Development of an Anthropogenic Emissions Inventory for Annual Nationwide Models-3/CMAQ Simulations of Ozone and Aerosols

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ABSTRACT

The U.S. EPA is undertaking a “Proof-of-Concept” effort which includes applications of the Models-3/CMAQ (Community Multiscale Air Quality) modeling system for a domain covering all of the continental United States as well as adjacent portions of Canada and Mexico. The intent is to determine the feasibility of applying this modeling system for a full year over a nationwide domain and to extend our knowledge of the behavior of the model for simulating ozone and aerosols. This paper describes the modifications and updates made in processing anthropogenic emissions used for performing national annual CMAQ simulations for ozone and aerosols.

The cornerstone of the U.S. anthropogenic emissions data base was the 1996 National Emissions Trends (NET) inventory Version 3.11. From this inventory, the primary emissions of VOC, NOX, CO, SO2, PM2.5, PM10, and NH3 were speciated into the chemical mechanism classes and directly emitted sulfate, nitrate, elemental carbon, organic aerosols, other fine particles <2.5 Fg/m^3, and coarse particles (particle diameters between 2.5 and 10 Fg/m^3). As part of this process a new methodology was developed and implemented for estimating gaseous sulfate emissions for certain combustion source categories based on SO2 emissions from these sources.

The species emissions from stationary area, highway vehicle, and nonroad source categories were gridded at 36 km resolution on a Lambert conformal projection with a centroid of 100 degrees West longitude and 40 degrees North latitude. Traditional procedures for gridding highway emissions, which had been based primarily on population, were substantially improved. The new method includes gridded surrogates for six general roadway types: urban primary, urban secondary, rural primary, rural secondary, urban local, and rural local. These surrogates were developed using the Census Topological Integrated Geographic Encoding Referencing (TIGER) line files and block-level 1990 census data. The result is a more representative spatial distribution of highway emissions in relation to the location of roadways.

The annual emissions were temporally disaggregated to hourly emissions by season/day type (i.e., typical weekday, typical Saturday, and typical Sunday) for stationary and nonroad sources and by

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month/day type for highway vehicles. New procedures were implemented for creating seasonally varying NH3 emissions as opposed to assuming a flat annual profile.

All of the emissions processing for anthropogenic sources was performed using the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system. As part of this process SMOKE was enhanced to incorporate user-defined reports intended to improve the quality assurance of each stage of emissions processing.

INTRODUCTION

It is generally recognized that pollutants in the atmosphere undergo chemical transformation, transport, and removal through highly complex interdependent processes over multiple time and space scales. The ability to simulate these processes in an air quality model is necessary to properly evaluate the relative effectiveness of emissions control strategies intended to mitigate unhealthy concentrations of pollutants such as ozone, fine particles, and toxics, as well as ecologically harmful levels of acid deposition and degradation in visibility. In this regard, the U. S. Environmental Protection Agency (EPA) has recently developed the CMAQ model[1] to provide a tool for integrated "one atmosphere" assessments of pollutants associated with these environmental problems. This model is being applied and evaluated by the EPA Air Quality Modeling Group as part of a "Proof of Concept" effort to examine the feasibility of using CMAQ for regulatory analyses involving annual nationwide simulations of ozone, fine particles, and regional haze. These model simulations are being made using a horizontal grid cell resolution of 36 x 36 km for a domain shown in Figure 1. The CMAQ simulations and results are described elsewhere[2]. The anthropogenic emissions inputs used for these simulations were derived from the 1996 NET, version 3.11[3] with revisions to highway mobile source emissions as developed for the EPA Heavy-Duty Diesel Rule[4]. This base data set provides source category specific emissions of criteria pollutants for individual point sources and county total emissions for stationary area, nonroad mobile, and highway mobile sources. Point, stationary area, and nonroad emissions are provided as annual totals, whereas highway emissions are available as monthly totals. The SMOKE[5] emissions model was used to process emissions of volatile organic compounds (VOC), oxides of nitrogen (NOX), carbon monoxide (CO), sulfur dioxide (SO2), ammonia (NH3), particles with a diameter less than 10 Fg/m^3 (PM10), and particles with a diameter less than 2.5 Fg/m^3 (PM2.5) into the hourly, speciated, gridded emissions needed for the CMAQ simulations[6]. A number of adjustments were made to the emissions data available from the NET inventory for the purposes of this modeling study. In addition, several enhancements were made to certain aspects of the emissions modeling and quality assurance procedures. The purpose of this paper is to provide an overview of the adjustments and enhancements made to the anthropogenic emissions as part of creating model-ready inputs to CMAQ for our annual simulations.

ADJUSTMENTS FOR MODELING THE NET INVENTORY

Three source types in the NET inventory were given special treatment for this modeling exercise. First, we made an adjustment to PM2.5 and PM10 emissions from certain fugitive source categories to remove what is termed the "non-transportable" component of these emissions. This component represents an approximation of the portion of fugitive emissions that settle out and are not dispersed more than a few meters from where they are emitted. Particulate emissions for the source categories listed in Table 1 were reduced by 75 percent to simulate the effects of this settling process. This adjustment was made because the emissions factors and activity data used in calculating fugitive emissions are designed to provide total emissions estimates whereas the nature of the processes which lead to such emissions (e.g., vehicles traveling on unpaved roads) result in much of the particle mass being deposited close to the location of the release[7].

Figure 1. The nationwide CMAQ modeling domain used for annual simulations.

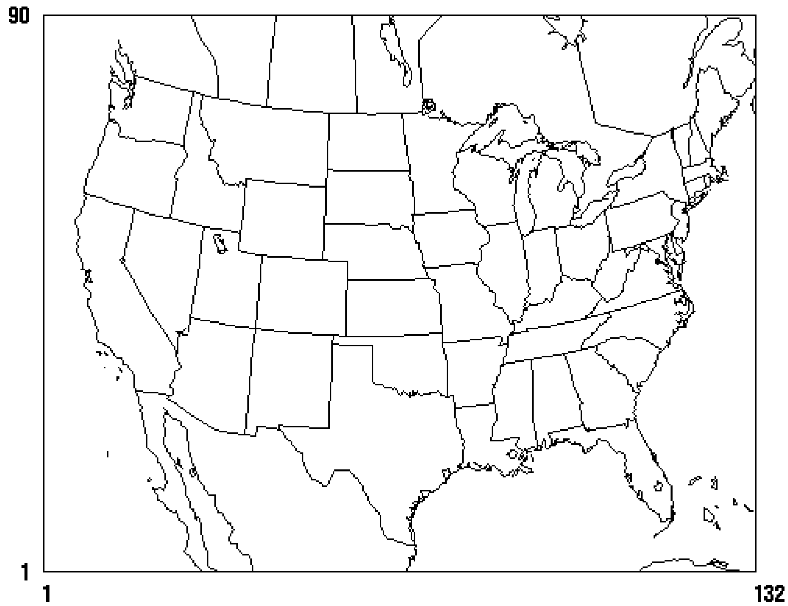


Table 1. Source categories for which the "non-transportable" reduction factor was applied to PM2.5 and PM10 emissions.

Source Category Description	**SCC[1]**
Unpaved Airstrips	22-75-08x-xxx
Paved Roads	22-94-xxx-xxx
Unpaved Roads	22-96-xxx-xxx
Construction/Wind Erosion	23-11-000-1xx
Agriculture Production-Crops	28-01-0xx-xxx
Agriculture Production-Fertilizer Application	28-01-7xx-xxx
Agriculture Production-Livestock	28-05-xxx-xxx

1. "x" is used to indicate that all applicable sub-SCCs are included.

In addition, emissions from two source types in the NET were not carried forward into the model simulations. These include wind blown dust (i.e., SCC 27-30-100-000 and -001) and wild fires (i.e., SCC 28-10-001-000). Emissions of wind blown dust were removed as they occur during episodic conditions such as dust storms, whereas the emissions for this category in the NET represent an annual total. The occurrence of episodes during the year, as well as the severity and duration of individual episodes, are not captured in the NET. Thus, we decided not to include wind blown dust emissions in our modeling as (a) there was no adequate method to temporally distribute these emissions to specific episodes and (b) we felt that it would be inappropriate to simply distribute the emissions uniformly throughout the year. The wild fires category was not included because it was determined that the1996 emissions for some States were calculated by growing emissions from an earlier year. Comparing emissions across several States indicated that this process of estimating emissions when actual 1996 emissions were not available resulted in atypically high particulate emissions in certain counties and States.

NEW METHOD FOR ESTIMATING SULFURIC ACID EMISSIONS

The treatment of sulfate aerosol formation in CMAQ includes processes that are dependent, in part, on the formation rate and concentration of sulfuric acid vapor (SULF). The rate of production of SULF from the reaction of SO2 with hydroxyl radicals is used as a basis for determining the rate of sulfate particle formation through nucleation and condensation. In the presence of clouds, SULF is absorbed into the cloud water and added to the accumulation mode sulfate aerosol mass. In addition to the chemical formation of SULF in the atmosphere, this species is directly emitted in small amounts by some combustion processes, but is not inventoried in the NET. The following procedures were developed to estimate SULF emissions for the purposes of our CMAQ applications.

For certain combustion sources, gaseous sulfate emissions were calculated as the multiplicative product of SCC-specific ratios and SO2 emissions. In general the methodology is based on information in AP-42[8] which identifies the fractions of each fuel's sulfur that are emitted as sulfate and as SO2. Using these fractions and the amount of SO2 reported in the NET we estimated the emissions of sulfate. In these calculations we assumed that gaseous sulfate, which actually includes a number of components, is primarily sulfuric acid. The equation for calculating SULF emissions is given below.

SULF (as H2SO4) = SO2 * (fraction of sulfur emitted as sulfate) / (fraction of sulfur emitted as SO2) * (98 / 64),
where 98 and 64 are the molecular weights (g/mol) of H2SO4 and SO2, respectively.

Note that the reported emissions of SO2 are not reduced as part of this methodology. The source categories and relevant fractions of fuel sulfur emitted as SO2 and as sulfates which were used for estimating gaseous sulfate emissions from SO2 emissions are provided in Table 2 for bituminous coal combustion, sub-bituminous coal combustion, lignite coal combustion, and oil combustion. Note that the numbers in the table do not sum to 1.0 for each fuel type. This reflects the fact that a small fraction of sulfur in the fuel is not emitted as SO2 or gaseous sulfate, but becomes part of the ash from the combustion process and is either emitted as part of the source's particulate emissions or is retained in the bottom ash.

Table 2. Fraction of sulfur emitted as SO2 and gaseous sulfate.

Fuel Type	Fraction as SO2	Fraction as gaseous sulfate
bituminous coal	0.950	0.014
sub-bituminous coal	0.875	0.014
lignite coal	0.750	0.014
residual oil	0.990	0.010
distillate oil	0.990	0.010

UPDATED APPROACH FOR SEASONAL AMMONIA EMISSIONS

The formation of nitrate and sulfate aerosols in the atmosphere is strongly dependent upon the availability of NH3. In the presence of water vapor NH3 reacts with nitric to form ammonium nitrate and reacts with sulfuric acid to form ammonium sulfate. The chemical pathways (i.e., gas-phase versus aqueous phase) and reaction rates for nitrate and sulfate formation are, in part, temperature and moisture dependent. In this regard, the extent of nitrate and sulfate formation is highly seasonal, with the highest nitrate levels observed during the cooler months and the highest sulfate levels observed during the warmer months. Traditionally, a flat profile has been used to temporally allocate annual NH3 emissions. This results in 25 percent of the annual total emissions being allocated to each season. However, given the seasonality in meteorological conditions favoring nitrate versus sulfate formation, it is important to consider how NH3 emissions change by month or by season in order to properly simulate aerosol formation over an annual time period. For our model applications we used a set of seasonal profiles for the animal husbandry operations and fertilizer application source categories derived from the results of a cursory analysis by the EPA Office of Research and Development[9]. These two categories are the major sources of NH3 emissions in the NET. Animal husbandry is the largest category, representing 74 percent of the total annual mass of NH3 emissions, with fertilizer applications contributing another 12 percent. There are still considerable gaps in the state-of-knowledge needed to adequately estimate the annual amount of NH3 emissions from these sources categories, as well as, how these emissions vary during the year. Estimating seasonal variations in emissions from these categories is quite complex for a number of reasons. For example, ammonia is very volatile and thus, emissions from sources like animal waste lagoons are very sensitive to temperature and moisture. Also, the time and extent of fertilizer applications in the Spring vary geographically as well as by crop type.

In brief, the factors developed for animal husbandry take into consideration that most of the emissions come from cattle, swine, and poultry. These animals are generally kept in housing, stockyards, or pastures. Emissions from housing were assumed to be constant given the relatively stable environmental conditions and nutrient intake. It was also assumed that emissions from housing represent half of the total from this sector, based on a review of published studies. The remaining emissions which come from waste storage lagoons, slurry applications, and when animals are in the fields tend to be more subject to variations associated with changes in ambient temperature and moisture. Emissions from waste storage and slurry applications were assumed to be largest during warm, dry conditions when ammonia volatilization is greatest. The result of these considerations lead to a seasonal distribution of 15 percent in the Winter (i.e., December, January, and February), 25 percent in the Spring, 40 percent in the Summer, and 20 percent in the Fall. Table 3 provides the animal husbandry source categories for which we applied these seasonal factors.

The seasonal distribution of NH3 emissions from fertilizer applications was estimated using the amount of fertilizer applied by crop type in conjunction with information on when these applications typically occur for the major crop types of corn, wheat, cotton, and soybeans. Approximately 70 percent of the fertilizer is applied to corn and cotton and occurs mainly during the Spring and early Summer. Most of the remaining fertilizer is applied to plantings of winter or summer wheat which was assumed to be evenly distributed throughout the year. In view of these considerations a seasonal allocation of 10 percent in the Winter, 50 percent in the Spring, 30 percent in the Summer, and 10 percent in the Fall was used for emissions from the fertilizer source categories (i.e., SCC 28-01-700-001 thru -010). The net effect of applying the new seasonal factors to both animal husbandry and fertilizer applications is that 16 percent of annual total NH3 is allocated to the Winter, 28 percent to the Spring, 37 percent to the Summer, and 19 percent to the Fall.

Table 3. Animal husbandry categories for which new seasonal factors were applied.

Animal Husbandry Categories (SCC)
Cattle (28-05-020-000)
Hogs (28-05-025-000)
Poultry (28-05-030-000)
Sheep (28-05-040-000)
Goats (28-05-045-000)
Horses and Ponies (27-10-020-030)

REVISED METHOD FOR SPATIAL ALLOCATION OF MOBILE EMISSIONS

Traditionally, the process followed by EPA for spatially allocating vehicle miles traveled (VMT), or mobile emissions from countywide totals to modeling grids has relied upon urban and rural population as a spatial surrogate for all roadway types, except for interstates. For interstate VMT or emissions, urban and rural interstate surrogates have been used. For the other roadway types, (i.e., urban and rural arterial and collectors) this procedure can result in mobile emissions for individual roadway types being placed in grids where there are no roadways of that type. To avoid this problem a new spatial allocation method was developed that combines 1990 census block rural and urban population with Geographic Information System (GIS) coverages for primary roads, secondary roads, and local roads based upon the TIGER data base of roadway links. This method resulted in gridded surrogates for six roadway types: urban primary, rural primary, urban secondary, rural secondary, rural local, and urban local. To calculate these surrogates, the 1990 census block rural population was subtracted from the total population to calculate urban population. Urban areas were then defined as those areas with more than 50 percent of the population in the urban category. Next, data for TIGER Census Feature Class Codes (CFCC) were consolidated into three groups: primary roads, secondary roads, and the remainder as local roads. This information was then merged using GIS techniques with urban area designations and the 36 km grid to create urban and rural gridded surrogates for each roadway type. Details on the procedures for generating these surrogates as well as the effects on estimated emissions are provided in a companion paper[10].

IMPROVED EMISSIONS PROCESSING QUALITY ASSURANCE TOOLS

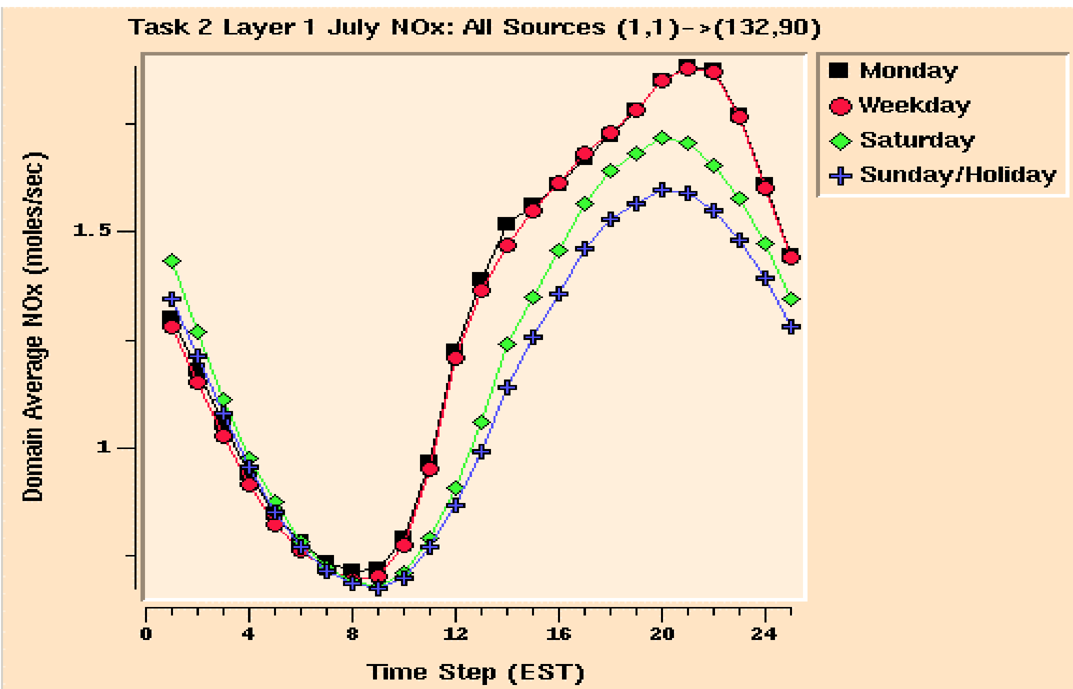
In order to enhance quality assurance during the emissions modeling portion of this effort, the SMKREPORT module in SMOKE was enhanced to provide the user with the option of generating additional reports summarizing emissions data during different stages of the processing.[11] These reports can be generated at the time of data import, as well as during speciation and/or temporal and spatial allocation of emissions data. The structure of these reports has been designed to permit easy import into spread sheets and in the preparation of printed reports. Reports can be generated with various logical combinations of the breakouts listed below.

- geographic: State, county, domain, grid cell(s)

- temporal: annual, day-specific, hour
- pollutant: speciated, non-speciated
- category: point, area, mobile, SCC, road class

The SMKREPORTs together with graphical displays of the data were used to help ensure the quality of the emissions processing. In brief, qualitative analyses of gridded and temporalized visualizations of different pollutants were examined to determine whether there were problems with the input data. In addition, the location of urban centers and highways, temporal profiles for the different source categories, and the omission of areas within the domain were all checked in this analysis. Time series plots were created to compare emissions for different months and day types to identify any atypical patterns or relationships which might indicate a potential problem in the data and/or processing. Figure 2 shows an example time series plot that displays the hourly profiles of July NOx emissions for each day type for a portion of the modeling domain in the Northeast. These profiles confirm that, as expected, the highest daytime emissions occur on weekdays with lesser emissions on Saturdays and Sundays. Quantitative analyses of the inventory reports were also prepared to ensure that SMOKE correctly processed the information contained in the raw inventories. Inventory reports were generated by the SMKREPORT module to check the emissions generation process from the SCC level up to the level of fully merged emissions for the entire domain. Random and frequent checks of the SMOKE log files were made to uncover problems with the SMOKE inputs, the processing, and scripting. The combination of these three steps led to highly quality-assured emissions and emissions modeling data sets in the context of SMOKE processing.

Figure 2. Diurnal profiles of weekday, Saturday, and Sunday NOx emissions for July.



CONCLUSIONS

As part of a project to generate model-ready emissions estimates for nationwide annual CMAQ simulations of ozone and primary and secondary fine particles, EPA made a series of adjustments and improvements to various sectors of the emissions inventory. These changes were made to enhance the quality of the emissions driving the model simulations, in view of the nature of the available data.

Certain adjustments including the reduction in emissions made to account for the "non-transportable" component of fugitive PM and the seasonal profiles for NH3 are considered interim and will be revised in time as newer information becomes available. Similarly, it may be possible, in the long term, to include estimates of emissions from wild fires and episodes of wind blown dust as more specific data on the occurrence of these emissions in time and space can be determined. It is hoped that other groups will consider evaluating the new techniques used in this project for estimating sulfuric acid emissions and for gridding mobile emissions. Finally, the new reporting features developed for SMOKE are expected to help others to more readily quality assure emissions inputs for grid-based air quality model applications.

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KEYWORDS

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SMOKE
Spatial Surrogates