

Manuscript ID: acp-2012-818

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

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**Overview:**

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. I found it difficult to understand the methods used for inventory development, particularly those used for estimating emission factors for on-road vehicles. Also, the lack of any discussion of uncertainty in the manuscript is troubling. 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. Specific comments on the manuscript are included below. I do not recommend this manuscript for publication in ACP without major revisions.

**Specific Comments:**

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.

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.

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

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.

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

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 NO<sub>x</sub> 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.

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.

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.

Page 30336, line 2: A description of  $\eta$  is given, however, this variable does not appear in equation 6.

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

Page 30343, line 16-24: 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.

Page 30344, lines 4-8: The discussion of the relative importance of different vehicle categories to NO<sub>x</sub> and CO emissions is oversimplified. For example, the authors conclude that, because NO<sub>x</sub> emission factors for heavy-duty vehicles are 5-10 times higher than light-duty vehicles, heavy-duty vehicles are a larger source of NO<sub>x</sub> 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 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 NO<sub>x</sub> 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 NO<sub>x</sub> emissions, while light-duty vehicles & trucks account for 20.6% of total emissions.

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 omission. 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.

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

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.

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.

#### **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”