Author Comments
May 6, 2011

We greatly appreciate the instructive and detailed *comments from Dr. P. Matrai*. Please see our replies below.

A general comment: Please make sure that every abbreviation is spelled out the first time it is used throughout the text and including the figure legends. Also, please check that all refs are there - the very first one I looked for was not on the list-; I did not cross check the others. Clearly, some sections were moved around and such details not re-checked, suggesting some sloppiness.
I recommend publication, once comments are attended to.

We thank the reviewer for checking so closely and have corrected for such errors.

Specific comments: P2875L12-15 indicate that this is for the offshore region; L15-16, inshore and/or offshore?

L15-16 refers to the offshore region.

L25 update the K&A'99 estimate with Lana et al. GLOBAL BIOGEOCHEMICAL CYCLES, VOL. 25, GB1004, doi:10.1029/2010GB003850, 2011, of 28.1 (17.6–34.4) TgS/yr; does not change the point.

Thanks for the reference.

P2876L5 spell out MBL

Suggestion accepted.

L28 do you mean that all CCN, as a size fraction, represent a large fraction of the TOTAL aerosol number?

Yes.

P2877L8 nss SO4 in both cases?

Yes. Unless preceded by 'seasalt', SO_4^{2-} in this manuscript indicates non-seasalt sulfate.

L15 spell out FT

Suggestion accepted.

L25 from or towards the SE?

From.

P2878L10 November

Corrected.

L11 add latitude of Arica

Suggestion accepted.

P2879L16 AMS sulfate concentration? data? missing word

Concentration.

L18 end of sentence, please insert (data not shown)

P2881L8 Benedict et al 2011 missing from bibliography; any others?

Reference added.

L18 replace 'to' with 'into' ... 2 regions P2882L4 shows

Corrected.

L6 SO4 concentration?

Yes.

in fig 2, where is the marker size scale?

Marker size range is indicated in the figure legend.

L8, is RF14 later in the season? any flight characteristic that makes this RF14 different in any way? Just because there was more pollution doesn't make it any less real. There is no later discussion of such events

RF14 was the last flight of the campaign, and was mostly around 80W, 20~22S. Away from the coast, the SO_2:CO ratio was 2~4 times higher on this flight than on any other flight. Back trajectory analysis also suggests a different air mass origin for this flight compared to the typical remote MBL, hence our exclusion in the manuscript.

L13 insert 'on' before 'aircraft'

Suggestion accepted.

L18-22 this section on seawater DMS needs references (Yang et al 2009? Hind et al

2011? Other?) and it would benefit from a sentence that indicates this is a brief summary of seawater concentration and fluxes ... OR add
C1958references for each of the next statements in this paragraph

Hind et al., 2011 is an appropriate reference here, thanks.

L26 fluxes; P2883L5 SO2 and SO4 concentrations;

Suggestions accepted.

L25 what proportion of the sampling time, hence data, were cloud free or POC?

The shipboard W-band radar measured an average cloud fraction of ~70% in the offshore region. The ship only encountered POCs on a couple of occasions. More observations of POCs were made on the C-130 aircraft per flight plans.

L27 'new particle nucleation' from what? DMS? what is/are possible source(s) for the 30nm particles seen above the cloud layer? Is the difference bw 64ppbv and 74ppbv that huge for only one to be high and related to long distance transport?

Under favorable conditions (low aerosol surface area due to precipitation scavenging, etc), nucleation could occur from either naturally derived sulfur (i.e. DMS) or pollution sulfur. This will be discussed in detail in an upcoming Clarke et al. paper. The 30 nm mode is likely from aerosols that were formed fairly recently and had not grown for very long, whereas the 80 nm mode indicates aged aerosols that had grown for much longer during transport. A CO concentration of ~60 ppbv is close to the expected background marine level for the Southern Hemisphere. Concentrations higher than that usually indicate combustion pollution.

P2885L27 very interesting; L29 evokes or invokes?

Invoke is more appropriate. Thanks.

P2886L2 would be, rather than was- this is a speculation, as indicated later

Suggestion accepted.

L21, L23 spell out LHS and RHS, respectively

Suggestion accepted.

P2889L1-3 were the resulting SO2 concentrations higher because of higher oxidation rates or higher DMS concentration or flux during the day time, ie, faster or more substrate?

As shown in Fig. 13, SO_2 concentrations were higher during the day because of the

greater DMS oxidation, and hence SO_2 production rate (due to the OH profile).

Is the diel DMS cycle shown in Yang et al 2009; certainly not in this manuscript? From later in the text, it appears that a diel cycle of OH is seen with mid-day max and a range of gamma values. Did observed fluxes (independent of wind speeds??) or atmospheric concentrations show a diel cycle?

DMS had a pronounced diel cycle, which is shown in Yang et al. 2009; a version of that will be added to the revision of this manuscript (see first author response). DMS sea-to-air flux did not vary substantially on a diel cycle; flux might be slightly higher during the day due to the higher wind speed.

P2890L9 if MSA WERE (not was);

L11 WERE formed from DMS

L12 please replace 'implies' with ' would imply'

Suggestions accepted.

L17 using the same range for gamma?

Yes.

P2891L12 R? is this the same R as defined later? a different one? make sure that all symbols are defined the first time they are used in the text;

R is the gas constant through out the manuscript.

P2894L2 please indicate explicitly why it would be 'unrealistic'; L should not exceed P, given that advection is negligible (?) as an alternate source OR unless there is another unaccounted source (?). Even to summarize the arguments made earlier in the discussion;

In this case, we think L should not exceed P.

L11-12 whose data? First time these MODIS data are mentioned

The MODIS images from this region are described in Wood et al. 2011.

L22 what is meant by "qualitatively" in this sentence?

We meant that because in both cases SO_2 is limiting, the relative depletion in H_2O_2 due to heterogeneous chemistry is much smaller than the depletion in SO_2.

P2897L29 and is the magnitude of this factor (0.43) comparable to something? too high?

too low? what would have happened if a closed system had been chosen? even if unrealistic? P2898L21 represents P2902L9 ten?

While a closed system would have been more realistic, implementing it over a diel cycle is difficult given the complicated cloud dynamics. The calculation on page 2893 suggests that over an hour, using the closed system gives a SO_2 depletion rate that is about 80% of the open system rate. Moreover, the SO_2 depletion rate calculation utilizes measured SO_2 concentration at cloud height (a first order dependence). However, with respect to MBL SO_2 budget, the portion of the depletion rate due to SO_2 entrained into the MBL from the FT should not be included, hence a need to further reduce the calculated depletion rate.

P2904L18 allowed us TO estimate

Suggestion accepted.

Table 1: format last section like others underlining subtitles

Fig 3: pollution, as indicated by xx and yy, or see text for details...

Suggestions accepted.

Fig 6: is the difference between day and night for inversion height statistically significant?

Yes. Multiple measurements (radiosondes, aircraft profiles, W-band radar) indicate a greater inversion height at night compared to during the day on average.

Fig 13 how does the implied DMS compare to the measured DMS? Or was it prescribed? Then say so. Same as indicated above.

Implied DMS matches measured DMS very well (again, see first author response).