France clearly peak in July while emissions in southern Italy are equally important in June-July-August. Moreover, while the European isoprene emission in 2010 were about 20% higher than in 2006 (Figure 15) for the 2 individual examples used here, the situation was exactly the opposite with about 20% higher emissions in 2006 than in 2010 (Figure 16). The variability can be explained by local to regional climate variability (between years and locations) over Europe.

5.2 North American Domain

The biogenic emission estimates used for the North American domain comparison in this work were based on the Biogenic Emission Inventory System (BEIS) version 3.14. Since the coupled WRF-CMAQ system was used to generate the emissions, these emission estimates were based on the meteorological fields from the coupled simulations. Domain-wide biogenic emission estimates have already been included Table 9 for 2006 and Table 10 for 2010. Although the total biogenic VOC emissions in 2006 were 2% higher than in 2010, there were important spatial differences in the biogenic emission estimates. Figure 17 shows the summer of 2006 and the summer 2010 state-wide temperature rankings. We see that in 2006, the entire country was warmer than average but more so in the west than in the east. However in 2010, in the east and

529 southeast the temperatures were at exceptionally warm but below normal in the west. As a result,
530 in Figure 18, when we subtract 2006 biogenic emission estimates from 2010 we get large
531 positive differences in the east and southeast and negative differences in the west. The summer
532 of 2010 was characterized by exceptional warmth in the east/southwest and generally cooler
533 temperatures in the west whereas the entire Contiguous United States was warm in 2006. This
534 implies more biogenic VOCs in the east for 2010 (compared to 2006) and more biogenic VOCs
535 in the west in 2006 (compared to 2010).
536
537 **6. Conclusions and Recommendations**
538 In this paper, we summarize the preparation of the emissions for both the EU and NA domains.
539 We have provided a summary of the differences in the emission estimates between phase 1 and
540 phase 2 of the AQMEII project. For the EU domain, there was substantial decrease in CO (-
541 19%), NH_3 (-11%) , and SO_2(-12%) emissions between phase 2 and phase 1 emissions for 2006.
542 For the NA domain, there were decreases in CO (-10%), non-methane hydrocarbons (-5%),
543 $PM_{2.5}$ (-8%), PM10 (-18%), SO_2 (-12%), with an increase of 4% in NOx. We have provided the
544 example of residential wood combustion emissions as an emission change between the two
545 phases that would impact model performance of $PM_{2.5}$ during the nighttime winter in urban
546 areas. We have also provided a summary of the emission changes between 2010 (NA
547 domain)/2009 (EU domain) and 2006 for the two modeling years that were used in phase 2 of the
548 AQMEII project. Between 2006 and 2009, considerable emission reductions were achieved for
549 17 EU countries, Norway and Switzerland as well as EU-Non-Member States, for all emissions
550 classes aside from NH_3, which remained stable. Non-EU countries showed little change in
551 emissions levels, though this may be a result of poor data quality. Shipping emissions decreased

552 for PM and SO_2, while increasing for other species. Between 2006 and 2010, estimated US NOx
553 emissions decreased by 17%, SO_2 by 29%, CO by 21%, $PM_{2.5}$ by 12%, PM10 by 7%, NMHC by
554 4% and NH_3 by 2%. We have provided a useful reference for all the modeling studies and
555 analysis associated with the AQMEII project. Finally, we have provided a brief summary of the
556 differences between the two years in two of the biogenic models used for the EU and NA
557 domains. Between 2006 and 2010, biogenic emissions in North America increased in the east
558 and decreased in the west, due to regional temperature differences between the years. The
559 European emissions of isoprene peaked earlier in 2006, but achieved higher levels in 2010.
560
561
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563
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References

Adelman, Z., G. Arora, A. Xiu, B. Baek, M. Houyoux, A. Eyth, and M. Strum, 2010. Development and preliminary results for a model of temporal variability in residential wood combustion emissions, paper presented at 19th Annual International Emission Inventory Conference" Emissions Inventories-Informing Emerging Issues" San Antonio, Texas.

Appel, K. W., G. A. Pouliot, H. Simon, G. Sarwar, H. O. T. Pye, S. L. Napelenok, F. Akhtar, and S. J. Roselle (2013), Evaluation of dust and trace metal estimates from the Community Multiscale Air Quality (CMAQ) model version 5.0, Geosci. Model Dev., 6(4), 883-899, doi:10.5194/gmd-6-883-2013.

Beltman, J.B., Hendriks, Lillyman, C., Buset, K., 2009.Tum, M., Schaap, M., 2013. The *2008 Canadian*impact of large scale biomass production on ozone air pollution in Europe, Atmospheric Environment, 71.

CEIP, 2012 Centre for Emission Inventories and Projections: Reported emissions by Parties under the Convention for Long-Range Transboundary Air Pollution, http://www.ceip.at/webdab-emission-database/officially-reported-emission-data/, date of download 12 March 2012, 2012.

CRC, 2010. Review of the 2009 Draft Motor Vehicle Emissions Simulator (MOVES) Model, Final*Assessment of Agricultural Ammonia*. National Agri-Environmental Standards Technical Series Report No. 4-1, Environment Canada, Gatineau, Quebec, Canada, 295 pp.

Curci, G. M. Beekmann, R. Vautard, G. Smiatek, R. Steinbrecher, J. Theloke, R. Friedrich, Modelling study of the impact of isoprene and terpene biogenic emissions on European ozone levels, Atmospheric Environment, 43 (2009), pp. 1444–1455

Environment Canada (EC), June 2008, 2006 Criteria Air Contaminants Emissions Inventory - Air Quality Modeling data, Ottawa, Ontario. Unpublished Internal Report. [see http://www.ec.gc.ca/inrp-npri/]

EPA, 2006. Documentation for the final 2002 nonpoint sector national emission inventory for criteria and hazardous air pollutants.

EPA, 2010. MOVES Highway Vehicle, Temperature, Air Conditioning, and Inspection and Maintenance Adjustments." US Environmental Protection Agency, EPA-420-R-10-027.

Giglio, L., van der Werf, G. R., Randerson, J. T., Collatz, G. J., and Kasibhatla, P., 2006. Global estimation of burned area using MODIS active fire observations, Atmos. Chem. Phys., 6, 957–974. .

Guenther, A., 1995. A global model of natural volatile organic compound emissions. Journal of Geophysical Research 100, 8873-8892.

Guenther, A.B., Zimmerman, P.R., Harley, P.C., Monson, R.K., Fall, R., 1993. Isoprene and monoterpene emission rate variability: model evaluations and sensitivity analysis. Journal of Geophysical Research 98D, 12609-12617.

IIASA (International Institute for Applied Systems Analysis), 2012: GAINS detailed emissions by source and activity, http://gains.iiasa.ac.at/gains/EUN/index.login?logout=1, PRIMES baseline scenario 2009, download date 19 June 2012, 2012.

Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baro, R., Bellasio, R., Brunner, D., Chemel, C., Curci, G., Flemming, J., Forkel, R., Giordano, L., Jimenez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L., Jorba, O., Knote, C., Kuenen, J.J.P., Makar, P.A., Manders-Groot, A., Neal, L., Perez, J.L., Pirovano, G., Pouliot, G., San Jose, R., Savage, N., Schroder, W., Sokhi, R.S., Syrakov, D., Torian, A., Tuccella, P., Werhahn, K., Wolke, R., Yahya, K., Zabkar, R., Zhang, Y., Zhang, J., Hogrefe, C., Galmarini, S., 2014a. Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part I: Ozone. Atmospheric Environment (this issue).

JRC (Joint Research Centre), Emission Database for Global Atmospheric Research (EDGAR) v4.2, http://edgar.jrc.ec.europa.eu (last access 24 May 2012), 2011.

Koeble, R., Seufert, G., 2001. Novel maps for forest tree species in Europe. In: Proceedings of the Conference “A Changing Atmosphere”.

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.

Kuenen, J. J. P., A. J. H. Visschedijk, M. Jozwicka, and H. A. C. Denier van der Gon (2014), TNO-MACC_II emission inventory: a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling, Atmos. . Discuss., 14(5), 5837-5869, doi:10.5194/acpd-14-5837-2014.

Makar, P.A., Moran, M.D., Zheng, Q., Cousineau, S., Sassi, M., Duhamel, A., Besner, M., Davignon, D., Crevier, L.-P., and Bouchet, V.S., 2009. Modelling the impacts of ammonia emissions reductions on North American air quality, Atmos. Chem. Phys.., 9, 7183-7212.

Mason, R., A. Zubrow, and A. Eyth, 2012. Technical Support Document (TSD) Preparation of Emissions Inventories for the Version 5.0, 2007 Emissions Modeling Platform. Available at http://epa.gov/ttn/chief/emch/2007v5/2007v5_2020base_EmisMod_TSD_13dec2012.pdf

Moran, M. D., Zheng, Q., Zhang, J., Pavlovic, R., Ménard, S., and Sassi, M., 2012. Recent improvements to the representation of anthropogenic emissions in the Canadian operational air quality forecast model (GEM-MACH), *4th International Workshop on Air Quality Forecasting Research*, 12-14 Dec., Geneva, Switzerland.

Pouliot, G., T. Pierce, H. Denier van der Gon, M. Schaap, M. Moran, and U. Nopmongcol, 2012 Comparing emission inventories and model-ready emission datasets between Europe and North America for the AQMEII project, Atmospheric Environment, 53(0), 4-14, doi:10.1016/j.atmosenv.2011.12.041.

Pulles, T. H. Kok, U. Quass, Application of the emission inventory model TEAM: Uncertainties in dioxin emission estimates for central Europe, Atmospheric Environment, Volume 40, Issue 13, April 2006, Pages 2321-2332

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.

Roe, Stephen and Holly C. Lindquist, 2004. MANE-VU Residential Wood Combustion Emission Inventory, E.H. Pechan & Associates, Inc, June 22, 2004.

Sassi, M., Chen, J., Samaali, M., Davignon, D., Moran, M. D., Taylor, B., Zheng, Q., 2010. 2006 Canadian emissions for air quality modelling. *19th International Emission Inventory Conference*, 27-30 Sept., San Antonio, Texas.
[http://www.epa.gov/ttn/chief/conference/ei19/session2/sassi.pdf]

Soares, J., Sofiev, M., Prank, M, San Jose, R., Perez, J.L. On uncertainties of wild-land fires emission in AQMEII case study, Atmospheric Environment, (this issue).

Schaap, M., Sauter, F., Timmermans, R.M.A., Roemer, M., Velders, G., Beck, J., Builtjes, P.J.H., 2008. The LOTOS-EUROS model: description, validation and latest developments, International Journal of Environment and Pollution 32 (2), 270–290

Schaap, M. A.A.M. Manders, E.C.J. Hendriks, J.M. Cnossen, A.J.S. Segers, H.A.C. Denier van der Gon, M. Jozwicka, F.J. Sauter, G.J.M. Velders, J. Matthijsen, P.J.H. Builtjes, Regional Modelling of Particulate Matter for the Netherlands, (2009) PBL Report 500099008, Bilthoven, The Netherlands. Available at: http://www.rivm.nl/bibliotheek/rapporten/500099008.pdf

Schwede, D., G. Pouliot, and T. Pierce (2005), Changes to the biogenic emissions inventory system version 3 (BEIS3), in 4th Annual CMAS Models-3 Users' Conference.

Sofiev, M., Vankevich, R., Lotjonen, M., Prank, M., Petukhov, V., Ermakova, T., Koskinen, J., and Kukkonen, J., 2009. An operational system for the assimilation of the satellite information on wild-land fires for the needs of air quality modelling and forecasting, Atmos. Chem. Phys., 9, 6833-6847

Steinbrecher, R., G. Smiatek, R. Köble, G. Seufert, J. Theloke, K. Hauff, P. Ciccioli, R. Vautard, G. Curci, Intra- and inter-annual variability of VOC emissions from natural and semi-natural vegetation in Europe and neighbouring countries, Atmospheric Environment, 43 (2009), pp. 1380–1391

Stoeckenius, T., J Zagunis, T Sturtz and T Sakulyanontvittaya, 2014. A Comparison between 2010 and 2006 Air Quality and Meteorlogical Conditions, and Emissions and Boundary Conditions used in Simulations of the AQMEII-2 North American Domain, Atmospheric Environment, (this issue).

Tingey, D.T., Manning, M., Grothaus, L.C., Burns, W.F., 1980. Influence of light and temperature on monoterpene emissions rates from slash pine. Plant Physiology 65, 797-801

Visschedijk, A.H.J. and H.A.C. Denier van der Gon, 2005. Gridded European anthropogenic emission data for NO_x, SO_2, NMVOC, NH3, CO, PM10, PM2.5 and CH4 for the year 2000, TNO B&O-A Rapport 2005/106, 2nd version November 2005.

Wang, C., J. J. Corbett, and J. Firestone, 2008. Improving spatial representation of global ship emissions inventories. Environ. Sci. Technol., 42:193 – 199.

Wolf, M.E., Paula G. Fields, P.G., Manne, G.K., López Villegas, M.T., Garibay Bravo, V., Iniestra Gómez, R., 2009. Developing Mexico national emissions inventory projections for the future years of 2008, 2012, and 2030. *18th International Emission Inventory Conference*, April 14-17, Baltimore, Maryland, 29 pp. [www.epa.gov/ttnchie1/conference/ei18/session2/wolf.pdf]

Zhang, J., Zheng, Q., Moran, M. D., Gordon, M., Liggio, J., Makar, P., Taylor, B., Stroud, C., 2012. Improvements to SMOKE processing of Canadian on-road mobile emissions. *20th International Emission Inventory Conference*, 13-16 Aug., Tampa, Florida, 13 pp. http://www.epa.gov/ttn/chief/conference/ei20/session1/jzhang.pdf

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748 Table 1. Source categories for the Contiguous United States emissions

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
"c1c2rail"	Area source emissions from locomotive and marine (class 1 & class 2 vessels only) This sector was named "alm_no_c3" in phase 1. Aircraft emissions are included in "ptnonipm".
"agfire"	Crop residue burning. This sector was part of the "nonpt" sector in phase 1
Beis	Biogenic VOC and soil NO emissions estimated with BEIS3.14 and meteorology from the WRF/CMAQ coupled system.
C1c2marine	Class 1 and class 2 commercial marine port and inter-port vessels plus railroads
C3marine	Commercial marine port and inter-port Class 3 (C3) vessels defined as having displacement greater than 30 liters per cylinder. This sector was called "seca_c3" in phase 1
Nonpt	Area source emissions not included in other sectors
Nonroad	Off-road mobile source emissions from EPA's NONROAD model
Onroad	On-road mobile source emissions from the SMOKE/MOVES system
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) and including aircraft emissions (takeoff, landing, support equipment)
"ptfire"	Wildland fires and prescribed burning fire emissions identified as point sources
"rwc"	Residential Wood Combustion. This sector was part of the "nonpt" sector in phase 1.

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751 Table 2. Source categories for Canadian/Mexican emissions

Source type	Description
“canafdust”	Area source fugitive dust from anthropogenic sources, PM10 & $PM_{2.5}$ only This sector was part of “othar” in phase 1
“canag”	Area source emissions from agricultural operations, NH_3 only. This sector was part of “othar” in phase 1
“canar”	Area source emissions from Canada not included in “canafdust” or “canag”. These were part of “othar” in phase 1
“canon”	On-road mobile source emissions from Canada. These were part of “othon” in phase 1
“canpt”	Point Source emissions from Canada. These were part of “othpt” in phase 1
“mexar”	Area source emissions from Mexico. These were part of “othar” in phase 1
“mexon”	On-road mobile Source Emissions from Mexico. These were part of “othon” in phase 1
“mexpt”	Point Source emissions from Mexico. These were part of “othpt” in phase 1

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Table 3 European emissions excluding International shipping (Gg/yr) for the year 2006 in AQMEII-1 and AQMEII-2.

Region[a]	Data set	CO	NH_3	NMVOC	NO_x	PM_{10}	SO_2
EU15+ Norway + Switzerland	AQMEII-1	23,731	3,315	7,756	9,112	1,690	4,584
	AQMEII-2	22,168	3,170	6,984	9,281	1,666	4,333
	Change	-7%	-4%	-10%	2%	-1%	-5%
EU-NMS[b]	AQMEII-1	8,198	843	2,257	2,040	724	3,688
	AQMEII-2	7,115	872	1,954	2,116	795	3,685
	Change	-13%	3%	-13%	4%	10%	0%
Non EU	AQMEII-1	20,750	2,127	5,088	5,625	2,646	7,103
	AQMEII-2	13,583	1,581	5,452	5,161	2,551	5,556
	Change	-35%	-26%	7%	-8%	-4%	-22%
Europe	AQMEII-1	52,678	6,286	15,101	16,777	5,059	15,376
	AQMEII-2	42,865	5,624	14,389	16,558	5,011	13,575
	Change	-19%	-11%	-5%	-1%	-1%	-12%

[a] For a listing of the countries in the country groups see Supplementary Table S1.
b NMS = New Member States of the European Union

Table 4 Comparison of total EU-27 emissions (Gg) excluding International shipping for 2009 and 2010. Data downloaded from Centre of Emission Inventories and Projections (CEIP http://www.ceip.at; April 2014).

Pollutant	2009	2010	Increase 2010
CO	23,534	24,985	6.2%
NH_3	3,679	3,613	-1.8%
NMVOC	7,077	7,115	0.5%
NO_x (as NO_2)	9,240	9,096	-1.6%
PM_{10}	1,782	1,842	3.4%
SO_x (as SO_2)	4,841	4,559	-5.8%

[a)] EU-27 Member States include: Austria, Belgium, Bulgaria, Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, the Netherlands, Poland, Portugal, Romania, Slovak Republic, Slovenia, Spain, Sweden and the United Kingdom.

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769 Table 5 Annual 2006 Emission Estimates for the North American Study Domain for phase 1

770 (Gg/Year except NMHC which is Gg C/yr)

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	CO	NH_3	NMHC	NO_x	$PM_{2.5}$	PM_{10}	SO_2
"afdust"					935	8,036	
"ag"		2942					
"alm_no_c3"+ "ptnonipm"	3,168	145	926	3,785	453	647	2,061
"seca_c3"	82		27	996	76	82	629
"othar"	3,740	720	1,146	811	403	1,361	164
"othon"	5,063	22	198	588	14	18	10
"othpt"	1,184	19	386	1,059	128	194	2,190
"nonpt"	6,685	122	5,199	1,525	976	1,223	1,134
"nonroad"	18,004	2	2,050	1,935	182	192	181
"onroad"	43,332	266	2,988	5,812	122	173	130
"ptfire"	17,401	286	2,029	257	1,515	1,788	137
"ptipm"	551	26	31	3,418	458	557	9,409
"beis"	8,784		41,846	1,260			
Totals	107,994	4549	56,826	21,446	5,262	14,271	16,045

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Table 6. Annual 2006 Emission Estimates for the North American Study Domain for phase 2 (Gg/Year except NMHC which is Gg C/yr)

Sector	CO	NH_3	NMHC	NO_x	$PM_{2.5}$	PM_{10}	SO_2
"afdust"					823	5,846	
"ag"		3,336					
"c1c2rail"+ "ptnonipm"	2,856	62	746	3,096	412	573	1,486
"c3marine"	90		30	1,072	82	90	678
"canar"+ "mexar"+ "canag"+ "canafdust"	3,844	587	1,186	832	402	1,529	139
"canon"+ "mexon"	4,800	23	293	592	19	24	7
"canpt"+ "mexpt"	1,202	19	742	1,161	144	218	2,270
"nonpt"+ "rwc"+ "agfire"	4,140	143	4,283	1,118	648	731	367
"nonroad"	16,803	2	1,990	1,873	170	180	91
"onroad"	37,440	134	2,557	7,857	309	387	164
"ptfire"	17,402	286	1,941	257	1,515	1,788	137
"ptipm"	635	23	28	3,156	298	394	8,700
"beis"	8,424		40,237	1,251			
Totals	97,636	4,615	54,033	22,265	4,823	11,757	14,040

Table 7 Percent Difference between Phase 2 and Phase 1 (2006) Emissions for North American study domain. See Text for explanation of text colors

Sector	CO	NH_3	NMHC (Gg C/yr)	NO_x	$PM_{2.5}$	PM_{10}	SO_2
"afdust"					-12%	-27%	
"ag"		13%					
"c1c2rail"+ "ptnonipm"	-10%	-57%	-19%	-18%	-9%	-11%	-28%
"c3marine"	9%		9%	8%	9%	9%	8%
"canar"+"mexar"+ "canag"+"canafdust"	3%	-18%	3%	3%	0%	12%	-15%
"canon"+"mexon"	-5%	6%	48%	1%	34%	30%	-36%
"canpt"+"mexpt"+offshore points	1%	0%	92%	10%	13%	12%	4%
"nonpt"+"rwc"+'agfire"	-38%	18%	-18%	-27%	-34%	-40%	-68%
"nonroad"	-7%	0%	-3%	-3%	-7%	-6%	-50%
"onroad"	-14%	-50%	-14%	35%	154%	124%	27%
"ptfire"	0%	0%	-4%	0%	0%	0%	0%
"ptipm"	15%	-12%	-8%	-8%	-35%	-29%	-8%
"beis"	-4%		-4%	-1%			
Net change	-10%	1%	-5%	4%	-8%	-18%	-12%

Table 8 European emissions of CO, NH3, NMVOC, NO_x, PM10 and SO_2 in Gg for the years 2006 and 2009 as used in AQMEII phase 2

Region	Year	CO	NH3	NMVOC	NO_x	$PM_{2.5}$	PM_{10}	SO2
EU15+NOR+CHE	2006	22,168	3,170	6,984	9,281	1,111	1,666	4,333
	2009	17,830	3,094	5,862	7,542	998	1,499	2,559
change 2006 to 2009		-20%	-2%	-16%	-19%	-10%	-10%	-41%
EU-NMS	2006	7,115	872	1,954	2,116	502	795	3,685
	2009	6,609	836	1,767	1,886	450	684	2,553
change 2006 to 2009		-7%	-4%	-10%	-11%	-10%	-14%	-31%
NONEU	2006	13,583	1,581	5,452	5,161	1,782	2,551	5,556
	2009	13,281	1,613	5,168	4,830	1,773	2,537	5,631
change 2006 to 2009		-2%	2%	-5%	-6%	0%	-1%	1%
SEA	2006	405	-	136	3,770	300	317	2,568
	2009	436	-	146	3,990	292	309	2,446
change 2006 to 2009		8%		8%	6%	-3%	-3%	-5%
European	2006	43,271	5,624	14,525	20,328	3,695	5,328	16,143
Domain	2009	38,156	5,543	12,943	18,247	3,513	5,029	13,189
change from 2006		-12%	-1%	-11%	-10%	-5%	-6%	-18%

Table 9 Annual 2006 Emission Estimates for the North American Study Domain for phase 2 (Gg/Year except NMHC which is Gg C/yr)

Sector	CO	NH_3	NMHC	NO_x	$PM_{2.5}$	PM_{10}	SO_2
"afdust"	0	0	0	0	823	5,846	0
"ag"	0	3,336	0	0	0	0	0
"agfire"	346	0	18	16	45	45	3
"c1c2rail"	197	1	36	1,218	39	42	46
"c3marine"	90	0	30	1,072	82	90	678
"canafdust"	0	0	0	0	171	1,231	0
"canag"	0	462	0	0	0	0	0
"canar"	3,411	5	857	652	179	218	88
"canon"	4,202	20	222	507	12	17	2
"canpt"	1,036	19	644	781	62	106	1,598
"mexar"	433	121	329	180	52	80	51
"mexon"	598	3	71	85	7	7	5
"mexpt"	166	0	98	380	83	112	672
"nonpt"	1,810	126	4,074	1,074	319	401	356
"nonroad"	16,803	2	1,990	1,873	170	180	91
"onroad"	37,440	134	2,557	7,857	309	387	164
"ptfire"	17,402	286	1,941	257	1,515	1,788	137
"ptipm"	635	23	28	3,156	298	394	8,700
"ptnonipm"	2,659	62	710	1,879	373	531	1,440
"rwc"	1,984	17	190	28	284	285	8
"beis"	8,424	0	40,237	1,251	0	0	0
Totals	97,636	4,615	54,033	22,265	4,823	11,757	14,040

798 Table 10: Annual 2010 Emission Estimates for the North American Study Domain for phase 2
799 (Gg/Year except NMHC which is Gg C/yr)

Sector	CO	NH_3	NMHC	NO_x	$PM_{2.5}$	PM_{10}	SO_2
"afdust"	0	0	0	0	793	5,633	0
"ag"	0	3,337	0	0	0	0	0
"agfire"	346	0	18	16	45	45	3
"c1c2rail"	197	1	36	1,218	39	42	46
"c3marine"	90	0	30	1,072	82	90	678
"canafdust"	0	0	0	0	182	1,294	0
"canag"	0	461	0	0	0	0	0
"canar"	3,412	5	858	652	179	218	88
"canon"	4,205	20	222	508	12	17	2
"canpt"	1,036	19	644	781	62	106	1,598
"mexar"	433	121	329	181	52	80	51
"mexon"	598	3	71	85	7	7	5
"mexpt"	166	0	98	380	83	112	672
"nonpt"	1,810	126	4,074	1,074	319	401	356
"nonroad"	14,294	2	1,733	1,575	151	159	15
"onroad"	24,950	111	1,914	5,702	229	303	32
"ptfire"	11,946	196	1,333	179	1,042	1,230	95
"ptipm"	636	23	28	1,960	298	394	4,919
"ptnonipm"	2,659	62	710	1,879	373	531	1,440
"rwc"	1,984	17	190	28	284	285	8
"beis"	8,182	0	39,500	1,218	0	0	0
Totals	76,944	4,503	51,788	18,507	4,231	10,945	10,008

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804 Table 11 Percent Difference in Emission Estimates for the North American Study Domain from
805 2010 to 2006. Only those sectors and pollutants with a non-zero difference are shown.

Sector	CO	NH_3	NMHC	NO_x	$PM_{2.5}$	PM_{10}	SO_2
"afdust"					-4%	-4%	
"canafdust"					+6%	+5%	
"nonroad"	-15%	+7%	-13%	-16%	-11%	-12%	-84%
"onroad"	-33%	-17%	-25%	-27%	-26%	-22%	-81%
"ptfire"	-31%	-31%	-31%	-30%	-31%	-31%	-31%
"ptipm"				-38%			-43%
"beis"	-3%		-2%	-3%			
Net Change	-21%	-2%	-4%	-17%	-12%	-7%	-29%

806

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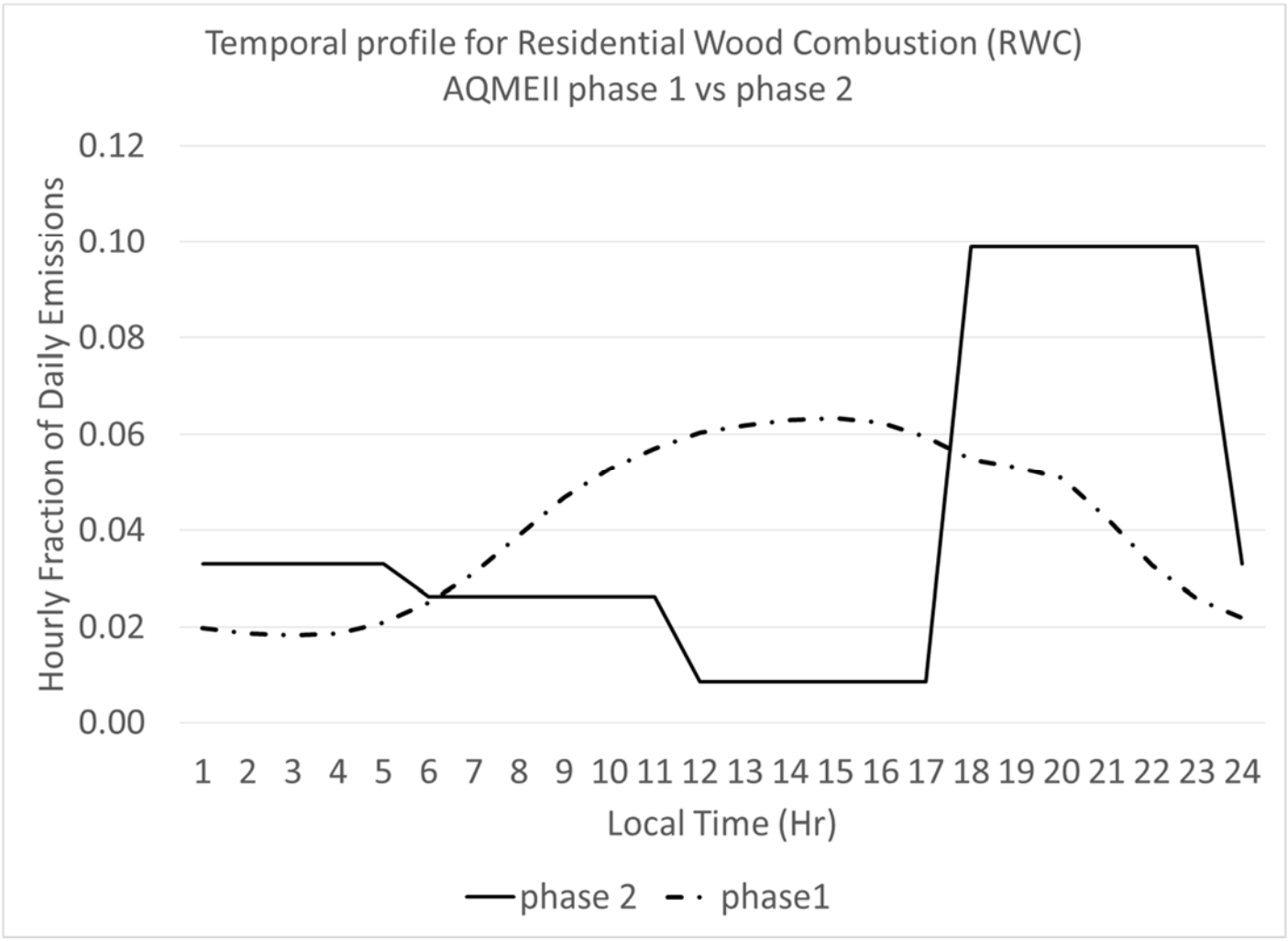
808

List of Figures

831 Figure 11. Spatial Map of absolute emission difference and percent difference in $PM_{2.5}$ between
832 2006 and 2010 for NA domain
833 Figure 12. Annual time series of US daily EGU emissions SO_2 for NO_x 2006 vs. 2010
834 Figure 13. Annual time series of US daily EGU emissions NO_x forSO_2 2006 vs. 2010
835 Figure 14. Annual time series of daily biogenic terpene emissions (left) and isoprene
836 emissions(right) for the European domain
837 Figure 15. The percentage fraction of annual biogenic isoprene emissions emitted per month
838 averaged for the European domain(left) and for two specific locations (right) for the years 2006
839 and 2010.
840 Figure 16. Biogenic isoprene emissions calculated for a site in the East of France (left) and
841 Southern Italy (left) for the years 2006 and 2010.
842 Figure 17. Statewide temperature ranks Jun-Aug. 2006 (left) and Jun-Aug. 2010 (right) for the
843 continental U.S. Contiguous United States from the National Climatic Data Center
844 Figure 18. Spatial Map of Absolute Biogenic VOC Emission Difference (2010-2006)(top) and
845 percent difference from 2006 (bottom) for NA domain
846

847 Figure 1



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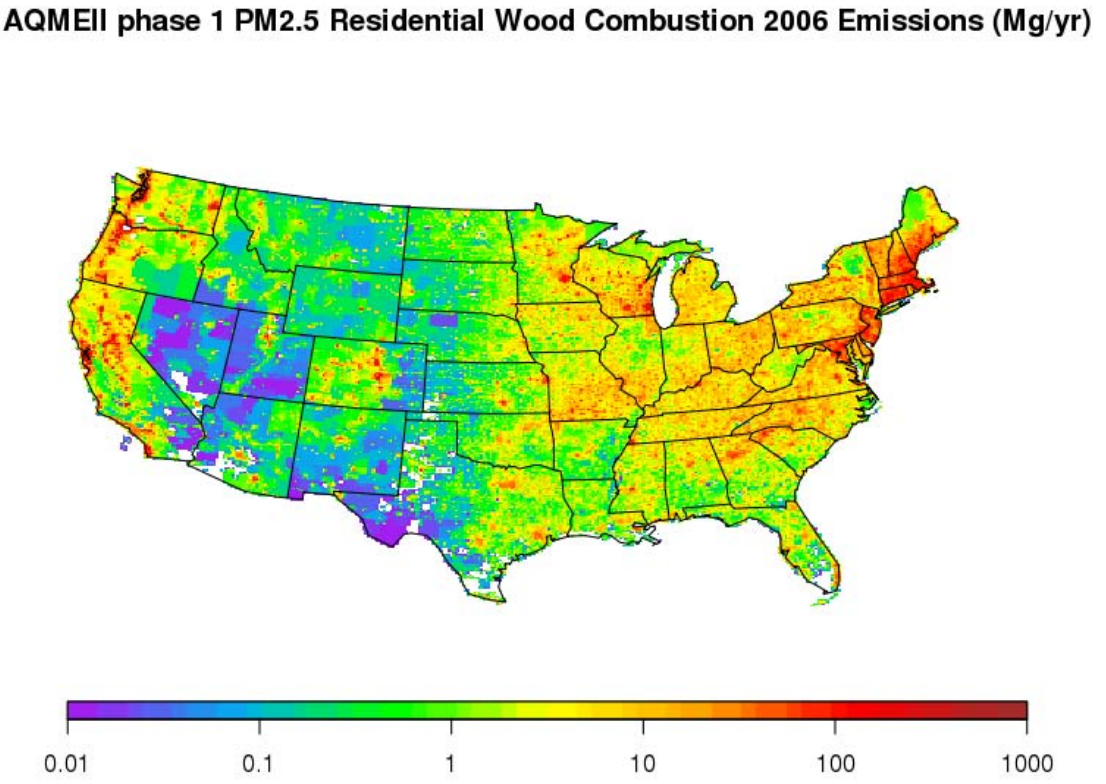
853 Figure 2



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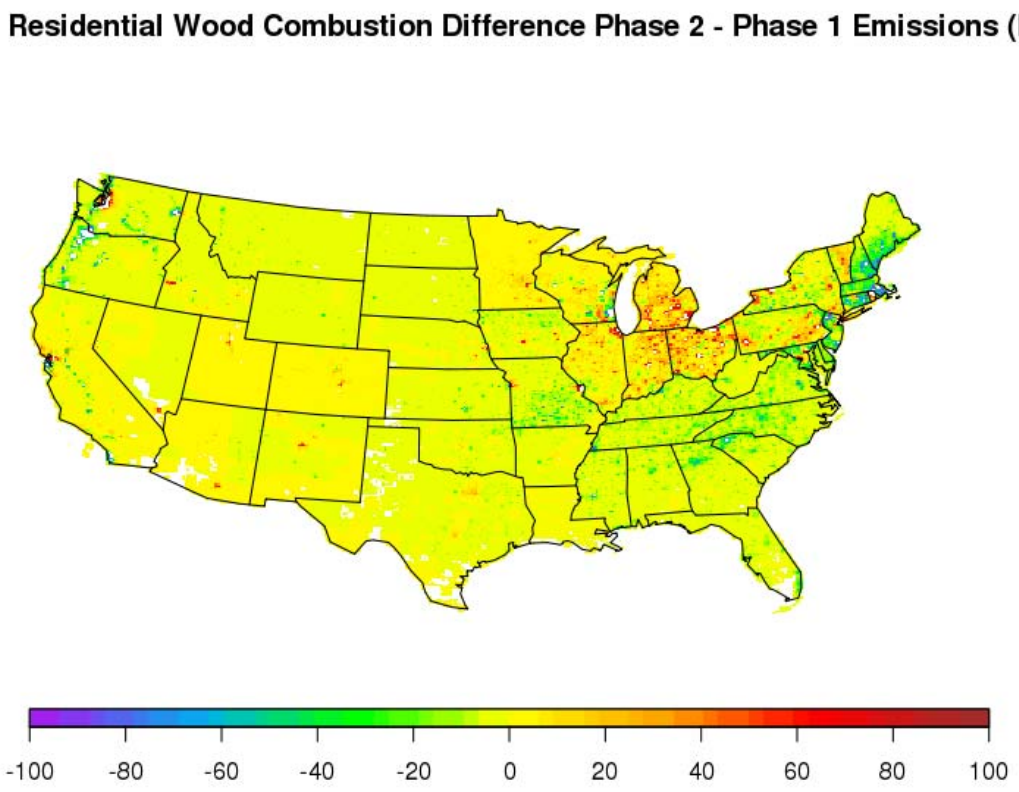

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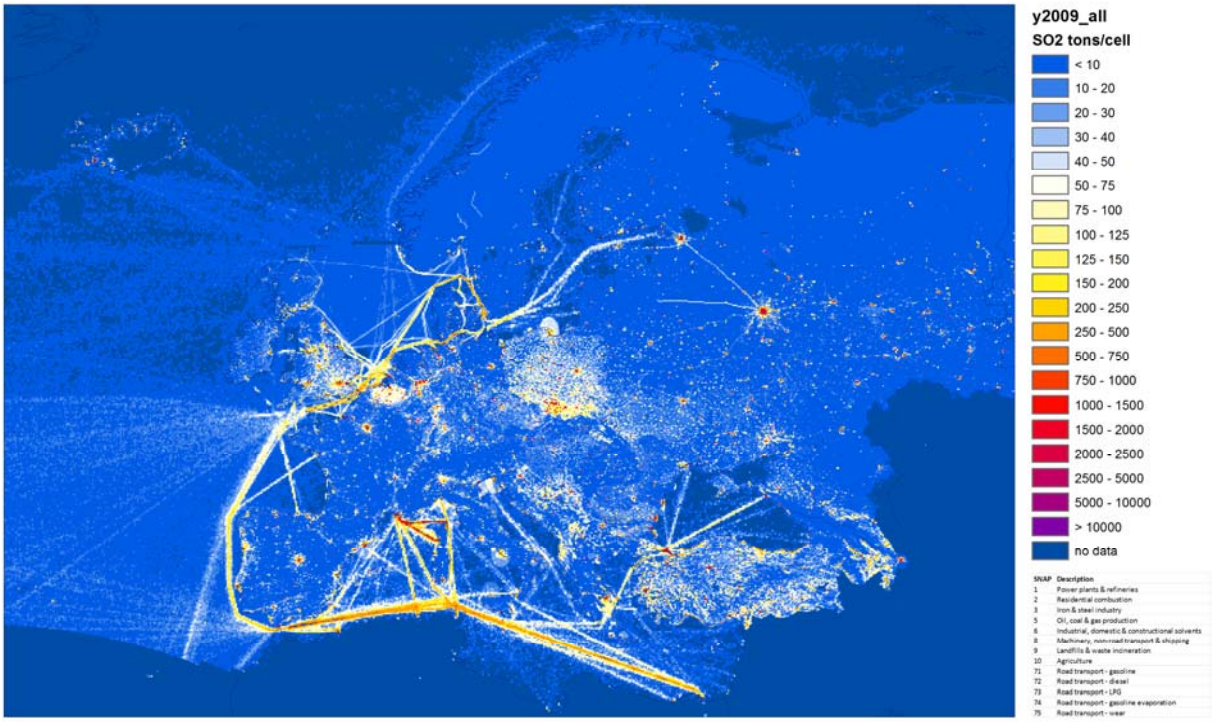
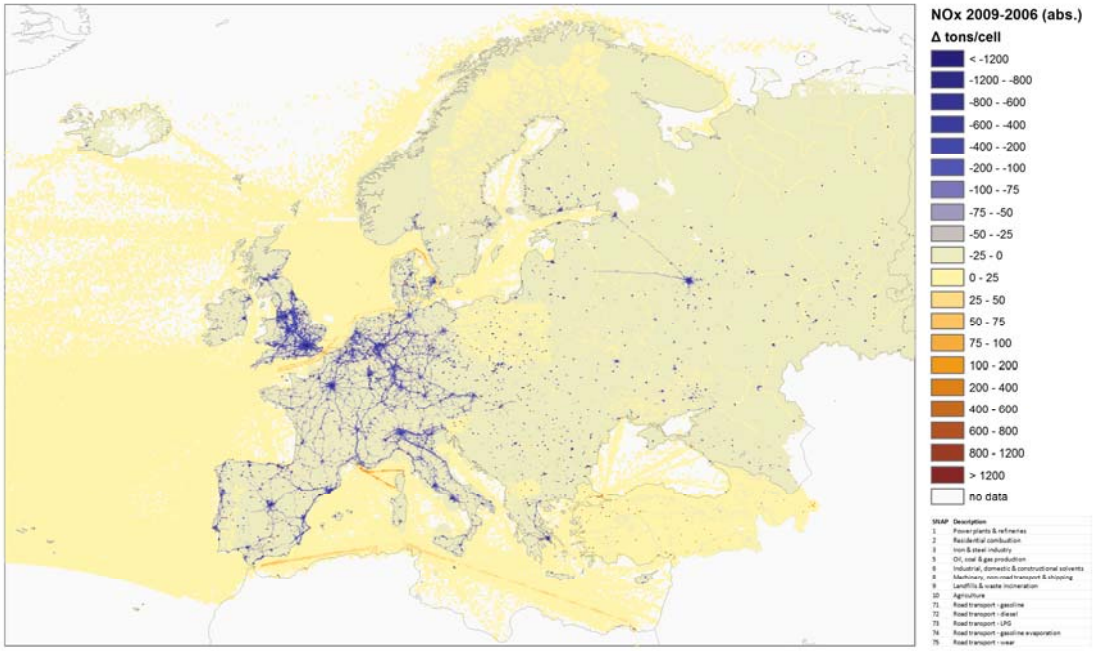
Figure 4


y2009_all
SO2 tons/cell
< 10
10 - 20
20 - 30
30 - 40
40 - 50
50 - 75
75 - 100
100 - 125
125 - 150
150 - 200
200 - 250
250 - 500
500 - 750
750 - 1000
1000 - 1500
1500 - 2000
2000 - 2500
2500 - 5000
5000 - 10000
> 10000
no data
SNAP Description
1 Power plants & refineries
2 Residential combustion
3 Iron & steel industry
5 Oil, coal & gas production
6 Industrial, domestic & constructional solvents
8 Machinery, non-road transport & shipping
9 Landfills & waste incineration
10 Agriculture
71 Road transport - gasoline
72 Road transport - diesel
73 Road transport - LPG
74 Road transport - gasoline evaporation
75 Road transport - wear

862 **Figure 5**



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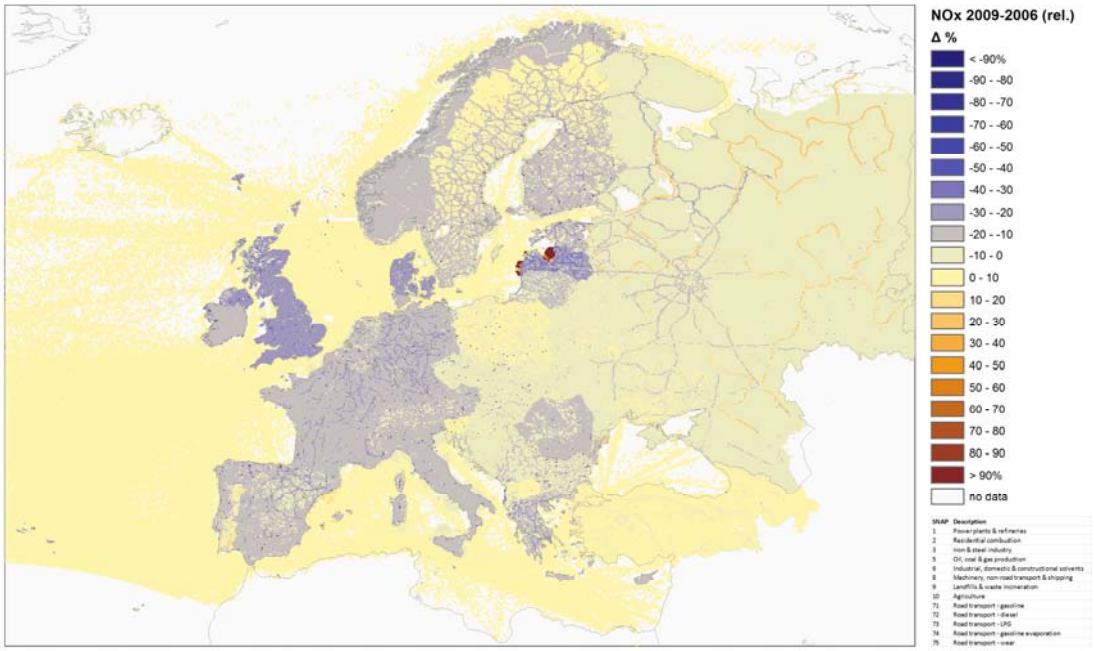

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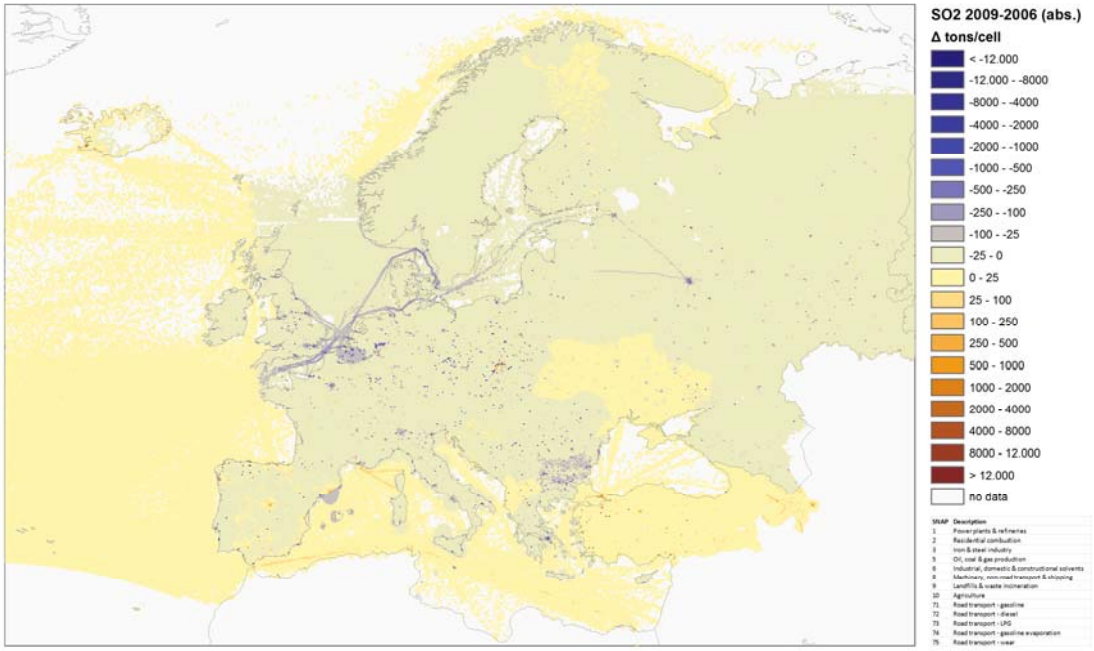

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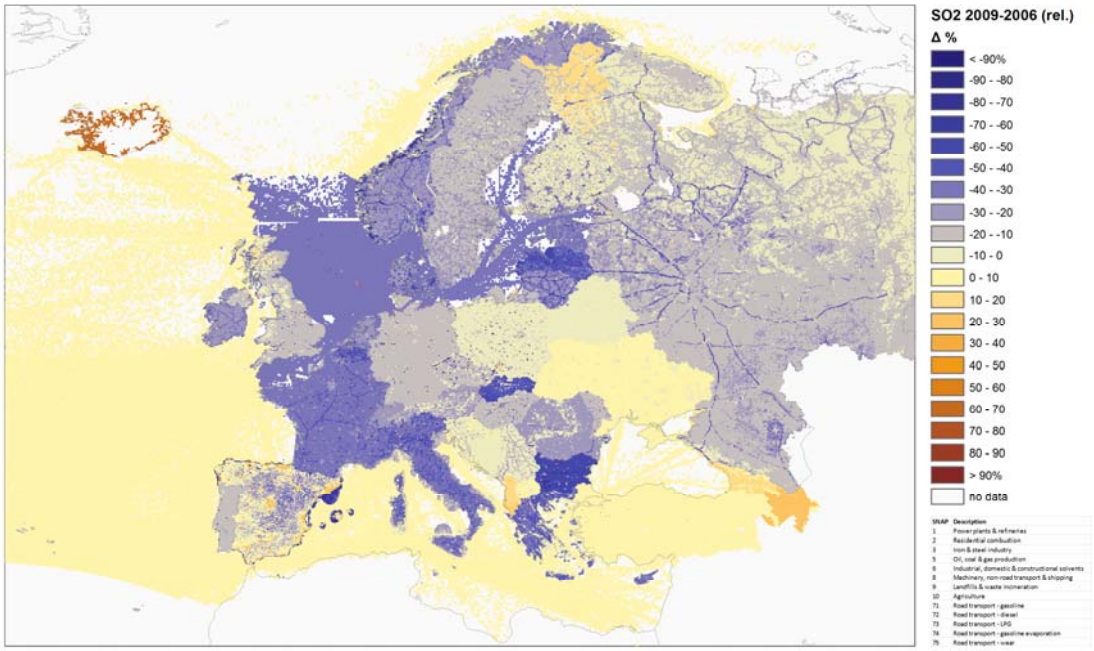

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870 Figure 7

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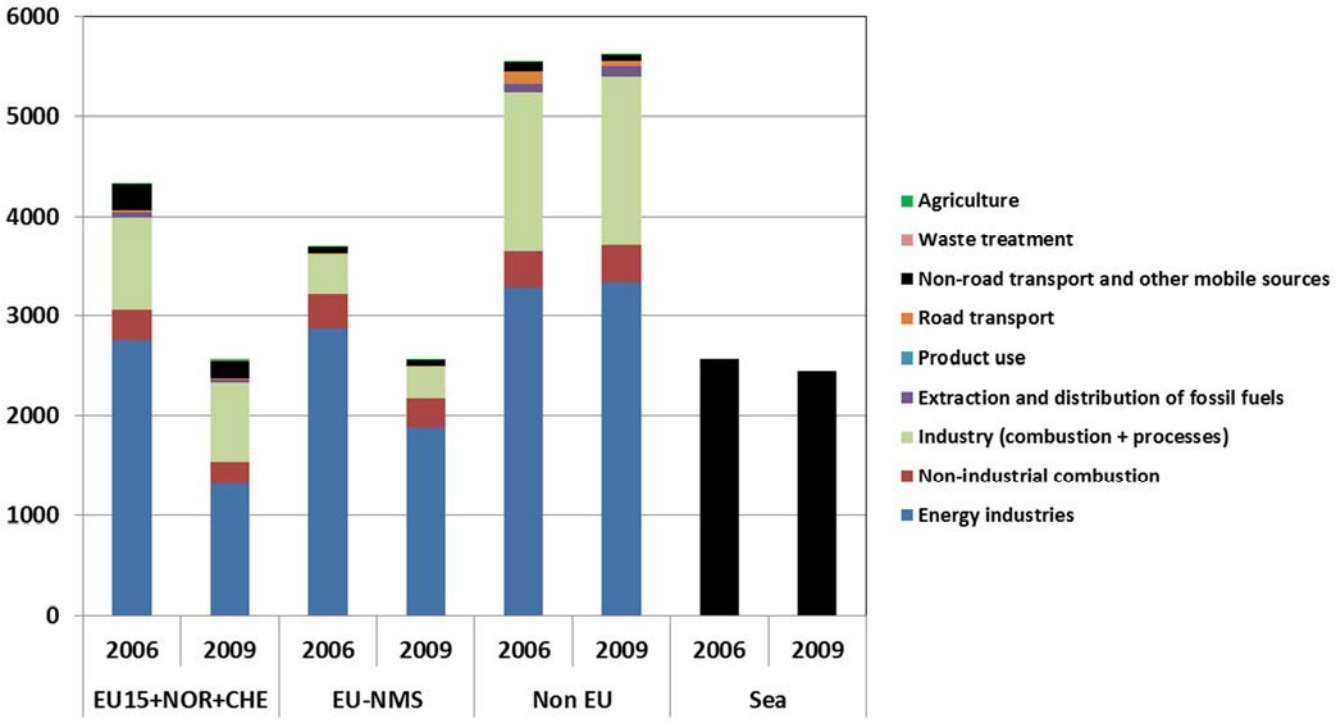

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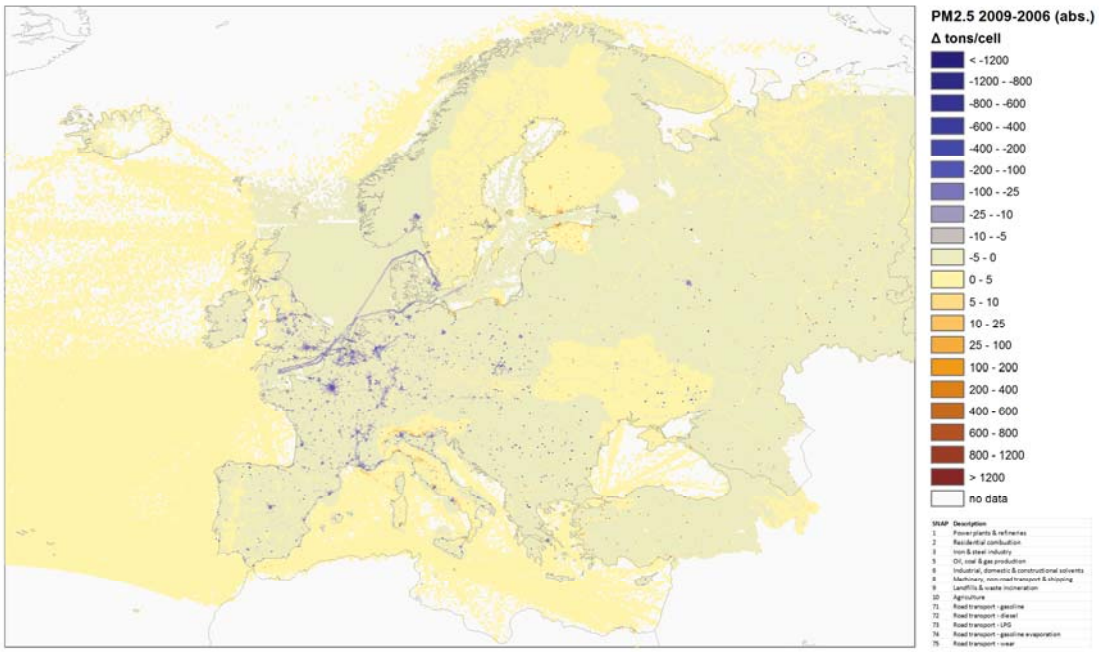
875 Figure 8



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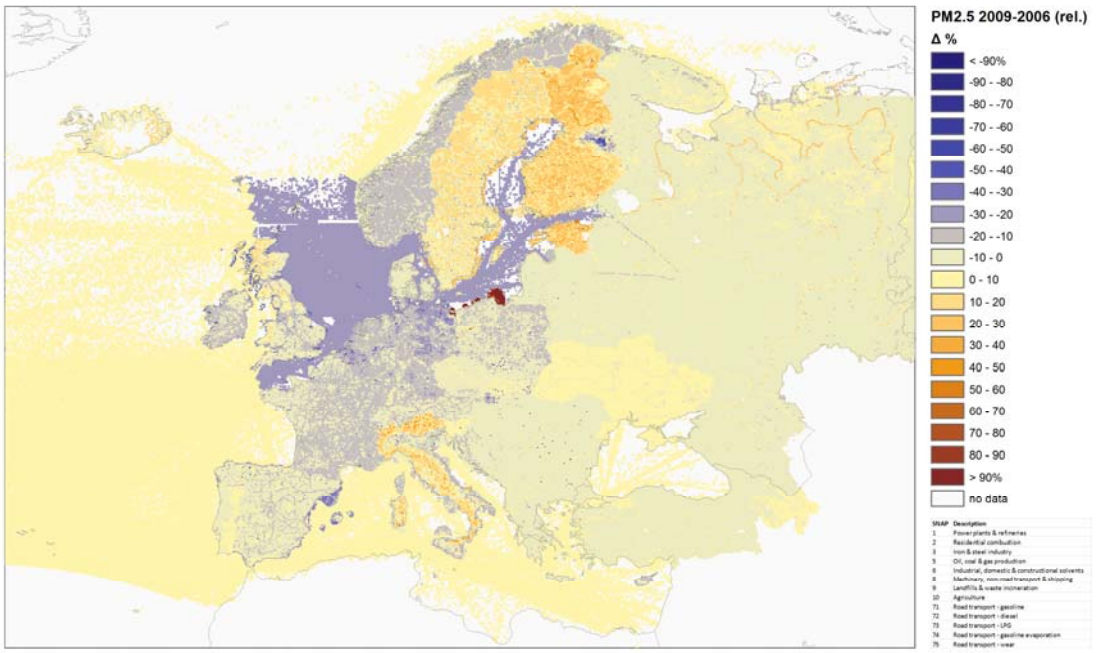

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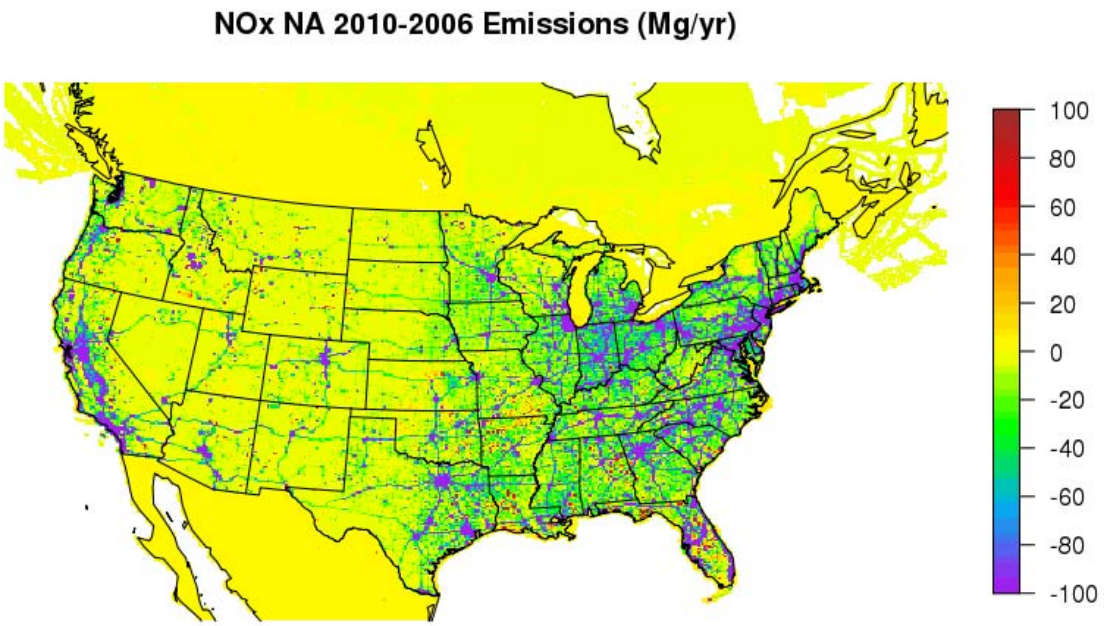
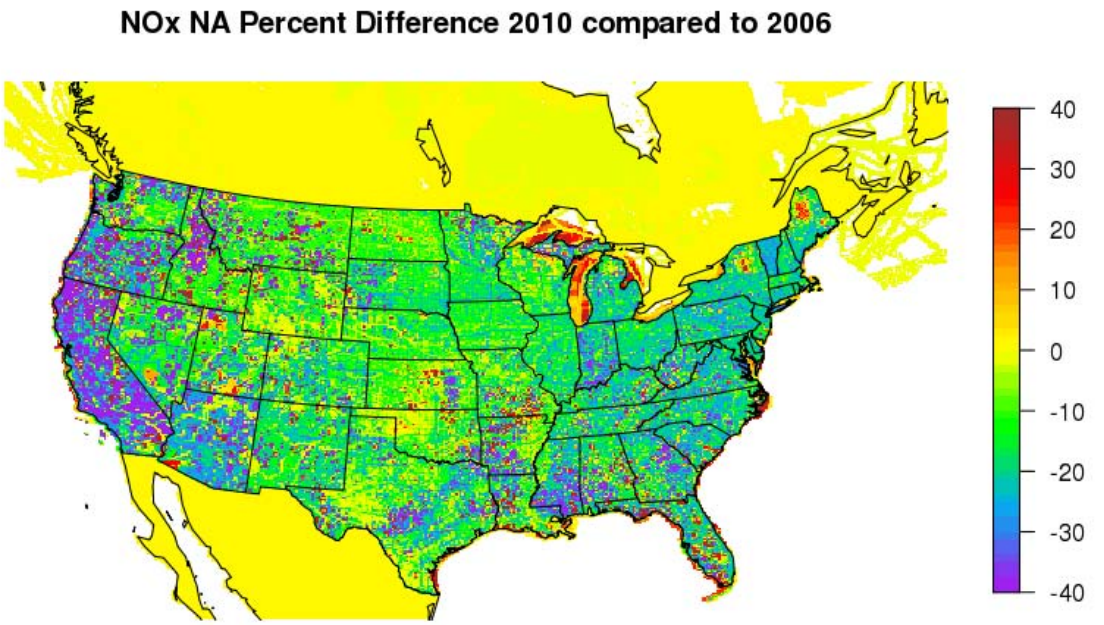
880 Figure 9





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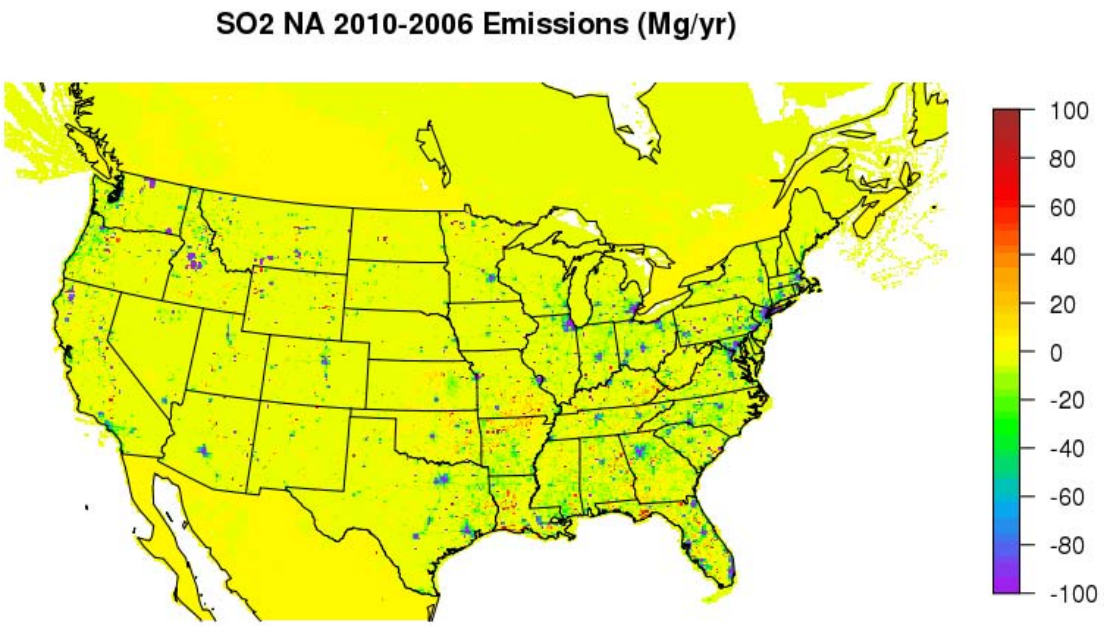

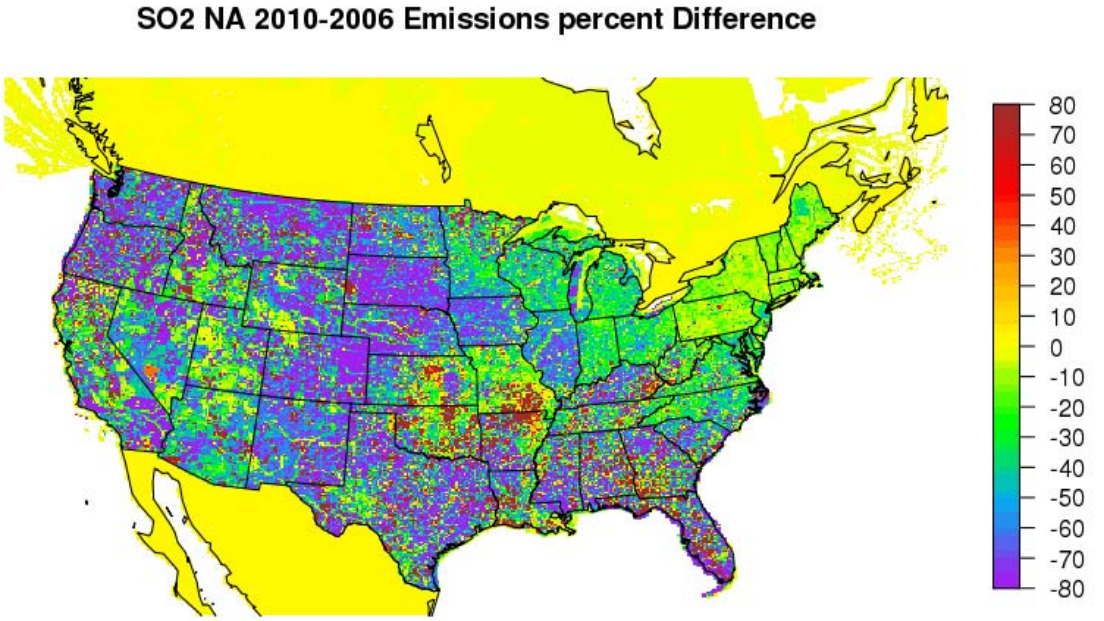

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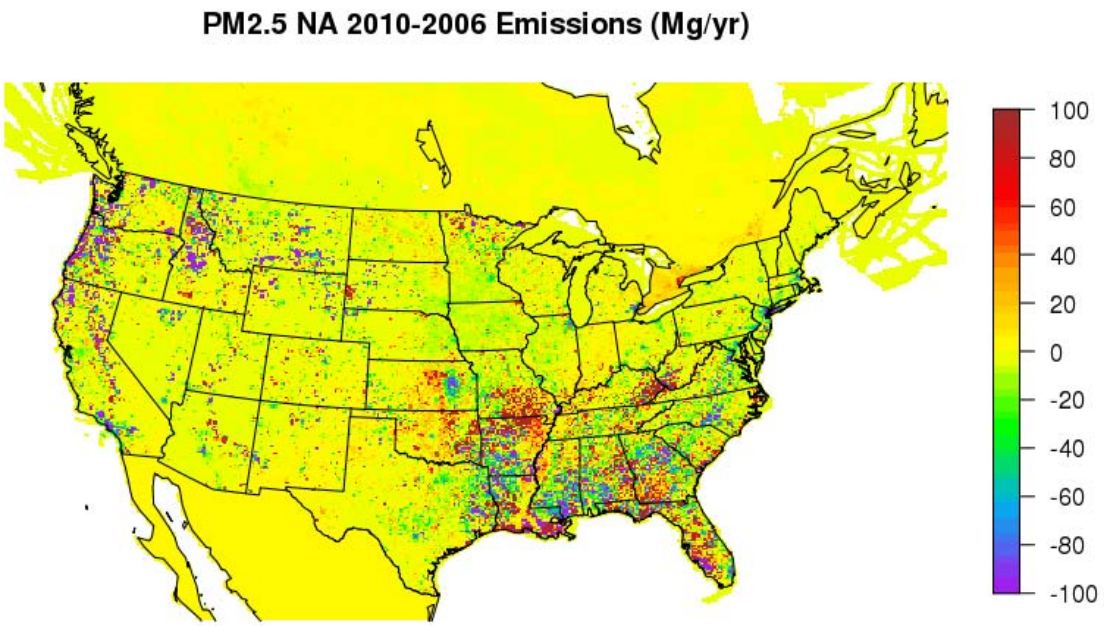

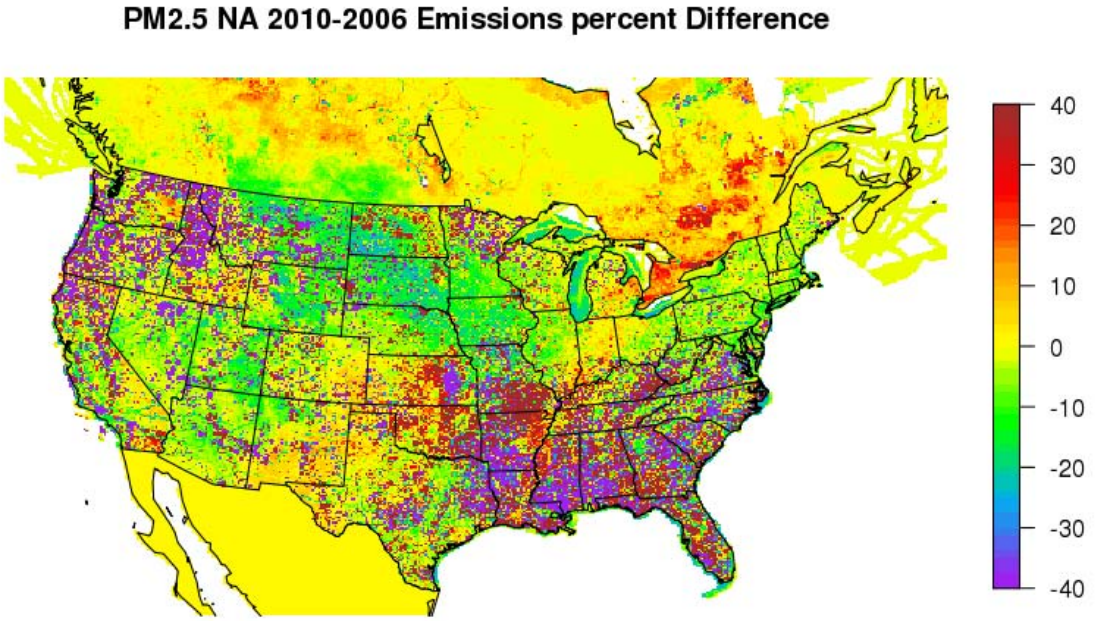

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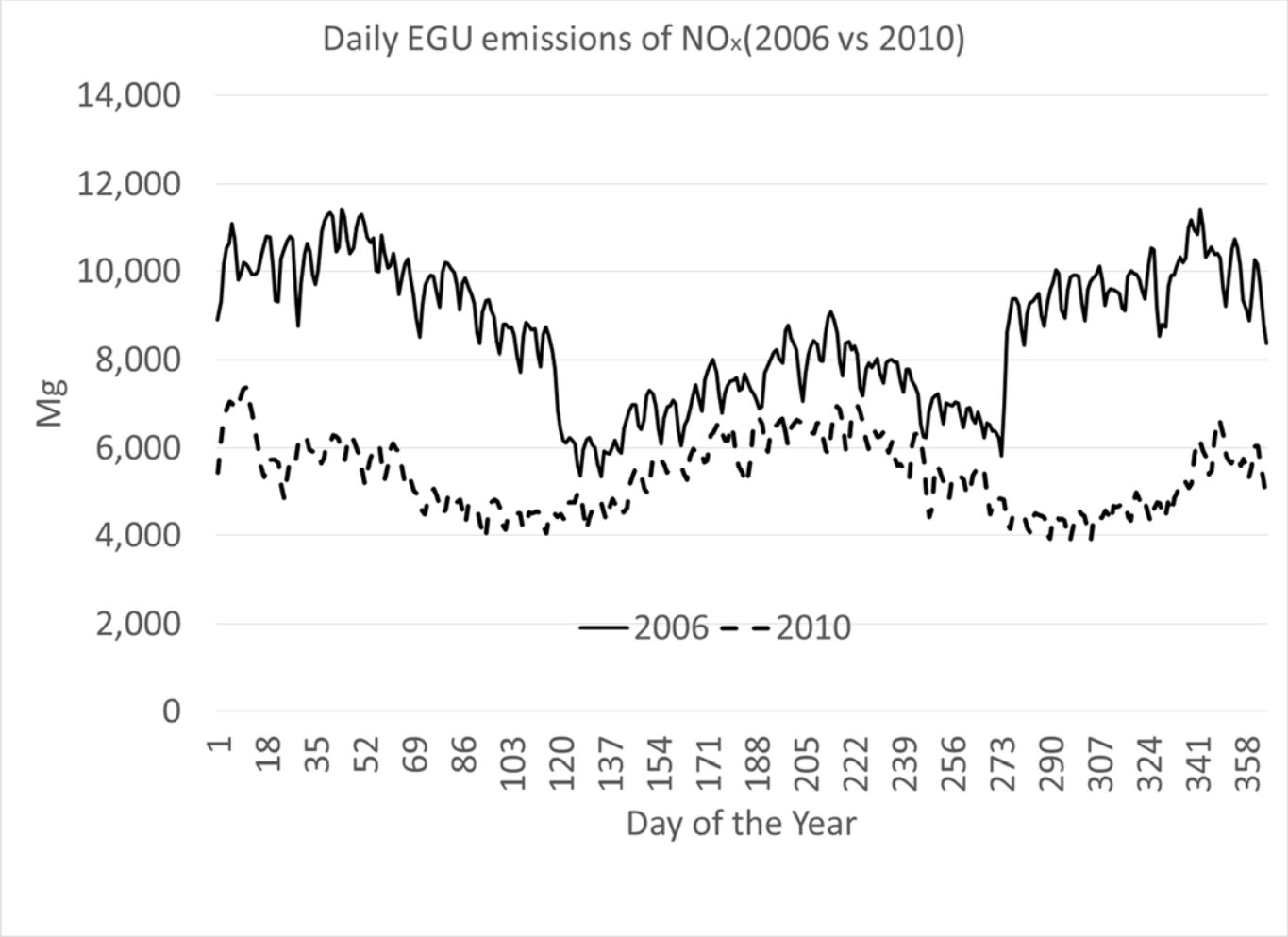

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892 Figure 13

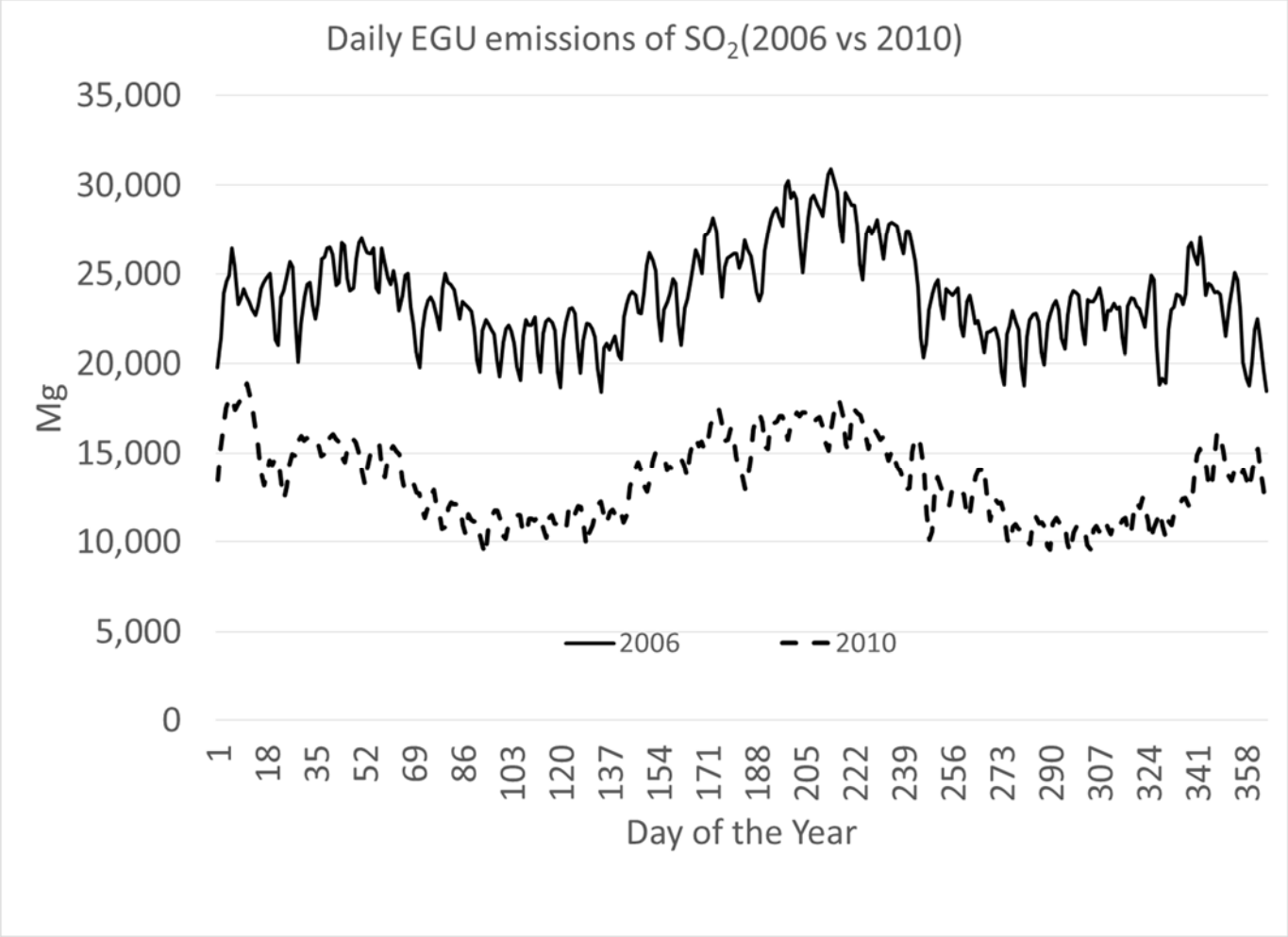

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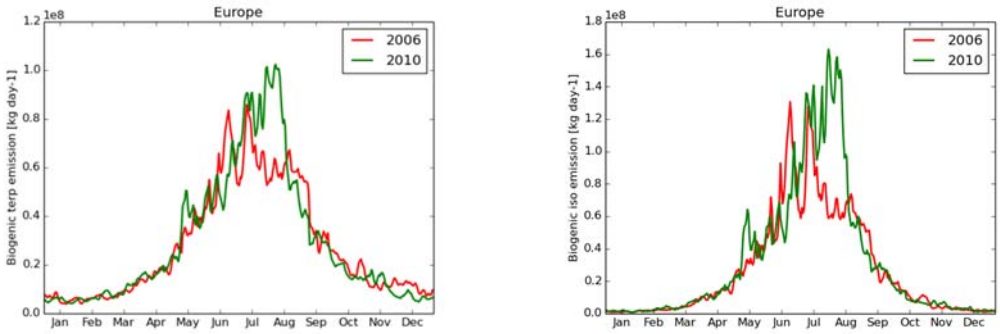
Figure 14



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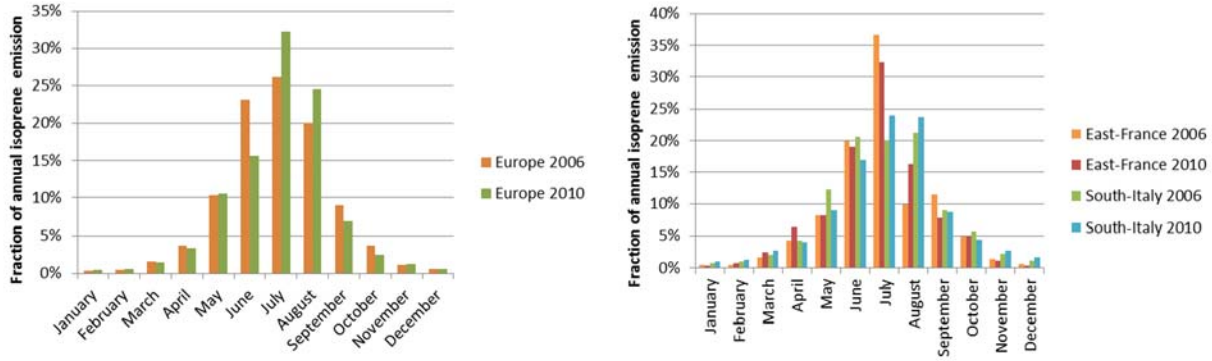
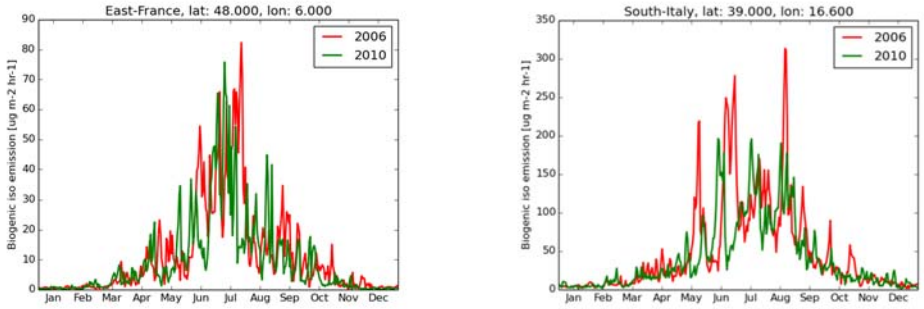


Figure 16



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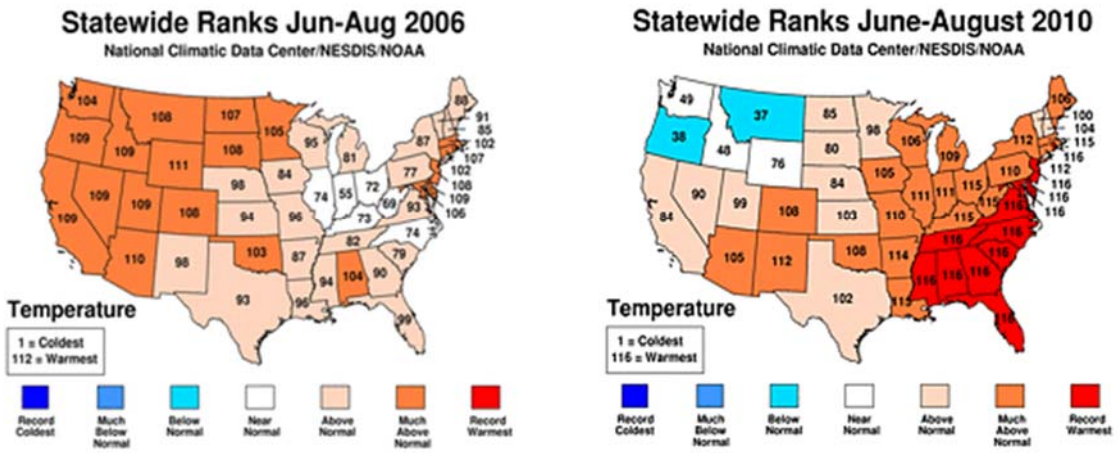

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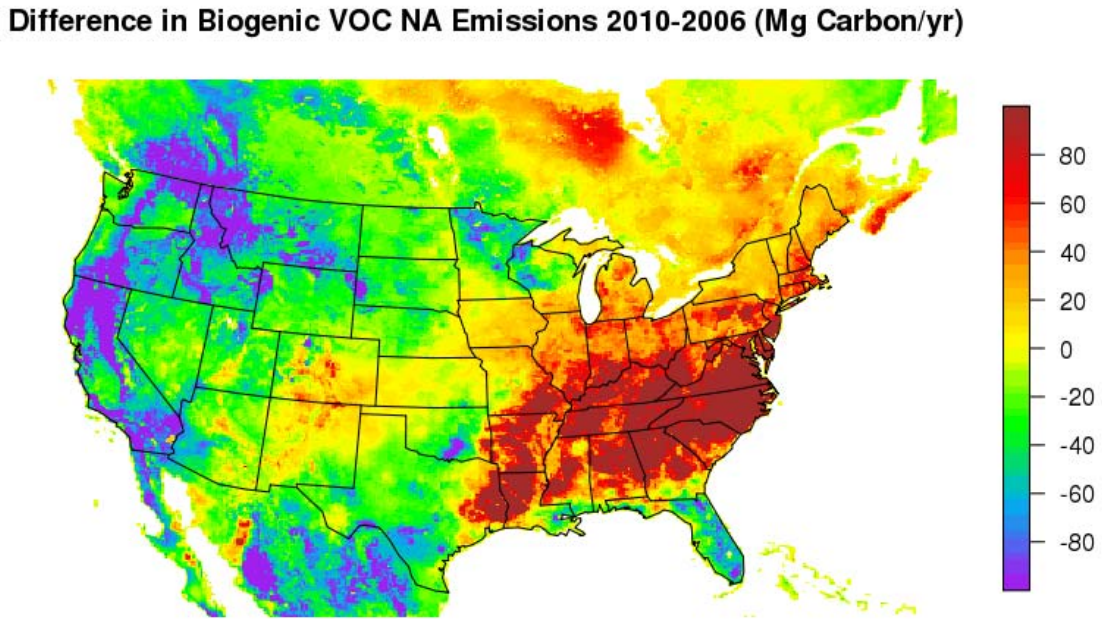

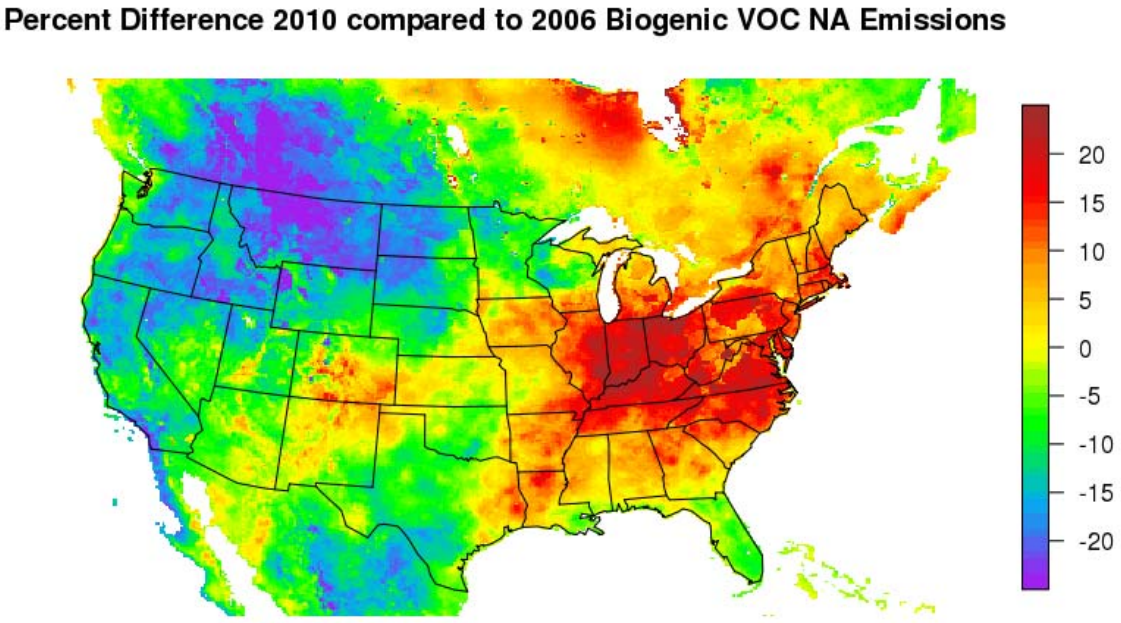

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Supplementary Tables

Table S1 Country groups distinguished in presenting the European emissions data.

Group	Country	Group	Country	Group	Country
EU15 + CHE,NOR	Austria	EU-NMS[a]	Bulgaria	Other Non EU	Albania
	Belgium		Cyprus		Armenia
	Denmark		Czech Republic		Azerbaijan
	Finland		Estonia		Belarus
	France		Hungary		Bosnia and Herzegovina
	Germany		Latvia		Georgia
	Greece		Lithuania		Iceland
	Ireland		Malta		Macedonia
	Italy		Poland		Moldova
	Luxembourg		Romania		Russian Federation
	Netherlands		Slovak Republic		Serbia
	Portugal		Slovenia		Turkey
	Spain		Croatia (HRV)		Ukraine
	Sweden				
	United Kingdom				
	Norway (NOR)				
	Switzerland (CHE)				

[a] NMS = New Member States

Table S2 Selected Nomenclature for Air Pollution (SNAP) level 1 source categories and the link to CRF/NFR[a]

SNAP (level 1)	SNAP name	CRF/NFR[a] codes
01	Combustion in energy and transformation industries	1.A.1
02	Non-industrial combustion plants	1.A.4, 1.A.5
03[b]	Combustion in manufacturing industry	1.A.2
04[b]	Production processes	2
05	Extraction and distribution of fossil fuels and geothermal energy	1.B
06	Solvent and other product use	3
07	Road transport	1.A.3.b
08	Other mobile sources and machinery	1.A.3 (excl. 1.A.3.b)
09	Waste treatment and disposal	6
10	Agriculture	4

[a)] CRF = Common Reporting Format; NFR = Nomenclature For Reporting

[b)] In TNO-MACC-II and AQMEII phase 2 the SNAP sectors 3 and 4 are combined into a new category SNAP 34. This was done because the separation between industrial combustion (SNAP 03) and industrial process emissions (SNAP 04) is often difficult to establish, creating considerable uncertainty when comparing reported emissions by various countries.