

We thank the reviewer for carefully reading our manuscript and for giving detailed comments and suggestions that have been helpful to improve the manuscript. We addressed all of the reviewer's comments. Detailed responses are given below (reviewer's comments are in italics and smaller font size).

General Comments

The authors claim to have performed a “direct” detection of glyoxal, yet their method relies on spectral fitting, which requires a model function to be fitted to observed radiances, and air mass factor (AMF) calculations, for which assumptions on the state of the atmosphere must be made, including the impact of aerosol scattering (cloudy data are not considered in the analysis). Furthermore, their MAX-DOAS technique does not provide a target-free reference spectrum, hence there is bound to be residual CHOCHO loading in their “Fraunhofer reference” against which differential glyoxal columns are retrieved. Admittedly, in ship-based measurements the observing hardware is much closer to the source than in observations made from satellites, but other than that, the authors' method is very much akin to satellite-based spectral retrievals of CHOCHO, and nobody would call those “direct measurements”.

The title of the manuscript should be changed to “(Ship-Based) Detection of Glyoxal over the Remote Tropical Pacific Ocean”. The only other reference to “direct” detection appears in the Conclusions (page 16, line10), which should be changed to “Ours are the first CHOCHO measurements in the remote marine boundary layer (MBL).”

As suggested by the reviewer we had already changed the title to ‘Ship-based Detection ...’ for the ACPD publication and removed ‘direct’ from the conclusions.

Detailed Comments

Note: Page numbers are that of the referee, since no page numbers were included in the manuscript.

General editing remark: Double parentheses as in “(5x10-19cm2 (Volkamer et al., 2005b))” or “(R(3.8))” are awkward and should be avoided.

We removed all double parentheses in the manuscript.

Page 1

line 21: “the open ocean must be a source for”

We changed it as suggested by the reviewer.

Page 3

line 26: change “inherently” to “intrinsically”

We changed it as suggested by the reviewer.

line 27: delete “unequivocally”; it would be nice if this was the case (and it may even be so, for perfect spectra), but spectral-fitting retrievals are often anything else but unequivocal, particularly when cross-correlation with other constituents come into play.

We deleted it as suggested by the reviewer.

Page 4

line 12: “from ships are rare mainly due to the challenge”; or “particularly” instead of “mainly”

The authors do not mean to necessarily create a relation between the two statements. So, this sentence has been kept unaltered.

line 24: change “Solar stray light spectra” to “Spectra of scattered sunlight”

line 27: “Two inclinometers, arranged perpendicular to each other,”

We changed both as suggested by the reviewer.

Page 5

line 13: sampling frequency should be given in addition to FWHM.

The mercury emission line was measured at the beginning and at the end of the measurement campaign without significant change between them. We added this information in the manuscript.

line 20: "CCD capacity of at least one wavelength channel were automatically projected."

We changed it as suggested by the reviewer ('rejected' instead of 'projected' though).

Page 6

line 8: "product of the MAX-DOAS spectral analysis"; DOAS in general does not necessarily use a Fraunhofer reference that contains the target gas.

lines 9/10: "(dSCD, ... Fraunhofer reference spectrum, FRS)"

line 14: "distribution, radiative"

We changed all as suggested by the reviewer.

line 23: is this Ph.D. thesis publicly available? Does a published reference exist?

This thesis is publicly available at

http://hci.iwr.uni-heidelberg.de/publications/dip/2006/Kraus_PhD2006.pdf.

We added the link in the reference list.

line 28: "FRS measured prior to the acquisition of the measurement spectrum."

We changed it as suggested by the reviewer.

Page 7

line 3/4: "strong band around"

We (including a native English speaker) think 'strong band at around' is correct.

line 6: the Brion/Malacet ozone cross-sections are being more and more widely accepted as the gold standard. Is there a particular reason for using Bogumil?

Thanks to the reviewer for the hint that the Brion/Malacet ozone cross-section might be something like the gold standard for ozone measurements. However, we do not think that this is crucial for the measurements in this publication. We neither quantified ozone nor did we retrieve ozone in a wavelength range where the differential ozone absorption is relatively strong. Furthermore, our retrieval approach is based on cancelling out stratospheric absorption, i.e. ozone absorption in the stratosphere. However, for future analysis we will consider including Brion/Malacet ozone cross-section.

Line 10: change "cros sections" to "line parameters"

We changed it as suggested by the reviewer.

Line 10: the HITRAN 2008 water vapor line parameters are significantly improved over the 2004 values. For the next re-analysis, the authors should consider to give these a try.

Actually, the HITRAN 2008 water vapor line parameters were used. We apologize for the wrong reference and updated the reference list.

line 29: delete "likely to be"

line 30: "CHOCHO data set, the"

We changed both as suggested by the reviewer.

Page 8

line 5: replace "Then," with "In those cases,"

We changed it as suggested by the reviewer.

lines 9/10: can you state a typical value for the detection limit?

As mentioned in the 'Discussion' section, the typical detection limit for CHOCHO amounts to a few 10 ppt.

lines 15-17: "for each spectrum, to serve as a measure of how 'blue' the sky is. Higher values indicate a sky with fewer and thinner clouds, lower values represent more and thicker clouds."

We changed it as suggested by the reviewer.

lines 20/21: "Thus, the ration of the color indices at R(3.8) (3.8o elevation angle) and R(25) provide"

We changed it to: 'Thus, the ratio of the color index at the 3.8° elevation angle, R(3.8), and the one at the 25° elevation angle, R(25), provide'

line 22: "in particular"

We changed it as suggested by the reviewer.

line 22: what is meant by "all of the area of measurement viewing directions"?

We admit that this expression is misleading and removed it from the manuscript.

line 26: "was found to be consistent"

We changed it as suggested by the reviewer.

Page 9

lines 11-13: this sentence belongs in to Section 5, since it relates to the components included in/excluded from the DOAS analysis. It should be preceded by a sentence like "Effects from liquid water absorption were not included in the DOAS fit."

We changed this as suggested by the reviewer.

line 14: delete "in arbitrary units"; $\text{cm}^5/\text{molecule}^2$ are the proper units for the $\text{O}_2\text{-O}_2$ cross-sections, hence units of $\text{molecule}^2/\text{cm}^5$ are required to make the product a dimensionless optical thickness.

line 16: "requires sunlight, no"

line 17: "For clarity, only"

line 22: "lower elevation angles, indicating"

line 30: "other, while values"

We changed all as suggested by the reviewer.

Page 10

line 8: "In principle, this effect"

line 12: "The effect of clouds is, typically, to"

line 13: "Under cloud free conditions, the"

lines 29/30: "in path lengths, which usually arise within different elevation angles, are"

We changed all as suggested by the reviewer.

Page 11

line 7: "O4 dSCD, provide the conversion of CHOCHO dSCDs"

line 8: "different solar zenith angles (SZAs)."

We changed both as suggested by the reviewer.

line 18: what exactly is "ground aerosol extinction"?

‘Ground aerosol extinction’ means the aerosol extinction at ground level. As mentioned in the same section, we assumed a constant aerosol load with height relative to the air pressure. Consequently the aerosol extinction decreases with height. In order to clarify this better, we changed it to ‘aerosol extinction at ground level’.

line 22: “for different SZAs.”

line 26: delete “(in arbitrary units)”

line 27: change “presuming” to “assuming”

lines 30/31: “cloud cover makes a significant difference compared to the cloud-free scenario on which”

We changed all as suggested by the reviewer.

Page 12

line 1: “used to remove all 1.5o and 178.5o dSCD values that were”

line 3: “values that did not deviate”

line 5: “The 14% that did”

We changed all as suggested by the reviewer.

lines 7/8: “the assumptions of especially the mixing layer hight and is at most 30%, including the DOAS fit error.”

We changed it to ‘the assumptions of the approach, especially of the mixing layer height, and’

line 24: I may have missed this - are effective path length influences considered in the VMR error analysis?

The calculation of the effective path length is part of the VMR retrieval and therefore is a parameter for the correction factor. Thus, effective path length influences are considered.

Page 13

line 9: “OH-initiated CHOCHO loss.”

line 15: “For the following discussion, we”

line 27: “as a source of CHOCHO.”

changed

Page 14

line 30: “molecular identity than CHOCHO.”

Page 15

line 17-18: remove double parentheses by separating references with a “;”

Page 16

line 10: “Ours are the first CHOCHO measurements in”

We changed all as suggested by the reviewer.