Review

ACP Ms# 2010-325
Authors R. Sinnreich, S. Coburn, B. Dix, and R.Volkamer
Title Direct Detection of Glyoxal over the Remote Tropical Pacific Ocean

Overview

Sinnreich et al. present the first ship-based measurements of glyoxal (CHOCHO) in the remote Pacific. This study is of high relevance for glyoxal, VOC, and SOA research, particularly given the currently still sparse observation record for glyoxal and the discrepancies exhibited by several satellite retrievals of CHOCHO. The paper is well organized and fairly well written, and only requires minor edits. However, the retrieval method presented here does not represent a "direct" retrieval, since it depends on radiative transfer and assumptions on the state and composition of the atmosphere to derive CHOCHO columns and VMRs. The title of the paper must be changed to reflect that.

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 sate 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)."

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 "($5x10^{-19}cm^2$ (Volkamer et al., 2005b))" or "(R(3.8))" are awkward and should be avoided.

Page 1
line 21: "the open ocean must be a source for"

Page 3
line 26: change "inherently" to "intrinsically"

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.

Page 4
line 12: "from ships are rare mainly due to the challenge"; or "particularly" instead of "mainly"

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

line 27: "Two inclinometers, arranged perpendicular to each other,"

Page 5
line 13: sampling frequency should be given in addition to FWHM.

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

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”

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

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

Page 7
line 3/4: “strong band around”

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

Line 10: change “cros sections” to “line parameters”

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.

line 29: delete “likely to be”

line 30: “CHOCHO data set, the”

Page 8
line 5: replace “Then,” with “In those cases,”

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

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.”

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

line 22: “in particular”

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

line 26: “was found to be consistent”

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.”

line 14: delete “in arbitrary units”; $cm^5/molecule^2$ are the proper units for the O_2-O_2 cross-sections, hence units of $molecule^2/cm^5$ 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”

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”

Page 11
line 7: “O_4 dSCD, provide the conversion of CHOCHO dSCDs”

line 8: “different solar zenith angles (SZAs).”

line 18: what exactly is “ground aerosol extinction”?

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”

Page 12
line 1: “used to remove all 1.5° and 178.5° dSCD values that were”

line 3: “values that did not deviate”

line 5: “The 14% that did”

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

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

Page 13
line 9: “OH-initiated CHOCHO loss.”

line 15: “For the following discussion, we”

line 27: “as a source of CHOCHO.”

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”