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Response to Referee #1

Ksakousti Skyllakou et al.

Author comment on "Changes in PM_{2.5} concentrations and their sources in the US from 1990 to 2010" by Ksakousti Skyllakou et al., Atmos. Chem. Phys. Discuss.,
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(1) *What do authors mean by exposure and how is it estimated differently when compared to concentration? Is the exposure estimated considering the variable populations in 1990, 2001 and 2010 or was the population kept fixed?*

We define exposure here following Walker et al. (1999) as the product of the average concentration times the population in each grid cell. The population distribution is based on the US census bureau data (www.census.gov/) and is different for 1990, 2001 and 2010. The population distribution of 2001 is assumed to be the same with that of 2000. These clarifications have been added to the revised paper.

(2) *The authors represent 2 decades 1990-2010 with 3 representative years: 1990, 2001, 2010. Are 3 individual years enough to give a complete picture of the effect of change in emissions on concentrations? This question becomes even more important when authors estimate the exposure and hence authors should also include observed vs predicted exposure in grids where observations are available in Table 3.*

The changes in concentrations during these years are due to both changes in emissions and also to the year-to-year variability of the concentrations due to meteorology. We have chosen these years that are a decade from each other because in almost all cases the effect of the significant changes in emissions is expected to dominate the year to year to variability. This is consistent with the analysis the emissions used by Xing et al. (2013). We do not argue that each year is mathematically representative of the corresponding decade (e.g., 1990 of 1985-1995). They should be viewed as three snapshots of US air quality in time that reflect mostly changes in emissions plus some year to year meteorological variability. Please note that because of the way that exposure is defined (concentration times population) and the population is measured, the evaluation metrics of our exposure predictions are exactly the same as the evaluation metrics of our concentration predictions. We have added the above discussion to the paper.

(3) *The SOA exposure is in single digits in Table 3 which is much smaller when compared to concentrations. What is the reason behind the same?*

The referee probably refers to the changes in exposure in Table 1, given that Table 3

presents predicted and observed changes in concentrations. The relationship between the changes in concentrations and exposure is strongly affected by the corresponding distributions in space. The more similar these distributions are, the closer to each other the corresponding changes. For example, for the SOA from road traffic the SOA concentration was reduced by 71% from 1990 to 2010 and the exposure was reduced by 66%. On the other hand, for non-road sources with a lot of them located in low population density agricultural areas, the corresponding SOA concentrations (according to PMCAMx) were reduced by 17%, but the exposure by only 6%. A significant part of the corresponding SOA reductions took place in areas with only a few people. The explanation of this important point has been added to the discussion of the results.

(4) *Population plays an important role when estimating exposure; viz: higher population will result in greater no people being exposed to the same concentration when compared to smaller population. Since population increased during 1990-2010 in USA, reduction in pollutant emissions should be high enough so as to negate the factor of increased population in order to indicate an overall reduction in emission of a pollutant. Is it the same case here?*

This is a good point and indeed this is the case here. The US population increased by 23.7% from 1990 to 2010 and if the emissions and concentrations had remained constant, the total exposure would have increased by the same percentage. The fact that the exposure to PM was reduced in most areas indicates that the reductions in concentrations were sufficient to overcome this effect and still lead to significant overall population exposure reductions. This point is now made both in the abstract and in the conclusions of the paper.