Response to Referee # 2

Overall Response: We thank the referee for the comments that are helpful in improving the manuscript. The detailed response is provided below following the reviewer's specific comments.

General comments

The manuscript shows WRF-Chem sensitivity simulations of anthropogenic and oceanic emissions in order to show their impact on the predictions of aerosols, clouds and radiative forcing. It uses state of the art measures to inter-compare the different simulations and successfully attempts to provide explanations to the responses of the system. The paper is scientifically sound, very well written and is in the scope of the journal. I recommend publication after some changes. My main concerns are related to a lack of explanation of why the anthropogenic effects are completely different depending if emissions x1 or x5 the base emissions, and the attempts of the authors to separate the aerosol indirect and direct radiative forcings in this highly coupled system.

Response 1: The two concerns from the referee are addressed as follows.

The responses of cloud and precipitation to AnthroEmis and ScaledEmis are similar over the polluted region P (e.g., Fig. 6). The reason "why the anthropogenic effects are completely different depending if emissions x1 or x5 the base emissions" over the remote region is primarily related to how drizzle frequency is affected by the addition of anthropogenic and enhanced anthropogenic emissions over this region. The effects of AnthroEmis are obtained by contrasting REF and 0ANT simulations. With negligible changes in anthropogenic aerosols in the two simulations over this region, the changes (REF-0ANT) in drizzle rate and frequency are relatively small (e.g., in Fig. 4 bottom panels, black versus green lines). On the contrary, when contrasting 5ANT and REF to estimate the effect of ScaledEmis, there are significant anthropogenic aerosol perturbations (5ANT-REF), which induce substantial changes in drizzle rate and frequency (e.g., in Fig. 4 bottom panels, red versus black lines). This has been discussed in the manuscript. Precipitation is important since it removes cloud liquid water and aerosols from the atmosphere. The near-coast region has low drizzle rates, and therefore anthropogenic aerosols have limited effect in inhibiting the precipitation; however, the drizzle rate is significant over the remote ocean, and aerosol could effectively impact precipitation. Thus, the impacts of aerosols on precipitation also affect cloud water amount and aerosol concentrations. Nevertheless, in response to the referee's comment we have added to the summary, "The clean region is heavily drizzling and the important role of drizzle in modifying distributions of water and static energy as well as aerosol sources (e.g., entrainment) and sinks in the MBL is tightly related to the high sensitivity over this region."

Regarding the direct forcing estimates, we first clarify the methodology. When calculating the direct shortwave radiative forcing, the offline code replicates the WRF-Chem radiation calculation using hourly WRF-Chem output files as input for aerosol and other atmospheric state variables. The offline radiative transfer module is called four times: with and without aerosol scattering and absorption, as well as for clear-sky (no clouds) and all-sky (i.e., using the WRF-Chem simulated cloud fraction, liquid water, and effective radii) conditions, respectively. We agree that all-sky aerosol direct forcing (-2.6 and -2.0 W m^{-2} over regions P and C) might be more representative than the clear-sky aerosol direct forcing. The current version of WRF-Chem does not calculate forcing estimates on-line. However, since the aerosol and atmospheric state variables are updated hourly in our offline calculation, the calculation should be quite close to that from an on-line calculation. We also agree that the residual obtained by subtracting the aerosol all-sky direct forcing from the total forcing is not exactly the indirect forcing, as this includes the semi-direct effect, indirect effect, other feedbacks, and likely non-linearity in the forcing estimate. Note that we did not mention these forcing estimates in the summary or abstract because they are a minor aspect in the manuscript. It is after all challenging to accurately estimate direct and in-direct forcings, and the accurate direct, semi-direct, and indirect forcing estimations such as those from Ghan et al. (2012) are beyond the scope of this paper. Also, see our response (Response 15) to the referee's later comments on section 3.5, final paragraph.

Specific comments

Abstract. "The reduction of precipitation due to this increase in anthropogenic aerosols more than doubles the aerosol lifetime in the clean marine boundary layer." What's the uncertainty on this given that there is no aerosol re-suspension after raindrops evaporate in the wet deposition parameterization? First, demonstrate directly that aerosol lifetime in the modeled MBL is simulated skillfully before considering this change.

Response 2: The estimation of wet-scavenging time-scale uses precipitation rates, and for precipitation prediction in the model, number concentrations are more important. The rain drops that fall out of the cloud base are aggregated from many small cloud droplets thus containing hundreds to thousands of CCN, however the resuspension only releases one aerosol per raindrop that completely evaporates before it reaches the surface. Thus, this resuspension effect would likely have very minor impact on aerosol and cloud droplet number concentrations and the wet scavenging timescale estimates for the remote region. The impact of the resuspension process on aerosol mass in the WRF-Chem is likely to be more uncertain, and is an area of ongoing research within our group. Currently, some of the co-authors are involved with related research within the framework of climate models. It is most likely that the impacts of resuspension with regional models are even smaller than with global models due to the significant aerosol sources from boundary conditions in regional models. As far as we know there are no direct measurements of wet-scavenging rates or aerosol lifetime available from VOCAL-REx measurements, thus it is difficult to directly assess the accuracy of the aerosol lifetime prediction skills using observations as the referee mentioned in his/her later comments. Also note that, due to the complex sources and sinks within the regional model, the aerosol lifetime cannot be calculated directly. Rather the wet-scavenging timescale is used to approximate aerosol lifetime. We agree that the accuracy of the calculated wet-scavenging timescale is limited by the accuracy of wet-scavenging parameterization in the model, which is yet to be assessed in our future research. However, the wet-scavenging parameterization used in WRF-Chem is typical of current regional and global climate models. Uncertainties in the wet-scavenging are expected to be primarily due to uncertainties in the cloud microphysics (cloud water removal rate), cloud macrophysics (cloud fraction), and aerosol activation (e.g., updraft velocity, and how much aerosol enters clouds via updrafts versus entrainment). The simulated cloud properties (cloud water, cloud fraction, precipitation rate) were evaluation in Q. Yang et al. (2011) and found to compare reasonably well with observations. The prediction of precipitation rate is also discussed in the last paragraph of Sect. 3.4. In addition, by contrasting the timescales within two simulations, it is likely that the systematic errors in relative changes in wet-scavenging estimates are smaller than those of the absolute values themselves.

Page 14628, lines 19-22: "To our knowledge, this is the first use of a regional model with prognostic aerosols and coupled aerosol-cloud-radiation to study the relative contributions of oceanic and anthropogenic aerosols to changes in cloud properties and radiative forcings over the SEP under realistic meteorological conditions at cloud-system resolving scale." I recommend removing this claim. There has been some work on this that the authors should probably be aware of, as they presented in the same meetings. From the VOCALS publication page (http://www.eol.ucar.edu/projects/vocals/publications/publications.html):
Spak, S., M. Mena-Carrasco, and G.R. Carmichael (2010). Simulating contemporary and preindustrial atmospheric chemistry and aerosol radiative forcing in the Southeast Pacific (Invited), Abstract A54B-07 presented at 2010 Fall Meeting, AGU, San Francisco, Calif., 13-17 Dec. From the VOCALS 3rd Meeting:
Spak, S., Simulating aerosol radiative forcing and impacts on marine stratocumulus, http://www.eol.ucar.edu/projects/vocals/meetings/2011/miami/presentations/tuesday/spak_the_u_032211.pdf

Response 3: We thank the referee for pointing out the two conference presentations by Spak, S. N. and coauthors. We meant to refer to studies in peer-reviewed publications, rather than studies from conference presentations. However, in response to the reviewer's comment, we have replaced, "the first use" with "among the first".

Page 14630, lines 13-26. The emissions inventory appears to also contain volcanic emissions (http://www.cgrer.uiowa.edu/VOCA_emis/). For the 0ANT, 5ANT, did you separate these emission sources so you wouldn't apply the 0x and 5x scaling factors?

Response 4: We did not separate the volcanic emissions from the anthropogenic emissions in VOCA emission inventory because the effect of volcanic emissions is relatively minor during our model simulation period. We have clarified this in the related discussion as follows. "The continental emissions are primarily anthropogenic but also contain volcanic emissions. The continental emissions are also referred to as anthropogenic emissions due to the dominance of anthropogenic sources and the negligible impacts of volcanic emissions to our results."

Page 14630, lines 23-26. "The results are, therefore, an extreme condition, and the magnitude of the influence would likely be smaller for changing anthropogenic emissions under most scenarios, although we expect that the tendency of the results will likely be the same." You results clearly show that when going from no anthro to 1xanthro and 5xanthro they generate very different responses, sometimes even with a different sign (radiation). So the tendency is not the same.

Response 5: It is that remote and clean regions have different responses (in direction/sign) to emissions rather than there are different (in direction) responses to AnthroEmis and ScaledEmis over the same region. Also note that anthropogenic emissions (AnthroEmis) have negligible impact over the remote ocean, and in Fig. 5a, the SW response to AnthroEmis is not statistically different from 0 at the 5% significance level (this has been pointed out in the revised manuscript) as shown by the error bars. Please see Fig. 6 for the detailed illustration of SW forcing for AnthroEmis and ScaledEmis. The shortwave forcing is by far the largest forcing compared to the changes in other energy fluxes, thus it is discussed in more details. To avoid confusion, the sentence has been rephrased to "The results are, therefore, a somewhat extreme scenario, and the magnitude of the response would likely be smaller for scenarios having smaller changes to anthropogenic emissions, although we expect that in each region the sign of the response will likely be the same as that of the 5ANT."

Page 14631, lines 11-12: "The aerosol direct radiative forcing at the surface is calculated as the difference in surface shortwave fluxes under cloud-free conditions with and without aerosols." What about cloudy conditions? Those are the ones that dominate here. Please quantify the percentage of gridcell-hours during the study period contain cloud-free conditions and their location to assess the representativeness of this value.

Response 6: Please see the third paragraph of Response 1 that addresses this question.

Section 3.1. I believe there is a missing explanation. For region C, you show almost no contribution to Nacc, CCN and AOD from anthropogenic, but then when anthropogenic is scaled by 5 it does show a contribution. Why? My first thought was that anthro aerosol doesn't get to region C so there is no contribution, but this is not correct as the scaled emissions wouldn't show an increase as well if this were true. As there is no anthropogenic contribution here, then there are also no sensitivities for cloud properties (Sect. 3.2).

Response 7: The transport of continental aerosols to the remote ocean MBL is mainly influenced by advection by mean winds, vertical turbulent mixing and cloud-top entrainment. With the same winds, mixing, and cloud-top entrainment, whether aerosols can reach the remote region or not depends on aerosol lifetime, or in other words, whether or not the aerosols get scavenged before they reach the remote region. In 5ANT, precipitation is strongly inhibited due to the enhanced anthropogenic aerosols within the MBL, thus aerosols in 5ANT have longer lifetime compared to those of the REF and are more likely to reach the remote region. Secondly, in 5ANT there are increases in cloud-top entrainment, which will lead to more tropospheric aerosols being entrained into the MBL, thus increasing aerosol concentrations. Therefore, with 5ANT, over the ocean (including both near-coast and remote regions) there are prolonged aerosol lifetime, increased aerosol sources, and decreased aerosol sinks, thus there are significant increases in aerosol concentrations over the remote region compared to the REF. In response to the reviewer, the following discussion has been added to the revised manuscript to clarify: "Over the remote region, the 25% increase in N_{CCN} due to the enhanced anthropogenic emissions (ScaledEmis) compared to the negligible impact of AnthroEmis in REF can be explained by the reduced precipitation (weaker aerosol sink and longer aerosol lifetime) and the increased cloud-top entrainment (stronger source of MBL aerosol) in 5ANT, which are discussed later."

Page 14635, Lines 23-25. You say DMS by itself generates an 8% increase in Nd, but before you said DMS generates not more than 6% increase in Naccum. I would expect increases in Naccum to always be higher than the increases in Nd. Can you elaborate on this and explain why this is happening?

Response 8: The 8% increase in N_d (ΔN_d = 11 cm^{-3}) is relative to the N_d concentration in the reference simulation (133 cm-3), and the 6% increase in accumulation mode aerosols (ΔN_{acc} = 23 cm^{-3}) is relative to the N_{acc} (384 cm-3) in the reference simulation. So the ratio of changes in N_d to N_{acc}, $\Delta N_d/\Delta N_{acc}$**,** is about 0.48, which is less than 1. We have modified the related text by providing not only a percentage change but also the changes in number concentration to avoid confusion.

Page 14638, Lines 28-29. "(_ 65% reduction in cloud-base and near-surface rain rate) due to anthropogenic aerosols leads. . .." I see almost no cloud sensitivity to anthro emissions in region C (figs 2-3), but you are saying that anthropogenic emissions generate a 65% rain reduction? You mean 5x anthropogenic? It would be helpful to show an additional plot of rain rates (cloud base will do) in Fig. 3.

Response 9: The whole paragraph is in the context of discussing responses to ScaledEmis over the remote region. We agree that just reading this paragraph by itself without the context is confusing. We have changed "anthropogenic aerosols" to "enhanced anthropogenic emissions (ScaledEmis)".

Section 3.3. You say there are changes in entrainment. I don't see a discussion about the additional/less aerosol that is being entrained and their impacts. It might be helpful to show the changes in the aerosols that are being entrained with their respective base quantities, maybe an additional panel in Fig. 2 showing delta Naccum on a layer above cloud top. Also, the changes in MBL height generated should generate changes in aerosol concentration as well (higher MBL for the same aerosol means dilution) which should to be discussed.

Response 10: We have mentioned on Page 14637, Line 14 that 'entrainment is an important process that is related to transport of aerosols from the free troposphere'. The calculated entrainment rates are likely to be biased high due to the relatively coarse vertical resolution used in the model compared to those in LES (e.g., 5 m). The use of relative changes in entrainment (in percentage) between two simulations minimizes the likely systematic high bias in the estimate. However, calculating changes to the aerosols due to entrainment would require using the raw entrainment rate, with its associated bias, and therefore would have a large uncertainty. Because of this, we are uncomfortable to provide the estimates of changes in aerosols due to entrainment.

The figure the referee suggested reflects the net changes in transported aerosol concentrations above cloud-top (due to changes in emissions and all associated processes) between two simulations with standard versus with enhanced anthropogenic emissions, but the subtle changes due to the entrainment alone cannot be distinguished in this type of plot.

In response to the referee's comment regarding "the MBL height increase has an effect of diluting aerosols in the MBL", the following discussion has been added to the revised manuscript: "The enhanced entrainment increases the amount of aerosols entrained from the free troposphere, although increases in MBL height dilute the MBL aerosols to some extent."

Page 14640, Lines 24-26. "Anthropogenic emissions increase daytime maximum decoupling frequencies to 33% and 61% over regions P and C." This is 5x anthro? Or no anthro vs base?

Response 11: This refers to ScaledEmis. We have modified 'Anthropogenic emissions' to 'Enhanced anthropogenic emissions (ScaledEmis)' to clarify.

Page 14643, lines 20-21. "Note that this neglects below-cloud wet-scavenging, but it is negligible for accumulation-mode number." This sentence makes sense as impaction efficiency is the lowest for accumulation mode aerosol, but I don't know if it's negligible considering that the wet scavenging parameterization in WRF-Chem does not consider aerosol re-suspension after rain evaporation. What fraction of simulated and observed accumulation mode loss in the MBL is due to wet deposition?

Response 12: Please see our response (Response 2) to the referee's previous comments regarding resuspension and aerosol lifetime. Again, we are not aware of measurements of aerosol wet deposition rates during VOCALS-REx. Therefore, we cannot compare model results to observations regarding 'what fraction of accumulation mode loss in the MBL is due to wet deposition'. Knowing what fraction of the simulated accumulation mode loss in the MBL is due to wet scavenging will allow more accurate estimates of aerosol lifetime. However, WRF-Chem does not include code for budget analysis of aerosols and our funding does not allow this level of code development at this time. Over the remote region, wet scavenging is the dominant loss process for sub-micron aerosol, and that is why we use the wet-scavenging timescale as an approximation of aerosol lifetime rather than directly calculate aerosol lifetime.

Section 3.5, first paragraph. This paragraph is very hard to read as it is right now, too many numbers. I think adding an additional panel to figure 5 showing the direct aerosol forcing would help.

Response 13: The estimate is only calculated based on the REF simulation, thus it is different from Fig. 5 that shows changes of energy fluxes in a sensitivity simulation compared to those of the REF simulation. To avoid confusion, we did not add it to Fig. 5. In addition, direct aerosol forcing is a minor aspect of this manuscript. That is why we did not mention it in the summary and abstract. We have revised this paragraph by deleting the direct forcing estimates to improve readability. See also the previous response (Response 1) to a related comment.

Section 3.5. You show results for the direct effect, but the model you use to compute it only considers clear-sky conditions, right? Is this computed for cloud free columns only and you assign 0 for the cloudy ones? What about the direct effect when there are clouds present (which is most of the time)? My concern is that you are not estimating the fraction of the total forcing due to the direct effect on a regional basis or over the stratocumulus deck when you compute this one without considering clouds. Maybe is not possible to separate the effects with this approach. Please discuss the presence or absence of non-additive effects of direct, semi-direct, and indirect radiative effects on low clouds in the model code for this set of physics schemes, those used in LES simulations for the same region and study period (e.g. Feingold et al., 2010; Kazil et al., 2011), and those included in GCMs, as well as results from empirical studies. Use box models if necessary to clearly isolate effects. If you can show a consistent process understanding and empirical evidence that the radiative effects are entirely additive, then the leave-one-out approach gains support, and the articIe and its results transcend the bounds of the specific model and configuration employed. If not, please reconsider the approach.

Response 14: This has been addressed in Response 1.

Page 14648, line 21. Again, it’s hard to quantify in an independent way direct and indirect effects in this coupled system. Direct effects could be overestimated by not considering clouds in the radiative transfer model used for this purpose.

Response: Please see our response in Response 1.

Page 14648, Lines 26-27. "and regional anthropogenic emissions have a negligible impact over this region." 1x Anthro doesn't have an impact but 5x anthro has a huge impact. You should rephrase this. Also, saying that it doesn't have an impact just by comparison to No anthro emissions scenario it's not appropriate, there should be an inflection point in between. Maybe it's something to explore.

Response 15: In response to the referee's comment, the sentence has been rephrased to "by contrasting 0ANT and REF simulations it is found that regional anthropogenic emissions (AnthroEmis) have a negligible impact over this region.". The reason why '5xAnthro reaches the remote region and yet 1xAnthro does not' has been addressed earlier (see Response 7).

Technical corrections

Page 14627, Line 17. Change "presentation" to "representation".

Response: Corrected.

Page 14634, Lines 10-11. "four-times" should be "five-times", right?

Response: It should be "four-times" there. Since 5ANT-1ANT = 4ANT, the response is expected to be 4 times of that with 1ANT perturbation. This has been clarified in the related text.

Fig 2. Caption. "sigma is the standard deviation of the mean changes." What does this mean? Why not state the overall standard deviation?

Response: We did not use the 'overall standard deviation' since the 'overall standard deviation' does not directly infer the statistical significance of the mean response presented. The ±2 x sigma of the mean change indicates the 98% confidence interval of the mean response.

Reference:

Ghan, S., X. Liu, R. Easter, P. Rasch, J. Yoon, and B. Eaton, 2012: Toward a minimal representation of aerosols in climate models: comparative decomposition of aerosol direct, semi-direct and indirect radiative Forcing. J. Climate. doi:10.1175/JCLI-D-11-00650.1, in press.