Response to Referee #1

Overall Response: We would like to thank the anonymous referee for the constructive comments. The detailed response is provided below following the reviewer's specific comments.

Yang et al. present a manuscript discussing four simulations using the WRF-Chem model where in sensitivity studies local aerosol emissions have been perturbed. No surprising results are found, and no new hypotheses are formulated. No comparison to observations is performed. Nevertheless, this study could be of some interest to a part of the community analysing the measurements from the VOCALS experiment.

Response: We thank the reviewer for this general comment. We are glad that the reviewer agrees that this study will be of interest to the community. The manuscript does report new and interesting findings specific to the Southeast Pacific.

The Southeast Pacific is an ideal location to study aerosol-cloud response under both polluted and clean marine conditions and is also a region of climate significance. As stated in our manuscript, "To our knowledge, this is the first use of a regional model at cloud-system resolving scale to study aerosol-cloud-precipitation interactions over the Southeast Pacific under realistic meteorological conditions. The analysis is based on month-long simulations. Thus, the results represent the response to emission changes under varying synoptic conditions over a longer time period than those in typical LES and MLM modeling studies, and hence can provide insights into the aerosol-cloud-precipitation interactions and their impact on climate.". Our result of a large aerosol lifetime effect over the clean marine region is a significant finding. Over the clean marine region, a 25% increase in CCN from the reference simulation more than doubles the aerosol lifetime over the clean marine boundary layer. Due to the positive feedback of aerosol lifetime on precipitation, there is a large response in cloud micro- and macro-physical properties. Compared to the polluted region, the large response to changes in anthropogenic aerosol over the remote region has implications towards slow manifolds and multiple cloud regimes in stratocumulus clouds (Bretherton et al., 2010).

Through quantifying and contrasting the response of cloud/precipitation to anthropogenic and natural aerosols over both clean and polluted marine regions we have presented a relatively complete picture of aerosol-cloud interactions over the Southeast Pacific using WRF-Chem, which sets the stage for future aerosol-cloud studies over this region using the regional model. Our results imply the important role of natural aerosol in accurately quantifying anthropogenic forcing. The simulated cloud responses and feedbacks to aerosol perturbations are in general agreement with those from previous studies using process-based models (e.g., more frequent daytime decoupling due to an increase in aerosols in Sandu et al., 2008 {page 14640 lines 15-29}; stronger entrainment in response to anthropogenic aerosols in Bretherton et. al., 2007 {page 14637 line 12 to page 14638 line 17}), although studies using process-based models in the literature are not directly comparable to ours because of different aerosol perturbations, our use of current emissions and different larger-scale meteorological conditions. This strengthens the credibility of the WRF-Chem regional model with prognostic aerosols and coupled aerosol-cloud-radiation processes in simulating aerosol-cloud interactions that will benefit the WRF-Chem community. We did not highlight this point in the manuscript, but will include this in the summary of the revised manuscript.

Regarding the lack of comparison to observations in the present paper, this manuscript is the follow-up study of our previous WRF-Chem model evaluation paper (Yang et al., 2011). The reference/standard simulation shown in the present study has been evaluated extensively against observations from VOCALS campaign and satellite observations in Yang et al. (2011). Using the evaluation paper as a foundation, this manuscript is able to focus on the modeling study of roles of natural and anthropogenic aerosols in the VOCALS domain. The known model biases from our previous model assessment work and their likely impacts on the simulated response are also discussed at various locations (e.g., page 14636 line 24 to page 14637 line 2; page 14639 lines 3-12; and page 14645 lines 10-17) in the manuscript.

In any case it would be necessary that the authors clearly discuss what is found simply due to the way it is parameterised as such in the model, and which results are found due to unexpected interactions of different parameterisations. For this

purpose, it would also be necessary that the relevant parameterisations are reported in this manuscript (ideally in terms of the equations).

Response: The relevant parameterizations are well documented in previous literature, such as Yang et al. (2011) for coupling between microphysics and aerosols, Morrison et al. (2009) for microphysics, and Zaveri et al. (2008) for the MOSAIC aerosol module. However, based on the reviewer's suggestion, in the revised manuscript we have added more details about the parameterizations that are important to our results and conclusions, as appropriate. We have also added discussion regarding what results may be due to the specific parameterizations and what may be due to complex interactions and feedback.

The authors present some "ACI" parameters. However, these are defined here in a way inconsistent with the published literature, and it would be useful to revise this.

Response: The "ACI" parameters used in this manuscript are partial derivatives approximated with finite differences from different model simulations and are used to quantify the sensitivity of cloud properties to changes in aerosol from either anthropogenic sources or natural sources over near-coast and remote regions. Similar use of such partial derivatives (approximated by finite differences) can be found in the literature such as Chen et al. (2011). This definition is different from that in some other papers (e.g., Feingold et al., 2003; Quaas et al., 2009), where the ACI is calculated by performing a linear regression of cloud parameters against aerosol parameters. In the revised manuscript, we have renamed the "ACI index" to "aerosol-cloud sensitivity factor" to avoid confusion and will point out the difference between the aerosol-cloud sensitivity factors and the ACI indices used in the literature.

The notion of "sea salt effects" is strange. Obviously, anthropogenic aerosols plus unspecified advected aerosols are taken as background, and sea salt aerosols, as an external perturbation. The relevance of this specific sensitivity study is very unclear.

Response: As discussed in the manuscript, "Sea-salt particles are hygroscopic and have larger surface areas than anthropogenic aerosols, such as sulfate. Therefore,

the condensation of gaseous sulphuric acid and water preferentially occurs on them, thus inhibiting new particle formation, lowering the maximum supersaturation in clouds, and suppressing the activation of anthropogenic aerosols in clouds (Ghan et al., 1998)."

It might be that what have been interpreted as "indirect effect" is what we intended to be "sensitivity " or "response" of cloud parameters to aerosol emissions/sources. The specific sensitivity simulation is to help illustrate the importance of locally emitted sea-salt particles. It might be taken for granted that with the presence of background sea-salt particles, the representation of local sea-salt emissions in a regional model is not important for the purpose of estimating the effect of anthropogenic aerosol over the polluted region. In addition, there are large uncertainties in sea salt emissions treated in contemporary models, and due to the dependence of sea-salt emissions on wind speed, the emissions have large temporal and spatial variability. For those reasons, a test of this "external perturbation" is meaningful. The remote clean marine region is an interesting region with about twice as high wind speed as the near-coast region and correspondingly higher sea-salt emission rates; however, the precipitation scavenging is also much stronger. This sensitivity study is also relevant to one of the VOCA project goals to determine sources of droplet number concentrations over the remote ocean. Results of this sensitivity simulation also suggest the importance of aerosol sources from boundary conditions (advected) over the remote region. These clarifications have been added to the revised manuscript.

Some specific remarks: p14625 l17: This concept of the "second indirect effect" is outdated. Microphysical feedbacks to a perturbation in aerosol concentrations are far more complex than the overly simplistic concept reported here.

Response: We agree with the reviewer that the aerosol effects on cloud microphysics and the associated feedbacks are far more complex than the conventional "second indirect effect". We have revised p14625 Line 17 as shown below to avoid using this simplistic concept when explaining such effects and feedbacks.

"*The second indirect effect describes how increases of CCN suppress rain formation, leading to longer cloud lifetime, larger liquid water path (LWP), and greater cloudiness (Albrecht, 1989). The large uncertainties related to aerosol direct and first indirect effects limit our understanding of the anthropogenic forcing on the climate system (Solomon, 2007). The second indirect effect is less well understood than the first and is even more difficult to quantify in climate models.*" has been revised to "The conventional second indirect effect describes how increases in CCN suppress rain formation, leading to longer cloud lifetime, larger liquid water path (LWP), and greater cloudiness (Albrecht, 1989). However, LES studies of aerosol perturbation have revealed complex microphysical and dynamical feedbacks that are more complicated than traditionally depicted as the second indirect effect (Wang and Feingold, 2009 and references therein). The large uncertainties related to aerosol direct and first indirect effects limit our understanding of the anthropogenic forcing on the climate system (Solomon, 2007). The processes and feedbacks through which aerosols affect clouds and precipitation are less well understood and are more difficult to quantify in climate models."

l24: It is probably better to refer to a commonly accepted review such as the one from IPCC (2007).

Response: As suggested, we have modified the sentence to only mention the range of first indirect effect from IPCC (2007). "*With varying representations of aerosols, clouds, and their interactions with radiation, previous modeling studies estimated global mean top-of-atmosphere (TOA) radiative forcings range from −0.5 to −1.9 W* m^{-2} *and from −0.3 to −1.4 W*m^{-2} *due to the first and second indirect effects, respectively (Thomas et al., 2011 and references therein).*" has been revised to "The Intergovernmental Panel on Climat Change (IPCC) fourth assessment report (AR4) provided a first indirect effect forcing estimate of -0.22 to -1.85 W m^{-2}, and its uncertainty was the largest among various contributors to anthropogenic forcing (Solomon, 2007)."

l25: This is a strange notion of aerosol indirect effects. Usually the effect of the anthropogenic perturbation of the aerosol on clouds is considered as indirect effect.

The background natural aerosol concentration is of course important for the magnitude of the indirect effect, but a "counteracting" is not happening.

Response: In response to the reviewer's comment, "*Over the ocean, the effect of anthropogenic aerosols is counteracted by that of large sea-salt particles.*" has been replaced with "Over the ocean, aerosols from natural emissions (e.g. sea salt and sulfate from DMS oxidation) play a critical role in determining cloud properties and the influence of anthropogenic aerosol on clouds and precipitation".

p14626 l7: It is unclear with respect to which reference the comparisons are done.

Response: The sentence has been revised to "Sea-salt particles could also serve as giant (e.g. > 2 μm in diameter) and ultra-giant (e.g., > 10 μm in diameter) CCN, and they have been found to promote drizzle production (Feingold et al., 1999), decrease total droplet number (Rosenfeld et al., 2002), and reduce LWP in polluted clouds (Lu and Seinfeld, 2005)." in the revised manuscript.

p14627 l9: Cloud microphysical processes are not resolved but parameterised in LES.

Response: Thanks for pointing this out. This has been revised by deleting 'cloud microphysical processes'.

p14629 l8: Since these parameterisations are essential to understand the results, they have to be reported here.

Response: More details are provided in the revised manuscript as described in an earlier response above.

p14633 l6: What is the parameterisation of Delta N_CCN /Delta N_acc, or rather: can this term not be inferred from the activation parameterisation?

Response: The $\Delta N_{CCN}/\Delta N_{acc}$ is calculated by differencing two separate simulations. N_{CCN} is part of the activation parameterization. The model uses a sectional representation for aerosols, with 8 size bins. Within each size bin, particles are assumed internally mixed. For calculating N_{CCN} at 0.1% S (or any other

supersaturation), we assume a narrow lognormal size-distribution for the particles within a size bin. The composition of the bin gives a volume-weighted hygroscopicity, from which the dry-diameter having critical supersaturation equal to 0.1% is calculated. The particles in the bin with dry-diameter exceeding this critical diameter contribute to N_{CCN}. As noted in the paper, N_{acc} is the number of particles with dry-diameter below 0.078–1.25 μm (bins 2–5). The $\Delta N_{CCN}/\Delta N_{acc}$ is calculated by differencing two separate simulations.

p14634 l27: So nDelta is the difference between the regional-temporal averages of two simulations? In the studies cited, it is rather a linear regression of instantaneous values for a certain (order of 100 km) pixel-size.

Response: Yes, the 'Δ' is the difference between two regional temporal averages of two simulations, which is different from the linear regression approaches used in the literatures cited. We have clarified this in the revised manuscript, noting that Penner et al. (2011) found a significant difference between these two methods.

p14635 l17: It is odd to speculate about model results. Is this effect parameterised?

Response: In Ghan et al. (1998), detailed numerical simulations were conducted to diagnose the water competition effect between sulfate and sea-salt aerosols. The model activation scheme (Abdul-Razzak and Ghan, 2000) used in that study "treats multiple aerosol modes, each composed of internal mixtures of material and each competing with each other for water with the use of a maximum supersaturation," and "the Kohler theory is used to relate the aerosol size distribution and composition to the number activated as a function of maximum supersaturation" (Abdul-Razzak and Ghan, 2000). Thus, the water competition effect is expected from the parameterization. We have rephrased the sentence to indicate that this effect is expected. An additional figure shown below illustrates the suppression of activation when sea-salt particles are added using the Abdul-Razzak and Ghan activation scheme. The related discussion in the original manuscript, "*Ghan et al. (1998) found that the competition for water vapor between large sea-salt particles and submicron particles (e.g., non-sea-salt sulfate) lowers the maximum supersaturation in a cloud*

updraft, and thus suppresses the activation of the more numerous submicron particles." will be rephrased to "When using the Abdul-Razak and Ghan (2000) aerosol activation scheme, the addition of sea-salt particles lowers the maximum supersaturation, and thus suppresses submicron sulfate particle activation. Therefore, it is expected that in the simulations the competition for water vapor between large sea-salt particles and submicron particles (e.g., non-sea-salt sulfate) lowers the maximum supersaturation in a cloud updraft, and thus suppresses the activation of more numerous submicron particles."

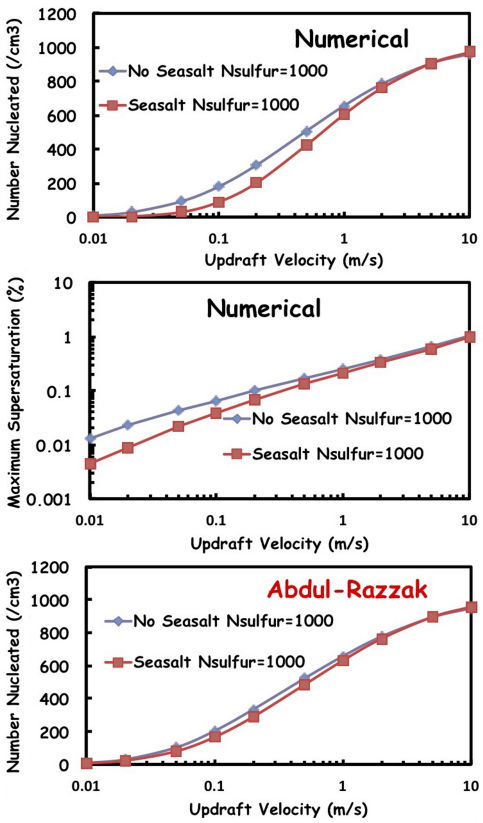

Fig. 1. Number concentrations of aerosols activated and maximum supersaturation as a function of updraft velocity with and without sea-salt particles based on detailed numerical simulations and the Abdul-Razzak and Ghan (2000) parameterized scheme. The sulfate aerosol is represented by a lognormal size distribution with a number modal radius of 0.05 μm and a geometric standard deviation of 2 (Quinn et al., 1990; Ghan et al., 1998). The sea-salt particles are represented by two lognormal size distributions with number mode radii of 0.1 and 1.0 μm, geometric standard deviations of 1.9 and 2.0, and number concentrations of

17 and 1.36 cm^{-3} for the film and jet modes, respectively (Ghan et al., 1998).

p14636 l2: A "mitigation" effect would usually imply a feedback. However, this is not what is discussed here.

Response: The sentence has been rephrased to "This implies that the presence of sea-salt particles reduces the sensitivity of cloud albedo to anthropogenic aerosol perturbations."

p14641 l28: L'Ecuyer

Response: Corrected.

p14644 l15: In order to judge on this, an assessment of the parameterisation of autoconversion in the model would be necessary. What is the approximate threshold put into the model?

Response: The autoconversion scheme used is based on Khairoutdinov and Kogan (2000) using an explicit autoconversion rate:

$$\frac{\partial q}{\partial t} = 1350 Q_c^{2.47} N_c^{-1.79}$$

where Q_c and N_c are cloud water mixing ratio and droplet number concentration, respectively. Unlike some other autoconverion parameterizations (e.g., Manton and Cotton, 1977), this formula does not include any threshold effective radius, but does implicitly depend on mean droplet radius given its dependence on both Q_c and N_c (i.e., mean droplet radius is proportional to $(Q_c/N_c)^{1/3}$). Thus, there is no explicit threshold effective radius applied in this autoconversion parameterization, but the dependence on Q_c and N_c helps to produce a sharp increase in probability of precipitation (POP) near ~ 12 microns. This will be clarified in the revised version.

p14663 "Changes" with respect to what? - It would be necessary to define these formally. The red numbers need more explanation here. The definition of the standard deviation is unclear. Is it the temporal standard deviation of the regional average values?

Response: We thank the reviewer for the suggestions. The caption and the related text have been revised to clarify. "Changes" are with respect to the reference simulation. The standard deviation is not the temporal standard deviation of the

regional average values. It includes both temporal and spatial variations within a region. The standard deviation is calculated as sqrt(var(x)/n); the var(x) is the variance of changes in a parameter in the sensitivity simulation relative to the reference simulation which is computed based on hourly model-grid outputs within the defined region for the simulated period; and n is the total number of data samples.

REFERENCE:

Abdul-Razzak and S. Ghan, A parameterization of aerosol activation 2. Multiple aerosol types, J. Geophys. Res., 105, D5, doi:10.1029/1999JD901161, 2000.

Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, Geophys. Res. Lett., 34, L03813, doi:10.1029/2006gl027648, 2007.

Bretherton, C. S., Uchida, J., and Blossey, P. N.: Slow manifolds and multiple equilibria in stratocumulus-capped boundary layers, J. Adv. Model. Earth Syst., 2, 14, doi:10.3894/JAMES.2010.2.14, 2010a.

Chen, Y.-C., Xue, L., Lebo, Z. J., Wang, H., Rasmussen, R. M., and Seinfeld, J. H.: A comprehensive numerical study of aerosol-cloud-precipitation interactions in marine stratocumulus, Atmos. Chem. Phys., 11, 9749–9769, doi:10.5194/acp-11-9749-2011, 2011.

Feingold, G., W. L. Eberhard. D. E. Veron, and M. Previdi, 2003: First measurements of the Twomey aerosol indirect effect using ground-based remote sensors. Geophys. Res. Lett., 30, No. 6, 1287, doi:10.1029/2002GL016633.

Khairoutdinov, M., and Y. Kogan (2000), A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus, Mon. Wea. Rev., 128(1), 229-243. Ghan, S. J., Guzman, G., and Abdul-Razzak, H.: Competition between sea salt and sulfate particles as cloud condensation nuclei, J. Atmos. Sci., 55, 3340–3347, 1998.

Penner, J. E., C. Zhou, L. Xu, and M. Wang (2011), Reply to Quaas et al.: Can satellites be used to estimate indirect climate forcing by aerosols?, Proc. Nat. Acad. Sci. U. S. A., 108, E1100–E1101.

Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey, A., Feingold, G., Hoose, C., Kristj ´ansson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P. A., Stier, P., Grandey, B., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevåg, A., Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol indirect eff ects – general circulation model intercomparison and evaluation with satellite data, Atmos. Chem. Phys., 9, 8697–8717, doi:10.5194/acp-9-8697-2009, 2009.

Quinn, P. K., T. S. Bates, J. E. Johnson, D. S. Covert, and R. J. Charlson,I nteractionsb etweent he sultbra nd the reduced nitrogen cycles over the central Pacific Ocean, J. Geophys. Res., 95, 405-416, 1990.

Manton, M. J., and W. R. Cotton, 1977: Formulation of approximate equations for modeling moist deep convection on the mesoscale, Colorado State University Technical Report, p.62.

Morrison, H., Thompson, G., and Tatarskii, V.: Impact of cloud microphysics on the development of trailing stratiform precipitation in a simulated squall line: comparison of one- and two- moment schemes, Mon. Weather Rev., 137, 991–1007, doi:10.1175/2008mwr2556.1, 2009.

Sandu, I., Brenguier, J. L., Geoffroy, O., Thouron, O., and Masson, V.: Aerosol impacts on the diurnal cycle of marine stratocumulus, J. Atmos. Sci., 65, 2705–2718, doi:10.1175/2008jas2451.1, 2008.

Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K., Tignor, M., and Miller, H. L. (eds.): IPCC, 2007: Summary for Policymakers, in: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, UK and New York, NY, USA, 2007.

Wang, H. and Feingold, G.: Modeling mesoscale cellular structures and drizzle in

marine stratocumulus. Part II: the microphysics and dynamics of the boundary region between open and closed cells, J. Atmos. Sci., 66, 3257–3275, doi:10.1175/2009jas3120.1, 2009a.

Yang, Q., Gustafson, W. I., Fast, J. D., Wang, H., Easter, R. C., Morrison, H., Lee, Y. N., Chapman, E. G., Spak, S. N., and Mena-Carrasco, M. A.: Assessing regional scale predictions of aerosols, marine stratocumulus, and their interactions during VOCALS-REx using WRFChem, Atmos. Chem. Phys., 11, 11951–11975, doi:10.5194/Acp-11-11951-2011, 2011.

Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), J. Geophys. Res.-Atmos., 113, D13204, doi:10.1029/2007jd008782, 2008.