

Referee #2

Specific Comments

First of all, I would like to compliment the authors on the successful creation of a 16-year H₂CO data record from multiple sensors. Formaldehyde is an extremely challenging atmospheric constituent to retrieve, and to do it consistently not only on a global scale but across platforms is quite a feat. I fully appreciate the GOME-2 specific modifications that had to be made to the retrieval approach. There is no “one-approach-fits-all-sensors” for small absorbers like formaldehyde, as instrument idiosyncrasies can easily introduce spectral features on the order of (or larger than) the H₂CO absorption features themselves. I am sure that this data set will find wide use in air quality research.

(i) My main questions regarding the retrieval approach for GOME-2 relates to the issue of BrO pre-fitting, as introduced in section 3.2, “Firstly, BrO slant columns are fitted in a wide wavelength interval (328.5–359 nm) that includes six BrO absorption bands and minimizes the correlation with H₂CO”:

1. Was H₂CO used as interfering absorber in the determination of BrO? The phrasing of the sentence seems to hint at that, but it isn't clear. The problem is, of course, that over strong H₂CO source regions like the Amazon or Indonesia (biomass burning), H₂CO can strongly correlated with BrO. If not simultaneously fitted, the derived BrO can be seriously over or underestimated and, when used as fixed input to a subsequent H₂CO fit, can lead to significant errors in formaldehyde.
2. Independent of whether H₂CO was used in the BrO retrievals, have the resulting BrO columns been checked for residual correlations with H₂CO, e.g., over strong H₂CO source regions? The (co-)authors of this manuscript have a strong history in BrO retrievals, so a comparison against the prime BrO product should be easy enough to do.
3. How are the BrO uncertainties used, either in the H₂CO fit itself or the quantification of errors of the final H₂CO product? Ideally, the H₂CO fit would be performed with linear constraints, where the pre-fitted BrO is allowed to vary within 1–2 σ fitting uncertainties. Very few retrieval codes have that option though. However, at a minimum, the BrO uncertainties have to be considered in the total H₂CO error.

Generally speaking, the spectral interference between BrO and H₂CO manifests itself as an overestimation of the BrO column in the Tropics where H₂CO concentrations are large. At the same time, the spectral cross-talk between the two molecules also tends to increase the noise of the H₂CO columns. This is generally accompanied by an underestimation of the H₂CO columns at large solar zenith angles. For this work, H₂CO is indeed fitted together with

BrO in the first spectral interval (328.5-359 nm). In this large window, there is no evidence of H₂CO impact on the BrO columns in the Tropics, while, as mentioned by the reviewer, H₂CO has a clear impact in the usual BrO fitting window (336-352 nm). This was one of the findings reported in the Theys et al. (2011) paper. For comparison, we show here Figure 1 of Theys et al (2011), together with the BrO geometrical columns obtained in our 328.5-359 nm retrieval interval, which is close to the final Theys et al. interval (332-359 nm). The BrO columns obtained in our large window have been systematically checked and found to be consistent with results published in Theys et al (2011).

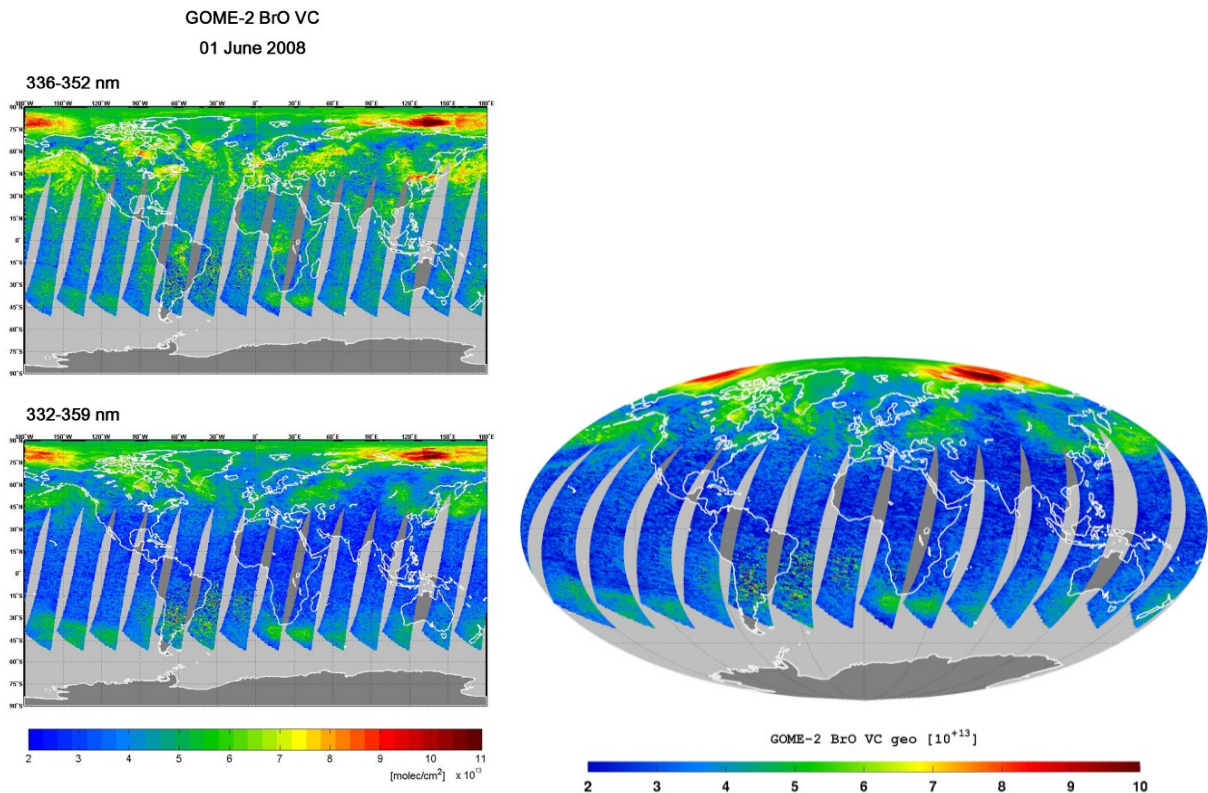


Figure 1: Left: Comparison of GOME-2 BrO vertical columns for 1 June 2008 using (upper map) the 336–352 nm fitting window from Theys et al. (2009a) and (lower map) the 332–359 nm interval introduced in the study of Theys et al. (2011). Right: GOME-2 BrO vertical columns for 1 June 2008 using the 328.5–359 nm interval (De Smedt et al., 2012). Simple geometrical air mass factors have been used to convert BrO slant columns into vertical columns. Note the reduced artefacts due to formaldehyde interference (e.g., in East China or Central Africa), the elimination of the viewing angle dependence and the reduced cloud-related structures compared to the 336–352 nm fitting window.

For the slant column error estimation, we use the formulation presented in De Smedt et al. (2008), which presents the advantage of taking into account the various correlations between the cross-sections, weighted by the slant column of each fitted parameter. For this work, we formulate the error as if BrO was fitted together with H₂CO in the second interval. We do not take into account the “de-correlation” offered by the pre-fit of BrO. This is indeed a limitation of the error estimate, and it probably results in an overestimation of the uncertainty.

(ii) In section 4: Air mass factors, a reference to cloud screening of “cloud fractions exceeding 40%” is made. It should be specified whether this is “effective”, “radiative”, or “geometric” cloud fraction.

The 40% refers to the effective cloud fraction provided by Fresco v6 (by opposition to the fractional cloud radiance that has been used in some other studies). This precision has been added in the text.

*No explicit correction is applied for aerosols but the cloud correction scheme accounts for a large part of the aerosol scattering (Boersma et al., 2004, 2011; De Smedt et al., 2008). In all cases, observations with **effective** cloud fractions exceeding 40% are filtered out. For larger cloud fractions, the error on the vertical column increases rapidly.*

Editorial Comments

Page 5575, line 4: “and retrieved consistently”

*To fully exploit the potential of satellite data, applications relying on tropospheric H₂CO observations require high quality long-term time series, provided with well characterised errors and averaging kernels, and **retrieved consistently** from the different sensors.*

Page 5581, line 8: “orbit on”

*Figure 5 illustrates the effects of the two corrections (dual fit for BrO and H₂CO, and O₃ correction) for one particular GOME-2 **orbit on** the 13 August 2007, which is typical of background level H₂CO concentrations.*

Page 5585, line 20: “10%”

*Over the Mediterranean Sea, vertical columns are **10 %** higher during summer time, due to lower reflectances over waters in the Kleipool climatology compared to Koelemeijer.*

Page 5589, line 7: “cloud and aerosols”

*A full treatment **of clouds and aerosols** in radiative transfer will only be possible if clouds and aerosols are represented separately as scattering layers and if detailed information on aerosol optical properties is available at the global scale (Valks et al., 2011).*

Page 5592, lines 8, 16: “H₂CO”

*Note that in India and China, the positive trends previously detected in the GOME and SCIAMACHY **H₂CO** columns (De Smedt et al., 2010) are supported by the GOME-2 observations. The GOME-2 columns are about 10% larger than SCIAMACHY in northern and equatorial Africa, such as in southeastern US and Europe. Larger differences are found in Australia, where the GOME-2 columns are 15% lower than SCIAMACHY, but appear more consistent with the GOME time-series. The discrepancy between SCIAMACHY and GOME-2 in this region is mostly important during the winters of 2009 and 2010, when the amplitude of the seasonal variations in the SCIAMACHY data is significantly reduced. An instrumental degradation effect in SCIAMACHY time-series cannot be excluded. As already mentioned, instrumental degradation has noticeable effects on the **H₂CO** random error (see Fig. 3), but no significant impact on the H₂CO columns was detected so far (except over Australia for SCIAMACHY).*

Page 5593, line 19: delete “an” in “an excellent”

*The resulting SCIAMACHY and GOME-2 formaldehyde columns, monthly averaged over the main NMVOC emission region of between 2007 and 2011, **show excellent agreement**, with correlation coefficients higher than 0.8 and mean column differences lower than 10% almost everywhere.*