Inter-annual and Seasonal Variability of Meteorologically Influenced Emissions

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ABSTRACT

The U.S. Environmental Protection Agency (EPA) is a participant in the U.S. Global Change Research Program (CGRP). The air quality portion of the GCRP addresses the effect on air quality attributable to climate change in the intermediate future (e.g., 2050). The first phase of the program examines the change in air quality with respect to climate change from 2000 to 2050, using a static emission inventory for 2001, except for biogenic and mobile source emissions which are modeled to vary with meteorology. Between five and ten years of modeled meteorology data will be used to define the climate of 2000 and 2050, respectively. This paper is a preliminary examination of the inter-annual and inter-seasonal variability of meteorologically-influenced emissions over a five-year base study period centered on 2000. Emission data for 2001 were used for all five years. Biogenic isoprene emission modeling results suggest definite east to west differences in seasonal and peak emission variability in the United States , and inter-regional peak emission differences in the east (up to approximately 30 percent), a reflection of both vegetation composition and long-term circulation patterns. Biogenic nitrogen oxide emissions follow a similar, but less variable pattern. Mobile source emissions within a region vary less between years than biogenic emissions within a region because the mobile source model is less sensitive to temperature, and the competing influence of other model variables such as prescribed diurnal behavior and vehicle fleet composition. There is a pronounced east to west variability difference in peak hourly emissions (an order of magnitude) related to differences in the density and composition of motor vehicle use. This study is intended to provide further information on the amount of emission changes necessary to provide a signal in climate change beyond background meteorological variability.

INTRODUCTION

The potential effects of climate change over the next hundred years and beyond include possible changes in air quality.[1] Accordingly, the U.S. Environmental Protection Agency (EPA) has embarked on a Climate Impact on Regional Air Quality (CIRAQ) project.[2] The project is coordinated with the EPA CGRP and the U.S. Climate Change Research Program.[3] CIRAQ is focused on the characterizing the effects of climate change on air quality between the present and around 2050. The purpose of this paper is to examine the spatial and temporal variability of diurnal patterns of modeled meteorologically-dependent emissions (biogenic and mobile source emissions) for a five-year base period (roughly 1999-2003) with the mesoscale meteorology driven exclusively by initial and boundary conditions from a large-scale Global Climate Model (GCM). The purpose is also to provide emission estimates and statistics that can be compared against observations of air chemistry constituents to determine the nature and extent of meteorological data bias related to modeling, and its effects on emissions. This will provide baseline information for detection and description of any emission variations due to longer term climate changes when meteorology (without changes in emission drivers) is projected to 2050. The second phase of the project will examine the effects when emissions are also projected.

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APPROACH

Examination of the temporal and spatial variability of the diurnal emissions was accomplished by modeling hourly (episodic) emissions for a five-year period, representing meteorological conditions circa 2000. Separate high-quality inventories for each year to be modeled were not available. Reported emissions (e.g., meteorologically independent) in inventories normally vary little between consecutive years. Changes depend on modeling inputs and methodology rather than meteorology.

Consequently, the inventory applied to all five years was the 2001 version of the modeling inventory, compiled by the EPA Office of Air Quality Planning and Standards.[4] The inventory was the best available for the United States and contains the most recent information on emission sources. The modeling inventory was prepared for use with the Community Multiscale Air Quality (CMAQ) model,[5] by using the Sparse Matrix Operator Kernel Emission (SMOKE) modeling system, Version 2.1, with the Statewide Air Pollution Research Center (SAPRC) chemical mechanism for gaseous criteria pollutants.[6,7] In order to address meteorologically-dependent emissions, SMOKE incorporates the Biogenic Emission Inventory System (BEIS), Version 3.12 for biogenic sources, and MOBILE Version 6.[8,9] for modeling of emissions from mobile sources. Gridded meteorology inputs were generated by providing initial and boundary conditions from the National Aeronautical and Space Administration Global Institute for Space Studies (GISS) Version II' GCM to the Mesoscale Meteorology (MM5) model.[10, 11,12] This dynamic downscaling was performed for the EPA GCRP through a partnership with the Department of Energy Pacific Northwest National Laboratory. No observational or re-analysis data were used to nudge MM5, which approximates the manner in which meteorology data will be simulated for 2050. As a result, the regional climate scenarios will not match MM5 model runs where observational nudging was used. The meteorology data and emissions were modeled for the North American modeling domain shown in Figure 1 at a gridded spatial resolution of 36 km.

Although this paper focuses on meteorologically-influenced emissions, a full range of criteria and precursor chemical compounds were modeled, including isoprene (ISO), nitrogen oxides (NOx), nitrogen oxides (NO), nitrogen dioxide (NO^2), sulfur dioxide (SO^2), ammonia (NH^3), coarse particulate matter (PM^{10}) and fine particulate matter ($PM^{2.5}$) for point and area sources in addition to biogenic and mobile sources emissions. In addition, particulate organic aerosols (POA) and particulate elemental carbon (PEC) derived from $PM^{2.5}$ were examined for mobile sources. Emissions of each of the compounds are examined by geographic regions chosen to represent divisions used in past air quality analyses: These regions are:

- An Eastern Domain (Figure 1). This region typifies the east-west division historically used in air quality modeling because of topographic and atmospheric chemistry differences.

- A Western Domain (Figure 1).

- Each of the five ozone regions of the eastern United States as defined by Lehman et al. based on an analysis of monitoring data using principal component analysis (Figure 2).[13] The regions represent areas of relative consistency but overlap geographically.

Emission data were examined for each region by grouping the emission data as follows:

- Average annual hourly emission summaries (for the five year period) of each of the above emission compounds for biogenic source and mobile sources by domain and the above geographic regions.

- Average seasonal hourly diurnal emission values (hourly) for the modeling domain and each region by season for each biogenic and mobile source emission compound. For this study seasons are defined as the calendar quarters (e.g., January -March (Winter), April - June (Spring), July - September (Summer), October - December (Fall). Seasonal definition may have an effect on the emission statistics.

Standard deviation, variance, and inter-quartile range (IQR) were computed for each of the above groupings to quantify the dispersion from the mean and departure from central tendency for average hourly emission values for each month and season between years (e.g., all Januarys, all Februarys, etc.) and between average hourly emission values on a annual basis between years.

ANALYSIS

The modeled emission fluxes were normalized to emission per square kilometer for each region to aid comparison. The resulting annual and seasonal average hourly values and the statistics were graphed for analysis. The figures shown in this paper are illustrative examples and summaries.

Biogenic Emissions

Isoprene (ISO) is the principle biogenic gaseous emission contributor to ozone formation. Biogenic emissions of ISO driven by dynamically downscaled regional meteorology, demonstrate expected patterns of emissions reflecting the influence of the diurnal temperature and insolation cycle over the contiguous United States (Figure 3). The variability of biogenic emissions differs with temporal scale. For example, annual aggregate ISO emissions vary from the annual mean for the Eastern Domain (0.20 percent) and Western Domain (0.23 percent) relatively little. However, for hourly emissions on annual and seasonal bases, the range of the diurnal emission cycle is an order of magnitude greater in the Eastern Domain than in the Western Domain, and the peak hourly emissions vary by approximately 17 percent from the mean. This reflects vegetation species density and composition differences between the domains. However, the year of greatest emission flux is not the same for both regions.

When diurnal emissions by season are examined, the expected change with season is evident, reflecting temperature and vegetation cover (Figure 4). However, the amount of seasonal change in flux among years and regions also varies, reflecting the influence of regional meteorology. Examination of the family of average hourly curves of ISO emissions by season among the five ozone regions, similar to Figure 4 which shows a variation of about 23 percent from the median at mid-day in summer and less than 2 percent during winter, again demonstrates that the peak year of ozone emissions varies both spatially and temporally. The average IQR of ISO emissions dominates variability of gaseous emissions from all sources in the contiguous United States in all seasons (Figure 5) and illustrates a significantly greater departure from the median during the summer and spring seasons (approximately 60 percent and 40 percent of the total IQR, respectively). The variability of ISO described by the IQR is also greater in the Eastern Domain than in the West (Figure 6). Among the ozone regions, Region 3 (Mid-Atlantic) and to a lesser degree, Ozone Region 4 (South Central), consistently demonstrate greater ISO variability (Figures 7 and 8) in the spring and summer seasons. This translates to a variability of approximately 38 and 28 percent, respectively. The pattern is consistent with an oscillating high pressure feature present during summer periods in the downscaled MM5 700 mb meteorological circulation simulation (Figure 9).[14] The overall variability of hourly emissions of ISO between regions and seasons is summarized in Table 1.

Biogenic nitrogen oxide (NO) diurnal emissions also show an expected diurnal pattern reflecting a combination of air temperature, precipitation, vegetation, and agricultural land cover. Biogenic NO modeling calculates emissions during the growing season from agricultural lands as a function of rainfall and temperature. During the balance of the year, NO is computed as a function of temperature. Values are greater for the Eastern Domain, both annually and seasonally, as well as greater inter-annual variability (Figure 10 and Table 2). For example, in Figure 10 summer variability approximately 6 percent for the Eastern Domain and less than 2 percent for the Western Domain. Again, the year of greatest flux varies between Eastern and Western domains and the seasons. The variability of emission flux of NO among seasons is also illustrated by the standard deviation, variance, and inter-quartile range (IQR) given in Table 1. The statistics (computed on normalized seasonal means) show less variability of NO than ISO, probably because of the greater sensitivity to temperature of the ISO algorithm relative to the NO algorithm. However, the ozone regions demonstrate substantial variability in NO emissions, and the peak year of NO emissions varies with region. The standard deviation is twice as great for Ozone Region 3 as for Ozone Region 1 (New England) and 2 (Great Lakes); and much less for the other ozone regions. The IQR for NO tends to increase while standard deviation decreases during the winter season, denoting increased sensitivity to processes causing outliers in the data, such as short-term warming, while in some areas NO is not produced because of end of the growing season and some vegetation is dormant. Note that the variability of the ozone regions is an order of magnitude larger than for the contiguous United States or Eastern or Western Domains.

Mobile Source Emissions

Mobile source gaseous emissions are, in part, temperature dependent. Modeled mobile emissions are subject to several other variables including local (usually county) level specifications of emission controls, fuels, and fleet mix, vehicle fleet behavior by road type, and specified diurnal allocations of vehicle miles traveled (VMT) activity data, Consequently, the influence of meteorology (temperature and humidity for mobile sources modeled with Mobile 6) is expected to be less important than for biogenic emissions. In particular, the averaging of temperatures in Mobile 6 affects the variability due to temperature in emissions. For this climate-oriented work a monthly averaging time was used, which has the effect of significantly smoothing the temperature variability. The Mobile 6 model runs substantially slower for shorter temperature averaging periods. A weekly or daily averaging period would accentuate the short-term variability. This is borne out in examining the average annual diurnal emission curves of NOx for the Eastern and Western Regions (Figure 11). The diurnal curves for each pollutant for each of the five years almost overlay each other. Variability from the median is approximately 2 percent. The curves for the Eastern Domain for NOx are shifted upward, reflecting the overall greater average density of mobile sources than for the Western Domain. The apparent diurnal time difference is attributable to differences in local time relative to Greenwich Mean Time (GMT). The amount of NOx emitted is dominant relative to other modeled gaseous mobile emissions, which also display little variability (Figure 12). The more limited variability for mobile source emissions is also apparent in the average seasonal diurnal emission NOx curves for the five ozone regions (approximately 15 percent among the regions) (Figure 13). The curves for $PM^{2.5}$, POA, and PEC also show the east to west regional differences (not shown), although they are not temperature dependent; and are controlled by seasonal profiles in the Mobile 6 model.

There is less difference in variability in emission fluxes among the ozone regions than between the Eastern and Western domains, again reflecting principally the amount of vehicle use and secondarily the latitudinal temperature differences with season manifested by regional meteorology. In addition, IQR increases slightly in the winter season while standard deviation and variance decrease. This is consonant with the many estimated variables in addition to meteorology that are used in modeling mobile emissions, and may reflect a greater seasonal meteorological variability.

CONCLUSIONS

An analysis of the annual and seasonal variability of modeled meteorologically-influenced average diurnal biogenic emissions for different regions within the United States using downscaled regional climate scenarios in the absence of nudging illustrates expected general diurnal and seasonal temporal patterns. However, a more detailed examination highlights distinct spatial differences. Peak emission fluxes of ISO during any season are an order of magnitude greater in the Eastern United States than in the West, and are more statistically variable in the East during all seasons, especially during the summer maxima. The variation of biogenic NO emissions demonstrated by variability statistics follows a similar regional and temporal trend, but is generally more stable less variable. This reflects the influence of agricultural land cover, precipitation, and temperature. Five ozone regions in the eastern half of the United States exhibit additional variability geographically annually and seasonally, with the greatest peak emission variance and IQR in the Mid-Atlantic (Ozone Region 3) and Southwest (Ozone Region 4). The Northeast (Ozone Region 1) had the least variable peak emissions. This pattern is consistent with persistent modeled circulation patterns. Average annual and seasonal hourly mobile gaseous emissions and variability statistics exhibit a distinct difference between the Eastern and Western United States, and to a lesser degree between ozone regions in the Eastern United States.

In addition to further statistical analyses of modeled emission and meteorology data, CIRAQ plans include further examination of the performance of the downscaled MM5 meteorology relative to observations and baseline MM5 modeled meteorology subjected to observational nudging. The results will have further implications for the meteorologically-influenced base period emissions. In addition, the temperature dependency for different regions and periods is being examined more throughly.

ACKNOWLEDGMENTS

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DISCLAIMER

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KEY WORDS

Emission modeling
Emission variability
Climate change effects
Biogenic emissions
Mobile emissions

Figure 1. CIRAQ modeling domain including Eastern and Western Regions

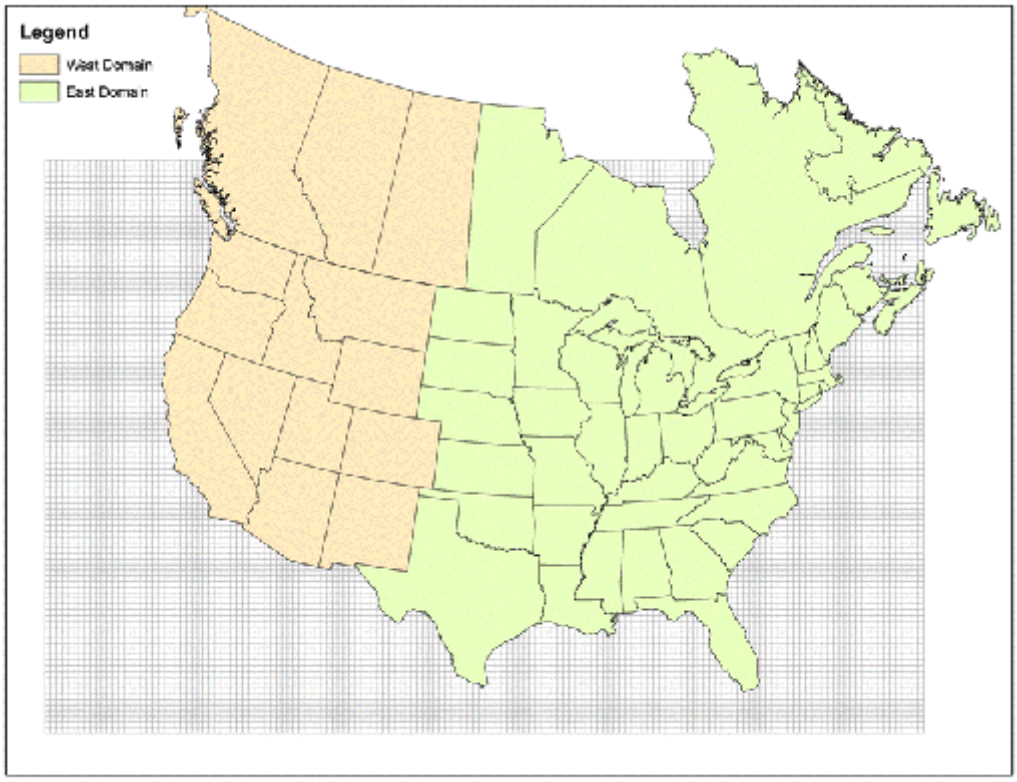

Figure 2. Ozone Regions of the Eastern United States

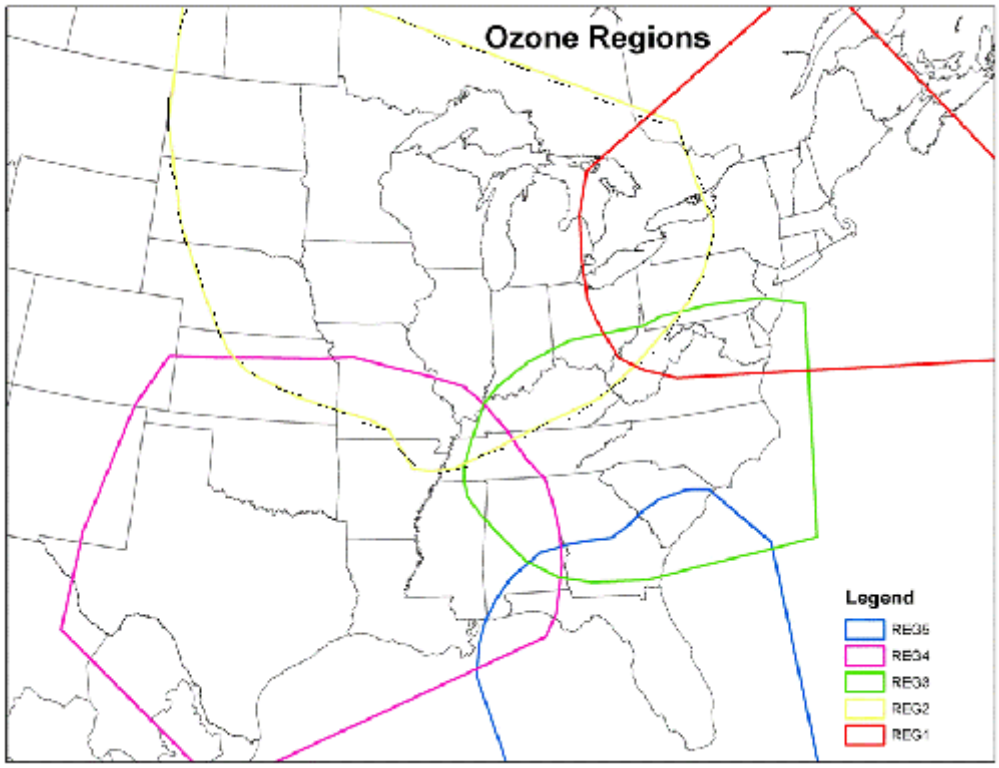

Figure 3. Average annual diurnal curves of hourly isoprene emissions (EY is Eastern Domain Year 1, WY1 is Western Domain Year 1, etc.)

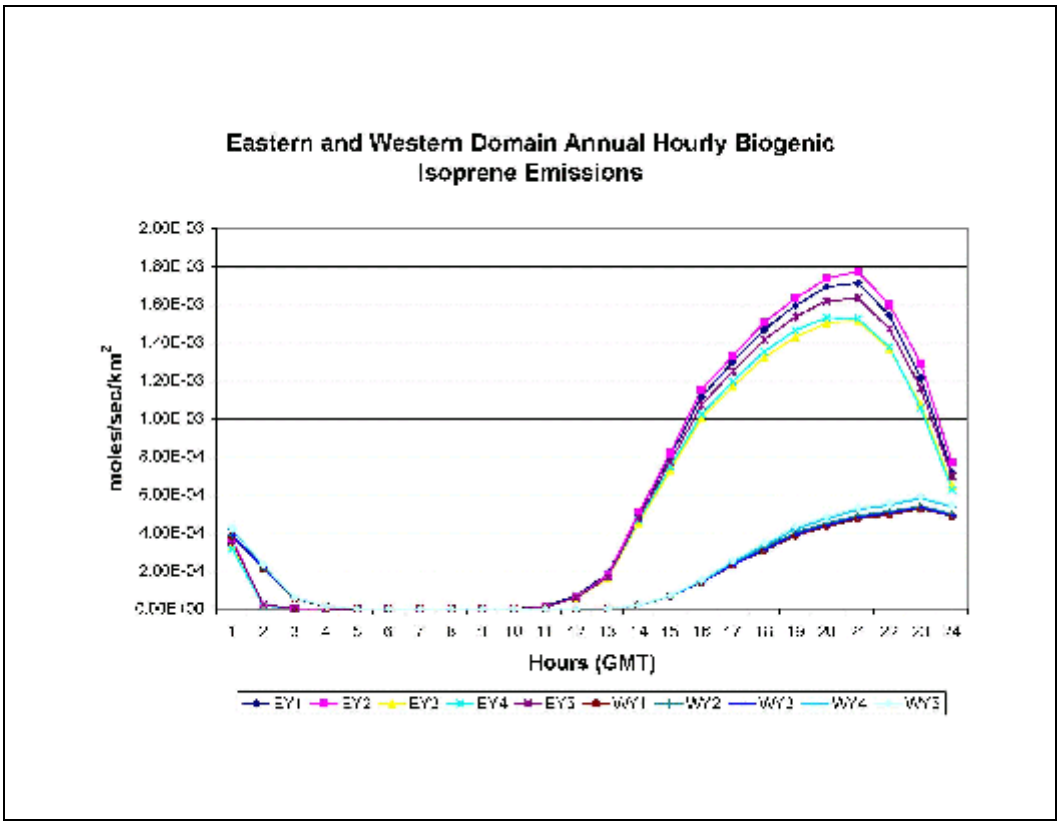

Figure 4. Example of seasonal variation of average hourly isoprene emissions (WinY1 is Winter in Year 1, SumY1 is Summer in Year 1, etc.)

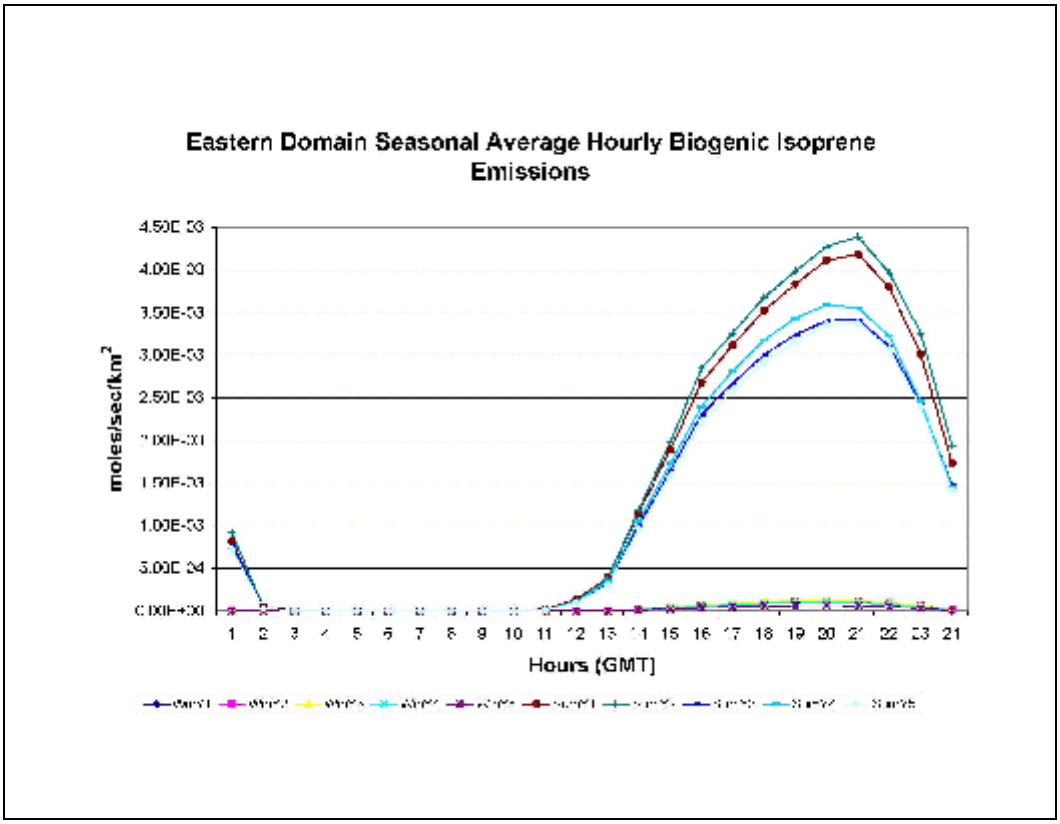

Figure 5. Inter-quartile range of gaseous emissions illustrating variability from all sources over a five-year period.

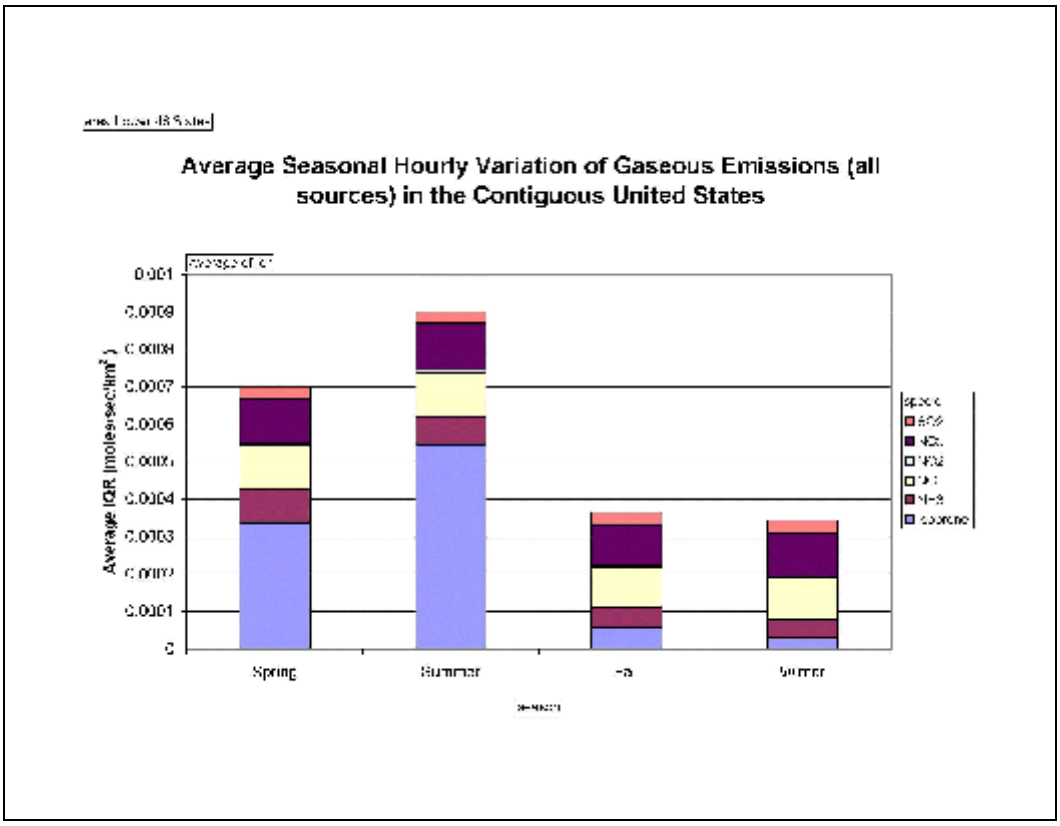

Figure 6. Average seasonal inter-quartile range of isoprene emissions for the Eastern and Western Domains of the United States.

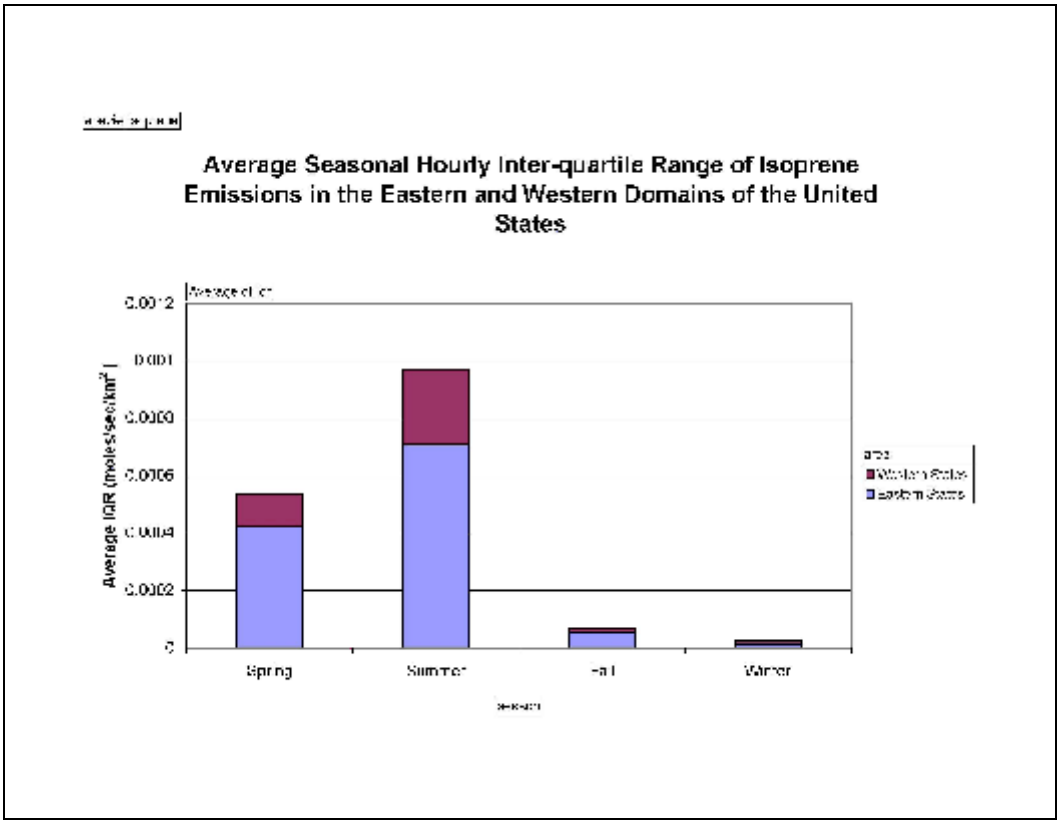

Figure 7. Average seasonal hourly inter-quartile range of isoprene emissions across the ozone regions of the Eastern United States.

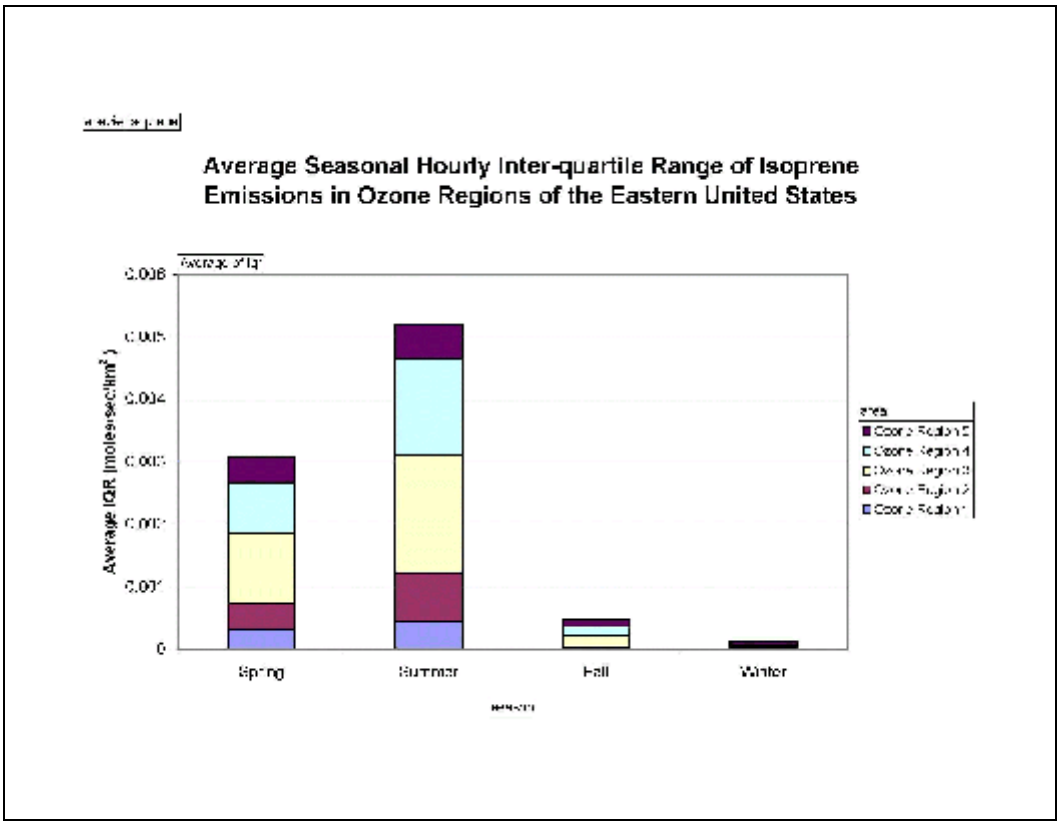

Figure 8. Average standard deviation of seasonal isoprene emissions across ozone regions of the Eastern United States.

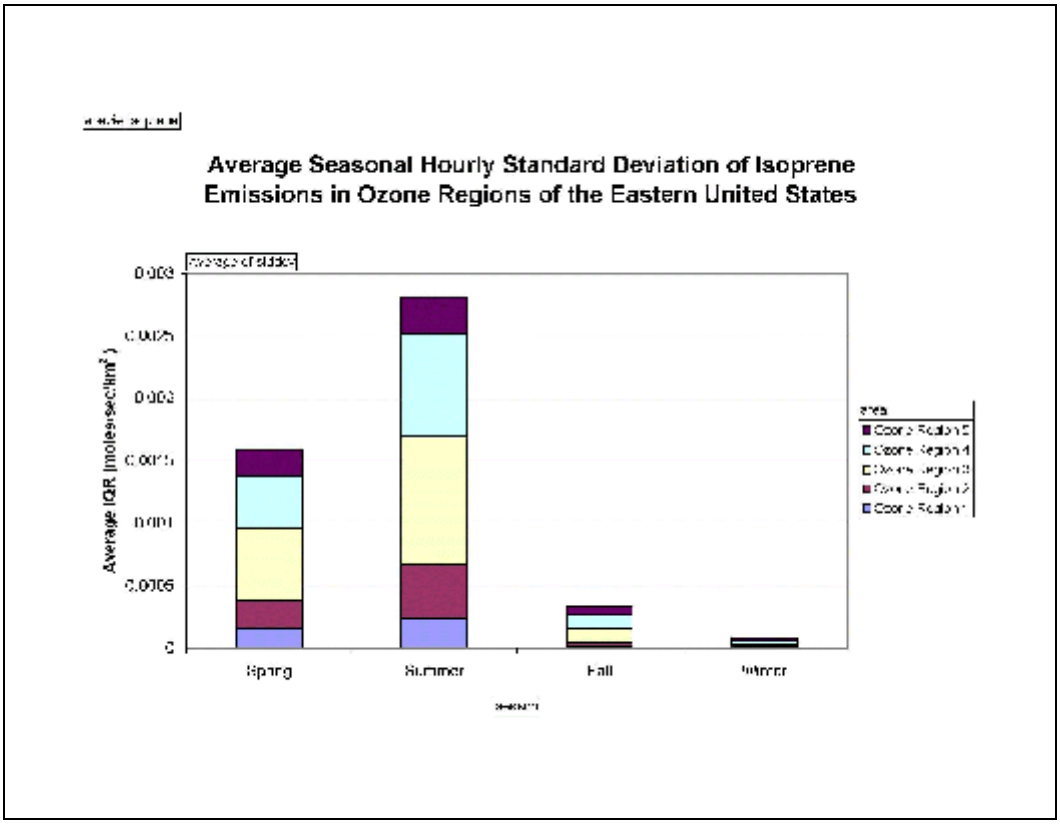

Figure 9. Dominant (43 percent) summer 700 mb temperature and wind pattern. The general circulation holds during other regimes except for the circulation center over the southern United States. This coincides with greater variability in Ozone Regions 3 and 4.

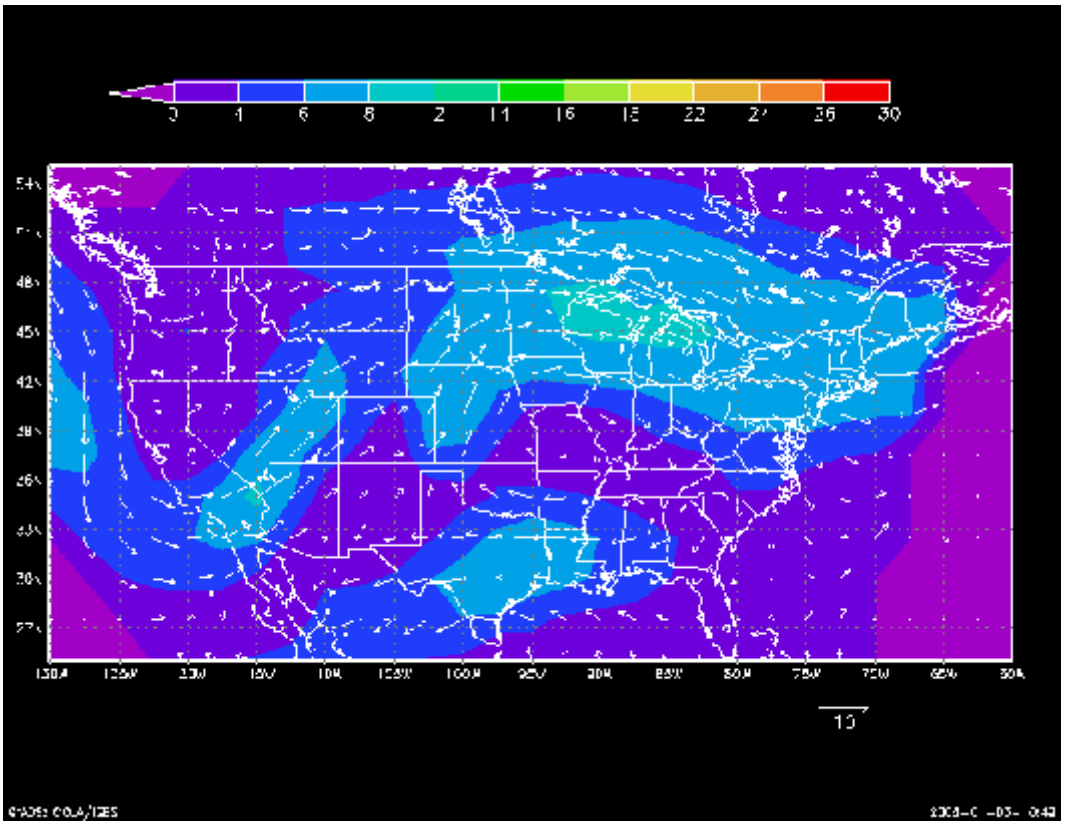

Figure 10. Illustration of variability in emission rates between the Eastern and Western Domains of the United States. (EY1 refers the first year in the Eastern Domain, WY3 is the third year in the Western Domain, etc.)

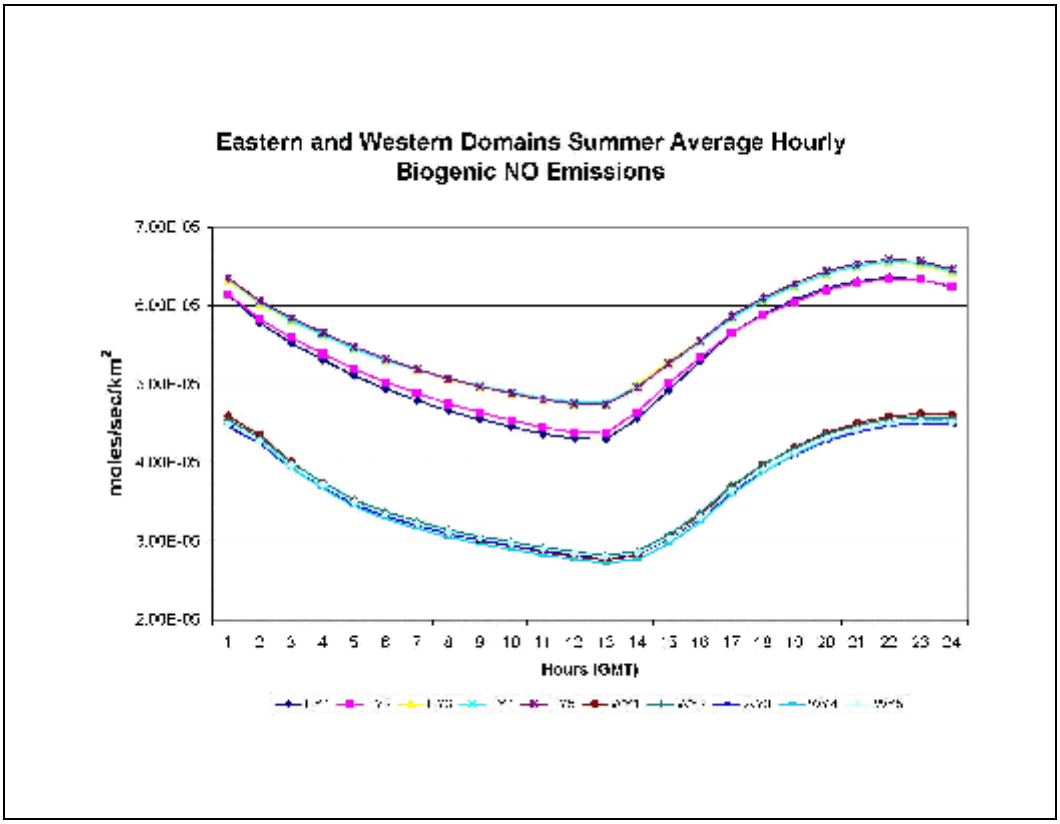

Figure 11. Examples of average annual hourly mobile source emission fluxes of NOx for the Eastern and Western Domains.

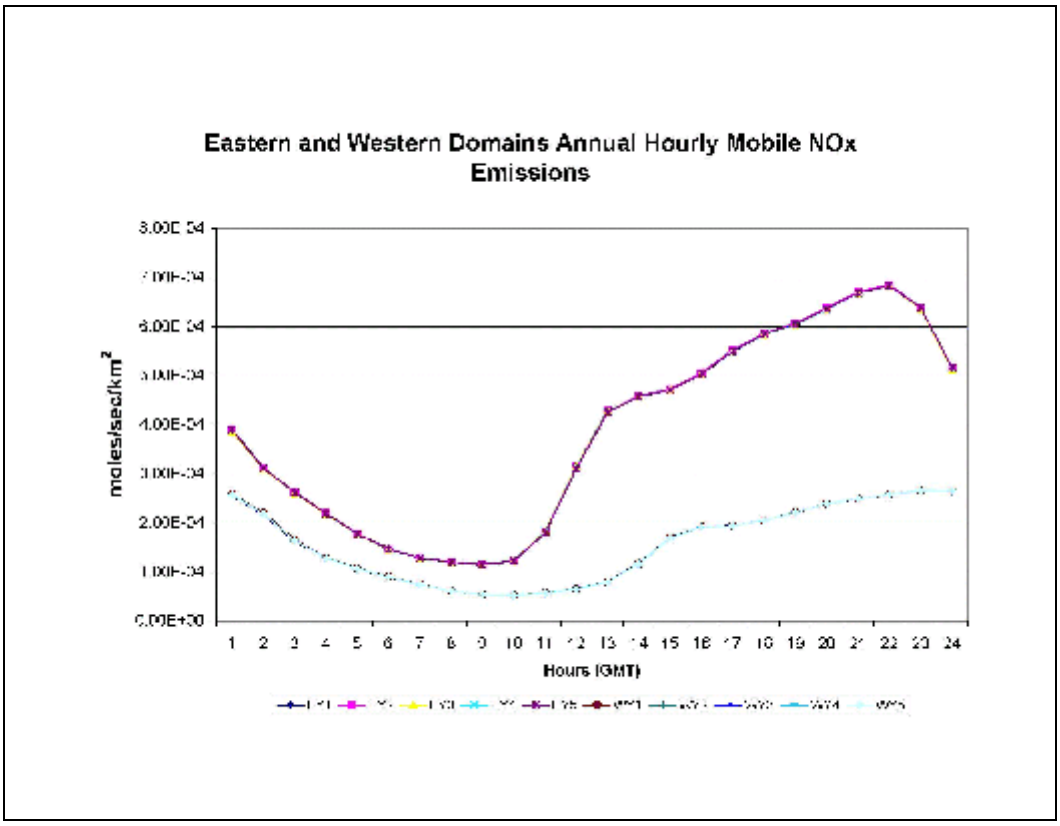

Figure 12. Example of average annual hourly emissions of mobile source gaseous emissions for two years.

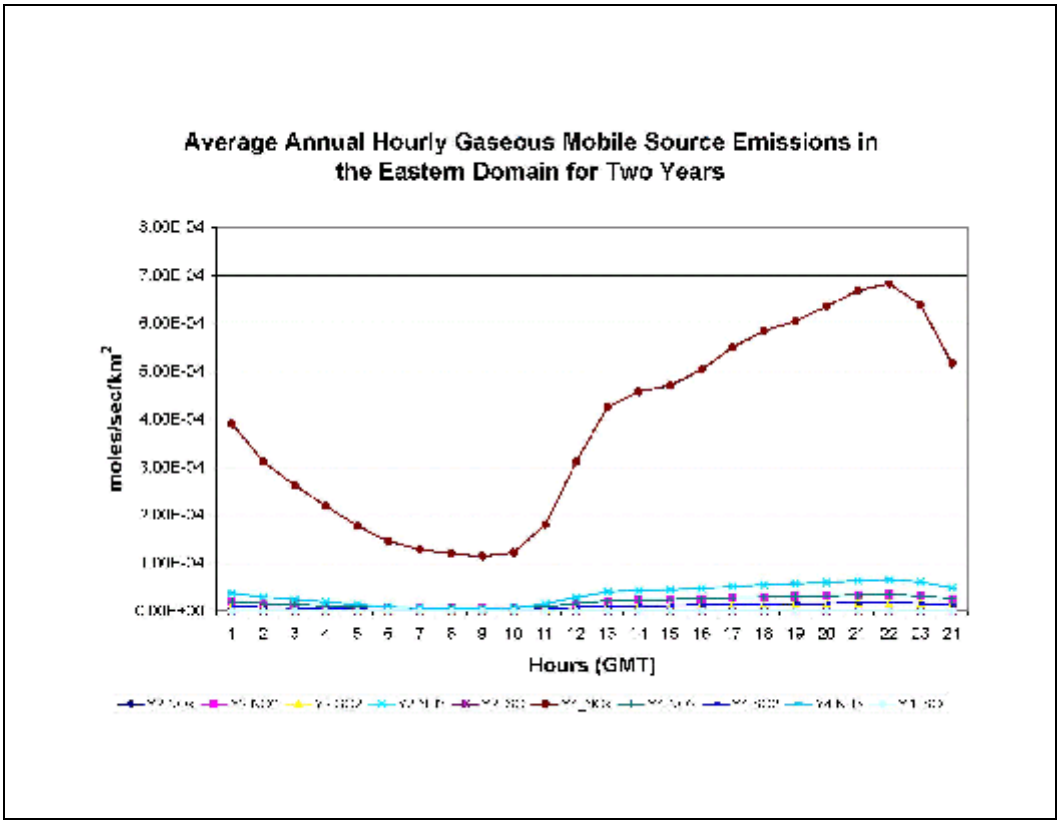

Figure 13. Example of average seasonal hourly emission curves for NOx for Ozone Regions 1 and 3 (OZ-Y1-Fall means Ozone Region 1, Year 1, fall season).

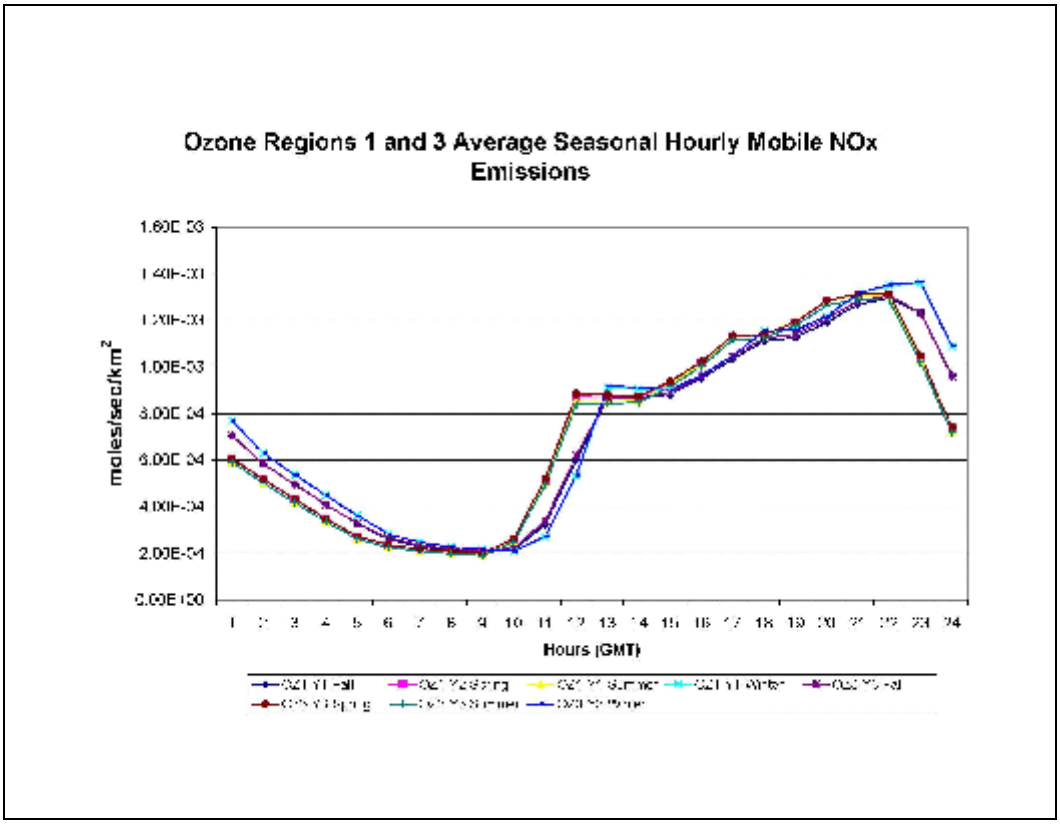

Table 1. Summary table of variability statistics of isoprene and NO emissions by season and ozone region.

Ozone Region Biogenic Emission Variability Statistics for a Five-Year Period													
		Average Spring Values			Average Summer Values			Average Fall Values			Average Winter Values		
Geographic Area	Species	Interquartile Range	Standard Deviation	Variance	Interquartile Range	Standard Deviation	Variance	Interquartile Range	Standard Deviation	Variance	Interquartile Range	Standard Deviation	Variance
Ozone Region 1	Isoprene	3.11E-04	1.58E-04	2.50E-08	4.42E-04	2.38E-04	5.67E-08	1.75E-05	1.43E-05	2.04E-10	4.41E-06	2.95E-06	8.67E-12
	NO	3.09E-04	1.49E-04	2.21E-08	3.14E-04	1.50E-04	2.25E-08	2.94E-04	1.42E-04	2.00E-08	3.05E-04	1.48E-04	2.19E-08
Ozone Region 2	Isoprene	4.24E-04	2.22E-04	4.93E-08	7.78E-04	4.26E-04	1.81E-07	2.43E-05	2.69E-05	7.26E-10	3.31E-06	2.65E-06	7.00E-12
	NO	2.83E-04	1.39E-04	1.93E-08	2.88E-04	1.42E-04	2.01E-08	2.58E-04	1.27E-04	1.62E-08	2.63E-04	1.31E-04	1.71E-08
Ozone Region 3	Isoprene	1.12E-03	5.81E-04	3.38E-07	1.89E-03	1.03E-03	1.06E-06	1.66E-04	1.17E-04	1.37E-08	2.76E-05	2.26E-05	5.09E-10
	NO	4.10E-04	2.02E-04	4.09E-08	4.09E-04	2.03E-04	4.14E-08	3.89E-04	1.90E-04	3.61E-08	4.03E-04	2.00E-04	3.99E-08
Ozone Region 4	Isoprene	8.09E-04	4.20E-04	1.77E-07	1.55E-03	8.27E-04	6.84E-07	1.48E-04	1.04E-04	1.09E-08	3.32E-05	2.36E-05	5.56E-10
	NO	2.07E-04	1.02E-04	1.05E-08	2.11E-04	1.03E-04	1.06E-08	1.89E-04	9.15E-05	8.38E-09	1.95E-04	9.54E-05	9.10E-09
Ozone Region 5	Isoprene	4.06E-04	2.07E-04	4.27E-08	5.40E-04	2.91E-04	8.49E-08	1.09E-04	6.89E-05	4.75E-09	5.22E-05	2.98E-05	8.87E-10
	NO	1.21E-04	6.01E-05	3.61E-09	1.19E-04	5.87E-05	3.44E-09	1.08E-04	5.24E-05	2.75E-09	1.10E-04	5.48E-05	3.00E-09

Table 2. Summary table of variability statistics of isoprene and NO emissions by season and Eastern and Western Domains.

United States Eastern and Western Domain Variability Statistics for a Five-Year Period													
Geographic Area	Species	Average Spring Values			Average Summer Values			Average Fall Values			Average Winter Values		
		Interquartile Range	Standard Deviation	Variance	Interquartile Range	Standard Deviation	Variance	Interquartile Range	Standard Deviation	Variance	Interquartile Range	Standard Deviation	Variance
48 United States	Isoprene	3.37E-04	1.67E-04	2.80E-08	5.47E-04	2.81E-04	7.87E-08	5.74E-05	3.65E-05	1.33E-09	2.76E-05	1.69E-05	2.86E-10
	NO	1.15E-04	5.68E-05	3.23E-09	1.17E-04	5.75E-05	3.31E-09	1.04E-04	5.24E-05	2.74E-09	1.10E-04	5.43E-05	2.95E-09
Eastern Domain	Isoprene	4.22E-04	2.19E-04	4.80E-08	7.07E-04	3.70E-04	1.37E-07	5.43E-05	3.78E-05	1.43E-09	1.50E-05	9.71E-06	9.43E-11
	NO	1.61E-04	8.00E-05	6.40E-09	1.63E-04	8.08E-05	6.54E-09	1.49E-04	7.35E-05	5.40E-09	1.54E-04	7.64E-05	5.83E-09
Western Domain	Isoprene	1.13E-04	5.80E-05	3.37E-09	2.61E-04	1.37E-04	1.87E-08	1.51E-05	1.05E-05	1.09E-10	7.50E-06	4.84E-06	2.34E-11
	NO	6.52E-05	3.17E-05	1.00E-09	6.72E-05	3.26E-05	1.06E-09	5.95E-05	2.90E-05	8.44E-10	6.09E-05	2.97E-05	8.85E-10