Response to reviewer #1:

We thank Reviewer #1 for his/her valuable recommendations. Below are our responses to each comment. The reviewer's comments are shown in *italic*, while our responses are in plain font.

Review of "Air pollution and associated human mortality…" By Fang et al.
This paper discusses the simulated (using a single chemistry--climate model) changes from pre--industrial (1860) to present in mortality, looking at the specific roles of climate, emissions and methane. This paper is one in just a handful of papers dealing with estimating mortality changes using global chemistry--climate models. As such, there is considerable value in seeing it published. There are however a certain number of questions and loose ends that need to be addressed before publication.

General comments

1) *It seems that only present−day population maps are being used to assess the mortality. Maybe this is not the case and it needs to be made more explicit. If it is the case, then it should be justified. Population maps (regardless of their quality) are available for 1860 and should be used (http://themasites.pbl.nl/tridion/en/themasites/hyde/download/index-2.html).*

Response (1)
The reviewer is correct that we only use present-day population to assess mortality. This is because our study is intended to be a sensitivity study in which we evaluate the level of current air pollution and associated present mortalities resulting from emissions changes over the industrial period. In other words, our paper examines how "if air quality had remained at its preindustrial level, how many current-day mortalities associated with air pollution would have been avoided?" Within this context, we only utilize present-day population. This type of sensitivity study has been conducted in other air pollution-health modeling papers as well, such as *Anenberg et al.* [2010] and *Tagaris et al.* [2009].

We understand this reviewer's suggestion to additionally analyze the heath effect using preindustrial population, in which case, the reviewer is expecting us to evaluate the preindustrial premature mortalities associated with air pollution. However, if we were to calculate mortalities during the preindustrial period, in addition to the preindustrial population distribution, we also need to know the baseline mortality, age structure, and disease-based mortality for the preindustrial population. To our knowledge, this kind of information is not available; therefore, we prefer to keep our study as a sensitivity study that focuses on present-day mortality.

2) *The discussion on OH and H2O2 is of very little use in this paper. While there is a coupling to PM2.5, this is not discussed specifically enough o justify the space used by this discussion. I would strongly suggest removing altogether (page 22725, second paragraph, page 22727, lines 16--28 and section 4.3.3).*

Response (2)

We included discussion of OH and H2O2 in the first place because they were important to explain PM2.5 changes associated with increased CH4. We then discussed OH and H2O2 when examining other factors to keep consistency between sections. Finally, we also noticed that among all three factors (CLIM, EMIS and TCH4), CH4 plays either a primary or secondary role in determining H2O2 and OH, which argues that CH4 increase is very important not only because of its contribution to mortalities and climate change, but also because of its effect on the atmospheric oxidizing power.

However, we understand the reviewer's concern and agree that OH and H2O2 are not strongly and directly associated with the health effect, the major topic of this paper. Therefore, we have removed the paragraphs in the places suggested by the reviewer.

3)Figures 3 and 5 are too small to be useful.

Response (3)
We agree with the reviewer that these two figures are small in the ACPD publication. The size of original plots is reasonable, however, when they were formatted for ACPD paper size, they shrank. We will work with the publication staff to be certain they are of legible size in ACP.

Specific Comments:

4)Page 22714, Line 4: write "preindustrial"
5)Page 22714, Line 6: change "go beyond" to "extend"
6)Page 22714, Line 21: mention explicitly that this is global number
7)Page 22715, Line 18: change "apply" to "use"
8)Page 22716, Line 10: it is Anenberg, not Annenberg
9)Page 22716, Line 25: reference Shindell et al., Science, 2009.

Response (4 - 9)
We thank the reviewer for these suggestions and have made the recommended changes.

10) Page 22715, Line 12: this 3-4 increase is not true for BC (see Lamarque et al., 2010, page 7033)

Response (10)
Thanks to the reviewer for pointing this out. We have changed the sentence to: "Sulfate aerosol concentrations in Greenland ice cores suggest a factor of 3-4 increase from the mid-1860s to the present (Döscher et al., 1995; Fischer et al., 1998). Over the same period, European high-alpine glaciers indicate an increase in carbonaceous aerosols of a factor of 3 (Lavanchy et al., 1999), while Greenland ice-cores show little change (Lamarque et al., 2010)."

11) Section 2.2: no spinup?

Response (11)

Please refer to the last sentence in the second paragraph in Section 2.2, "All simulations were run for 11 years with the first year used as spin up." In order to further clarify simulation time and spin-up time, we additionally add one sentence near the end of the fourth paragraph of Section 2.2, "In order to distinguish signals, driven by changing emissions, CH_4 and climate, from internal model variability, we use annually-invariant SST, SIC and air pollutant emissions to drive 11-year model simulations and analyze averages of the last 10 years of each simulation."

12) Page 22722, line 16: change 10 ppbv to 15 ppbv, which by the way is not that small!

Response (12)
Thanks. We corrected the sentence by changing "10 ppbv" to "15 ppbv" and we remove the statement "relatively small".

13) Page 22724, line 8: what is the methane lifetime and how does it compare to Prather et. al. (2012)?

Response (13)
We estimate the total methane lifetime in our "2000" simulation as 9.0 years, consistent with Prather et al. (2012). We added this information to the paper. Detailed discussion on Methan

14) Page 22728, line 3: is that direct+indirect forcing?

Response (14)
Yes, it is the total radiative forcing.

15) Page 22728, lines 5-9: sentence does not belong to this section, if at all in a paper.

Response (15)
We removed the sentence.

16) Page 22728, line 25: this is not completely clear once the potential CO2 compensation is taken into account. Rephrase.

Response (16)

Here we mean the biogenic emissions of air pollutants. We revised the sentence to: "However, our historical climate change does not include the climate change induced effect on emissions of biogenic hydrocarbons, such as isoprene and terpenes, that are air pollutant precursors. If a biogenic air pollutant precursor emission response to the changing climate were included, it would likely increase our estimate of the changes in $PM_{2.5}$ and O_3 concentrations described below. "

17) Page 22729, line 5: correlated with what?

Response (17)

"The spatial distribution of PM2.5 changes, driven by climate change and driven by all factors, is only loosely correlated (R = 0.3)." Here, we mean that "The spatial distribution of PM2.5 change driven by climate change and that driven by all factors together are loosely correlated (R=0.3). " We have clarified the sentence.

18) Page 22733, line 9: does "anthropogenic" refer to all the changes or simply the changes in anthropogenic emissions. There is also a potential change in natural emissions and impact of methane on its lifetime, so "anthropogenic" might be a misnomer.

Response (18)
As in the *Fiore et al.* (2008) study, anthropogenic means the portion of CH4 emissions that are of anthropogenic origin. Their estimate is based on a scaling of emissions of CH4 and O3 responses from previous literature using CTM models (which should include the feedback of CH4 from anthropogenic sources on its own lifetime). To avoid confusion in the text, we now revise the sentence to "Although the impact of CH4 on O3 has been discussed in previous literature (Dentener et al., 2005; Fiore et al., 2002; Fiore et al., 2008; West et al., 2006), most of these studies focus on the potential benefit of future CH4 mitigation while our study examines the total change in O3 resulting from historic increases in CH4. However, the magnitude of CH4 impact on O3 in those studies are consistent with ours: for example, Fiore et al. (2008) estimate that anthropogenic CH4 emissions contribute 5 ppbv to global mean surface O3; West et al. (2006) and Anenberg et al. (2009) find that a 20% reduction in anthropogenic CH4 and a 20% reduction in global CH4 mixing ratio lead to approximately a 1 ppbv increase in global mean surface O3."

19) Page 22733, line 11: why can't the same measure be used instead of simply a "consistent" statement?

Response (19)
As mentioned in Response (18), their study is not exactly the same as our study, as we focus on the effect of historical changes in CH4 concentrations, while they focus on the impact of anthropogenic CH4 emissions (although we may expect that historical CH4 change may be mainly driven by anthropogenic emission changes during that time period). Our study focuses on the health and air quality effects due to changes during the industrial time period, including changes in climate, CH4 and air pollutant emissions. We revise the whole sentence to avoid the confusion. Please see Response (18).

20) Page 22738, line 29: there is really no discussion of cost prior to this? More information should be used to make this useful.

Response (20)
We revised the sentence from "As the benefit of CH4 reduction does not depend on its location, for cleaner regions, such as Europe, South America and Australia (where we find mortality burdens to be more sensitive to CH4 concentration than other regions), identifying the least cost CH4 mitigation option internationally can be a highly cost-effective way to reduce premature

mortalities from O3 exposure" to "As the benefit of CH4 reduction does not depend on its location, for cleaner regions, such as Europe, South America and Australia (where we find mortality burdens are more sensitive to CH4 concentrations than other regions), identifying low-cost CH4 mitigation options internationally may be an effective method of reducing local premature mortalities associated with O3 exposure."

References:

Anenberg, S. C., Horowitz, L. W., Tong, D. Q., and West, J. J.: An estimate of the global burden of anthropogenic ozone and fine particulate matter on premature human mortality using atmospheric modeling, Environ. Health Persp., 118, 1189–1195, doi:10.1289/ehp.0901220, 2010.

Tagaris, E., Liao, K.-J., DeLucia, A. J., Deck, L., Amar, P., and Russell, A. G.: Potential Impact of Climate Change on Air Pollution-Related Human Health Effects, Environ. Sci. Technol., 43, 4979–4988, doi:10.1021/es803650w, 2009.