Historical gaseous and primary aerosol emissions in the United States from 1990-2010

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Abstract

An accurate description of emissions is crucial for model simulations to reproduce and interpret observed phenomena over extended time periods. In this study, we used an approach based on activity data to develop a consistent series of spatially resolved emissions in the United States from 1990 to 2010. The state-level anthropogenic emissions of SO_2, NO_x, CO, NMVOC, NH_3, PM_{10} and $PM_{2.5}$ for a total of 49 sectors were estimated based on several long-term databases containing information about activities and emission controls. Activity data for energy-related stationary sources were derived from the State Energy Data System. Corresponding emission factors reflecting implemented emission controls were calculated back from the national emission inventory (NEI) for seven years (i.e., 1990, 1995, 1996, 1999, 2001, 2002 and 2005), and constrained by the AP-42 (US EPA's Compilation of Air Pollutant Emissions Factors) dataset. Activity data for mobile sources including different types of highway vehicles and non-highway equipments were obtained from highway statistics reported by the

Federal Highway Administration. The trends in emission factors for highway mobile source were informed by the 2011 National Transportation Statistics. Emissions for all non-energy related sources were either scaled by the growth ratio of activity indicators or adjusted based on the NEI trends report.

Because of the strengthened control efforts, particularly for the power sector and mobile sources, emissions of all pollutants except NH_3 were reduced by half over the last two decades. The emission trends developed in this study are comparable with the NEI trend report and EDGAR (Emissions Database for Global Atmospheric Research) data, but better constrained by trends in activity data. Reductions in SO_2, NO_x, CO and EC (speciation of $PM_{2.5}$ by SMOKE, Sparse Matrix Operator Kernel Emissions) emissions agree well with the observed changes in ambient SO_2, NO_2, CO and EC concentrations, suggesting that the various controls on emissions implemented over the last two decades are well represented in the emission inventories developed in this study. These inventories were processed by SMOKE and are now ready to be used for regional chemistry transport model simulations over the 1990-2010 period.

Key words

Emission inventory, United States, emission trends, gaseous, primary aerosol

1. Introduction

Quantification of long-term historic emissions is necessary to assess their impacts on atmospheric chemistry and composition through model simulation and analysis. Recent studies suggest that the observed transition from decadal "dimming" to "brightening" during the 1990s in the continental United States was strongly influenced by the reductions in anthropogenic emissions of aerosol precursors (Streets et al., 2006; Stern, 2006; Wild, 2009). SO_2 and NO_x emissions in particular were required to be reduced by ten and two million tons respectively from their 1980 levels by Title IV of the U.S. Clean Air Act Amendments enacted in the year of 1990. It is believed that such reductions have had considerable effects on anthropogenic aerosol loading and regional radiation budgets over the past two decades. Regional chemistry or climate models are good tools for improving our understanding of the role of aerosols in the decadal changes of solar radiation. However, using such simulations to reproduce and interpret the observed phenomena requires knowledge of changes in the magnitude as well as the spatial and temporal patterns of emissions (Streets et al., 2003).

Some studies have generated global emissions over extended time periods (Lamarque et al., 2010; Smith et al., 2011). One example is the Emissions Database for Global Atmospheric Research (EDGAR) (European Commission, 2011). Because these inventories typically are resolved at the country or region level, they cannot adequately support regional-scale chemistry or climate model simulations over areas such as the continental United States. The National Emission Inventory (NEI) data for specific years prepared by the U.S. EPA is a comprehensive and detailed estimate of regional emissions based on detailed information provided by state, local and tribal air agencies for sources in their jurisdictions, it is hereafter referred to as the "NEI data". Because of the continual development of emission-estimation methodology, the inconsistency between different years of NEI data is an obstacle for their use in long-term air quality model simulations. For example, on-road NOx emission estimated from MOVES (www.epa.gov/otaq/models/moves/index.htm) used in 2005 NEI (current newest version 4.2) is much higher than that estimated from its predecessor model MOBILE6 which were used in previous NEI (Lindhjem et al., 2012; McDonald et al., 2012). Besides, when performing long-term analysis of decadal-scale variations and trends, accuracy of trends is as important as accuracy of absolute values. Though national-level sector-based emissions for each year are available from the NEI trends report (U.S. EPA, 2000, http://www.epa.gov/ttnchie1/trends/, referred to as "NEI trends" in this paper), the necessary harmonization of these coarser data with the detailed information available in the periodic NEI data as well as the interpolation for years for which no detailed NEI data is available is challenging (Hogrefe et al., 2009).

In general, emissions from a certain source are calculated by using a specific activity indicator (e.g., fuel consumption) multiplied by source-specific emission factor. Changes of emissions over a period of time can be caused by changes in both activity and emission factors due to emission controls, and such changes are usually fairly well constrained over long periods (Lamarque et al., 2010). For example, the State Energy Data System (http://www.eia.gov/state/seds/) provides a long historic record back to 1960 about the state-level energy use by broad energy-related sectors (i.e., combustion in electric power, industrial, domestic, transportation) which account for up to 90% of total emissions of SO_2 and NO_x (the major two species associated with the reduction in anthropogenic aerosol loading) in the United States. Therefore, the approach of scaling the emissions based on the changes in activities and controls would be a good choice to generate a consistent series of emissions particularly for such extended time periods.

To support multi-decadal regional-scale air quality simulations, we developed a consistent series of spatially resolved emission inventories (generated at the county level in SMOKE format, Sparse

Matrix Operator Kernel Emissions, http://www.smoke-model.org/data.cfm) in the United States from 1990 to 2010, by using an approach based on several long-term databases containing information about activity data and emission controls. Our goal is to have a single consistent methodology across the 20 year period for estimating the primary criteria pollutants for all the major sectors in the NEI. This work is a significant improvement over the NEI data and the NEI trends reports because our estimates capture the annual changes in emissions using a consistent methodology for each sector and we remove artificial step changes found in the NEI and in the trends data due to changes in methods. The state-level anthropogenic emissions of SO_2, NO_x, CO, NMVOC, NH_3, PM_{10} and $PM_{2.5}$ were estimated. In addition, trends of SO_2, NO_x, CO and EC (speciation of $PM_{2.5}$ by SMOKE) emissions from 1990 to 2010 were compared with changes in observed ambient SO_2, NO_2, CO and EC concentrations.

2. Method

2.1 Development of 1990-2010 emission inventories

The U.S. EPA National Emission Inventory (NEI) data are considered as the most comprehensive and detailed estimates of pollution emissions in the United States and have been widely used in modeling studies. Therefore, NEI data serves as the primary reference database in this study. Specifically, this study uses the most recent NEI data as reference and then scales emissions up or down for the other years based on the trends of activity data and emission controls over the entire period. Seven years of detailed NEI data were collected, including those developed for the more recent years of 1999, 2001, 2002 and 2005 which could be directly download from the EPA website (http://www.epa.gov/ttn/chief/, the 2008 NEI was not yet ready for SMOKE processing when our study of inventory development was initiated in 2011) and the three earlier years of 1990, 1995 and 1996 which were developed for previous studies (U.S. EPA, 1993; Adelman and Houyoux, 2001).

The approach we used to develop the long-term emission inventory is given in Figure 1.

First, to better organize each sector, all point, area and mobile emission sources (obtained from individual files in NEI data) were combined into three major groups (i.e. energy-related stationary sources, mobile sources, non-energy related sources) with 49 subsectors based on the SCC (Source Classification Codes). Details about the combination can be found in Table S1 of the supplementary material. All sectors were aggregated at the state level for trend purposes. The 2005 county-level NEI data was used as the reference for most sectors. The 2002 county-level NEI data was used as the reference for some sectors for which the 2005 NEI data was missing (e.g. aircraft) or inconsistent (for example, the on-road NO_x emission in 2005 NEI is significantly higher than that reported in NEI trends due to the methodology change from MOBILE to MOVES. However, mobile emission estimates by MOVES were unavailable for previous 20 years back to 1990s. Recent analysis by McDonald et al. (2012) suggests that overall MOBILE6 estimates were closer to EDGAR than MOVES only except for the past few years. For the purpose of this study, we selected the most recent NEI data which were based on MOBILE6 (i.e. 2002 NEI, instead of 2005 NEI) as the reference for on-road sector.

Additionally, since all sectors have noticeable contributions to total emissions of one or more pollutants (as seen in Table 1), to properly interpolate the emissions, corresponding activity and control information in each sector needs to be collected thoroughly, as shown in Table 2. Details about the approach applied to each sector are described in section 2.1.1-2.1.3.

Finally, emissions in each sector were scaled by the ratio (relative to the baseline) calculated for each year between 1990 and 2010 at the state level, to generate inventory files for each specific year. SMOKE was then run to generate the spatially and temporally resolved emissions. This is further clarified in the discussion in section 2.2.

2.1.1 Energy-related stationary sources

1) Activity

Following the structure of the energy data provided by State Energy Data System, emission sources in NEI data were grouped into four major energy-related stationary sources by seven types of fuel categories (total 20 sectors), as seen in Table 1a. Annual time series estimates of state-level energy use by broad energy-related sectors from 1960 to 2010 were directly derived from the State Energy Data System.

2) Emission factor

Trends in emission factors for each sector are also needed for the 20 year period in order to calculate the historical emissions. Originally, the AP-42 emission factors (http://www.epa.gov/ttn/chief/ap42/index.html) were used to calculate the emissions for each source in NEI data. There may be significant differences due to different control levels as well as sorts of combustion technologies (the range of unabated emission factors are given in Table 3). Since it's extremely difficult to obtain such detailed information about the absolute percentage of each control or combustion technology applied in each individual state during 20 yrs, in this study we just ensured all the averaged emission factors were within a reasonable range informed by AP-42 dataset. Also we attempted to back-calculate these emission factors from NEI data for seven years to quantify the evolution of emission controls.

The emission factor for each sector was calculated based on equation E1:

$$EF_{p,s,i,y} = \sum_{n} Emis_{p,s,i_n,y} \Big/ Act_{s,i,y} \qquad \text{(E1)}$$

Where, $EF_{p,s,i,y}$ is the calculated emission factor for pollutant p from sector i in State s in the year of y; $Emis_{p,s,i_n,y}$ is the emission amount in NEI data for pollutant p from sub-sector i_n of sector i in State s in the year of y; $Act_{s,i,y}$ is the energy consumption in sector i in State s in the year of y; sub-sector i_n means the original sector defined in NEI. For example, the sector "coal combustion in power plant" defined in this study is actually grouped from "sub-sectors" in NEI data (e.g., subbituminous Coal combustion in power plant using Wet/Dry Bottom technology).

The emission factors calculated for the seven years should satisfy the following rules:

a) All emission factors should be within the range from AP-42, i.e., equal or smaller than uncontrolled-level, and equal or greater than the maximally controlled-level;

b) The emission factor varies with the application of control technologies, thus for any given year it should be no larger than the one for the previous year;

c) If there is no evidence of controls, a consistent emission factor should be applied to all years during the study period.

The rules can be described as equation E2:

$$\min\left(EF_{p,s,i,y-1}, \max\_EF_{p,i_n}\right) \geq EF_{p,s,i,y} \geq \max\left(EF_{p,s,i,y+1}, \min\_EF_{p,i_n}\right) \quad \text{(E2)}$$

Where, $\max\_EF_{p,i_n}$ and $\min\_EF_{p,i_n}$ are respectively uncontrolled and maximum controlled emission factor for pollutant p in sub-sector i_n.

There are two cases when interpolating the emission factor for those years when NEI-data is unavailable:

For uncontrolled sectors, the changes of emissions are only related to changes in activity, therefore the emission factor is kept the same over the period.

For controlled sectors, the emission factor between two available years was constrained by national-level NEI trends information, described as equation E3.

$$\begin{cases} EF_{p,s,i,y} = EF_{p,s,i,py} - \dfrac{Act_{s,i,py}}{Act_{s,i,y}} \times \left(EF_{p,s,i,py} - EF_{p,s,i,ny}\right) \times \left(\dfrac{ef_{p,i,py} - ef_{p,i,y}}{ef_{p,i,py} - ef_{p,i,ny}}\right) & (py < y < ny \leq 2005) \\ EF_{p,s,i,y} = EF_{p,s,i,py} \times \dfrac{ef_{p,i,y}}{ef_{p,i,py}} & (y > py \geq 2005) \end{cases} \quad \text{(E3)}$$

Where, $ef_{p,i,y}$ is the national-averaged emission factor for pollutant p from sector i in the year of y calculated from NEI trends; py and ny are the two available years around the year of y;

2.1.2 Mobile sources

a) Activity

Activity data for mobile sources by types of highway vehicles and non-highway equipment were obtained from the highway statistics reported by Federal Highway Administration (http://www.fhwa.dot.gov/policyinformation). On-road sources from NEI data were grouped into four types of on-road vehicles: light-duty vehicle, light-duty truck, heavy-duty vehicle/truck and motorcycles. Vehicle Miles Traveled (VMT) rather than energy consumption was used as the activity indicator for each type of vehicle, in order to match with the estimated emission factors and emissions certification standards in which the given unit is grams per mile (obtained from 2011 National Transportation Statistics).

The VMT was calculated by using vehicle population multiplied by the average distance traveled per vehicle (DPV), as shown in equation E4, where, $VMT_{s,i,y}$ is the calculated annual VMT for type i of vehicle in State s in the year of y; $Population_{s,i_n,y}$ is the vehicle populations for sub-type i_n of type i in State s in the year of y; and $DPV_{i_n,y}$ is the DPV for sub-type i_n of vehicle type i in year y.

$$VMT_{s,i,y} = \sum_{n} \left(Population_{s,i_n,y} \times DPV_{i_n,y}\right) \quad \text{(E4)}$$

The state-level population of each type of vehicle is provided by the highway statistics. The evolution of DPV over the past two decades was obtained from the 2011 National Transportation Statistics.

The fuel usage by vehicle type was also estimated by E5 to examine the consistency in aspect of energy consumption provided by both highway statistics and State Energy Data System.

$$ENE_{s,i,y} = \sum_{n}\left(Population_{s,i_n,y} \times DPV_{i_n,y} \times FE_{i_n,y}\right) \qquad \text{(E5)}$$

Where, $ENE_{s,i,y}$ is the estimated annual fuel usage for type i of vehicle in State s in year y and $FE_{i_n,y}$ is the fuel efficiency for sub-type i_n of vehicle type i in year y, representing gallons of gas/diesel per average miles traveled.

The fuel efficiency for each type of vehicle in 1990-2010 was obtained from 2011 National Transportation Statistics. Since the fuel efficiency presents an increase trend from 1990 to 2010 due to the improvement of technology, the increase in fuel usage is a little smaller than that in VMT. Trends in annual fuel consumption for each mobile sector from 1990 to 2010 are presented in Figure S1. The mobile gasoline and diesel consumption estimated in this study agrees well with the one in State Energy Data System and Dallmann and Harley (2010).

To derive activity data for off-road sources, the sources were grouped by fuel type (i.e., residual oil, natural gas, LPG and jet fuel), and diesel and gasoline off-road equipments were further divided into 6 sectors based on more detailed information provided by highway statistics, as seen in Table 1b. Diesel fuel consumption for other off-road equipments (except railroad and marine) was calculated based on the method provided by Kean et al 2000, since such diesel fuel used is excluded from taxable fuel sales reported by highway statistics. Energy consumption in each sector is regarded as the activity indicator, which was obtained from State Energy Data System and highway statistics.

b) Emission factor

Following the approach described above for stationary sectors, emission factors for each mobile sector by state were back-calculated from NEI data. The difference is that we only chose the calculated emission factor from the 2002 NEI (the reason that we didn't use 2005 NEI was given in previous section 2.1) as a reference. The evolution of emission factors from 1990 to 2010 was informed by 2011 National Transportation Statistics which gives the estimated national average emission rates by vehicle type from the results of MOBILE6, the same model as the one used in 2002 NEI. We scaled the emission factors for the whole period by the ratios obtained from the 2011 National Transportation Statistics (the diesel fraction informed by MOBILE6 default data was used to average the trends of emission factors of gasoline and diesel for the same vehicle size), as function E6, where, $EF_{p,s,i,y}$ is the calculated emission factor for pollutant p from vehicle-type i in State s in year y; $ef_{p,i,y}$ is the emission factor obtained from National Transportation Statistics, and py is the baseline year as 2002.

$$EF_{p,s,j,y} = EF_{p,s,j,py} \times \frac{ef_{p,i,y}}{ef_{p,i,py}} \qquad \text{(E6)}$$

For nonroad sources, unfortunately AP-42 does not provide emission factors that we can directly use for comparison, since it's all embedded in the nonroad model. In order to ensure that those back-calculated emission factors are within the normal range, the calculated emission factor for each sector was validated through comparison with the corresponding factor in GAINS (The Greenhouse Gas and Air Pollution Interactions and Synergies model, http://gains.iiasa.ac.at/index.php/gains-europe, developed by IIASA, International Institute for Applied Systems Analysis), which provides a full range of emission factors from unabated to maximally controlled for each non-road sector. The evolution of emission factors for nonroad diesel and gasoline equipment was informed by NEI trends and Dallmann and Harley (2010). Dallmann and Harley (2010) suggested that NO_x and PM emission factors for off-road diesel-powered engines decreased significantly between 1996 and 2006. According to their results and NEI trends, we assumed the average NO_x and PM emission factors from off-road sources decreased by 25% and 18% over the past two decades. Due to the introduction of oil with lower sulfur content, the average emission factor of SO_2 in transportation decreased by 40%.

2.1.3 Non energy-related sources

Using the categories defined in NEI trends as well as the importance of each emission source, all non-energy related sources were combined into two groups with 16 sectors, as seen in Table 1c.

The first group is industrial processes which contributes less than 10% of total emissions for all pollutants. Considering the difficulties in collecting 20-years activities for those numerous industrial processes, we simply estimated their historic emissions by scaling the 2005 NEI data with national-level NEI trends, using linear fit method shown in equation E7.

$$Emis_{p,s,i,y} = Emis_{p,s,i,ry} \times \frac{emis_{p,i,y}}{emis_{p,i,ry}} \qquad \text{(E7)}$$

where $emis_{p,i,y}$ and $emis_{p,i,ry}$ are the national emissions from NEI trends for year y and ry (reference year).

The rest of non energy-related sources were grouped into 12 sectors, each of which primarily contributes to one or two specific pollutants. There are two cases when interpolating their emissions to the other years from the reference year:

First one, for uncontrolled sectors, the 20-year emissions were simply scaled by following the trend of activities, as equation E8:

$$Emis_{p,s,i,y} = Emis_{p,s,i,ry} \times \frac{Act_{p,s,i,y}}{Act_{p,s,i,ry}} \qquad \text{(E8)}$$

where, $Act_{p,s,i,y}$ and $Act_{p,s,i,ry}$ are the activities in sector i in State s for year y and ry (reference year).

Second one, for controlled sectors, an additional constraint was introduced by using national-level NEI trends, described as equation E9:

$$Emis_{p,s,i,y} = Emis_{p,s,i,ry} \times \frac{Act_{p,s,i,y}}{Act_{p,s,i,ry}} \times \frac{\left(emis_{p,i,y} / act_{i,y} \right)}{\left(emis_{p,i,ry} / act_{i,ry} \right)} \qquad \text{(E9)}$$

where $Act_{i,y}$ and $Act_{i,ry}$ are the national activities in sector i for year y and ry (reference year).

The sources of the activity data for the different sectors are described below.

a) VOC related sources

Solvent utilization accounts for up to 25% of total VOC emissions and half of non-energy related VOC emissions. Activity data were obtained from the national Quantity of Shipments of Paint and Allied Products from 1990-2010 (http://www.paint.org/about-our-industry/item/310.html). Emissions were constrained by NEI trends.

Storage and transport account for 8% of total VOC emissions. As with industrial processes, we estimated historic emissions by scaling the 2005 NEI data with national-level NEI trend using equation E6.

b) NH_3 related sources

The activity data used for fertilizer application (i.e., Agriculture crop shown in Table 1c) were the sum of the number of acres harvested multiplied by the nitrogen fertilizer application rate for each crop, obtained from Department of Agriculture National Agricultural Statistics Service (http://www.nass.usda.gov/).

The trend of NH_3 emission from livestock was estimated by summing the number of head obtained from the 1990-2010 Suvey of Agriculture (http://www.nass.usda.gov/) weighted by the NH_3 emission factor for each animal. These data included state-level estimates of number of head for the following livestock: cattle, hogs, poultry and sheep.

c) PM related sources

Construction dust accounts for 7% and 4% of total PM_{10} and $PM_{2.5}$ emissions in 2002. Previous studies suggested that the activity could be represented by the acres of land under construction estimated from the dollars spent on construction (U.S. EPA, 1998). Net values of construction work in residential, highway, street, and bridge construction, and other non-residential in year 2002 and 2007 by states as well as national trends from 1992 to 2010 were used for scaling. In order to remove the effects of inflation, the earnings data were converted to 1990 constant dollars using the implicit price deflator for personal consumption expenditures. (http://www.bea.gov/national/nipaweb/DownSS2.asp)

Mining and quarrying dust accounts for 5% and 2% of total PM_{10} and $PM_{2.5}$ emissions in 2002. According to the methodology described in NEI trends, the historic emissions in this sector are estimated by the sum of the amount of crude ore and coal handled at surface mines weighted by their corresponding PM_{10} emission factors. The amount of regional crude ore by state was obtained from the U.S. Geological Survey (http://minerals.usgs.gov/minerals/pubs/commodity/m&q/index.html#myb). The coal productions at surface mines were obtained from annual coal industry report (http://www.eia.gov/cneaf/coal/cia/cia_sum.html).

Agriculture tilling accounts for 19% and 15% of total PM_{10} and $PM_{2.5}$ emissions in 2002. According to the methodology in NEI trends, the activity for agriculture tilling is the number of acres of each crop in production multiplied by its corresponding number of passes and tillings. The acres planted for each crops for year 1990-2010 by state were obtained from the National Agricultural Statistics Service (http://www.nass.usda.gov/). Parameters of its corresponding number of passes & tillings for each type of crops were informed by NEI trends report.

Forest fires account for 7% and 17% of total PM_{10} and $PM_{2.5}$ emissions, including wildland and prescribed fires. Their activities were obtained from acres burned state and national data by National Interagency Fire Center (http://www.nifc.gov/fireInfo/fireInfo_statistics.html).

Paved and unpaved road emissions are the most important PM sources, accounting for 40% and 20% of total PM_{10} and $PM_{2.5}$ emissions. Its emission trend was estimated by State-based vehicle miles traveled with constraint of NEI trends.

d) Others

Information about the evolution of emissions from waste disposal and recycling and other miscellaneous sources was not available. We simply set their emissions to be the same over the period.

2.2 Processing for regional-scale model simulations and comparison with measurements

To support regional-scale air model simulations, the 20-year county-level emission inventories developed as described in Section 2.1 were processed by SMOKE to generate the spatially and temporally resolved emissions over a 12×12km CONUS (Continental United States) modeling domain.

a) The most recent NEI inventory files (in IDA or ORL format) were split into 49 sectors by SCC. Emissions in each sector were scaled by the ratio (to baseline) calculated for year 1990 to 2010 at the state level, to generate inventory files for each year. Spatial and temporal reference files were obtained from the most recent NEI dataset and applied for all the years.

b) Spatial allocation: Point sources are assigned to grid cells using the geographic coordinates. For area and mobile sources, a cross-reference file was used to match the gridding surrogates to the source level emissions.

c) Temporal allocation: Hourly Continuous Emissions Monitoring (CEM, http://ampd.epa.gov/ampd/) data from 1995 to 2010 were used for point source emissions. Similar temporal cross-reference and profile files were used for other sources.

Some studies (Lu et al., 2010; Wang et al., 2012) indicate that there is a highly linear relationship between the ambient concentrations of short-lived species (like SO_2 and NO_2) to their local emissions because their regional transport impacts are negligible. In this study, we collected long-term trends of observed SO_2, NO_2, CO and EC concentrations to compare with the emission trends developed as described in Section 2.1.

The 1990-2010 annual mean SO_2 concentrations over United States were downloaded from the CASTNET dataset (The Clean Air Status and Trends Network, http://epa.gov/castnet/javaweb/index.html). Data for SO_2, NO_2, CO concentrations monitored by Air Quality System (AQS) were downloaded from the EPA website (Air Quality Trends by City, 1990-2010, http://www.epa.gov/airtrends/factbook.html). Data for EC was obtained from IMPROVE network (Interagency Monitoring of Protected Visual Environments, http://vista.cira.colostate.edu/IMPROVE/). Trends of observed SO_2, NO_2, CO and EC concentrations respectively monitored at 39 CASTNET/96 AQS, 69 AQS, 89 AQS and 30 IMPROVE sites, having 20 yrs of completeness were used to compare with the trends of SO_2, NO_x, CO and EC emissions at the same spatial location. For the purpose of this analysis, emissions from the 81 grids located nearest to each monitor were summed; thus, the emissions in a roughly 100km × 100km area around each monitor are assumed to impact the concentrations measured at the monitor.

3. Results

3.1 Emission inventory from 1990-2010

Based on the method we discussed in section 2.1, the 20-years emission inventories were developed. The following sections give the discussion about the results by sector.

3.1.1 Power plants

Coal-fired power plants which in 2002 accounted for 69% and 20% of total SO_2 and NO_x

emissions respectively were the major control targets in the Acid Rain Program that started in 1990 (http://www.epa.gov/airmarkets/progsregs/arp/). Later, advanced SO_2 control technologies such as flue gas desulfurization (FGD) have been widely applied. The FGD application ratio in coal-fire power plants increased from 21% in 1990 to 56% in 2010, as seen in Figure S2. Because of the market-based initiative program that reduced SO_2 emissions in a cost-efficient manner by using emission trading, most FGD controls were applied to the units which consumed higher sulfur content coal to obtain the maximum cost benefit. From our estimates, average SO_2 emission factor in coal-fired power plants decreased by more than 70% during last two decades, as seen in Table 3. Average SO_2 emission factor in oil-fired units also decreased by around 30-60%, partly because the sulfur content of oil decreased due to the "spillover effect" from the impact of lowering the sulfur content on highway diesel (Bookhart and Zien, 2003).

Control efforts also reduced NO_x emissions in power plants during the Acid Rain Program as well as from the NO_x Budget Trading Program (NBP) that started in 2003 (http://www.epa.gov/airmarkt/progsregs/nox/sip.html). Figure 2 gives the application ratios of NO_x control technologies (weighted by unit capacity) in power plants from 1990-2010, which increased dramatically for all types of fuel combustion in power plants. More advanced post-combustion control technologies like selective catalytic reduction (SCR), selective noncatalytic reduction (SNCR) as well as their combinations with traditional combustion modification like overfire air (OFA), low NO_x burners (LNB) have been widely applied in coal-fired and natural gas-fired units since Phase II Stage (starting January 1, 2000) of the Acid Rain Program. In 2010, NO_x control application ratios reached 86% and 70% of total coal-fired and natural gas-fired units, respectively. Almost half of the control units applied SCR. LNB is the most prevalent control technology used in residual oil-fired units and its application ratio is around 20%. From our estimation, average of NO_x emission factors for both coal-and natural gas-fired units has decreased by around 70% over last two decades, and decreased by 28% for residual oil-fired units, as shown in Table 3. In addition, SO_2 and NO_x emission factors used in this study are all within the reasonable range of AP-42.

Compared to the trends in energy consumption from power plants in Figure 3a, trends of SO_2 and NO_x emission estimated in this study are within the constraint of energy evolution (i.e., below the energy trends). Also, majority of the emissions in this study agree with the original NEI data, except for NO_x emissions from distillate fuel oil combusted in power plant. Since the increase during 1996-1999 and 2001-2002 shown in the NEI data is hardly explained by the change in activities, which also means the emission factors during that period don't meet the rules (i.e., any given year it should be no larger than the one for the previous year; and all emission factors should be within the range from AP-42, i.e. equal or smaller than uncontrolled-level, and equal or greater than the maximally controlled-level;). The modified emission factors (which were set to be equal as the one for the previous year and within the uncontrolled-level) were used in this study; these agree better with the energy trends. The discrepancies in energy and emission trends, evident in the charts for coal-fired units for both SO_2 and NO_x, natural gas for NO_x and oil-fired units for SO_2, indicate the progress of emission controls which was informed by seven years NEI data. Besides, the consumption of natural gas in power plants increased much faster than coal, suggesting that cleaner fuel (i.e., natural gas) was used to replace coal and oil, which resulted in further reduction of emissions (See Figure 3a, consumption of natural gas increased by 200% while coal increased by 20%).

PM emissions from power plants have been well controlled since 1990. The application ratio of post-combustion control technology (mainly from electrostatic precipitators) reached 90% of total capacity in coal-fired units in 1990, and further increased to 96% in 2010, because of the wide application of advanced control technologies like baghouses and electrostatic precipitators as seen in

Figure S3. Such strengthened control efforts reduced the average of PM emission factors by 27% for coal-fired power plants.

Since there were no significant controls on CO and NMVOC, the emissions increased by 53% and 18% along with the growth in fuel consumption. The increase in NH_3 emissions was caused by the application of NO_x controls (e.g. SCR) that use NH_3 or urea.

3.1.2 Other combustion

Fuel combustion from industrial, commercial and residential sources accounted for 11% and 14% of total NO_x and SO_2 emissions in 2002, respectively. Use of low sulfur coal as well as innovative technologies to clean high sulfur were promoted by the Acid Rain Program, resulting in a reduction of average SO_2 emission factor in coal-fired boilers by 40-60%. Average SO_2 emission factor in oil-fired boilers was also decreased by 30-60% which was caused by the "spillover effect" from the impact of lowering the sulfur content of highway diesel. NO_x controls in coal- and natural gas-fired industrial boilers reduced their emission factors by 37% and 23% respectively, as seen in Table 3. SO_2 and NO_x emission factors used in this study are all within the reasonable range of AP-42.

As seen in Figure 3b-d, SO_2 and NO_x emission trends estimated in this study are better constrained by energy trends than that in the NEI data. For example, the SO_2 emission from industrial natural gas combustion increased by 100% from 1990 to 2000 in NEI which is doubtful because the energy consumption only increased by 20% during that period. Similar excessive increases in NEI are also shown in NOx emissions in 2000-2005 industrial distillate fuel combustion and 2000-2002 commercial coal combustion. This suggests that the emission factors during that period don't meet the rules (i.e., any given year it should be no larger than the one for the previous year). Besides, the residential NOx emissions decreased sharply from 1996 to 1999 in NEI data. Information about such reduction is unavailable, so in our estimates we followed the rule (i.e., if there is no evidence of controls, a consistent emission factor should be applied to all years during the study period) to modify the trends of residential NO_x emissions to be the same as the trends in energy.

Though the control effort taken in these combustion sectors is not as much as that in the power sector, all pollutant emissions from other combustion were significant reduced, mainly caused by the decline in consumptions of coal and oil which were replaced by cleaner fuels like natural gas.

3.1.3 On-road mobile sources

On-road transportation is one of the most important emission sources, contributing 41%, 68% and 31% to total anthropogenic NO_x, CO and NMVOC emissions in 2002, respectively. Light-duty vehicles and trucks, which have a larger vehicle population, contribute more to CO and NMVOC emissions, while heavy-duty vehicles and trucks, the majority of which is powered by compression ignition engines using diesel fuel, contribute a comparable percentage as light-duty vehicles and trucks to NO_x emissions because of their higher NO_x emission factor that is 5~10 times higher than that of light-duty vehicles and trucks, as shown in Table 1b.

As shown in Figure 4, along with the progress in strengthening the federal exhaust emissions certification standards for newly manufactured vehicles starting from 1970s (see Figure S4), the average emission factors of NO_x, CO and NMVOC in highway transportation significantly decreased by 80% between 1990 and 2010. Besides, all the emission factors used in this study are within the range between the current standards and the one about 20 years ago (assuming the vehicle lifetime was about 20 years), and exhibit comparable values but a smoother decline (agreed well with the estimated national average emission rates in 2011 National Transportation Statistics) compared to those calculated from NEI data. Particularly, NO_x emission factors of light-duty trucks and heavy-duty vehicles and trucks in 2005 NEI are much higher than those in 2002 NEI which caused by the method change from MOBILE to MOVES.

Though highway transportation accounts for only a small part of total SO_2 and PM emissions (about 2%), efforts have been made by U.S. EPA to lower the sulfur content of diesel oil used for transportation in the 1990s, as well as to reduce on-road dust emissions (Dallmann and Harley, 2010).

As seen from the comparison of emission and activity (i.e., VMT) trends in Figure 5, our estimations agree well with NEI data. With strengthened control efforts, all pollutants (except NH_3) exhibit declining trends despite a growth of activity for all types of vehicles. The on-road NOx emission was decreased by 58% which is close to the NEI trends data which is 55%. McDonald et al. (2012) suggested relatively lower reduction ratio as 33%. That's mainly because in McDonald et al. (2012) study, the reduction in NOx emission factor for heavy duty diesel vehicles is ~36%, which is relatively smaller than the one suggested by National Transportation statistics, as 70%.

3.1.4 Off-road mobile sources

Off-road transportation is another important emission source for NO_x, CO and NMVOC (17%, 12 and 11% respectively). NO_x emissions are primarily attributable to diesel-powered engines, including off-road transportation, rail and marine vessels which have higher emission factors, while CO and NMVOC are largely attributable to gasoline-powered equipments (see in Table 1b). The emission factors used in this study are comparable with those in GAINS (see in Table 3), as well as the studies on U.S. mobile emissions from 1996-2006 by Dallmann and Harley (2010), suggesting that the activity we selected and emission factor we calculated in this study are suitable to use.

3.1.5 Non-energy related sources

Emissions from industrial processes were processed by using a linear fit method calculated from NEI trends, as shown in Figure 6. Significant reductions by 20-80% are found in sectors such as chemical manufacturing and metals processing for all pollutants. The linear-fit trends in this study roughly agree well with NEI trends. However, since it is extremely difficult to obtain activity indicators for each of those numerous processes over such an extended time period, the trend estimates for these sectors are highly uncertain and warrant further investigation.

Each of the other non-energy related sources typically contributes to one or two specific pollutant, as shown in Figure 7. NH_3 emissions from fertilizer applications and PM emissions from mining and quarrying, and forest wildfires increased significantly in NEI data, even larger than the activity trend. Those emissions were scaled by growth ratios of corresponding activities alone without controls. Compared to the trends of activity data, though NEI data presented decrease in several sectors including livestock NH_3 and PM from agriculture tilling and prescribed forest management, information about such decreases is unavailable, so those emissions were also scaled by growth ratios of corresponding activities.

The declining trends evident in VOC emissions from solvent utilization, PM emissions from construction processes, and paved/unpaved road dust shown in NEI data, suggest that control efforts have been made in these sectors. Thus their emissions were additionally adjusted based on NEI data.

3.2 Comparison and Validation

Table 4 summarizes the changes in total emissions from 1990 to 2010. Significant reductions are shown for SO_2, NO_x, CO, NMVOC, PM_{10} and $PM_{2.5}$; these reductions are 67%, 48%, 60%, 48%, 50% and 34% respectively. Most sectors contribute to these reductions. Strengthened controls on power plants are the dominant factor for SO_2 reductions and are also one of the major contributors to NO_x reductions. Controls on mobile sources largely contribute to the reductions of NO_x, CO and NMVOC emissions. The reductions in PM emissions are mainly from the controls on the on-road dust sources. Seen from the spatial distribution of those changes presented in Figure 8, those reductions were widely distributed over the U.S., but much more strengthened in the north area and California where emission intensities were higher. Due to the growth of livestock activities, NH_3 emissions were increased by

11%, particularly in North Carolina and Iowa due to significant increases in the activity of livestock and agriculture. The growth of application of NO_x controls (e.g. SCR) which using NH_3-rich material as the reducing agent also contributes some to the NH_3 increases.

The 20-year emissions calculated in this study were compared with NEI trends and EDGAR data, as seen in Figure 9. In general, the results of this study agree well with the other two trends, particularly for SO_2. Relative lower emissions were shown in EDGAR for NO_x, CO, VOC and PM. The emission trends developed in this study are closer to NEI trends, but with a smoother trend.

The trends of SO_2, NO_x, CO and EC emissions were compared with the observed trends in ambient surface SO_2, NO_2, CO and EC concentrations to evaluate the 20 yr of emission inventories. The spatial distributions of trends generally agree well with the observations, as seen in Figure 10a. The results indicate that the declining emission trends manifest themselves in decreasing observed concentrations for all species, and that those reductions were widely distributed across the whole continental US domain.. The average reduction of SO_2, NO, CO and EC emissions in the grid cells near monitors are 69%, 47%, 58% and 36% respectively, which agrees well with the observed decrease of SO_2, NO_2, CO and EC concentrations, as 63%, 33%, 71% and 50%, respectively.

As seen in Figure 10b, Emission trends of SO_2, NO, CO and EC are given by sector (the same as the one defined in SMOKE), i.e., (1) ptipm- NEI point source EGUs (Electric Generating Units) mapped to the Integrated Planning Model model using the National Electric Energy Database System database; (2) ptnonipm- All NEI point source records not matched to the ptipm sector; (3) mobile; (4) non-road and (5) other area sources. It can be seen that deceases in different species were driven by reductions in different source sectors. At the national level, EGUs are the dominant source of SO_2. The trend of observed SO_2 concentration closely follows the EGU trend, with decreases during the period of 1990-1995 and after 1998 and increases during the period of 1995-1998. Since the dominant sources may be different at different locations, we also conducted the comparison at a sub-regional scale. The sub-regions used in this analysis were the same as defined in Hand et al. (2012), i.e., West, Great Plains, Southwest, Northeast, Midsouth and Southeast, as shown in Figure 11.

For SO_2 (see Figure 12a), the comparisons for the Northeast, Midsouth and Southeast show similar results as the analysis at the national level. The decrease in emission trends after 2006 is 10-30% larger than that in observed trends. EGU is also the dominant sources in the southwest area, but its reduction is more significant after 1998. In West and Great Plains, the comparison is not as good as the other regions. In West, non-EGU point and area sources are the dominant sources. Emissions generally present similar decreasing trends, but the decrease in emission trends after 2006 is 10-30% smaller than that in the observed trend. In Great Plains, SO_2 emission was dominated by non-EGU point sources. Though a decreasing trend was shown in both emissions and observed concentrations, the SO_2 concentration before 1996 is extremely high but the SO_2 emission rate is even lower than other regions. Some important sources in that area may be missing during that period (the baseline inventory is more recent may not include sources that are now shut off).

Mobile sources are the dominant contributor to NO_x emissions in all regions (see Figure 12b). The national emission trend agrees better with the trends of observed NO_2 concentration before 2000 than with the trends after 2000. The decrease in emission trends after 2000 is 10-20% smaller than that in observed trends. Similar results are also found in Northeast, Midsouth and Southeast. NO_x reductions in mobile sources may be over-predicted by 10-20% in those areas. In West and Southwest, observed trends during 1997-2006 are 10-20% lower than the emission trends, which suggests that NO_x mobile controls in those regions may be several years ahead of the nation level. Further improvement of this study may consider using different trends of mobile emission factors for different regions.

Mobile is also the dominant CO emissions sources for all regions (see Figure 12c). The national emission trend agrees well with observed CO concentration before 1999, but 10-30% higher after 2000. The trends of mobile CO emission factors might under-predict the control effectiveness after 2000, particular in the West region.

EC emissions are contributed by various sources. The observed trend is more variable than other species, suggesting that the changes of meteorological conditions and wildfire activity may contribute to that variation. Even through, the EC emission trend roughly agrees with the observed trend of EC concentration. Such decrease is mainly driven by the reduction in mobile sources.

4. Conclusion

A consistent series of spatially resolved anthropogenic emissions of SO_2, NO_x, CO, NMVOC, NH_3, PM_{10} and $PM_{2.5}$ in the United States from 1990 to 2010 was developed by using an approach based on several long-term databases containing information about changes in activity data and emission controls. These inventories were processed by SMOKE and are ready to be used for regional chemistry transport model simulations.

The set of inventories developed in this study is internally consistent, constrained by activity trends, within reasonable range of emissions and controls, and comparable with previous studies. However, due to the lack of a detailed historic record of control information over such an extended time period (except for the power sector), our estimations on control efforts for other sectors highly depended on NEI data or NEI trends. Besides, since this study mainly focused on trends rather than the absolute value in each individual year, some sectors (e.g., industrial processes) and sub-sectors (types of combustion and stoves) may not have been well considered or examined. For example, Heald et al (2012) suggested that ammonia emissions in 2005 NEI (which they used for their simulations) are underestimated in California and in the springtime in the Midwest. Since the new emission inventory is based on 2005 NEI, it may also suffer such underestimation. Therefore future improvement in the accuracy of baseline emission inventory (i.e., 2005 NEI in this study) is also necessary. Mobile emissions were only based on MOBILE6 results rather than the new-developed MOVES module. Further improvement on those details is still necessary.

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Table 1 Defined sectors and percentage of their contributions to total anthropogenic emissions (calculated from the 2002 NEI data, Unit: %)

(a) Energy-related stationary sources

Sector		Pollutant						
Group	Type	NO_x	SO_2	CO	NMVOC	PM_{10}	$PM_{2.5}$	NH_3
Power plants	Coal	20.2	69.4	0.4	0.2	4.5	9.5	0.3
	Residual oil	0.6	2.4	0.0	0.0	0.2	0.3	0.0
	Distillate oil	0.4	0.2	0.0	0.0	0.0	0.1	0.0
	Natural gas	1.4	0.4	0.1	0.1	0.1	0.2	0.3
	Total	**22.6**	**72.4**	**0.6**	**0.3**	**4.8**	**10.2**	**0.7**
Industrial Combustion	Coal	1.7	7.4	0.1	0.0	1.5	1.3	0.1
	Residual oil	0.3	1.3	0.0	0.0	0.1	0.2	0.0
	Distillate oil	0.6	0.9	0.0	0.0	0.1	0.2	0.1
	Natural gas	5.2	0.2	0.4	0.4	0.1	0.2	0.2
	Total	**7.7**	**9.9**	**0.6**	**0.5**	**1.8**	**1.9**	**0.4**
Commercial Combustion	Coal	0.2	0.9	0.0	0.0	0.3	0.2	0.0
	Residual fuel oil	0.1	0.6	0.0	0.0	0.0	0.0	0.0
	Distillate oil	0.2	0.9	0.0	0.0	0.1	0.2	0.1
	Natural gas	0.8	0.0	0.1	0.1	0.0	0.0	0.0
	LPG	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Kerosene	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Total	**1.3**	**2.5**	**0.2**	**0.1**	**0.4**	**0.4**	**0.1**
Residential Combustion	Coal	0.0	0.1	0.1	0.0	0.0	0.1	0.0
	Distillate oil	0.3	0.9	0.0	0.0	0.1	0.1	0.0
	Natural gas	1.2	0.0	0.1	0.1	0.0	0.0	0.0
	LPG	0.2	0.0	0.0	0.0	0.0	0.0	0.0
	Kerosene	0.0	0.1	0.0	0.0	0.0	0.0	0.0
	Wood	0.2	0.0	2.9	8.5	3.2	8.2	0.2
	Total	**1.9**	**1.2**	**3.1**	**8.7**	**3.3**	**8.4**	**0.3**

(b) Energy-related mobile sources

Sector: Group	Sector: Type	Pollutant: NO_x	SO_2	CO	NMVOC	PM_{10}	$PM_{2.5}$	NH_3
Highway	Light Duty Vehicle, gasoline & diesel	11.2	0.6	34.2	16.1	0.4	0.5	4.0
	Light Duty Truck, gasoline & diesel	9.4	0.5	29.7	12.2	0.3	0.4	2.6
	Heavy Duty, gasoline & diesel	20.2	0.6	3.7	2.4	0.9	2.1	0.3
	Motorcycle, gasoline	0.1	0.0	0.2	0.2	0.0	0.0	0.0
	Total	**40.8**	**1.7**	**67.8**	**30.9**	**1.6**	**3.0**	**6.9**
Off-highway	Agriculture, gasoline	0.0	0.0	0.1	0.0	0.0	0.0	0.0
	Construction, gasoline	0.0	0.0	0.5	0.2	0.0	0.0	0.0
	Other gasoline	0.5	0.0	9.3	9.4	0.3	0.7	0.0
	Marine, diesel	4.0	0.6	0.1	0.1	0.2	0.6	0.0
	Rail, diesel	5.5	0.5	0.1	0.3	0.2	0.6	0.0
	Other diesel	5.0	0.7	0.5	0.7	0.8	1.9	0.0
	Marine, residual oil	1.2	1.1	0.0	0.0	0.1	0.3	0.0
	All natural gas	0.2	0.0	0.1	0.0	0.0	0.0	0.0
	All LPG	1.0	0.0	0.8	0.3	0.0	0.0	0.0
	All Jet fuel	0.4	0.1	0.5	0.3	0.2	0.4	0.0
	Total	**17.9**	**3.0**	**12.0**	**11.5**	**1.9**	**4.5**	**0.1**

(c) Non energy-related sources

Sector: Group	Sector: Type	Pollutant: NO_x	SO_2	CO	NMVOC	PM_{10}	$PM_{2.5}$	NH_3
Industry processes	Chemical manufacturing	0.5	1.8	0.4	1.5	0.3	0.6	0.6
	Metals processing	0.4	1.6	1.0	0.3	0.7	1.2	0.1
	Petroleum & related industries	2.2	2.6	0.4	3.6	0.2	0.4	0.1
	Other industry processes	2.5	2.6	0.5	2.6	3.2	5.1	4.6
	Total	**5.6**	**8.7**	**2.3**	**8.1**	**4.4**	**7.4**	**5.3**
Other	Solvent Utilization	0.0	0.0	0.0	24.7	0.1	0.1	0.0
	Storage & Transport	0.0	0.0	0.0	8.4	0.0	0.0	0.0
	Waste disposal & recycling	0.5	0.2	1.5	2.2	2.2	5.4	0.7
	Construction processes	0.0	0.0	0.0	0.0	6.9	4.0	0.0
	Paved/Unpaved road	0.0	0.0	0.0	0.0	40.1	19.5	0.0
	Mining & Quarrying	0.0	0.0	0.0	0.0	4.7	2.4	0.0
	Agriculture crop	0.3	0.1	1.9	1.0	19.3	15.4	30.0
	Agriculture livestock	0.0	0.0	0.0	0.2	0.8	0.4	54.4
	Forest Wildfires	0.9	0.3	7.7	2.2	6.2	13.8	1.0
	Prescribed Burning for Forest Management	0.2	0.1	1.6	0.5	1.3	3.0	0.2
	Miscellaneous	0.1	0.0	0.8	0.8	0.1	0.3	0.0

Total	**2.1**	**0.8**	**13.5**	**40.1**	**81.8**	**64.3**	**86.2**

Table 2 Summary of data sources used for scaling emissions

Sector			Emission constrains: Activity indicator	Emission constrains: Control info.
Energy-related Stationary	Power plants	Coal, Residual oil, Distillate oil, Natural gas, LPG, Kerosene, Wood	State-based energy combustion by fuel types[1]	Unit level database[2]
	Industrial			NEI[3]
	Commercial			
	Residential			
Energy-related Mobile	On-road	Light Duty Vehicle	State-based vehicle miles traveled by vehicle types[4]	Evolution of emission factors by vehicle types[5]
		Light Duty Truck		
		Heavy Duty		
		Motorcycle		
	Off-road	Agriculture, gasoline	State-based gasoline consumption by equipment types[4]	NEI
		Construction, gasoline		
		Other gasoline		
		Marine, diesel	National diesel consumption by equipment types[4]	
		Rail, diesel		
		Other diesel	Total Distillate Fuel Oil and Kerosene Sales by End Use[6]	
		Marine, Residual oil	State-based energy consumption by fuel types[1]	
		All natural gas		
		All LPG		
		All Jet fuel		
Non energy-related	Industry process	Chemical manufacturing	NEI	
		Metals processing	NEI	
		Petroleum industries	NEI	
		Other processes	NEI	
	Other	Solvent Utilization	Quantity of Shipments of Paint and Allied Products[7]	NEI
		Construction processes	The dollars spent on construction[8]	NEI
		Paved/Unpaved road	State-based vehicle miles traveled[4]	NEI
		Storage & Transport	NEI	
		Waste disposal & recycling	Keep the same over the period	
		Mining & Quarrying	crude ore and coal handled on surface mine[9,10]	
		Agriculture tilling and Fertilizer Application	number of acres harvested for each crop[11]	

		Livestock	Livestock Operations[9]
		Forest fires	Acres Burned[12]
		Miscellaneous	Keep the same over the period

Data obtained from:

1. State Energy Data System, http://www.eia.gov/state/seds/
2. Air Markets Program Data, http://camddataandmaps.epa.gov/gdm
3. NEI trends report, http://www.epa.gov/ttnchie1/trends
4. Highway statistics, http://www.fhwa.dot.gov/policyinformation/index.cfm
5. 2011 National Transportation Statistics, http://www.bts.gov/
6. U.S. Energy Information Administration, http://www.eia.gov/petroleum/fueloilkerosene/
7. American Coating Association, http://www.paint.org/about-our-industry/item/310.html
8. United States Census Bureau, http://www.census.gov/construction/c30/totpage.html
9. Minerals Yearbook, http://minerals.usgs.gov/minerals/pubs/commodity/m&q/index.html#myb
10. Annual Coal Report, http://www.eia.gov/cneaf/coal/cia/cia_sum.html
11. Department of Agriculture National Agricultural Statistics Service, http://www.nass.usda.gov/
12. National interagency fire center, http://www.nifc.gov/fireInfo/fireInfo_statistics.html

Table 3 Summary of NO_x and SO_2 emission factors in energy-related stationary and nonroad sectors (Unit: 0.01 lb/MMBtu)

Sector	Fuel	NO_x Emission factor This study 1990	NO_x This study 2010	NO_x Δ[2]	NO_x Unabated[1]	SO_2 Emission factor This study 1990	SO_2 This study 2010	SO_2 Δ	SO_2 Unabated
Power plants	Coal	79	23	-71%	7~127	211	59	-72%	3~512[3]
	Residual oil	31	23	-28%	21~31	105	77	-27%	105[4]
	Distillate oil	34	34	-	17~34	73	25	-66%	101~107
	Natural gas	28	7.6	-72%	10~28	0.06	0.06	-	0.06
Industrial Combustion	Coal	57	36	-37%	7~127	192	102	-47%	3~512
	Residual oil	37	37	-	27~37	104	78	-25%	105
	Distillate oil	24	24	-	14~34	46	23	-50%	101~107
	Natural gas	27	20	-23%	10~28	0.06	0.06	-	0.06
Commercial Combustion	Coal	66	66	-	7~127	472	296	-37%	3~512
	Residual fuel oil	37	37	-	31~37	92	67	-27%	105
	Distillate oil	17	17	-	14~17	89	42	-52%	101~107
	Natural gas	12	12	-	10~28	0.06	0.06	-	0.06
	LPG	14	14	-	13~16	0.02	0.02	-	0.02
	Kerosene	17	17	-	7~17	68	68	-	101~107
Residential Combustion	Coal	66	66	-	7~127	498	225	-55%	3~512
	Residual oil	13	13	-	13	75	34	-55%	101
	Natural gas	12	12	-	10~28	0.06	0.06	-	0.06
	LPG	16	16	-	13~16	0.02	0.02	-	0.02
	Kerosene	13	13	-	13	57	57	-	101
	Wood	15	15	-	15	2.3	2.3	-	2.3
Off-road Transport	Gasoline	46	46	-	7~200	1.4	0.8	-42%	0.1~1.0
	Diesel	300	224	-25%	209~326	55	35	-35%	0.1~55
	Residual fuel oil	77	77	-	326	55	55	-	0.1~55
	Natural Gas	6.8	6.8	-	7~200	0.01	0.01	-	0.00
	LPG	6.8	6.8	-	7~196	0.01	0.01	-	0.00

	Jet Fuel	8.0	8.0	-	9.0	0.7	0.7	-	0.7

Notes:

[1] The unabated emission factors for stationary sectors are from the AP-42 dataset. The unit has been changed to lb/MMBtu, divided by the heating value. The range represents all the situations under different types of firing configuration. For off-road transport, the values referred to GAINS datasets.

[2] The relative change from the year 1990, i.e., $(EF_{2010}-EF_{1990})/EF_{1990}\times100\%$

[3] The value was calculated based on 3.4% sulfur content in coal, applied to all other sectors.

[4] The value was calculated based on 1% sulfur content in oil, applied to all other sectors.

Table 4 Sector-specific emissions in the United States for 1990 and 2010 estimated in this study (unit: Gg·year^{-1})

Sector	SO_2			NO_x			CO			NMVOC			PM_{10}			$PM_{2.5}$			NH_3		
	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ
Power plants	16117	5018	-69%	6431	2237	-65%	294	462	57%	39	46	20%	654	559	-15%	529	462	-13%	11	19	70%
Other Comb.	3764	1215	-68%	2625	1772	-32%	4686	2598	-45%	841	428	-49%	1048	511	-51%	767	389	-49%	26	28	6%
On-road	499	35	-93%	10972	4643	-58%	136586	38627	-72%	9798	2828	-71%	392	118	-70%	333	92	-72%	145	276	91%
Off-road	619	452	-27%	3428	2768	-19%	12257	11870	-3%	2388	1950	-18%	245	188	-23%	222	172	-22%	2	2	-21%
Ind. Process	2147	873	-59%	1412	1227	-13%	5498	2133	-61%	2291	1686	-26%	637	318	-50%	469	287	-39%	126	126	0%
Other	100	104	5%	390	400	2%	12922	12992	1%	8450	5277	-38%	17130	8394	-51%	3814	2675	-30%	3181	3414	7%
All	23246	7697	-67%	25258	13048	-48%	172243	68682	-60%	23807	12215	-49%	20105	10088	-50%	6134	4077	-34%	3491	3864	11%

Note: $\Delta = (Emis_{2010} - Emis_{1990})/Emis_{1990} \times 100\%$

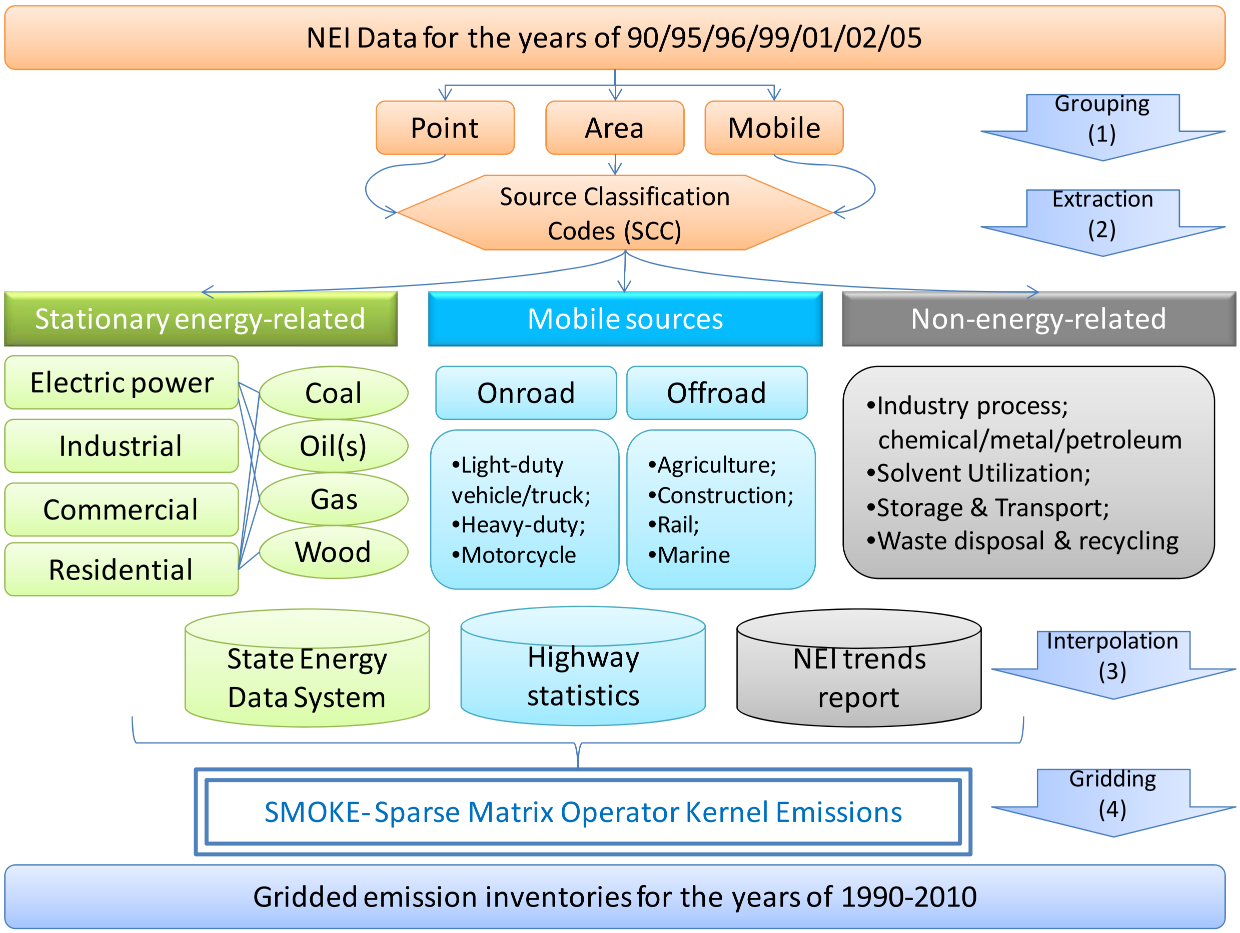
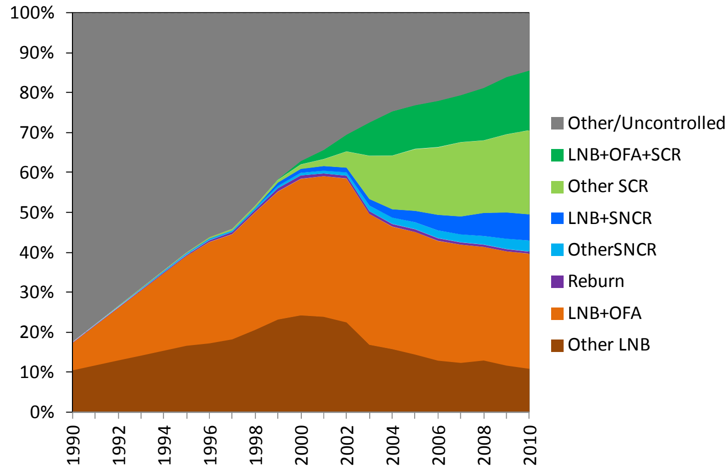
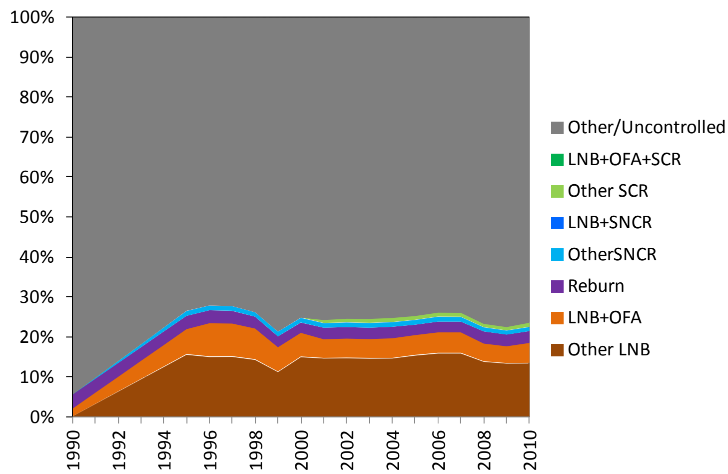


Fig. 1 The framework of the 20 years emission inventory development

(a) Coal-fired



(b) Oil-fired



(c) Gas-fired

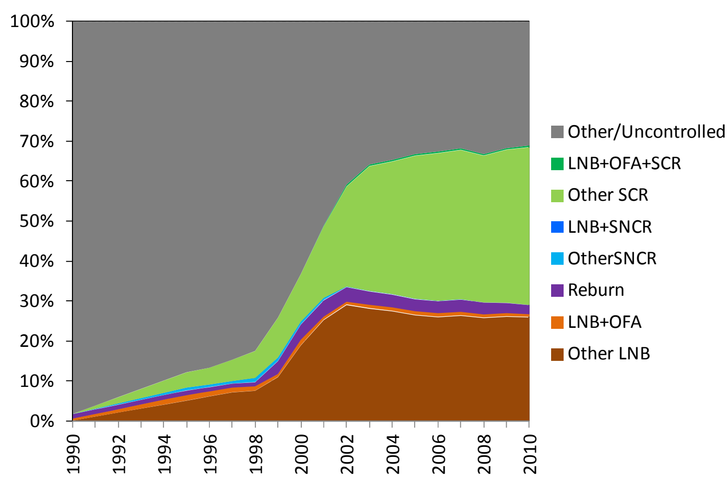


Fig. 2 Application of NO_x control technologies in power plants from 1990-2010 by primary fuel type (weighted by unit capacity, LNB- low NO_x burners; OFA- overfire air; SNCR- selective noncatalytic reduction; SCR- selective catalytic reduction; based on the Clean Air Markets data, http://ampd.epa.gov/ampd/)

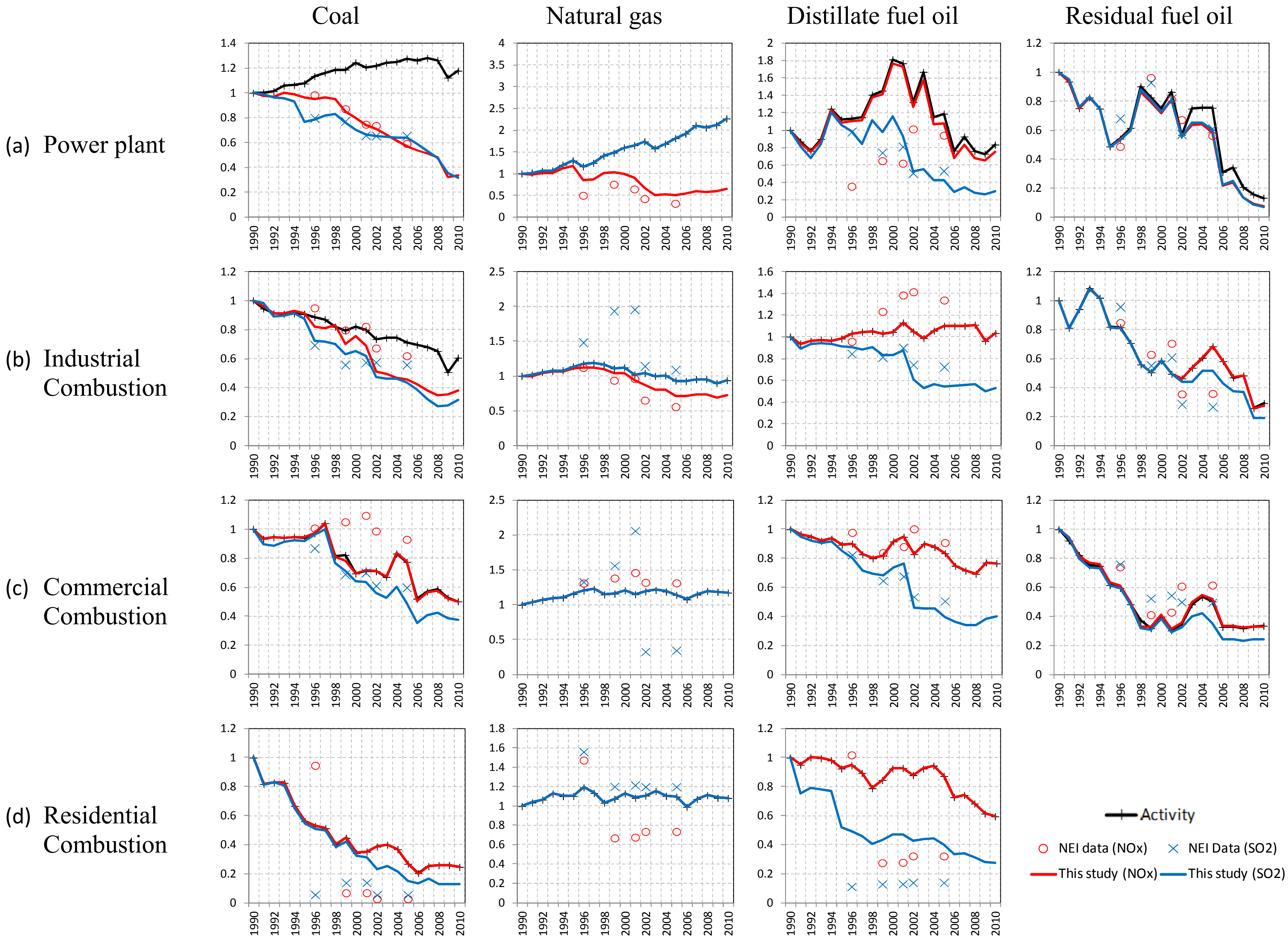

Coal
Natural gas
Distillate fuel oil
Residual fuel oil
(a) Power plant
(b) Industrial Combustion
(c) Commercial Combustion
(d) Residential Combustion
Activity
NEI data (NOx)
NEI Data (SO2)
This study (NOx)
This study (SO2)

Fig. 3 Activity (fuel use) and NO_x and SO_2 emission trends during 1990-2010 for energy-related stationary sources (Year 1990=1, absolute values are provided in Table S2a)

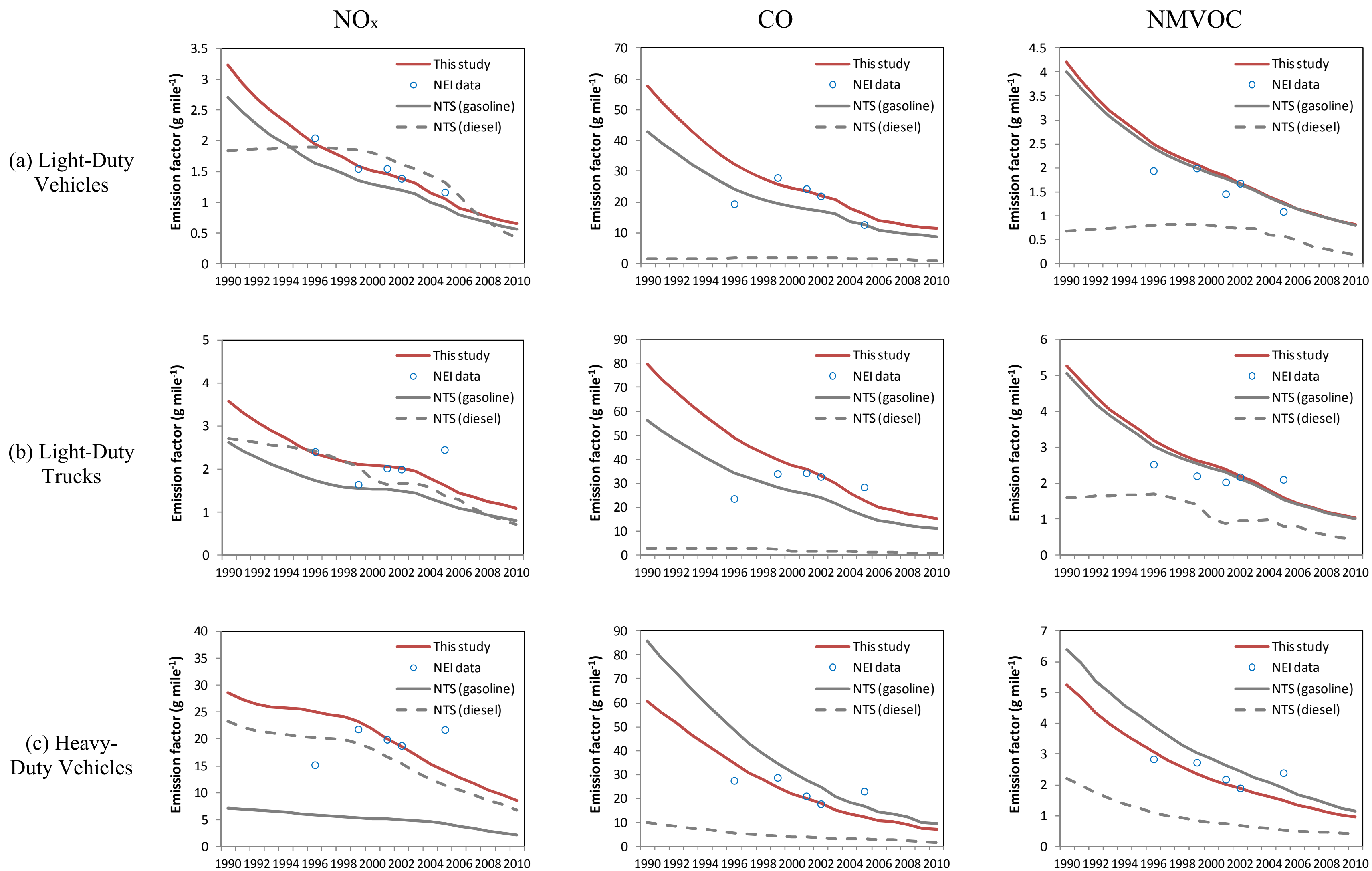


Fig. 4 On-road NO_x, CO and NMVOC emission factors in this study (NTS-National Transportation Statistics)

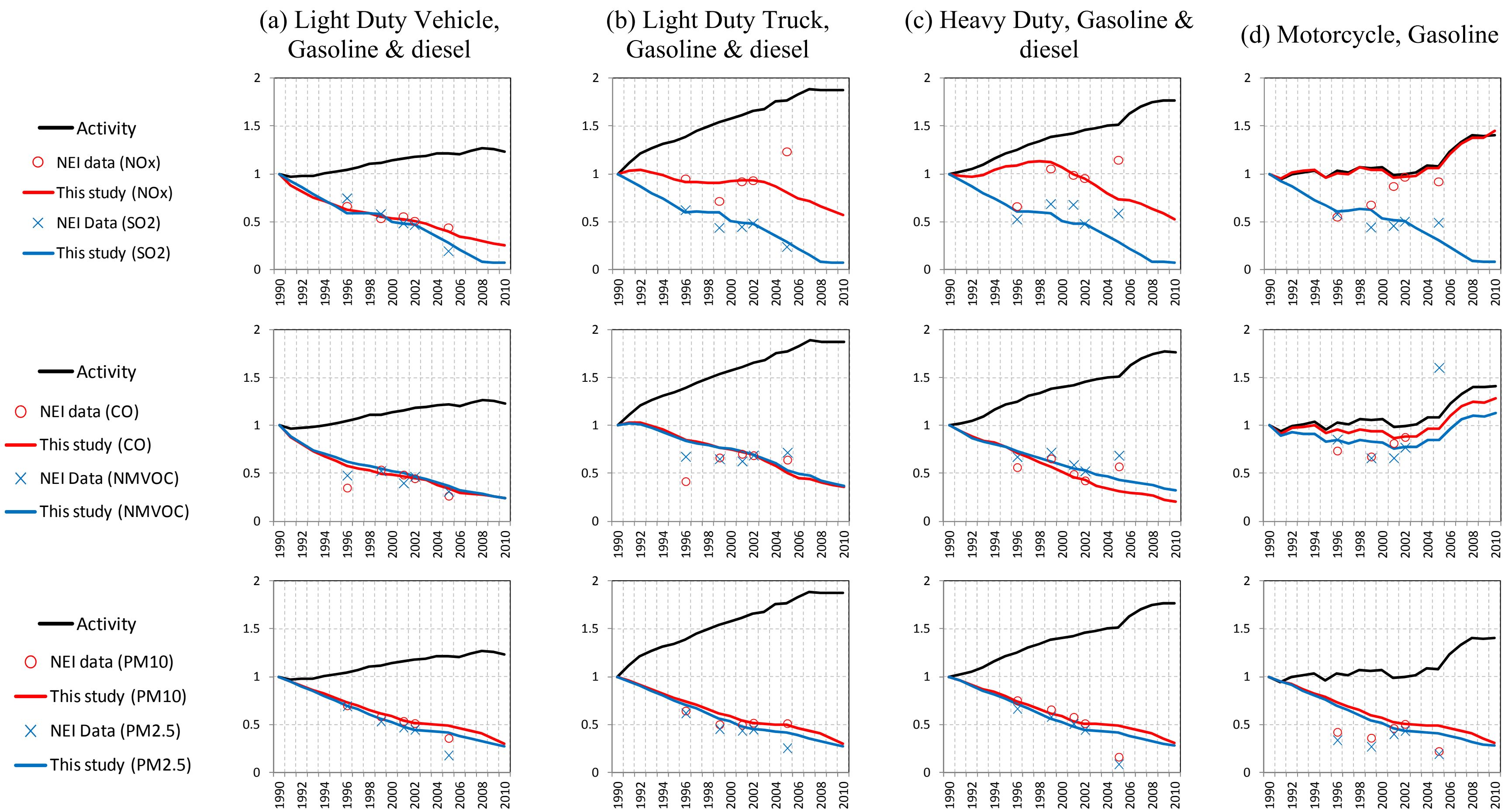


Fig. 5 Activity and emission trends during 1990-2010 for on-road mobile sources (Year 1990 =1, absolute values are provided in Table S2b)

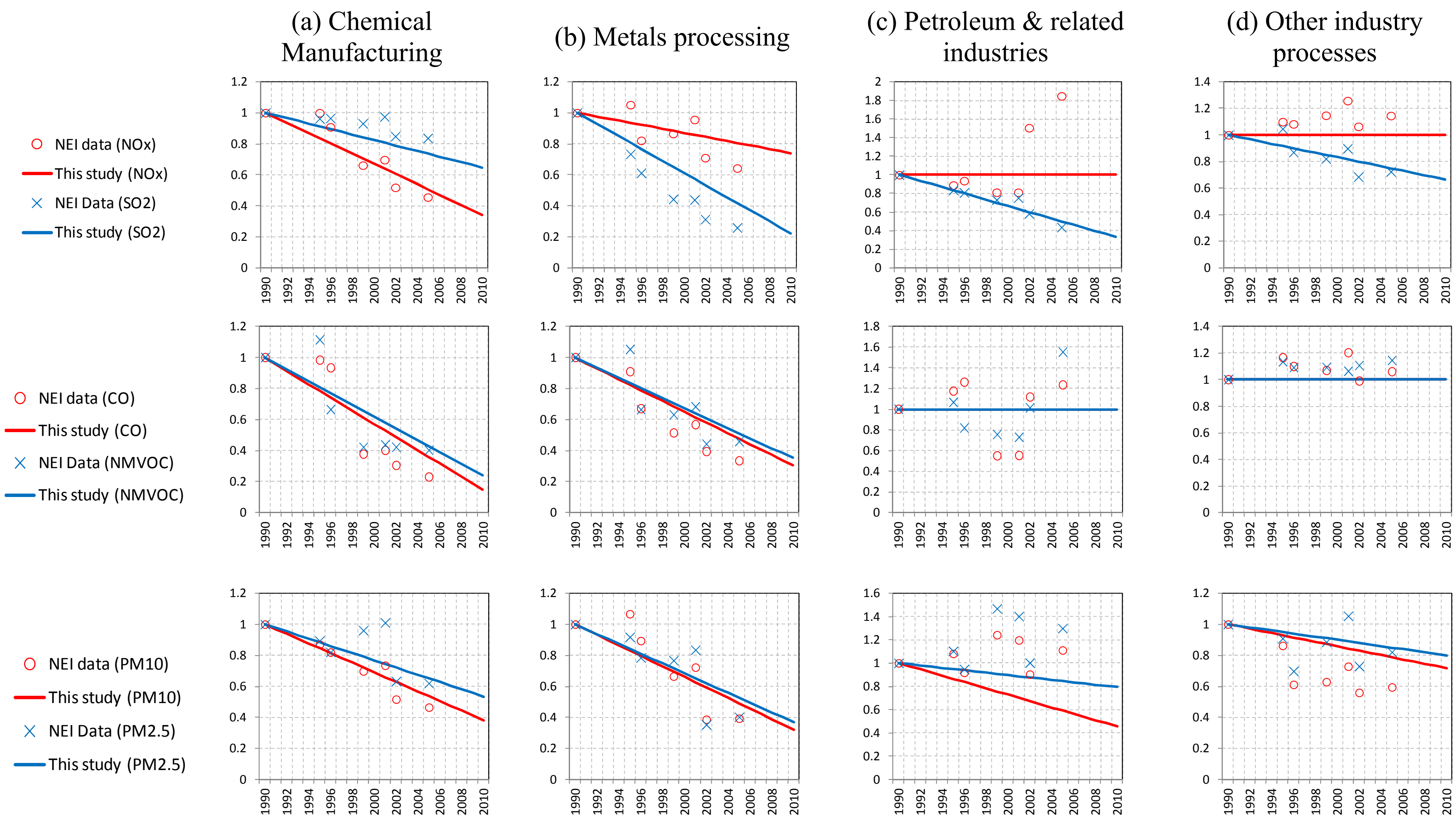


Fig. 6 Emission trends during 1990-2010 for industrial process sources (Year 1990=1, absolute values are provided in Table S2c)

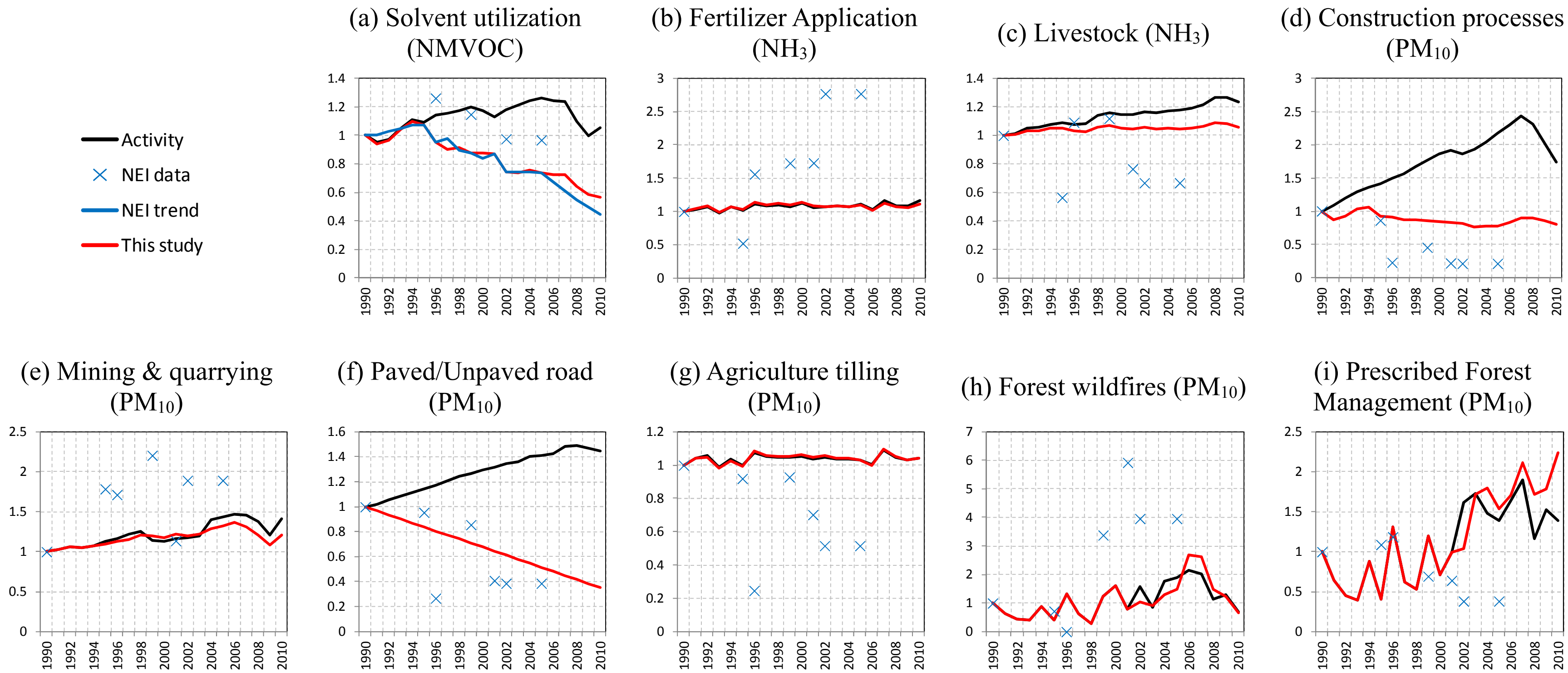


Fig. 7 Activity and emission trends during 1990-2010 for other non-energy related sources (Year 1990=1, absolute values are provided in Table S2d)

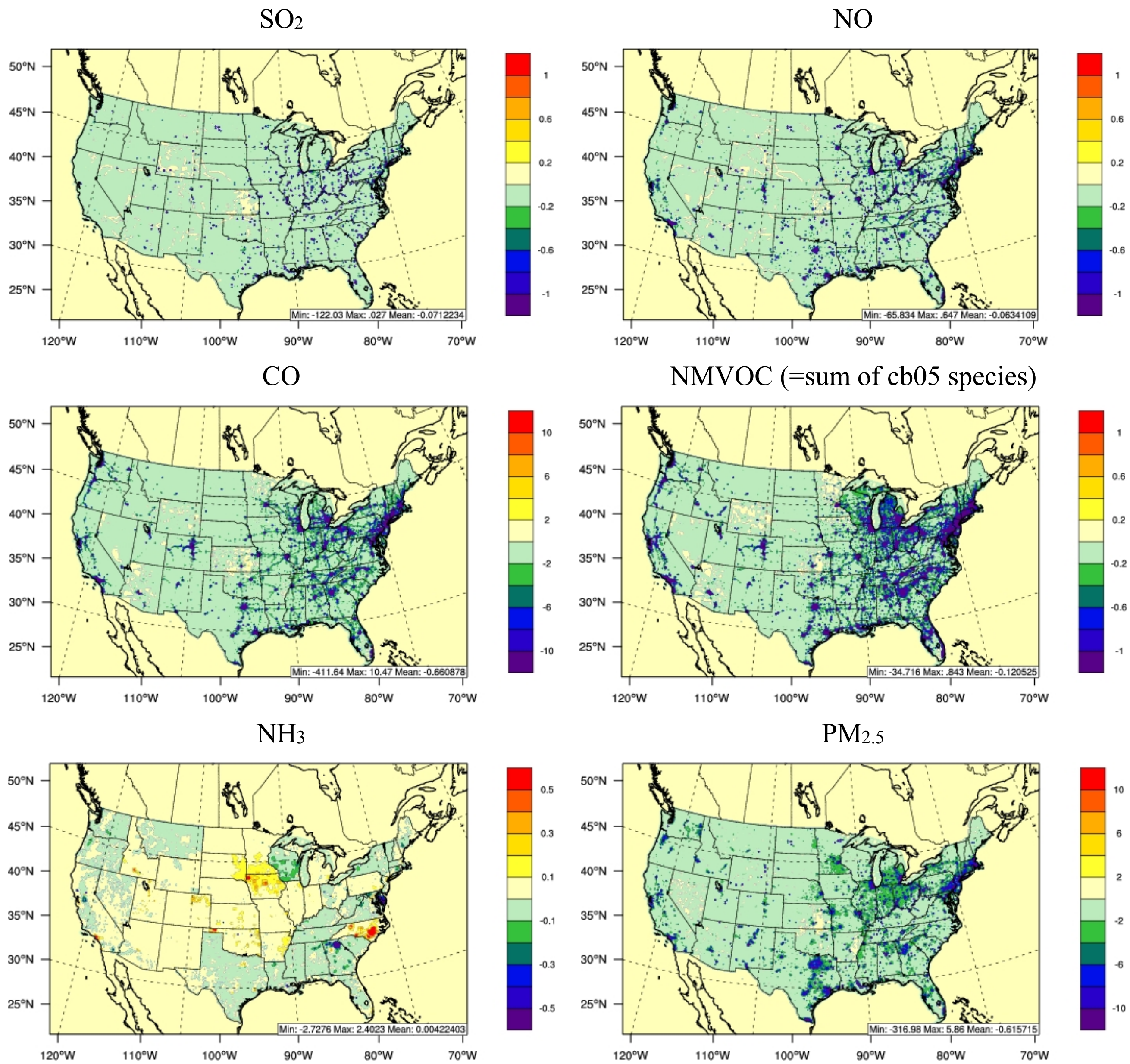


Fig. 8 Changes in emission rates over the 12×12km CONUS domain from 1990 to 2010 (unit: moles/s·grid for gaseous species, g/s·grid for $PM_{2.5}$)

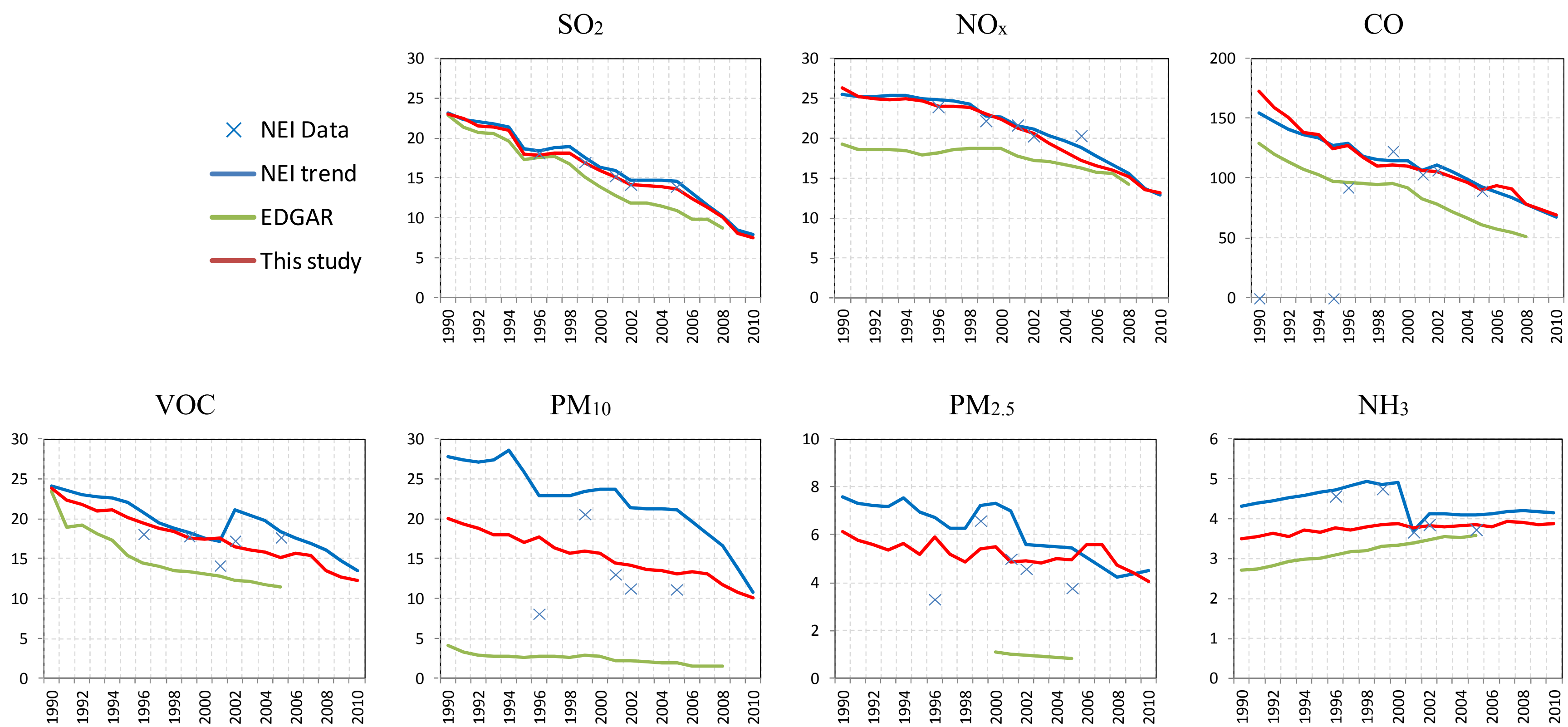


Fig. 9 Emission trends during 1990-2010 (unit: Tg•year[-1])

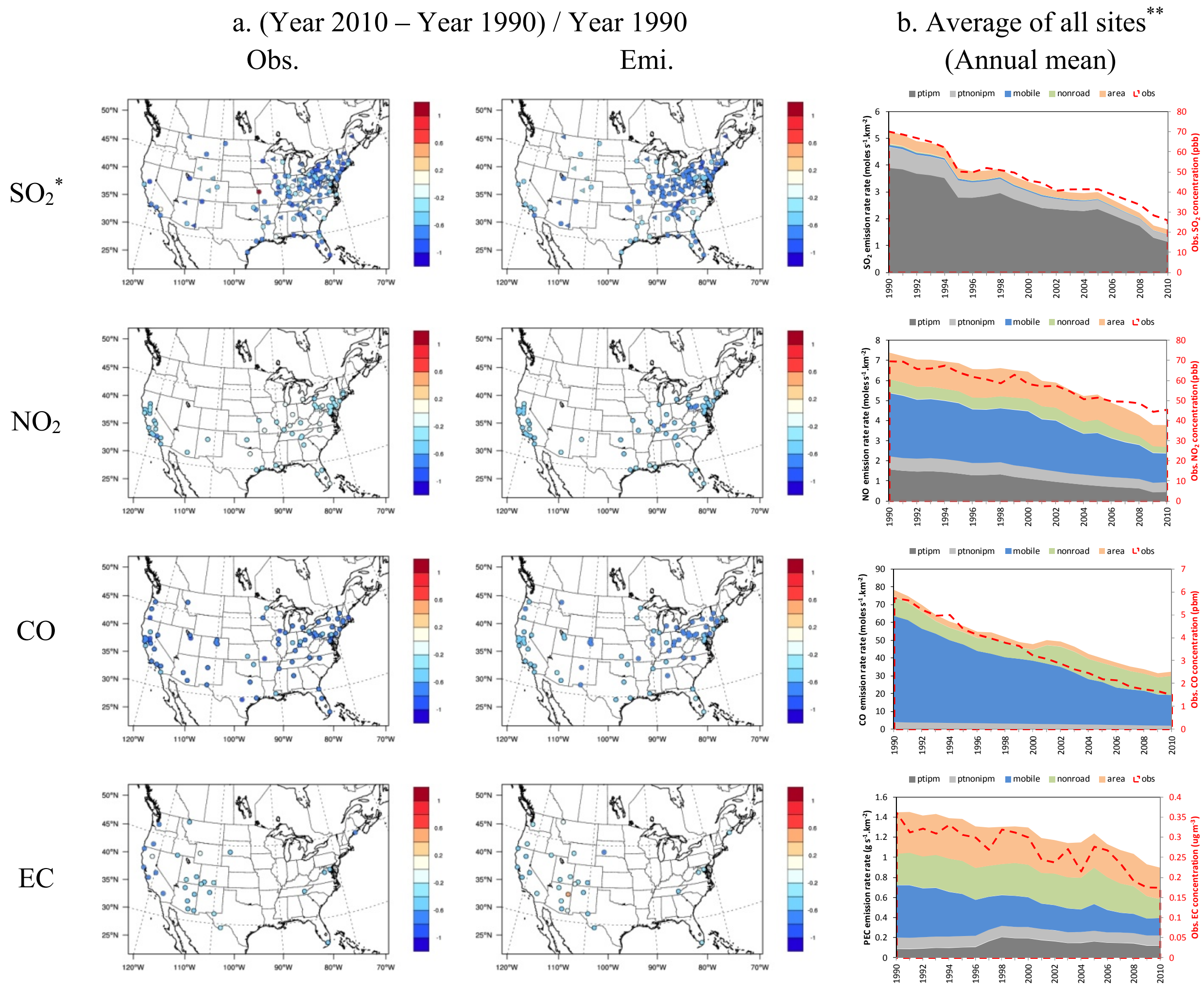


Fig. 10 Comparison of historic trends between emissions and observed concentration from 1990 to 2010 (*for SO_2, Circles represent AQS sites, Triangle represent CASTNET sites; ** SMOKE sectors include (1) ptipm- NEI point source EGUs mapped to the Integrated Planning Model using the National Electric Energy Database System database; (2) ptnonipm- All NEI point source records not matched to the ptipm sector; (3) mobile; (4) non-road and (5) other area sources.)

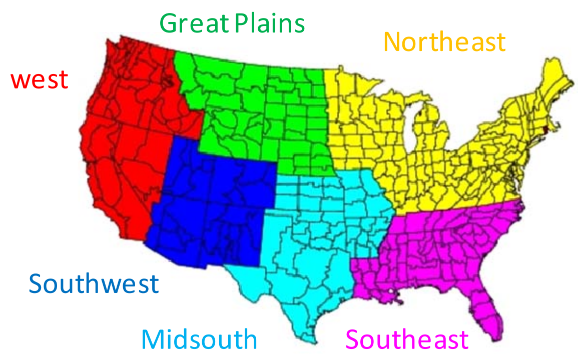
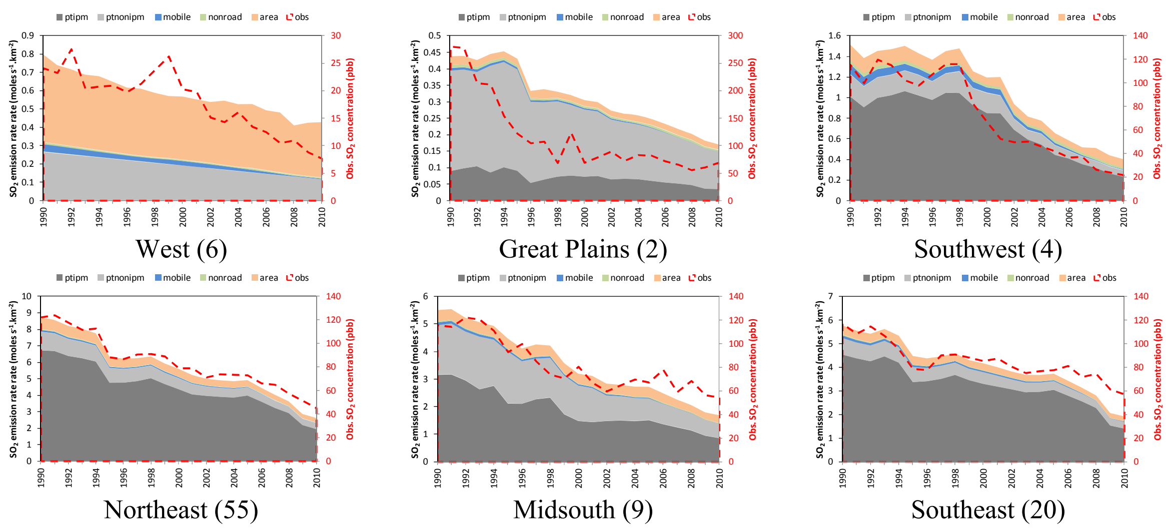
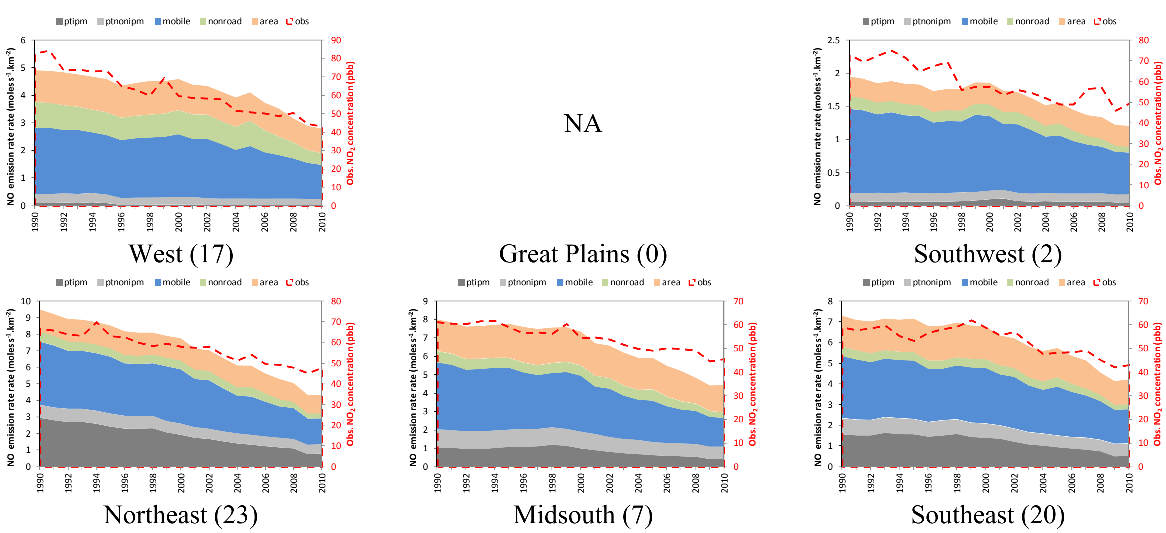


Fig. 11 Definition of sub-regions used in analysis



a. SO_2



b. NO_2

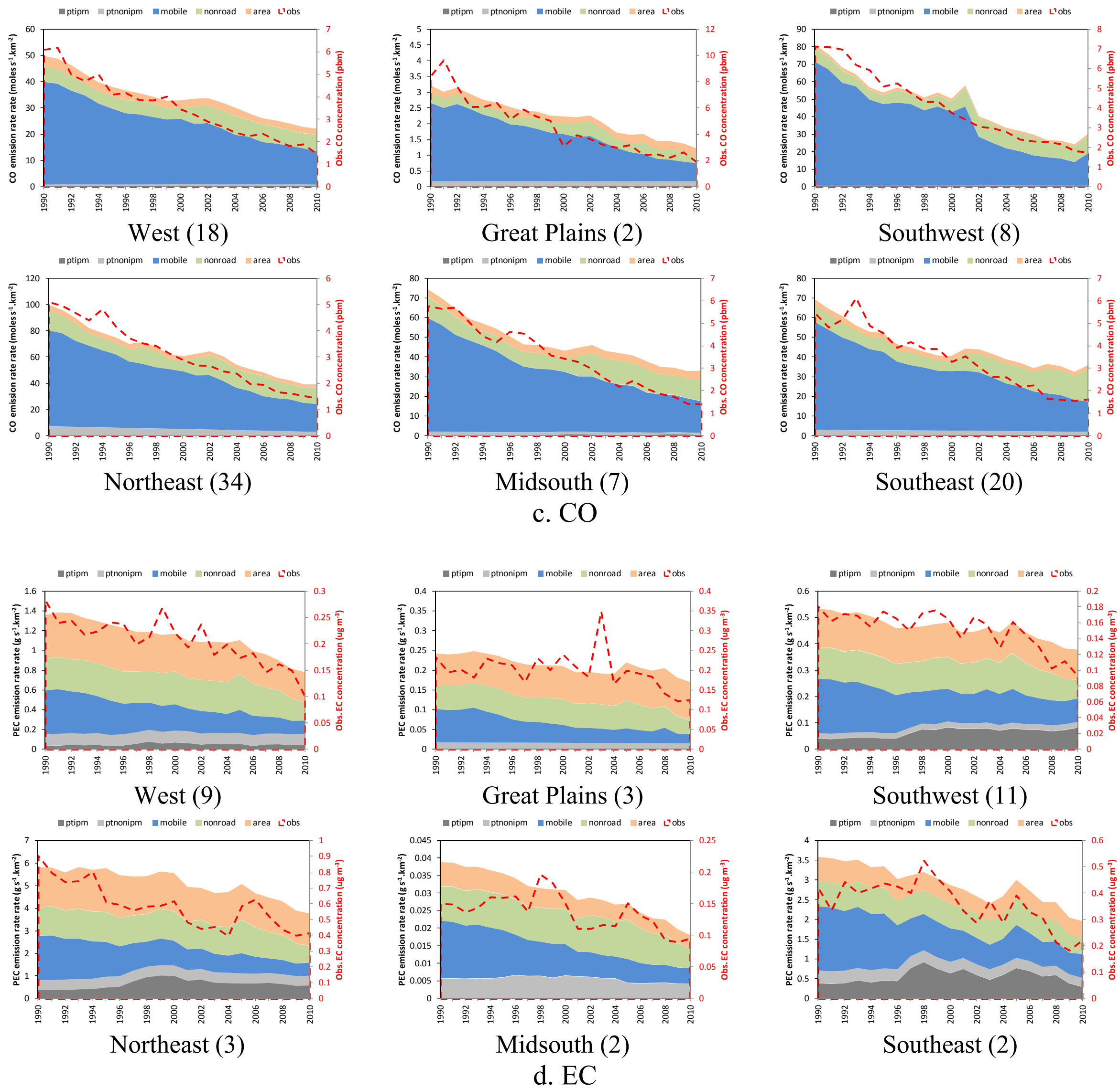


Fig. 12 Comparison of historic trends between emissions and observed concentration from 1990 to 2010 by sector and region (number of sites are shown in brackets)

Supporting Information for

Historical gaseous and primary aerosol emissions in the United States from 1990-2010

Table S1 Sectors grouped by NEI SCC in this study

(a) Power plants

Fuel	SCC	Explanation		Controlled species
Coal	101001XX	External Combustion Boilers	Anthracite Coal	NO_x, SO_2, PM
	101002XX		Bituminous/ Subbituminous Coal	
	101003XX		Lignite	
	2101002XXX	Electric Utility	Bituminous/ Subbituminous Coal	
Residual oil	101004XX	External Combustion Boilers	Residual Oil	NO_x, SO_2, PM
	2101005XXX	Electric Utility		
Distillate oil	101005XX	External Combustion Boilers;	Distillate Oil	SO_2, PM
	201001XX	Internal Combustion Engines		
	2101004XXX	Electric Utility		
Natural gas	101006XX	External Combustion Boilers	Natural Gas	NO_x
	201002XXX	Internal Combustion Engines		

(b) Industry combustion

Fuel	SCC	Explanation		Controlled species
Coal	102001XXX	External Combustion Boilers	Anthracite Coal	NO_x, SO_2, PM
	102002XXX		Bituminous/ Subbituminous Coal	
	102003XXX		Lignite	
	2102001XXX	Stationary Source	Anthracite Coal	
	2102002XXX		Bituminous/ Subbituminous Coal	
Residual oil	102004XXX	External Combustion Boilers	Residual Oil	SO_2, PM
	2102005XXX	Stationary Source		
Distillate oil	102005XXX	External Combustion Boilers	Distillate Oil	SO_2, PM
	202001XXX	Internal Combustion Engines		
	2102004XXX	Stationary Source		
Natural	102006XXX	External Combustion	Natural Gas	NO_x

gas		Boilers	
	202002XXX	Internal Combustion Engines	
	2102006XXX	Stationary Source	

(c) Commercial combustion

Fuel	SCC	Explanation		Controlled species
Coal	103001XXX	External Combustion Boilers	Anthracite Coal	SO_2, PM
	103002XXX		Bituminous/ Subbituminous Coal	
	2103001XXX	Stationary Source Fuel Combustion	Anthracite Coal	
	2103002XXX		Bituminous/ Subbituminous Coal	
Residual oil	103004XXX	External Combustion Boilers	Residual Oil	SO_2, PM
	2103005XXX	Stationary Source Fuel Combustion		
Distillate oil	103005XXX	External Combustion Boilers	Distillate Oil	SO_2, PM
	2103004XXX	Stationary Source Fuel Combustion		
Natural gas	103006XXX	External Combustion Boilers	Natural Gas	-
	2103006XXX	Stationary Source Fuel Combustion		
LPG	103010XXX	External Combustion Boilers	Liquified Petroleum Gas (LPG)	-
	2103007XXX	Stationary Source Fuel Combustion		
Kerosene	2103011XXX	Stationary Source Fuel Combustion	Kerosene	PM

(d) Residential combustion

Fuel	SCC	Explanation		Controlled species
Coal	2104001XXX	Stationary Source Fuel Combustion	Anthracite Coal	SO_2, PM
	2104002XXX		Bituminous/ Subbituminous Coal	
Distillate oil	2104004XXX		Distillate Oil	SO_2, PM
Natural	2104006XXX		Natural Gas	-

gas				
LPG	2104007XXX		Liquified Petroleum Gas (LPG)	-
Wood	2104008XXX		Wood	PM
Kerosene	2104011XXX		Kerosene	PM

(e) On road transportation

Type	SCC	Explanation	Controlled species
Light Duty Vehicle	220100XXXX	Light Duty Gasoline Vehicles	NO_x, SO_2, CO, VOC, PM
	223000XXXX	Light Duty Diesel Vehicles	
Light Duty Truck	220102XXXX	Light Duty Gasoline Trucks 1 & 2	NO_x, SO_2, CO, VOC, PM
	220104XXXX	Light Duty Gasoline Trucks 3 & 4	
	223006XXXX	Light Duty Diesel Trucks 1 thru 4 (M6) (LDDT)	
Heavy duty	220107XXXX	Heavy Duty Gasoline Vehicles	NO_x, SO_2, CO, VOC, PM
	223007XXXX	Heavy Duty Diesel Vehicles (HDDV) Class 2B	
Motorcycle	220108XXXX	Motorcycles (MC)	NO_x, SO_2, CO, VOC, PM

(f) Off road transportation

Fuel	Type	SCC	Explanation	Controlled species
Gasoline	Industry & Commercial	2260003XXXX 2265003XXXX 2265010XXXX	Industrial Equipment	SO_2
		2260006XXXX 2265006XXXX	Commercial Equipment	
	Construction	2260002XXXX 2265002XXXX	Construction and Mining Equipment	
	Agriculture	2260005XXXX 2265005XXXX	Agricultural Equipment	
	Marine, gasoline	2280004XXXX	Marine Vessels, Gasoline	
	Other	2260008XXXX 2265008XXXX	Airport Ground Support Equipment	
		2260001XXXX 2265001XXXX	Recreational Equipment	
		2260004XXXX 2265004XXXX	Lawn and Garden Equipment	
		2260007XXXX 2265007XXXX	Logging Equipment	

Distillate oil	Rail	2285XXXXXX	Class I Operations	NO_x, SO_2, PM
	Marine, diesel	2280002XXXX	Marine Vessels, Diesel	
	Other diesel	2270XXXXXX	Other diesel	
Residual oil	Marine	2280003XXXX	Marine Vessels, Residual oil	-
Natural gas	All Transport	2268XXXXXX	CNG	-
LPG	All Transport	2267XXXXXX	LPG	-
Jet fuel	Aviation	2275XXXXXX	Aircraft	-

(g) Industry processes

Category	SCC	Explanation	
Chemical Manufacturing	301XXXXX	Chemical Manufacturing	Point
	2301XXXXXX		Area
Metals processing	303XXXXX	Primary Metal Production	Point
	304XXXXX	Secondary Metal Production	
	309XXXXX	Fabricated Metal Products	
	2303XXXXXX	Primary Metal Production	Area
	2304XXXXXX	Secondary Metal Production	
	2309XXXXXX	Fabricated Metal Products	
Petroleum & related industries	306XXXXX	Petroleum Refining	Point
	310XXXXX	Oil and Gas Production	
	2306XXXXXX	Petroleum Refining	Area
	2310XXXXXX	Oil and Gas Production	
Other industry processes	302XXXXX	Food and Agriculture	Point
	305XXXXX	Mineral Products	
	307XXXXX	Pulp and Paper and Wood Products	
	308XXXXX	Rubber and Miscellaneous Plastics	
	311XXXXX	Building Construction	
	312XXXXX	Machinery	
	313XXXXX	Electrical Equipment	
	314XXXXX	Transportation Equipment	
	315XXXXX	Photo Equip/Health Care/Labs/Air Condit/SwimPools	
	32XXXXXX	Leather and Leather Products	
	33XXXXXX	Textile Products	
	36XXXXXX	Printing and Publishing	
	38XXXXXX	Cooling Tower	
	39XXXXXX	Miscellaneous	
	2302XXXXXX	Food and Kindred Products	Area
	2305XXXXXX	Mineral Processes	
	2307XXXXXX	Wood Products	
	2308XXXXXX	Rubber/Plastics	

	2312XXXXXX	Machinery
	2313XXXXXX	Electrical Equipment
	239XXXXXXX	Miscellaneous

(h) Others

Category	SCC	Explanation		Controlled species
Solvent Utilization	401XXXXX	Organic Solvent Evaporation	Point	VOC
	402XXXXX	Surface Coating Operations		
	405XXXXX	Printing/Publishing		
	410XXXXX	Dry Cleaning		
	425XXXXX	Loss		
	49XXXXXX	Organic Solvent Evaporation		
	24XXXXXXXX	Solvent Utilization	Area	
Storage & Transport	403XXXXX	Petroleum Product Storage at Refineries	Point	-
	404XXXXX	Petroleum Liquids Storage (non-Refinery)		
	406XXXXX	Transportation and Marketing of Petroleum Products		
	407XXXXX	Organic Chemical Storage		
	408XXXXX	Organic Chemical Transportation		
	25XXXXXXXX	Storage and Transport	Area	
Waste disposal & recycling	5XXXXXXX	Waste Disposal	Point	-
	26XXXXXXXX	Waste Disposal, Treatment, and Recovery	Area	
Miscellaneous	6XXXXXXX	MACT Source Categories	Point	-
	2294XXXXXX	All Paved Roads	Mobile	PM
	2296XXXXXX	All Unpaved Roads		
	2311XXXXXX	Construction	Area	PM
	2325XXXXXX	Mining and Quarrying		-
	2801XXXXXX	Agriculture Production - Crops		-
	2805XXXXXX	Agriculture Production - Livestock		NH_3
	2810001XXX	Forest Wildfires		-
	2810015XXX	Prescribed Burning for Forest Management		-
	2810XXXXXX	Other Combustion		-
	2820XXXXXX	Cooling Towers		-
	2830XXXXXX	Catastrophic/Accidental Releases		-
	284XXXXXXX	Repair Shops		-
	285XXXXXXX	Health Services		-

Table S2 Activity and emission trends during 1990-2010

(a) energy-related stationary sources (absolute values for Fig. 3)

Power plant			1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Coal	Activity		16.259	16.248	16.464	17.194	17.259	17.465	18.428	18.903	19.216	19.279	20.220	19.614	19.783	20.185	20.305	20.737	20.461	20.807	20.513	18.226	19.133
	NOx	NEI	5.638					5.579	5.539			4.909		4.208	4.150			3.335					
		This study	5.837	5.694	5.677	5.843	5.753	5.629	5.550	5.637	5.553	4.953	4.667	4.325	4.156	3.886	3.604	3.335	3.158	2.981	2.811	1.878	1.943
	SO2	NEI	15.219					11.603	12.136			11.746		10.004	9.970			9.936					
		This study	15.565	15.396	14.963	14.853	14.511	11.982	12.274	12.696	12.873	11.839	10.941	10.341	10.134	10.029	9.926	9.935	9.153	8.328	7.442	5.528	4.970
Natural Gas	Activity		3.333	3.399	3.535	3.559	4.000	4.327	3.882	4.147	4.698	4.924	5.318	5.496	5.789	5.259	5.609	6.036	6.394	7.028	6.849	7.044	7.550
	NOx	NEI	0.691					0.650	0.344			0.519		0.445	0.292			0.217					
		This study	0.414	0.405	0.417	0.417	0.467	0.487	0.354	0.362	0.420	0.425	0.409	0.375	0.276	0.210	0.219	0.208	0.224	0.249	0.241	0.247	0.271
	SO2	NEI	0.002					0.013	0.008			0.179		0.211	0.060			0.060					
		This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.002	0.001	0.001	0.002	0.002	0.002	0.002	0.002	0.002
Distillate fuel oil	Activity		0.097	0.084	0.074	0.086	0.120	0.108	0.109	0.111	0.136	0.140	0.175	0.171	0.127	0.161	0.111	0.115	0.074	0.089	0.073	0.070	0.080
	NOx	NEI	0.084					0.033	0.029			0.054		0.052	0.084			0.078					
		This study	0.015	0.012	0.011	0.013	0.018	0.016	0.016	0.016	0.020	0.021	0.026	0.025	0.019	0.023	0.016	0.016	0.010	0.012	0.010	0.010	0.011
	SO2	NEI	0.057					0.056	0.056			0.042		0.046	0.029			0.030					
		This study	0.031	0.025	0.021	0.026	0.037	0.033	0.031	0.026	0.035	0.031	0.036	0.029	0.016	0.017	0.013	0.013	0.009	0.011	0.009	0.008	0.009
Residual fuel oil	Activity		1.163	1.085	0.872	0.959	0.869	0.566	0.628	0.715	1.047	0.959	0.871	1.003	0.659	0.869	0.879	0.876	0.361	0.397	0.240	0.181	0.154
	NOx	NEI	0.192					0.094	0.094			0.185		0.156	0.129			0.108					
		This study	0.165	0.154	0.124	0.136	0.123	0.080	0.089	0.098	0.143	0.131	0.119	0.134	0.091	0.105	0.105	0.096	0.036	0.040	0.022	0.015	0.012
	SO2	NEI	0.603					0.408	0.410			0.559		0.492	0.345			0.369					
		This study	0.520	0.495	0.396	0.433	0.389	0.251	0.278	0.312	0.459	0.421	0.379	0.422	0.287	0.339	0.339	0.313	0.116	0.130	0.072	0.046	0.037

Industrial Combustion			1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Coal	Activity		2.754	2.600	2.512	2.500	2.507	2.500	2.438	2.396	2.254	2.188	2.259	2.194	2.020	2.044	2.046	1.954	1.914	1.864	1.792	1.394	1.666
	NOx	NEI	0.511					0.560	0.485			0.407		0.419	0.343			0.316					
		This study	0.694	0.665	0.633	0.634	0.643	0.632	0.569	0.560	0.576	0.487	0.524	0.477	0.355	0.342	0.325	0.317	0.293	0.262	0.239	0.246	0.263
	SO2	NEI	0.082					0.091	0.103			0.109		0.113	0.126			0.119					
		This study	2.368	2.323	2.111	2.126	2.160	2.076	1.714	1.695	1.656	1.494	1.541	1.461	1.120	1.093	1.091	1.025	0.918	0.762	0.644	0.662	0.748
Natural Gas	Activity		8.520	8.637	8.996	9.129	9.202	9.678	9.999	10.109	9.882	9.438	9.550	8.674	8.865	8.510	8.573	7.930	7.881	8.098	8.102	7.629	7.982
	NOx	NEI	1.648					1.775	1.844			1.543		1.583	1.071			0.917					
		This study	1.023	1.023	1.065	1.085	1.091	1.136	1.144	1.149	1.119	1.062	1.064	0.959	0.886	0.817	0.819	0.731	0.727	0.748	0.748	0.704	0.738
	SO2	NEI	0.031					0.035	0.046			0.060		0.061	0.035			0.034					
		This study	0.002	0.002	0.002	0.002	0.002	0.003	0.003	0.003	0.003	0.003	0.003	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.002	0.002
Distillate fuel oil	Activity		1.150	1.078	1.107	1.117	1.111	1.131	1.187	1.203	1.211	1.187	1.200	1.300	1.204	1.136	1.214	1.264	1.263	1.265	1.277	1.107	1.188
	NOx	NEI	0.080					0.086	0.077			0.099		0.111	0.113			0.107					
		This study	0.125	0.117	0.121	0.122	0.121	0.123	0.129	0.131	0.132	0.129	0.131	0.142	0.131	0.124	0.132	0.138	0.137	0.138	0.139	0.121	0.129
	SO2	NEI	0.181					0.177	0.153			0.148		0.163	0.135			0.131					
		This study	0.242	0.215	0.225	0.228	0.225	0.220	0.219	0.214	0.218	0.202	0.201	0.212	0.147	0.127	0.136	0.131	0.133	0.136	0.137	0.120	0.128
Residual fuel oil	Activity		0.411	0.334	0.387	0.446	0.419	0.337	0.335	0.291	0.230	0.207	0.241	0.203	0.190	0.220	0.249	0.281	0.239	0.193	0.199	0.106	0.120
	NOx	NEI	0.158					0.147	0.133			0.099		0.111	0.056			0.056					
		This study	0.068	0.055	0.064	0.074	0.069	0.056	0.055	0.048	0.038	0.035	0.040	0.034	0.032	0.037	0.041	0.047	0.040	0.032	0.033	0.018	0.019
	SO2	NEI	0.651					0.683	0.623			0.360		0.396	0.185			0.174					
		This study	0.195	0.158	0.183	0.211	0.198	0.159	0.158	0.138	0.109	0.098	0.114	0.096	0.086	0.086	0.101	0.101	0.084	0.073	0.072	0.037	0.037

Commercial Combustion			1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Coal	Activity		0.124	0.115	0.117	0.117	0.117	0.116	0.120	0.129	0.101	0.102	0.086	0.088	0.088	0.083	0.103	0.096	0.064	0.070	0.072	0.065	0.062
	NOx	NEI	0.035					0.038	0.035			0.036		0.038	0.034			0.032					
		This study	0.037	0.034	0.035	0.035	0.035	0.035	0.036	0.038	0.030	0.029	0.026	0.026	0.026	0.025	0.031	0.029	0.019	0.021	0.021	0.019	0.018
	SO2	NEI	0.211					0.199	0.183			0.145		0.147	0.129			0.126					
		This study	0.225	0.202	0.198	0.205	0.207	0.206	0.216	0.224	0.172	0.160	0.144	0.143	0.126	0.118	0.135	0.110	0.079	0.092	0.095	0.087	0.084
Natural Gas	Activity		2.698	2.807	2.883	2.944	2.978	3.117	3.251	3.306	3.098	3.132	3.261	3.109	3.223	3.271	3.211	3.083	2.908	3.095	3.235	3.199	3.172
	NOx	NEI	0.123					0.162	0.162			0.170		0.180	0.163			0.162					
		This study	0.147	0.153	0.157	0.160	0.162	0.170	0.177	0.180	0.169	0.170	0.178	0.169	0.175	0.178	0.175	0.168	0.158	0.168	0.176	0.174	0.173
	SO2	NEI	0.006					0.007	0.008			0.010		0.013	0.002			0.002					
		This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Distillate fuel oil	Activity		0.536	0.517	0.507	0.493	0.501	0.479	0.483	0.444	0.429	0.438	0.491	0.508	0.444	0.481	0.470	0.447	0.401	0.384	0.372	0.413	0.410
	NOx	NEI	0.050					0.049	0.049			0.042		0.044	0.050			0.045					
		This study	0.041	0.040	0.039	0.038	0.039	0.037	0.037	0.034	0.033	0.034	0.038	0.039	0.034	0.037	0.036	0.035	0.031	0.030	0.029	0.032	0.032
	SO2	NEI	0.238					0.222	0.196			0.154		0.161	0.128			0.121					
		This study	0.209	0.199	0.193	0.189	0.192	0.178	0.168	0.149	0.145	0.143	0.154	0.160	0.096	0.095	0.095	0.083	0.076	0.071	0.072	0.080	0.083
Residual fuel oil	Activity		0.230	0.212	0.189	0.173	0.172	0.141	0.137	0.111	0.085	0.073	0.092	0.070	0.080	0.111	0.122	0.116	0.075	0.075	0.073	0.076	0.077
	NOx	NEI	0.035					0.041	0.026			0.014		0.015	0.021			0.022					
		This study	0.037	0.035	0.030	0.029	0.028	0.023	0.023	0.018	0.012	0.012	0.015	0.012	0.013	0.018	0.020	0.019	0.013	0.013	0.012	0.012	0.012
	SO2	NEI	0.186					0.174	0.142			0.098		0.102	0.093			0.093					
		This study	0.093	0.087	0.074	0.068	0.068	0.057	0.055	0.045	0.030	0.029	0.036	0.027	0.030	0.037	0.039	0.033	0.023	0.023	0.022	0.022	0.023

Residential Combustion			1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Coal	Activity		0.031	0.025	0.026	0.026	0.021	0.017	0.016	0.016	0.012	0.014	0.011	0.011	0.012	0.012	0.011	0.008	0.006	0.008	0.008	0.008	0.008
	NOx	NEI	0.106					0.111	0.100			0.007		0.007	0.003			0.003					
		This study	0.009	0.008	0.008	0.008	0.006	0.005	0.005	0.005	0.004	0.004	0.003	0.003	0.004	0.004	0.003	0.002	0.002	0.002	0.002	0.002	0.002
	SO2	NEI	0.371					0.029	0.021			0.052		0.052	0.021			0.021					
		This study	0.070	0.057	0.058	0.056	0.045	0.038	0.035	0.035	0.027	0.030	0.022	0.022	0.016	0.018	0.015	0.010	0.009	0.011	0.009	0.009	0.009
Natural Gas	Activity		4.519	4.684	4.820	5.098	4.981	4.984	5.391	5.125	4.671	4.857	5.104	4.902	5.006	5.224	4.993	4.958	4.483	4.849	5.018	4.899	4.893
	NOx	NEI	0.326					0.519	0.481			0.219		0.221	0.240			0.241					
		This study	0.246	0.255	0.262	0.277	0.271	0.271	0.293	0.279	0.254	0.264	0.278	0.267	0.272	0.284	0.272	0.270	0.244	0.264	0.273	0.267	0.266
	SO2	NEI	0.001					0.002	0.002			0.001		0.001	0.001			0.001					
		This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Distillate fuel oil	Activity		0.978	0.930	0.980	0.974	0.960	0.905	0.926	0.874	0.772	0.828	0.905	0.908	0.860	0.905	0.924	0.854	0.712	0.726	0.669	0.602	0.583
	NOx	NEI	0.191					0.210	0.194			0.053		0.053	0.062			0.062					
		This study	0.057	0.054	0.057	0.057	0.056	0.053	0.054	0.051	0.045	0.048	0.053	0.053	0.050	0.053	0.054	0.050	0.042	0.042	0.039	0.035	0.034
	SO2	NEI	0.962					0.144	0.108			0.125		0.126	0.135			0.135					
		This study	0.333	0.251	0.262	0.260	0.255	0.172	0.164	0.154	0.135	0.144	0.157	0.157	0.142	0.145	0.147	0.133	0.111	0.112	0.104	0.093	0.091

Activity – energy combustion, unit: 10^6 MMBtu year^{-1}

Emission – unit: Tg year^{-1}

(b) on-road mobile sources (absolute values for Fig. 4)

Light Duty Vehicle		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		1.404	1.360	1.367	1.375	1.407	1.438	1.466	1.505	1.556	1.563	1.603	1.624	1.658	1.669	1.705	1.707	1.692	1.742	1.776	1.765	1.731
NOx	NEI							3.001			2.421		2.512	2.295			1.986					
	This study	4.529	3.990	3.682	3.410	3.229	3.031	2.840	2.751	2.675	2.485	2.427	2.385	2.295	2.189	1.964	1.807	1.537	1.461	1.370	1.238	1.138
CO	NEI							28.573			43.816		39.590	36.482			21.699					
	This study	81.111	71.175	64.940	59.222	54.969	51.045	47.127	44.698	43.008	40.409	39.145	38.160	36.482	34.844	30.425	27.712	23.709	23.055	22.220	20.934	19.926
SO2	NEI							0.129			0.101		0.084	0.081			0.034					
	This study	0.173	0.160	0.149	0.137	0.125	0.114	0.102	0.102	0.101	0.101	0.087	0.083	0.081	0.070	0.059	0.048	0.036	0.025	0.014	0.013	0.012
VOC	NEI							2.851			3.126		2.377	2.783			1.861					
	This study	5.916	5.210	4.779	4.400	4.153	3.920	3.664	3.521	3.411	3.230	3.104	2.976	2.783	2.611	2.375	2.166	1.934	1.829	1.715	1.555	1.412
PM10	NEI							0.062			0.051		0.047	0.045			0.032					
	This study	0.088	0.084	0.080	0.076	0.072	0.069	0.065	0.062	0.058	0.054	0.052	0.048	0.045	0.044	0.044	0.043	0.041	0.038	0.036	0.031	0.026
PM2.5	NEI							0.038			0.029		0.026	0.024			0.010					
	This study	0.054	0.052	0.049	0.046	0.043	0.041	0.038	0.036	0.033	0.031	0.029	0.026	0.024	0.024	0.023	0.023	0.021	0.019	0.018	0.016	0.015

Light Duty Truck		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		0.578	0.644	0.698	0.731	0.756	0.776	0.802	0.834	0.860	0.889	0.910	0.929	0.955	0.970	1.012	1.022	1.055	1.089	1.083	1.082	1.081
NOx	NEI							1.962			1.478		1.903	1.925			2.549					
	This study	2.068	2.129	2.158	2.106	2.049	1.954	1.896	1.883	1.867	1.874	1.902	1.925	1.925	1.893	1.794	1.662	1.534	1.480	1.360	1.270	1.175
CO	NEI							19.282			30.659		32.404	31.732			29.613					
	This study	45.993	47.361	47.460	45.858	43.839	41.383	39.191	38.090	36.817	35.439	34.290	33.197	31.732	29.127	26.435	23.090	20.887	20.298	18.689	17.481	16.464
SO2	NEI							0.096			0.067		0.069	0.074			0.037					
	This study	0.153	0.143	0.133	0.122	0.113	0.103	0.092	0.092	0.091	0.091	0.078	0.075	0.074	0.064	0.054	0.044	0.033	0.023	0.013	0.012	0.010
VOC	NEI							2.065			1.989		1.920	2.104			2.195					
	This study	3.045	3.109	3.074	2.959	2.841	2.696	2.546	2.482	2.405	2.347	2.293	2.221	2.104	1.981	1.834	1.620	1.514	1.460	1.297	1.199	1.114
PM10	NEI							0.043			0.034		0.033	0.035			0.034					
	This study	0.067	0.064	0.061	0.058	0.055	0.052	0.050	0.047	0.044	0.041	0.039	0.036	0.035	0.034	0.034	0.033	0.031	0.029	0.028	0.024	0.020
PM2.5	NEI							0.028			0.020		0.019	0.020			0.011					
	This study	0.044	0.042	0.040	0.038	0.036	0.034	0.031	0.030	0.027	0.025	0.024	0.021	0.020	0.020	0.019	0.019	0.017	0.016	0.014	0.013	0.012

Heavy Duty		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		0.152	0.155	0.159	0.166	0.177	0.185	0.190	0.198	0.203	0.210	0.213	0.216	0.221	0.225	0.228	0.229	0.247	0.258	0.266	0.269	0.268
NOx	NEI							2.873			4.585		4.295	4.149			4.980					
	This study	4.353	4.241	4.214	4.299	4.543	4.707	4.729	4.874	4.928	4.896	4.656	4.352	4.149	3.834	3.479	3.205	3.149	3.011	2.774	2.573	2.299
CO	NEI							5.219			6.056		4.552	3.941			5.291					
	This study	9.226	8.695	8.165	7.764	7.558	7.156	6.563	6.121	5.641	5.200	4.716	4.269	3.941	3.418	3.117	2.860	2.703	2.632	2.444	2.038	1.911
SO2	NEI							0.091			0.118		0.117	0.082			0.101					
	This study	0.172	0.161	0.150	0.137	0.128	0.117	0.104	0.104	0.102	0.102	0.088	0.083	0.082	0.072	0.061	0.050	0.038	0.026	0.014	0.013	0.012
VOC	NEI							0.538			0.575		0.471	0.420			0.549					
	This study	0.796	0.750	0.690	0.661	0.640	0.619	0.580	0.555	0.521	0.496	0.466	0.438	0.420	0.390	0.370	0.340	0.328	0.318	0.300	0.275	0.255
PM10	NEI							0.177			0.155		0.136	0.121			0.038					
	This study	0.236	0.226	0.216	0.205	0.198	0.188	0.176	0.167	0.156	0.146	0.138	0.127	0.121	0.120	0.118	0.115	0.109	0.103	0.096	0.084	0.071
PM2.5	NEI							0.155			0.135		0.118	0.104			0.020					
	This study	0.233	0.223	0.211	0.199	0.191	0.180	0.166	0.156	0.143	0.132	0.123	0.111	0.104	0.102	0.099	0.096	0.089	0.082	0.075	0.070	0.065

Motorcycle		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Activity		0.010	0.009	0.010	0.010	0.010	0.009	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.010	0.011	0.011	0.012	0.013	0.014	0.014	0.014
NOx	NEI							0.012			0.015		0.019	0.021			0.020					
	This study	0.022	0.021	0.022	0.023	0.023	0.021	0.022	0.022	0.023	0.023	0.023	0.021	0.021	0.022	0.023	0.023	0.026	0.029	0.030	0.030	0.032
CO	NEI							0.189			0.173		0.209	0.225			0.691					
	This study	0.255	0.234	0.249	0.251	0.256	0.235	0.245	0.236	0.245	0.239	0.240	0.221	0.225	0.226	0.246	0.248	0.282	0.308	0.318	0.317	0.327
SO2	NEI							0.000			0.000		0.000	0.000			0.000					
	This study	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
VOC	NEI							0.036			0.028		0.028	0.032			0.067					
	This study	0.042	0.037	0.039	0.038	0.038	0.035	0.036	0.034	0.036	0.035	0.034	0.032	0.032	0.033	0.035	0.035	0.040	0.044	0.046	0.046	0.047
PM10	NEI							0.000			0.000		0.000	0.000			0.000					
	This study	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
PM2.5	NEI							0.000			0.000		0.000	0.000			0.000					
	This study	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

Activity – vehicle miles traveled, unit: 10^{12} miles year^{-1}

Emission – unit: Tg year^{-1}

(c) industrial process sources (absolute values for Fig. 6)

Chemical Manufacturing		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.186					0.186	0.169			0.123		0.129	0.096			0.085					
	This study	0.233	0.226	0.218	0.210	0.203	0.195	0.187	0.180	0.172	0.164	0.157	0.149	0.141	0.134	0.126	0.118	0.111	0.103	0.095	0.088	0.080
CO	NEI	1.266					1.245	1.182			0.478		0.508	0.385			0.291					
	This study	1.655	1.585	1.515	1.444	1.374	1.303	1.233	1.162	1.092	1.021	0.951	0.881	0.810	0.740	0.669	0.599	0.528	0.458	0.388	0.317	0.247
SO2	NEI	0.312					0.300	0.301			0.291		0.304	0.265			0.261					
	This study	0.310	0.304	0.299	0.293	0.288	0.282	0.277	0.271	0.266	0.260	0.255	0.249	0.244	0.239	0.233	0.228	0.222	0.217	0.211	0.206	0.200
VOC	NEI	0.618					0.688	0.410			0.260		0.270	0.261			0.248					
	This study	0.669	0.644	0.618	0.593	0.567	0.542	0.516	0.491	0.465	0.440	0.414	0.389	0.363	0.338	0.312	0.287	0.261	0.236	0.210	0.185	0.159
PM10	NEI	0.078					0.069	0.067			0.057		0.060	0.043			0.039					
	This study	0.083	0.081	0.078	0.076	0.073	0.071	0.068	0.065	0.063	0.060	0.058	0.055	0.052	0.050	0.047	0.045	0.042	0.040	0.037	0.034	0.032
PM2.5	NEI	0.048					0.044	0.041			0.048		0.050	0.032			0.032					
	This study	0.051	0.050	0.049	0.048	0.047	0.045	0.044	0.043	0.042	0.041	0.039	0.038	0.037	0.036	0.035	0.033	0.032	0.031	0.030	0.029	0.027

Metals processing		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.126					0.133	0.104			0.109		0.121	0.090			0.081					
	This study	0.119	0.118	0.116	0.114	0.113	0.111	0.110	0.108	0.107	0.105	0.104	0.102	0.101	0.099	0.097	0.096	0.094	0.093	0.091	0.090	0.088
CO	NEI	2.642					2.400	1.771			1.358		1.498	1.041			0.883					
	This study	2.812	2.714	2.616	2.518	2.420	2.322	2.225	2.127	2.029	1.931	1.833	1.736	1.638	1.540	1.442	1.344	1.246	1.149	1.051	0.953	0.855
SO2	NEI	0.729					0.535	0.447			0.323		0.320	0.229			0.189					
	This study	0.787	0.756	0.726	0.695	0.665	0.634	0.604	0.573	0.542	0.512	0.481	0.451	0.420	0.390	0.359	0.329	0.298	0.267	0.237	0.206	0.176
VOC	NEI	0.130					0.136	0.086			0.082		0.088	0.057			0.059					
	This study	0.146	0.141	0.137	0.132	0.127	0.122	0.118	0.113	0.108	0.104	0.099	0.094	0.089	0.085	0.080	0.075	0.070	0.066	0.061	0.056	0.051
PM10	NEI	0.231					0.247	0.207			0.154		0.167	0.090			0.091					
	This study	0.234	0.226	0.218	0.210	0.202	0.194	0.186	0.178	0.170	0.162	0.154	0.147	0.139	0.131	0.123	0.115	0.107	0.099	0.091	0.083	0.075
PM2.5	NEI	0.164					0.151	0.129			0.126		0.137	0.058			0.066					
	This study	0.172	0.167	0.161	0.156	0.150	0.145	0.140	0.134	0.129	0.123	0.118	0.112	0.107	0.101	0.096	0.091	0.085	0.080	0.074	0.069	0.063

Petroleum & related industries		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.301					0.267	0.282			0.244		0.244	0.453			0.556					
	This study	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564	0.564
CO	NEI	0.363					0.427	0.459			0.200		0.201	0.407			0.449					
	This study	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457	0.457
SO2	NEI	0.643					0.538	0.520			0.470		0.484	0.375			0.281					
	This study	0.608	0.588	0.568	0.548	0.527	0.507	0.487	0.466	0.446	0.426	0.406	0.385	0.365	0.345	0.324	0.304	0.284	0.264	0.243	0.223	0.203
VOC	NEI	0.617					0.658	0.505			0.466		0.450	0.625			0.957					
	This study	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023	1.023
PM10	NEI	0.030					0.033	0.028			0.038		0.036	0.027			0.034					
	This study	0.066	0.064	0.063	0.061	0.059	0.057	0.055	0.054	0.052	0.050	0.048	0.046	0.045	0.043	0.041	0.039	0.037	0.036	0.034	0.032	0.030
PM2.5	NEI	0.022					0.024	0.021			0.033		0.031	0.022			0.029					
	This study	0.036	0.036	0.036	0.035	0.035	0.035	0.034	0.034	0.033	0.033	0.033	0.032	0.032	0.032	0.031	0.031	0.030	0.030	0.030	0.029	0.029

Other industry processes		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
NOx	NEI	0.487					0.534	0.526			0.558		0.612	0.516			0.556					
	This study	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495	0.495
CO	NEI	0.590					0.688	0.649			0.631		0.710	0.584			0.626					
	This study	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575	0.575
SO2	NEI	0.552					0.578	0.480			0.453		0.496	0.378			0.400					
	This study	0.442	0.435	0.427	0.420	0.412	0.405	0.398	0.390	0.383	0.375	0.368	0.361	0.353	0.346	0.338	0.331	0.324	0.316	0.309	0.301	0.294
VOC	NEI	0.408					0.463	0.445			0.445		0.434	0.451			0.467					
	This study	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452	0.452
PM10	NEI	0.734					0.633	0.449			0.462		0.535	0.411			0.437					
	This study	0.253	0.249	0.246	0.242	0.239	0.235	0.231	0.228	0.224	0.221	0.217	0.214	0.210	0.206	0.203	0.199	0.196	0.192	0.188	0.185	0.181
PM2.5	NEI	0.346					0.315	0.242			0.306		0.365	0.253			0.284					
	This study	0.209	0.207	0.205	0.203	0.201	0.198	0.196	0.194	0.192	0.190	0.188	0.186	0.184	0.182	0.180	0.178	0.175	0.173	0.171	0.169	0.167

Emission – unit: Tg $year^{-1}$

(d) other non-energy related sources (absolute values for Fig. 7)

		1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
(a) Solvent utilization (NMVOC)	Activity	1.093	1.038	1.060	1.144	1.212	1.192	1.248	1.263	1.283	1.311	1.281	1.235	1.291	1.326	1.362	1.381	1.360	1.353	1.201	1.089	1.153
	NEI data	4.400					6.186	5.541			5.039		5.016	4.282			4.247					
	NEI trend	2.099	2.110	2.154	2.196	2.249	2.257	1.999	2.052	1.879	1.838	1.763	1.829	1.561	1.557	1.553	1.549	1.415	1.280	1.145	1.040	0.935
	This study	5.752	5.422	5.538	5.975	6.330	6.186	5.479	5.191	5.276	5.038	5.057	5.014	4.280	4.231	4.349	4.247	4.183	4.163	3.695	3.349	3.243
(b) Fertilizer Application (NH3)	Activity	12.671	12.953	13.561	12.286	13.494	12.814	13.979	13.672	13.845	13.516	14.156	13.440	13.589	13.784	13.610	14.113	13.080	14.760	13.784	13.673	14.716
	NEI data	0.420					0.219	0.656			0.726		0.727	1.164			1.164					
	This study	1.086	1.134	1.176	1.076	1.161	1.123	1.233	1.189	1.224	1.188	1.238	1.168	1.164	1.172	1.162	1.187	1.101	1.214	1.166	1.145	1.211
(c) Livestock (NH3)	Activity	0.199	0.202	0.210	0.210	0.215	0.217	0.215	0.215	0.228	0.231	0.228	0.228	0.233	0.231	0.234	0.235	0.238	0.243	0.253	0.253	0.246
	NEI data	3.160					1.784	3.451			3.536		2.419	2.112			2.112					
	This study	2.024	2.030	2.087	2.091	2.121	2.122	2.082	2.069	2.134	2.161	2.123	2.108	2.135	2.112	2.119	2.112	2.130	2.149	2.196	2.189	2.141
(d) Constructio n processes (PM10)	Activity	0.260	0.282	0.310	0.337	0.355	0.369	0.390	0.407	0.434	0.459	0.486	0.498	0.482	0.503	0.530	0.566	0.595	0.632	0.601	0.525	0.453
	NEI data	4.249					3.654	0.960			1.923		0.919	0.894			0.894					
	This study	1.151	1.000	1.064	1.184	1.219	1.069	1.053	0.998	1.011	0.994	0.971	0.961	0.935	0.881	0.900	0.894	0.963	1.028	1.033	0.985	0.920
(e) Mining & quarrying (PM10)	Activity	2.897	2.961	3.071	3.048	3.113	3.257	3.357	3.514	3.626	3.308	3.269	3.369	3.410	3.450	4.062	4.156	4.265	4.219	3.983	3.507	4.094
	NEI data	0.321					0.573	0.550			0.708		0.365	0.608			0.608					
	This study	0.462	0.472	0.489	0.486	0.496	0.507	0.519	0.531	0.557	0.550	0.542	0.560	0.550	0.562	0.593	0.608	0.630	0.604	0.560	0.498	0.560
(f) Paved/Unp aved road (PM10)	Activity	2.137	2.174	2.254	2.310	2.373	2.436	2.498	2.582	2.651	2.705	2.770	2.812	2.878	2.912	2.995	3.011	3.048	3.168	3.185	3.139	3.090
	NEI data	13.353					12.752	3.578			11.443		5.470	5.169			5.169					
	This study	13.353	12.919	12.486	12.052	11.618	11.185	10.751	10.317	9.884	9.450	9.016	8.583	8.149	7.715	7.282	6.848	6.414	5.981	5.547	5.113	4.680
(g) Agriculture tilling (PM10)	Activity	0.167	0.174	0.177	0.166	0.173	0.167	0.180	0.176	0.175	0.175	0.176	0.173	0.175	0.173	0.173	0.173	0.168	0.183	0.175	0.172	0.174
	NEI data	4.826					4.441	1.191			4.482		3.391	2.491			2.491					
	This study	2.418	2.521	2.529	2.375	2.480	2.403	2.625	2.557	2.548	2.544	2.576	2.533	2.551	2.513	2.514	2.491	2.416	2.653	2.541	2.485	2.513
(h) Forest wildfires (PM10)	Activity	1.664	1.063	0.745	0.647	1.466	0.663	2.184	1.028	0.876	1.988	1.189	1.645	2.686	2.875	2.462	2.306	2.721	3.148	1.942	2.531	2.318
	NEI data	0.447					0.487	0.530			0.310		0.287	0.171			0.171					

	This study	0.165	0.105	0.074	0.064	0.145	0.066	0.216	0.102	0.087	0.197	0.118	0.163	0.171	0.283	0.297	0.253	0.282	0.349	0.282	0.294	0.369
(i) Prescribed Forest Management (PM10)	Activity	4.602	2.941	2.061	1.790	4.057	1.833	6.041	2.845	1.324	5.603	7.363	3.556	7.186	3.962	8.098	8.686	9.871	9.302	5.254	5.922	3.233
	NEI data	0.202					0.145	0.000			0.683		1.196	0.800			0.800					
	This study	0.712	0.455	0.319	0.277	0.627	0.283	0.934	0.440	0.205	0.866	1.139	0.550	0.726	0.640	0.915	1.055	1.909	1.876	1.063	0.865	0.470

Activity –

(a) Estimated Quantity of Shipments of Paint and Allied Products, unit: 10^9 gallons year^{-1}

(b) Fertilizer (NITROGEN) Application, unit: 10^9 lb year^{-1}

(c) Weighted sum of livestock, unit: 10^9 Head year^{-1}

(d) Value of Construction Put in Place, unit: 10^{12} Head dollars year^{-1}

(e) Weighted sum of crude ore and coal handled on surface mine, unit: 10^9 Ton year^{-1}

(f) Vehicle miles traveled, unit: 10^{12} miles year^{-1}

(g) Weighted sum of number of acres harvested, unit: 10^9 acres year^{-1}

(h, i) Acres Burned, unit: 10^6 acres year^{-1}

Emission – unit: Tg year^{-1}

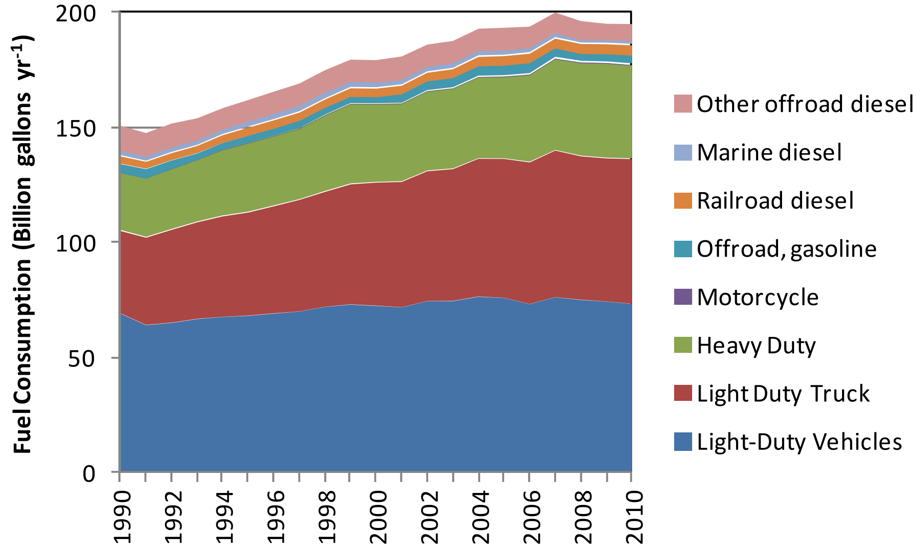


Fig. S1 Mobile source fuel consumption in the United States, 1990–2010

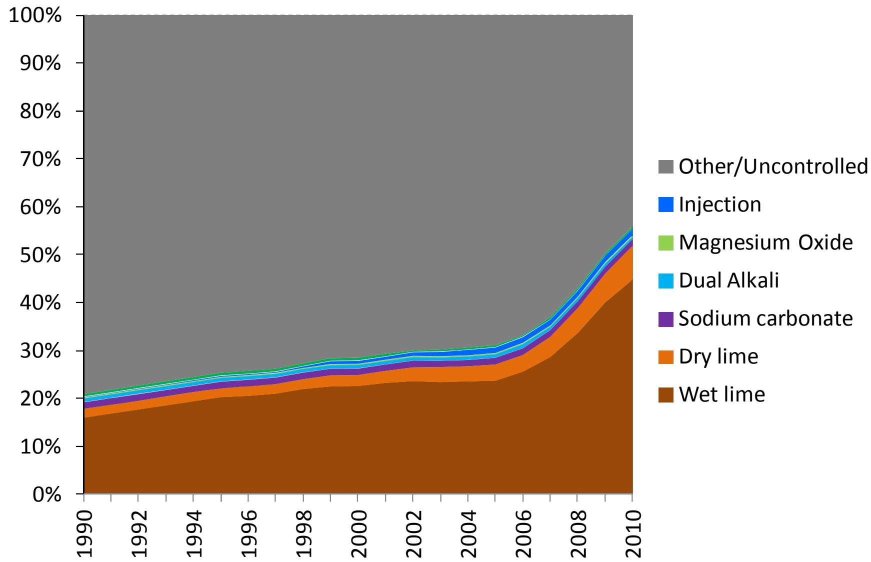


Fig. S2 Application of post-combustion SO_2 control technologies in coal-fired power plants from 1990-2010 (weighted by unit capacity, based on the Clean Air Markets data, http://ampd.epa.gov/ampd/)

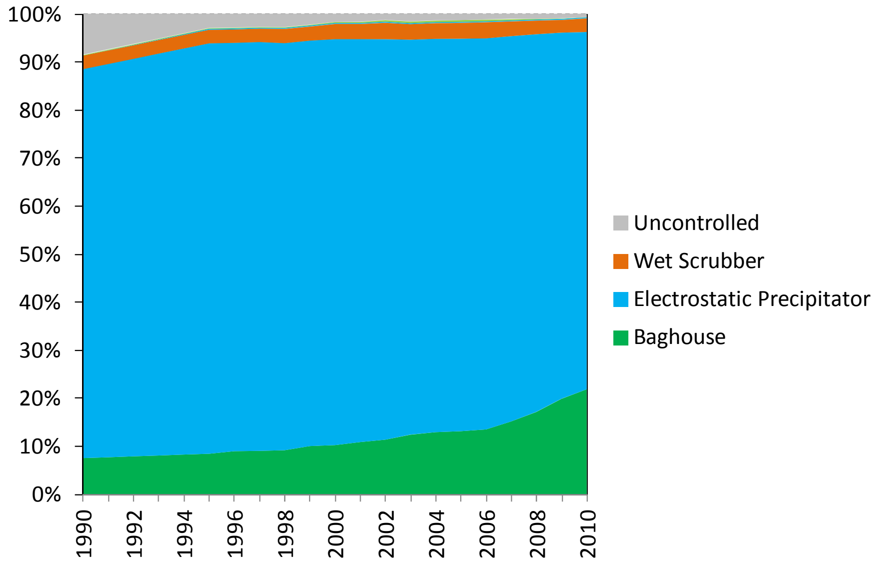


Fig. S3 Application of post-combustion PM control technologies in coal-fired power plants from 1990 to 2010 (weighted by unit capacity, based on the Clean Air Markets data, http://ampd.epa.gov/ampd/)

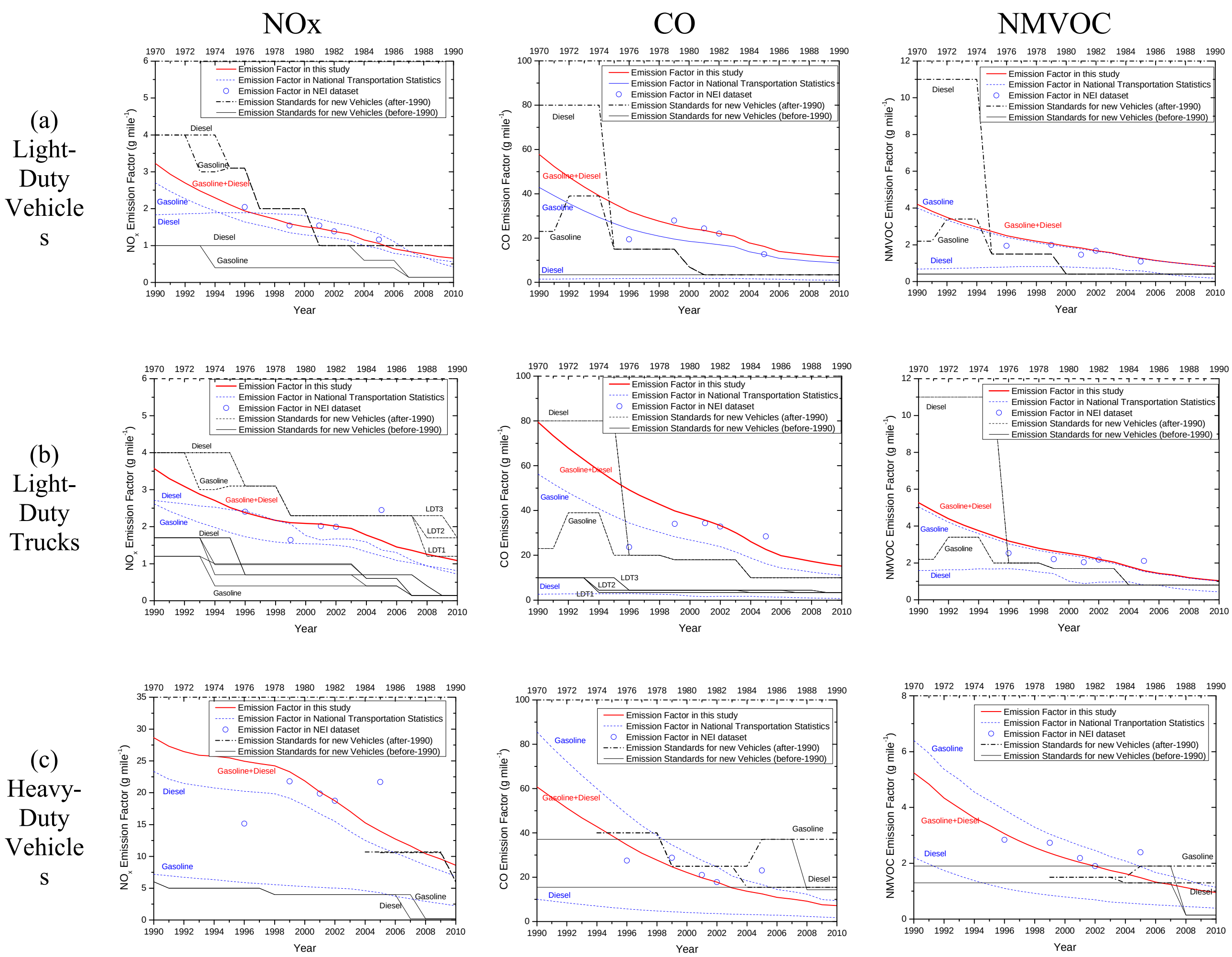


Fig. S4 Comparison of on-road NO_x, CO and NMVOC emission factors with the federal exhaust emissions certification standards for newly manufactured vehicles