We thank the reviewer for carefully reading our manuscript and for giving valuable comments. In this reply, we address all of the reviewer's comments. The detailed responses are given below (reviewer's comments are in italics and smaller font size).

The paper by Sinreich et al. describes measurements of glyoxal (CHOCHO) in the remote Eastern Pacific. Since this study supports recent studies on satellite observations which have shown elevated levels of CHOCHO above water bodies, it is of high relevance for several research topics like VOC oxidation, SOA or CTM. In general, glyoxal observations are quite sparse and this data set has the potential to fill some of the gaps in our knowledge. The study comprises three major sections: 1) the description of the instrument itself, the analysis and the data filtering (sections 2 to 6, I would prefer to have subsections 2.1 to 2.5). 2) The description of the retrieved data (section 7). 3) The detailed discussion of possible sources for the observed glyoxal amounts (section 8).

We changed the numbering as suggested by the reviewer into the main chapters Introduction, Measurements, Results, Discussion.

In general the paper is an interesting and scientific important piece of work and merits publication in ACP. The introduction as well as the discussion section are well written and reflect (with some minor exceptions) the current knowledge. However, I have some serious concerns about the presentation of the data and its interpretation. These concerns are explained below.
First of all, I agree with referee #1 that the authors should avoid to claim that they did direct observations of glyoxal. They are describing remote-sensing data in the UV/Vis and that's never direct.

We had already changed the title to 'Ship-based Detection …' for the ACPD publication and removed 'direct' from the conclusions as suggested by reviewer #1.

Furthermore one should avoid terms like "inherently calibrated" or "unequivocally identifies" (p15079) to characterise the DOAS technique. If this would be the case the interpretation of DOAS data would be much easier. In particular for small absorbers there is always the risk that radiation processes not proper accounted for, induce false signals.

We followed the suggestion of reviewer #1 and removed 'unequivocally' and changed 'inherently' to 'intrinsically'. Notably, a fundamental advantage of DOAS is that it alerts of 'false signals' by residual structures that remain if not all the absorbers are accurately accounted for in the retrieval. Within the accuracy claimed we can rule out that 'false signals' affect our retrievals.

The spectral "proof" for the detection is given in Figure1. The authors have calculated a RMS of the residual of about 3.7e-4 which is 4 times smaller than the signal of glyoxal shown here. However, I prefer to define the residual as peak-to-peak and in this case this would lead to the same number as the glyoxal signal (1.5e-3). In fact only the main band of the glyoxal absorption cross section close to 455 nm is visible in the residual. In general the residual seems to have several systematic features. How does the residual looks like, when more than 30s are integrated? Just to clarify: I'm not challenging the detection of glyoxal in general but I'm really sceptical about the very optimistic assumptions in section 7 leading finally to 30% total error for the calculated VMR including the fitting error!

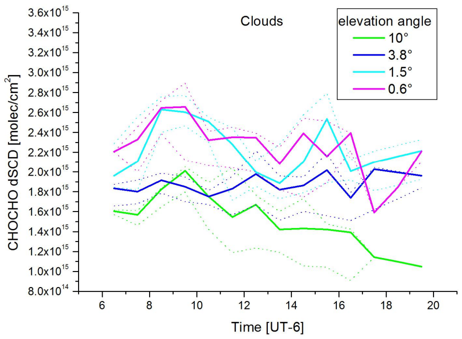
The RMS is a suitable criterion for evaluating DOAS retrievals (Stutz and Platt, 1996). We agree that the residual might not be fully statistical. Thus, we applied a 2σ detection limit for the estimate of the VMR. Sensitivity studies during the analysis varying settings like polynomial, offset and wavelength range showed only small variations in the retrieved dSCD (typically <10%), which substantiates the robustness of the retrieval. Furthermore the spectral fit shown in the manuscript is not smoothed at all, and it is often observable also in other retrievals that the absorption band at 440nm is not retrievable in the fit. Unfortunately we did not integrate longer than 30s for a measurement, and scanned 16 angles towards both sides of the ship sequentially. This gives rise to significant time differences between two observations at the same elevation angle. Adding up spectra measured at the same elevation angle did not lead to significant further reductions in the residuals due to the long time difference between two measurements of the same elevation angle.

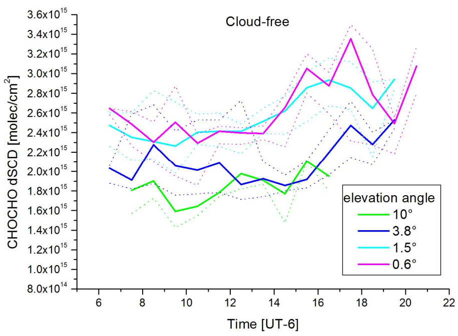
Figure 2: Here DSCDs for CHOCHO (2a) and O4 (2b) are shown for one single week only, from a total of ten weeks. What is the reason that only this week is shown? If there is a good reason, it would be helpful for the reader if the authors could stretch the x-axis in order to be able to identify more details. The diurnal variation of the glyoxal looks a bit strange. It is always u-shape for this week, independent of cloud coverage having highest values close to sunrise and sunset. This cannot be explained with changes in radiative transfer. In fact O4 shows a completely different behaviour. Could the authors comment on that?

Figure 2 shows an example week for the whole measurement period depicting CHOCHO being inside the MBL. This week was picked because the week was relatively cloud free, which substantiated the conclusion of the location of CHOCHO inside the MBL. We follow the suggestion of the reviewer to stretch the x-axis. Taking a closer look shows that only the data on 12/15 is u-shaped, whereas all other days show rather an increase in the dSCDs in the evening, which can also be observed in the O_4 dSCDs. We followed the reviewer's suggestion to generate median profiles (see below), which substantiates the lack of a u-shape. We have not included the below graphs in the paper, as we feel they are captured well in the example period chosen shown in Figure 2.

Is it possible to calculate mean diurnal variations for cloud/non-cloud scenarios for the whole data set? This information would be also helpful for the discussion of possible sources of the glyoxal.

We created median diurnal variation plots for cloudy and cloud-free conditions of the TAO leg, which comprises mainly the cruise through the remote Pacific Ocean. The medians are based on 1 hour periods at a standardized time stamp (in respect to 90°W) and include all values above a 2σ detection limit. The two plots show elevation angles of 0.6°, 1.5°, 3.8° and 10° (solid lines). The signal-to-noise ratio of the 25° elevation angle values is too small to be shown here. The dashed lines represent the 25 and 75 percentiles of the values of the 1 hour periods.





Values measured with an elevation angle of 25° should be a good proxy for the total vertical column of the trace gas, independent of the aerosol content in the atmosphere (dAMF ~ 1). This has been shown by several other studies. For O4 this is the case (Figure 2b), but for CHOCHO (Fig 2a) in particular those values measured under clear sky conditions, are very close to zero and not in the range of a few 10e14 molec/cm2 as expected. Again, this is difficult to explain with changes in the light path. Comments?

We agree with the reviewer that, in general, the 25° dSCD values provide a good proxy for vertical columns. However, in this study the signal-to-noise ratio of the 25° values is too small for a useful quantification of the vertical column. For this reason, we chose the lower 1.5° elevation angle, which inhibits a leverage effect in terms of the signal-to-noise ratio, for retrieving VMR values. From the VMR we calculated a mean vertical column in the MBL. We have slightly modified language in the manuscript to reflect the uncertainty of the 25° dSCD values.
Recent measurements of boundary layer heights in the study area (Rahn and Garreaud, 2010) and also earlier observations in the equatorial Pacific Ocean (Kley et al., 1996) find a slightly

higher MBL height (1-2km) compared to the 1km we had assumed in our inverse approach. Thus, we recalculated the VMRs for a mean height of 1.5 km resulting in 17% smaller VMR values (in average), and changed this throughout the manuscript.

Rahn, D. A. and Garreaud, R.: Marine boundary layer over the subtropical southeast Pacific during VOCALS-REx – Part 1: Mean structure and diurnal cycle, Atmos. Chem. Phys., 10, 4491-4506, doi:10.5194/acp-10-4491-2010, 2010.

The authors explain in detail the overlapping of DSCDs for different (but small) elevation angles. They are right that this is most probably due to aerosol extinction. But this is well-known and discussed in detail by many other studies (before Volkamer 2009b). But there is another possible explanation: the profile shape. If the trace gas layer is located a bit higher up in the troposphere or even in the boundary layer it leads to an overturning point for the DSCD similar to the aerosol effect. For that reason most state of-the-art retrievals for MAXDOAS use as much information as possible (e.g. DSCDs for several elevation angles) to retrieve the trace gas information. It is not really clear for me, why the authors did not try to benefit from all their different observation modes.

We respectfully disagree with the reviewer's comment. A different profile shape in glyoxal as cause for the overlap at lower elevation angles seems to be very unlikely, since the overlapping dSCDs are seen in both CHOCHO and O_4 dSCDs. For O_4 the vertical profile shape is well known, with its bulk concentration in the lower troposphere. The sensitivity in the lower angles towards elevated layers is greatly reduced, and largely canceled out by our analysis using always a zenith reference spectrum which is close in time to the measurement spectrum. If indeed glyoxal aloft was enhanced to a point that it caused a turnover point, it would be visible in higher dSCDs measured in the higher elevation angles and decreasing dSCDs in the lower elevation angles. This is not observed (see Fig. 2 in the paper and above). We added this point in the discussion of Fig. 2 in the manuscript.
In our research group we continuously work on retrieving profile information from MAX-DOAS measurements. We agree that the fact that dSCDs collapse at small elevation angles might have previously been noted, but it had – to our knowledge – not yet been exploited systematically for VMR conversion. The approach described in Volkamer et al. (2009b) and applied in this study is a fast and simplified method to retrieve reasonable VMRs and, within the error range, independent from the mixing height.

Discussion: Since it is much simpler to retrieve tropospheric vertical columns instead of VMR from MAXDOAS observations, I would expect at least a simple comparison of MAXDOAS CHOCHO columns with satellite results. These results have originally triggered this study, or not?

This paper is to present ship-based CHOCHO measurements. The need for these measurements was indeed triggered by the mismatch between model results and satellite observations in the remote oceans. A comparison with satellite measurements would require a much deeper look than just comparing VCDs. For example, there is some question about the a-priori assumptions about glyoxal vertical distributions that are at present used to invert satellite retrievals (see e.g., http://www.atmos-chem-phys-discuss.net/10/C9262/2010/acpd-10-C9262-2010.pdf). We agree with the reviewer about the potential of our data to add to this discussion, but this is not the focus of this paper.

In contrast to the authors opinion most of the satellite retrievals are quite consistent (even those just published in ACPD for GOME2). The only exception might be the OMI data set, but this has never published.

As mentioned in the paper, there have not been many CHOCHO satellite retrievals so far, and actually only one of them was published in a peer-reviewed journal. The recent publications in ACPD for GOME2 were published after the submission of this paper. We changed the respective paragraph in the introduction as follows:
'Although CHOCHO has been measured for over two decades (Kieber et al., 1990), CHOCHO measurements continue to be scarce. Also, almost all observations were performed

over land and in the northern hemisphere (Volkamer et al., 2010). Information over the remote ocean is only available from several satellite instruments. Wittrock et al. (2006) and Vrekoussis et al. (2009) published global maps of CHOCHO using the SCanning Imaging Absorption SpectroMeter for Atmospheric CartograpHY (SCIAMACHY). Worldwide CHOCHO distribution maps also were derived from the Ozone Monitoring Instrument (OMI, available at http://www.cfa.harvard.edu/~tkurosu/SatelliteInstruments/OMI/SampleImages/CHOCHO/index.html; Kurosu et al., 2005) and from the Global Ozone Monitoring Experiment 2 (GOME-2; Lerot et al., 2010; Vrekoussis et al., 2010). However, they are not consistent in the amount of CHOCHO, which reflects the fact that measurements from space are inherently difficult to conduct due to low albedo over oceans and cloud coverage (specifically over the tropics).‘

Kurosu, T., Chance, K., and Volkamer, R: Global Measurements of OClO, BrO, HCHO, and CHO-CHO from the Ozone Monitoring Instruments on EOS Aura. Eos Trans, AGU Fall Meet. Suppl., Abstract A54B-01 (ORAL), December 5-9, San Francisco, CA, USA, 2005.

Lerot, C., Stavrakou, T., De Smedt, I., Müller, J.-F., and Van Roozendael, M.: Glyoxal vertical columns from GOME-2 backscattered light measurements and comparisons with a global model, Atmos. Chem. Phys. Discuss., 10, 21147-21188, doi:10.5194/acpd-10-21147-2010, 2010.

Vrekoussis, M., Wittrock, F., Richter, A., and Burrows, J. P.: GOME-2 observations of oxygenated VOCs: what can we learn from the ratio glyoxal to formaldehyde on a global scale?, Atmos. Chem. Phys., 10, 10145-10160, doi:10.5194/acp-10-10145-2010, 2010.

Minor point: Instrumentation: For the correction of the pointing inclinometers are used. For which time period the standard deviation is calculated?

It was calculated for about 2 minutes. We added this information in the manuscript.