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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Southeast Pacific atmospheric composition and variability sampled along 20 S during VOCALS-REx" *by* G. Allen et al.

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Response to Reviewer 2:

Please also see response to reviewer 1 and general comments

Reviewer 2 - General comments:

1/ Sampling diurnal variability: The reviewer is quite correct to note that diurnal variability may be a factor which could conceivably affect systematic biases in atmospheric composition, especially with respect to photochemically active species such as ozone and cloud-sensitive aerosol populations. The reviewer is also correct to note that the majority of VOCALS flights were conducted in the local morning hours. In addressing this potential source of bias, we should first make a distinction between this hypothesis

for the MBL and FT.

In the MBL, diurnal modulation of below-cloud composition, especially offshore would be expected to be small for the following reasons: 1/ Rapid mixing - surface heat fluxes are tied to a near-constant sea surface temperature and so efficient boundary layer mixing (which typically occurs over the order of hours) can be expected to be a constant day and night feature, especially with respect to the lifetimes of ozone and CO (days and weeks, respectively). 2/ Photochemistry –CO can be essentially considered to be inert over the diurnal cycle (lifetime of months in the troposphere). In the case of ozone, the Tropical Pacific marine boundary layer is well known to be subject to net removal of ozone due to both photochemical destruction in a low NOx environment and surface deposition (e.g. Johnson et al., 1990, Monks, 2000). The typical range of net ozone destruction rates calculated for marine NOx-limited environments, such as those typical of the SEP, e.g. during the SOAPEX-1 campaign (Table 5 of Monks et al., 2000) are 1 to 2 ppbv/day. Typical diurnal cycles in ozone in the remote MBL have been found to have a peak-to-peak amplitude of 1 to 2 ppb from both observations and a chemical model (Thompson and Lenschow, 1984). Given this general loss trend for MBL ozone superposed with an expected diurnal cycle of the order 1-2 pppv, we can conclude that any intra-day variation that we might be biased to by flying mostly in the local morning might be expected to be less than a few ppbv and therefore much less than the sampling variability typical for any given longitude as plotted in our Figure 4 (5 ppbv).

As a further test, we have compared trace gas and aerosol concentrations on flight B408 (a 20 south transect between 7 am and 1 pm local time on 26 Oct 2008) and B409 (a 20 South transect between 4 pm and 10:30 pm the following day) to examine systematic biases versus longitude. There was no evidence of such bias, insofar as the difference between longitudinally-gridded mean concentrations for each longitude bin were much less than the corresponding sampled standard deviation of measurements for both flights. In summary, this comment has shown us that diurnal variability is

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both expected and observed to be negligible versus other sources of observed natural variability – a useful result and one which we have now included in our discussion at the recommendation of the reviewer.

2/ Choice of spatial gridding: The reviewer suggests that a 2.5-degree gridding for our composition statistics is too coarse and that modellers would benefit from a finer resolution, perhaps up to 1-degree. This would certainly be optimal. However, we choose this lower resolution for 2 important reasons: 1) we are limited by our sampling frequency and to get robust statistically significant data, we are required to grid at 2.5 degrees; 2) This gridding is consistent with that used by Bretherton et al., 2010, and as our results are intended to be directly relevant to an investigation of effects on cloud bulk properties, which Bretherton et al., 2010 documents, then we prefer to retain this spacing so that direct comparisons can be readily made between atmospheric composition and observed cloud properties.

3/ Aerosol optical properties: As the reviewer notes, measurements of aerosol optical properties recorded during VOCALS do indeed warrant their own companion paper and those measurements will be discussed in forthcoming work by Clarke et al and others.

4/ Complex vertical interleaving of airmasses: This is an important conclusion and the reviewer is correct that it does warrant further figures. This is a result that was carefully examined by the team by plotting vertical profiles from all flight data. We feel that to include further plots documenting this from measurements in this paper is beyond the scope of presenting longitudinal statistics and that such analysis deserves its own paper documenting free tropospheric transport processes, which will be the subject of advanced forthcoming work by Clarke et al and Allen et al. For the purposes of this paper, we present back trajectories which do show this interleaving of airmasses and we now reference to these when discussing vertical interleaving.

5/ Linkage to VOCALS hypotheses: Thank you for this suggestion. We notw directly

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link to these hypotheses in our discussion and conclusions section.

Reviewer 2 - Specific comments:

Free tropospheric organic aerosol: We make airmass history conclusions based on back trajectory analysis, which consistently show that layers of enhanced organic aerosol in the free troposphere originate from the tropical western Pacific surface, uplifted and carried long range in the subtropical jet stream. Since the tropical western Pacific (Indonesia and northern Australia) are areas of wide biomass burning during October/November, we make the conclusions that the observed organics originating there are from a biomass burning source. For back trajectories in the MBL, we do not observe airmass history tracing back long range and instead observe long time periods of contact with the coast, where we expect local industrial, biogenic and agricultural processes to be the dominant source of organics observed in the MBL. We express this conclusion already but we have further modified our discussion to more succinctly state this. We have also included a new figure showing MODIS fire maps for October and November over the tropical western pacific to support our conclusion for long range transport of organics in the FT.

P.708, ln 9-10: The reviewer is correct to point out that we cannot attribute point sources for common (simultaneous) emissions of sulphate, ammonium and CO. Therefore we have amended our discussion to state that this observed correlated source is simply the coastal environment in general so as not to potentially mislead the reader that there may necessarily be individual point sources which emit all three tracers simultaneously.

Figure 8 – It is not possible to create a box and whiskers plot which conveys simultaneous information content about molar ratio and sulphate concentration at the same time. We would therefore prefer to leave this figure as is, as we believe it is useful to illustrate that gradients in molar ratio are not generally correlated with gradients in sulphate concentration.

Reviewer 2: Minor comments:

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P. 717, line 18: We have removed the redundant “rapidly” from this sentence and we are grateful for the more appropriate referenced provided which are now incorporated.

References used in this response:

Monks, P.S. et al., A seasonal comparison of ozone photochemistry in the remote marine boundary layer, Volume 34, Issue 16, 2000, Pages 2547-2561 Thompson and Lenschow, 1984

Johnson et al., Ozone in the marine boundary layer over the Pacific and Indian Oceans: latitudinal gradients and diurnal cycles, JGR, 1990

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 681, 2011.