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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.

Anonymous Referee #2

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Overview:

"Southeast Pacific atmospheric composition and variability sampled along 20° S during VOCALS-REx" presents an overview of zonal gradients in trace gas and aerosol properties observed by aircraft and marine platforms on a representative zonal transect of the Southeast Pacific. This article warrants publication in ACP, as it represents a long-term resource of great importance for several reasons:

1. Novel data: it provides the first in-situ multi-platform description of trace gases and aerosols over one of the most under-observed, poorly understood climatic zones in the world, and one of the most important for global climate.

2. One-stop shopping: it represents a canonical reference and starting point for future

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explorations of atmospheric chemistry in the region.

3. Key documentation: on a practical level, it's an important deliverable, the first-order direct observational results of one of three core aspects of a very large and resource-intensive field campaign.

The article generally succeeds in each of these capacities. The authors thoughtfully aggregate observations to develop a useful unified climatology from this extensive dataset, and provide more detailed metadata than the experiment's field catalog. The manuscript's primary limitations at this point are the choice to aggregate at a coarse spatial resolution, the limited aerosol optical information presented, and a conclusions section that summarizes results rather than framing them with regards to the VOCALS hypotheses that motivated the study. Also, one of the primary conclusions is not fully supported by the data presented. Minor revision is recommended.

General comments:

As the authors acknowledge, the Southeast Pacific region features very strong diurnal patterns in meteorology. However, very few flights sampled the afternoon and evening (16:00 UTC - 6:00 UTC), so the observations do not capture the known diurnal variability in many of the trace gas, aerosol, and meteorological properties observed. Moreover, since the flight schedule correlated distance from the coast with time of day, the observed longitudinal profiles could contain signals that reflect diurnal cycles (i.e. ozone photochemistry) as much as they do longitudinal gradients. These limitations should be acknowledged, and implications on findings considered. Are there observed time-of-day dependencies in any of the trace gas and aerosol features presented? How large are they, compared to the longitudinal gradients? Discussion of the ozone longitudinal gradient, in particular, should address this.

The choice to grid at 2.5° is a limitation for comparison with modeling and remote sensing, and should be explained or improved upon. The article employs trajectories on a 1.125° grid, and since spatial features must be resolved in models to capture the

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ocean-land-atmosphere coupling in the region, many of the dynamical modeling studies for which this article will inform will be conducted at higher than the 2.5° resolution used in legacy global weather, climate, and chemical transport models. Likewise, most remote sensing products are provided at resolution of ∼0.1°. This choice of binning is also a clear limitation in the results, as few of the longitudinal gradients are smooth and fully explained by distance from the coast, and the variability might be better captured at higher resolution. Since the article goes on to further aggregate to three "zones," the seven longitudinal bins seems especially limited. It's understandable if the choice was made to ensure large enough sample sizes, but this should be quantified: given the number of research flights in each longitude bin, is the statistical significance of the variability observed in the 2.5° bins different from 1° bins or 0.5° bins?

The presentation of aerosol optical properties is limited to one figure on wet nephelometer f(RH), although these platforms did observe many other aerosol properties. Typical aerosol optical properties (AOD, SSA) are not presented, and likely deserve their own companion paper. These properties may have even sharper gradients than the trace gas and aerosol conentrations. In that case, ensuring that the gridding convention to be used in follow-up papers adequately resolves observed gradients becomes even more important.

What evidence is presented to support the observation of "complex vertical interleaving of airmasses" in the free troposphere, as mentioned in the abstract and at three places in the article? This is an important conclusion, and if it represents an important feature of the data set, then it warrants additional figures, tables, and discussion.

The article provides evidence that may support several of the primary VOCALS hypotheses listed in Table 1 of the article by Wood et al. in the same special issue of ACP. A direct consideration of the results in light of these hypotheses may be a useful addition to the conclusions section.

Specific comments:

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What evidence is there that "local and regional agriculture and biology" contribute substantially to observed organic carbon concentrations? If, as suggested, organic carbon in the free troposphere is due to biomass burning, what differences support a different attribution for coastal MBL organic carbon?

page 708, line 9-10: "This perhaps suggests a common source for CO, sulphate and ammonium aerosol in the coastal region." That source IS the coastal region. Motor vehicles are the only emissions sector that emits all of these species, but emissions inventories suggest that this is not likely a strong enough source to dominate the signal. Far more likely is that CO, SO2, and NH3 are all emitted by different sources along the Pacific coast, in the narrow strip between the ocean and Andes. That's the extent of attribution the 1.125° trajectory analysis supports, and evidence would need to be presented for any more specific conclusion.

Figure 8 might be more informative as box-and-whiskers plots binned by longitude as in the other concentration-longitude figures.

Minor comments:

page 717, line 18: rephrase "characterised dynamically by both rapid descent of airmasses transported rapidly"

More representative citations for background tropospheric ozone, with less emphasis on episodic wintertime stratospheric-troposphere ozone exchange:

Fiore, A. (2002), Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes, J. Geophys. Res., 107, 4275.

Fiore, A. (2003), Variability in surface ozone background over the United States: Implications for air quality policy, J.. Geophys Res., 108, 4787.

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