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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Historical gaseous and primary aerosol emissions in the United States from 1990–2010" *by* J. Xing et al.

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We would like to thank the referee for a very thoughtful and detailed review of our manuscript that helped to improve the paper. Below we provide a point-by-point response to the reviewer's comments and how we have addressed them in the revised manuscript. General Comments:

[Comment]: This is an important study that developed long-term emission inventories in a consistent way and still held detailed process level information based on EPA National Emission Inventories (NEIs). The emission inventories developed in this study are on high demands and will be well used for regional model simulations of air quality and climate. It is convenient to see the trends of major gaseous and aerosol emissions and the details on controls of the emissions in one paper although the trends in this

study are similar to those in the NEI trend report and may not be something new. My concern on the emission inventories in this study is uncertainties that many users may have to deal with and spend lots of time to understand. Especially, the philosophy of the authors shown in line 2-4 in page 30330 makes me nervous. Major difference of the inventory in this manuscript from the NEI trend report would be spatially-resolved (gridded) emissions for regional model simulations, which in turn helps the evaluation of the emission inventory at much finer scale than the U.S. total.

[Response]: We thank the reviewer for recognizing the importance of developing a consistent series of spatially resolved emissions from 1990 to 2010 that is not subject to trend artifacts due to method changes that often affect databases such as the EPA NEI and EPA trends report. We believe that our approach of using a consistent set of activity data, emission factors, and emission control information represents a significant improvement over the information currently available, and we hope that we have addressed the concerns raised by the reviewer in our revised manuscript as further discussed below.

[Comment]: I suggest the authors to revise the section of comparison and validation in the manuscript by focusing on the emission inventories at finer scales for more species if possible (CO and nonreactive or relatively nonreactive hydrocarbons). The plots of contours and circles in Figure 13 are not clear and more monitoring sites can be included. Dominant sector to the total emission would be different depending on the location of the sites. Thus, looking at the finer spatial resolution and year-to-year changes may help to evaluate the emissions for different sectors and species.

[Response]: We appreciated the reviewer's suggestion. In the revised manuscript, we provided analysis of trends for additional species and observations at more monitoring sites. Besides the CASTNET data for SO2 used in the original manuscript, Air Quality System (AQS) data for SO2, NO2 and CO concentrations 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 SO2, NO2, CO and EC concentrations monitored at 39 CASTNET&96 AQS, 69 AQS, 89 AQS and 30 IMPROVE sites, having 20 yr of completeness were used to compare with the trends of SO2, NOx, CO and EC emissions at the same spatial location. The 20 yrs emissions were gridded to 12km$\times$12km resolution grid over CONUS domain through SMOKE processing. For the purpose of this analysis, emissions from the 81 grids located nearest to each monitor were summed; thus, the emissions in a roughly 100km$\times$100km area around each monitor are assumed to impact the concentrations measured at the monitor. As the reviewer suggested, the comparison was conducted at a finer scale (six regions over CONUS). The regions were defined as in Hand et al. (2012), i.e., West, Great Plains, Southwest, Northeast, Midsouth and Southeast, as shown in Fig. C1. Emission trends of SO2, NO, CO and EC (speciation of PM2.5 by SMOKE) are given by sector (the same as the one defined in SMOKE), i.e., (1) ptipm- NEI point source EGUs mapped to the Integrated Planning Model (IPM) model using the National Electric Energy Database System (NEEDS) database; (2) ptnonipm- All NEI point source records not matched to the ptipm sector; (3) mobile; (4) non-road and (5) other area sources. We revised the third paragraph of Section 3.2 as follows: "The trends of SO2, NOx, CO and EC emissions were compared with the observed trends in ambient surface SO2, NO2, CO and EC concentrations to evaluate the 20 years of emission inventories. The spatial distributions of trends generally agree well with the observations, as seen in Fig. C2. 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 SO2, 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 SO2, NO2, CO and EC concentrations, as 63%, 33%, 71% and 50%, respectively. 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 SO2. The trend

of observed SO2 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 those defined in Hand et al. (2012), i.e., West, Great Plains, Southwest, Northeast, Midsouth and Southeast, as shown in Fig. C1. For SO2 (see Fig. C3a), 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, SO2 emission was dominated by non-EGU point sources. Though a decreasing trend was shown in both emissions and observed concentrations, the SO2 concentration before 1996 is extremely high but the SO2 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 NOx emissions in all regions (see Fig. C3b). The national emission trend agrees better with the trends of observed NO2 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. NOx 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 NOx 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, but is beyond the scope of the current study. Mobile is also the dominant CO emissions sources for all regions

(see Fig. C3c). 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."

[Comment]: It would be helpful if newly calculated and compiled activity data, emission factors, and emissions for each species, sector, state, and year are provided as supplementary material. Most of results presented in the manuscript are normalized emission trends. The absolute values should be presented in supplementary material or the values used for normalization should be provided in the figures or figure captions. Overall the results section is too short compared to the presented figures. More explanations, discussions and references can be added.

[Response]: We intend to make all data publicly available. Unfortunately the volume is far too large to attach as supplementary material to this manuscript. We plan to share these data thought the clearinghouse of the CMAS center. Also, the validation of the new emission inventory is very limited in this paper since only primary pollutants were considered. We are currently conducting 20 yrs air quality model simulations driven by these emissions. Additional species will be involved in comparison with observation data, such as ozone and fine particles. As the reviewer suggested, we now provide the absolute values used in Figure 5, 6, 9 and 10 in the supplementary material, and additional discussion has been added to the revised manuscript (see our detailed answers to the specific comments related to Figure 5)

Specific Comments: [Comment]: Line 27-30, 30329: MOBILE 6 could be used for 2005 NEI. When the 2005 NEI was first released, it was at transition from MOBILE 6 to MOVES.

[Response]: We agree that it would be better to use the old MOBILE 6 version for 2005 NEI; however it is not available to the public any more. Future development could use the newly released MOVES model for the entire 20-year period, but is beyond the scope of the current study. To add clarity we have modified the sentence in the revised manuscript to "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"

[Comment]: Line 2-4, 30330: This should be deleted. It is not possible to make accurate model simulations of gas and aerosols without knowledge on both absolute value and trend.

[Response]: The statement has been revised as follows: "when performing long-term analysis of decadal-scale variations and trends, accuracy of trends is as important as accuracy of absolute values."

[Comment]: Line 24, 30330: What is the finest resolution in the emission inventories in this study? 36 km? 4km?

[Response]: The inventories in this study are generated at the county level and are available in SMOKE format. To generate emission fields for air quality modeling, SMOKE allocates these county-level emissions to a regular grid based on spatial surrogates such as population, roadways, railroads, etc.; these attributes typically are available at very high spatial resolution. For our study, we processed these county-level emissions to a 12 km modeling grid, but processing could certainly be performed for finer grids such as 4 km as well. We modified that sentence to "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) in the United States from 1990 to 2010"

[Comment]: Line 3, 30331: It is not clear what Figure 1 explains. For example, why

is the grouping to point, area, and mobile source necessary? What is interpolated in "interpolation" step?

[Response]: We revised and added some discussion about Figure 1 as follows: "The approach we used to develop the long-term emission inventory is given in Fig. 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 the Supplement 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 NOx 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.

[Comment]: Line 14-17, 30332: It takes long time to understand the reason why the authors recalculated emission factors, before looking at equations etc. It may be helpful to explain why this step is necessary.

[Response]: The purpose for the emission factor (EF) recalculation is to quantify the evolution of emission controls, since we need both the activity and EFs to calculate the emissions. To ensure the accuracy of those recalculated EF, we set up following restrictions: a) EF present decreasing trends for controlled sector; b) EF kept the same for uncontrolled sector; c) all EFs are within the range from the AP-42. Thus trends in emission factors for each sector are also needed for the 20 year period in order to calculate the historical emissions. This discussion is now detailed in the revised manuscript.

[Comment]: Line 22, 30332: What is the definition of "sub-section" here?

[Response]: Because we grouped all NEI sectors into several sectors based on SCC, the "sub-sector" 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). To provide clarity, we have now provided this additional explanation in the revised manuscript.

[Comment]: Eq. (5) or definition of "FE" (line 2, 30335) needs to be checked to get a right unit.

[Response]: This represents the gallons of gas/diesel per average miles traveled. We changed the definition of "FE" to "gallons of gas/diesel per average miles traveled" in the revised manuscript.

[Comment]: Line 1, 30336: "ny" was not used in Eq. (6). Either Eq. (6) or "ny" needs to be corrected.

[Response]: We thank the reviewer for pointing this out; this has been corrected.

[Comment]: Figure 4: Web-link to http://camddataandmaps.epa.gov/gdm in the caption does not work.

[Response]: The updated weblink is http://ampd.epa.gov/ampd/

[Comment]: Figure 5: Was the same normalization factor used for NEI data and the emissions in this study? What caused large differences between NEI data and the emissions in this study? Can differences in the activities mainly explain the differences in the emissions because the emission factors were calculated from the NEI data?

[Response]: We chose the 2005NEI (not 1990NEI) as the baseline, therefore most of the normalization factors used in this study aren't the same as the one used for NEI data. That's because our emission factors calculated from NEI before 2005 were modified to satisfy the rules (e.g., any given year it should be no larger than the one for the previous year). That is also the reason that caused large differences between NEI data and the emissions in this study, i.e., the emission factors used in this study originally calculated from NEI data, but they were modified to meet the rules to be more consistent over time. It is important to note that this approach circumvents the issue of methodological inconsistencies between the specific NEIs.

[Comment]: Can more discussions be added to sections 3.1.1 and 3.1.2?

[Response]: Following the reviewer's suggestion Section 3.1.1has been expanded to include the following additional discussion: "Compared to the trends of energy consumption from power plants in Fig. 5a, trends of SO2 and NOx 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 NOx 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." Section 3.1.2 was also modified to include the following: "As seen in Fig. 5b–d, SO2 and NOx emission trends estimated in this study are better constrained by energy trends than that in the NEI data. For example, the SO2 emission from industrial natural gas combustion increased by 100% from 1990 to 2000 in NEI data 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 NOx emissions to be the same as the trends in energy."

[Comment]: Figures 5, 8, 9, 10, and 13 provide the normalized trends. The absolute values need to be provided as Tables in supplementary material.

[Response]: We provided the absolute values used in Figure 5, 8, 9, 10 as supplementary material. We also revised Figure 13 to now provide the absolute values rather than the normalized trends.

[Comment]: The trends in activity in these plots are not always discernible (black line can not be seen). Comments on this need to be added in the text or the figure captions.

[Response]: We have revised Figure 5 so that the trends in activity are now clearly visible, as shown in Fig. C4.

[Comment]: Section 3.1.3. On-road mobile sources: More explanations on Figure 7

would be necessary. Also discussions based on comparison of this study with recent publication on on-road NOx emission in the U.S., McDonald et al. (2012) would be helpful.

[Response]: Since the Figure 7 is too complicated, we moved the emission standards to supporting information, and only focused on the annual fleet-average emission factors. 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 $\sim$36%, which is relatively smaller than the one suggested by National Transportation statistics, as 70%.

[Comment]: Table 3: Was the same unit (0.01 lb MMBtu-1) used for Off-road Transport?

[Response]: Yes.

[Comment]: Section 3.1.4. Off-road mobile sources: Are the values in GAINS well evaluated? Why is GAINS used here?

[Response]: The baseline emission factors in 2005 for each non-road sector by state were back-calculated from 2005 NEI data. But we need to ensure that those back-calculated emission factors are within the normal range. Unfortunately AP-42 doesn't provide the emission factor that we can directly use for comparison, since it's all embedded in the nonroad model. Therefore, we have to use alternate estimates (i.e., GAINS model which provides a full range of emission factors from unabated to maximally controlled for each non-road sector). In fact all values of emission factors are within the range inferred from GAINS, suggesting that the activity we selected and emission factor we calculated in this study are suitable to use.

[Comment]: Figure 13: Figure caption should include the explanations about circles in the map. Do the circles simply represent the locations of the site or both the locations and the trend values? I think there are more available sites for the evaluation, for

example, the sites over California. It would provide new insights if the line plots on the right in Figure 13 are provided for each state or each region because year-to-year changes are influenced by controls on the dominant sectors in each region (e.g., power sector and industrial sector).

[Response]: We have revised Figure 13 so that the trends in both observation and emission are now clearly visible.

[Comment]: Ammonia emissions: does new emission inventory in this study suggesting large reduction from 2005 NEI and other NEI data (Figure 10) agree with the results by Heald et al. (2012)? Heald et al., 2012, Atmospheric ammonia and particulate inorganic nitrogen over the United States, Atmospheric Chemistry and Physics

[Response]: Ammonia emissions in this study followed the same level as 2005 NEI. Since there're no clue about the "intensive reduction" from 1999 NEI to 2001 NEI, we scaled from 2005 back to 1990 only following the activity trends. 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. We added the explanation in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 30327, 2012.

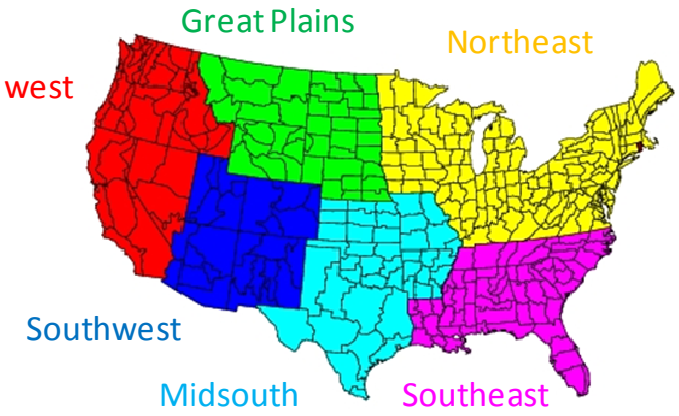


Fig. 1. Fig. C1 Definition of sub-regions used in analysis

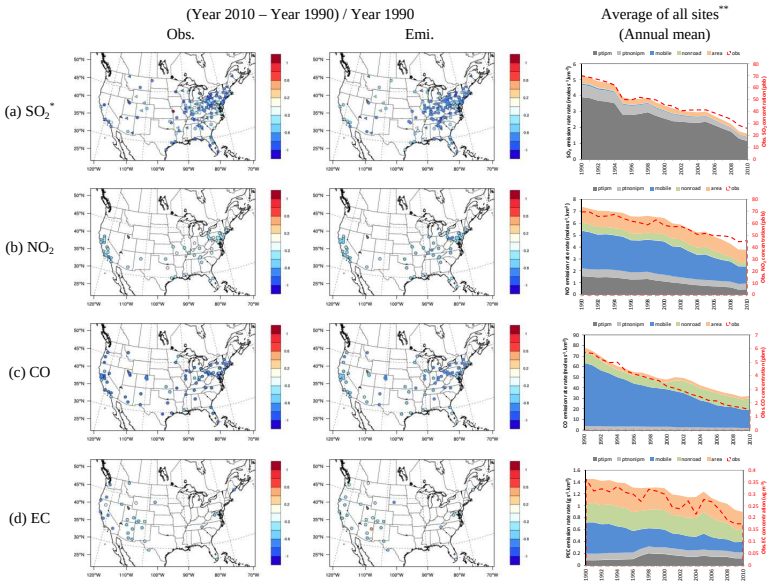
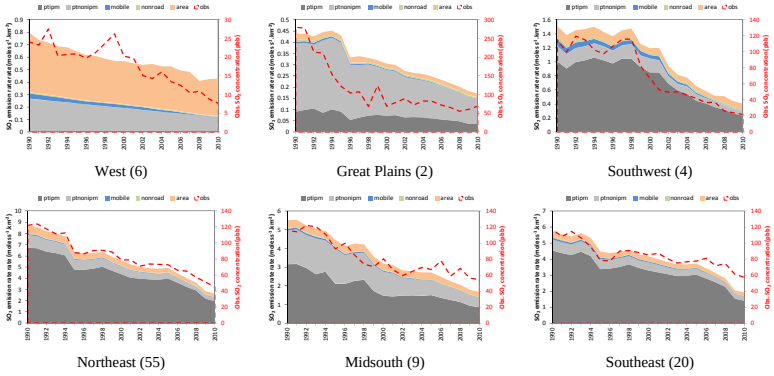
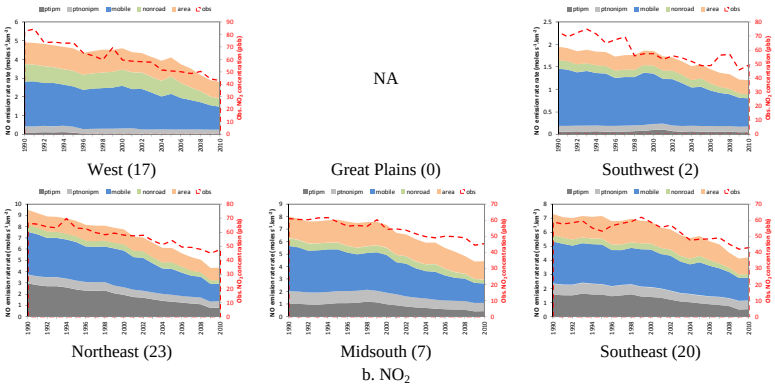


Fig. 2. Fig. C2. Comparison of historic trends between emissions and observed concentration from 1990 to 2010 (*for SO2, Circles represent AQS sites, Triangle represent CASTNET sites; ** SMOKE sectors)



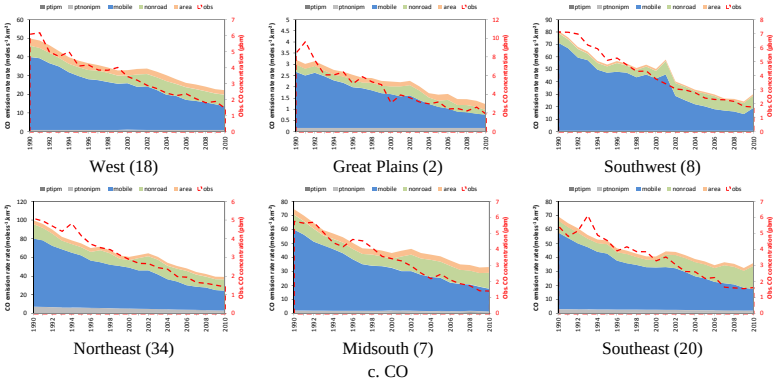
a. SO_2

Fig. 3. Fig. C3. Comparison of historic trends between emissions and observed concentration from 1990 to 2010 by sector and region (number of sites are shown in brackets)



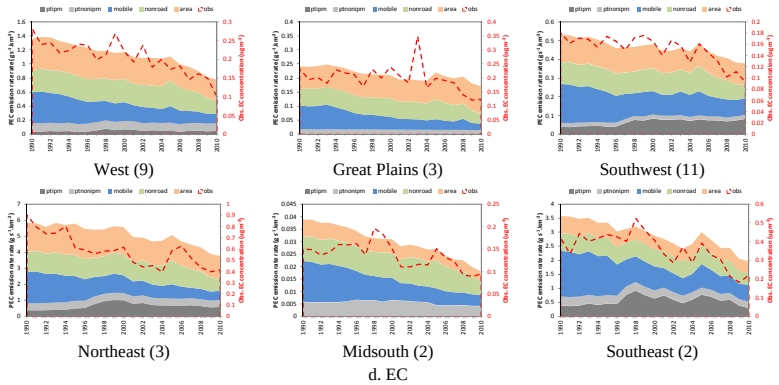
b. NO_2

Fig. 4. Fig. C3. (continued)



c. CO

Fig. 5. Fig. C3. (continued)



d. EC

Fig. 6. Fig. C3. (continued)

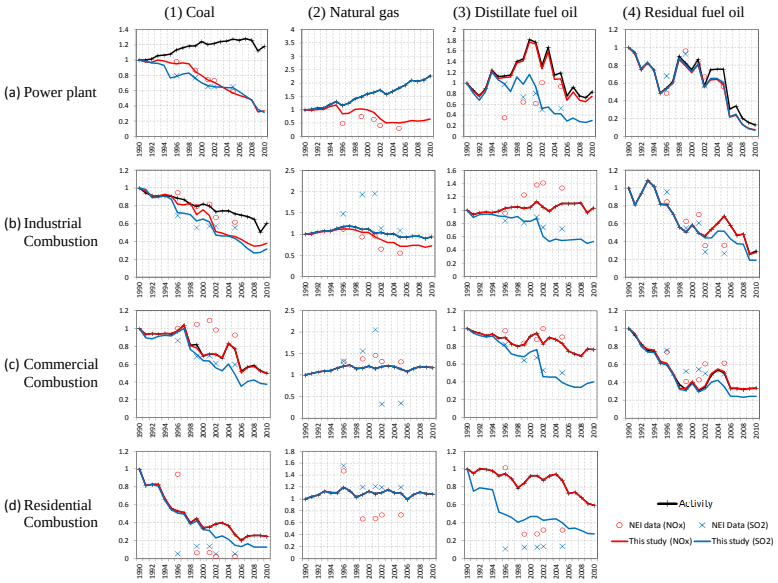


Fig. 7. Fig. C4. Activity (fuel use) and NOx and SO2 emission trends during 1990-2010 for energy-related stationary sources (Year 1990=1)