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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.

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. Incorporation of the reviewer's suggestions has led to a much improved manuscript. Below we provide a point-by-point response to the reviewer's comments and how we have addressed them in the revised manuscript.

Overview: [Comment]: In this manuscript the authors estimate trends in anthropogenic emissions of a number of pollutants for the years 1990-2010. The authors combine activity estimates with emission factors derived from NEI data to estimate annual state-level emissions for 49 source categories. State-level emissions are further resolved spatially and temporally using the SMOKE model. While the objectives of this research are interesting and potentially important, I have a number of concerns with the manuscript.

[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 represesents 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.

[Comment]: I found it difficult to understand the methods used for inventory development, particularly those used for estimating emission factors for on-road vehicles

[Response]: We revised the discussion about our methods as below:

"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

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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." In this study, the trends of emission factor provided in the National Transportation Statistics are only limited to the national level. So we averaged the trends of emission factors of gasoline and diesel for the same vehicle size, basing on the diesel fraction (informed by MOBILE6 default data). We added some explanation in the revised manuscript as below: "The emission factor used for on-road vehicles was scaled 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)"

[Comment]: Also, the lack of any discussion of uncertainty in the manuscript is troubling.

[Response]: We agree with the reviewer that characterization of uncertainty in emissions is important. To address this shortcoming, in the revised manuscript we have provided additional finer-scale comparisons of the estimated emission trends against observations for several species and for several regions. These comparisons provide a measure of the uncertainties associated with the pollutant trends in this new inventory. However, an in-depth analysis of the uncertainties in the underlying data sources (such as activity data and emission factors) used to compute these trends is beyond the scope of this study. The following information has been added to the revised manuscript: "The trends of SO_2, NO_x, 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. C1. 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, of 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 subregions 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. C2. 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]: Finally, the results of the emission trends analysis presented in the manuscript do not contribute significant new scientific understanding beyond what is currently available in the EPA's NEI Air Pollutant Emissions Trends Data.

[Response]: We strongly disagree with this assessment. Our approach in developing a historical record of gas and aerosol emissions is a significant improvement beyond what is available in the NEI Air Pollutant Emission Trends Data because the Trends Data includes confounding changes in methods as well as changes in emission trends. EPA's NEI air emission trends do not use consistent methods across the years but rather interpolate between the NEI years and do not account for method changes in

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the emission estimates. Figure 12 for the pollutant NH3 demonstrates the improvement in our estimation of NH3 over the 20 year period because we eliminate the artificial step change in NH3 emissions seen the NEI data and NEI trends to a smooth year to year annual changes (red line). Improvements for the other pollutants in Figure 12 as less apparent because the downward trends in the emissions is a stronger signal than changes due to step changes. However, our approach removes all step changes in the emission trends because consistent methods are used for each major sector over the 20 year period. We have added the following explanation to our revised manuscript: "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."

Specific Comments: [Comment]: Page 30329, line 27: The authors indicate the MOVES model was used to develop the 2005 NEI. It is my understanding that the MOBILE6 model was used in the development of the 2005 NEI, and that the MOVES model was not used for NEI development until the 2008 NEI. This distinction is important, as the authors later state their selection of the 2002 NEI as the reference year for the on-road sector is because the 2002 NEI is the most recent version using the MOBILE6 model (page 30331, lines 20-30). This explanation does not seem to be supported by actual NEI development methods. Thus, it is not clear why the 2005 NEI, which is the most recent NEI using the MOBILE6 model, was not selected as the reference year for the on-road sector as it was for other sectors considered in this study. The authors should clarify these discrepancies and provide a clear explanation of why the 2002 NEI was selected as the reference year for the on-road sector.

[Response]: In this study, we used the SMOKE-ready NEI data which only provides the current 2005 NEI with MOVES version (please see http://www.epa.gov/ttn/chief/emch/index.html#2005). We agree that it would be better to use the old MOBILE 6 version for 2005 NEI; however it is not publicly available any more. Future studies could use the newly released MOVES model to apply 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]: Page 30331, lines 7-14: Were there specific reasons for excluding 2008 NEI data from this study? I believe this is the most recent version of the NEI. Perhaps a sentence here explaining why the 2008 NEI data wasn’t used in this analysis would be useful for the reader.

[Response]: When our study of inventory development was initiated in late 2011, the 2008 NEI at that time (version v1.5) was not yet ready for SMOKE processing. Following the reviewer’s suggestion, we now have clarified the aspect by including the following: “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 (US EPA, 1993; Adelman and Houyoux, 2001).

[Comment]: Page 30333, line 1: The authors lay out a number of rules constraining values for emission factors calculated using equation 1. It would be helpful to include discussion of whether results from the application of equation 1 violated these rules and if so, how often. In cases where violations did occur what values were used for emission factors?

[Response]: Following the reviewer’s suggestion Section 3.1.1has been expanded to

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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]: What is the variability amongst individual states in emission factors calculated using equation 1 for a single source category?

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[Response]: There may be significant differences due to different control levels and combustion technologies. Table 3 gives the range of unabated emission factors. Because it is beyond the scope of this study to try and obtain such detailed information about the absolute percentage of each control or combustion technology applied in each individual state during 20 yrs, we instead ensured that all the averaged emission factors are within a reasonable range informed by the AP-42 dataset. We further clarified this aspect in the revised manuscript as below: "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 in this study, we attempted to back-calculate these emission factors from NEI data for seven years to quantify the evolution of emission controls."

[Comment]: Page 30333, line 14: It would be useful to know what sectors are considered to be uncontrolled. Perhaps an additional column with this information could be included in Table S1.

[Response]: This information has been added to Table S1.

[Comment]: Page 30335, line 2: In equation 2, how is fuel economy estimated? A source for these data should be cited.

[Response]: The data of fuel economy was obtained from National Transportation Statistics. We added in the following information to the revised manuscript: "The fuel efficiency for each type of vehicle in 1990-2010 was obtained from 2011 National Transportation Statistics."

[Comment]: Pages 30335-30336: I find it difficult to follow the steps used to calculate emissions from mobile sources. My understanding is that emission factors for each of the four on-road vehicle categories are calculated for a single reference year (2002) using emissions data published in the NEI and activity data calculated using equation 4. Emission factors for other years are then calculated by scaling the reference year emission factor using separate emission factor data published in the National Transportation Statistics. If this is the case, my concern is that the authors do not differentiate between gasoline and diesel fueled vehicles in their selection of on-road source categories and in emission factor calculations. Rather, emission factors calculated using equation 6 combine emission rates from gasoline and diesel vehicles into one "gasoline+diesel" category (shown in Figure 7). This is problematic due to the potentially large differences in pollutant emission factors for gasoline and diesel engines. For example, diesel engines tend to have higher NOx and lower CO emission rates than similarly sized gasoline engines. I suggest splitting each on-road source category by fuel type, as is done for the energy-related stationary sources. This could potentially simplify the calculation of emission factors, as the National Transportation Statistics data could then be used directly.

[Response]: We acknowledge that the calculation of mobile emissions in this study is simplified compared to using the original onroad model. This simplification is due to the unavailability of more detailed datasets such as state-level diesel fractions for each type of vehicle under different control levels for those 20 years. In this study, the trends of emission factor provided in the National Transportation Statistics are only limited to the national level. So we averaged the trends of emission factors of gasoline and diesel for the same vehicle size, basing on the diesel fraction (informed by MOBILE6 default data). We added the following clarification to the revised manuscript: "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)"

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[Comment]: Page 30336, lines 5-7: The authors state “necessary adjustment was made to ensure the calculated emission factors to be comparable with the vehicle emission standards and references.” The types of adjustments that were made and the reasons for making them need to be explained here. As written, this statement is ambiguous.

[Response]: The statement has been removed from the revised manuscript. We believe the additional clarification included in response to the specific reviewer’s comments will help remove the ambiguity.

[Comment]: Page 30336, lines 8-15: It is not clear what methods were used to calculate emission factors for nonroad mobile sources. Similar to my last comment, the statement “the evolution of emission factors for nonroad diesel and gasoline equipment was informed by NEI trends and Dallmann and Harley (2010)” is vague and needs to be expanded upon to give the reader a full understanding of the methods used here. Also, I am uncertain as to what is meant when the authors state nonroad emission factors were validated through comparison with the GAINS model.

[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 does not provide emission factors 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 that range we obtained from GAINS, suggesting that the activity we selected and emission factor we calculated in this study are suitable to use. Dallmann and Harley (2010) suggested that NOx and PM emission factors for off-road diesel-powered engines decreased significantly between 1996 and 2006. According to their results and the decrease ratio given in NEI trends, we assumed the average NOx and PM emission factors from off-road sources decreased by 25% and 18% over the past two decades.

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Due to the introduction of oil with lower sulfur content, the average emission factor of SO2 in transportation decreased by 40 %.

[Response]: We have added this explanation in the revised manuscript.

[Comment]: Page 30336, line 2: A description of ny is given, however, this variable does not appear in equation 6.

[Response]: We have corrected this error in the revised manuscript.

[Comment]: Page 30340, line 16-18: Reference needed for this statement.

[Response]: The following references were added to the revised manuscript: "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 SO2 and NO2) to their local emissions because their regional transport impacts are negligible." Lu, Z., Streets, D. G., Zhang, Q., Wang, S., Carmichael, G. R., Cheng, Y. F., Wei, C., Chin, M., Diehl, T., and Tan, Q.: Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000, Atmos. Chem. Phys., 10, 6311-6331, doi:10.5194/acp-10-6311-2010, 2010. Wang, S. W., Zhang, Q., Streets, D. G., He, K. B., Martin, R. V., Lamsal, L. N., Chen, D., Lei, Y., and Lu, Z.: Growth in NOx emissions from power plants in China: bottom-up estimates and satellite observations, Atmos. Chem. Phys., 12, 4429-4447, doi:10.5194/acp-12-4429-2012, 2012.

[Comment]: Page 30343, line16-24: Here and elsewhere in the manuscript (including in Figures 5, 8-9) the authors compare their emission trends results with NEI data. Methods for NEI development have been continually refined over the past ∼20 yrs, and are thus not consistent from year to year. I feel a better comparison would be with the NEI air pollutant emission trends data, which use a more consistent set of methods to estimate emissions from the source categories considered in this study.

[Response]: We cannot get such detailed sectoral emissions from NEI trends data (no fuel type was considered in NEI trends data). As stated earlier, the estimation of sector-based, spatially resolved, internally consistent emissions over a 20 year time period as introduced in this study represents an improvement over what is currently available in either NEI data or NEI trends.

[Comment]: Page 30344, lines 4-8: The discussion of the relative importance of different vehicle categories to NOx and CO emissions is oversimplified. For example, the authors conclude that, because NOx emission factors for heavy-duty vehicles are 5-10 times higher than light-duty vehicles, heavy-duty vehicles are a larger source of NOx emissions. This fails to account for differences in activity levels between vehicle types. For on-road vehicles, both emission factors and activity levels change over time and temporal trends may be vary amongst vehicle types. To understand changes in emissions over time, trends in both activity and emission factors must be considered. Also, the statement that heavy-duty trucks contribute more NOx emissions than light-duty vehicles and trucks in 2002 is not supported by data shown in Table 1b. In this table heavy-duty vehicles account for 20.2% of total NOx emissions, while light-duty vehicles & trucks account for 20.6% of total emissions.

[Response]: This sentence has been reworded to convey the fact that heavy-duty trucks contribute more NOx emissions than each of the other two sectors individually (not their sum). We revised that as below: “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 NOx emissions because of their higher NOx emission factor that is 5-10 times higher than that of light-duty vehicles and trucks, as shown in Table 1b.”

[Comment]: General Comment: The authors do not include uncertainty estimates for any of the results presented in this manuscript. In my opinion this is a major emission. At the least, the authors should include a section discussing sources of uncertainty in the data sources and methods used to estimate activity, emission factors, and emission trends.

[Response]: Please see our response to the general comments above. In the revised manuscript, we have provided additional comparisons of the estimated emission trends against observations for several species and regions. These comparisons provide a measure of the uncertainties associated with the pollutant trends in this new inventory. However, as noted above, an in-depth analysis of the uncertainties in the underlying data sources (such as activity data and emission factors) used to compute these trends is beyond the scope of this study.,

[Comment]: Table 3: Why are emission factors for on-road vehicles excluded from this table? This is an important energy-related sector.

[Response]: The mobile emission factors by type of vehicle were given in Figure 7. We modified the title of Table 3 as follows: "Summary of NOx and SO2 emission factors in energy-related stationary and nonroad sectors"

[Comment]: Figures 2-3, 6: These figures could be moved to the supporting information or removed from the manuscript entirely with little impact on the presented analysis.

[Response]: We moved those figure to supporting information.

[Comment]: Figure 7: I found this figure to be very confusing. I recommend removing the emission standards from the figure and focusing on annual fleet-average emission factors for 1990-2010.

[Response]: We moved the emission standards to supporting information.

[Comment]: Technical Comments: Page 30330, line 16: change "combustions" to "combustion" Page 30331, line 8: change "basing" to "based" Page 30331, line 20: delete "of" Page 30333, line 5: change "pervious" to "previous" Page 30339, line 16: change "importation" to "important" Page 30342, line 27: change "widely" to "wide" Page 30343, line 12: change "on" to "of" Page 30343, line 17: change "constraint" to "constrained" Page 30344, line 3: change "MMVOC" to "NMVOC"

[Response]: These typos have been corrected in the revised manuscript

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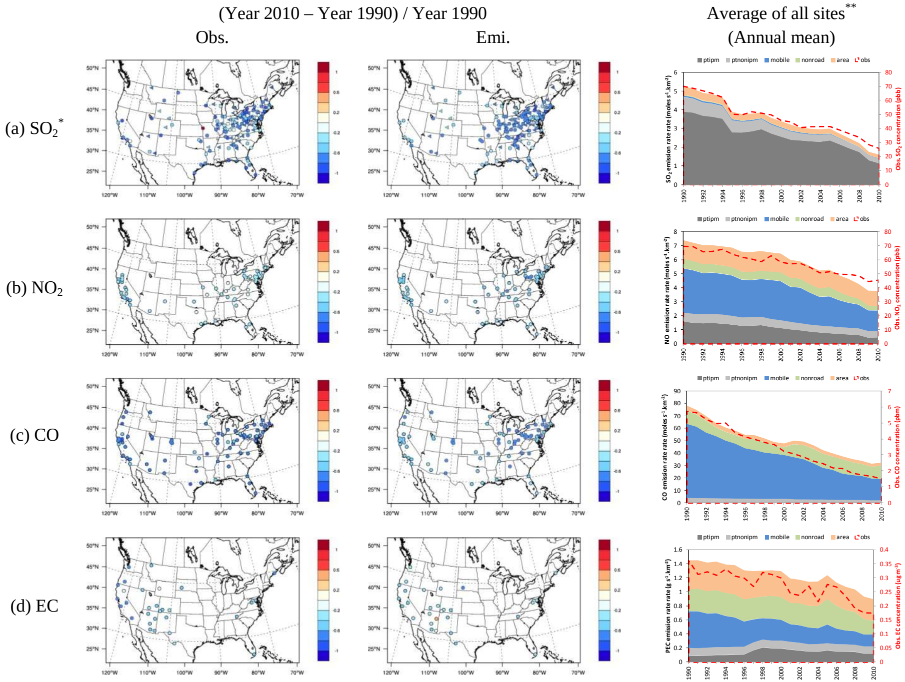

Fig. 1. Fig. C1. Comparison of historic trends between emissions and observed concentration from 1990 to 2010

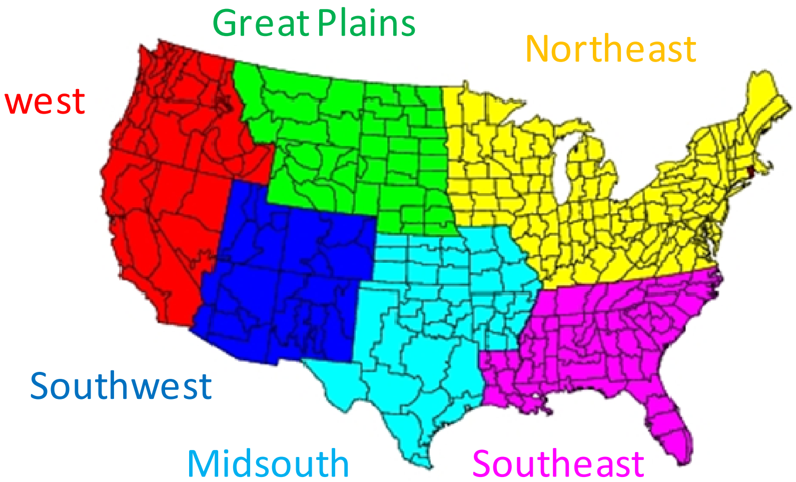

Fig. 2. Fig. C2. Definition of sub-regions used in analysis

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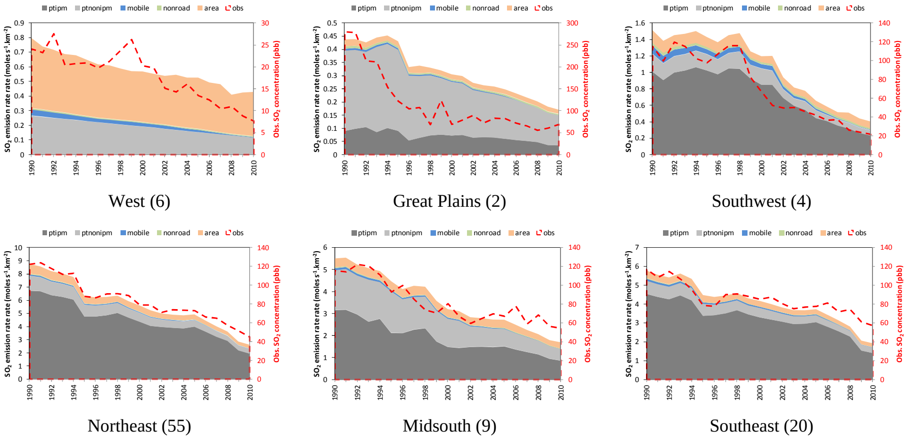

a. SO_2

Fig. 3. Fig. C3. Comparison of historic trends between emissions and observed concentration from 1990 to 2010 by sector and region

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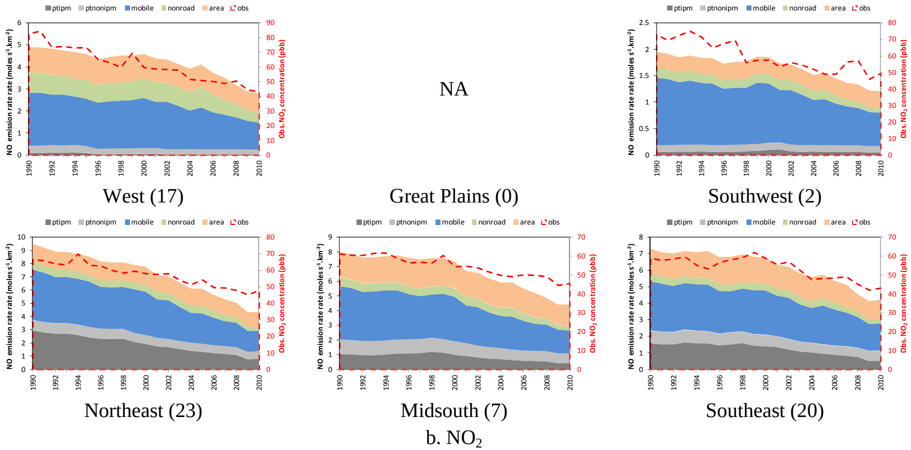

Fig. 4. Fig. C3. (continued)

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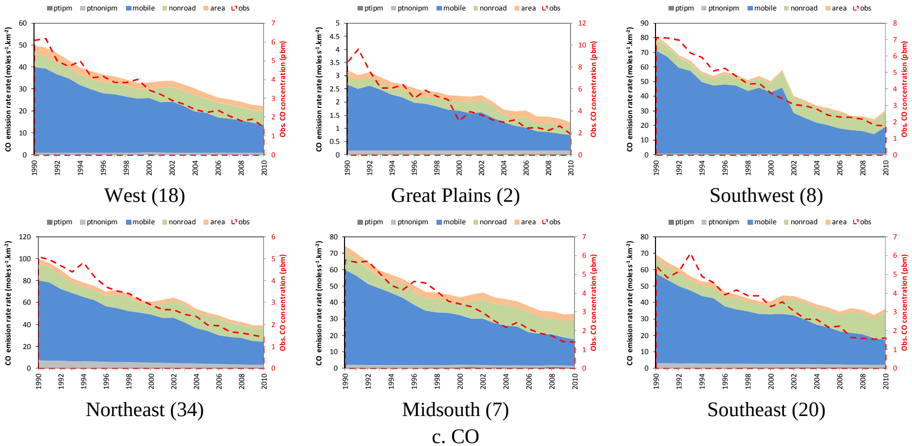

Fig. 5. Fig. C3. (continued)

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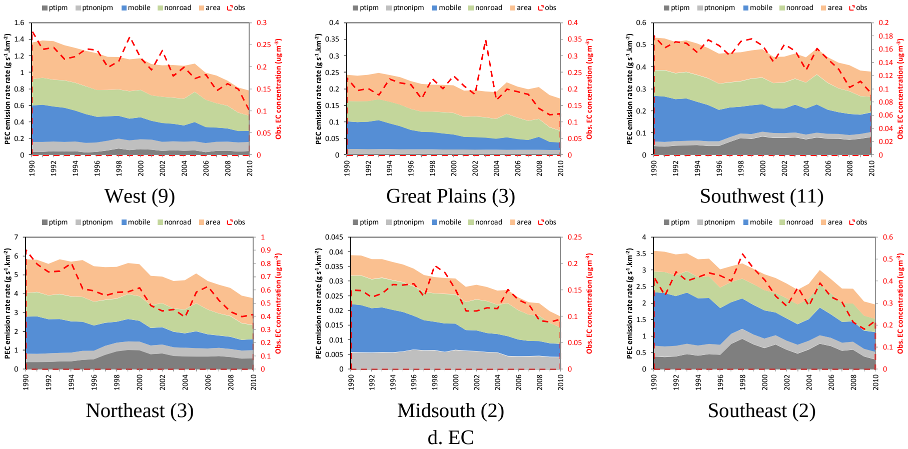

Fig. 6. Fig. C3. (continued)