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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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General Comments::

This is an interesting and useful model study of the impact of pollution aerosol transported over the SEP during the VOCALS experiment and the characterization of model aerosol entrainment in modifying SEP cloud features. It is well suited to ACP and will be of interest to the atmospheric science and radiation community.

This paper shows that WRFchem can capture the nature and transport of some of the aerosol layers observed above cloud during VOCALS including the influence of the Santiago pollution on cloud structure (hooks) and CCN. Important resulting influ-

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ences of this FT pollution are estimated for droplet number, cloud albedo and cloud fraction. The paper also points out discrepancies found in the model when used for this objective for select cases. However, direct observations during VOCALS revealed other examples of FT pollution transport and cloud interactions than those examined in these select cases [eg. Combustion Aerosol, Entrainment and Clouds in the VOCALS RegionCLIVAR Exchanges, v15, 2, 2010, Clarke et al.]. Moreover, the model did not yield consistent results for all of the 3 selected “hook” cases, so the generality of the conclusions regarding these aerosol/cloud interactions is not clear. The paper could be improved through a more detailed discussion/assessment of the model uncertainties and more complete investigation of the nature of identified disagreements. This could also be furthered by taking greater advantage of the in-situ aircraft data available.

Although most of the approach and the modeling is communicated well, there are a number of places the presentation can be improved. Some are simple changes in sentence structure but others are more substantive. Generally, more quantitative statements could be employed throughout and greater discussion of uncertainties and their impacts upon conclusions. Specific issues/comments are outlined below. Although I like the thrust of the paper and I support publication, I feel that most of these comments need to be addressed before the paper is accepted for ACP.

Specific Comments – using the page and line numbers in ACPD paper follow:

L13 “..the model suggests that high concentrations of ...” Because pollution aerosol with high number but low mass (compared to the MBL) were regularly observed above cloud during VOCALS it is not informative to say the WRFchem model “suggests” their presence. It would be more relevant to say something like “...the model captures the high concentrations of pollution aerosol observed above cloud during VOCALS.”

L16 “...originate mainly from a pulse of offshore flow that transports Santiago...” This may occur frequently and be true for the limited cases discussed in this paper but pollution was often observed above cloud for more than just the cases discussed in

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this paper and sometimes originated from very different sources. Rephrase.

P2498 L19 What justification exists for this choice of the %S of 0.5? This is about twice the %S for VOCALS stratus as evident in the observed dry-size aerosol Hopple minimum. This choice will lead to larger absolute model numbers and a different sensitivity to diurnal fluctuations in %S (discussed later) than would normally occur. Given this choice has a direct impact in assessing CCN, I feel that authors need to clearly show model size distributions explicitly and indicate where the activation threshold for 0.5 %S (and 0.25 %S) would be expected. Were any runs done at %S less than 0.5 and, if so, how different were they? Given that this choice is fundamental to the results in the paper, this needs to be discussed accordingly.

P2499 L4 “..midday retrievals reasonably match. . .” If this result does not merit a figure then a more quantitative assessment of “reasonably” needs to be specified in the text. How well do they match and in what way?

L21 “although accumulated trajectory errors could potentially account. . . .” This statement is premature and not an observation. A discussion of trajectory performance and associated errors is needed before it can be interpreted.

P2500 L5 It is stated— “The two most western trajectories do not quite intersect the coast, the clearly connected nature of the observed hook suggest these locations have similar sources.” However, if one looks at tracers like CO, black carbon and organics etc, these indicators are lower for these two outermost longitudes than the others, suggesting there is a real difference.

L18 The authors say “. . . fire related aerosol are unlikely to cause the heavily polluted Nd. . .”. However, the above mentioned trajectories further east did have these high Nd concentrations above cloud and had enhanced organic and black carbon concentrations. Most pollution has a combustion source (fire) as evident in the general relation observed aerosol BC and CO during VOCALS.

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L24 I do not think the data mentioned demonstrates a “. . .lack of an FT source” or perhaps better stated “lack of an FT supply of pollution derived CCN”. This kind of FT/MBL comparison of CCN and potential FT contributions requires great care and cannot be explored without a discussion of uncertainties, winds and wind shear, subsidence and timing. Model layers and model inversion heights have to match reality too. Observed above-cloud layers were often patchy, variable and often thin. Infrequent CCN measurements in the FT also limit the assessment of these aerosol fields in the FT. Also, for an entrainment rate of say 0.4 cm/s would take 10 hours to entrain a 100m layer. Hence, what was above the inversion during the past 10’s of hours is more important than what was observed above cloud but yet to be entrained. Given the wind directional shear often present across the inversion, it is the upwind time-integrated influence of the FT layers on the MBL that controls the resulting MBL concentration. This “exposure” is something the model can be interrogated to provide. On this flight, enhanced pollution aerosol at CCN sizes in the 200-300 /cm³ were measured above cloud along 20S between 80W and 77W (CCN measurements were limited).

In general, more effort to compare with other in-situ data should be made and some accounting for possible uncertainties should be discussed before assuming the above cloud data at west end of flight path implies the MBL there had not been influenced by prior entrainment. Also, I would avoid using “an FT source” as it is not a source of the aerosol in the same sense as say Santiago. Maybe an FT aerosol layer of enhanced CCN - or something like that.

P2501 L10 What is model resolution in the vertical just above the inversion where transport and entrainment needs to be resolved in this paper and what does this imply for uncertainties? For example a 50m layer resolution could be associated with an uncertainty of about 5 hours for an entrainment rate of say 0.4 cm/s. L19 factor of 100? Warrants a little more information. Seems drastic, is there some problem here, maybe model removal is way off?

P2502 L18 How consistent are these model assumptions with the DMS flux assess-

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ment made by Yang et al. (Atmos. Chem. Phys., 11, 5079–5097, 2011) for the VOCALS region at this time? L23 IMPORTANT – As mentioned earlier, size distributions are critical to modeling CCN and they can be compared directly with measurements. The WRFchem model distributions mentioned need to be shown in a figure along with the sizes expected to be activated at the assumed 0.5%S. This is particularly true as later diurnal fluctuation in %S and activation are discussed in some detail. This figure can be then referenced directly in the context of sensitivity of the model to diurnal fluctuations in %S. .

P2503 L16 “. . .magnitude underestimated by 10-30%.” This seems odd as the %S used in the model is higher than evident from the MBL size distributions (Hoppel minimum). Later, when comparisons to other measurements and albedo etc. is made, reference to this uncertainty should be included in interpretation. L18 etc. Comparisons to Saide et al. are mentioned but not for DMS. This is argued to be overestimated by Saide et al. in WRFchem. More generally, this section merits a more complete discussion of similarities and differences in models and performance (and with observations) rather than alluding to “reasonably reproduces REX mean conditions”. It is not clear what that means. Is this “reasonable” in the MBL or in the FT or both? How reasonable is it? More importantly, some discussion of how the differences (or uncertainties) in absolute or relative quantities might impact (or not) the assessment of model CCN in the FT or MBL should be included and/or to what extent these differ from the Saide et al. WRF performance assessment for various key model parameters. How well (quantitatively) does this WRFchem get the inversion height and cloud top correct as this will affect reliability of timing influences for advected FT aerosol interactions with clouds compared to reality. What are implications of any key differences found?

P2504 L7 -10 This shift of several degrees in model/observed behavior merits a greater discussion in the context of uncertainties. Horizontal and vertical uncertainties are coupled in a 3-D subsiding aerosol field and need more discussion. For example, what does horizontal displacement error imply in terms of altitude uncertainties for when

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or where a pollution layer will encounter the inversion and start to have an effect on CCN? This discussion is important for this paper and the strategy for choosing when or whether or over what scales the in-situ measurements or satellite retrievals can be compared to a model output etc.

P2506 L20-25 Here again reference needs to be made to assumed size distributions and the CCN activated under the influence of different %S. Are these model distribution behaviors consistent observations? Similarly, is sensitivity to TKE in the model consistent with that expected for observed sizes and variability actual typical cloud supersaturation (eg. width of Hoppel minimum).

P2508 L1 “. . .only a small increase in MBL aerosol mass occurs as the FT aerosols are entrained. . .”. IMPORTANT –If this is really what the model shows, I worry about how the model is handling mass conservation. Entrainment of air with lower aerosol mass concentrations can only dilute (lower) the MBL mass concentration and not increase it. Please explain.

L2 “Thus the FT is comprised of numerous small aerosol. . .”. This is not a finding as it was observed directly during VOCALS in the airborne aerosol size measurements. A more relevant concern is how well model and measured number distributions compare with model estimates for different air mass/aerosol types in the FT? Some evidence that model is getting aerosol sizes correct is needed.

P2509 L9-12 Please clarify discussion. In a typically divergent region with constant or increasing mean wind (typical) and with cloud base constant but with cloud top growing, it seems like significant entrainment of FT air is required for mass conservation. Or am I misreading the argument.

P2512 L8-15 Here is another example of model uncertainty that will affect comparisons with measurements or satellite retrievals near locations of expected entrainment. It is good that such examples are included but it would be more useful to have uncertainties collected and discussed clearly in one section (such as pg. 2503) that then propagates

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them into uncertainties in some of the findings reported.

P2514 L19 Is this claimed rapid coagulation of Aitken mode number over 12 hr. and subsequent stabilization consistent with expected coagulation in the FT for these sizes and concentrations? This appears to be too fast for a non-cloud environment. How did model size distributions change?

P2516 L1-10 There are certainly other, perhaps less pronounced, hooklike features present during VOCALS that also have pollution above clouds than the ones selected in this paper. Line 26 it indicates the model is not correctly simulating offshore advection. This implies that many examples or features may not be seen in the model or captured correctly for different sources. L17 “The transition ...”. Is this transition in the MBL of FT? L21 “...lack of FT support...” Again, what is above MBL for any measurement profiles does not mean that this is what has been above it and entraining into it for the past day.

P2517 L13-19 The authors should provide more complete discussion/clarification/evaluation of the merits of their speculations on why WRFchem does not consistently produce hooks. Text Reason 1) As removal of CCN takes place in the MBL, if entrainment is the source then these should show up in the cloudiness changes before they can be removed by precip. Or are authors suggesting entrainment is suppressed? Text Reason 2) nucleation requires time and a precursor source strong enough to grow them to CCN sizes. Again, a comparison of model and measured size distributions and associated CCN in the plume seems warranted before speculation. Text Reason 3) Errors in altitude, latitude and magnitude of model flow were mentioned at several points in the text. As noted previously, a separate section that provides a more quantitatively discussion of expected and observed uncertainties (eg. altitude) in the context of predicting CCN and hooks would be helpful. It is not constructive to speculate on these possibilities without following up. On Nov. 2, the in-situ measurements show pollution aerosol sizes at high concentrations in the MBL (not removed) out to beyond 80W. Given the extensive aerosol, cloud, chemistry,

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meteorological data etc. available on the C-130 one expects that model altitude fidelity and uncertainty could be better demonstrated/constrained and the likelihood of some of these reasons eliminated. Does the model get the inversion height correctly for these flights? etc..

Technical Corrections:

Title could be made more clear Perhaps something like –Large droplet concentrations in arcs of cloudiness over the remote southeast Pacific: Their origin and impact.

P2494 L3 “hook shaped arcs” is redundant (–like “round circles”). Why not use just arcs throughout as they are less ambiguous in meaning?

P2495 L18 change to “To provide pollution CCN that can sustain hooks. . .”

L23 change to “ LWP also increases as the hook evolves over time. . .”

P2496 L22 change “topographic transport restrictions” to “transport restricted by topography”

P2543 Nice figures (13,14) in principle but very hard to read numbers.

P2545 Not clear this Fig is needed.

P2546 Switching to SO₂ for 16c is confusing. At least show CCN (black) along with the SO₂.

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

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