A Spatially and Temporally Explicit Life Cycle Inventory of Air Pollutants from Gasoline and Ethanol in the United States

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S *Supporting Information*

ABSTRACT: The environmental health impacts of transportation depend in part on where and when emissions occur during fuel production and combustion. Here we describe spatially and temporally explicit life cycle inventories (LCI) of air pollutants from gasoline, ethanol derived from corn grain, and ethanol from corn stover. Previous modeling for the U.S. by Argonne National Laboratory (GREET: Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation) suggested that life cycle emissions are generally higher for ethanol from corn grain or corn stover than for gasoline. Our results show that for ethanol, emissions are concentrated in the Midwestern "Corn Belt". We find that life cycle emissions from ethanol exhibit different temporal patterns than from gasoline, reflecting seasonal aspects of farming activities. Enhanced chemical speciation beyond current GREET model capabilities is also described. Life cycle fine particulate matter emissions are higher for ethanol from corn grain than for ethanol from corn stover; for black carbon, the reverse holds. Overall, our results add to existing state-of-the-science transportation fuel LCI by providing spatial and temporal disaggregation and enhanced chemical speciation, thereby offering greater understanding of the impacts of transportation fuels on human health and opening the door to advanced air dispersion modeling of fuel life cycles.
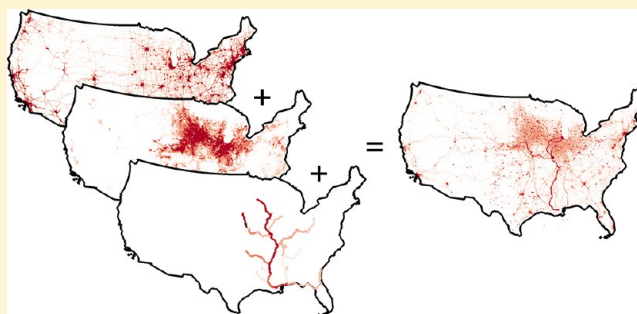
INTRODUCTION

On-road transportation accounts for approximately 20% of United States energy consumption.[1] Associated tailpipe emissions alone account for 40−60% of ground-level ozone (O_3) precursors, 6% of fine particulate matter ($PM_{2.5}$),[2] and 22% of greenhouse gases (GHGs)[3] emitted. Upstream processes involved in fuel production also contribute to overall environmental impacts. Life cycle assessment (LCA) has been used extensively to quantify the combined effects of fuel production and use,[4,5] but descriptions of where and when emissions occur are typically not reported in life cycle inventories (LCI). Such information is generally not relevant for long-lived GHGs or for fossil fuel depletion, which together have received overwhelming attention among extant LCAs of transportation fuels. For many non-GHG pollutants, knowledge of spatial and temporal aspects of emissions is critical for understanding life cycle impacts; such information has been identified as a priority for inclusion in future analyses.[6−8]

Many extant LCAs incorporate spatial and temporal information to some degree (e.g., spatially explicit treatment of a single process within the life cycle,[9−16] country, region, or state-specific impact factors,[17−27] disaggregation by urban versus rural locations,[28] county-specific information for the whole life cycle,[1,29] or spatial and temporal information for aggregated groups of processes.)[30,31] Here we add process-specific spatial and temporal information to an existing attributional life cycle inventory (LCI) so as to reveal patterns in the geographic distribution and intra-annual timing of emissions. We focus on transportation fuels in the U.S. and analyze three fuels pathways: gasoline, ethanol from corn grain, and cellulosic ethanol from corn stover. One goal of our work is to set the stage for future air quality modeling in the preparation of advanced life cycle impact assessments (LCIA). For example, our approach uses existing chemical speciation factors[32] to describe pollutant emissions by chemical group. Another goal is to explore effects of model spatial resolution on the apparent distribution between urban and rural emissions.

MATERIALS AND METHODS

The approach presented here builds on the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model, version 1.8d1, from Argonne National Laboratory.[33] GREET models the energy use and air emissions of pollutants from activities that occur during fuel production and use. GREET, as configured for this analysis, is an attributional life cycle model, meaning that it includes
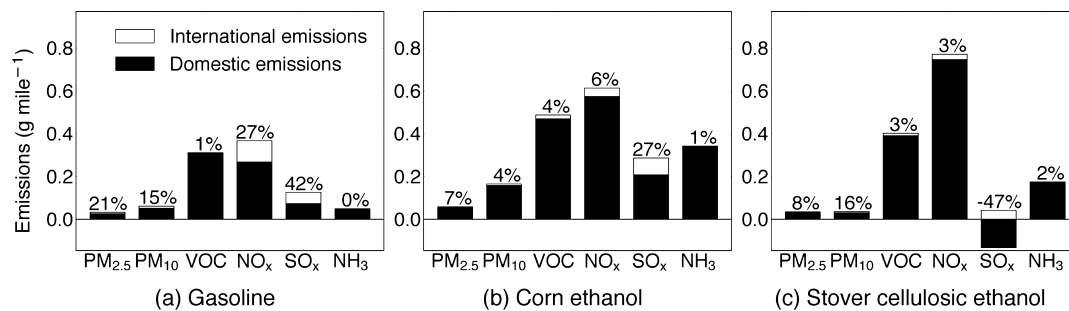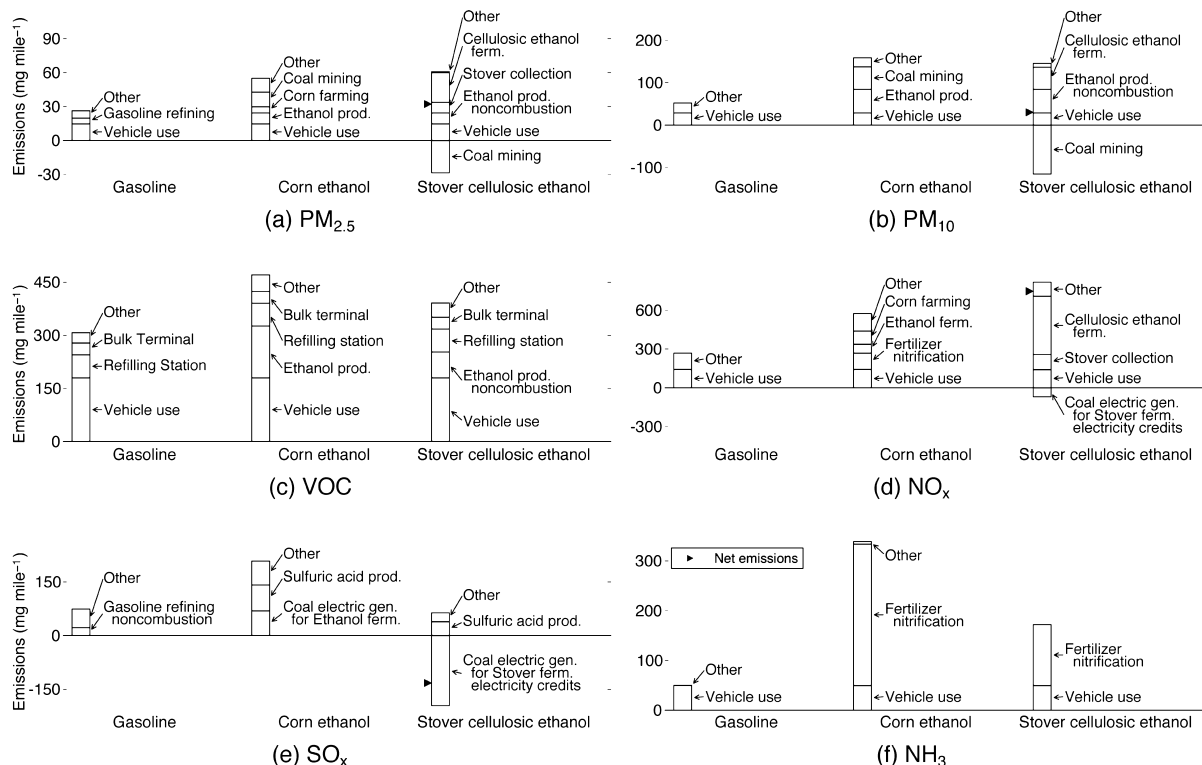
Figure 1. Amounts of emissions inside (domestic) and outside (international) the spatial modeling domain in units of grams emitted per vehicle mile traveled. Numeric labels indicate percent of life cycle emissions that are international. Our results are for domestic emissions only.
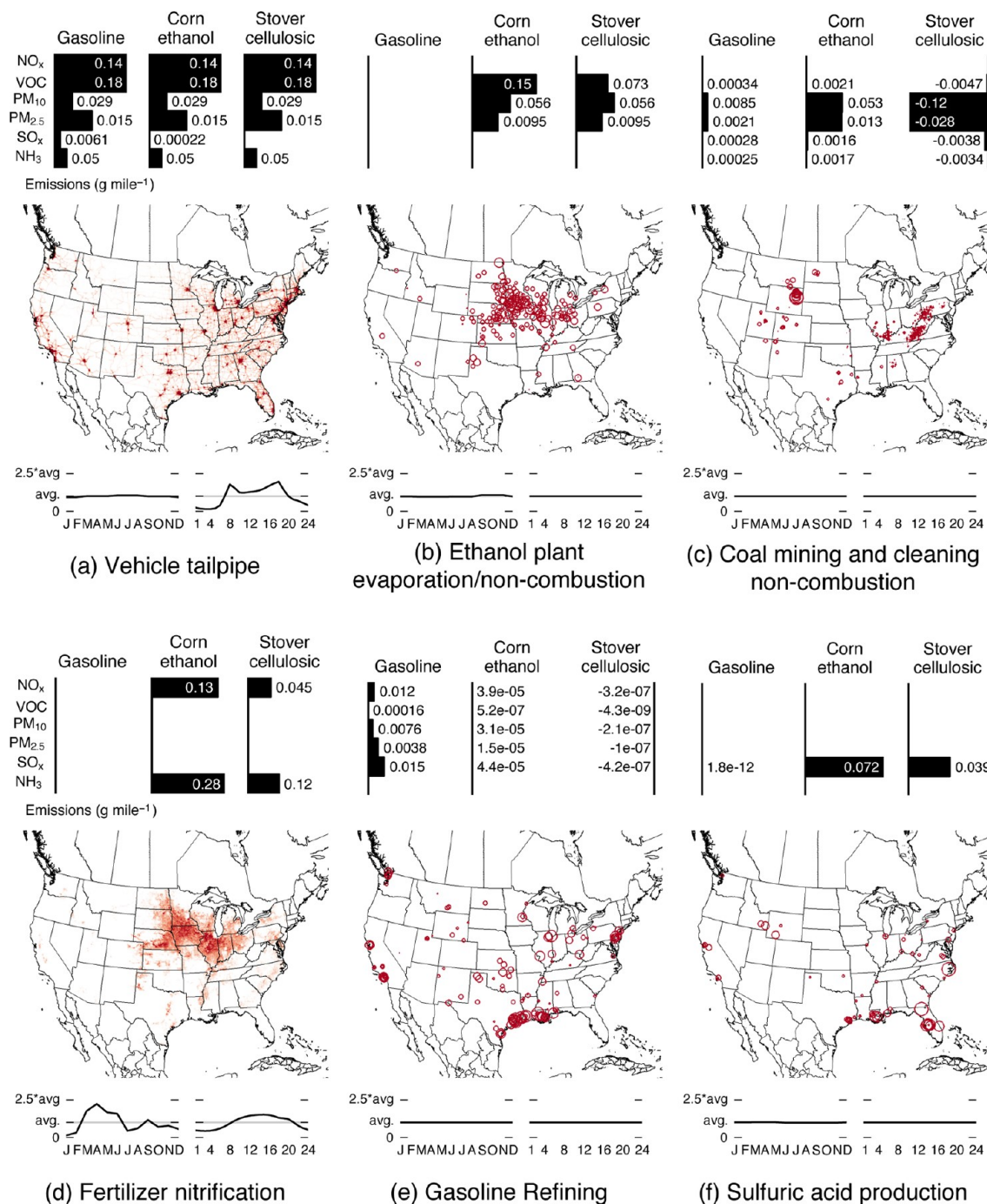

Figure 2. Fuel life cycle emissions, disaggregated by process. Plots exclude international emissions. For visual clarity, processes with emissions too small to display individually are lumped into an "Other" category; details regarding "Other" emissions can be found in SI S1. When a coproduct of a fuel production process displaces a competing product, the emissions from the life cycle of the displaced product are treated as negative emissions. For life cycles with negative emissions (e.g., stover cellulosic ethanol in plot (a)), net emissions are indicated by a small triangle on the left side of that bar. Abbreviations: trans. = transportation, ferm. = fermentation, gen. = generation, prod. = production.

emissions in fuel supply chains, but not those caused indirectly by market-mediated effects. Additional information about GREET is available elsewhere.[33]

GREET models five groups of air pollutant emissions: oxides of nitrogen (NO_x), nonmethane volatile organic compounds (VOCs), primary particulate matter less than 2.5 and 10 micrometers in diameter ($PM_{2.5}$ and PM_{10}, respectively), and sulfur oxides (SO_x). Following prior work,[29,40] we also include a sixth pollutant, ammonia (NH_3). $PM_{2.5}$ and PM_{10} inhalation can cause human mortality[34,35] and can decrease visibility. Particulate matter can be directly emitted ("primary $PM_{2.5}$") or can form in the atmosphere ("secondary $PM_{2.5}$"). VOCs can cause respiratory symptoms,[36] and NO_x can increase mortality risk.[37] VOCs and NO_x are precursors for ground-level ozone, which has also been linked to human mortality.[38,39] VOCs, ammonia, SO_x and NO_x can each contribute to secondary $PM_{2.5}$. SO_x and NO_x can cause acid rain.

The number of fuels and fuel-processes one could study is large; here, we focus on a small number of fuels that are societally relevant, widely studied in previous research, and representative of a range of fuel types. Specifically, we present emission estimates for the production and use of gasoline, ethanol from corn grain through dry milling ("corn ethanol"), and ethanol from corn stover through cellulosic fermentation ("stover cellulosic ethanol"). Stover cellulosic ethanol is not currently produced on a large scale; we assume processes for the farming and refining of stover cellulosic ethanol to be colocated with the corresponding processes for corn grain ethanol. We present results using a functional unit of a vehicle-mile traveled.

We focus on adding to an existing life cycle inventory rather than refining its existing data. Accordingly, we use default GREET settings with the following exceptions: we assume that (1) corn ethanol plants use 100% natural gas process heat; (2)

Panel (a) Vehicle tailpipe — Emissions (g mile⁻¹)

	Gasoline	Corn ethanol	Stover cellulosic
NO_x	0.14	0.14	0.14
VOC	0.18	0.18	0.18
PM_{10}	0.029	0.029	0.029
$PM_{2.5}$	0.015	0.015	0.015
SO_x	0.0061	0.00022	
NH_3	0.05	0.05	0.05

Panel (b) Ethanol plant evaporation/non-combustion

	Gasoline	Corn ethanol	Stover cellulosic
VOC		0.15	0.073
PM_{10}		0.056	0.056
$PM_{2.5}$		0.0095	0.0095

Panel (c) Coal mining and cleaning non-combustion

	Gasoline	Corn ethanol	Stover cellulosic
NO_x	0.00034	0.0021	-0.0047
VOC	0.0085	0.053	-0.12
PM_{10}	0.0021	0.013	-0.028
SO_x	0.00028	0.0016	-0.0038
NH_3	0.00025	0.0017	-0.0034

Panel (d) Fertilizer nitrification — Emissions (g mile⁻¹)

	Gasoline	Corn ethanol	Stover cellulosic
NO_x		0.13	0.045
NH_3		0.28	0.12

Panel (e) Gasoline Refining

	Gasoline	Corn ethanol	Stover cellulosic
NO_x	0.012	3.9e-05	-3.2e-07
VOC	0.00016	5.2e-07	-4.3e-09
PM_{10}	0.0076	3.1e-05	-2.1e-07
$PM_{2.5}$	0.0038	1.5e-05	-1e-07
SO_x	0.015	4.4e-05	-4.2e-07

Panel (f) Sulfuric acid production

	Gasoline	Corn ethanol	Stover cellulosic
SO_x	1.8e-12	0.072	0.039

Figure 3. An overview of the spatial and temporal disaggregation for six of the highest emitting processes in the life cycles of the fuels discussed here. (Over 400 processes are described for each life cycle; see SI S1 and S3 for processes not shown here.) The bar charts show emissions by pollutant in grams per vehicle mile traveled; each pollutant has its own bar length scale, and scales are consistent among panels. The maps show the points or areas where the process occurs, with either the grid cell color intensity or the area of the circle proportional to the fraction of total activity occurring at each location. Line plots indicate the relative amount of emissions by month of the year and hour of the weekday. For fertilizer nitrification, temporal profiles vary by location; the average values are plotted for each month. "Stover cellulosic" = stover cellulosic ethanol.

the ethanol produced is 100% ethanol without denaturant; however, because tailpipe emissions depend on blend level, we use tailpipe emission factors for E10 (i.e., a mixture of 90% gasoline, 10% ethanol) for the ethanol fuels (results for E85 [85% ethanol, 15% gasoline] can be found in the Supporting Information (SI) S1); (3) gasoline production is 100% conventional (i.e., not reformulated) gasoline; and (4) crude oil production is 100% conventional crude (most oil sands production occurs outside of our spatial modeling domain and is therefore excluded from our spatial analyses). Vehicle energy-efficiency and emissions are the same for all fuels, except SO_x emissions, which are lower for ethanol vehicles. Efficiencies and emissions factors reported here are for year 2010.

Transportation fuels used in the U.S. are delivered by a global supply chain, and their associated pollution likewise occurs worldwide. GREET includes hundreds of unit processes involved in fuel production and use, and reports life cycle emissions as global totals. Here, we focus on the fraction of life

cycle emissions that occurs in the continental U.S. and its neighboring waters. To estimate GREET emissions inside that boundary, we multiply individual GREET unit process emissions by the ratio of domestic production of their products (refs 41−43, excluding Alaska and Hawaii) to total consumption. We use the same factor to adjust upstream emissions for each process. For ocean tanker emissions, we consider emissions within the continental United States' Exclusive Economic Zone (within 200 nautical miles from shore) to be within the spatial modeling domain. We assume that ocean tankers acquire 50% of their fuel outside of the spatial modeling domain. Figure 1 shows emissions allocated within and outside of the spatial modeling domain. Most emissions are domestic. The median value in Figure 1 is 94% domestic and 6% international. SI S2 provides similar details for all processes; as in Figure 1, in all cases, most emissions are domestic. Figure 2 shows domestic emissions for select processes; further details are in SI S1.

Spatial Disaggregation. In this attributional approach, emissions are assumed to come from existing infrastructure. The fraction of emissions allocated to each facility is assumed to be equal to the fraction of total U.S. production at that facility. For example, a refinery that currently produces 5% of the gasoline in the U.S. would have 5% of gasoline production emissions allocated to it. We obtain information on the locations and average U.S. production of coal mines,[44] crude oil and natural gas extraction,[45] natural gas processing,[46] petroleum and natural gas pipelines,[2] fertilizer production,[47] sulfuric acid production,[48] pesticide production,[2] biorefineries,[49] petroleum refineries,[50] corn and soy farm locations,[51] and vehicle use,[52,53] and use it to spatially distribute the emissions by life cycle stage to a user-defined grid or within geographical polygons (e.g., states, counties, regions, etc.).[54] A 2010 calendar year industry is modeled using available data from between 2002 and 2011 (see SI S2 for more detail). To accomplish this spatial (and later temporal) assignment of emissions, GREET data and equations are rewritten as a program in the Python language to allow process-specific emissions tracking for at least 97% of emissions for each fuel. We present results for a 12-km grid resolution, with comparisons to 4- and 36-km resolutions in SI S2. References for the spatial data used for each process, and the year each data set was collected, are in SI S2. For processes that use electricity, we allocate the electrical generation emissions to generators from the U.S. EPA's eGRID database[55] located in the same North American Reliability Corporation (NERC) region as the end-use process. We allocate emissions from transportation via truck, rail, and barge using a combination of geographic network analysis[54] and linear optimization[56] techniques. The method used is described in more detail in SI S2. Pipeline and ocean tanker emissions are allocated to existing emission locations.[2] Coproducts, such as distiller's grain with solubles (DGS) production for corn ethanol and excess electricity production for stover cellulosic ethanol, are treated in GREET using a system expansion approach and are assumed to displace emissions from competing products (i.e., soy production or electricity generation). We allocate negative emissions resulting from displacement using the same methodology as for positive emissions. Each emission source is additionally classified as ground-level or elevated; elevated pollutant releases are assigned average values for U.S. electricity generating units: height, 23 m; diameter, 3 m; temperature , 456 K; exit velocity, 1.8 m s^{-1}.[2] Refer to SI S1 for the height classification of each

process. Process-specific spatial and temporal information is summarized in Figure 3 for six of the more than 400 processes; additional details are in SI S3.

Temporal Disaggregation. The time at which air pollutant emissions occur can be an important determinant of their ultimate impacts. For instance, conversion rates of VOCs and NO_x into ground-level ozone are greater during hot summer days than at other times. In contrast, emissions of $PM_{2.5}$ and its precursors may result in higher concentrations during winter than during other times because dilution rates tend to be lower in winter.[57,58] We link each of the processes shown in SI S1 with process-specific temporal profiles[2,59] to allocate emissions by month, day-of-week, and hour-of-day, with different allocations for weekdays and weekends. Figure 3 shows temporal profiles for several processes.

Chemical Disaggregation. The species categories that GREET outputs (VOCs, NO_x, SO_x, $PM_{2.5}$, and PM_{10}) are aggregates of many individual chemicals or pollutant types. Such broad categories can limit the accuracy of an impact analysis. For instance, the combustion of gasoline and ethanol both produce VOCs, but they produce different types of VOCs, with varying toxicity, reactivity, and ozone-production potential. The combustion of gasoline and ethanol both produce $PM_{2.5}$ and its precursors, but certain types of $PM_{2.5}$ cause atmospheric cooling (e.g., sulfate aerosols) while others cause atmospheric warming (e.g., black carbon aerosols).[60] To develop the inventory presented here, we link each of the processes shown in SI S1 with chemical speciation profiles[32] to disaggregate the emissions into 34 chemical species groups according to the Carbon Bond 2005 (CB05[61]) chemical mechanism. CB05 speciates emissions into the following groups:

- **VOCs**: acetaldehyde, higher aldehydes, benzene, methane, ethene, ethane, ethanol, formaldehyde, internal olefins, isoprene, methanol, olefins, paraffins, sesquiterpenes, terpenes, toluene, xylene, nonreactive VOCs, nonvolatile VOCs, and unknown/other VOCs
- **$PM_{2.5}$**: black carbon, nitrate particulates, organic particulates, sulfate particulates, and unclassified $PM_{2.5}$
- **NO_x**: NO and NO_2
- **SO_x**: SO_2 and $SO_{4,g}$.

PM_{10} and NH_3 are included without speciation.

■ RESULTS

Figure 1 shows fractions of emissions occurring outside of the spatial modeling domain for each fuel. (These and all other results show emissions of air pollutants, which may or may not be directly correlated with changes in ambient pollution levels.) As mentioned above, emissions that occur outside of the spatial modeling domain (contiguous U.S. and surrounding waters), are excluded from results below. Excluding international emissions has the largest effect on the gasoline life cycle. This result is because we model 96% of emissions from transportation of crude oil by ocean tanker as occurring outside the spatial modeling domain. International tanker emissions account for 34% of SO_x, 16% of NO_x, 14% of $PM_{2.5}$, and 8% of PM_{10} gasoline life cycle emissions. However, these ocean tanker emissions occur over the open ocean where human exposure is very low,[26] so the exclusion of these emissions will likely not noticeably affect estimates of total human exposure. Other notable spatial modeling domain exclusions include 28% of sulfuric acid production for fertilizer,[62] accounting for 12% of
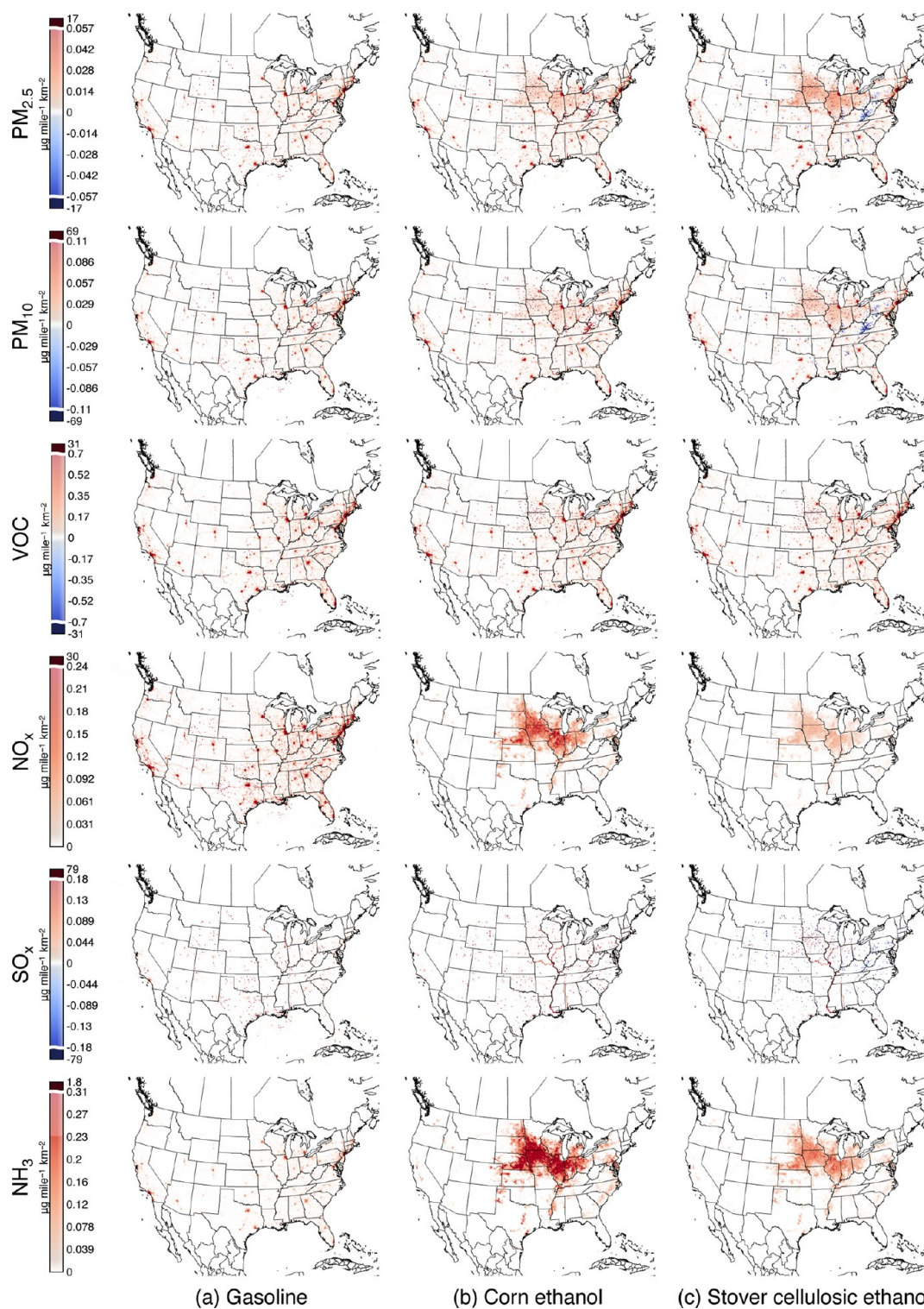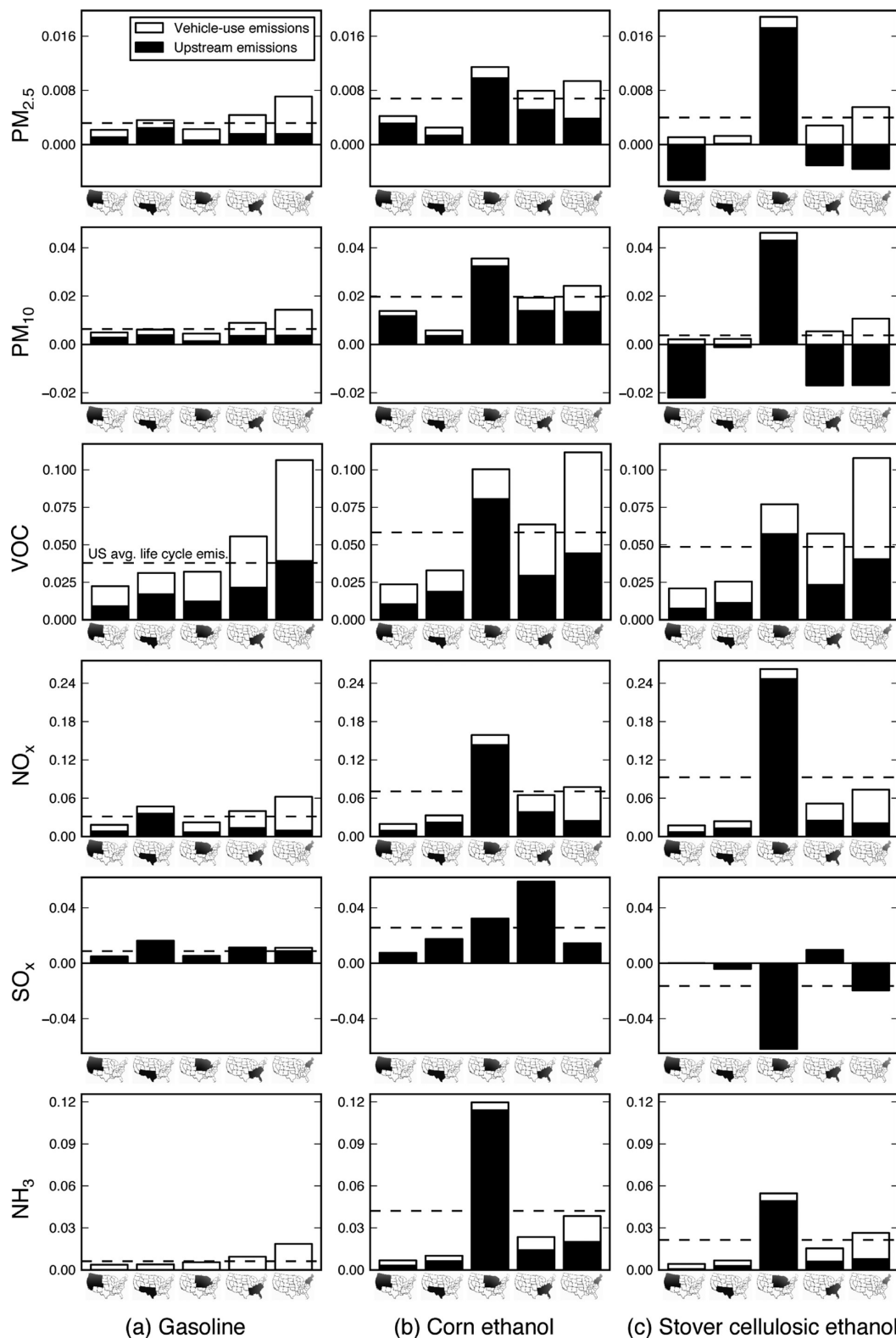
Figure 4. Annual total life cycle emissions for three fuels. For ease of viewing, each linear color scale contains a discontinuity at the 99th percentile of emissions.

corn ethanol and 20% of stover cellulosic ethanol SO_x life cycle emissions, and 56% of crude oil extraction,[63] accounting for 6% of NO_x emissions from the gasoline life cycle. All other processes either occur completely within the modeling domain or comprise a negligible fraction of the life cycle total emissions.

Figure 4 shows 12 km resolution gridded emissions for the gasoline, corn ethanol, and stover cellulosic ethanol fuels. In general, gasoline emissions tend to be correlated with vehicle use and so are distributed in or near urban centers. Ethanol emissions tend also to be correlated with ethanol production and so are concentrated in the Midwest "Corn Belt". The area along the Kentucky/Virginia border extending into West Virginia experiences a reduction in $PM_{2.5}$ emissions (shown in blue in Figure 4) owing to reduced coal mining activity caused by excess electricity generation in biorefineries. Excess

Figure 5. Contributions of U.S. regions to total life cycle emissions for three fuels (micrograms emitted per vehicle-mile traveled per square kilometer land area). Dashed lines show U.S. average emissions. Refer to SI S2 for a version of this figure with units of g mi^{-1}. Key: Northeast ▧, Midwest ▧, Southeast ▧, Southwest ▧, West ▧.

electricity is sold to the electrical grid and assumed to offset electricity produced elsewhere.

Figure 5 shows emissions contributions by region of the U.S. For all three fuels, the greatest intensity of emissions per land area occurs in the Northeast for VOCs, owing to the large portion of total vehicle miles traveled per land area occurring there. See SI S2 for the fraction of vehicle miles traveled in each region. The Midwest receives a large amount of emissions for both ethanol fuels, owing to ethanol fermentation plants and ammonia emissions from fertilizer nitrification. For the

Midwest, emissions are lower for gasoline than for ethanol, with the exception that SO_x emissions are negative (i.e., reduced) for stover cellulosic ethanol owing to excess electricity generation at fermentation plants. SO_x emissions in the Southeast for corn ethanol are mainly attributable to Florida-based sulfuric acid production for phosphate fertilizer. The Southwest and West regions generally do not receive large proportions of pollutant emissions for any of the three fuels (exception: SO_x emissions for gasoline).

Emissions are spatially disaggregated by allocating them to cells in a raster grid. As has been previously noted,[15] we observe that the resolution of the grid used can influence the apparent locations of emission. SI Figures S2-1 and S2-2 show the dependence of emissions allocated to urban areas on the spatial scale of the grid used for allocation and compares the results to the urban emissions given by GREET.[28] Overall, increasing grid resolution leads to an increased allocation of emissions to urban areas, suggesting that a coarse grid tends to artificially dilute urban emissions to the surrounding rural areas. See S2 for further discussion.

Figure 6 shows temporal profiles for fuels and pollutants discussed here. Pollutant emissions from the gasoline life cycle do not vary appreciably by month. For the corn ethanol life cycle, however, there is a spike in NH_3 and NO_x emissions in the spring, corresponding to fertilizer application. This pattern is also true to a lesser extent for stover cellulosic ethanol. All fuels show a slight decrease in emissions on weekends. Weekday emissions commonly show a bimodal distribution around the morning and evening rush hours owing to increased vehicle tailpipe emissions at those times. For fuels and pollutants where farming activities are a major contributor, however, the weekday and weekend emissions are unimodally distributed around the daylight farming hours.

Our chemical speciation reveals the following. Although stover ethanol emits the lowest total amount of $PM_{2.5}$ of all three fuels, it emits more black carbon (15 mg mi^{-1}), a species of fine particulate matter that contributes to atmospheric warming, than either of the other two fuels (gasoline: 4.5 mg mi^{-1}, corn ethanol: 10). Stover cellulosic ethanol also emits the lowest amount of sulfate aerosols (-1.3 mg mi^{-1}; gasoline: 1.9, corn ethanol: 3.8), which cause atmospheric cooling. Emissions of ethanol are 30 000−40 000 times higher for the ethanol fuels than for gasoline; ethanol in the atmosphere may be oxidized to form acetaldehyde (a carcinogen). However, emissions of benzene (another carcinogen[64]) are higher for gasoline than for the ethanol fuels (the relative amounts by fuel depend on the ethanol feedstock and the blend level of the final fuel). Full results for chemical speciation, including for 85% ethanol blends (E85), are in SI S1.

■ DISCUSSION

We have presented a spatially and temporally explicit life cycle inventory for transportation fuels. Prior life cycle inventories were typically presented at global, national, or regional levels, which is sufficient for understanding global processes such as climate change and fossil fuel depletion, but is insufficient for the analysis of local processes such as air pollution. The spatially and temporally explicit LCI presented here not only provides the level of detail necessary to perform detailed LCIA of air pollutant emissions, it also gives information on spatial and temporal trends that can be useful in policy making and regulation. For instance, in the U.S., implementation plans for coming into compliance with air quality standards are generally determined by individual states. Spatially and temporally explicit LCA can help state-level policy makers identify potential sources of air pollutant emissions within their jurisdictions and create appropriate regulations. Such information is not delivered by conventional LCA approaches.

The framework presented here outputs gridded, time-resolved emissions files, which is an important step toward photochemical dispersion modeling. For example, U.S. production and consumption of stover cellulosic ethanol would increase emissions of NO_x, NH_3, and $PM_{2.5}$ in the Midwest, but would decrease SO_x emissions in the same region. All four of these species influence ambient concentrations of $PM_{2.5}$, it is unclear a priori whether the net change in $PM_{2.5}$ concentrations will be positive or negative in this region. Photochemical dispersion modeling can help answer this question.

Our choice of spatial modeling domain (continental U.S.) by definition restricts our study. However, we do not see this aspect as a major limitation because most emissions occur within our boundary, and because most of the excluded emissions occur over the open ocean, for which human exposures are much lower than for emissions on land.[26] This same domain has been used in prior regulatory analysis.[65] At present, lack of computational power and accurate input data

Legend:
- · Gasoline
― Corn ethanol
··· Stover cellulosic ethanol
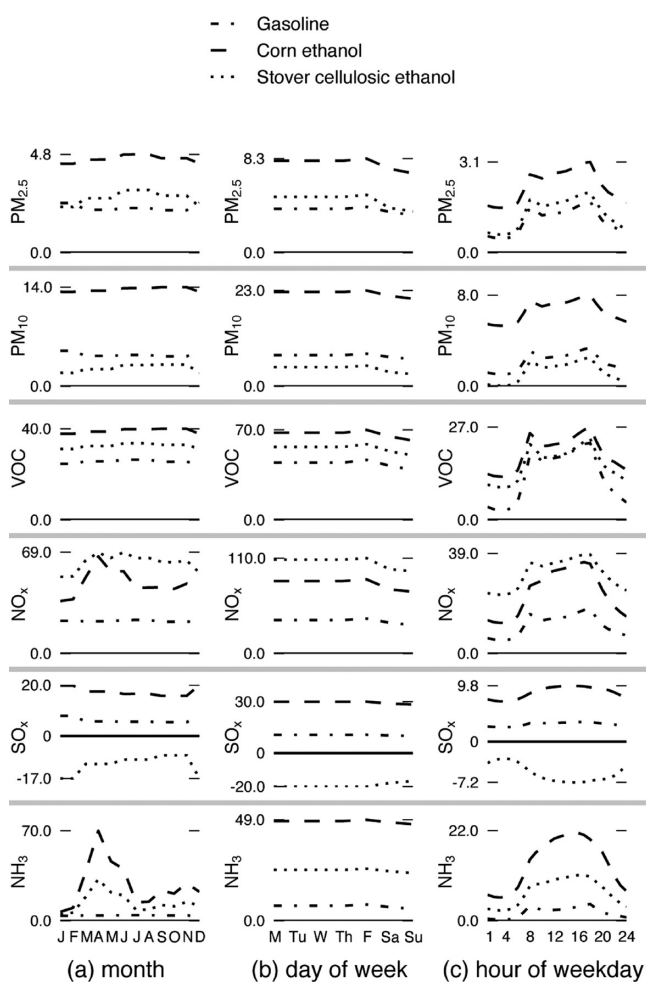
Figure 6. Temporal profiles of life cycle emissions (mg emitted per vehicle-mile traveled) by month of year, day of week, and hour of day (weekday). Results by hour of day (weekend) are in SI S2.

make it impractical to extend the system boundaries to the entire world. For future work, a nested approach (detailed spatial treatment of emissions within the modeling domain; coarser resolution outside of the domain) could prove useful. A further limitation of this study is that owing to limited data availability, the spatial data used here represent a range of years (2002–2011; see SI S2 for details). Finally, uncertainty and variability in the spatial, temporal, and chemical speciation data used here contribute to the overall uncertainty in the life cycle inventory, but this information is generally unknown or unreported.

Our work considers supply chain emissions of existing production. GREET, and by extension the framework presented here, performs an exclusively attributional (static) LCA for non-GHG air pollutants. We do not include indirect (market-mediated) effects. An example of an indirect effect would be if corn ethanol production in the U.S. increases global grain prices, causing emissions from the burning of tropical rainforest to grow crops. A consequential (dynamic) LCA, which we do not do, would aim to capture indirect effects. We assume here that all processes occur at existing production locations; in reality, new production may cause new facilities to open, or existing facilities to close or change locations. We assume that cellulosic biorefineries for stover cellulosic ethanol are colocated with existing corn ethanol biorefineries; in reality, the difficulty of transporting corn stover may cause biorefineries to be smaller and closer to cornfields.

We have focused here on the air pollutant implications of the choice between ethanol and gasoline as a transportation fuel. In general, methods presented here can provide insight into any spatially or temporally inhomogeneous environmental impact categories, such as water quality and availability, soil properties, or wildlife habitats. They can also be expanded to study specific processes that affect those impact categories, such as agriculture and food production, building construction, or electricity generation; the possible applications are only limited by the availability of data.

■ ASSOCIATED CONTENT

ⓢ Supporting Information

Process-specific, chemically speciated, and state and urban area specific emissions data (S1a–c, Excel format), supplemental text and figures, and process-specific maps (S2 and S3a–c, pdf format) for gasoline, corn ethanol, and stover cellulosic ethanol. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

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Supplemental Information 2

A spatially and temporally explicit life cycle inventory of air pollutants from gasoline and ethanol in the United States

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1 Transportation emissions allocation

To allocate emissions from transportation, we first calculated the shortest routes along major road, rail, and waterway networks [1] between all combinations of source and destination locations having a viable route between them over the network for each product transported. For computational expediency, in some cases we only calculated routes between source and receptor pairs with substantial contributions to total production. We then optimized the amount of product transported along each route [2] to minimize total product-distance traveled. Finally, we weighed the fraction of product transported along each route by the route distance to determine the fraction of emissions allocated to each route.

2 Urban vs. rural emissions

Because emissions are spatially disaggregated by allocating them to cells in a raster grid, the grid resolution can influence the fraction of emissions in urban areas. Figures S3-1 and 2 show emissions allocated to urbanized areas as defined by the 2000 US Census [3] using three different grid resolutions for the GREET output pollutants plus ammonia, and compare them to the urban emissions output by the GREET spreadsheet [4]. This dependence of results on spatial scale has also been noted elsewhere [5]. In general, gasoline emissions are more concentrated in urban areas. Most (80%) petroleum refinery production, but only 10% of current biorefinery production, is located in urban areas. Overall, increasing grid resolution leads to an increased allocation of emissions to urban areas, suggesting that a coarse grid tends to artificially dilute urban emissions to the surrounding rural areas.

The urban emissions fractions for the 4km grid generally agree with those reported by Huo *et al.* [4] (as incorporated in GREET spreadsheet) with the exception of SO_x emissions from the two ethanol fuels. A large portion of the SO_x emissions from these fuels come from sulfuric acid production for phosphate fertilizer. Huo *et al.* assume 0% of sulfuric acid production occurs in urban areas, whereas we use data that allocates 52% of sulfuric acid production to urban areas [6], specifically the Tampa, Florida metropolitan area, which is near the United States' largest phosphoric rock mine.
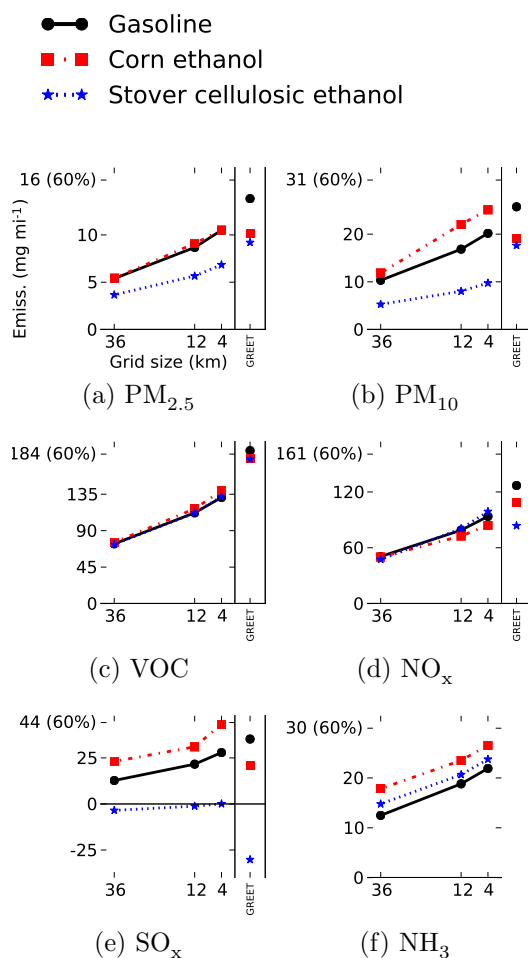
Figure S2-1: Life cycle emissions allocated to urbanized areas as calculated using three different grid resolutions (left section of each plot) and urban emissions calculated using the default GREET spreadsheet (right section of each plot). To allow comparison among pollutants, all plots are scaled to 60% of the gasoline total emissions. In general, more precise estimates (smaller grid sizes) suggest a greater proportion of emissions are urban. For gridded calculations (left sections), emissions occurring outside the spatial system boundary are excluded. Calculations using the GREET spreadsheet (right sections) include all emissions. The GREET model does not calculate ammonia emissions. International emissions are excluded. See Figure S2-2 for a version of this figure calculated without first excluding international emissions.
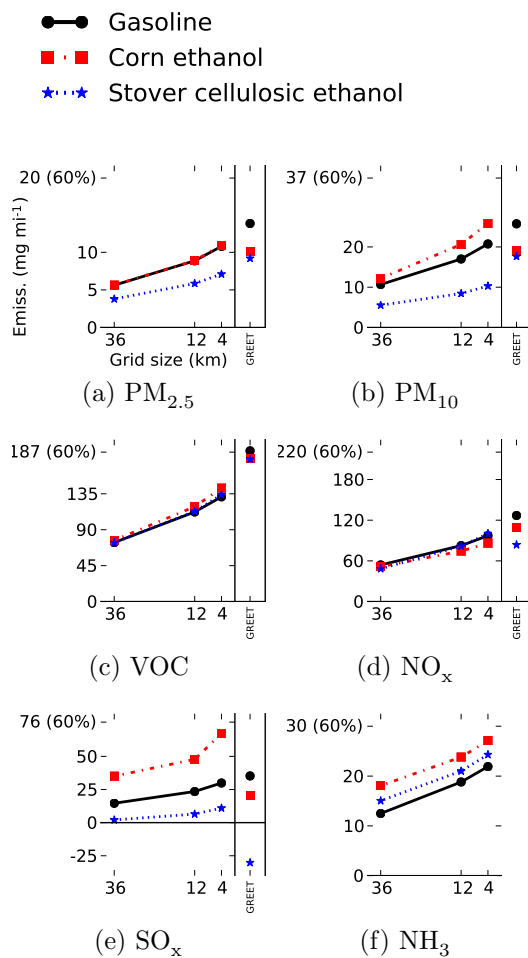
Figure S2-2: Life cycle emissions allocated to urbanized areas as calculated using three different grid resolutions (left section of each plot) and urban emissions calculated using default GREET spreadsheet (right section of each plot). To allow comparison between pollutants, all plots are scaled to 60% of the gasoline total emissions. Calculations include international emissions fractions. The GREET model does not calculate ammonia emissions.

3 Additional tables and figures

Table S2-1: Fractions of processes occurring within the spatial modeling domain. Processes that are not listed are assumed to occur 100% within the spatial modeling domain. Emissions occurring outside of the modeling domain are excluded from the analysis by multiplying emissions from the processes below, as well as the emissions upstream of the process, by the corresponding fraction.

Process	Domestic fraction	Data source [a] [b] [c]
Natural Gas	77.9%	EIA – adjusted to exclude Alaska production
Sulfuric Acid	72.0%	USGS: Sulfuric acid
Nitrogen–all	56.5%	USGS: Nitrogen (ammonia)
Phosphoric Rock and acid	85.0%	USGS: Phosphate Rock
Potash	17.0%	USGS: Potash
CaCO3	98.0%	USGS: Lime
Pesticides–all	90.2%	EPA Market Estimate, Year 2001
Coal	100.0%	EIA
Crude Extraction	43.6%	EIA – adjusted to exclude Alaska production
Gasoline Refining	100.0%	EIA
Diesel Refining	100.0%	EIA
Residual Oil Refining	100.0%	EIA

[a]EIA: US Energy Information Administration. Independent Statistics & Analysis. Available at: http://www.eia.gov [Accessed July 3, 2012]

[b]USGS: US Geological Survey. Mineral Commodity Summaries. 2010. Available at: http://minerals.usgs.gov/minerals/pubs/mcs/ [Accessed July 3, 2012]

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Table S2-2: Full names of chemical species found in SI1a-c [7]

Abbreviation	Name
ALD2	Acetaldehyde
ALDX	Higher aldehydes
BENZENE	Benzene
CO	Carbon Monoxide
ETH	Ethene
ETHA	Ethane
ETOH	Ethanol
FORM	Formaldehyde
HONO	Nitrous acid
IOLE	Internal olefins
ISOP	Isoprene
MEOH	Methanol
NH3	Ammonia
NO	Nitrogen monoxide
NO2	Nitrogen dioxide
NVOL	Nonvolatile organic chemicals
OLE	Olefins
PAR	Paraffins
PEC	Elemental carbon
PM10	Particulate matter less than 10 micrometers in diameter
PMFINE	Particulate matter less than 2.5 micrometers in diameter; unclassified
PNO3	Nitrate particulates less than 2.5 micrometers in diameter
POC	Organic particulates less than 2.5 micrometers in diameter
PSO4	Sulfate particulates less than 2.5 micrometers in diameter
SO2	Gaseous sulfur dioxide
SULF	Gaseous sulfate
TERP	Terpenes
TOL	Toluene
UNK	Unknown VOCs
UNR	Unreactive VOCs
XYL	Xylene

Table S2-3: Sources of spatial data and the year the data was collected. The "Map name" column refers to the column of the same name in Supporting Information *S1*, which cross-references each unit process in the life cycle to one of the spatial data sources described below.

Map name	Data year	Source
Ammonia, AmmoniumNitrate, K2O, NitricAcid, PhosphoricAcid, PhosphateRock, Urea	2011	International Fertilizer Development Center (IFDC). *North America Fertilizer Capacity*. 2011. Available at: `http://www.ifdc.org/getdoc/5fee0591-47b9-4226-9338-cf43a980e15a/Fertilizer_Market-Related_Reports_(1)`.
CoalMining	2007	US EIA. *Coal Production by MSHA ID, Mine Operation, Union Status, and Average Number of Employees and Hours*. 2007. Available at: `http://www.eia.gov/cneaf/coal/page/database.html`.
CurrentBiorefineries	2010	Renewable Fuels Association. *Biorefinery Locations*. 2010. Available at: `http://www.ethanolrfa.org/bio-refinery-locations/` [Accessed August 2, 2010].
ElectricGeneration (all types)	2007	US EPA. *eGRID2007 Version 1.1*. Available at: `http://www.epa.gov/cleanenergy/energy-resources/egrid/index.html`.
FarmAreaCornTotal, FarmProduction-CornTotal	2010	US Department of Agriculture. *National Agricultural Statistics Service*. 2010. Available at: `http://www.nass.usda.gov/Data_and_Statistics/index.asp` [Accessed January 5, 2011]. *and* Johnson DM, Mueller R. The 2009 *Cropland Data Layer*. 2010. Available at: `http://www.nass.usda.gov/research/Cropland/SARS1a.htm`.
GasPipelines, LNG, OceanTankers, OilPipelines, Pesticides	2005	US Environmental Protection Agency. *2005 National Emissions Inventory (NEI)*. 2009. Available at: `http://www.epa.gov/ttn/chief/emch/index.html` [Accessed March 7, 2012].
NaturalGasExtraction, OilExtraction	2009	US Energy Information Administration. *Top 100 Oil and Gas fields of 2009*. 2010. Available at: `http://www.eia.gov/pub/oil_gas/natural_gas/data_publications/crude_oil_natural_gas_reserves/current/pdf/top100fields.pdf`.
NGProcessing	2004	US Energy Information Administration. *Natural Gas Processing: The Crucial Link Between Natural Gas Production and Its Transportation to Market*. 2006. Available at: `http://www.arcticgas.gov/sites/default/files/documents/2006-eia-ng-processing.pdf`.
OilRefineries	2009	US Energy Information Administration. *Ranking of U.S. Refineries*. 2009. Available at: `http://www.eia.gov/neic/rankings/refineries.htm` [Accessed March 3, 2010].
SulfuricAcid	2005	ICIS Chemical Market Intelligence. Chemical Profile: Sulfuric Acid. *Chemical Business*. 2005. Available at: `http://www.icis.com/`.
VMT	2002	US Environmental Protection Agency. *EPAs National Inventory Model (NMIM), A Consolidated Emissions Modeling System for MOBILE6 and NONROAD*. 2005. Available at: `www.epa.gov/otaq/models/nmim/420r05024.pdf`.
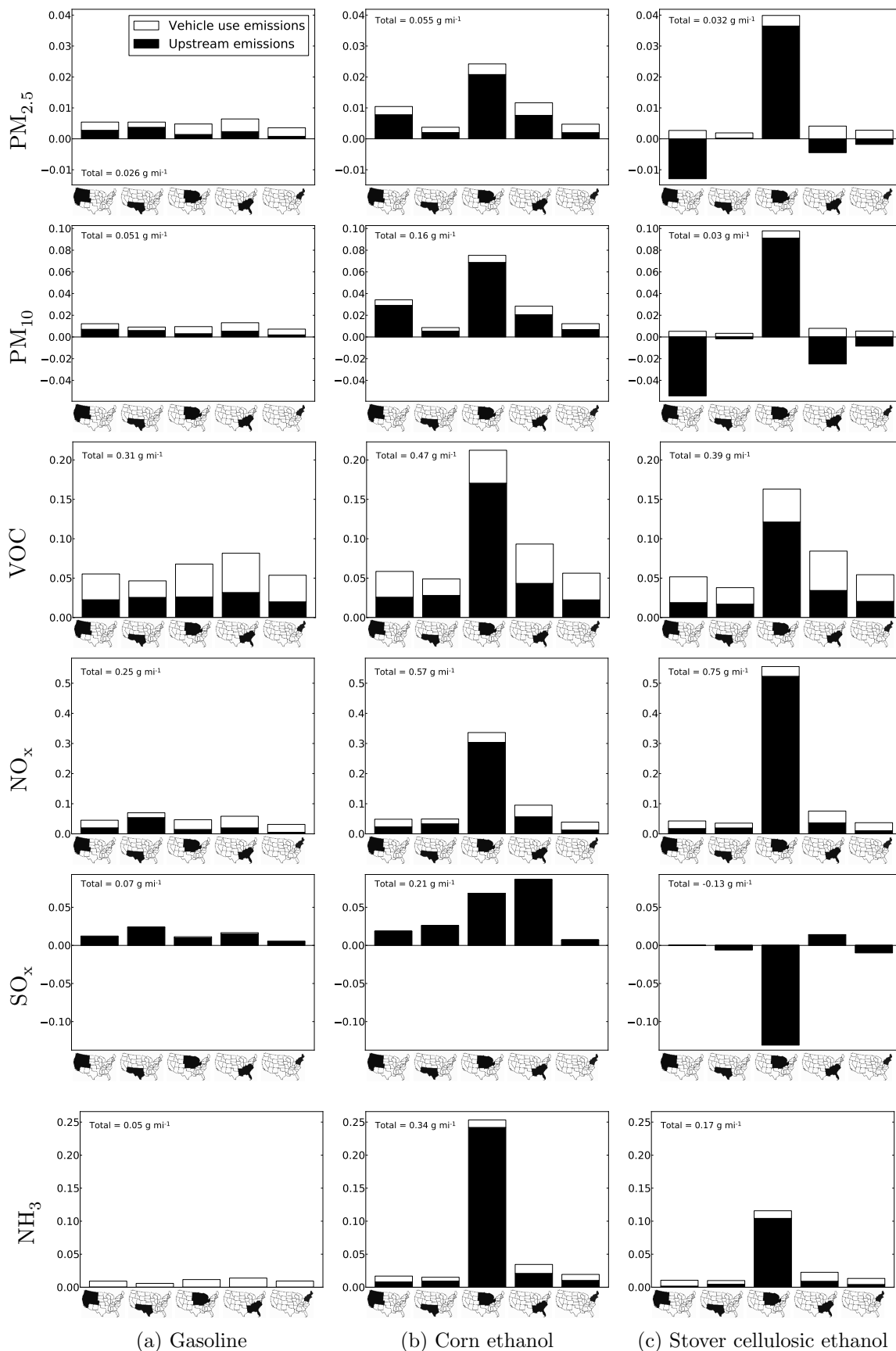
Figure S2-3: Contributions of US regions to total life cycle emissions (grams emitted per vehicle mile traveled). Emissions occurring outside of all regions (e.g., offshore emissions and international emissions outside of the modeling domain) are not included.

Table S2-4: Fractions of vehicle miles traveled occuring in each region (see above for data source) and total land area in each region.

Region	% VMT	% Land area
	18%	31%
	12%	19%
	23%	26%
	28%	18%
	19%	6%
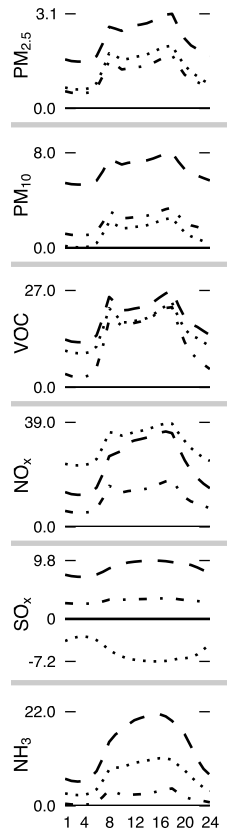
(a) hour of day

Figure S2-4: Temporal profiles of life cycle emissions (mg emitted per vehicle mile traveled) by hour of day (weekend).

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[3] US Census Bureau, *Census 2000 Urbanized Areas Cartographic Boundary Files*, 2001, URL: http://www.census.gov/geo/www/cob/ua2000.html.

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Supplemental Information 3a: Gasoline

A spatially and temporally explicit life cycle inventory of air pollutants from gasoline and ethanol in the United States

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This document contains maps of emissions contributions from those processes and pollutants that contribute more than 1% of life cycle total emissions. The amount of emissions is indicated by color intensity for line and area sources, and by rings for point sources, with the area of the ring being proportional to total emissions at each site.

Figure S3a-1: Color scales for area and line maps below.

(a) CO

(b) PM$_{10}$

(c) VOC

(d) PM$_{2.5}$

(e) NO$_x$

(f) NH$_3$

(g) SO$_x$

(h) Temporal
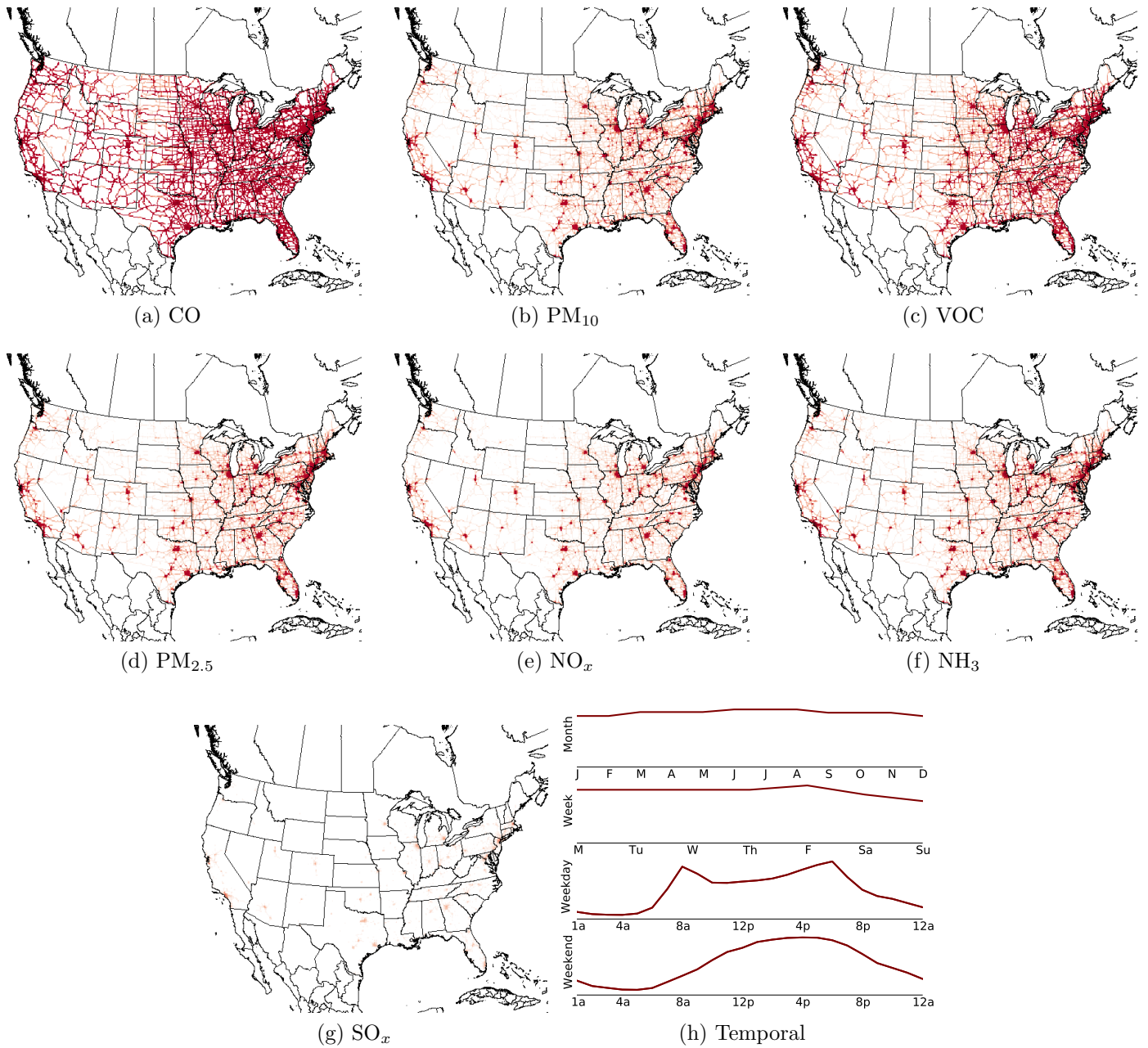
Figure S3a-2: **Process:** Vehicles.Emissions; **Technology/subprocess:** Light Duty Vehicle, Gasoline - Exhaust; **Emissions: mg mile^{-1} (% life cycle total):** CO: 3.7e+03 (98%), PM$_{10}$: 29 (54%), VOC: 1.8e+02 (58%), PM$_{2.5}$: 15 (56%), NO$_x$: 1.4e+02 (52%), NH$_3$: 50 (99%), SO$_x$: 6.1 (8%); **Map name**: VMT.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) NO$_x$

(d) SO$_x$

(e) Temporal
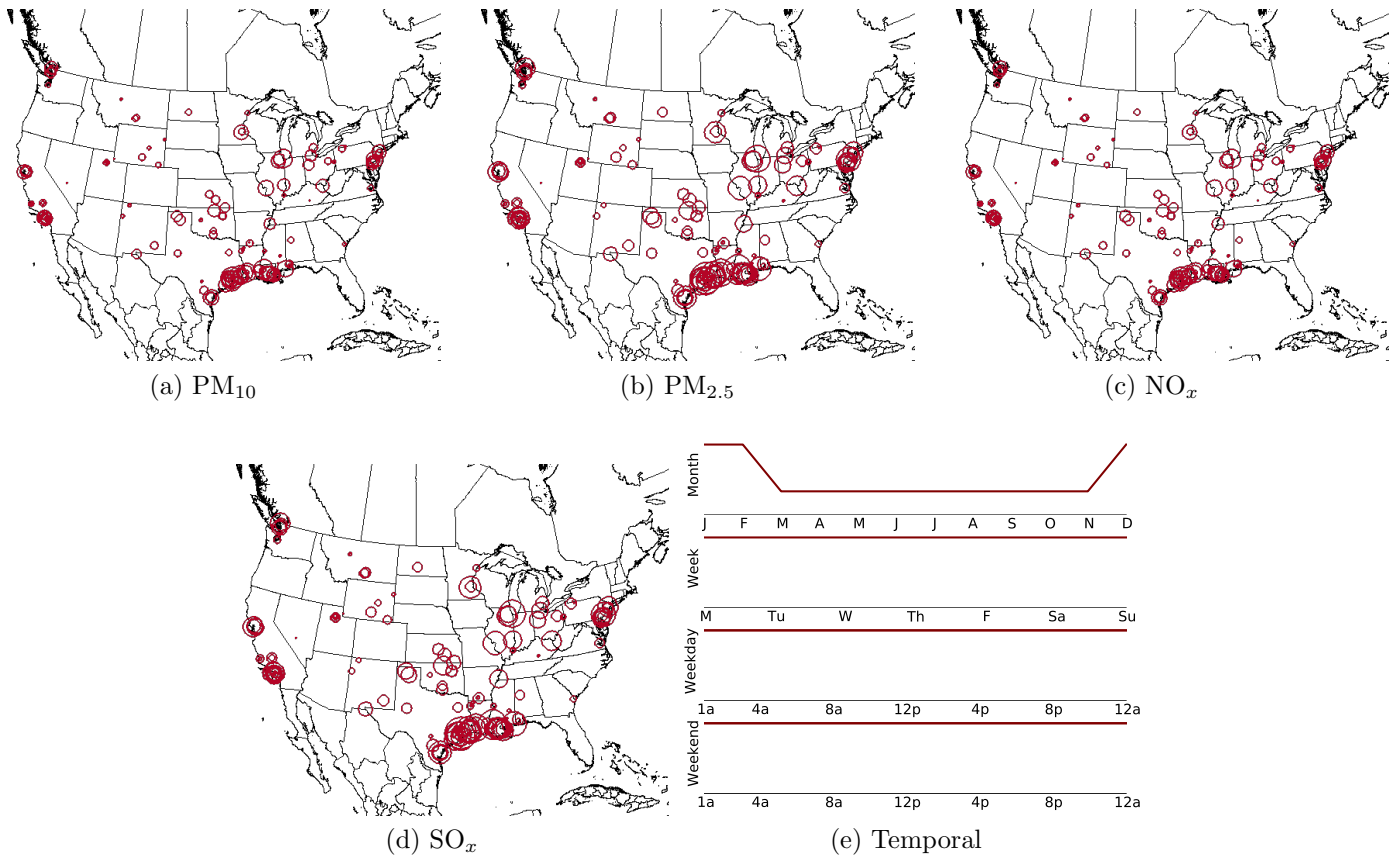
Figure S3a-3: **Process:** Petroleum.CGRefining; **Technology/subprocess:** Pet Coke, Industrial Boiler; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 7.6 (14%), PM$_{2.5}$: 3.8 (14%), NO$_x$: 12 (4%), SO$_x$: 15 (20%); **Map name**: OilRefineries.

(a) PM$_{10}$

(b) VOC

(c) PM$_{2.5}$

(d) NO$_x$

(e) SO$_x$

(f) Temporal
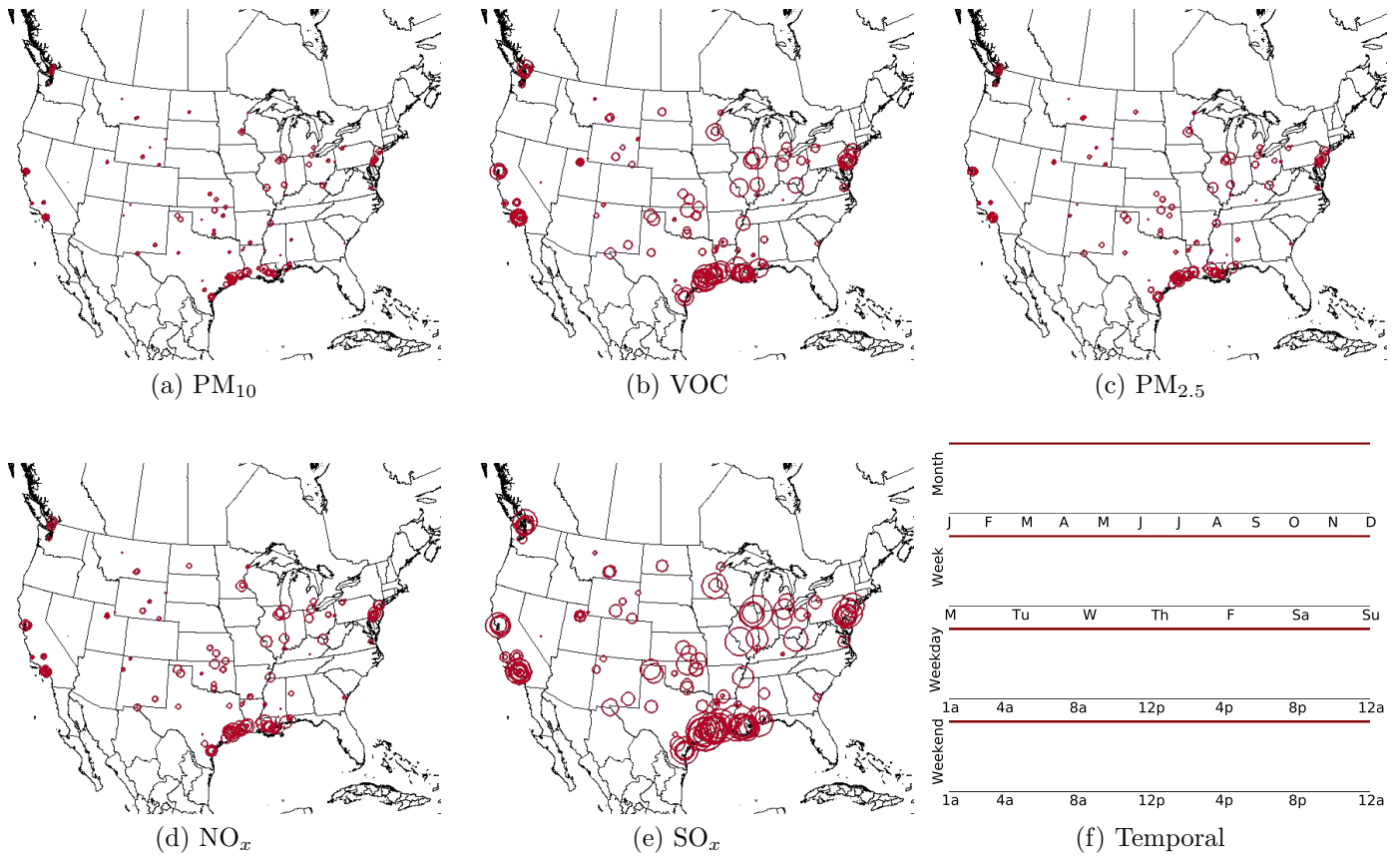
Figure S3a-4: **Process:** Petroleum.ConventionalGasolineCGRefining:Non-CombustionEmissionsnoncombustion; **Technology/subprocess:** Petroleum, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 1.6 (3%), VOC: 11 (3%), PM$_{2.5}$: 0.79 (3%), NO$_x$: 6.8 (2%), SO$_x$: 22 (29%); **Map name**: OilRefineries.

(a) PM$_{2.5}$

(b) PM$_{10}$

(c) Temporal
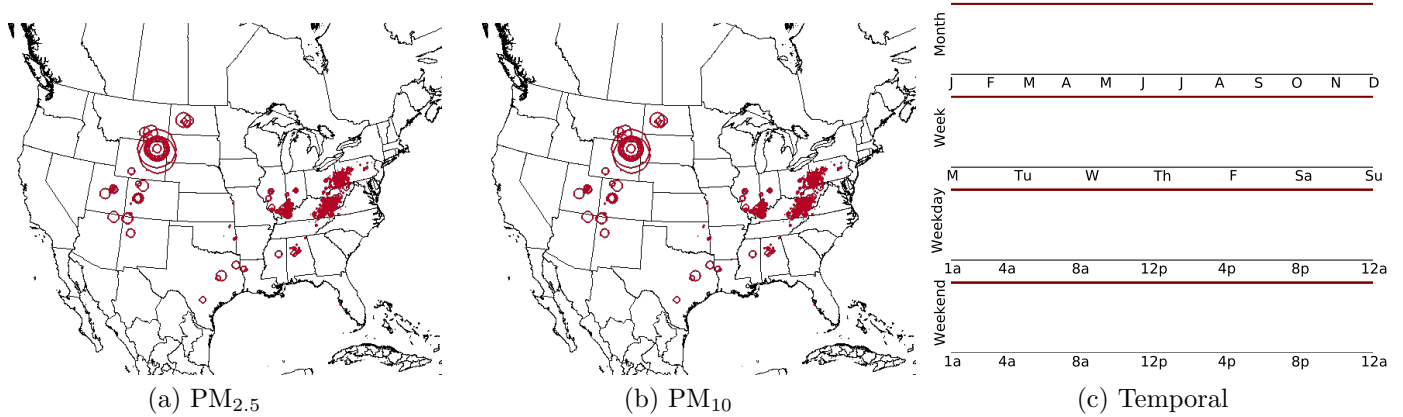
Figure S3a-5: **Process:** Coal.MiningNoncombustion; **Technology/subprocess:** Coal, Mining and Cleaning Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 2.1 (7%), PM$_{10}$: 8.5 (16%); **Map name**: CoalMining.

(a) VOC

(b) Temporal
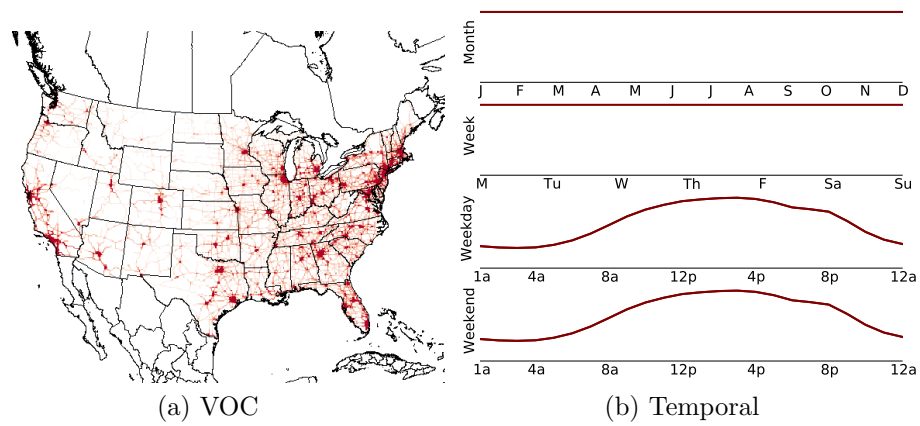
Figure S3a-6: **Process:** Petroleum.ConventionalGasolineCGTransportationandDistributionRefStation; **Technology/subprocess:** Petroleum, Refilling Station; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 65 (21%); **Map name**: VMT.

(a) NO$_x$

(b) SO$_x$

(c) Temporal
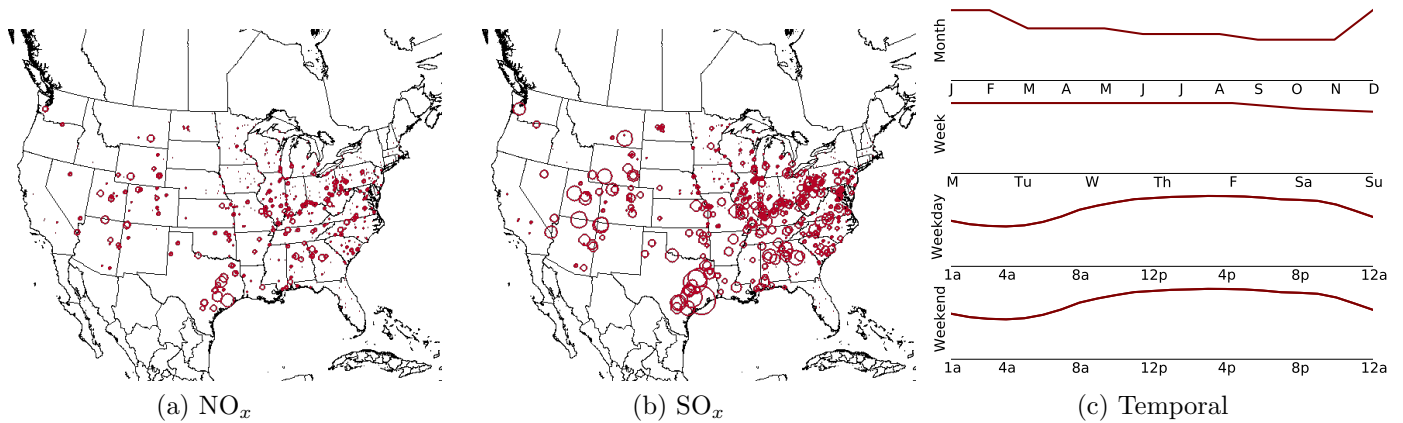
Figure S3a-7: **Process:** Electric.CoalFiredEF FOR Petroleum.CGRefining; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 3.4 (1%), SO$_x$: 9.4 (12%); **Map name**: ElectricGenerationCoal FOR OilRefineries.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) NO$_x$

(d) Temporal
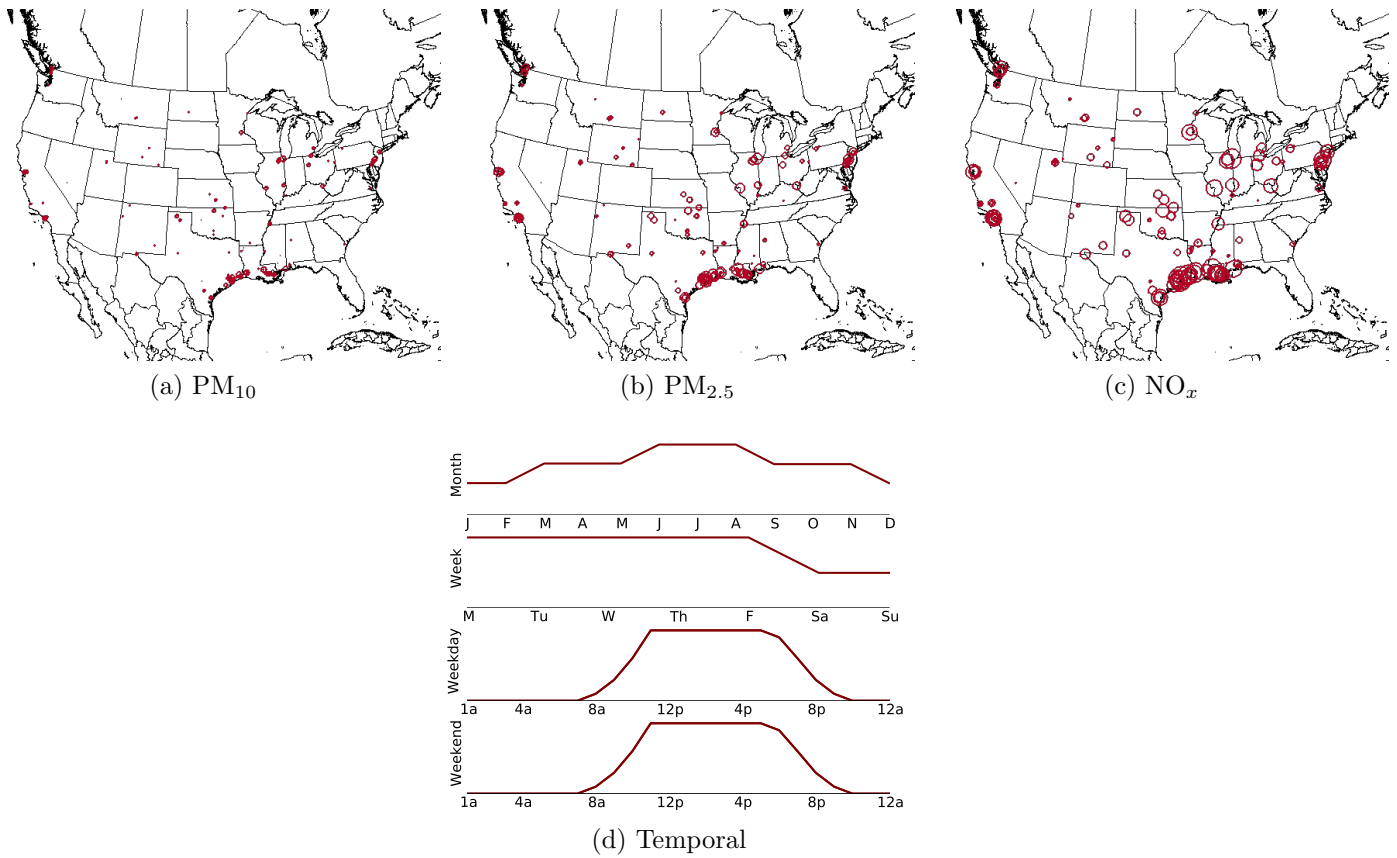
Figure S3a-8: **Process:** Petroleum.CGRefining; **Technology/subprocess:** Natural Gas, Utility/ Industrial Boiler (¿100 mmBtu/hr input); **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 0.69 (1%), PM$_{2.5}$: 0.69 (2%), NO$_x$: 12 (4%); **Map name**: OilRefineries.

(a) VOC

(b) Temporal
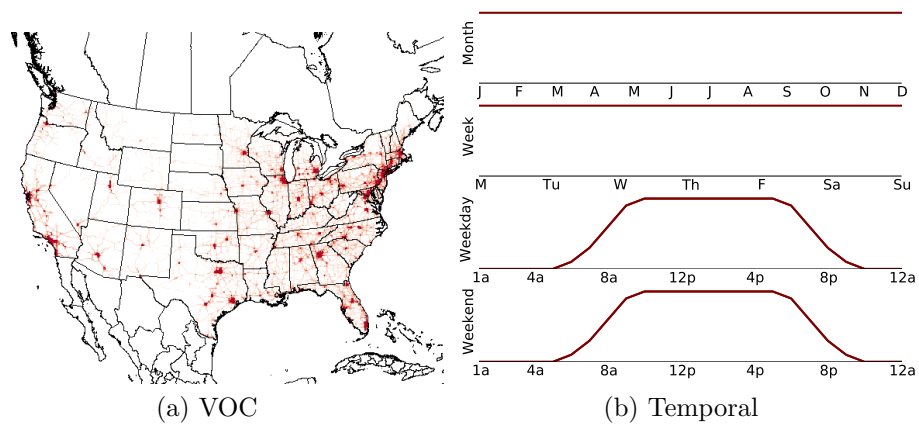
Figure S3a-9: **Process:** Petroleum.Conventional GasolineCG Transportation and DistributionBulkTerminal; **Technology/subprocess:** Petroleum, Bulk Terminal; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 33 (10%); **Map name**: VMT.

(a) PM$_{10}$ (b) PM$_{2.5}$ (c) Temporal
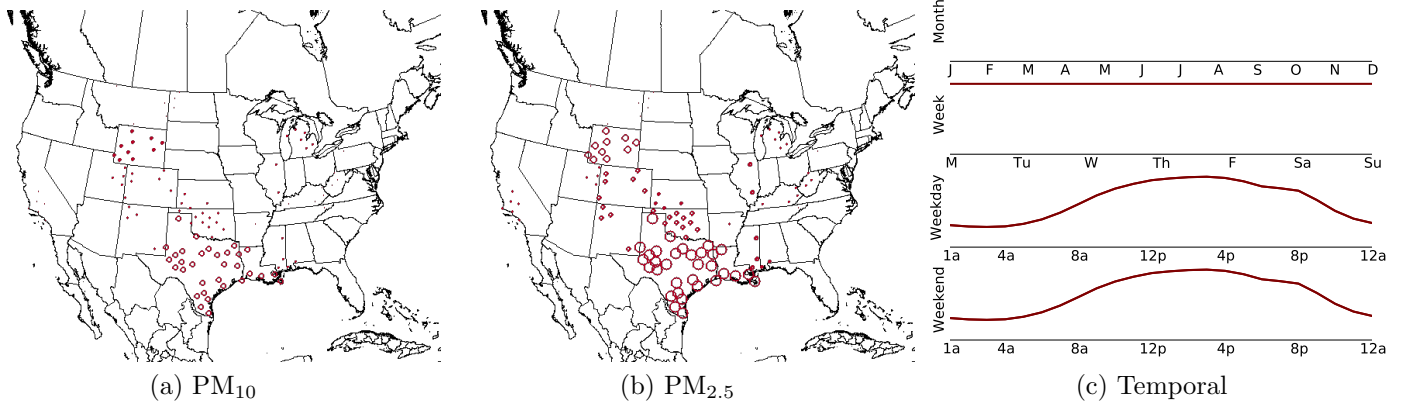
Figure S3a-10: **Process:** Hydrogen.H2centralNGorFGproductionnoncombustion; **Technology/subprocess:** NG, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 1.3 (2%), PM$_{2.5}$: 1.3 (4%); **Map name**: NGProcessing.

(a) NO$_x$ (b) Temporal
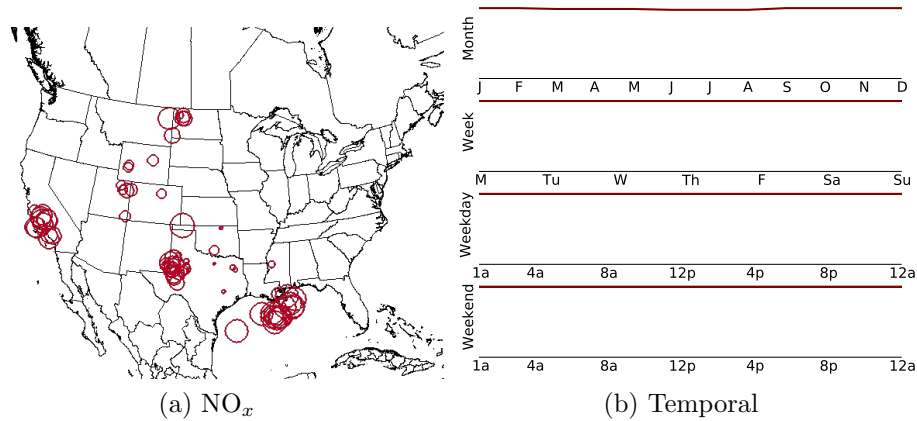
Figure S3a-11: **Process:** Petroleum.ConventionalCrudeRecovery; **Technology/subprocess:** Natural Gas, Stationary Reciprocating Engine; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 17 (6%); **Map name**: OilExtraction.

(a) PM$_{2.5}$ (b) NO$_x$ (c) Temporal
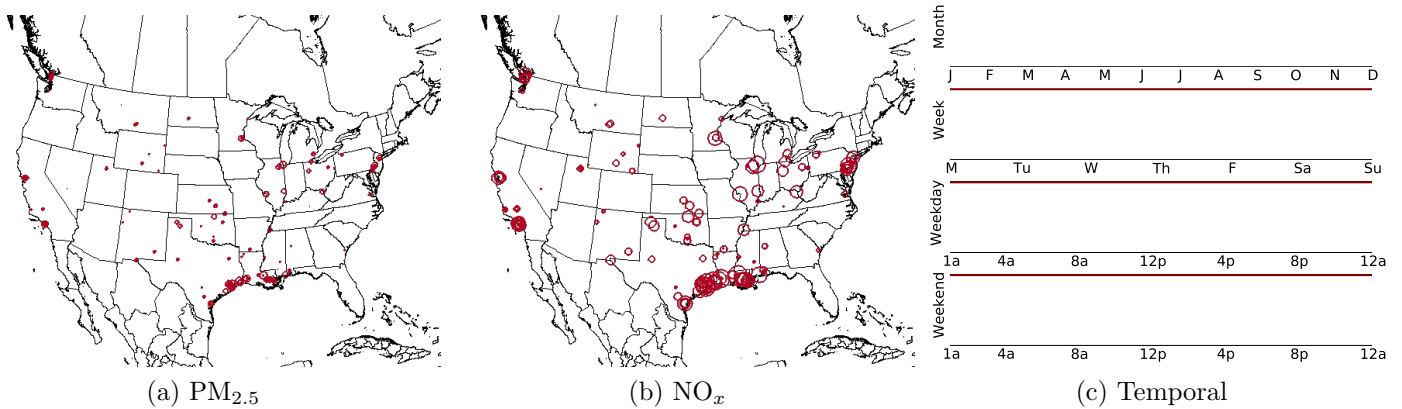
Figure S3a-12: **Process:** Petroleum.CGRefining; **Technology/subprocess:** Natural Gas, Large Gas Turbine; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 0.28 (1%), NO$_x$: 10 (3%); **Map name**: OilRefineries.

(a) SO$_x$ (b) NO$_x$ (c) Temporal
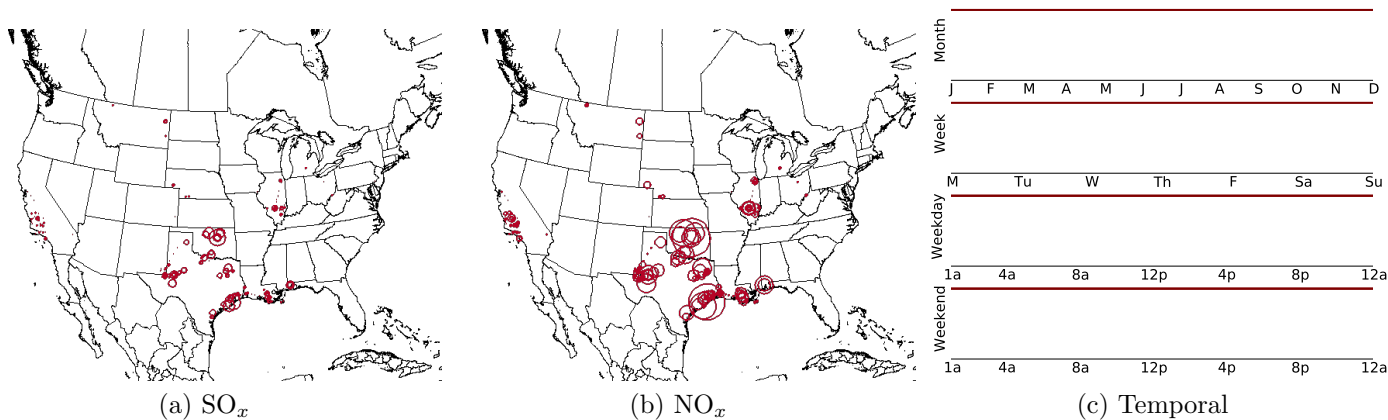
Figure S3a-13: **Process:** TandD.UScrude Pipelines; **Technology/subprocess:** Pipeline Reciprocating Engine: Current, Residual Oil; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 1.3 (1%), NO$_x$: 10 (3%); **Map name**: OilPipelines.

(a) PM$_{10}$ (b) SO$_x$ (c) Temporal
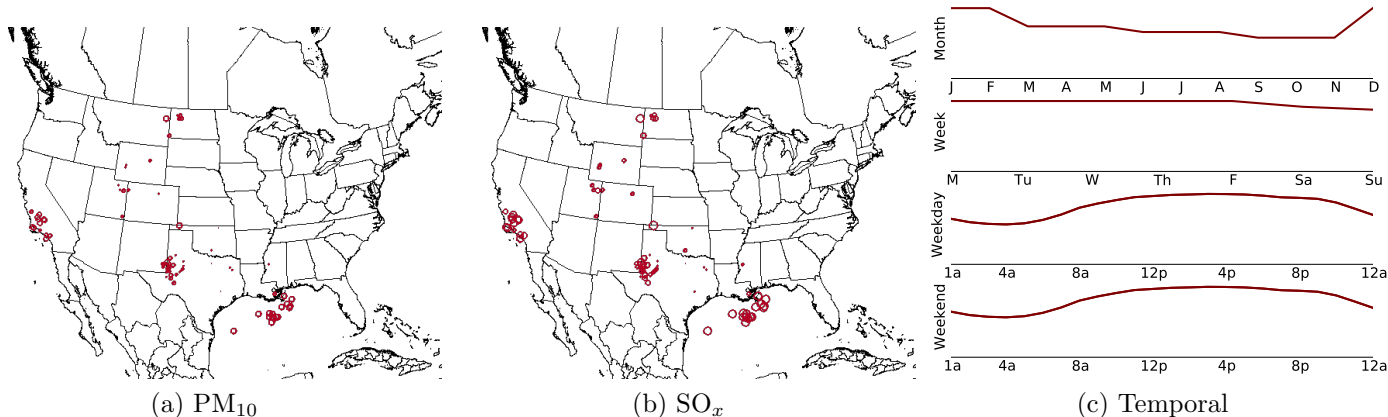
Figure S3a-14: **Process:** Petroleum.PetCokeTotal; **Technology/subprocess:** Undefined, Using Default SCC; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 0.76 (1%), SO$_x$: 1.4 (1%); **Map name**: OilExtraction.

(a) SO$_x$ (b) Temporal
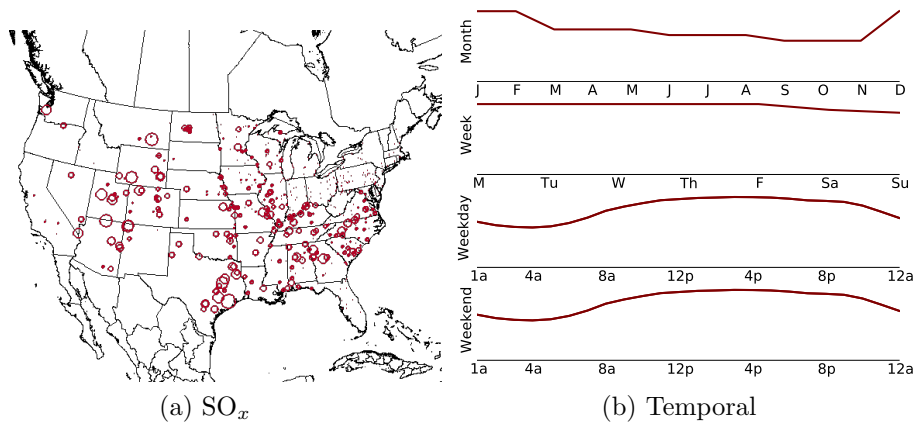
Figure S3a-15: **Process:** Electric.CoalFiredEF FOR Petroleum.ConventionalCrudeRecovery; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 3.6 (4%); **Map name**: ElectricGenerationCoal FOR OilExtraction.

| (a) NO$_x$ | (b) SO$_x$ | (c) Temporal |
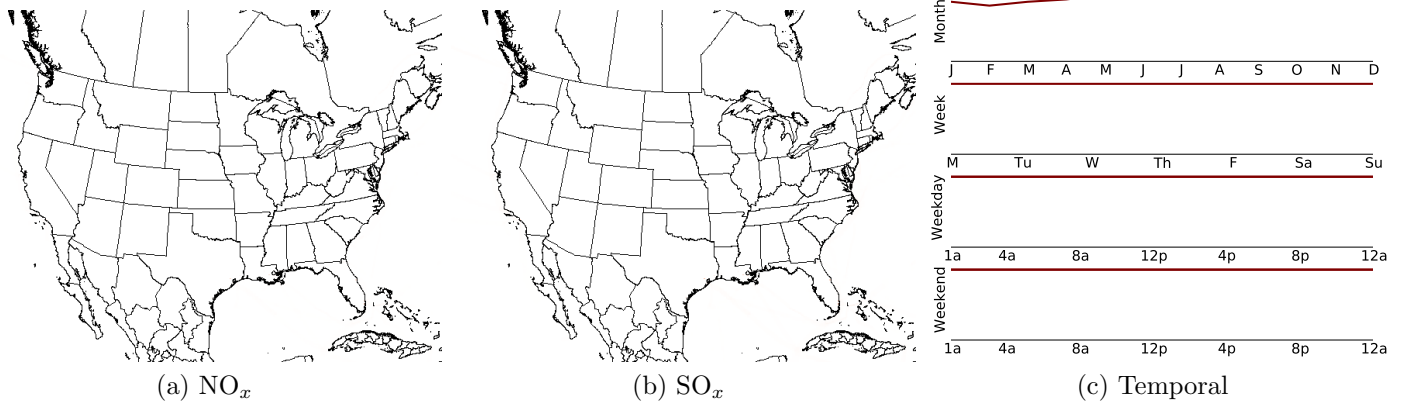
Figure S3a-16: **Process:** TandD.UScrude OceanTankers; **Technology/subprocess:** Ocean Tanker, Bunker Fuel; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 2.8 (1%), SO$_x$: 2 (2%); **Map name**: OceanTankers.

| (a) SO$_x$ | (b) Temporal |
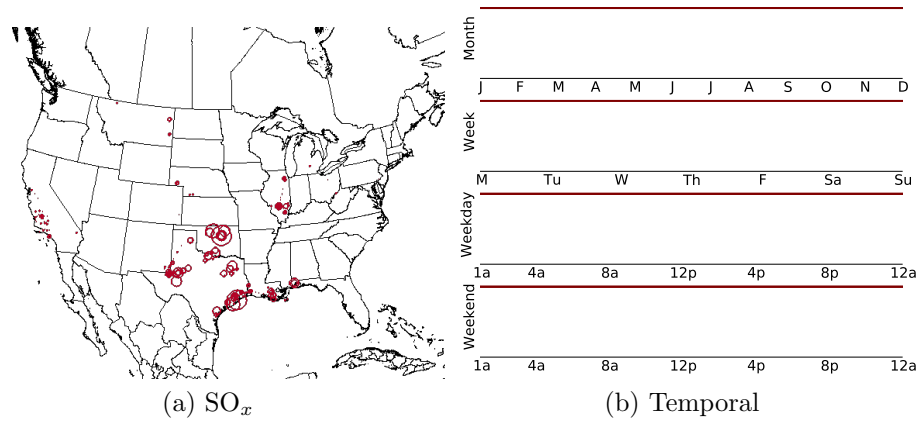
Figure S3a-17: **Process:** TandD.UScrude Pipelines; **Technology/subprocess:** Pipeline Turbine, Residual Oil; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 1.9 (2%); **Map name**: OilPipelines.

| (a) SO$_x$ | (b) Temporal |
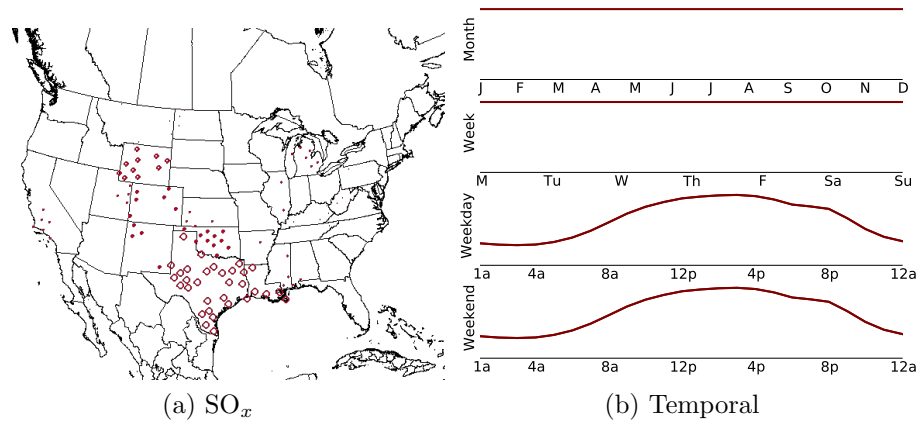
Figure S3a-18: **Process:** NG.asProcFuelNoncombustion; **Technology/subprocess:** NG, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 2.1 (2%); **Map name**: NGProcessing.

(a) VOC

(b) Temporal
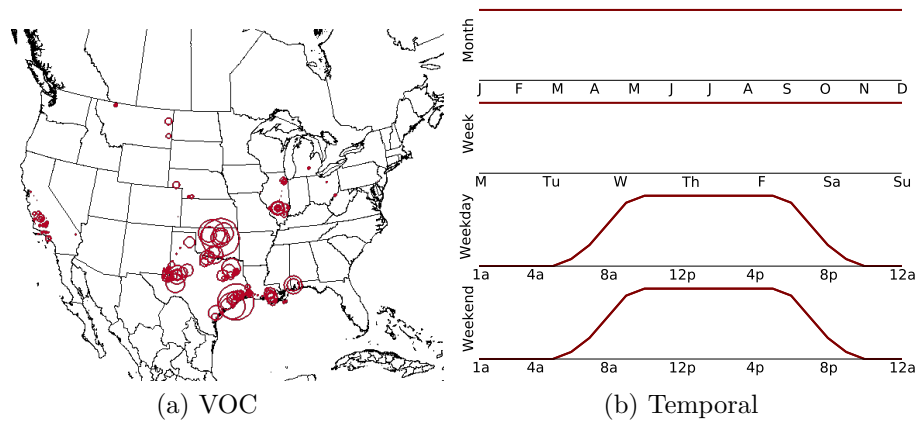
Figure S3a-19: **Process:** Petroleum.Crude OilTransportation to U.S. RefineriesBulkTerminal; **Technology/subprocess:** Petroleum, Bulk Terminal; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 7.8 (2%); **Map name**: OilPipelines.

(a) NO$_x$

(b) Temporal
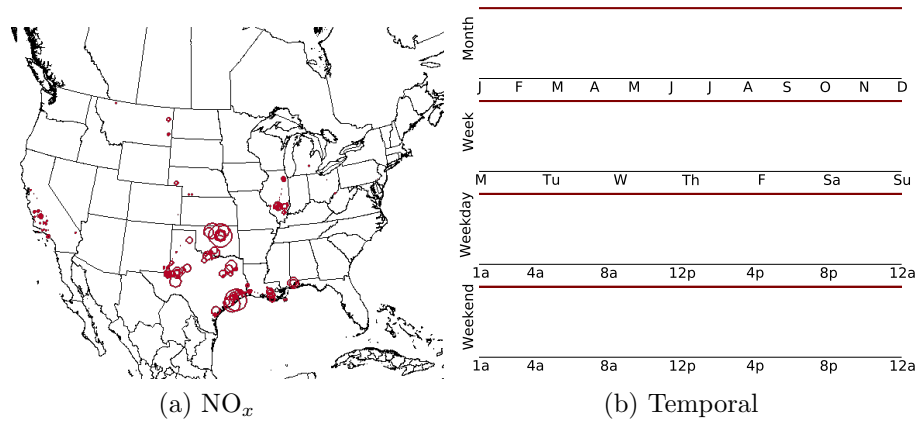
Figure S3a-20: **Process:** TandD.CGTrans Pipelines; **Technology/subprocess:** Pipeline Reciprocating Engine: Current, Residual Oil; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 3.8 (1%); **Map name**: OilPipelines.

(a) NO$_x$

(b) Temporal
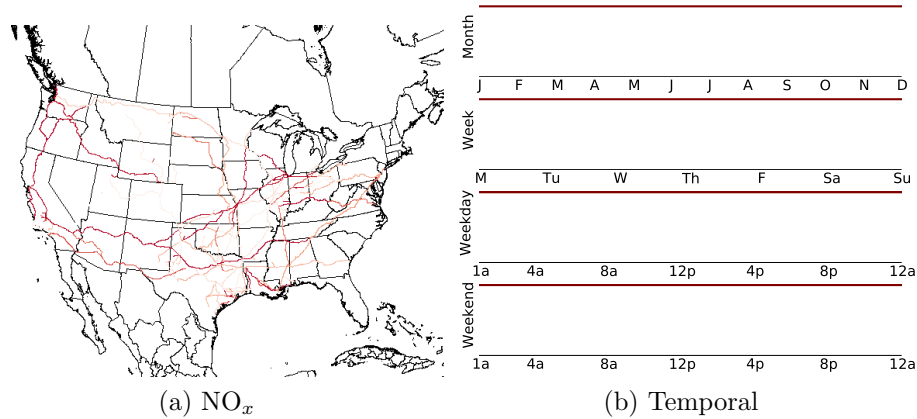
Figure S3a-21: **Process:** TandD.CGTrans Rail; **Technology/subprocess:** Locomotive, Diesel; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 3.5 (1%); **Map name**: OilExtraction TO OilRefineries RailNetwork.

(a) NO$_x$ (b) Temporal
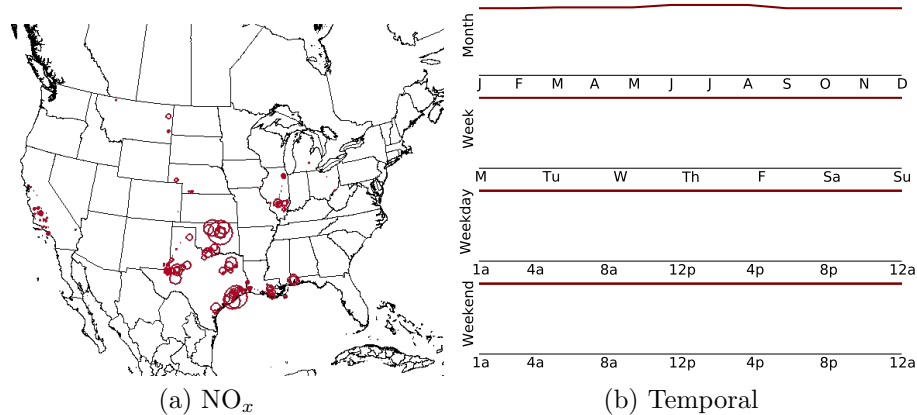
Figure S3a-22: **Process:** TandD.UScrude Pipelines; **Technology/subprocess:** Pipeline Reciprocating Engine: Current, Diesel; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 4.1 (1%); **Map name**: OilPipelines.

(a) SO$_x$ (b) Temporal
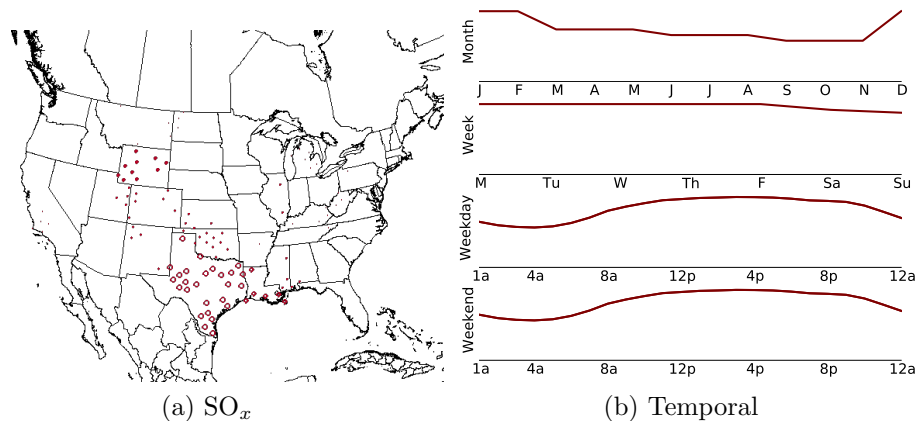
Figure S3a-23: **Process:** NG.NANGProcessingNoncombustion; **Technology/subprocess:** Undefined, Using Default SCC; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 1.1 (1%); **Map name**: NGProcessing.
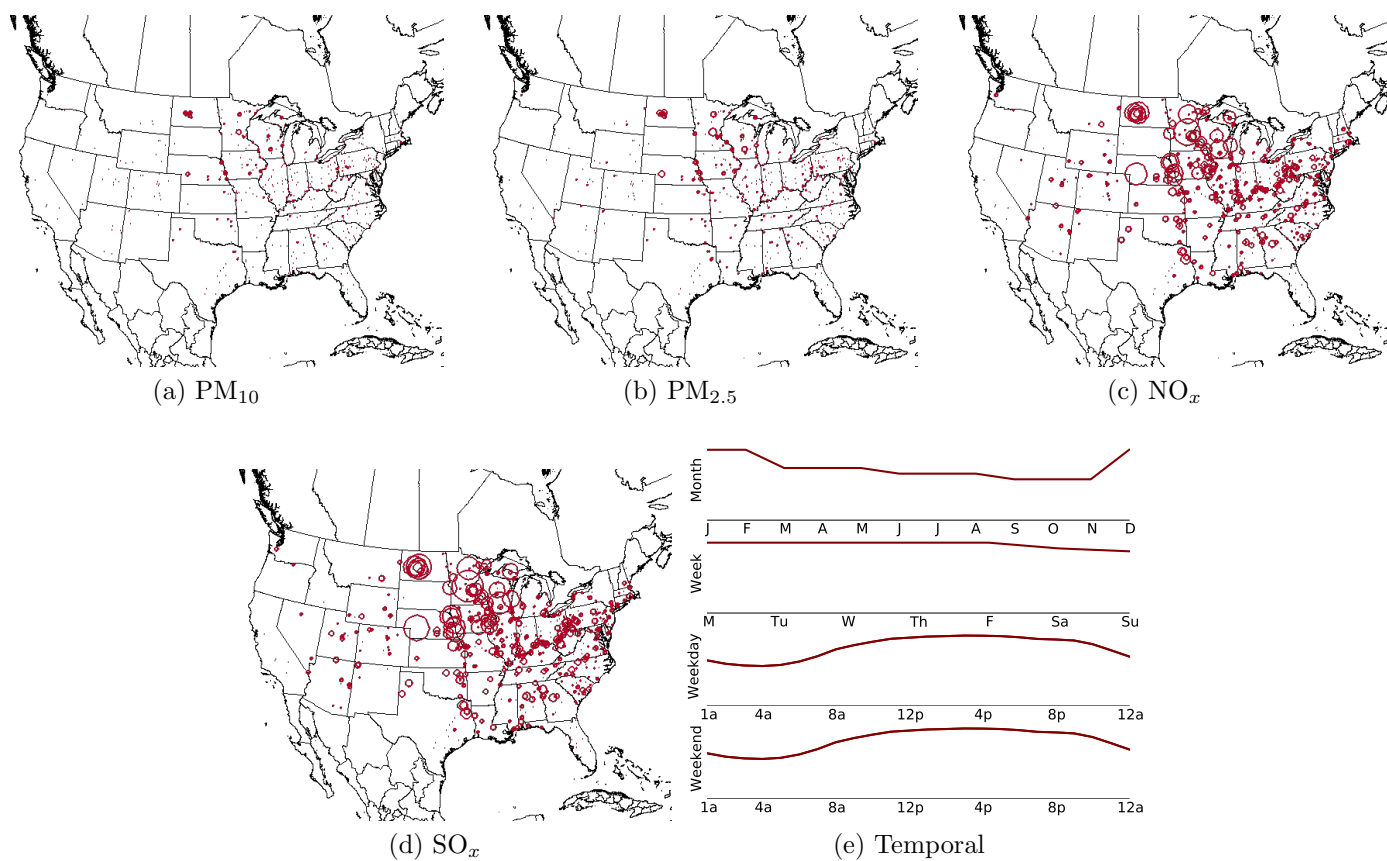

(a) SO$_x$ (b) Temporal
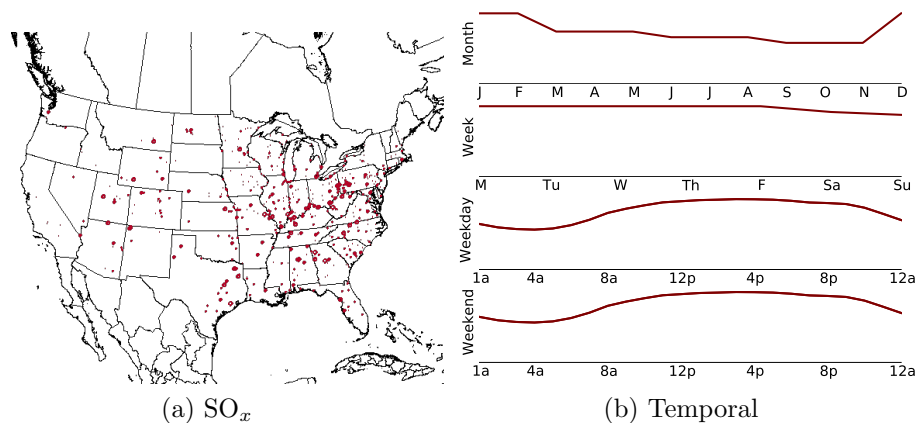
Figure S3a-24: **Process:** Electric.CoalFiredEF; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 0.94 (1%); **Map name**: ElectricGenerationCoal.

(a) VOC

(b) Temporal
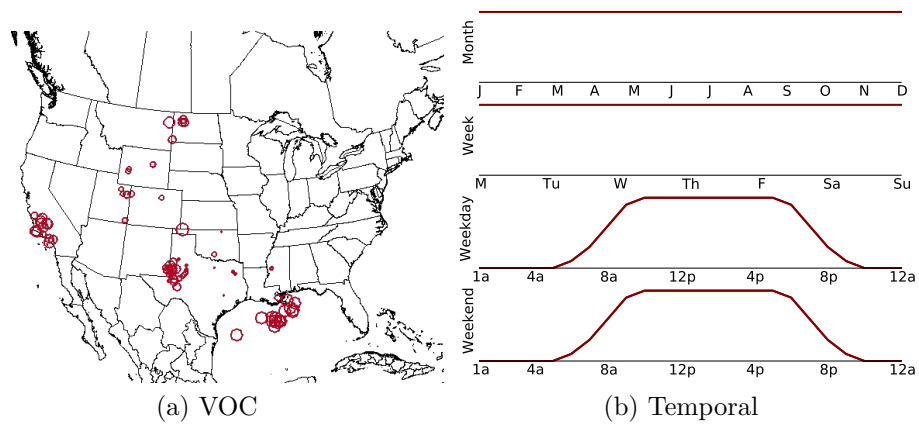
Figure S3a-25: **Process:** Petroleum.Crude OilRecoveryBulkTerminal; **Technology/subprocess:** Petroleum, Bulk Terminal; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 3.6 (1%); **Map name**: OilExtraction.

Supplemental Information 3b: Corn Grain Ethanol

A spatially and temporally explicit life cycle inventory of air pollutants from gasoline and ethanol in the United States

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This document contains maps of emissions contributions from those processes and pollutants that contribute more than 1% of life cycle total emissions. The amount of emissions is indicated by color intensity for line and area sources, and by rings for point sources, with the area of the ring being proportional to total emissions at each site. In all cases, red indicates positive emissions, and blue indicates negative emissions.

Figure S3b-1: Color scales for area and line maps below.

(a) CO

(b) PM$_{10}$

(c) VOC

(d) PM$_{2.5}$

(e) NO$_x$

(f) NH$_3$

(g) Temporal
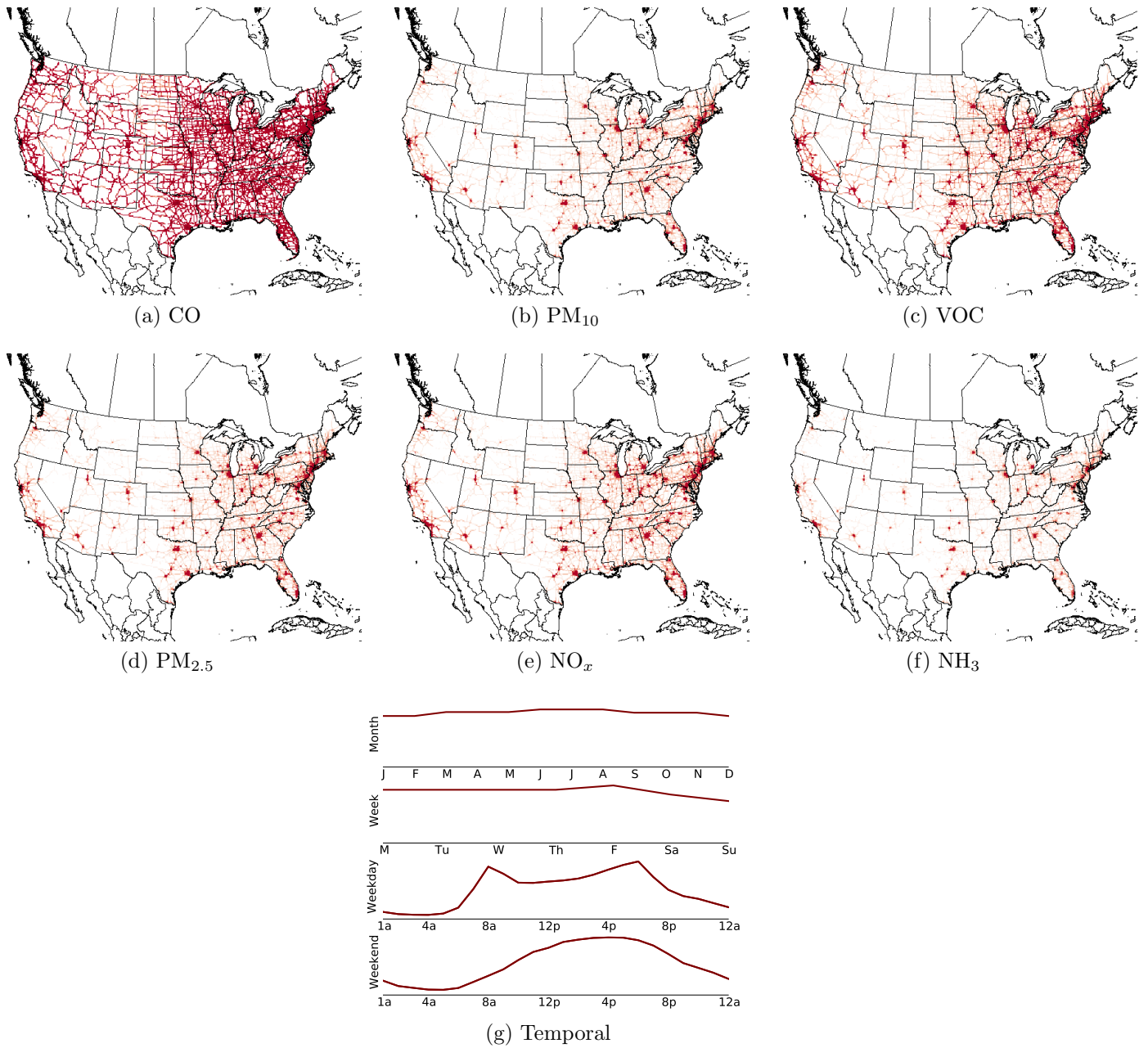
Figure S3b-2: **Process:** Vehicles.Emissions; **Technology/subprocess:** Light Duty Vehicle, Gasoline - Exhaust; **Emissions: mg mile^{-1} (% life cycle total):** CO: 3.7e+03 (96%), PM$_{10}$: 29 (17%), VOC: 1.8e+02 (38%), PM$_{2.5}$: 15 (26%), NO$_x$: 1.4e+02 (24%), NH$_3$: 50 (14%); **Map name**: VMT.

(a) NO$_x$ (b) NH$_3$ (c) Temporal
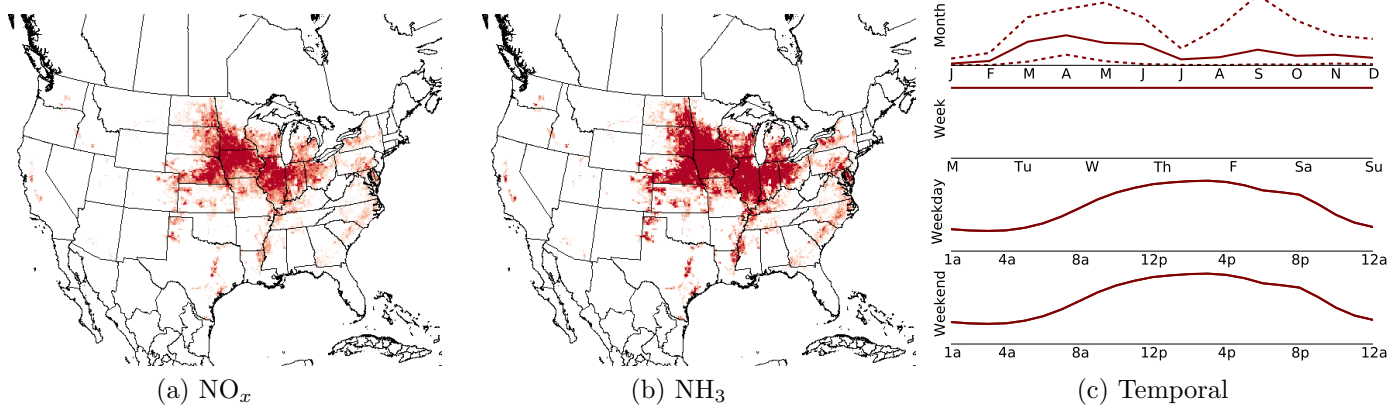
Figure S3b-3: **Process:** ETOH.CornNFertilizerNitrification; **Technology/subprocess:** Fertilizer, Nitrification Emissions; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 1.3e+02 (22%), NH$_3$: 2.8e+02 (83%); **Map name**: FarmAreaCornTotal.

(a) PM$_{2.5}$ (b) PM$_{10}$ (c) VOC

(d) Temporal
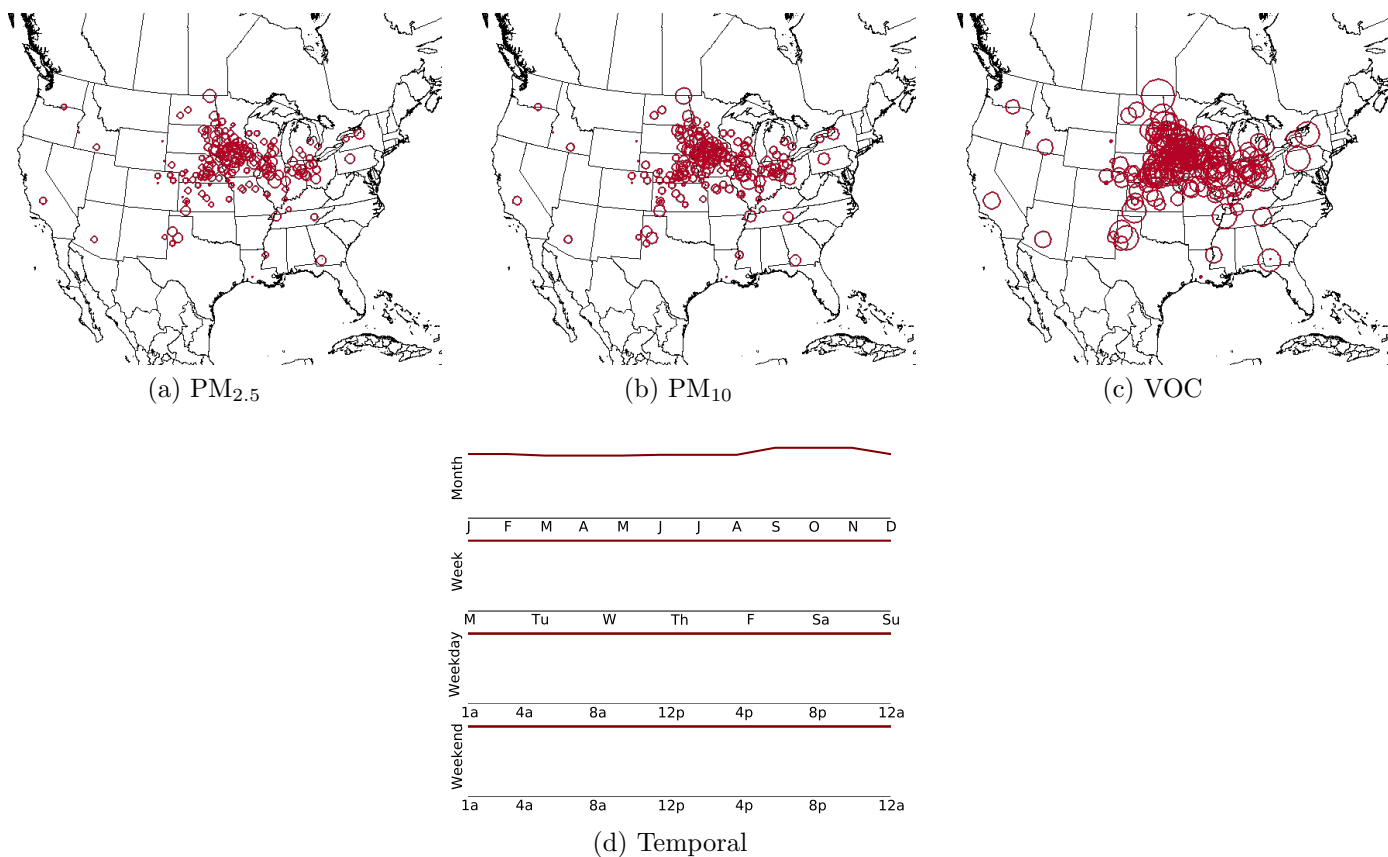
Figure S3b-4: **Process:** ETOH.BiorefineryNoncombustion; **Technology/subprocess:** Ethanol, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 9.5 (17%), PM$_{10}$: 56 (35%), VOC: 1.5e+02 (30%); **Map name**: CurrentBiorefineries.

(a) PM$_{2.5}$ (b) PM$_{10}$ (c) Temporal
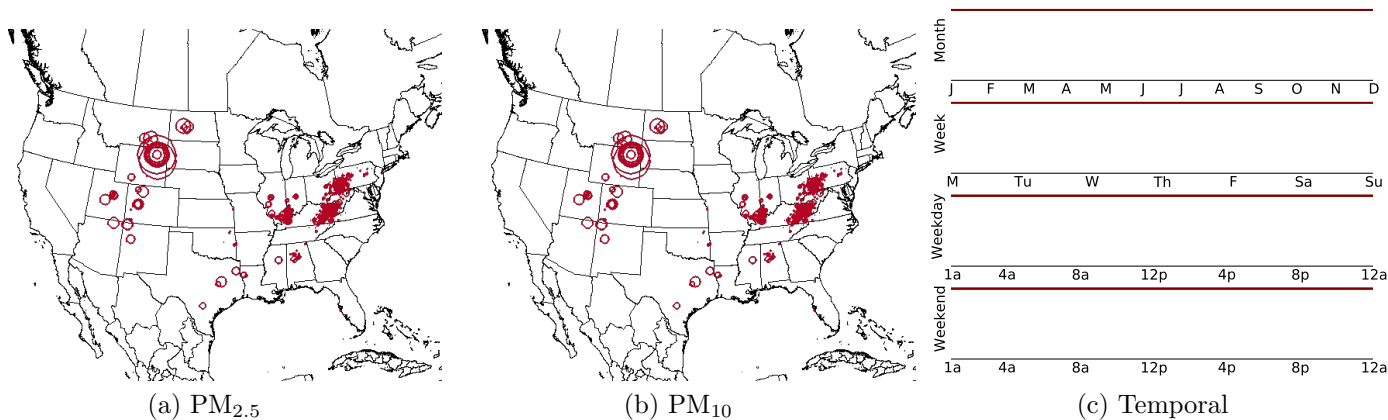
Figure S3b-5: **Process:** Coal.MiningNoncombustion; **Technology/subprocess:** Coal, Mining and Cleaning Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 13 (23%), PM$_{10}$: 53 (33%); **Map name**: CoalMining.

(a) PM$_{10}$ (b) PM$_{2.5}$ (c) NO$_x$

(d) SO$_x$ (e) Temporal

Figure S3b-6: **Process:** Electric.CoalFiredEF FOR ETOH.DryMillEthanol; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 1.8 (1%), PM$_{2.5}$: 0.89 (1%), NO$_x$: 25 (4%), SO$_x$: 69 (33%); **Map name**: ElectricGenerationCoal FOR CurrentBiorefineries.

(a) SO$_x$ | (b) Temporal
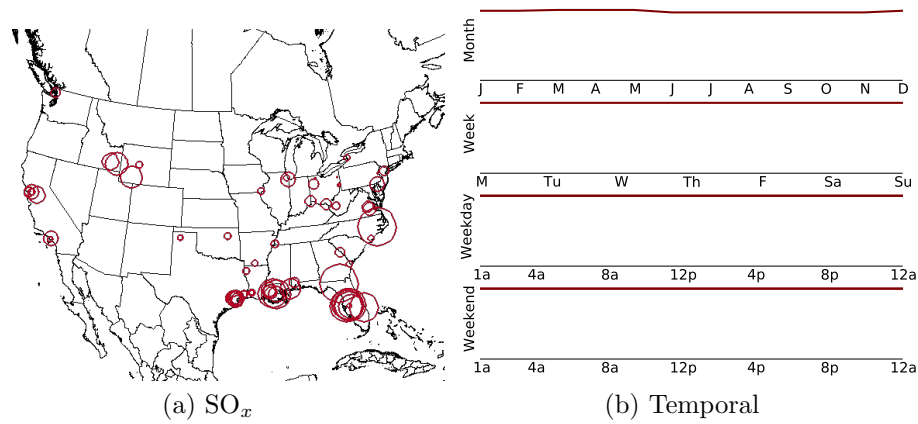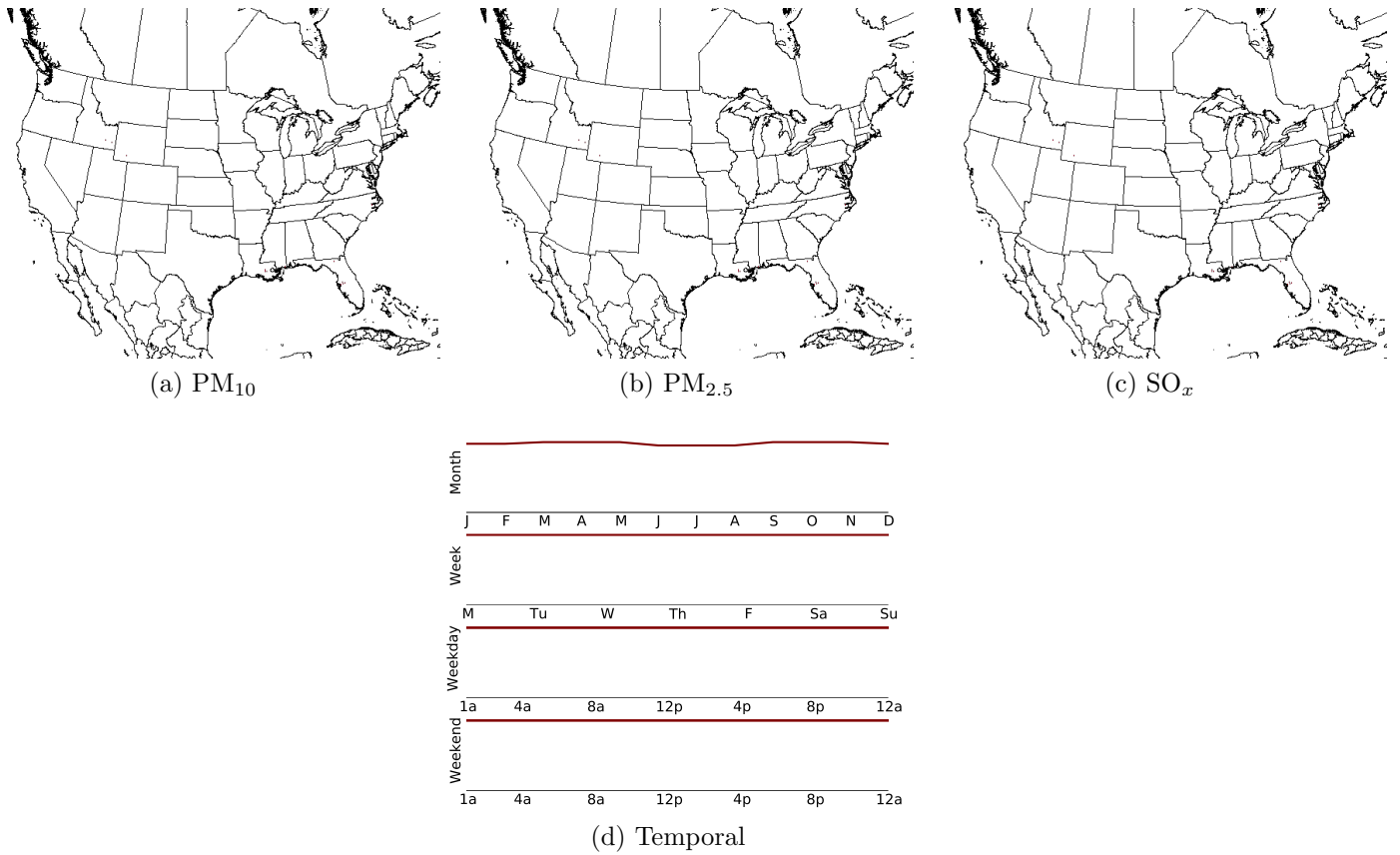
Figure S3b-7: **Process:** Ag Inputs.SulfuricAcidProcessEmis; **Technology/subprocess:** Sulfuric Acid, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 72 (34%); **Map name**: SulfuricAcid.

(a) PM$_{10}$ | (b) PM$_{2.5}$ | (c) NO$_x$

(d) Temporal
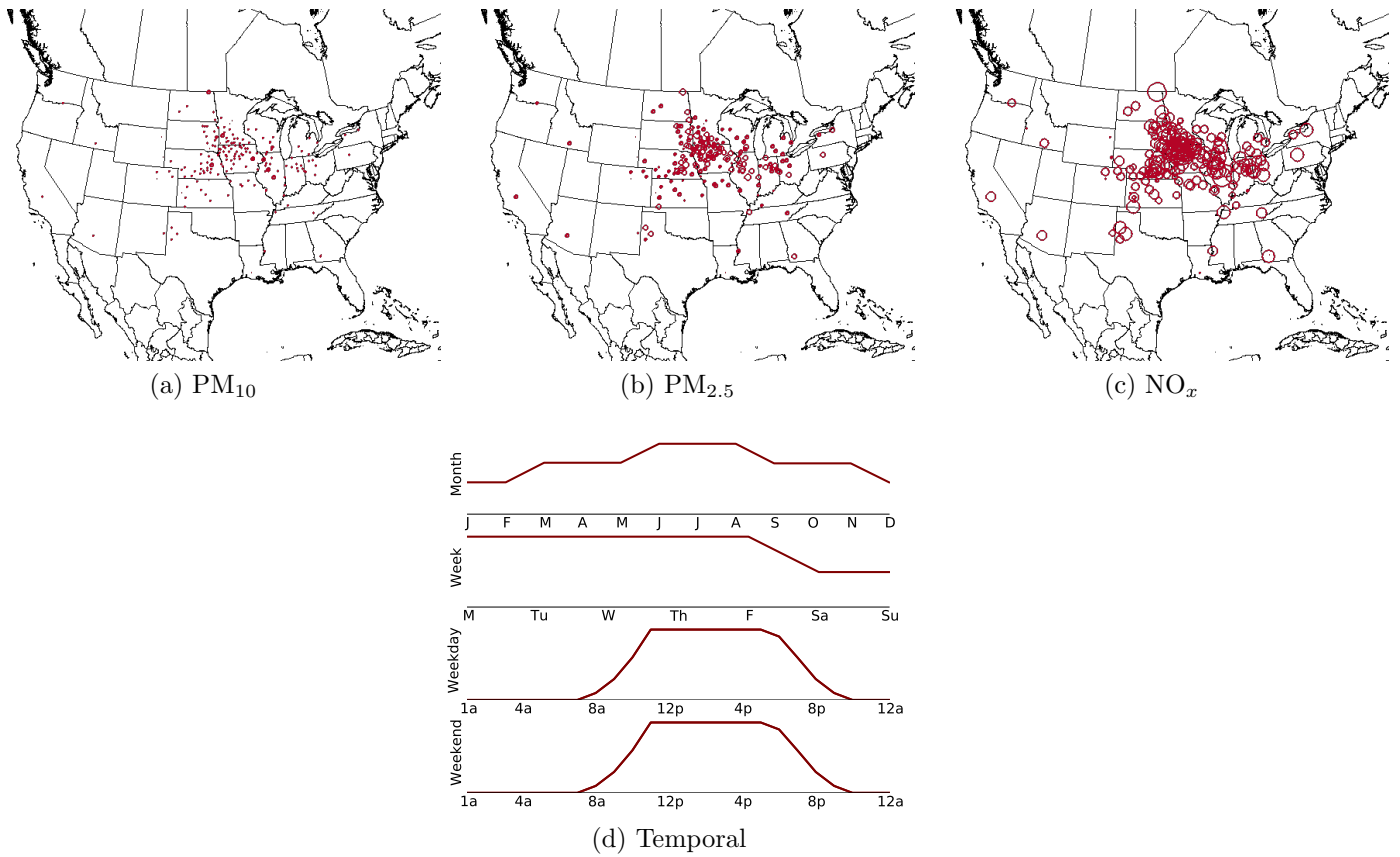
Figure S3b-8: **Process:** ETOH.DryMillEthanol; **Technology/subprocess:** Natural Gas, Utility/ Industrial Boiler (¿100 mmBtu/hr input); **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 2.5 (1%), PM$_{2.5}$: 2.5 (4%), NO$_x$: 44 (7%); **Map name**: CurrentBiorefineries.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) NO$_x$

(d) Temporal
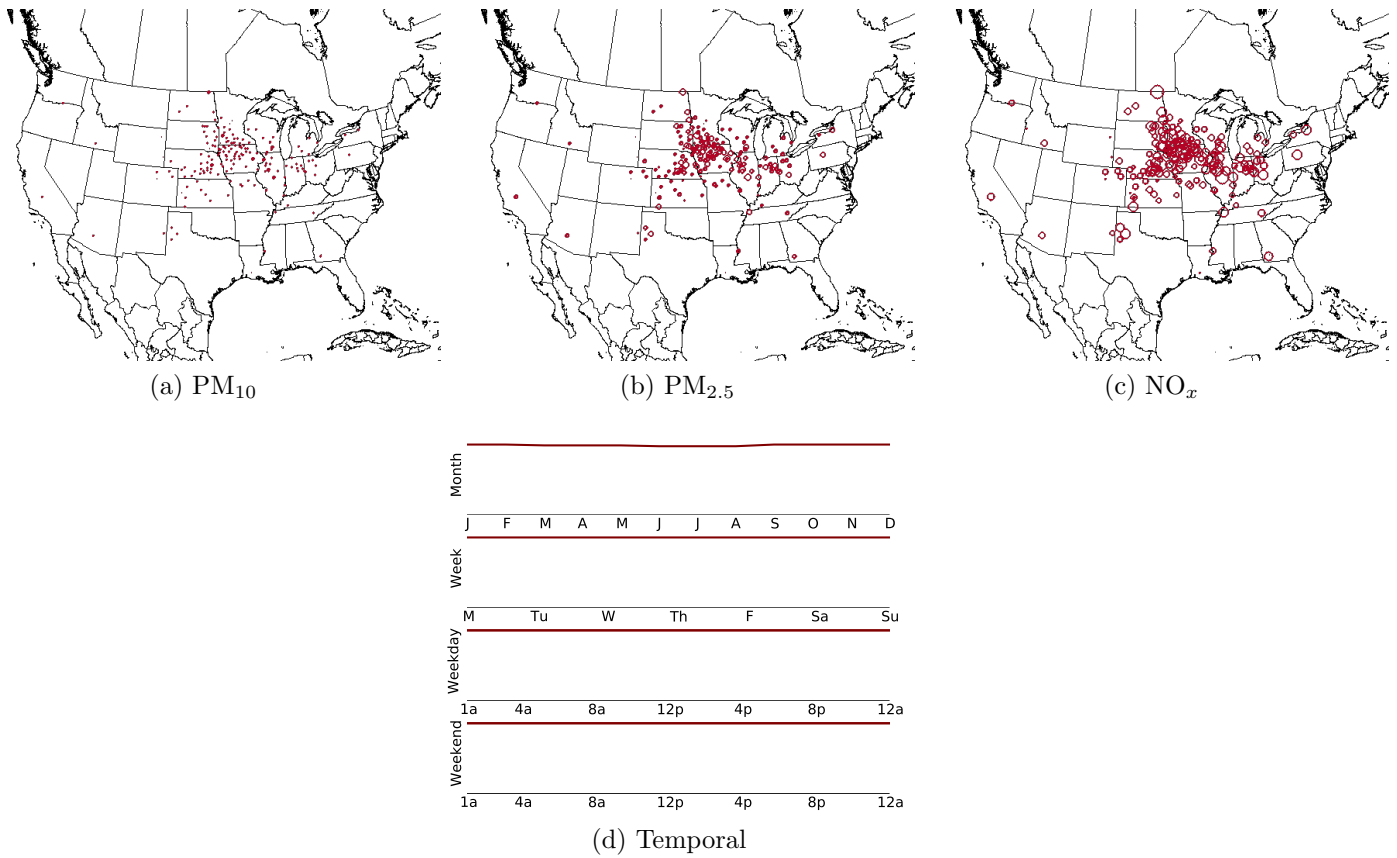
Figure S3b-9: **Process:** ETOH.DryMillEthanol; **Technology/subprocess:** Natural Gas, Small Industrial Boiler (10-100 mmBtu/hr input); **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 2.3 (1%), PM$_{2.5}$: 2.3 (4%), NO$_x$: 23 (4%); **Map name**: CurrentBiorefineries.

(a) PM$_{10}$

(b) VOC

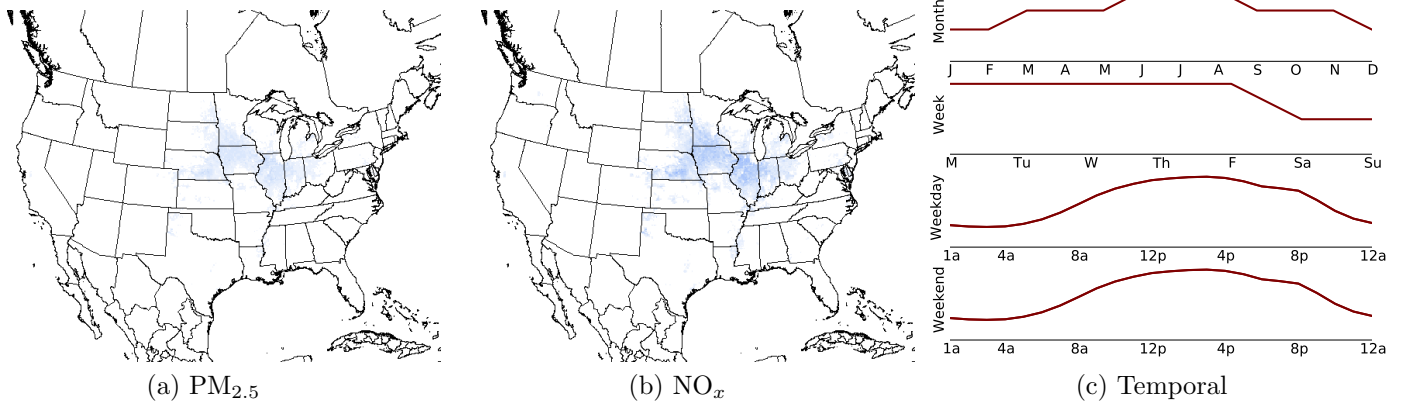(c) PM$_{2.5}$

(d) NO$_x$

(e) Temporal
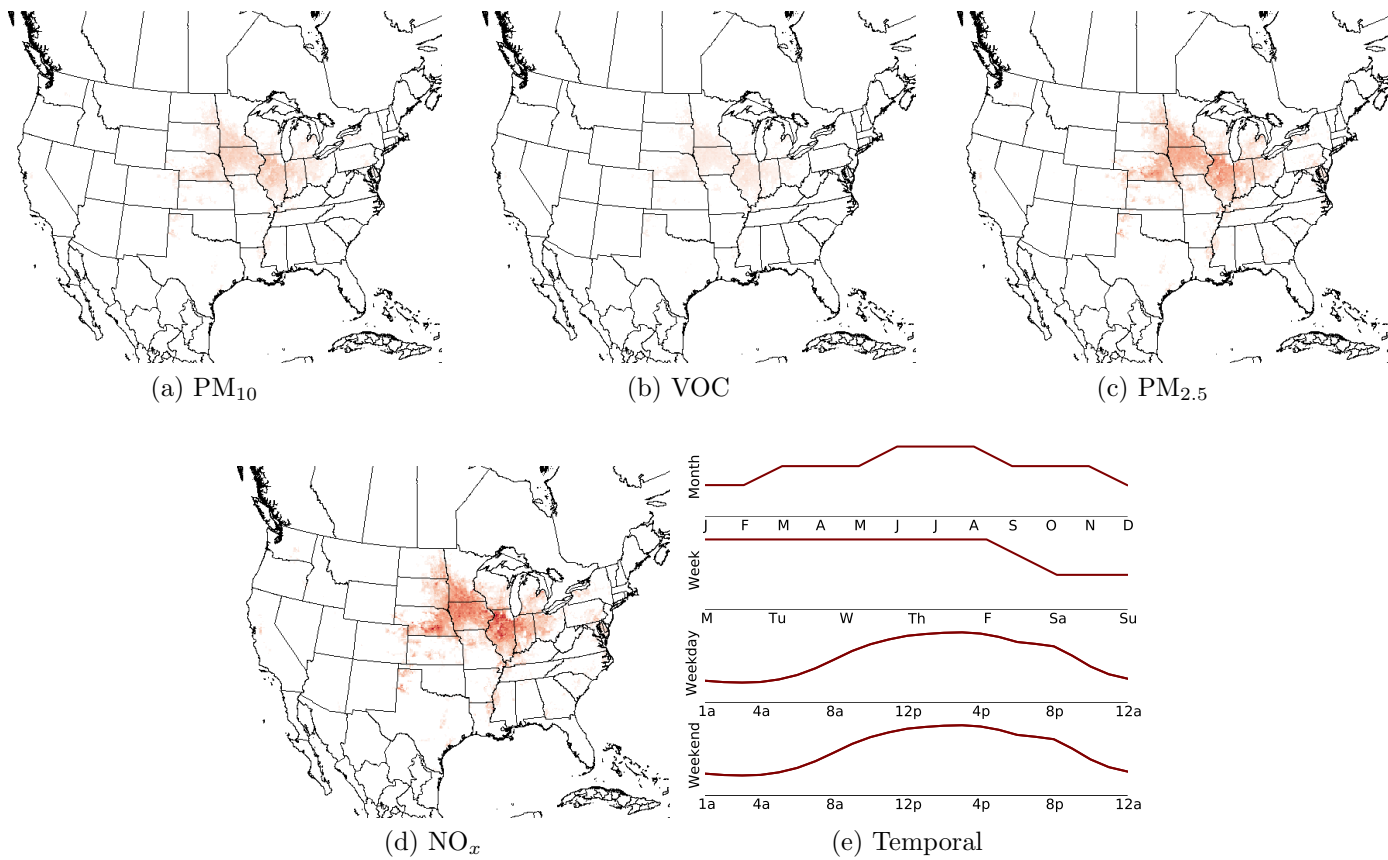
Figure S3b-10: **Process:** ETOH.CornFarming; **Technology/subprocess:** Diesel Fuel, Farming Tractor; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 4.3 (2%), VOC: 4.8 (1%), PM$_{2.5}$: 3.8 (6%), NO$_x$: 47 (8%); **Map name**: FarmProductionCornTotal.

(a) PM$_{2.5}$

(b) NO$_x$

(c) SO$_x$

(d) Temporal
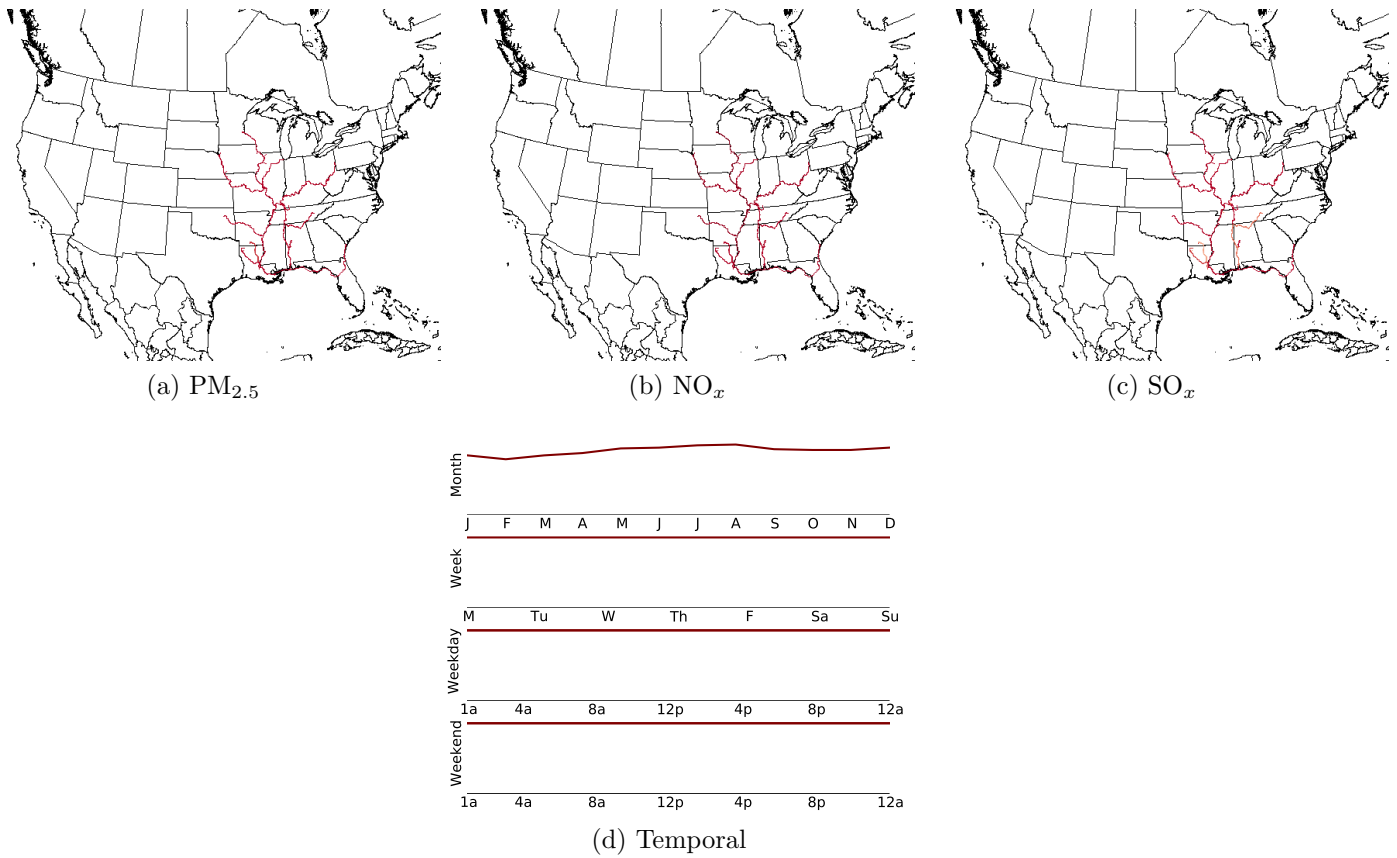
Figure S3b-11: **Process:** TandD.USETOHtrans Barge; **Technology/subprocess:** Barge, Residual Oil; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 1.2 (2%), NO$_x$: 35 (6%), SO$_x$: 8.4 (4%); **Map name**: CurrentBiorefineries TO OilRefineries WaterNetwork.
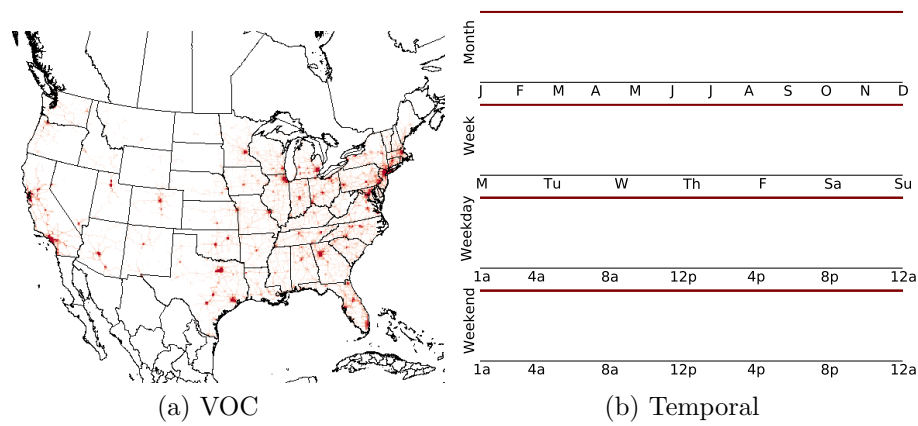

(a) VOC

(b) Temporal
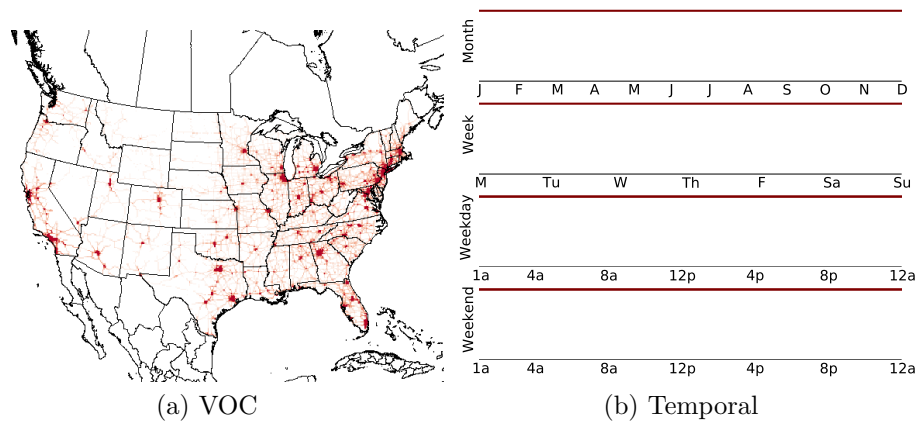
Figure S3b-12: **Process:** ETOH.Corn Ethanol and Cellulosic Ethanol TandD Ethanol Transportation and Distribution RefStation; **Technology/subprocess:** Ethanol, Refilling Station; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 65 (13%); **Map name**: VMT.

(a) VOC

(b) SO$_x$

(c) Temporal
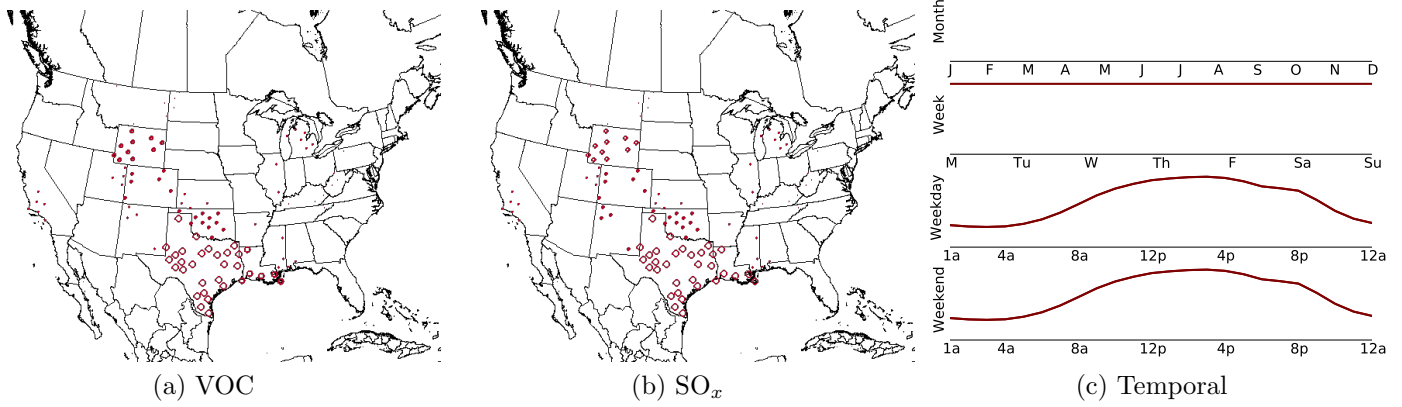
Figure S3b-13: **Process:** NG.asProcFuelNoncombustion; **Technology/subprocess:** NG, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 8 (1%), SO$_x$: 18 (8%); **Map name**: NGProcessing.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) SO$_x$

(d) Temporal

Figure S3b-14: **Process:** Ag Inputs.PhosphoricAcidProcessEmis; **Technology/subprocess:** Phosphoric Acid, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 2.2 (1%), PM$_{2.5}$: 1.6 (2%), SO$_x$: 13 (6%); **Map name**: PhosphoricAcid.

(a) PM$_{2.5}$

(b) NO$_x$

(c) Temporal
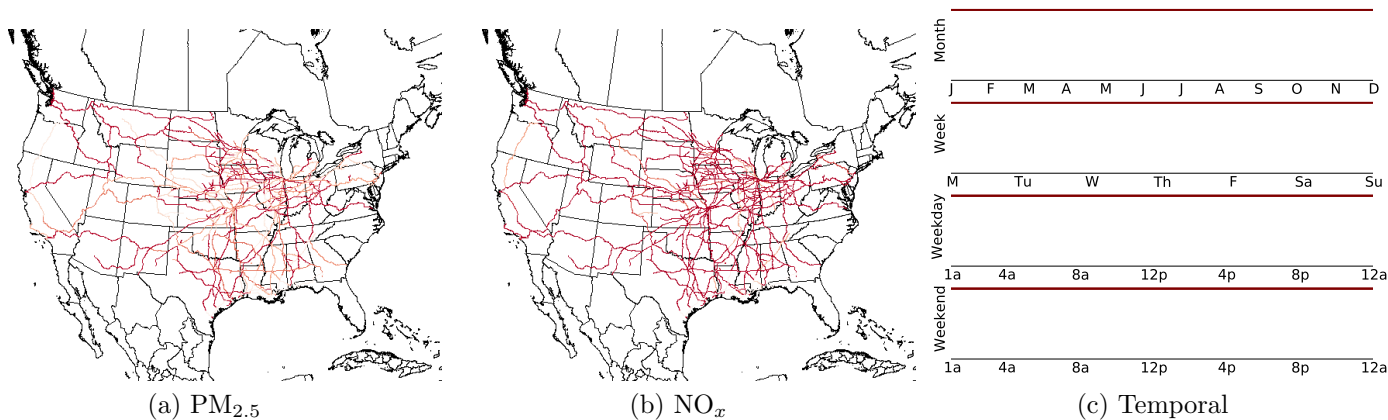
Figure S3b-15: **Process:** TandD.USETOHtrans Rail; **Technology/subprocess:** Locomotive, Diesel; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 0.89 (1%), NO$_x$: 32 (5%); **Map name**: CurrentBiorefineries TO OilRefineries RailNetwork.
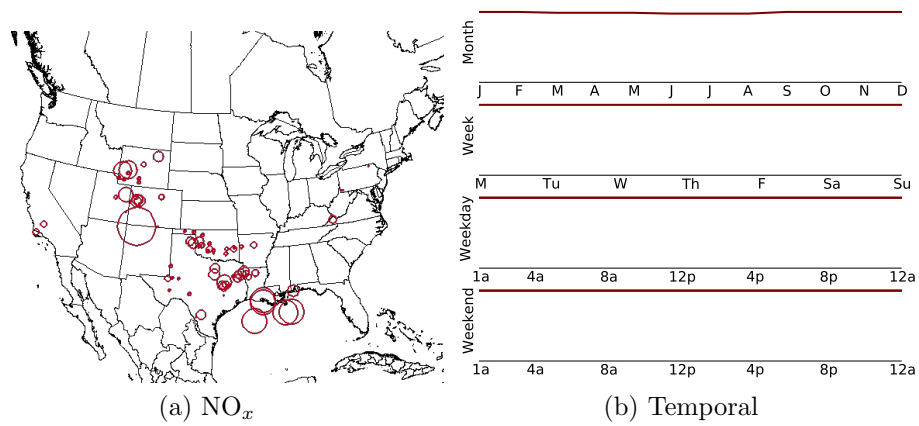

(a) NO$_x$

(b) Temporal
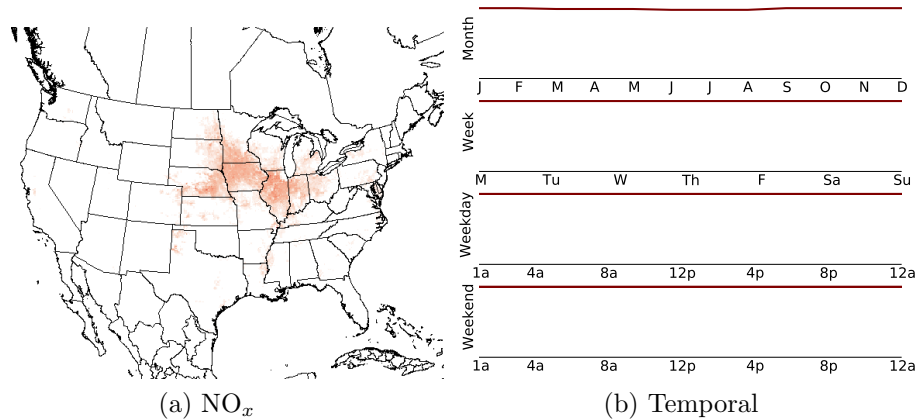
Figure S3b-16: **Process:** ETOH.CornFarming; **Technology/subprocess:** Natural Gas, Stationary Reciprocating Engine; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 28 (4%); **Map name**: FarmProduction-CornTotal.
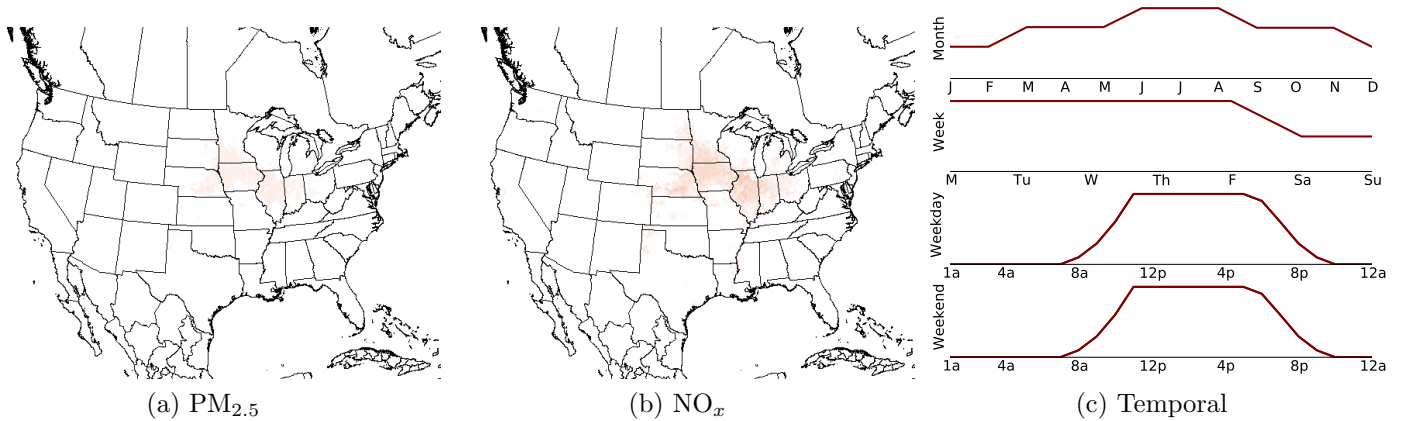
(a) PM$_{2.5}$ (b) NO$_x$ (c) Temporal

Figure S3b-17: **Process:** BD.SoybeanFarming; **Technology/subprocess:** Diesel Fuel, Farming Tractor; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: -1.2 (-2%), NO$_x$: -19 (-3%); **Map name**: Farm-ProductionCornTotal.
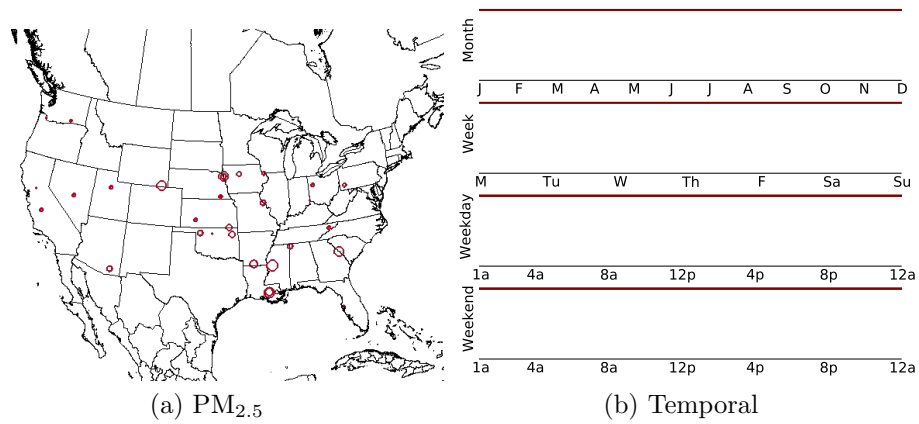

(a) NH$_3$ (b) VOC   (c) Temporal
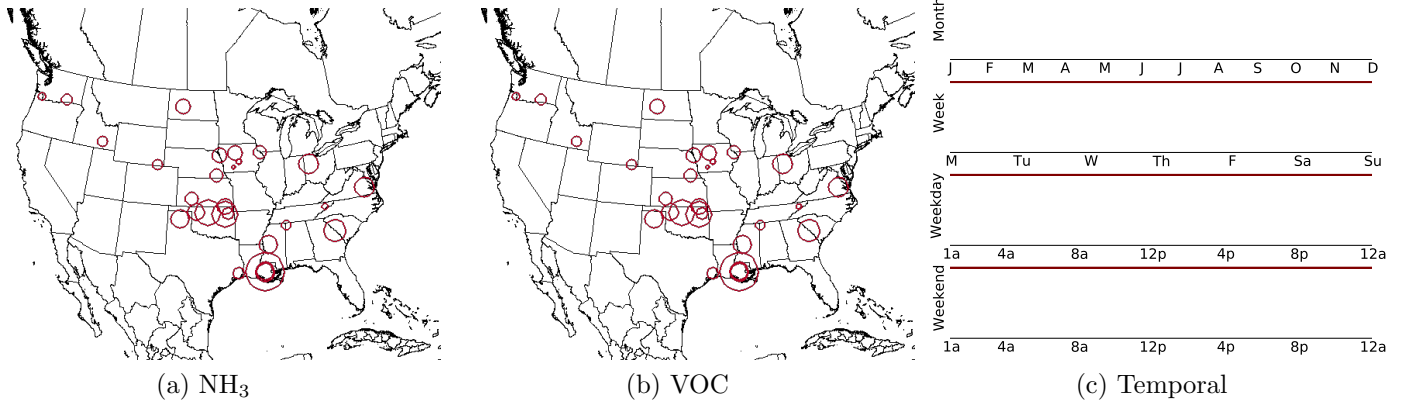
Figure S3b-18: **Process:** Ag Inputs.AmmoniaProcessEmis; **Technology/subprocess:** Ammonia, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** NH$_3$: 4 (1%), VOC: 18 (3%); **Map name**: Ammonia.

(a) VOC

(b) Temporal

Figure S3b-19: **Process:** ETOH.Corn Ethanol and Cellulosic Ethanol TandD Ethanol Transportation and Distribution Bulkterminal; **Technology/subprocess:** Ethanol, Bulk Terminal; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 33 (7%); **Map name**: VMT.

(a) PM$_{2.5}$

(b) NO$_x$

(c) Temporal
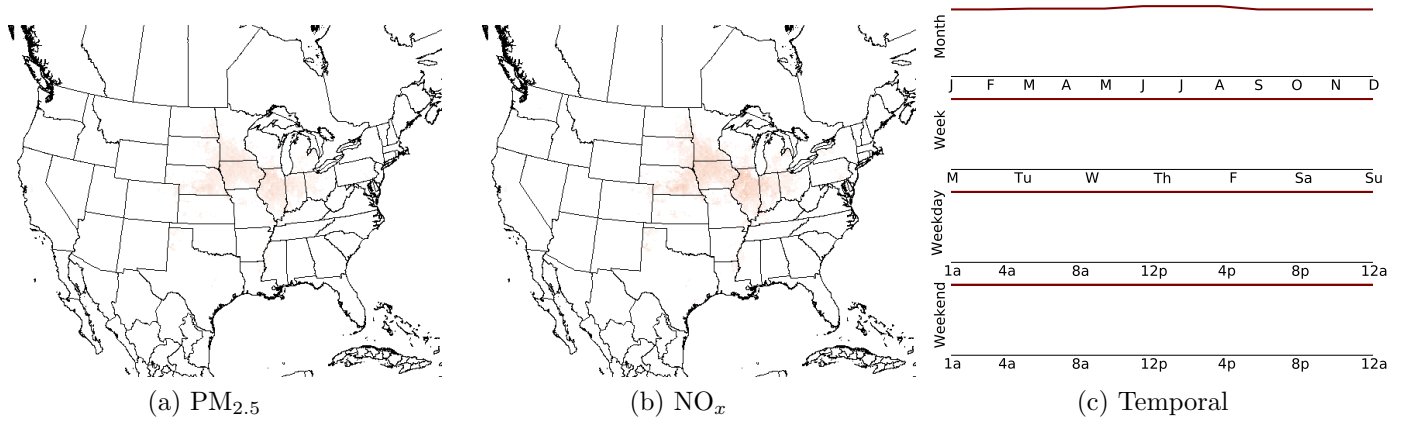
Figure S3b-20: **Process:** ETOH.CornFarming; **Technology/subprocess:** Diesel Fuel, Stationary Reciprocating Engine; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 0.95 (1%), NO$_x$: 12 (2%); **Map name**: FarmProductionCornTotal.

(a) NO$_x$ (b) Temporal

Figure S3b-21: **Process:** NG.asProcFuelRecovery; **Technology/subprocess:** Natural Gas, Stationary Reciprocating Engine; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 19 (3%); **Map name**: NaturalGasExtraction.

(a) PM$_{2.5}$ (b) NO$_x$ (c) Temporal

Figure S3b-22: **Process:** ETOH.CornFarming; **Technology/subprocess:** Gasoline, Farming Tractor; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 0.59 (1%), NO$_x$: 12 (2%); **Map name**: FarmProduction-CornTotal.

(a) NO$_x$ (b) Temporal
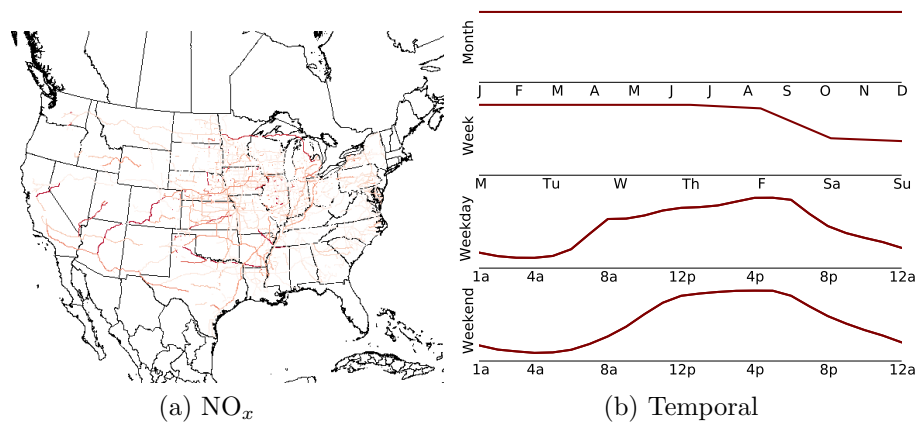
Figure S3b-23: **Process:** TandD.CornStack2Plant Truck; **Technology/subprocess:** Heavy Heavy-Duty Truck: grams per MMBtu, Diesel; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 6.8 (1%); **Map name**: FarmProductionCornTotal TO CurrentBiorefineries RoadNetwork.

(a) SO$_x$ (b) Temporal
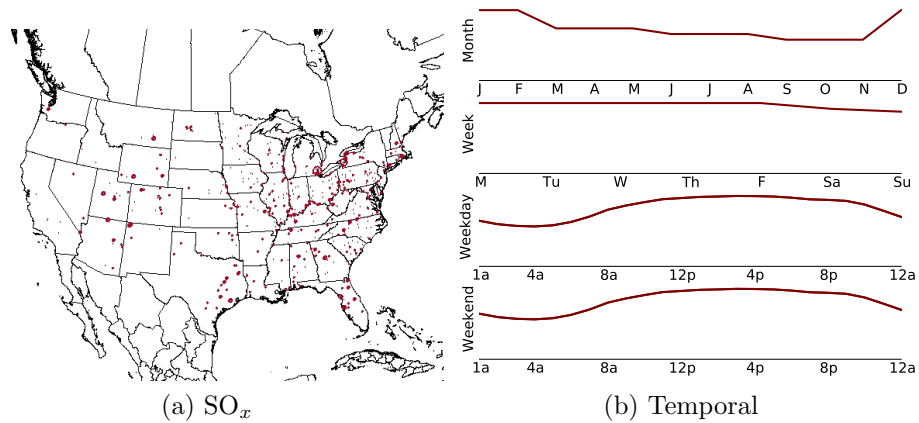
Figure S3b-24: **Process:** Electric.CoalFiredEF FOR AgInputs.CaCO3Total; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 5.7 (2%); **Map name**: ElectricGenerationCoal FOR VMT.

(a) PM$_{2.5}$

(b) Temporal

Figure S3b-25: **Process:** Ag Inputs.AmmoniumNitrateProcessEmis; **Technology/subprocess:** Ammonium Nitrate, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 0.92 (1%); **Map name**: AmmoniumNitrate.

(a) SO$_x$

(b) Temporal
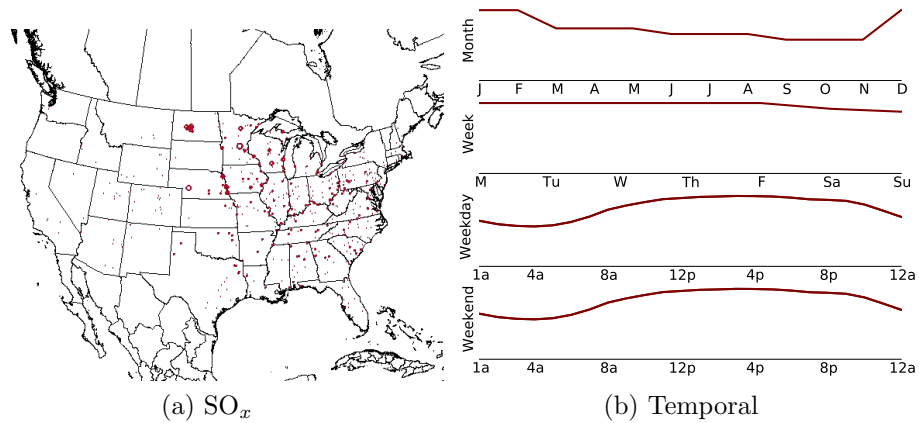
Figure S3b-26: **Process:** Electric.CoalFiredEF FOR ETOH.CornFarming; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 3.4 (1%); **Map name**: ElectricGenerationCoal FOR FarmProductionCornTotal.

(a) SO$_x$ (b) Temporal
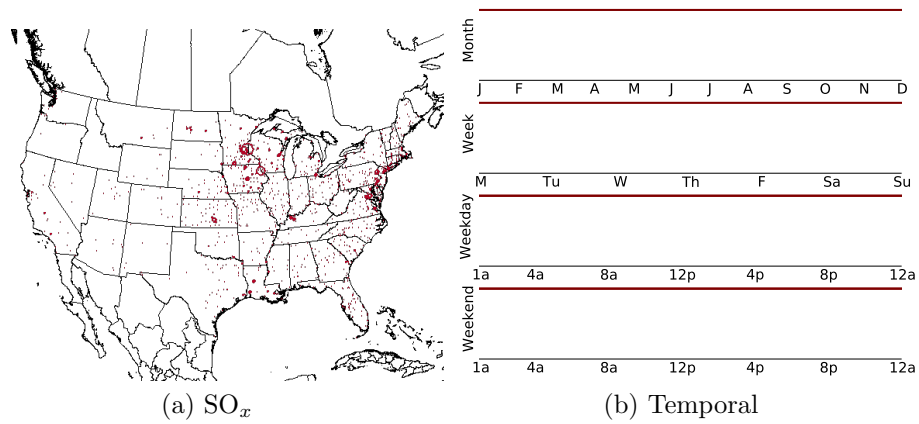
Figure S3b-27: **Process:** Electric.OilFiredEF FOR ETOH.DryMillEthanol; **Technology/subprocess:** EPA Electric Generation, Oil Boiler; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 3.1 (1%); **Map name**: ElectricGenerationOil FOR CurrentBiorefineries.

Supplemental Information 3c: Corn Stover Cellulosic Ethanol

A spatially and temporally explicit life cycle inventory of air pollutants from gasoline and ethanol in the United States

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This document contains maps of emissions contributions from those processes and pollutants that contribute more than 1% of life cycle total emissions. The amount of emissions is indicated by color intensity for line and area sources, and by rings for point sources, with the area of the ring being proportional to total emissions at each site. In all cases, red indicates positive emissions, and blue indicates negative emissions.

Figure S3c-1: Color scales for area and line maps below.

(a) CO

(b) PM$_{10}$

(c) VOC

(d) PM$_{2.5}$

(e) NO$_x$

(f) SO$_x$

(g) Temporal
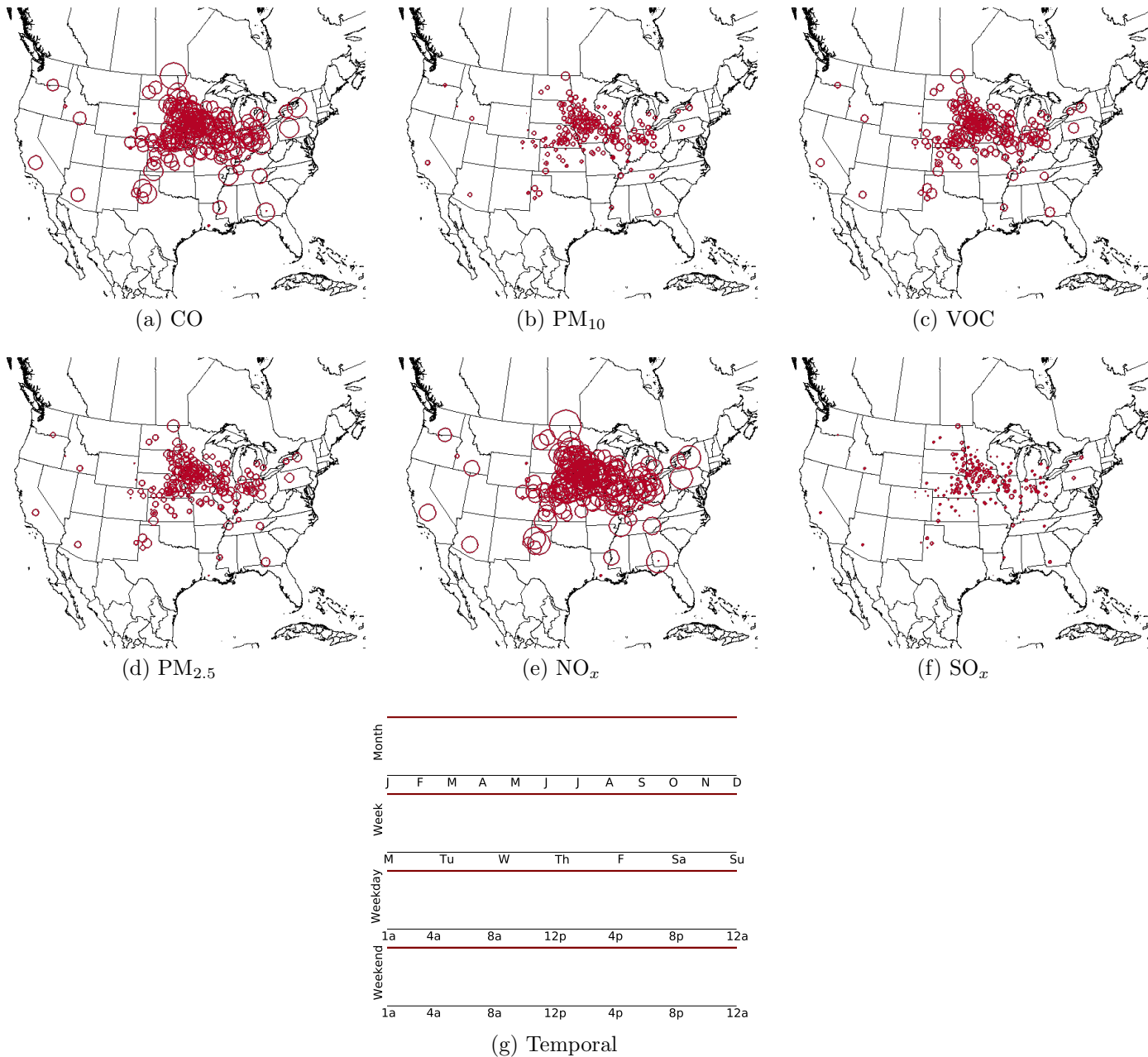
Figure S3c-2: **Process:** ETOH.StoverEthanolFermentation; **Technology/subprocess:** Corn Stover, Small Industrial Boiler; **Emissions: mg mile^{-1} (% life cycle total):** CO: 3.1e+02 (7%), PM$_{10}$: 52 (169%), VOC: 22 (5%), PM$_{2.5}$: 26 (79%), NO$_x$: 4.5e+02 (59%), SO$_x$: 17 (-12%); **Map name**: CurrentBiorefineries.
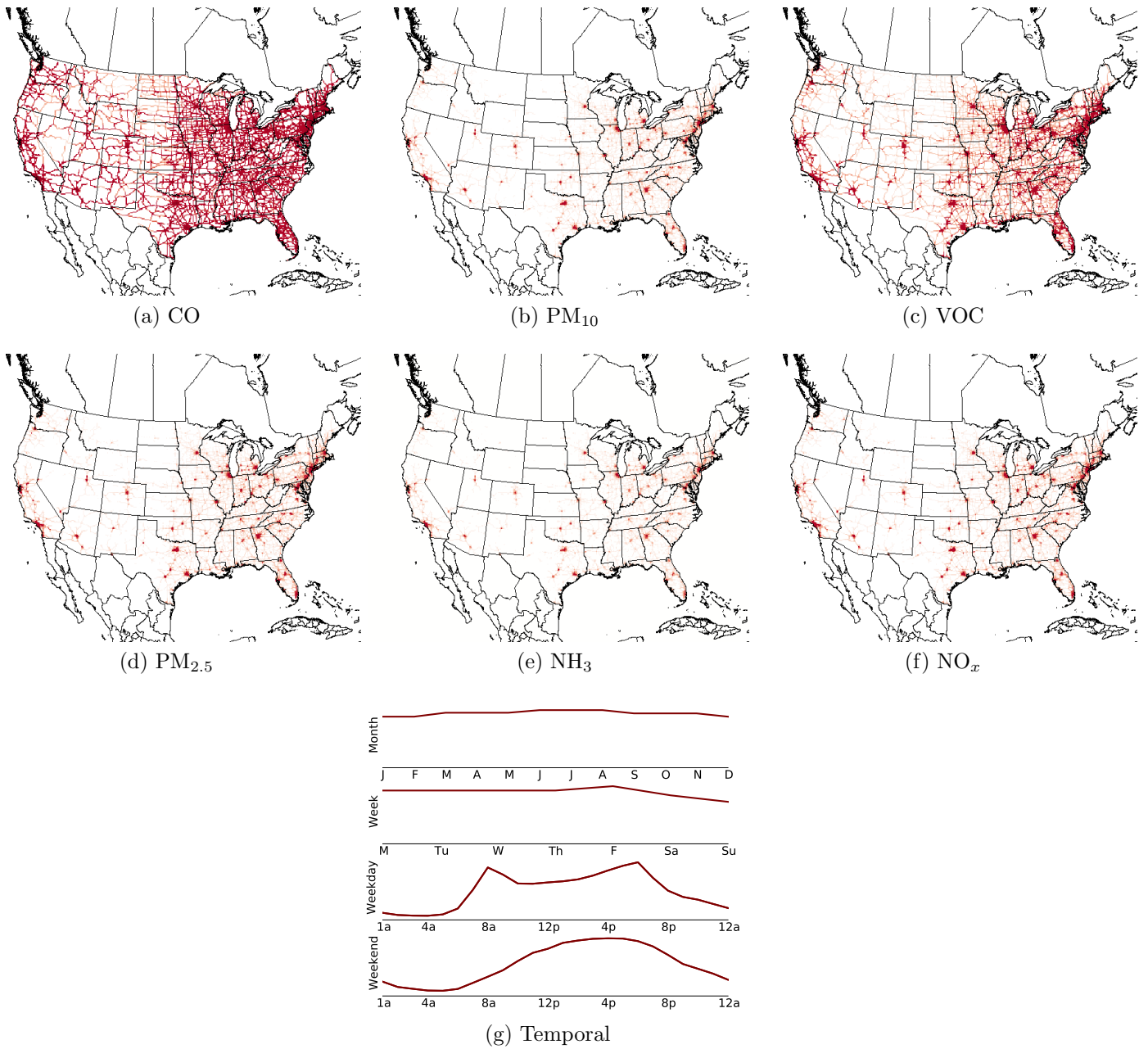
(a) CO

(b) PM$_{10}$

(c) VOC

(d) PM$_{2.5}$

(e) NH$_3$

(f) NO$_x$

(g) Temporal

Figure S3c-3: **Process:** Vehicles.Emissions; **Technology/subprocess:** Light Duty Vehicle, Gasoline - Exhaust; **Emissions: mg mile^{-1} (% life cycle total):** CO: 3.7e+03 (90%), PM$_{10}$: 29 (94%), VOC: 1.8e+02 (45%), PM$_{2.5}$: 15 (45%), NH$_3$: 50 (28%), NO$_x$: 1.4e+02 (18%); **Map name**: VMT.

(a) PM$_{2.5}$

(b) PM$_{10}$

(c) NH$_3$

(d) Temporal
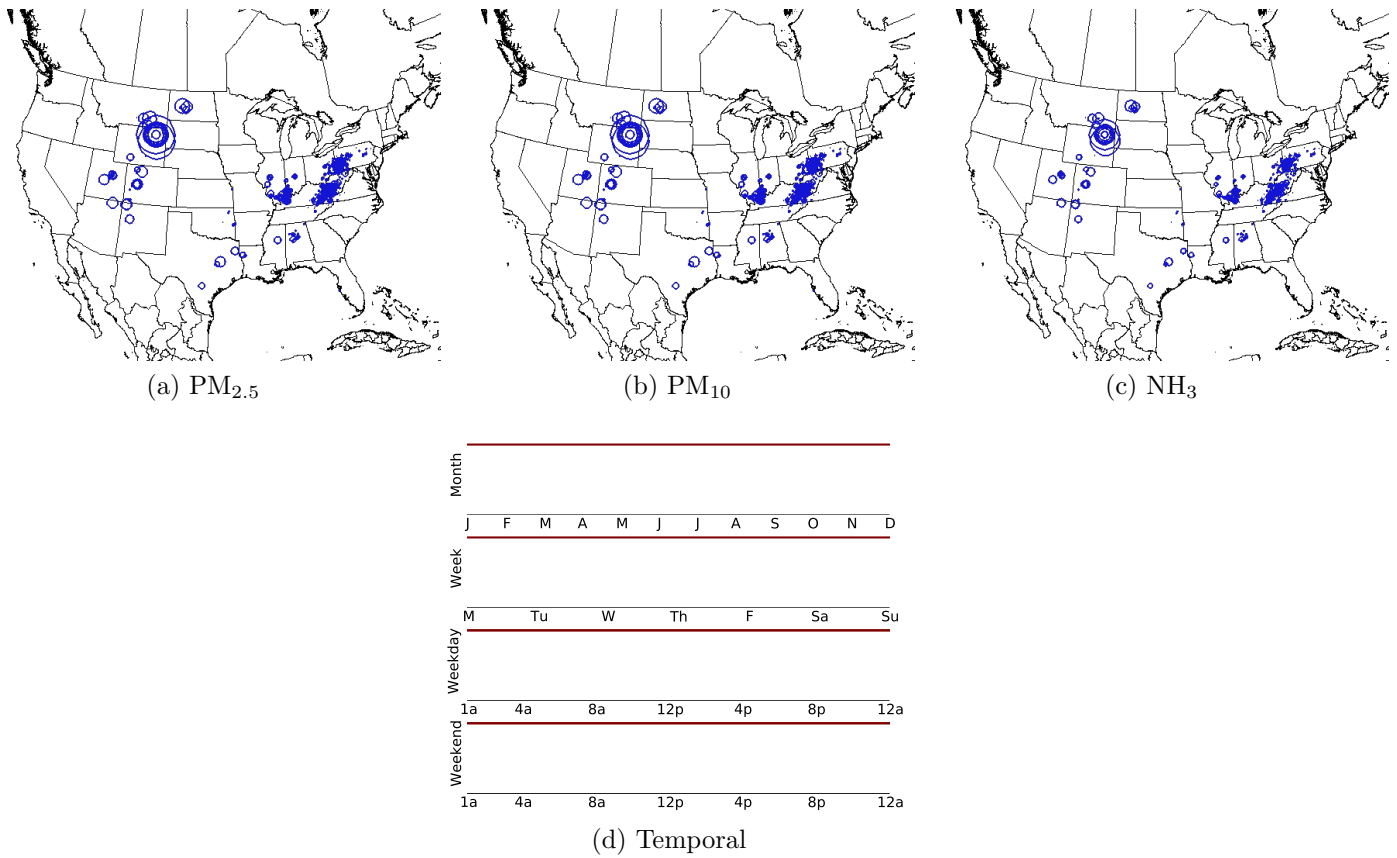
Figure S3c-4: **Process:** Coal.MiningNoncombustion; **Technology/subprocess:** Coal, Mining and Cleaning Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: -28 (-88%), PM$_{10}$: -1.2e+02 (-379%), NH$_3$: -3.4 (-1%); **Map name**: CoalMining.

(a) PM$_{2.5}$ (b) PM$_{10}$ (c) VOC

(d) Temporal
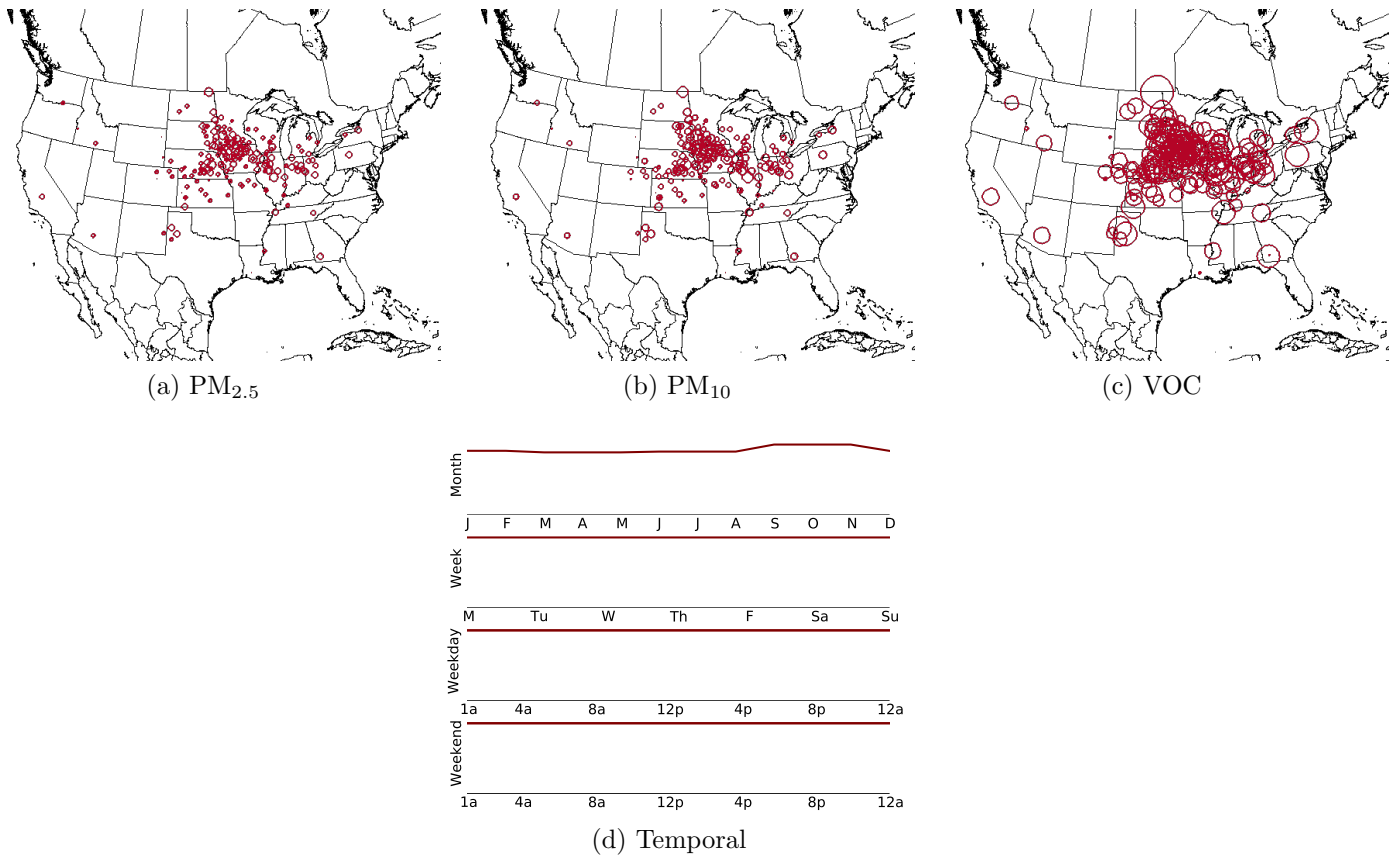
Figure S3c-5: **Process:** ETOH.StoverEthanolNoncombustion; **Technology/subprocess:** Ethanol, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 9.5 (29%), PM$_{10}$: 56 (183%), VOC: 73 (18%); **Map name**: CurrentBiorefineries.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) NO$_x$

(d) SO$_x$

(e) Temporal
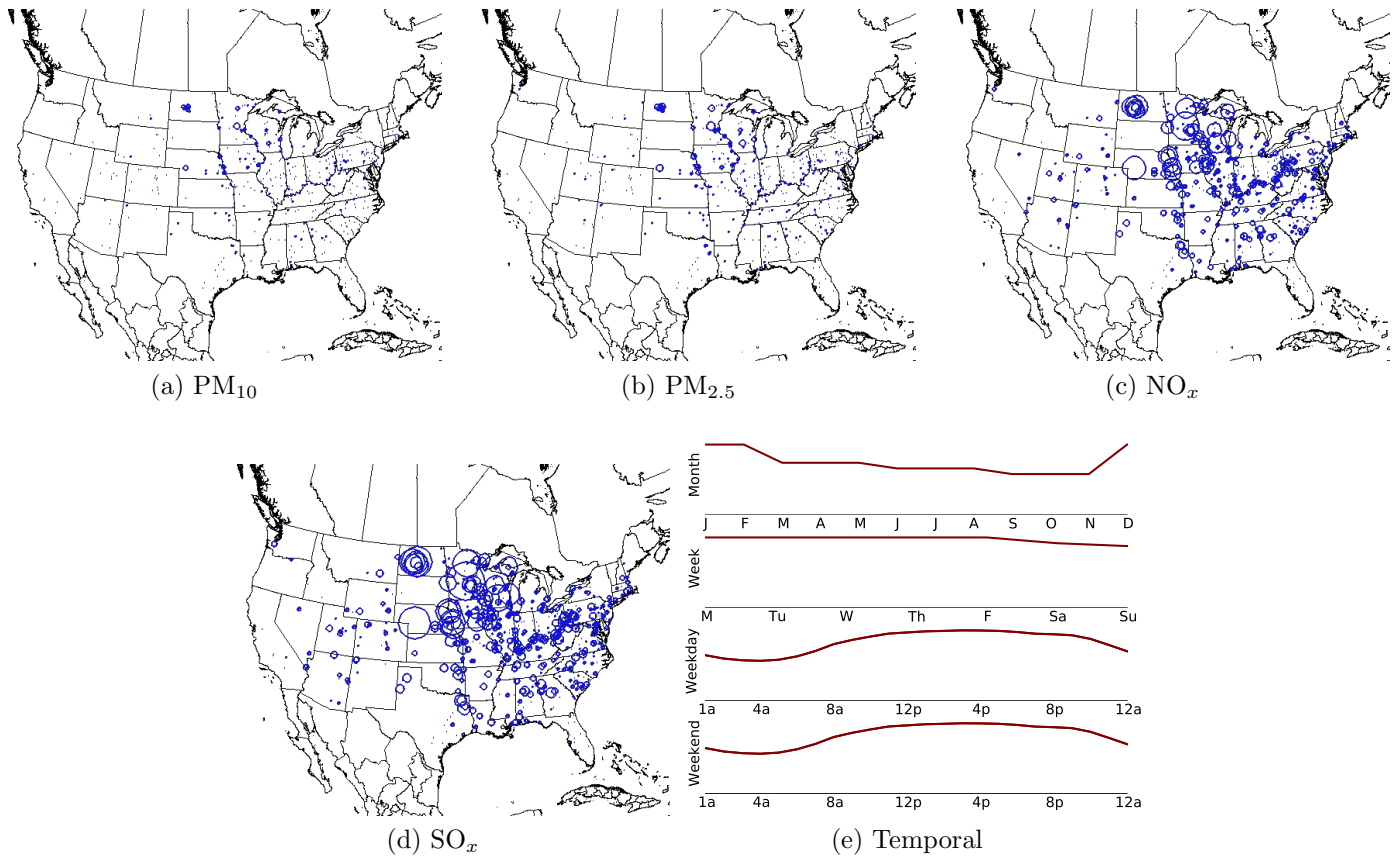
Figure S3c-6: **Process:** Electric.CoalFiredEF FOR ETOH.StoverFermentationElectricityCredits; **Technology/subprocess:** EPA Electric Generation, Coal Boiler; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: -5.1 (-16%), PM$_{2.5}$: -2.5 (-7%), NO$_x$: -70 (-9%), SO$_x$: -2e+02 (147%); **Map name**: ElectricGenerationCoal FOR CurrentBiorefineries.

(a) CO

(b) PM$_{10}$

(c) VOC

(d) PM$_{2.5}$

(e) SO$_x$

(f) NO$_x$

(g) Temporal
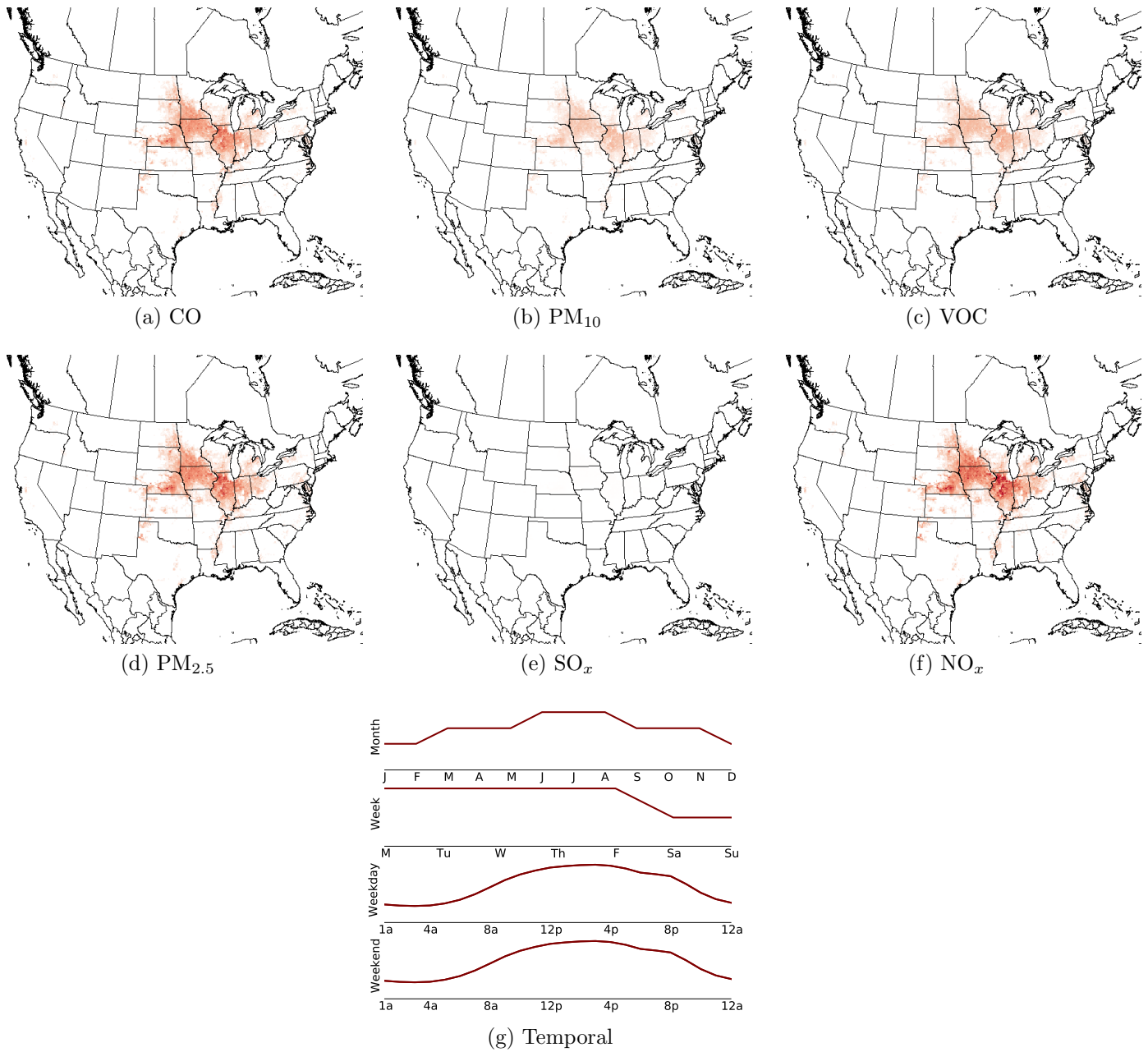
Figure S3c-7: **Process:** ETOH.StoverCollection; **Technology/subprocess:** Diesel Fuel, Farming Tractor; **Emissions: mg mile^{-1} (% life cycle total):** CO: 62 (1%), PM$_{10}$: 11 (34%), VOC: 12 (3%), PM$_{2.5}$: 9.5 (29%), SO$_x$: 1.4 (-1%), NO$_x$: 1.2e+02 (15%); **Map name**: FarmProductionCornTotal.

(a) NO$_x$ (b) NH$_3$ (c) Temporal
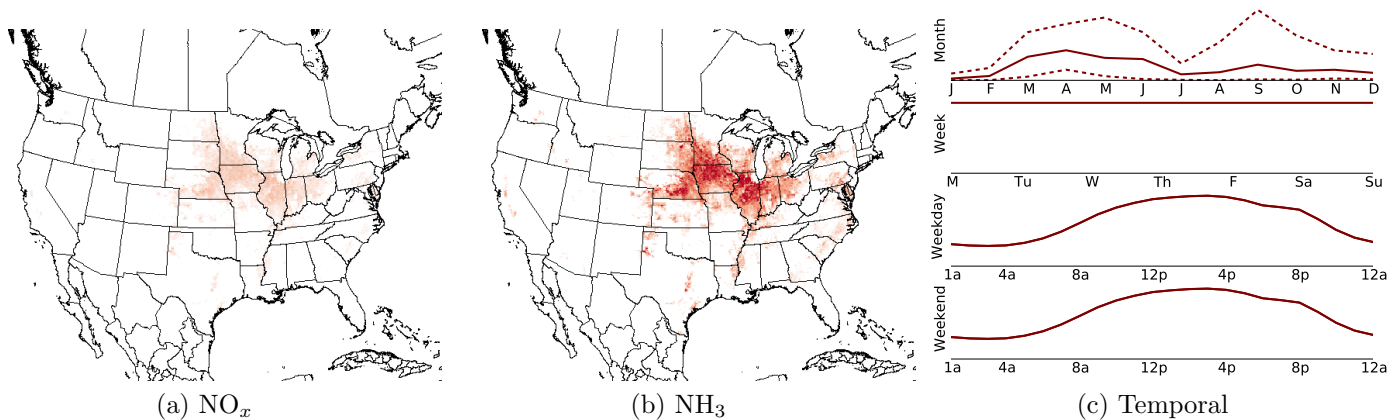
Figure S3c-8: **Process:** ETOH.StoverNFertilizerNitrification; **Technology/subprocess:** Fertilizer, Nitrification Emissions; **Emissions: mg mile^{-1} (% life cycle total):** NO$_x$: 45 (6%), NH$_3$: 1.2e+02 (71%); **Map name**: FarmAreaCornTotal.

(a) SO$_x$ (b) Temporal
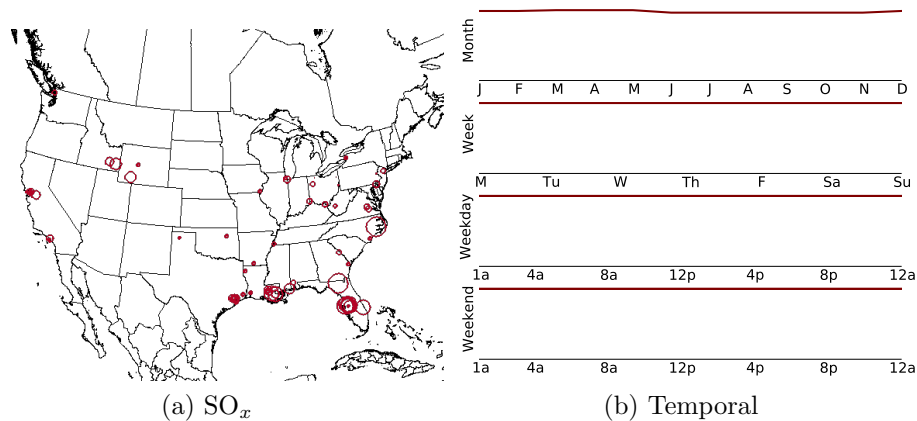
Figure S3c-9: **Process:** Ag Inputs.SulfuricAcidProcessEmis; **Technology/subprocess:** Sulfuric Acid, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: 39 (-29%); **Map name**: SulfuricAcid.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) NO$_x$

(d) SO$_x$

(e) Temporal
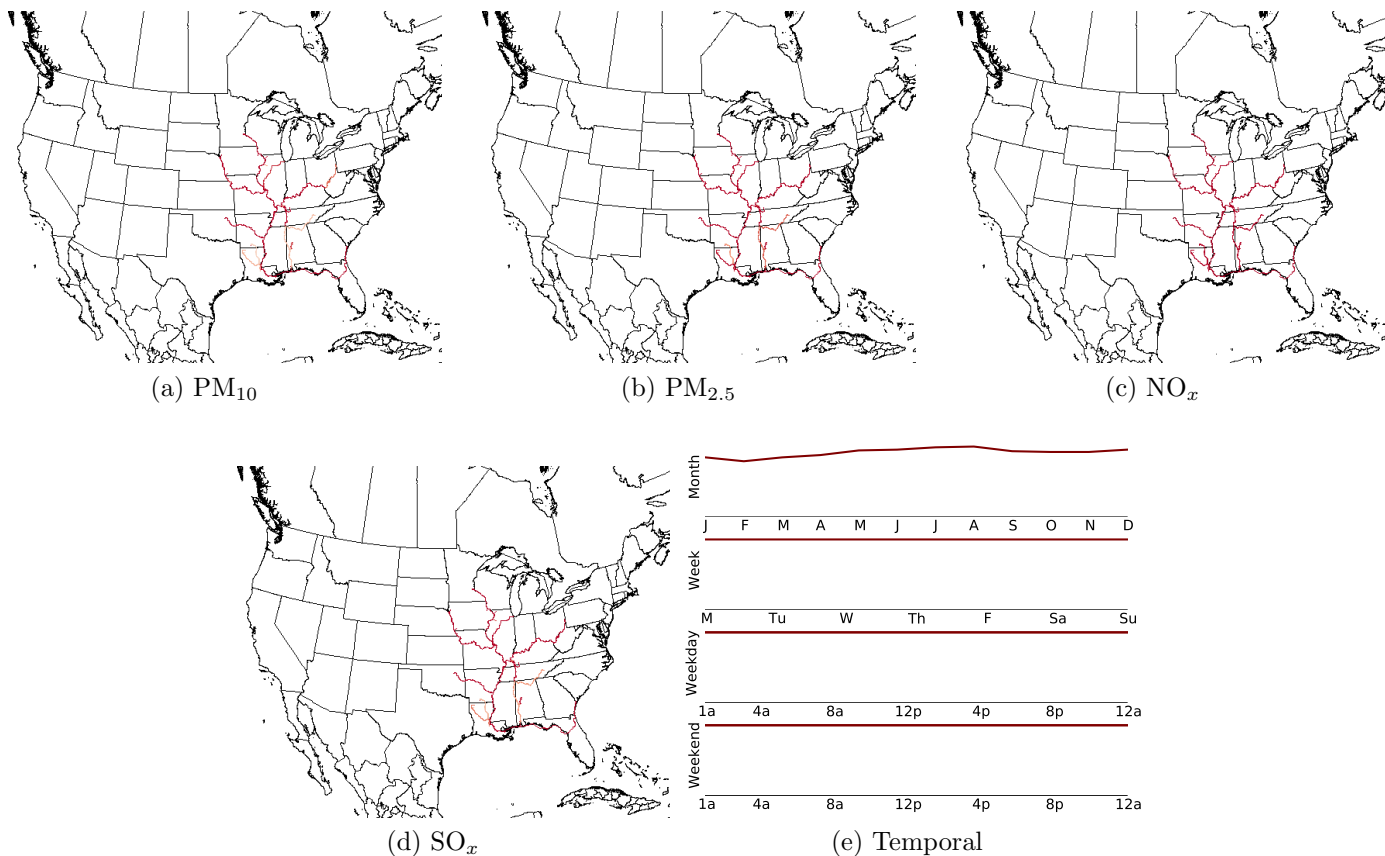
Figure S3c-10: **Process:** TandD.USETOHtrans Barge; **Technology/subprocess:** Barge, Residual Oil; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 1.3 (4%), PM$_{2.5}$: 1.2 (3%), NO$_x$: 35 (4%), SO$_x$: 8.4 (-6%); **Map name**: CurrentBiorefineries TO OilRefineries WaterNetwork.
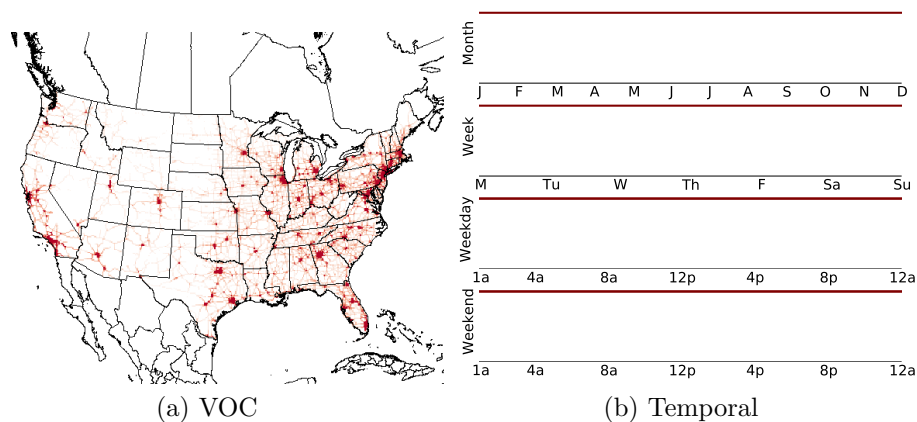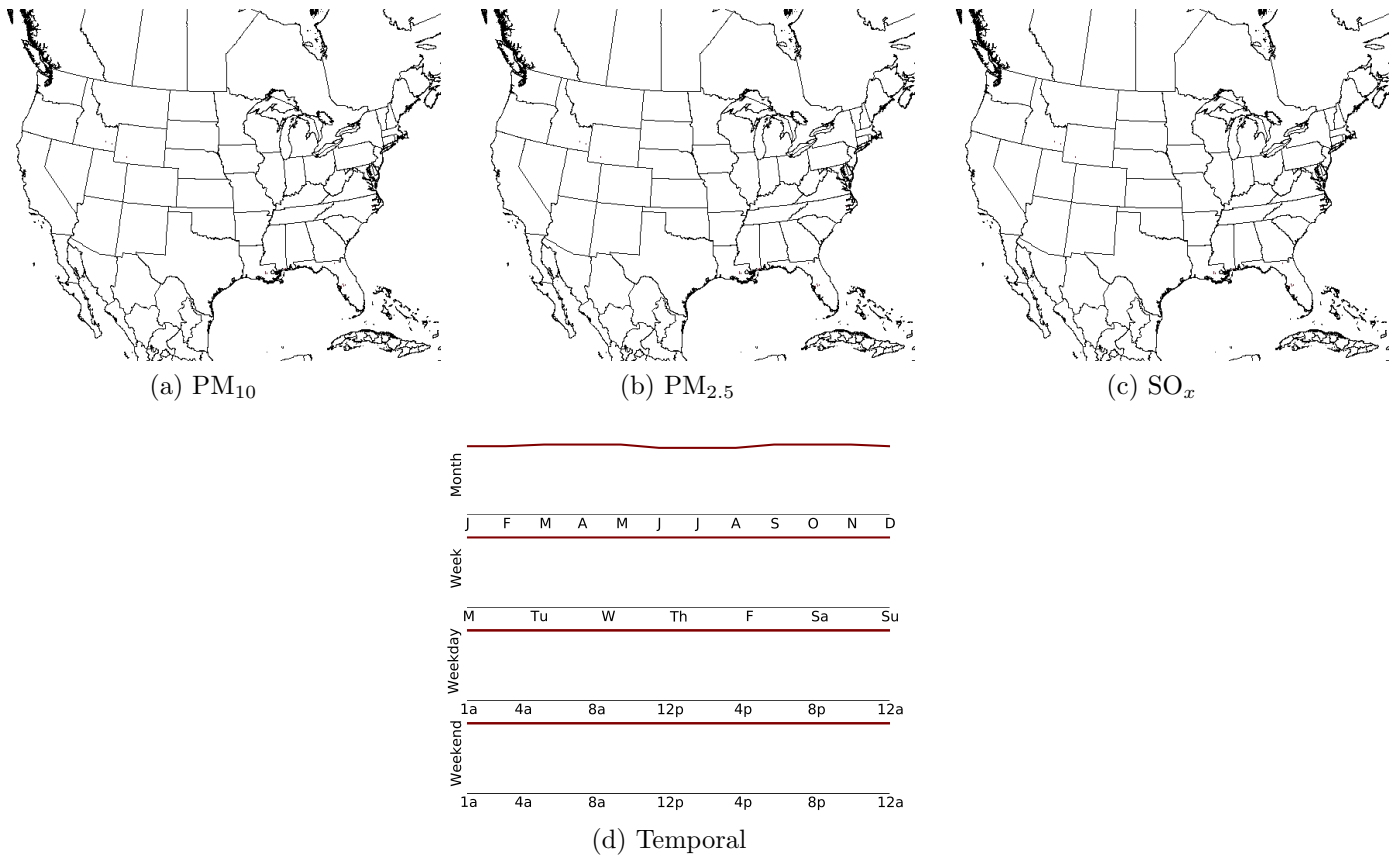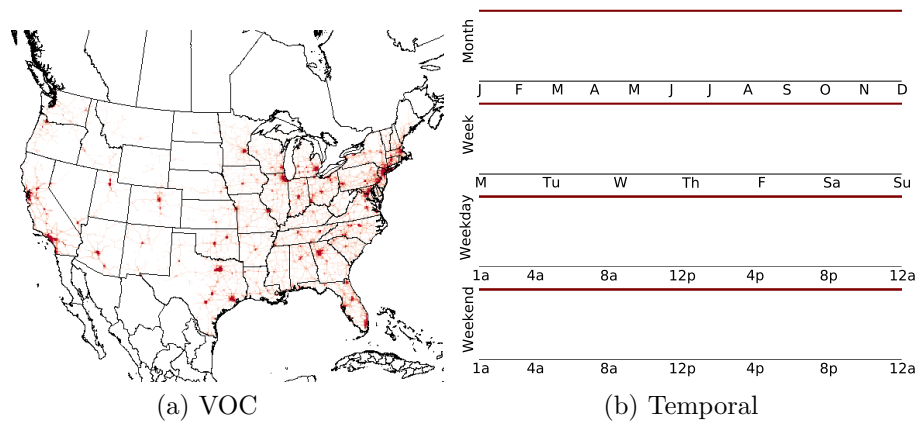

(a) VOC

(b) Temporal

Figure S3c-11: **Process:** ETOH.Corn Ethanol and Cellulosic Ethanol TandD Ethanol Transportation and Distribution RefStation; **Technology/subprocess:** Ethanol, Refilling Station; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 65 (16%); **Map name**: VMT.

(a) PM$_{10}$

(b) PM$_{2.5}$

(c) NO$_x$

(d) Temporal
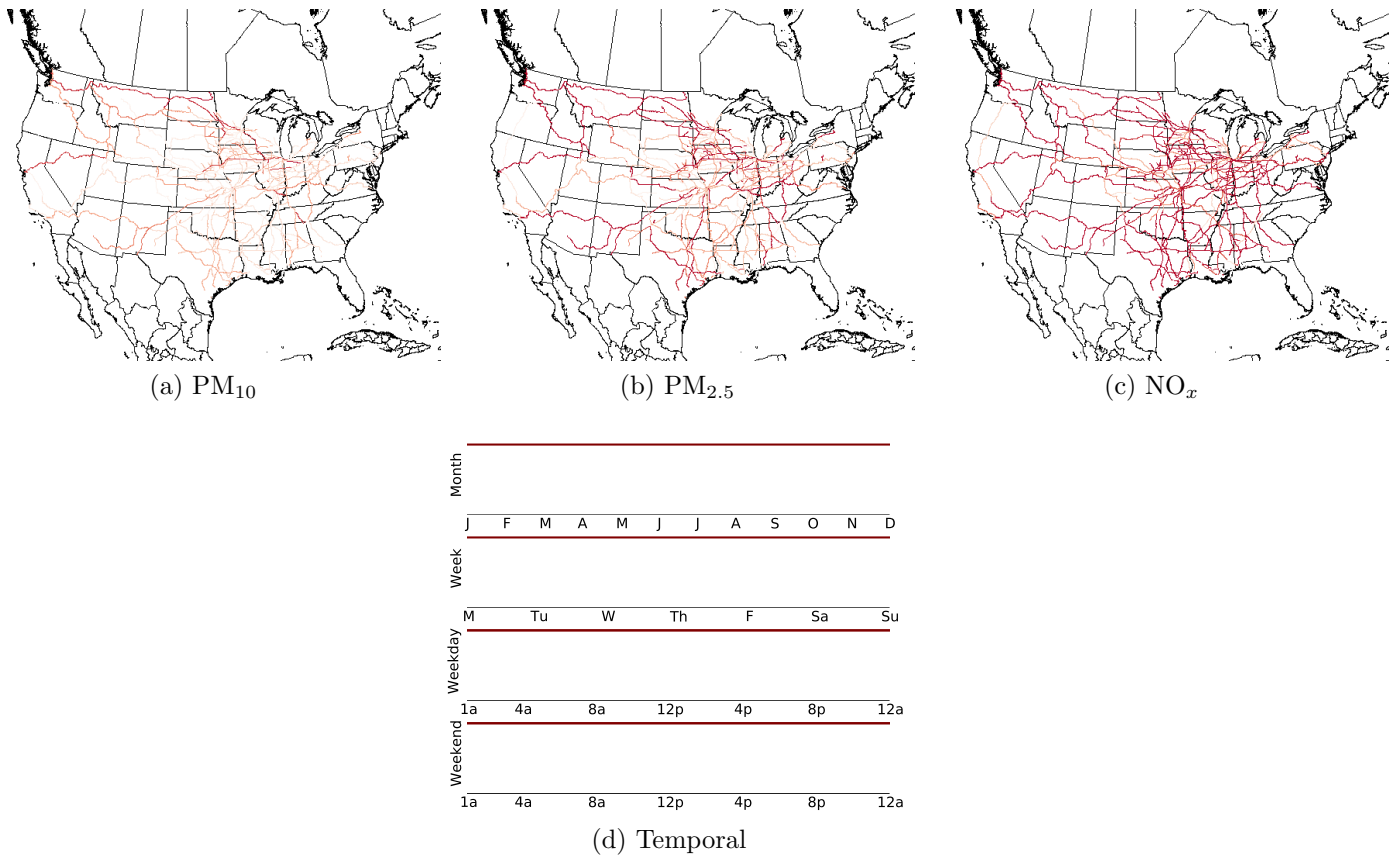
Figure S3c-12: **Process:** TandD.USETOHtrans Rail; **Technology/subprocess:** Locomotive, Diesel; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 0.92 (3%), PM$_{2.5}$: 0.89 (2%), NO$_x$: 32 (4%); **Map name**: CurrentBiorefineries TO OilRefineries RailNetwork.

(a) PM$_{10}$ (b) PM$_{2.5}$ (c) SO$_x$

(d) Temporal

Figure S3c-13: **Process:** Ag Inputs.PhosphoricAcidProcessEmis; **Technology/subprocess:** Phosphoric Acid, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 1 (3%), PM$_{2.5}$: 0.78 (2%), SO$_x$: 7 (-5%); **Map name**: PhosphoricAcid.

(a) VOC   (b) Temporal

Figure S3c-14: **Process:** ETOH.Corn Ethanol and Cellulosic Ethanol TandD Ethanol Transportation and Distribution Bulkterminal; **Technology/subprocess:** Ethanol, Bulk Terminal; **Emissions: mg mile^{-1} (% life cycle total):** VOC: 33 (8%); **Map name**: VMT.
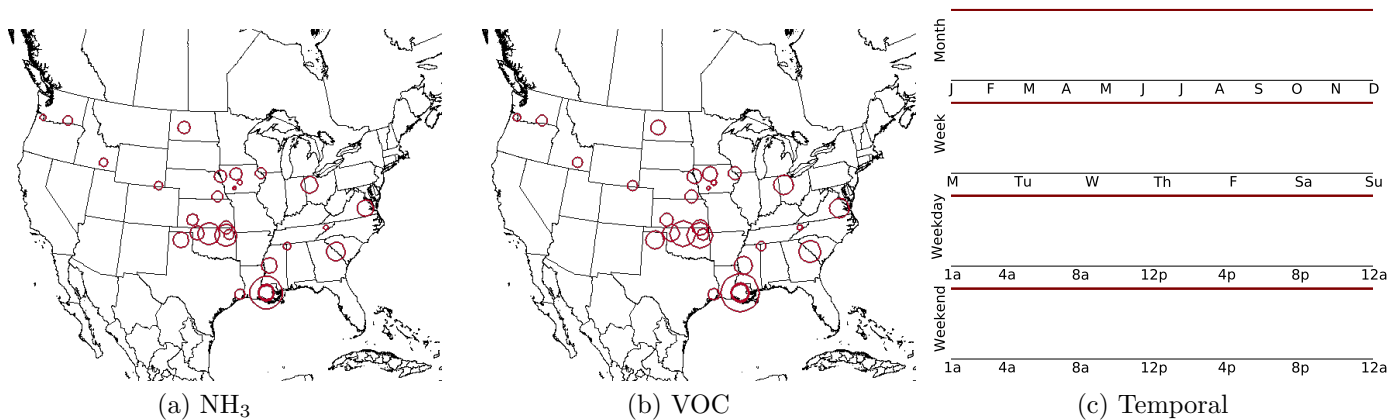
(a) NH₃ (b) VOC (c) Temporal

Figure S3c-15: **Process:** Ag Inputs.AmmoniaProcessEmis; **Technology/subprocess:** Ammonia, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** NH$_3$: 2 (1%), VOC: 9.3 (2%); **Map name**: Ammonia.
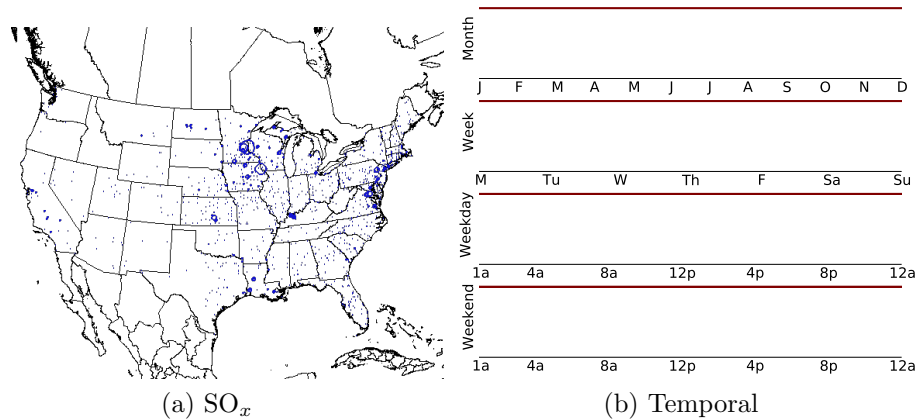

(a) SO$_x$ (b) Temporal

Figure S3c-16: **Process:** Electric.OilFiredEF FOR ETOH.StoverFermentationElectricityCredits; **Technology/subprocess:** EPA Electric Generation, Oil Boiler; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: -8.8 (6%); **Map name**: ElectricGenerationOil FOR CurrentBiorefineries.
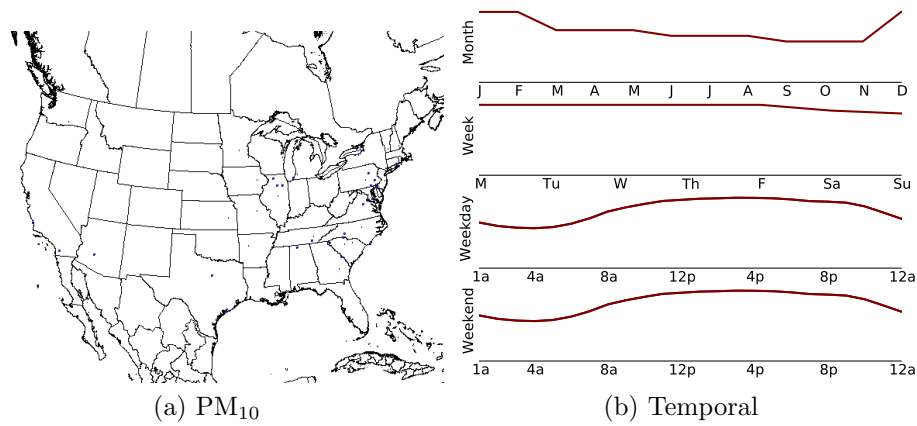
(a) PM$_{10}$ (b) Temporal

Figure S3c-17: **Process:** Uranium.TotalLWRElectricityProduction; **Technology/subprocess:** Undefined, Using Default SCC; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: -0.49 (-1%); **Map name**: ElectricGenerationNuclear.
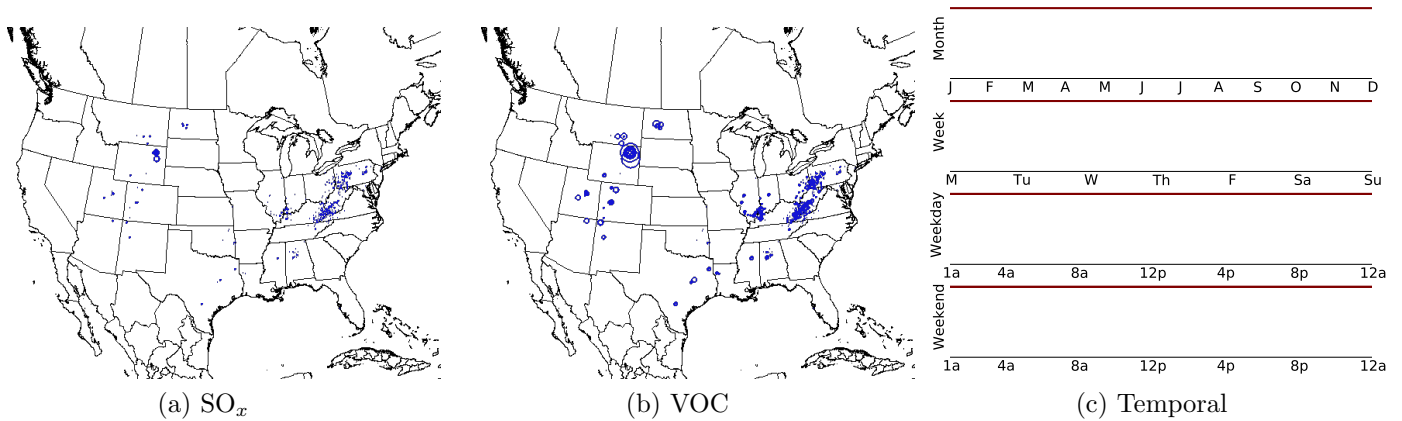

(a) SO$_x$ (b) VOC   (c) Temporal

Figure S3c-18: **Process:** Coal.CleaningNoncombustion; **Technology/subprocess:** Coal, Mining and Cleaning Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: -3.8 (2%), VOC: -4.7 (-1%); **Map name**: CoalMining.
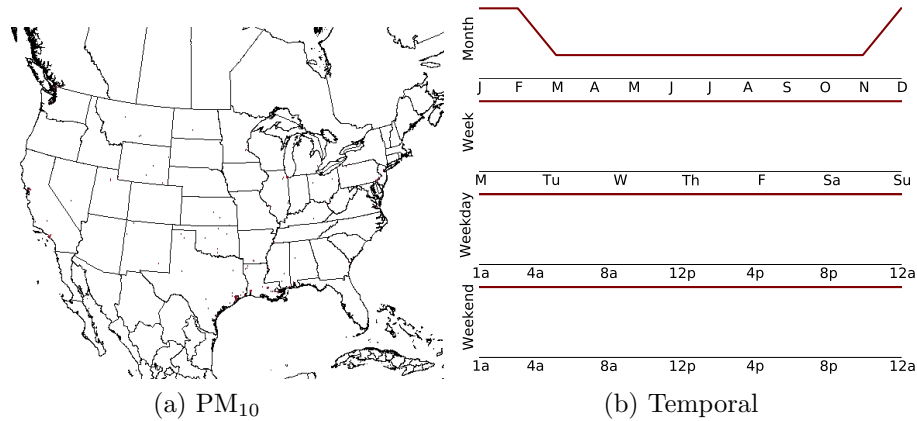

(a) PM$_{10}$ (b) Temporal

Figure S3c-19: **Process:** Petroleum.ConventionalDieselRefining; **Technology/subprocess:** Pet Coke, Industrial Boiler; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{10}$: 0.43 (1%); **Map name**: OilRefineries.

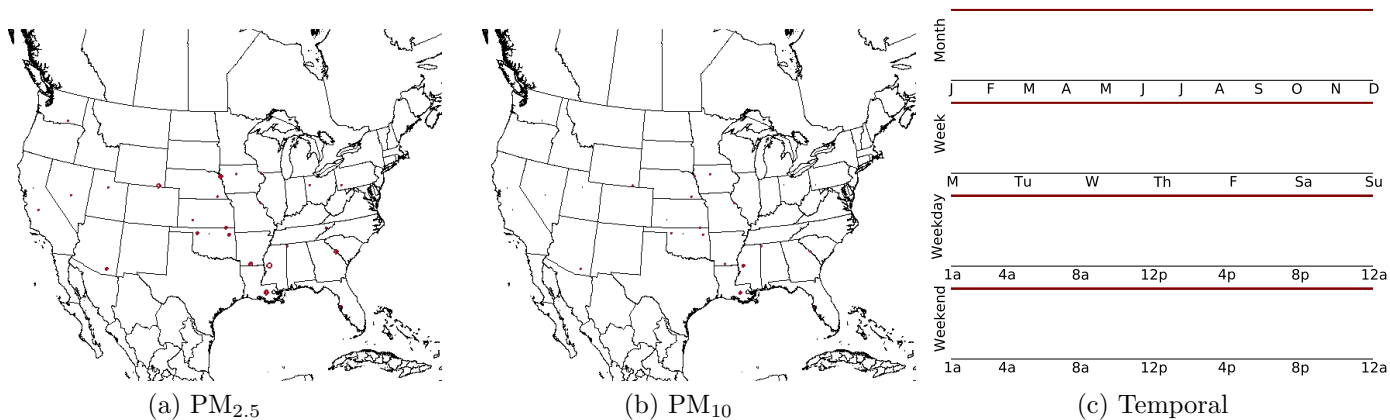(a) PM$_{2.5}$ (b) PM$_{10}$ (c) Temporal

Figure S3c-20: **Process:** Ag Inputs.AmmoniumNitrateProcessEmis; **Technology/subprocess:** Ammonium Nitrate, Production Process Emissions; **Emissions: mg mile^{-1} (% life cycle total):** PM$_{2.5}$: 0.4 (1%), PM$_{10}$: 0.5 (1%); **Map name**: AmmoniumNitrate.

(a) SO$_x$ (b) Temporal
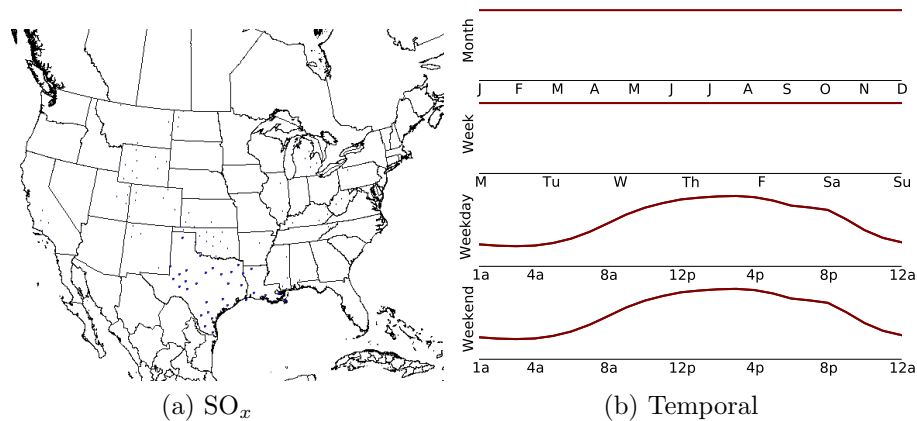
Figure S3c-21: **Process:** NG.asProcFuelNoncombustion; **Technology/subprocess:** NG, Noncombustion; **Emissions: mg mile^{-1} (% life cycle total):** SO$_x$: -2.3 (1%); **Map name**: NGProcessing.