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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 and NO_x emissions agree well with the observed changes in ambient SO_2 and NO_2 concentrations, suggesting that the various controls on SO_2 and NO_x 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.

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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 US 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 US 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 NO_x emission estimated from MOVES (www.epa.gov/otaq/models/moves/index.htm) used in 2005 NEI is much

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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 more important than accuracy of absolute values. Though national-level sector-based emissions for each year are available from the NEI trends report (US 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. combustions 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 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. 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 and NO_x emissions from 1990 to 2010 were compared with changes in observed ambient SO_2 and NO_2 concentrations.

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2 Method

2.1 Development of 1990–2010 emission inventories

The approach we used to develop the long-term emission inventory is given in Fig. 1. The US 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 basing 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/) and the three earlier years of 1990, 1995 and 1996 which were developed for previous studies (US EPA, 1993; Adelman and Houyoux, 2001).

In this study, all point, area and mobile sources 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 the Supplement in Table S1. All sectors were processed at the state level for trend purposes. The 2005 county-level NEI data was used as the reference for most of 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 (e.g. 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 years back to 1990s. Besides, the results from McDonald et al. (2012) suggested that overall MOBILE6 results were even closer to both EDGAR and results by McDonald et al. (2012) than MOVES only except for the past few years. For the purpose of this study, we selected most recent NEI data which were using MOBILE6 (i.e. 2002 NEI,

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instead of 2005 NEI) as the reference for on-road sector). As seen in Table 1, all sectors have noticeable contributions to total emissions of one or more pollutants. Thus, 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 the following sections.

2.1.1 Energy-related stationary sources

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.

Emission factor

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. In this study, 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 Eq. (1):

$$EF_{p,s,i,y} = \frac{\sum_{n} Emis_{p,s,i_n,y}}{Act_{s,i,y}} \quad (1)$$

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.

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

1. 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 maximumly controlled-level;
2. 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 pervious year;
3. 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 Eq. (2):

$$\min\left(\mathrm{EF}_{p,s,i,y-1}, \max\_\mathrm{EF}_{p,i_n}\right) \geq \mathrm{EF}_{p,s,i,y} \geq \max\left(\mathrm{EF}_{p,s,i,y+1}, \min\_\mathrm{EF}_{p,i_n}\right) \quad (2)$$

where, $\max\_\mathrm{EF}_{p,i_n}$ and $\min\_\mathrm{EF}_{p,i_n}$ 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 Eq. (3).

$$\begin{cases} \mathrm{EF}_{p,s,i,y} = \mathrm{EF}_{p,s,i,\mathrm{py}} - \frac{\mathrm{Act}_{s,i,\mathrm{py}}}{\mathrm{Act}_{s,i,y}} \times \left(\mathrm{EF}_{p,s,i,\mathrm{py}} - \mathrm{EF}_{p,s,i,\mathrm{ny}}\right) \times \left(\frac{\mathrm{ef}_{p,i,\mathrm{py}} - \mathrm{ef}_{p,i,y}}{\mathrm{ef}_{p,i,\mathrm{py}} - \mathrm{ef}_{p,i,\mathrm{ny}}}\right) \\ \qquad\qquad (\mathrm{py} < y < \mathrm{ny} \leq 2005) \\ \mathrm{EF}_{p,s,i,y} = \mathrm{EF}_{p,s,i,\mathrm{py}} \times \frac{\mathrm{ef}_{p,i,y}}{\mathrm{ef}_{p,i,\mathrm{py}}} \quad (y > \mathrm{py} \geq 2005) \end{cases} \quad (3)$$

where, $\mathrm{ef}_{p,i,y}$ 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.

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2.1.2 Mobile sources

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 Eq. (4), where, $\mathrm{VMT}_{s,i,y}$ is the calculated annual VMT for type i of vehicle in State s in the year of y; $\mathrm{Population}_{s,i_n,y}$ is the vehicle populations for sub-type i_n of type i in State s in the year of y; and $\mathrm{DPV}_{i_n,y}$ is the DPV for sub-type i_n of vehicle type i in year y.

$$\mathrm{VMT}_{s,i,y} = \sum_n \left(\mathrm{Population}_{s,i_n,y} \times \mathrm{DPV}_{i_n,y}\right) \quad (4)$$

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.

$$\mathrm{ENE}_{s,i,y} = \sum_n \left(\mathrm{Population}_{s,i_n,y} \times \mathrm{DPV}_{i_n,y} \times \mathrm{FE}_{i_n,y}\right) \quad (5)$$

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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 average miles traveled per gallon gas/diesel.

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 Fig. 2. 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.

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 did not use 2005 NEI was given in previous Sect. 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, as function Eq. (6), where, $EF_{p,s,i,y}$ is the calculated emission

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factor for pollutant p from vehicle-type i in State s in year y; $\mathrm{ef}_{p,i,y}$ is the emission factor obtained from National Transportation Statistics, and py and ny are the two available years near the year of y.

$$\mathrm{EF}_{p,s,j,y} = \mathrm{EF}_{p,s,j,\mathrm{py}} \times \frac{\mathrm{ef}_{p,i,y}}{\mathrm{ef}_{p,i,\mathrm{py}}} \quad (6)$$

Necessary adjustment was made to ensure the calculated emission factors to be comparable with the vehicle emission standards and references (Dallmann and Harley, 2010).

For nonroad sources, 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 consistent framework to calculate the emission inventories for Europe and Asia. The evolution of emission factors for nonroad diesel and gasoline equipment was informed by NEI trends and Dallmann and Harley (2010).

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-yr 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 Eq. (7).

$$\mathrm{Emis}_{p,s,i,y} = \mathrm{Emis}_{p,s,i,\mathrm{ry}} \times \frac{\mathrm{emis}_{p,i,y}}{\mathrm{emis}_{p,i,\mathrm{ry}}} \quad (7)$$

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where $\text{emis}_{p,i,y}$ and $\text{emis}_{p,i,\text{ry}}$ 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-yr emissions were simply scaled by following the trend of activities, as Eq. (8):

$$\text{Emis}_{p,s,i,y} = \text{Emis}_{p,s,i,\text{ry}} \times \frac{\text{Act}_{p,s,i,y}}{\text{Act}_{p,s,i,\text{ry}}} \tag{8}$$

where, $\text{Act}_{p,s,i,y}$ and $\text{Act}_{p,s,i,\text{ry}}$ 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 Eq. (9):

$$\text{Emis}_{p,s,i,y} = \text{Emis}_{p,s,i,\text{ry}} \times \frac{\text{Act}_{p,s,i,y}}{\text{Act}_{p,s,i,\text{ry}}} \times \frac{\left(\frac{\text{emis}_{p,i,y}}{\text{act}_{i,y}}\right)}{\left(\frac{\text{emis}_{p,i,\text{ry}}}{\text{act}_{i,\text{ry}}}\right)} \tag{9}$$

where $\text{Act}_{i,y}$ and $\text{Act}_{i,\text{ry}}$ 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.

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.

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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 Eq. (6).

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.

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 (US 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.

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The amount of regional crude ore by state was obtained from the US 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 and 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 importation 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.

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-yr county-level emission inventories developed as described in Sect. 2.1 were processed by SMOKE (Sparse

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Matrix Operator Kernel Emissions, http://www.smoke-model.org/data.cfm) to generate the spatially and temporally resolved emissions over a 36 × 36 km CONUS (Continental United States) modeling domain.

1. 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.

2. 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.

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

Many studies indicated 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 and NO_2 concentrations to compare with the emission trends developed as described in Sect. 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 NO_2 concentrations monitored by Air Quality System (AQS) were downloaded from the EPA website (http://www.epa.gov/airtrends/nitrogen.html). Trends of observed SO_2 and NO_2 concentrations respectively monitored at 39 CASTNET sites and 52 AQS sites having 20 yr of completeness were used to compare with the trends of SO_2 and NO_x emissions at the same spatial location. For the purpose of this analysis, emissions from the 9 grids located nearest to

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each monitor were summed; thus, the emissions in a roughly 100 km × 100 km 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 Sect. 2.1, the 20-yr 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 Fig. 3. 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 4 gives the application

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ratios of NO_x control technologies (weighted by unit capacity) in power plants from 1990–2010, which increased dramatically for all types of fuel combustions 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 1 January 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 of energy consumption in power plants in Fig. 5a, 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). 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 Fig. 5a, 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 widely application of advanced control technologies like baghouses and electrostatic precipitators as seen in Fig. 6. Such strengthened control efforts reduced the average of PM emission factors by 27 % for coal-fired power plants.

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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 on 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.

Seen in Fig. 5b–d, SO_2 and NO_x emission trends estimated in this study are better constraint by energy trends than that in 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 NO_x emissions in 2000–2005 industrial distillate fuel combustion and 2000–2002 commercial coal combustion. Besides, the residential NO_x emissions decreased sharply from 1996 to 1999 in NEI data. Information about such reduction is unavailable, so in our estimates the trends of residential NO_x emissions were the same as the trends of 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 combustions were significant reduced, mainly caused by the decline in consumptions of coal and oil which were replaced by cleaner fuels like natural gas.

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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 MMVOC 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 more to NO_x emissions because of their higher NO_x emission factor that is $5 \sim 10$ times higher than that of light-duty vehicles and trucks, as shown in Table 1b.

As shown in Fig. 7, along with the progress in strengthening the federal exhaust emissions certification standards for newly manufactured vehicles starting from 1970s, 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 yr ago (assuming the vehicle lifetime was about 20 yr), 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 US 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 Fig. 8, 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.

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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 US mobile emissions from 1996–2006 by Dallmann and Harley (2010).

Dallmann and Harley (2010) also suggested that NO_x and PM emission factors for off-road diesel-powered engines that decreased significantly from 1996 to 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 %.

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 Fig. 9. 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 Fig. 10. 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

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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 Fig. 11, those reductions were widely distributed over the US, 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-yr emissions calculated in this study were compared with NEI trends and EDGAR data, as seen in Fig. 12. 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.

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The trends of SO_2 and NO_x emissions were compared with the observed surface SO_2 and NO_2 concentrations to evaluate the 20 yr of emission inventories. The spatial distribution of trends generally agree well with the observations, as seen in Fig. 13. The results indicate that those reductions were widely distributed across the whole continental US domain. Larger SO_2 reduction ratio were found in north-east and southwest where more strengthened control efforts have been taken. Seen from the comparison of historic trends between emission and observations, the changes of emission reflect the declines in both SO_2 and NO_2 observations. The average reduction of SO_2 and NO_x emissions in the grid cells near monitors are 60 % and 43 % respectively, which agrees well with the observed decrease of SO_2 and NO_2 concentrations, 69 % and 39 %.

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.

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Mobile emissions were only based on MOBILE6 results rather than the new-developed MOVES module. Further improvement on those details is still necessary.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/12/30327/2012/acpd-12-30327-2012-supplement.pdf.

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

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Table 1b. Defined sectors and percentage of their contributions to total anthropogenic emissions (calculated from the 2002 NEI data, Unit: %). Energy-related mobile sources.

Sector		Pollutant						
Group	Type	NO_x	SO_2	CO	NMVOC	PM_{10}	$PM_{2.5}$	NH_3
Highway	Light Duty Vehicle, gasoline and diesel	11.2	0.6	34.2	16.1	0.4	0.5	4.0
	Light Duty Truck, gasoline and diesel	9.4	0.5	29.7	12.2	0.3	0.4	2.6
	Heavy Duty, gasoline and 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**

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Table 1c. Defined sectors and percentage of their contributions to total anthropogenic emissions (calculated from the 2002 NEI data, Unit: %). Non energy-related sources.

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 and 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 and Transport	0.0	0.0	0.0	8.4	0.0	0.0	0.0
	Waste disposal and 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 and 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**

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Table 2. Summary of data sources used for scaling emissions.

Sector			Emission constrains	
			Activity indicator	Control info.
Energy-related Stationary	Power plants Industrial Commercial Residential	Coal, Residual oil, Distillate oil, Natural gas, LPG, Kerosene, Wood	State-based energy combustion by fuel types[1]	Unit level database[2] NEI[3]
Energy-related Mobile	On-road	Light Duty Vehicle Light Duty Truck Heavy Duty Motorcycle	State-based vehicle miles traveled by vehicle types[4]	Evolution of emission factors by vehicle types[5]
	Off-road	Agriculture, gasoline Construction, gasoline Other gasoline	State-based gasoline consumption by equipment types[4]	NEI
		Marine, diesel Rail, diesel Other diesel	National diesel consumption by equipment types[4] Total Distillate Fuel Oil and Kerosene Sales by End Use[6]	
		Marine, Residual oil All natural gas All LPG All Jet fuel	State-based energy consumption by fuel types[1]	

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Table 2. Continued.

Sector			Emission constrains	
			Activity indicator	Control info.
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 and Transport	NEI	
		Waste disposal and recycling	Keep the same over the period	
		Mining and 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. US 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.

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Table 3. Summary of NO_x and SO_2 emission factors in energy-related sectors (Unit: 0.01 lb MMBtu^{-1}).

Sector	Fuel	NO_x Emission factor This study 1990	2010	Δ[2]	Unabated[1]	SO_2 Emission factor This study 1990	2010	Δ	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

[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. $(EF_{2010}–EF_{1990})/EF_{1990} \times 100\,\%$.

[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.

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Table 4. Sector-specific emissions in the United States for 1990 and 2010 estimated in this study (unit: $Gg\,yr^{-1}$).

Sector	SO_2			NO_x			CO			NMVOC		
	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ
Power plants	16 117	5018	−69 %	6431	2237	−65 %	294	462	57 %	39	46	20 %
Other Comb.	3764	1215	−68 %	2625	1772	−32 %	4686	2598	−45 %	841	428	−49 %
On-road	499	35	−93 %	10 972	4643	−58 %	136 586	38 627	−72 %	9798	2828	−71 %
Off-road	619	452	−27 %	3428	2768	−19 %	12 257	11 870	−3 %	2388	1950	−18 %
Ind. Process	2147	873	−59 %	1412	1227	−13 %	5498	2133	−61 %	2291	1686	−26 %
Other	100	104	5 %	390	400	2 %	12 922	12 992	1 %	8450	5277	−38 %
All	23 246	7697	−67 %	25 258	13 048	−48 %	172 243	68 682	−60 %	23 807	12 215	−49 %

Sector	PM_{10}			$PM_{2.5}$			NH_3		
	1990	2010	Δ	1990	2010	Δ	1990	2010	Δ
Power plants	654	559	−15 %	529	462	−13 %	11	19	70 %
Other Comb.	1048	511	−51 %	767	389	−49 %	26	28	6 %
On-road	392	118	−70 %	333	92	−72 %	145	276	91 %
Off-road	245	188	−23 %	222	172	−22 %	2	2	−21 %
Ind. Process	637	318	−50 %	469	287	−39 %	126	126	0 %
Other	17 130	8394	−51 %	3814	2675	−30 %	3181	3414	7 %
All	20 105	10 088	−50 %	6134	4077	−34 %	3491	3864	11 %

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

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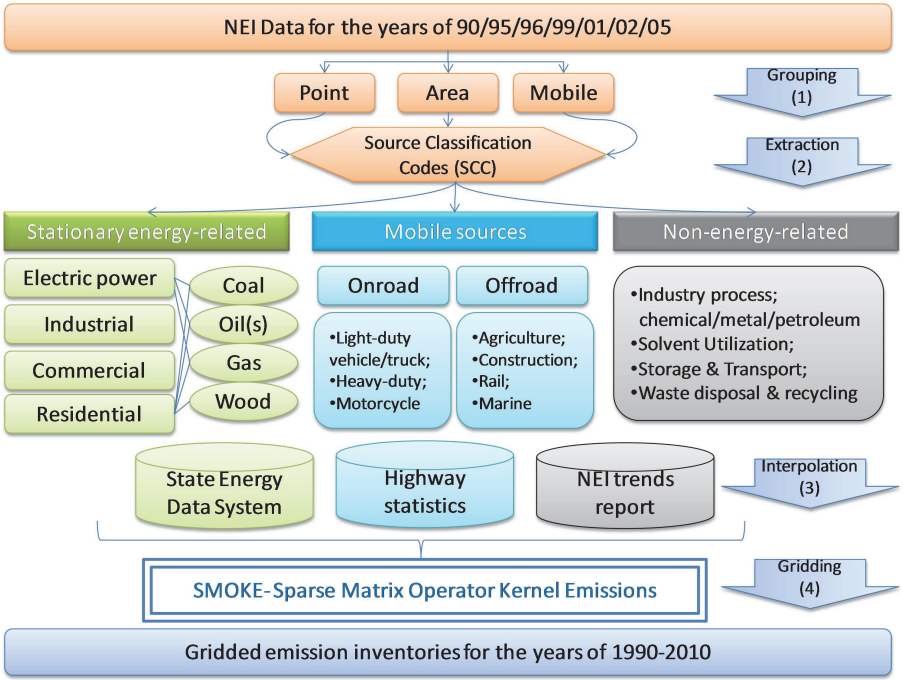

Fig. 1. The framework of the 20 yr emission inventory development.

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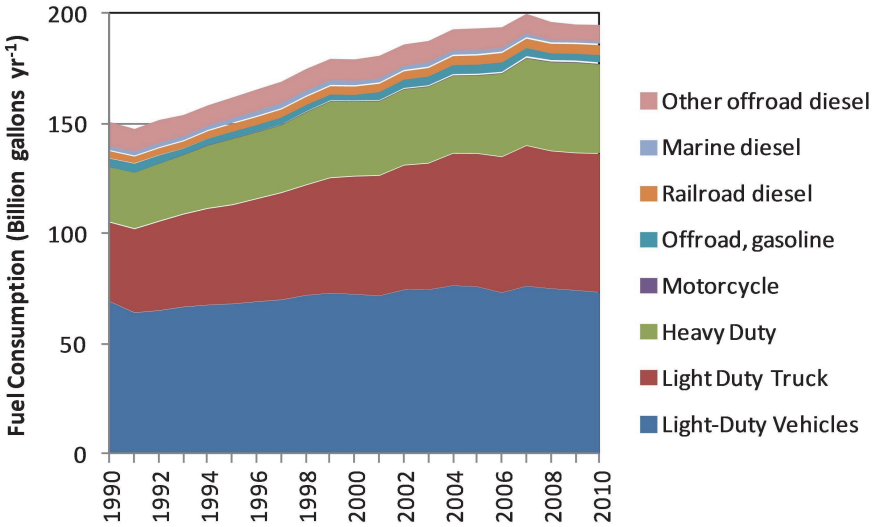

Fig. 2. Mobile source fuel consumption in the United States, 1990–2010.

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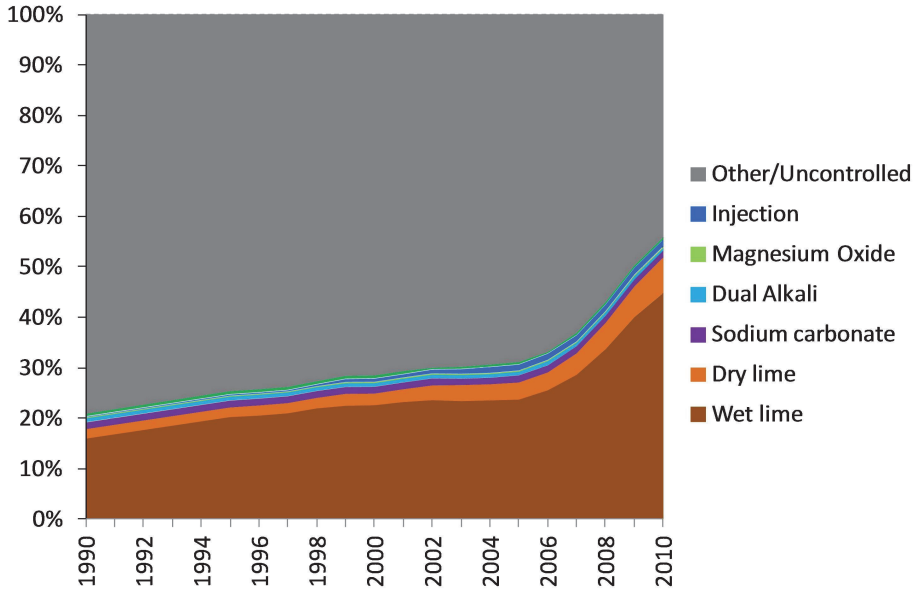

Fig. 3. Application of post-combustion SO_2 control technologies in coal-fired power plants from 1990–2010 (weighted by unit capacity, based on the clean air markets data, http://camddataandmaps.epa.gov/gdm/).

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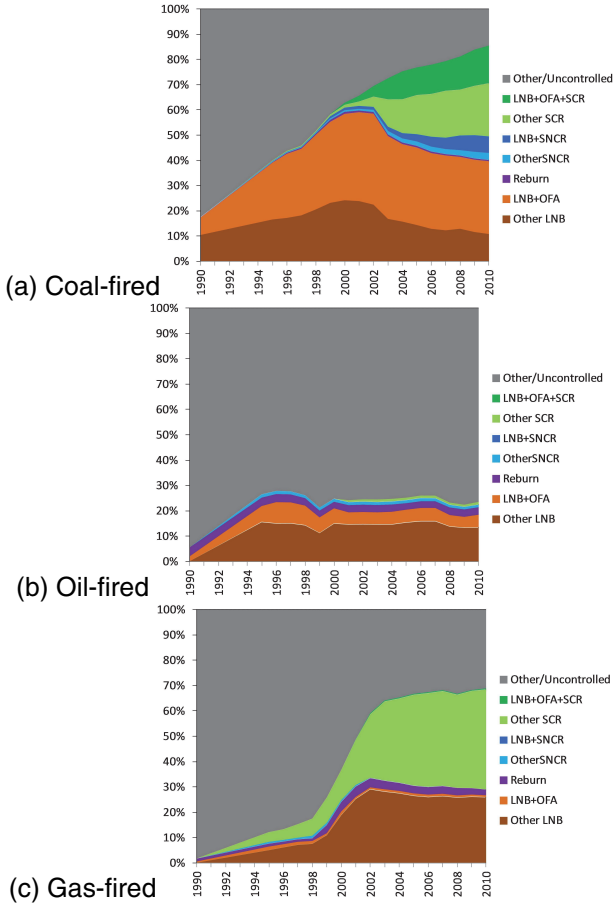

Fig. 4. Application of NO_x control technologies in power plants from 1990–2010 by primary fuel type (weighted by unit capacity, LNB-low NO_x burners; OFA-overfire air; SNCR- selective noncatalytic reduction; SCR-selective catalytic reduction; based on the clean air markets data, http://camddataandmaps.epa.gov/gdm).

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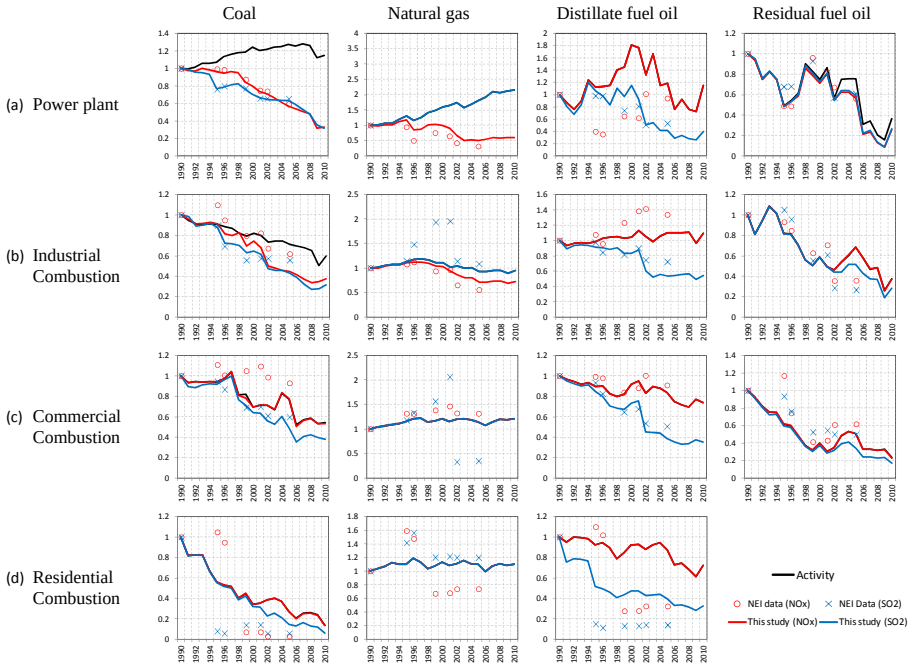

Fig. 5. Activity (fuel use) and NO_x and SO_2 emission trends during 1990–2010 for energy-related stationary sources (Year 1990 = 1).

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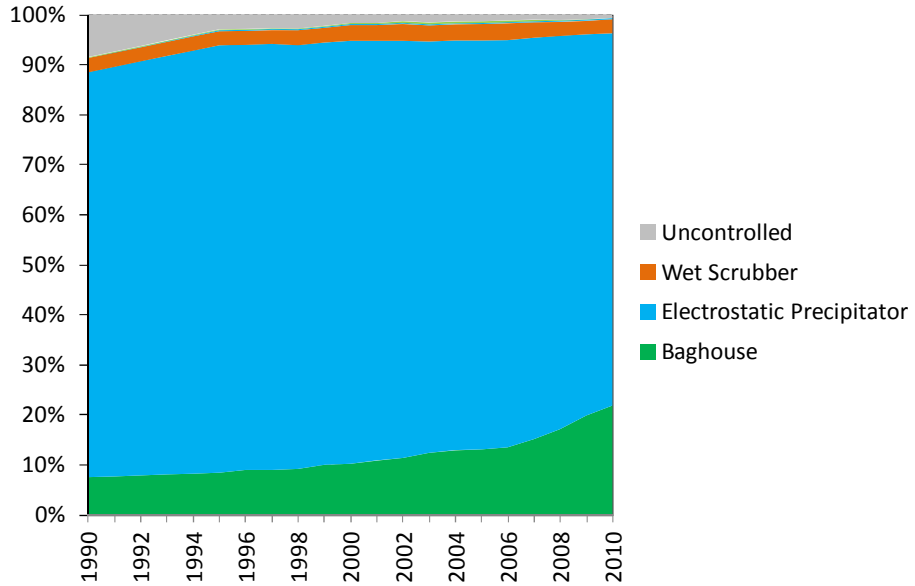

Fig. 6. Application of post-combustion PM control technologies in coal-fired power plants from 1990 to 2010 (weighted by unit capacity, based on the clean air markets data, http://camddataandmaps.epa.gov/gdm).

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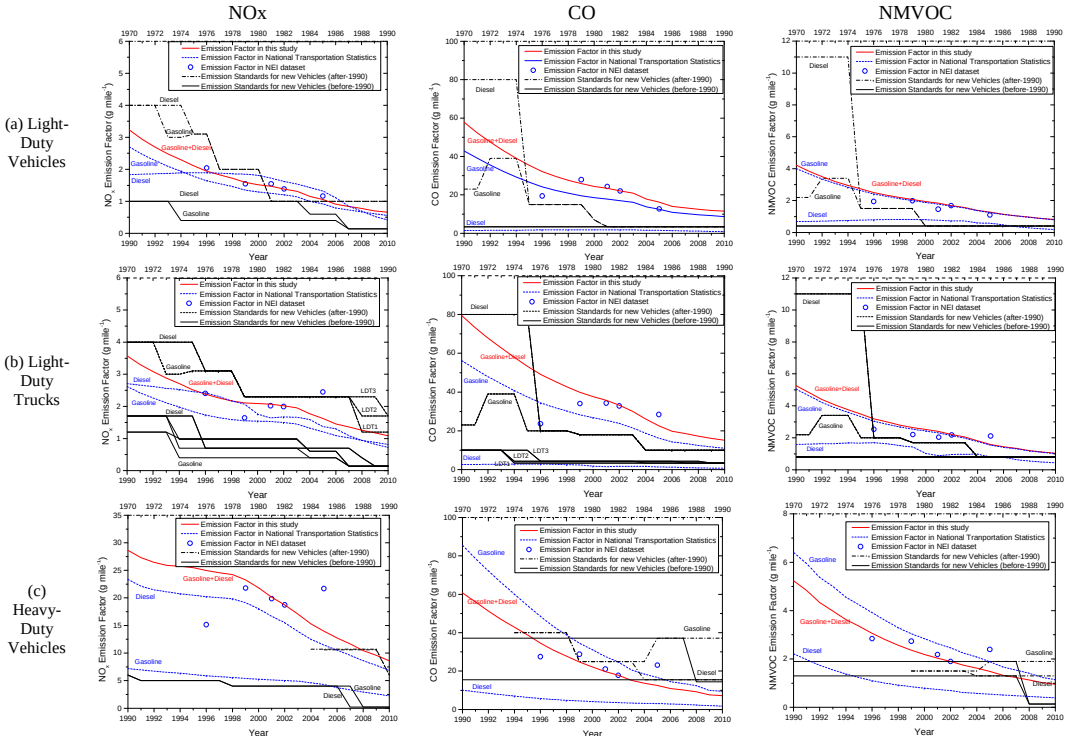

Fig. 7. Comparison of on-road NO_x, CO and NMVOC emission factors with the federal exhaust emissions certification standards for newly manufactured vehicles.

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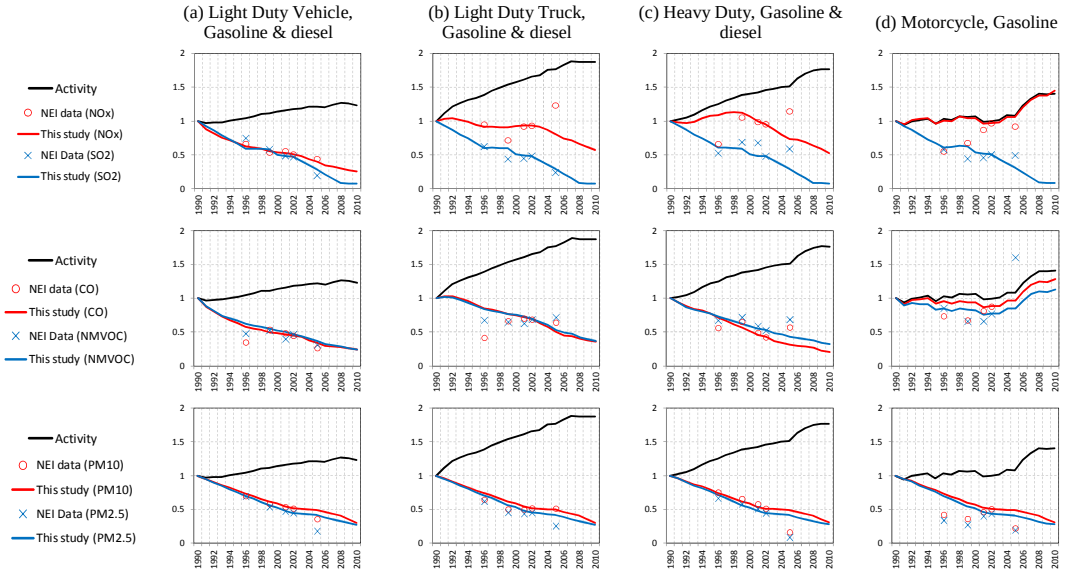

Fig. 8. Activity and emission trends during 1990–2010 for on-road mobile sources (Year 1990 = 1).

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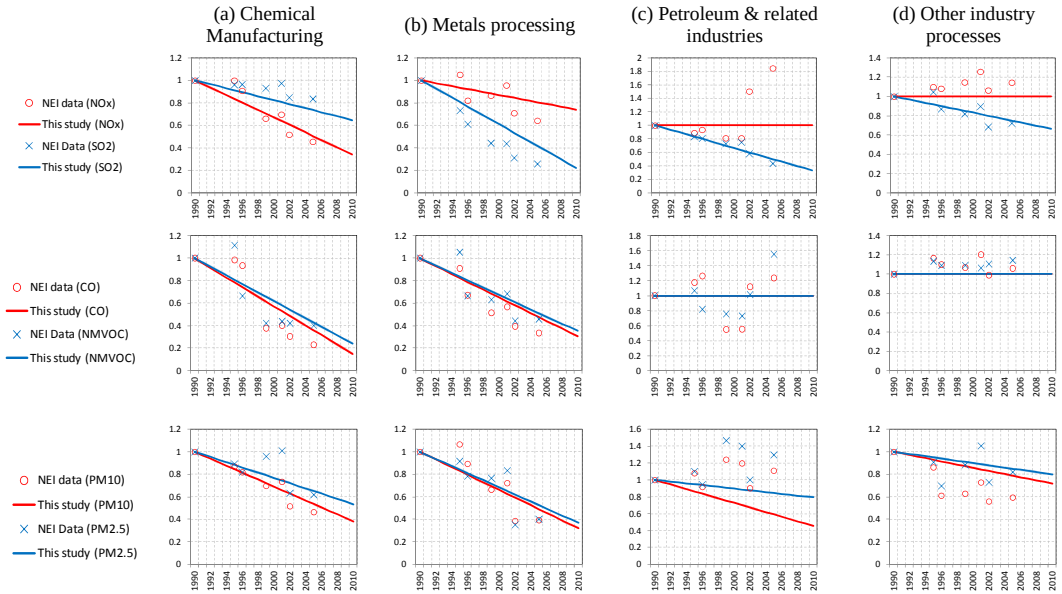

Fig. 9. Emission trends during 1990–2010 for industrial process sources (Year 1990 = 1).

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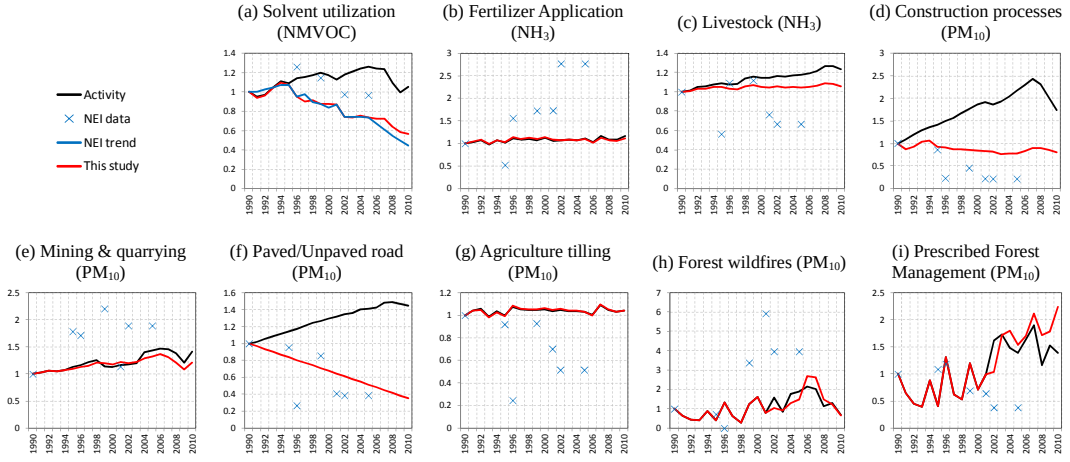

Fig. 10. Activity and emission trends during 1990–2010 for other non-energy related sources (Year 1990 = 1).

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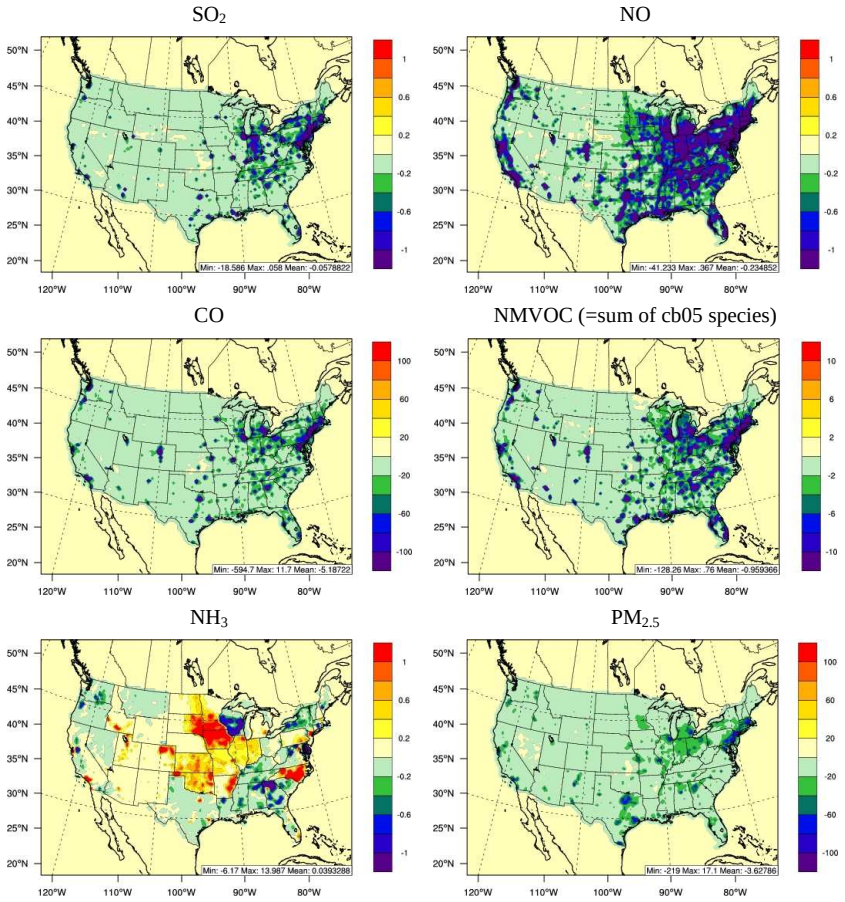

Fig. 11. Changes in emission rates over the 36 × 36 km CONUS domain from 1990 to 2010 (unit: moles s^{-1} $grid^{-1}$ for gaseous species, g s^{-1} $grid^{-1}$ for $PM_{2.5}$).

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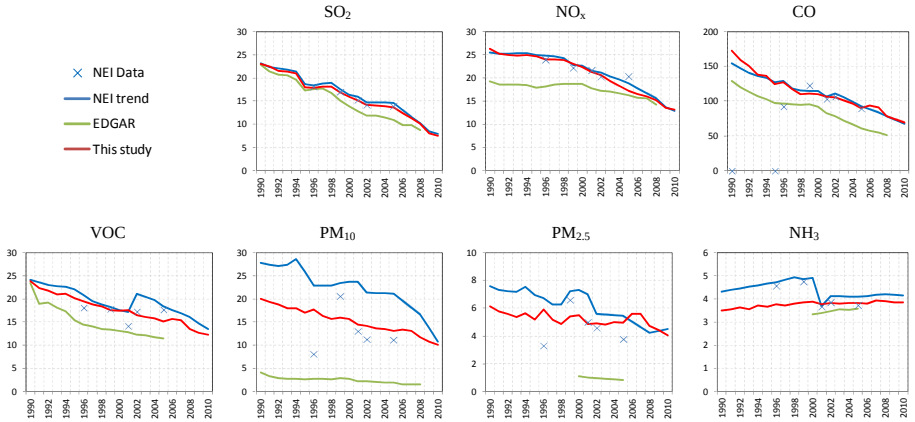

Fig. 12. Emission trends during 1990–2010 (unit: $Tg\,yr^{-1}$).

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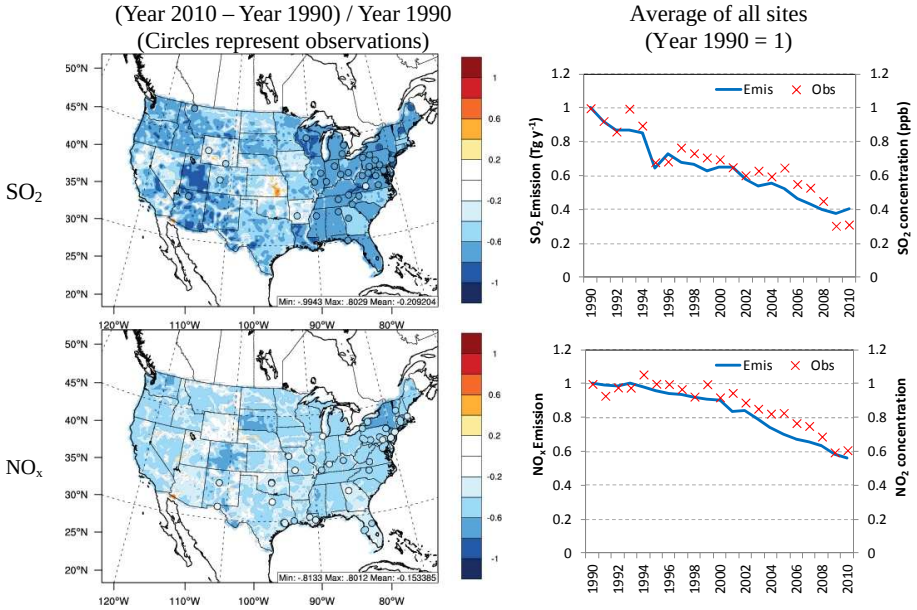

Fig. 13. Comparison of historic trends between SO_2 and NO_x emissions and observed SO_2 and NO_2 concentration from 1990 to 2010.

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