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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Development and impact of hooks of large droplet concentration on remote southeast Pacific stratocumulus" *by* R. C. George et al.

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Responses to Review 2:

Reviewer: The article by George et al (2013) talks about "hooks" structures produced in SEP stratocumulus performing a comprehensive study using different sources of information (in situ observations, satellite data, trajectories and 3D fully interactive model) explaining its sources, conditions when is produced, characteristics and impacts. It's very well written, very clear and thorough and makes great use of the available literature. These structures have been mentioned before, but a study like this one was necessary to more deeply explore this phenomenon. I support publication after some changes. I feel the paper would be further strengthened if an effort is made to improve model ability to reproduce the timing and extent of observed hooks by using recommendations found in the literature and by including further analysis of the properties of the aerosol plume that generates the hooks.

One of my main concerns is the partial inability of the model to represent the different observed hooks. The 1st hook has a 24 hour shift in the prediction, and for the other 2 the structures are not well represented. The authors should make the best of efforts to conïňĄgure the model to try to reproduce the events to the best extent that computational time and model limitations provide. Previous WRF-Chem modeling studies (Yang et al. and Saide et al.) both have used ïňĄner vertical resolution (64 and 72 layers respectively) with most of the layers in the ïňĄrst 1-3 km, which have been demonstrated to better reproduce the MBL dynamics. As the current study uses 27 layers, this could be a point of improvement. Another difference with the two previous studies is the use of a nested domain, where the previous ones were not nested and obtained boundary conditions directly from analysis. Having the analysis forcing away from the domain boundaries could generate inconsistencies as simulations are continuous and not re-initialized from analysis. As an important concern is the timing of the event, performance could improve if model meteorological initialization is made closer to the event (according to Saide et al (2012), only 3-4 days are needed for WRF to recover from biases in the global analysis). If the authors wanted to go a little further (and if its computationally feasible), they could include an additional 2-way nested 3rd domain centered in Central Chile extending to the south to resolve the complex ïňĆow that happens in the region that connects the point of emissions to the point where pollutants go over the ocean. An evaluation of improvement (or not) from adding additional levels of complexity should be made as it would be very useful for the community to know what's really needed to achieve different levels of accuracy.

Authors: The authors thank this reviewer for their support and thorough read of this work, as well as helpful suggestions.

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One point we feel is important to clarify is that this study was not intended to be a model evaluation study, and reproducing the hooks to a fine degree of precision was not the main aim of this work. Although several studies have shown good model representation of REx mean conditions in the SEP, no study has focused on the mechanisms driving particular events (Saide et al., 2012 did discuss mechanisms for a near-shore sulfate feature, but this was not the central focus of their paper). We spent two years developing the version of the model used here, and we made substantial effort to reduce the error between the model and observations.

We ran numerous simulations to find optimal results for both the REx mean and the hook cases. The results shown here represent the best of that effort and the model hooks produced are able to reproduce the MODIS cloud droplet concentrations of the strongest hooks fairly well, albeit with timing errors. We employed many sensitivity studies to improve the model representation, but most avenues resulted in additional problems and larger errors than the simulation shown here. We appreciate the suggested changes to make the model even better, but it is unclear which avenues would actually improve results. More importantly, although improving the model representation of the hook would of course strengthen the paper, it is highly unlikely it would change the results shown in this study.

It is indeed concerning that there are errors in the model representation of hooks, but given that most models struggle to even represent mean features in this region (e.g. Wyant et al, 2010 and upcoming second phase VOCA paper), we find it encouraging that the model can represent salient features of day-to-day variability. Many model studies tend to focus only on features that the model represents well, while this study was driven by a desire to understand the causes of observed hook features. This provides a stern test for the model. The second two hooks described in the paper are not well represented, but they are weaker and less extensive features. The model does reproduce other hooks well that are synoptically similar to Hook1, such as the hook that occurred November 16-17, just after the campaign ended. We limited our exploration to the REx period so that even if the model does not reproduce observations well, the observations can provide context.

Although the vertical resolution in our simulations is coarse compared to other WRF-Chem studies, the physical PBL parameterization (Bretherton-Park) we use is designed specifically to simulate stratocumulus well without needing many vertical levels. As such, our ability to simulate cloud properties is no worse than the WRF-Chem simulations described in previous studies despite their much higher vertical resolution. Our experiments with higher vertical resolution led to major problems in transport and Nd. Simply changing the vertical resolution is not enough. This is because much experimentation needs to be done to tune the model to whatever resolution is used to accommodate different combinations of physical parameterizations better suited to a high vertical resolution simulation. Also, it is not necessarily the case that higher vertical resolution, even one heavily tuned, would fix the model errors.

We disagree that using a nested domain worsens the model accuracy. The NCEP analysis used for boundary conditions is known to have errors in this region, is not necessarily better than the WRF-Chem dynamics, and is available at much lower horizontal resolution. Similarly, initializing the run closer to the hook time period would not necessarily improve results. As a check we compared wind fields from model output to NCEP reanalysis in various locations and the accuracy of the model did not degrade over time or worsen far away from the initial conditions, thus we do not think decreasing the spin-up time would improve the timing of the hook event. Additionally, we use a nested domain and a long spin up time to increase the time and space between MOZART chemical influence and the model values. Decreasing the spinup time would leave residual chemical influence from MOZART in the SEP, which has different mean patterns and magnitudes of SO2, sulfate etc. than the mean WRF-Chem conditions after sufficient spinup. Saide et al (2012) tuned the FT SO2 values to provide a better match with VOCALS observations likely because the aerosol boundary conditions are not good.

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We agree a simulation that focuses on Chile that could provide information to the SEP through a 2 way nest would be incredibly powerful, but we are indeed limited in computational resources and time.

Reviewer: The discussion of albedo and aerosol sources would beneïňĄt from including describing aerosol single-scattering albedo in the hooks, in-situ observed and modeled. Are the source aerosols for hooks in any way different from those in other parts of the region? Do they have different mass or number distributions or composition? Do they undergo changes during hook development that differ from typical aerosol aging? Are aerosol:cloud effects in any way accelerated in the hooks?

Author: While the single scattering albedo would be a very interesting quantity to compare, we feel this is beyond the scope of this work and that this paper is already quite extensive in length and analysis. We intended to focus on basic features of the aerosols, transport and meteorology that influences hooks rather than expanding upon many details in any one area.

The difference between source aerosols for the hook and aerosols in other parts of the region can be seen clearly in the FT properties following the hook Figure 8. The first half of the time period shown can be contrasted to the second half, when the FT source has been depleted. Differences in the size and composition of FT aerosol are shown in Figure 8e,f and described in the text. The FT aerosol mass is not noticeably different, but the hook source aerosols are smaller than aerosols in other regions. Because of this there is not a notable increase in FT sulfate mass associated with the hook source although the number concentration is high. The source can also be noted by the SO2 concentrations, which are higher in the hook source region than other parts of the region. We note in the text that following the FT source aerosols with 3D trajectories, the aerosol properties do not change much (besides evidence of coagulation affecting Aitken mode particle numbers). What is unique about hooks is that polluted aerosol concentrations reach the remote ocean, but otherwise we found no evidence of aerosol processes being any different than what happens closer to the coast on a

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regular basis, so we do not focus on this type of comparison. Establishing what the 'typical' aerosol aging is to compare the hook aerosols with is nontrivial. Do you mean aging of aerosols from natural sources or aging of anthropogenic aerosols that do not travel to the remote ocean? We note the aerosol-cloud effects by looking at changes in liquid water path with and without anthropogenic emissions, and in the REx mean and show the enhancement associated with the hook (Fig. 8d). The difference between Anth and NoAnth Nd (Fig. 8a) is also indicative of the aerosol-cloud impacts. .

Reviewer: Many aspects of the hooks described in this article were already brieïňĆy described and explained in Section 3.4 and Figs. 8 and 10 of Saide et al (2012) and in Spak et al., (2010) (e.g. attribution to anthropogenic central Chile sources, entrainment from free troposphere, conversion to SO4 once it reaches the MBL, DMS contribution, location of the subtropical high as source of the off shore ïňĆow, etc). Please reference accordingly. Of course a better and more thorough explanation, demonstration and description of these structures was needed, and the one that this article provides does an excellent job.

Authors: Thank you for pointing this out and we apologize for the unintentional oversight. We have now added more references to the Saide paper as well as the Spak CLIVAR Exchanges article. It is interesting to compare the feature discussed in Saide et al. (2012) to the mechanisms associated with hooks. The Saide et al. (2012) feature shown in their Figure 8 does not extend into the remote ocean, and the sulfate mass rather than number is identified. But, the importance of the continental source, transport in the FT, entrainment and conversion to SO4 are similar.

4.2.2 Reviewer: "Hook impact on cloud properties". Another interesting cloud property to track over the hook trajectory would be cloud base and top height with and without Anth emiss, where you should expect deeper cloud when the hook is present (Pincus and Baker, 1994).

Authors:We do show that LWP is larger in the polluted clouds associated with the hook.

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This larger LWP is likely driven by thicker clouds. However, cloud thickness, or cloud base and cloud height are themselves difficult model metrics to make sense of because of the coarse model resolution. Perhaps one of the other groups who have working WRF-Chem simulations at high vertical resolution could look at this.

4.3. Reviewer: Impact of hook on albedo (and other sections). A big inïňĆuence is found by DMS. Previous studies (Yang et al and Saide et al articles) have found overestimation of DMS against VOCALS observations. I think it would be good to include this comparison for this model coniïňĄguration to better interpret the results, since if this model is also overestimating observations, then model DMS effects would be ampliïňĄed compared to reality. Fig 9. The no DMS runs show narrower hooks, more similar to observation. Maybe because DMS is overestimated so is less realistic?

Authors: Other studies suggested the surface flux was overestimated, and we use the same values and parameterizations. Our DMS is also overestimated very similarly, but this is not shown because, as the reviewer points out, this is already a published finding that this happens with WRF-Chem in this region. That the model DMS impacts may be amplified compared to reality is a good point and we have now added text to that effect: The end of section 4.3 now has: “However, DMS is overestimated in the model compared to observations (Yang et al., 2011), so the impact of DMS on results is likely also overestimed.”

It is a great observation that adding DMS to the simulation actually makes the hook Nd comparison to observations worse. However, without DMS, the 20ïĆřS REx mean state SO_2 and CCN concentrations in the MBL are severely underestimated compared to VOCALS flights, so finding the optimal representation is more complex than just removing DMS. This may be due to a deficit in FT CCN from in the remote ocean SO_2 boundary conditions (that Saide et al, 2012 adjust in their model), so the excess in DMS may be compensating for the weakened FT source.

Added to section 3.2: “The sulfate mass bias is doubled without the addition of DMS to

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the chemical mechanism."

4.6. Reviewer: Sources of hook aerosols. It is mentioned here and in previous sections that when some sources are turned off this changes the location of the hook. Can you elaborate why this happens? Is it the large scale meteorology that changes because of the aerosol interactions? Or is related to having different plumes from different sources that when overlapped look like 1 single hook but when some are removed they just shrink and generate the shift impression?

Authors: There are several reasons why this may occur. The last statement you make is one possibility and is the one we gave in the text, that there may be contributions from other sources besides the identified pathway from the Santiago region. Although it is possible that there are aerosol-driven changes in the large scale meteorology, there are also meteorological differences between any two ensemble simulations due to uncertainty in the initial conditions. Indeed, we found that these can be so large that we have devoted an entire paper to this topic that is currently in preparation. The transport mechanism is very dependent of the meteorological conditions and it is possible that slightly changes in meteorology just due to running a new simulation may contribute to the location and strength of the aerosol transport.

Page 2494, Line 11. Reviewer: As said in the conclusion, I think is better to say "lower free troposphere" to avoid confusions of long-range transport.

Authors: Thank you, changed.

3.1 model conïňĄguration. Reviewer: Please state if wet deposition is used or not.

Authors: Thank you, we have now mentioned wet deposition.

Fig 1. Reviewr: Add tick marks of lat/lon to a) and d) as well, not only to c):

Authors: The dashed lines coincide with the markings of c) and adding numbers to all panels clutters the figure. The same domain is shown for all panels. There are several other figures like this in the paper with one panel labeling the lat/lon lines.

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Fig 1. Reviewer: 30-40S 90-100W has typical stratocumulus?

Authors: 30-40âĄřS, 90-100âĄřW is dominated by stratocumulus in a decoupled boundary layer (see e.g. Warren cloud atlas). Low cloud cover exceeds 60%, three quarters of which is stratocumulus or stratus during SON. We use it because it represents a very clean region of stratocumulus that can provide an estimate of Nd free of continental South American influence. Using a different region with unpolluted Nd does not change this figure noticeably.

Page 2521, line 9. Reviewer: Correct "al.edo"

Authors: Thank you, fixed.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 2493, 2013.