!st reviewer response

We thank the reviewer for the overall positive assessment of this study and for providing helpful comments that have helped us in improving the manuscript.

Major points

1) Section 3.2., trends in AOD and SW radiation, is particularly unclear and difficult to read with frequent changes from all sky to clear sky to direct and diffuse and back, in model and observations, and intermixed with (maybe a bit too much) speculations on why a certain feature may be seen in the observations or may not be captured well by the model. This section in particular would benefit from a more structured, point by point discussion (model, observation, east, west) of all sky SSR, clear sky SSR, direct, and diffuse.

Looking at Table 5, points that may be worthwhile addressing include:

Clear sky trends (observations show stronger trends in the west than in the east, the opposite is true for the models; observations are dominated by the diffuse component, while direct and diffuse essentially compensate in the model when aerosols are present; given the different relevance of diffuse and direct in model and observation, as well as the different sensitivity of diffuse and direct to aerosols, one may wonder whether aerosols indeed play a dominant role for observed clear sky trends).

These discrepancies between the model and the observations particularly the differing trends in clear-sky direct and diffuse SW are discussed in some detail in Section 3.2.

All sky trends (stronger in observations than model, independent of aerosol feedback in model; much stronger than clear sky trends except for observations in the western US; the direct component dominates except for observations in the west; comparing east and west, modeled direct and diffuse are comparable without aerosols but are clearly different if aerosols are included).

Following the reviewer's suggestion, Section 3.2 is reorganized and simplified to minimize repetition of some points. The discussion points suggested by the reviewer are included in the revised version of Section 3.2.

2) The paper is, as mentioned above, a follow up of Gan et al. (2014, ACP) and Xing et al. (2015, ACP). This should be stated more clearly. Figure 1 and Table 1 are identical copies from Gan et al. (2014, ACP). This may be acceptable in order to have a self-contained manuscript, but it should be made clear. Similarly, the observational trends presented in Tables 3, 4, and 5 are identical to results given in Table 2 of Gan et al. (2014, ACP). Section 2.3. follows most closely section 2.5 in Gan et al. (2014, ACP). It may be also worthwhile to mention that site specific time series can be found in Gan et al. (2014, ACP).

The reviewer is correct that the present study builds upon Gan et al. (2014a, ACP) and Xing et al. (2015, ACP). This has been clarified in the revised manuscript on page 3, page 5 and the caption of Figure 1.

Minor points:

p. 17718, line 16: "In general ... the model output and emissions agreed well with CASTNET measurements...". One may add (in line with Xing et al. 2015) that this is not true for NO3, where even the sign of the trend is different in observations and model.

The NO3 trends shown in Figure 2 and Table 3 are directionally consistent between observations and simulations for both the Eastern and Western U.S.. As noted by the reviewer, this is indeed different from the results of the analysis of hemispheric CMAQ simulations presented by Xing et al. (2015). A likely reason for this difference in model behavior is that the present study used a different emission inventory than Xing et al. 2015 to drive the model. In particular, the Xing et al. 2015 study used the global EDGAR inventory while the present study used the 1990 – 2010 emission inventory developed by Xing et al. (2013) for North America.

p. 17719, line 24: "One of the possible reasons... such as sea salt, wild fires and underestimation of secondary constituents...". It seems not obvious how deficiencies in sea salt or wild fire emissions could affect aerosol trends. Being natural sources they are likely more or less constant, no? And how about other possibilities, e.g. aerosol properties (internal / external mixing, hygroscopicity, optical properties)?

This statement in our original manuscript referred to the underestimation of AOD, not the underestimation of aerosol or AOD trends. We have clarified this point in the revised manuscript and have included additional explanation and a discussion of other possible causes for the underestimation of AOD in the revised manuscript: Page 8 line 221 – page 9 line 237

p. 17720, line 6: "One of the possible causes... aerosol indirect effects". How about inaccurate cloud representation already in the absence of aerosols? Due to inaccurate boundary conditions, nudging, etc.?

In general, these factors may have effects in the simulation but we believe they are not the main factors in this underestimated trend. We tried to minimize these effects by using boundary conditions from a hemispheric simulation (these BCs are likely more representative of long term changes in inflow conditions) and using a moderate nudging to ensure that the model dynamical state does not diverge relative to the observations (see Hogrefe et al., 2015). This is further clarified in the revised discussion on page 10.

p. 17720, line 9: "Aerosol indirect effects have recently been included...". You mention this again a bit later, p. 17722, line 2. It may be easier for a reader if you discuss these (and other) potential short comings in one place instead of repeating them several times / scattering them throughout the text. After all, the title of your paper is 'assessment of WRF-CMAQ. Admittedly, though, this may not make sense in each case.

Yes, we agree with the reviewer's concern that the current discussion can be confusing. This section is reorganized and simplified.

p. 17720, line 25: "These anomalies are likely associated with the very strong El Nino...". To me, it is not obvious how even a strong El Nino should affect clear sky SSR in the US. Could you give a reference?

To further clarify the point, we have added the text below in the revised manuscript, page 11.

These anomalies are likely associated with the very strong El Nino occurrence of 1998-1999 which had significant impact on continental US weather patterns. For example, El Nino affects (i.e. increases) the rain and snow fall, water vapor and temperature in the atmosphere. As discussed in Long et al. (2009) and Gan et al. (2014), we allow some amount of condensed water in the atmospheric column under the "clear sky" classification. Dupont et al. (2008) show that up to an optical depth of 0.15 of primarily elevated ice crystals are still typically classified as clear sky. Augustine and Dutton (2013) show using SURFRAD data that there exists a moderate correlation between ENSO, and surface air temperature and surface specific humidity at the SURFRAD sites. Their Figure 7 shows the 1998-1999 El Nino increasing the yearly average specific humidity, with Bonneville and Goodwin Creek sites exhibiting the greatest increase of almost 1 g/kg. This increased humidity likely also increased the occurrence and/or amount condensed water in the atmospheric column at levels still classified as clear-sky, yet as shown in Figure 8 had an impact on the partitioning of the downwelling clear-sky SW, significantly decreasing the downwelling direct SW while increasing the diffuse SW. In the west, the decreased direct SW anomaly is about balanced by the increased diffuse SW, but not so for the east where the decrease in the direct SW is much larger.

p. 17720, line 28: "... may also be due to errors in model representation of emissions...". If I understand correctly, you are still referring here to two years that do not match well between observations and model. Why should the emissions be wrong only for these two years?

This discussion is attempting to convey that estimates of emissions for recent years has benefitted from the availability of more detailed activity and other auxiliary data then estimates from the early 1990s.

The statement "... may also be due to errors in model representation of emissions...". has been rephrased on Page 11 of the revised manuscript

p. 17721, line 15: Same question as p. 17720, line 6. Why does it have to be indirect aerosol effects and not just "misrepresentation of clouds" in general?

We agree that the representation of clouds in general is subject to considerable uncertainties, this includes the aerosol indirect effect not currently included in our model. This sentence is no longer part of the rewritten Section 3.2 in the revised manuscript.

p. 17722, line 2 and line 16: Both comments (on indirect aerosol effects and "whitening") you already had just one page earlier. Maybe no need to repeat them in the same section so frequently.

Thank you for pointing this out. The discussion is modified and the repeated portion is removed.

p. 17722, line 8: "This is further verified through the comparison of the feedback (FB) case...". The sentence seems a bit daring, given that the observed clear sky trend is dominated by the diffuse component, while the modeled trend is dominated by the direct component. Personally, I think this disagreement between model and observation (dominance of either direct or diffuse component) is among the most interesting findings of your study. It points, as you write yourself, to other relevant factors for clear sky trends beyond (simple) direct aerosol effects.

This sentence is rewritten on page 10 line 284 – page 11 line 287

One of the indications that the aerosol direct effect is contributing to the "brightening" is shown in comparison of the feedback (FB) case with the no feedback (NFB) case. As illustrated by the data tabulated in Table 5, almost no trends is apparent in the no feedback case for clear-sky total, direct and diffuse SW radiation.

p. 17722, line 27: "...in particular in the last 3 years (i.e. both of them decrease)". In the west it would be only the last 2 years. More generally, I find it doubtful to rely on only two or three years of data here.

The overall conclusion was made based on several indications and references. On page 12 line 327 – 335 of the revised manuscript, we have revised the discussion to address the reviewer's concern as follows:

For example, as a result of the increasing air traffic, ice haze layers associated with aircraft emission contrails (Hofmann et al., 1998) can potentially increase the diffuse radiation. More support for this theory was presented by Gan et al. (2014a); the pattern of US air carrier traffic (i.e. steady growth of air traffic from 1996 to 2007, followed by a decrease after 2008) agreed well with the pattern inferred in the observed clear-sky diffuse radiation especially during the last 3 years (i.e. both of them decreased). Moreover, Haywood et al. (2009) and Gerritsen (2012) illustrated that increasing contrails do increase the diffuse radiation. This suggests that contrails or sub-visual cirrus clouds and ice haze can play a role in the increasing trend noted in the observed clear-sky diffuse SW radiation.

p. 17723, line 22: "In particular, analysis of model and observations of clear sky total SW trends... agree well...may be due to better estimates of recent emission data sets." The concrete formulation is certainly correct. However, again, personally I find the really interesting fact here the difference between diffuse / direct in model / observations (see above).

Please see our detailed response above.

2nd reviewer response

We thank the reviewer for the many helpful comments which have helped us in improving this manuscript.

The topic of the paper is well suited for ACP. However, I feel this paper does not deliver what the title says, that is, to use the WRF-CMAQ simulation for understanding direct aerosol effects on radiation brightening in the United States. Some important aerosol species (dust, organic aerosols) seem to be excluded in the model, the calculated AOD is much too low, only half of observed values in both west and east US, and the attribution of SW radiation change to aerosol change is not convincing. I recommend a major revision. My comments are given below.

The WRF-CMAQ modeling system used in this study treats all relevant aerosol species, including dust and organic aerosols. Likewise, the model also used comprehensive emission dataset which included dust, organic carbon and CO etc. Additional details on the aerosol speciation represented in the CMAQ model can be found in Carlton et al (2010; ES&T), Foley et al (2010; GMD) and Appel et al (2013; GMD). Further Gan et al. (2014b) detail how the optical properties and AOD is estimated using the CMAQ simulated aerosol size and composition. However, in the analysis in this paper, we mainly focus on the change (i.e. reduction) of sulfate and nitrate which are the aerosol species most affected by emission reductions under the Clean Air Act and its amendments over the past two decades. We agree that other species also contribute to the aerosol direct effect and SW radiation but in order to keep the manuscript focused on trends over time, most of the analysis pertains to the species most strongly affected by emission reductions. However, it is important to note that the estimation of the optical properties and radiative effects of the airborne PM was based on the predicted spatially and temporally varying compositional characteristics which included the full suite of inorganic and organic constituents.

General comments:

1. Aerosol simulations: It has not been made clear how many aerosol species are included in the model. The concentration figures showed sulfate, nitrate, and EC, but no indication on what are included in AOD and SW radiation calculations. I noticed the statement in Figure 2 and Figure 3 captions: “Note that for emission dataset, only SO2 and NOx are available”. Does it mean that other aerosols and precursors are not available? If the model omitted other important species, like dust and organic aerosol, then the AOD and SW radiation calculation would be incorrect, and the paper would not be appropriate for assessing the aerosol effects on surface radiation trends.

As mentioned above, all relevant aerosol species (including dust, sea-salt and primary and secondary organic aerosols) are included in WRF-CMAQ as well as the radiation calculation (Gan et al, 2014b), and the emission dataset also included all relevant PM and PM precursor species. The reason for focusing on sulfate and nitrate in our analysis was discussed in our response above. The statement in Figure 2 and 3 was meant to indicate that SO2 emissions are shown along with both SO4 and SO2 concentrations since most of the atmospheric SO4 burden is due to secondary formation from SO2 rather than primary emissions of particulate SO4. The coupled WRF-CMAQ model uses a Mie and Core-shell scattering scheme (Gan et al. 2014b) for estimating aerosol optical properties and which are then used in the RRTMG scheme for radiation calculation. Additional clarification and information is added in Section 2.2 in the revised manuscript.

For more detail aerosol and radiation calculation please refer to Table 2, Gan et al (2014b), Wong et al. (2012) and below links:

http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQ_version_5.0_%28February_2012_release%29_Technical_Documentation#Two-way_Coupled_WRF-CMAQ

http://rtweb.aer.com/rrtm_frame.html

2. Clear-sky and all-sky SW surface radiation trends: The all-sky brightening trends over both west and east US are evident from the SURFRAD stations, which are pretty much reproduced by the model. Even though there is little change of aerosol concentrations over the west US in the 16-year time period, both model and data show a clear brightening trend in the west under all-sky condition. On the other hand, the model is much less successful in reproducing the SURFRAD trends under clear-sky conditions when aerosol is supposed to be the driver for the clear-sky trends. Such results may suggest (1) aerosol is not the major factor responsible for the all-sky brightening trends, and (2) the change of anthropogenic aerosol simulated by the model cannot explain the observed clear-sky brightening trend. These issues are the core for this investigation and should be seriously addressed.

We agree that factors other than the changing aerosol burden may contribute to the observed "brightening". Nevertheless, as indicated by our analysis of direct aerosol effects, i.e. the differences in simulated trends between the feedback and no-feedback simulations, there is a clear association between the change in aerosol burden and changes in clear sky radiation which are not explained when aerosol direct effects are not considered (as in the base model simulation; see Xing et al., 2015 for additional analysis). Secondly, the change of anthropogenic aerosol in the model and clear-sky SW radiation simulated by the model cannot completely represent the observed clear-sky brighting effect because of several factors listed below which are also discussed in the manuscript.

First, the observed clear-sky SW radiation is influenced by the contrails and cirrus cloud, these phenomena are not included in the current WRF-CMAQ model. Second, the observed clear-sky SW radiation is an estimated product derived from the all-sky measurements so it does contain uncertainty which had been discussed in the previous study Gan et al. (2014a) and Long et al. (2009). For example, as discussed in Long et al. (2009) and Gan et al. (2014a), some amount of condensed water in the atmospheric column is allowed under the "clear sky" classification. Dupont et al. (2008) show that up to an optical depth of 0.15, elevated ice crystals are still typically classified as clear sky. However, in the model, clear-sky SW radiation is 100% cloud free which implies that discrepancies can be expected when they are quantitatively compared. Lastly, even though the modeled clear-sky SW radiation trend is underestimated direction of the trend (i.e. increasing especially eastern US) is captured only when the aerosol direct radiative effects are included in the model.

3. Multi-decadal: The analysis and comparisons in this work is from 1995 to 2010, i.e.,16 years, which is not "multi-decadal". The title should be modified.

The underlying WRF-CMAQ simulations were actually performed for the entire 1990-2010 time period, but due to the lack of observations covering the entire time period the analysis presented in this manuscript covered the 1995 – 2010 time period. Following the reviewer's suggestion the title has been changed from "multi-decadal" to "long-term".

4. Uncertainty and error estimation and range of data: Because of the omission of the aerosol species in the model, it is necessary to provide uncertainties and estimated errors in model calculations. The uncertainty of the data should also be presented. In addition, the comparisons are shown for the aggregation of sites over multiple sites, the range or standard deviation of both data and model should be shown in the comparison figures, as well as the statistic measures (e.g., correlation coefficients, biases, errors, etc.).

As stated previously in our response to the reviewer's comments, there is no omission of aerosol species in the model. The uncertainty in the estimated trends is quantified by the significance ratio and confidence levels included in Tables 3-5. Standard errors of trends for observations and models have been added to Table 3, 4 and 5. Measurement errors have been summarized in Gan et al. (2014a) but quantifying their impact on estimated trends is beyond the scope of our study. Data from individual sites (both observed and model) is not shown to avoid cluttering the figure and obscuring the "main" trends for comparison. However, the measurements from each sites were presented in a previous study (Gan et al. 2014a).

Specific comments:

Page 17713, line 2-3: Does this opening sentence suggest you only consider sulfate and nitrate?

No, please see our detailed response to the general comments.

Page 17713, line 19-21: I don't understand why the "irregular" aerosol distribution of aerosol would be a big challenge for measurements to quantify the aerosol forcing. To me, the big challenge is the clouds that may dominate the all-sky radiative flux and may still contaminate the "clear-sky" data. Besides, "heterogeneous" is a better word than "irregular".

Both factors (heterogeneous aerosol distributions and clouds effects on radiation) are challenging because they cannot be quantified separately in radiation measurements. It is difficult to state which one is more important. Thank you for the suggestion, "irregular" is replaced with "heterogeneous" in the revised manuscript.

Page 17716, line 5: What aerosol and precursor species are included in this "comprehensive emission data"? This sentence contradicts the sentence in the captions for Figure 2 and Figure 3 saying "for emission dataset, only SO2 and NOx are available", as I mentioned earlier. Does the wild fire emission included? And dust? Page 17717, line 15: Add "that" in between "estimated trend" and "is statistically".

Please see our detailed response above for clarification on the aerosol species included in the model and how the aerosol size and composition is used to estimate the optical and radiative properties as well as clarification on Fig 2 and 3.

Yes, both forest fires and dust were included in our emission dataset. As mentioned in Xing et al. (2013), forest fires account for 7 and 17% of the total PM10 and PM2.5 emission, including wildland and prescribed fires. The dust sources considered in the emission were windblown, construction, mining and quarrying dust. The captions for Figures 2 and 3 are meant to indicate that SO2 emissions are shown along with both SO4 and SO2 concentrations since most of the atmospheric SO4 burden is due to secondary formation from SO2 rather than primary emissions of particulate SO4.

Page 17717, line 21-23, sentence begins with "The size of the circle": This is a bit out of context – are you talking about the circles in some of the figures? You should talk about this together with the figures.

Yes, we agree with this suggestion. The sentence has been rewritten as

"The size of the circle in the Figures 4, 5, 6 and 9 represents the level of the significance (e.g. the bigger the circle, the higher the significance)."

Page 17718, line 4: "feedback simulations" – if the meteorological fields are "nudged", how would feedback change them? Does feedback matter?

Yes, despite nudging there still is a clear effect of aerosol feedbacks as evident from a comparison of the feedback and no-feedback simulations in Table 5. It should be noted that the nudging employed in this study is weaker than the nudging employed in typical uncoupled retrospective WRF simulations. Hogrefe et al. (2015) showed that this weak nudging led to only a small reduction of the simulated direct aerosol effect on temperature compared to a no-nudging simulation while improving model performance. This information has been added to page 5 line 124-128.

The "nudged" meteorological fields (e.g. temperature, wind and moisture) affect the calculation of aerosol fields in CMAQ, which are then transferred back to WRF where they affect the radiation calculation. The strength of the nudging was chosen so as not to squelch this feedback effect (Hogrefe et al., 2015). As shown in Table 5, radiation trends from feedback and without feedback were different. More detail for the feedback and without feedback comparison can be found in Wong et al. (2012).

Additional explanations regarding "feedback" simulation is added into section 2.2.

Page 17718, line 5-6: "...the observed and modeled surface aerosol concentrations... are presented in Figs 2 and 3" – SO2, shown in Fig. 2, is not aerosol. It is a gas.

Thank you for pointing out this oversight, the sentence is rewritten as follows:

"First, the observed and modelled surface aerosol and gas concentrations are assessed at the CASTNET and IMPROVE monitors."

Page 17718, line 20-22: "...the model predictions agree well with surface measured aerosol concentration" – there is no comparisons of actual concentrations, only the concentration anomalies are compared.

The sentence is rewritten as follows:

"Overall, the model trend predictions agree well with surface observation trends for both networks, especially SO_4^{2-}."

Page 17718, line 21 and 23, page 17719, line 2 and 12, and page 17723, line 23: "...well..." – how well is "well"? This is a subjective term and should be avoided. The degree of agreement should be presented by statistics, such as correlation coefficient, bias, and error.

Following the reviewer's suggestion, a new table 6 with statistical metrics has been added to the revised manuscript.

Page 17719, line 8-9, trends of PM2.5, AOD: They don't necessarily change together in the same directions, as several previous studies showed positive correlation in the eastern US for some seasons but weak or even negative correlations over the western US. The relationship depends on aerosol vertical distributions, compositions, among others.

Yes, we agree with this comment and have removed the sentence in question.

Page 17719, line 22-23: The observed trend is 9 times stronger than the model simulated trend. Such difference means that the model is not able to "capture trends similar to observations".

We agree that from a numerical perspective the observed trend in the west is nine times larger than the modeled trend (obs_west: 0.0009 $year^{-1}$, sim_west: 0.0001 $year^{-1}$) but would like to note that the absolute magnitude the trends are relatively small for both. In contrast, the comparison for the eastern US shows much closer agreement (obs_east: -0.0012 $year^{-1}$, sim_east: -0.0017 $year^{-1}$). The sentence has been modified.

Page 17719, line 24-27: Which one of these possibilities is the most important one? What fire emission data are you using? Since dust is an important aerosol component in the west, omission of dust would be a significant problem for model. Sea salt contributions should be negligible for the sites shown in Figure 1.

In this statement, we are providing several potential explanations for the underestimation of AOD. Yes, the seasalt contribution should not be the main reason for the underestimation of AOD in this study. Note that dust emissions are included in our study.

The source of wild fire emission is obtained from

https://www.nifc.gov/fireInfo/fireInfo_statistics.html

Page 17720, line 1-4: Yes both model and data show a brightening trend, but is this trend due to the change of aerosols?

As elaborated to in our response earlier, in the current calculations we only consider the aerosol direct effect, and our results indicate that aerosols do contribute to the brightening effect (especially in the eastern US) but in the measurements other factors can also contribute to this effect.

Page 17720, line 13-15: Is the PM concentration underestimated in the model? By how much? The AOD is underestimated by a factor of 2.

The underestimation of AOD cannot necessarily be attributed to an underestimation / incomplete representation of PM concentrations. As stated in Gan et al. (2014b), several possible reasons for this model AOD underestimation are the underestimation of specific aerosol constituents such as organic carbon, low sea-salt concentration in the accumulation mode and uncertainties in characterizing the water soluble potion of the organic carbon leading to poor representation of refractive indices of organic aerosol. Furthermore, the hygroscopic effects of water soluble OC and external mixing are not considered in the current version WRF-CMAQ model. The omitted effects and uncertainties in representation of mixing state can play an important role in the apportionment of extinction. (Gan et al. 2014b, Curci et al., 2014).

Another study, Hogrefe et al. (2015) also showed in their Figure 2 that the underestimation of AOD occurs throughout all season despite the fact that the analysis of 24-hr average surface $PM_{2.5}$ predictions (Table 1 in Hogrefe et al., 2015) indicate overestimations of $PM_{2.5}$ during winter but largely unbiased $PM_{2.5}$ prediction during summer. Note that the $PM_{2.5}$ concentration represents a ground-level measurement while the AOD is an integration of aerosol extinction over a vertical column. Model predicted extinctions in any one layer depends not only on aerosol concentrations and properties but also relative humidity; thus differences in the vertical distribution of modeled aerosol concentrations and relative humidity would yield different calculated AOD even if $PM_{2.5}$ column mass was consistent.

This sentence is removed since section 3.2 is restructured and simplified. Additional clarification is added on pages 8-9 of the revised manuscript.

Page 17720, line 25 to page 17721, line 2: Do you expect the anthropogenic emission should change with El Nino? How? Also, from Figure 6, the disagreement of modeled and observed AOD for 1998-1999 is similar to that for other years!

The anthropogenic emission should not change with El Nino. They are separate factors and are discussed separately in the revised manuscript. The discussion was attempting to convey that “in general, the emission estimates for more recent years (after 2000) are better because of improvements in measurement techniques and greater availability of information for constructing the emission dataset”.

This statement "... may also be due to errors in model representation of emissions...". has been rephrased.

Second, we were trying to explain the factors which affect the anomalies of observed clear-sky SW radiation for 1998-1999. Discussions for underestimated AOD and observed clear-sky SW radiation anomaly are seperated into two paragraphs in the revised manuscript to avoid confusion.

Please see page 11 in the revised manuscript.

Page 17721, line 10-14: Indeed, if the clear-sky radiation change is dominated by aerosols, with the decrease of aerosols, the total and direct radiation should increase but the diffuse radiation should decrease. How is “clear-sky” defined in the SURFRAD data? What is the uncertainty in the clear-sky radiation observations?

These continuous clear-sky radiation estimates are a product from measured clear-sky SW rad together with all-sky SW radiation by using Radiative Flux Analysis (RFA). RFA is a series of codes developed to examine the time series of the broadband radiation measurements and detect periods of clear (i.e. cloudless) skies, then use the detected clear-sky data to fit appropriate functions, interpolate the fit coefficients across cloudy periods and thus produce continuous clear-sky radiation estimates. The resultant measured and clear-sky data are then used to infer various atmospheric and cloud microphysical properties, including daylight fractional sky cover for an effective field of view of 160 degrees, effective cloudy sky SW transmissivity calculated as the ratio of the total downwelling SW over the corresponding clear-sky total SW, and visible optical depth for overcast periods. For the observations of downwelling direct, diffuse and all-sky SW, the approximated uncertainties are 3% or 4 W/m^2, 6% or 20 W/m^2 and 6% or 10 W/m^2, respectively (Stoffel, 2005).

However, in this RFA, there is some amount of condensed water in the atmospheric column under the “clear sky” classification. Dupont et al. (2008) show that up to an optical depth of 0.15 of primarily elevated ice crystals are still typically classified as clear sky.

Clarification is added in revised manuscript at page 11. Additional details on SW radiation measurements can be found at Gan et al. (2014a).

Page 17722, line 3-4: The model-data discrepancy is much larger for clear-sky than for all-sky. If the implementing aerosol indirect effect might help improve the all-sky simulation, it would not be helpful for clear-sky simulations.

Yes, we agree with this statement. For the clear-sky scenario, including aircraft contrails, cirrus cloud and air traffic emission might improve the clear-sky SW radiation calculation especially diffuse radiation. This will be explored in future studies.

Page 17721-17723 of Section 3.2: The comparisons and discussions of radiation trends should be better organized. It is said in line 3-4 that "this study focuses on clear-sky SW radiation in the following discussion", but then the all-sky and clear-sky results are shown alternately without much organization.

Yes, we agree with the reviewer that this discussion is confusing. In the revised manuscript we have reorganized and simplified this section.

Page 17723, line 25-27: There is no sufficient evidence to point that the inaccurate emission before 2000 is responsible for the model-data discrepancy of AOD and radiative fluxes. As I said earlier, the agreement of simulated AOD and concentration trend before 2000 are similar to that after 2000.

We do believe that more recent emission datasets benefit from improved or more detailed measurements / information available form field studies, industries or local agencies. For example, wild fire emissions are provided by states after 2002 instead of national totals. As mentioned in Xing et al. (2013), for earlier years, information for some sectors was not as detailed as recent data so scaling factors based on activities were used to estimate some of the emissions for earlier years. Additionally, for some sectors information was not available at the appropriate spatial resolution, so assumptions in spatial allocation of emissions could potentially influence model spatial distributions and thus comparisons with measurements paired in space and time. We agree that this does not necessarily imply that the underestimation of AOD and discrepancy in clear-sky diffuse radiation from the model can be attributed to emission uncertainties alone. Nevertheless uncertainties in the historical record of emissions need to be considered when interpreting long-term model results.

Clarification is added on page 13.

Page 17724, line 2-4: If the model cannot reproduce the trends over the available observation sites, will it be a mistake to use such model to fill the observational gaps?

As noted above, the model does reproduce the directionality of the trends in PM concentrations, AOD, total all-sky and total clear-sky radiation and also captures regional differences in trends. We believe that despite the differences in magnitude between observed and modeled trends, the modeled fields can still provide valuable information about trends at unmonitored locations and the model evaluation results presented in this study can serve as a reference point for interpreting these model results at unmonitored locations.

Page 17723, line 12: "...influence by local terrain influences" – how? What are the references? This is not mentioned anywhere in the previous sections.

This was discussed in previous study, Gan et al. (2014a). References and additional explanation are added on page 13 of the revised manuscript.

Table 3 and 4: What quantity is in the “significance” column? Needs clarification.

“Significance” in this study is the absolute trend relative to its uncertainty $\frac{|\hat{m}|}{\sigma_m}$, please see Section 2.3 page 6 for clarification.