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

## Anonymous Referee #1

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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

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(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. 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.

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. There are several parts in the manuscript that need clarifications and editorial corrections of typing errors. I suggest the manuscript to be accepted after major revision.

Specific comments 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.

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.

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

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?

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.

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

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

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

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

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? Can more discussions be added to sections 3.1.1 and 3.1.2? Figures 5, 8, 9, 10, and 13 provide the normalized trends. The absolute values need to be provided as Tables in supplementary material. 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.

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.

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

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

Figure 13: Figure caption should include the explanations about circles in the map.

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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).

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

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