

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

I. De Smedt et al.

Received and published: 30 June 2008

The authors would like to thank the three referees for their detailed reviews. The quality and readability of the paper has certainly been improved by answering their comments.

In reply to the main criticism raised by referees 1 and 2, we have tried to condense the manuscript by first limiting the theoretical introductions and associated redundancies, and second by suppressing three figures (fig. 6, 8 and 13). In addition, section 4.5 which was felt unnecessary was also suppressed. The second main comment expressed by referees 1 and 2, concerning the qualitative nature of the comparison between the satellite data sets has been addressed by providing additional quantita-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



tive statements where relevant and possible.

Replies to comments from Referee 1

Referee 1: question 1: p7558 - line 10: the sentence "discussion of differences with previously retrieved datasets" is partly misleading for the reader who will expect to find a detailed comparison between the different retrievals. I would suggest rewording this sentence.

The sentence has been modified as follows: "Section 5 presents the global distribution and temporal evolution of the CH₂O vertical columns derived in this study, as well as a brief comparison with previously retrieved datasets."

Referee 1: question 2: p7563 - reference to Fig. 4: Why the examples of CH₂O optical density fits are not given for the overlapping period between GOME and SCIAMACHY? If it is because of the degradation of GOME performances, it could be interesting to also provide this information to the reader.

Because it is important to compare the intrinsic quality of the fit that we can achieve with the two instruments, the satellite scenes have been chosen to illustrate fitting results under optimal conditions, i.e. for high formaldehyde columns and when both instruments performed at their nominal signal to noise ratio. After 2002, the degradation of the instrument results in poorer S/N ratio, hence larger fit residuals, however the fits remain of good quality when the signal of formaldehyde is high enough. Figure 5 shows two overlapping orbits of GOME and SCIAMACHY in 2003 together with fitting residuals. The impact of the degradation of GOME performances can be judged there. This section has been reformulated also to answer the question 4 of referee 1.

Referee 1: question 3: P7563 - caption of Fig. 5: There is an error in the standard deviation values given in the caption. In the text, the authors write that the standard deviation of SCIAMACHY is larger than the GOME one and it is the contrary in the caption.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

The caption has been corrected as follows: "The standard deviations of GOME and SCIAMACHY slant columns are 6.1015 molec./cm² and 1.1016 molec./cm² respectively."

Referee 1: question 4: P7563 - lines 14-17: In order to clarify the statement "the standard deviation in the SCIAMACHY slant columns ONLY exceeds GOME one by 30%, as a result...", it would be interesting to add how the standard deviations compared before the degradation of GOME performances.

The section has been reformulated as follows: "From photon statistical consideration, when the instruments perform at best, the ratio between the standard deviations should be about 0.38 because the SCIAMACHY ground pixels are 7 times smaller compared to GOME. We find a ratio of 0.4, the SCIAMACHY RMS being 60% higher than the one of GOME. This demonstrates that the noise in the retrieved columns is consistent with the signal to noise ratio of the instruments. Figure 5 shows the slant columns for two overlapping orbits of GOME and SCIAMACHY over Eastern China, on the 14th of April 2003. The mean CH₂O values agree very well and, in this case, the standard deviation of the SCIAMACHY slant columns exceeds GOME by only 30%, as a result of the long term degradation of the GOME instrument from 1996 until 2003 (see Subsect. 4.2)."

Referee 1: question 5 (and Referee 2: question 5) P7566 - lines 21-22: The vertical CH₂O profile shapes that the authors used are provided on a monthly based. Could the authors precise the variability of this profile shape and which the impact would be if a constant profile shape was used?

Recently, a new version of the CH₂O vertical columns has been generated using daily IMAGES profiles. These daily profiles have been obtained with an improved version of the model, in which emissions as well as relevant meteorological parameters (temperature, water vapor, boundary layer diffusion, convective updraft fluxes, cloud optical depth) vary on a daily basis. Based on these data, tests have been performed to estimate the influence of the temporal variability of the CH₂O profiles. The comparison

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

with our baseline vertical column product (obtained using monthly IMAGES profiles) shows that the profile variability results in negligible differences for the background CH₂O. In regions of emissions, the mean influence on the columns is around 5% in Africa, South America, Indonesia or India, and up to 10% in Europe, eastern US and China. In Millet et al., 2006, the error due to the use of a modeled air mass factor is estimated to be 7% over North America, which is consistent with our sensitivity tests. Note that the error due to the profile shape uncertainty has been determined by comparison of the IMAGES profiles with aircraft measurements in US and in the Tropics. From these comparisons, uncertainties on the altitude of the profile maximum and on the profile width have been stated. Using this method, the error due to profile shape uncertainties has been estimated to be 20%. The comparison between IMAGES profiles and aircraft data is limited to North America since intensive campaigns allowing relevant quantitative evaluation of the model profiles were so far only available for this region. Global distributions of the IMAGES model are further presented in a companion paper by Stavrou et al., 2008, submitted to JGR.

Referee 1: question 6: P7566 - lines 24-27: A better quantification of the comparison between the simulated and the observed profiles would help to support the statement "fairly good agreement".

A quantitative comparison has been added: "The modeled mixing ratios at altitudes higher than 1.5 km essentially lie between the values defined by the two datasets, whereas over the continental boundary layer they are more than 10% higher than the NCAR measurements (Stavrou et al., 2008)."

Referee 1: question 7: P7567 - Section 3.4: A detailed quantitative comparison would be valuable for the discussion. Will the comparison results differ if the horizontal resolution of SCIAMACHY is degraded to match GOME resolution? Moreover, this section is in the middle of the method description. It would make sense to include this paragraph in Section 5 and to develop it with more quantitative results. Concerning the 2 last sentences of this section: do some hypotheses exist to tempt to explain the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

differences?

Section 3.4 has been moved to the "Results and discussion" section and has been further developed to provide a more quantitative evaluation of the GOME and SCIAMACHY comparisons, as suggested by the referees 1 and 2. The main results of the comparison would not differ if SCIAMACHY columns were degraded to the GOME resolution because the data are already monthly averaged and/or re-gridded over larger regions (as considered for analysis of regional emissions).

Referee 1: question 7, referee 2: question 8 and referee 3 comment: Why is the South Atlantic anomaly more pronounced in SCIAMACHY than GOME data?

The large scatter in CH₂O SCDs observed in the region of the Southern Atlantic Anomaly (SAA) in the earth magnetic field, results from exposure of the satellite instruments to enhanced radiation and high energy particles. The reason for the difference of behaviour between GOME and SCIAMACHY is currently not fully understood. It might be related to differences in the design of the detectors used in both instruments (mainly the electronic gain and the instrument throughput for photons). If the detector gain is higher, and the throughput a corresponding factor lower, the SAA energetic particle impacts on the detector will be more prominent with respect to the detected light, while the detected radiance or reflectance signal will not be significantly different in both instruments. Alternatively differences in the instruments shielding could possibly explain the observed different sensitivities to SAA.

Referee 1: question 8: P7568 - section 4.1: the authors chose to estimate the random error on the slant column using the standard deviation of measured columns. How do this error compare to the direct fitting error of the retrieval?

In regions of low emissions and therefore weak variability in the C₂H₂O column, the random error on the slant columns can be expressed by the standard deviation of measured columns around the mean. On the other hand, and provided that fitting residuals are dominated by shot noise in the detection system, the random error on the

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



slant columns can be estimated from the properly weighted root mean square (RMS) of the fitting residuals (Taylor, 1982). We use RMS values as an estimate of the random error. This precision has been added to the manuscript, with reference to the Book by Taylor (1982). Ref: Taylor, John R., An introduction to error analysis, University Science Books, 81-91 and 153-159, 1982.

Referee 1: question 9: P7569 - line 11: Are the concentration profile or the slant columns of the species considered here fitted in the same time than the CH₂O columns or fixed during the fit? In the former case, the resulting error should be accounted for by the fitting (random) error.

The slant columns of all interfering species are fitted together with the CH₂O columns. Fitting residuals only account for random errors. Systematic errors which are due to cross-sections interferences and uncertainties need to be estimated independently, which is the purpose of the matrix analysis presented in the paper.

Referee 1: question 10: P7573 - section 4.4: a reminder of the random error for one pixel (in relative) would help the reader.

The sentence has been modified as follows: "Note that the total error is generally much larger (about 70%) and dominated by the random error when considering individual pixels."

Referee 1: question 11: P7574 - lines 20-21: A better quantification of the "very good agreement" is expected.

See question 7. This section has been further developed to provide a more quantitative assessment of the GOME - SCIAMACHY comparisons (see revised manuscript).

Referee 1: Technical corrections

P7562 - line 13: "the biggest differences" has been replaced by "the largest differences".

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

P7565 - line 2: The word "only" has been suppressed in "W depends only on..."

P7570: The lines 5-6 have been suppressed.

Replies to comments from Referee 2

Referee 2: question 1: 18 figures are excessive and in the interest of readability I suggest the authors seriously consider reducing this number.

See general comments.

Referee 2: question 2: P7557 - line 8. Should include Palmer et al, 2003 that developed the theory to infer VOC emissions from HCHO columns.

The reference to Palmer et al, 2003 has been added.

Referee 2: question 3: P7557 - line 19. "Despite SCIAMACHY provides..."??

The sentence has been modified as follows: "SCIAMACHY provides nadir UV-visible radiance measurements suitable to extend GOME observations. However, until now, only one study reports its use to retrieve CH₂O columns (Wittrock et al., 2006)."

Referee 2: question 4: P7562 - line 4: Later of latter?

The word "later" has been replaced by "latter".

Referee 2: question 5: A serious concern is that the model, used to determine the air-mass factors, is driven by monthly mean meteorology. Is this really good enough to capture daily variations in isoprene? The authors have provided only scant evidence of the model ability to reproduce mean statistics of aircraft profiles over North America and yet show global distributions. I appreciate that a companion paper might cover this in some detail but it might be worth summarising where (and why) the model perform well and badly.

See Referee 1: question 5.

Referee 2: question 6: There are qualitative (and hollow) statements about data sprin-

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

kled throughout the paper, e.g., "fairly good agreement" and "very satisfactory". I suggest the comparisons are quantified or the qualitative statements are removed.

See Referee 1: question 7

Referee 2: question 7: What is the origin of the localized hotspot of HCHO over the Highveld region in South Africa?

The localized hotspot of CH₂O over the Highveld region in South Africa corresponds to a highly industrialised region, which lies 1400-1700 m above sea level in the Mpumalanga Province. The CH₂O emissions are likely to be due to emissions of anthropogenic NMVOCs. Elevated concentrations of NO₂ are also observed in the same region. Emission densities are amongst the highest in the world. More than 60% of the emissions in the Highveld region occur from stacks taller than 200m, and most of these emissions originate from 8 large coal fired power plants and a synthetic fuel (from coal) processing complex (Wenig et al., 2003). The latter reference has been added in the paper.

Ref: Wenig, M., N. Spichtinger, A. Stohl, G. Held, S. Beirle, T.Wagner, B. Jahne, and U. Platt, Intercontinental transport of nitrogen oxide pollution plumes, Atmos. Chem. Phys., 3, 387-393, 2003.

Referee 2: question 8: Why is the South Atlantic anomaly more pronounced in SCIAMACHY than GOME data?

See Referee 1: question 7

Referee 2: question 9: To reduce the length of the paper, I suggest the authors consider removing/reducing some of the review information that is discussed in published papers, e.g., averaging kernels and air mass factor calculations.

The section and the figure about averaging kernel have been suppressed. Averaging kernels are mentioned at the end of section 3.3.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



Referee 2: question 10: P7575 - line 17. What is the reason for the smaller seasonal variation in the BIRA HCHO product?

The reasons for the difference with the Harvard dataset over Eastern US (and elsewhere) are under investigation. Preliminary work shows that the differences are mostly related to differences in the settings used for the DOAS retrievals (like the fitting windows or the handling of interfering species, mainly ozone and BrO), rather than to differences in air mass factor calculations. A comprehensive intercomparison of the retrievals is certainly desirable although it was felt beyond the scope of the present paper. Instead this will make the subject of a future focused publication involving other retrieval teams.

Replies to comments from Referee 3

Referee 3: Comments on the choice of molecular absorption cross-sections

Sensitivity tests have been performed based on a subset of GOME and SCIAMACHY orbits. Alternative cross section datasets have been selected for each molecule in order to estimate the error due to the choice of cross sections and also to validate the results obtained with the SCD error estimation presented in the paper. Note that the two methods (sensitivity analysis versus matrix analysis) were found to lead to consistent error estimates. This precision has been added in the section on the slant column errors. For O₃, we have compared the impact of using three different cross section data sets (Voigt et al., GOME FM 98 and SCIAMACHY PFM). In the 328.5–346 nm window, globally, we have obtained a sensitivity of the CH₂O slant columns of 2% in the case of GOME and of 5% in the case of SCIAMACHY. Of course, the sensitivity is function of the latitude. It is almost zero at the equator and increases to 15% at 60° of latitude (20% for SCIAMACHY). For NO₂, we have tested the Vandaele et al. cross section as an alternative to the flight-model reference data sets. The CH₂O fit was found to be insensitive to this change of cross section dataset, implying little sensitivity to uncertainties on the NO₂ absorption cross-sections. Nevertheless

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

we agree that for the sake of homogeneity it would be more satisfying to adopt a coherent common set of reference data applicable to both GOME and SCIAMACHY. This will be considered for future processing. Concerning BrO, the Wahner et al. cross section data set has generally been used in the literature (see e.g. Aliwell et al., 2002). In order to minimize the impact of the known wavelength shift, a pre-shift of +0.17 nm has been systematically applied in our CH₂O retrievals. This precision has been added in Table 1 in the paper. Further tests have been performed using the Wilmouth et al. cross section. The sensitivity to the change of BrO cross section is 3% for GOME and 2% for SCIAMACHY. Concerning the O₄ cross-sections, the Greenblatt et al. dataset also has known wavelength shifts which must be corrected for. In our study, we applied a shift of +0.2 nm (precision added in Table 1). In the fitting window selected here for CH₂O retrieval (328.5–346 nm), the O₄ absorption is weak in comparison to what typically happens e.g. in the SCIAMACHY BrO fitting interval (337–351 nm). Therefore, the sensitivity to O₄ cross section uncertainties is expected to be weak. This is the reason why the O₄ contribution has not been displayed in figure 9 of the paper. Nevertheless, sensitivity tests have been performed using the alternative Hermans et al. (2006, unpublished results) O₄ cross-section data set. For GOME, in the 2 fitting windows, the differences with the Greenblatt results are below the percent. The mean value above Sahara desert is also equivalent using the two O₄ datasets (and below the background in the 337–359 fitting interval). For SCIAMACHY, the sensitivity is below the percent in the UV-shifted fitting interval. Ref : Aliwell, S.R., M. Van Roozendael, P. V. Johnston, A. Richter, T. Wagner, et al., Analysis for BrO in zenith-sky spectra - An intercomparison exercise for analysis improvement, *J. Geophys. Res.*, 107, D14, doi: 10.1029/2001JD000329, 2002.

Referee 3: Editorial comments

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

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

should be pointed out.

In Section 3.3, a sentence has been added in the description of the shape factors, to explain that formaldehyde is mainly a tropospheric molecule: "Because of the short lifetime of CH₂O, there is no transport in the stratosphere and the shape factors vanish at lower pressures."

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

In the same section, the following sentence has been added together with a reference to FRESCO: "FRESCO provides Lambertian-reflecting cloud heights."

P7556 - L20: The following sentence has been added: "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.

The sentence "and plays a central role in tropospheric chemistry" has been deleted.

P7557 - L4: The word ", however" has been deleted.

P7557 - L15: The sentence "as a mean to provide" has been changed to "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.

The word "complement" as been replaced by "extend".

P7558 - L15: The sentence has been modified: "measuring sunlight back-scattered and reflected at Earth's atmosphere and surface".

P7558 - L24: The sentence has been changed to: "recorded continuously between".

P7559 - L1: Add "(in the nominal mode of observation in the HCHO retrieval window)".

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



SCIAMACHY on-ground spatial resolution depends on the spectral window and geolocation, hence a general statement like "60x30" is misleading.

The precision "(in the nominal mode of observation in the HCHO retrieval window)" has been added.

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

The mistake has been corrected; there are 16 pixels across track and not 14.

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 hence is due to the DOAS approach, not rotational Raman scattering.

It is explained that the Ring effect is important when the DOAS method is used.

P7563 - L16: The expression "exceeds GOME ones by 30%" has been changed to "exceeds GOME by 30%".

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

See Referee 1: question 7

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

Since FRESCO uses scattering properties to retrieve cloud top heights, the quantity generally retrieved does not correspond to the geometrical cloud top height but to some effective scattering height located towards the middle of the cloud. This is why we talk about the "cloud height" in the paper. Ref: Wang et al., An improved O₂ A-band cloud retrieval algorithm for tropospheric trace gas retrievals, Atmos. Chem. Phys. Discuss., submitted.

P7573 - L18: The acronym "GEOS-CHEM" has been changed to "GEOS-Chem".

P7580 - L22: The sentence has been changed to "better quantified since it may have".

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Caption of Fig 2: The sentence "Panel d shows" has been added.

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

ACPD

8, S4322–S4334, 2008

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

S4334

