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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Twelve years of global observation of formaldehyde in the troposphere using GOME and SCIAMACHY sensors" *by* I. De Smedt et al.

**Anonymous Referee #3**

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NOTE: The review mentions an image showing absorption cross-sections. Due to limitations of the ACPD web site, no such image could be included. It was communicated to the authors directly.

1. Overview

De Smedt et al. report on a long-time record of tropospheric formaldehyde retrieved from the GOME and SCIAMACHY instruments. The subject of the paper is suitable for publication in ACP. The paper is written clearly, and discussion of both retrieval approach and error analysis is exhaustive. Some minor editing, as outlined below, is required for the final version, but other than that I find nothing wrong with the manuscript

per se.

My major point of criticism to the approach concerns to the choice of cross sections used for some of the interfering absorbers (mainly O2-O2 and BrO, but also NO2 and O3). For this long-term data record of HCHO - extended in the future by OMI and GOME-2 observations - to be truly useful, the utmost care needs to be taken to ensure that an optimum set of common cross-sections are used for all the instruments. Most likely, this leads to small compromises for the individual instruments, but at the gain of having a truly concise data set of HCHO observations across multiple platforms. Since the study reported in this manuscript aims to provide a concise HCHO data record, the cross-section issue must be revisited in order to answer unequivocally any outstanding questions regarding, say, the influence of errors in wavelength registration of O3 cross-sections on HCHO retrievals (similar studies have been performed for BrO, with illuminating results). Details regarding these cross-section issues are given below.

Even though I expect that changing to a common set of molecular absorption cross-sections will have a significant impact on the HCHO retrievals form GOME and SCIAMACHY, I do not recommend that this be done before the publication of the manuscript (the required effort is rather large, since it involves a complete reprocessing of the entire 12-year data record). However, I strongly urge the authors to make this change before they pursue any further studies with the GOME and SCIAMACHY HCHO data record.

In summary, I recommend publication of the manuscript with minor editorial corrections.

2.Comments on choice of molecular absorption cross-sections

In Table 1, the authors list the set of absorption cross-sections used in the GOME and SCIAMACHY retrievals. They are the same for HCHO, OClO, Ring, BrO (Wahner), and O2-O2 (Greenblatt), while different sets are chosen for O3 and NO2 (Burrows 1998/9 for GOME, Bogumil 1999 for SCIAMACHY). It is particularly the selection of the BrO and O2-O2 cross-sections, as well as the adoption of different sets for different



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instruments that raise questions as to exactly how concise the HCHO data record is. Details follow below.

For illustration purposes, I have included a plot with all absorption cross sections concerned: Cantrell HCHO (convolved to GOME spectral resolution), Wahner and Wilmouth BrO, and Greenblatt and Hermans O2-O2. The rotational Raman (Ring") contribution is shown as well.

(I) NO2 and O3 cross-sections The particular choice of absorption cross-sections - Burrows 1998/9 for GOME, measured with the GOME flight model; and Bogumil 1999 for SCIAMACHY, measured with the SCIAMACHY proto flight model - is one often employed in GOME and SCIAMACHY retrievals. If optimum retrieval for an individual instrument is desired, this is a fair choice. However, this practice becomes questionable when the data records are to be combined, for it usually fails to quantify the effects those different cross-sections have on the retrieval of the target gas. In the case of BrO, Mohamed-Tahrin et al. (2001) showed that a small error in wavelength registration in the O3 cross sections can have significant effects on BrO retrievals. Not choosing a common set of O3 and NO2 cross sections in the GOME/SCIA HCHO retrievals opens the door to just these kinds of errors. Brion/Malicet for O3 and Vandaele for NO2 are perfectly fine choices that work well for both instruments. They require convolutions with the Instrument slit functions (as do the HCHO absorption cross sections!), but this is not problematic. If the common set performs no worse than the instrument-specific/instrument-measured cross-sections, there is no reason not to choose it, as it makes the much stronger case for a consistent HCHO record.

(ii) BrO cross-sections There are currently three sets of BrO absorption cross-section measurements: Wahner (used in this manuscript), Wilmouth, and Fleischmann (the authors are well familiar with those, hence I do not provide detailed references). Wilmouth and Fleischmann, both FTS measurements, are consistent to within 0.6% between the common GOME and SCIAMACHY BrO fitting windows, while Wahner differs from both by almost 30% between the windows and, in addition, exhibits a considerable wave-



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length registration error at shorter wavelengths (within the HCHO fitting window used in the present work). While the differences in cross-section values are more of an issue for BrO retrievals, the error in wavelength registration becomes important when BrO interference is to be removed from the HCHO fit: BrO and HCHO have overlapping absorption features, hence the best-possible wavelength registration is required to minimize mis-allocation of absorption features when both absorbers are varied simultaneously during the retrieval process. The authors should replace the Wahner cross-sections by either Fleischmann or Wilmouth as soon as possible.

(iii) O2-O2 cross-sections The manuscript mentions repeatedly the large impact that O2-O2 absorption can have on the HCHO retrieval. It is unfortunate that they then proceed by choosing a set of cross-sections that has an apparent wavelength registration error of about 1nm over the HCHO retrieval window used here. Comparison with Hermans O2-O2 cross-sections, measured with an FTS, indicate that indeed the error lies with Greenblatt: Sample retrievals using long light-path GOME spectra (large solar zenith angles) including the region around 360nm where O2-O2 has a strong absorption feature, using either Greenblatt or Hermans, show that the Hermans cross-sections substantially reduce the fittng RMS. This indicates that the Hermans data set better captures the O2 absorption peak, and that it is almost certain that the Greenblatt wavelength registration is in error. Given the interference that O2-O2 exudes on the HCHO retrievals, replacing the Greenblatt data set with Hermans should be of utmost importance for the derivation of a high-quality, consistent HCHO data record from GOME and SCIAMACHY.

As I have already indicated in the Overview, I do not recommend that all these changes are to be made prior to publication of the manuscript due to the large amount of work this requires. On the other hand, any check the authors can perform as to the impact of a more consistent (O3, NO3) or better-quality (BrO, O2-O2) set of cross-sections will improve the manuscript and raise confidence in the product they are reporting.

3. Editorial comments (Page and line numbers refer to the Print Edition.)



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Somewhere in the manuscript it should be mentioned why the "tropospheric" from the title is justified. The short lifetime of HCHO - meaning: no transport - plus the fact that the shape factors used in the AMF calculations most likely vanish at lower pressures should be pointed out.

Using FRESCO as source for cloud information will provide Lambertian-reflecting cloud tops. This should be mentioned in the discussion on AMFs.

P7556,L20: "Despite its short lifetime of about 1.5 hrs".

P7556,L21: delete "and plays a central role in tropospheric chemistry". HCHO is not on par with either, OH or O3, both of which more readily deserve that moniker.

P7557,L4: Delete ", however".

P7557,L15: "as a means to provide".

P7557,L20: Replace "complement" by "extend"; there is not enough temporal overlap between GOME and SCIAMACHY for the data sets to be truly complementary.

P7558,L15: "measuring sunlight back-scattered and reflected at Earth's atmosphere and surface".

P7558,L24: "recorded continuously between".

P7559,L1: Add "(in the nominal mode of observation in the HCHO retrieval window)". SCIAMACHY on-ground spatial resolution depends on the spectral window and geolocation, hence a general statement like "60x30" is misleading.

P7559,L1: If the swath width is 960km, then there have to be 16 pixels across track.

P7560,L19ff: The Ring effect (see plot above) has a fairly benign wavelength dependence. Any higher-oscillation structure is introduced by division with the irradiance spectrum and is hence is due to the DOAS approach, not rotational Raman scattering.

P7563,L16: "exceeds GOME by 30%".



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P7567,L10ff: Is it known why SCIAMACHY spectra would be more affected by the SAA?

P7572,L12: "cloud top is at 8km".

P7573,L18: "GEOS-Chem".

P7580,L22: "better quantified since it may have".

Caption Fig 2: "Panel d shows".

Reference

Mohamed-Tahrin, N., A.M. South, D.A. Newnham, and R.L. Jones, A new accurate wavelength calibration for the ozone absorption cross section in the near-UV spectral region, and its effect on the retrieval of BrO from measurements of zenith-scattered sunlight, JGR 106 (D9), 9897-9907, 2001

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 7555, 2008.