

Anonymous Referee #1

Page 5573, Line 9: Typo? Should “key actor” be “key factor”?

The sentence has been changed as:

*Monitoring the spatial and temporal variability of NMVOC emissions is essential for a better understanding of the processes that not only control the production and the evolution of tropospheric ozone, **playing a key role** in air quality and climate change, but also of the hydroxyl radical OH and secondary organic aerosols.*

Page 5574, Line 8: Phrasing. Change to “...variability of tropospheric emissions

The sentence has been changed as:

*Moreover, the higher spatial sampling of GOME-2 in comparison to GOME and SCIAMACHY is expected to allow for a better identification of the spatial structures of short-lived tropospheric species like H₂CO, and for a more precise determination **of the variability of tropospheric emissions** (Lerot et al., 2010; Richter et al., 2011).*

Page 5578 & Figure 2: Have the sensors been sampled in the same way, i.e. only using days/locations when coincident observations occurred? How do differences in the cloud products from SCIAMACHY and GOME-2 contribute (if at all) to these differences? Please elaborate.

For Figure 2, GOME-2 observations have been restricted to viewing angles smaller than 32°, to match the SCIAMACHY swath. To answer the question of reviewer 2, we have redone the figure, using only GOME-2 observations coincident with SCIAMACHY observations. The results are shown in Figure 1 of this document. The underestimation of GOME2 v07 compared to SCIAMACHY remains (and is even more pronounced), and increases with time.

As Figure 2 presents differential slant columns, there is no correction for clouds, except from a cloud filtering for cloud fractions larger than 40%. The cloud products used to filter the pixels are fully consistent and based on the Fresco v6 algorithm. We have done the same figure for different cloud fraction thresholds (from 10% to 60%, dotted lines of Figure 1), and we find very little differences in these monthly and latitudinal averages, both for SCIAMACHY and for GOME-2. Moreover, the impact of the different ground pixel sizes on the cloud selection is rather limited. Indeed, it has been shown in Krijger et al. (2007)¹ that the respective pixel areas of SCIAMACHY and GOME-2 (1800km² and 3200km² in nominal mode) result in about 19% and 21% of cloud free pixels at the global scale (if up to 20% of the footprint area is allowed to be filled by MODIS cloudy observations).

¹ Krijger, J. M., van Weele, M., Aben, I., and Frey, R.: Technical Note: The effect of sensor resolution on the number of cloud-free observations from space, Atmos. Chem. Phys., 7, 2881-2891, doi:10.5194/acp-7-2881-2007, 2007.

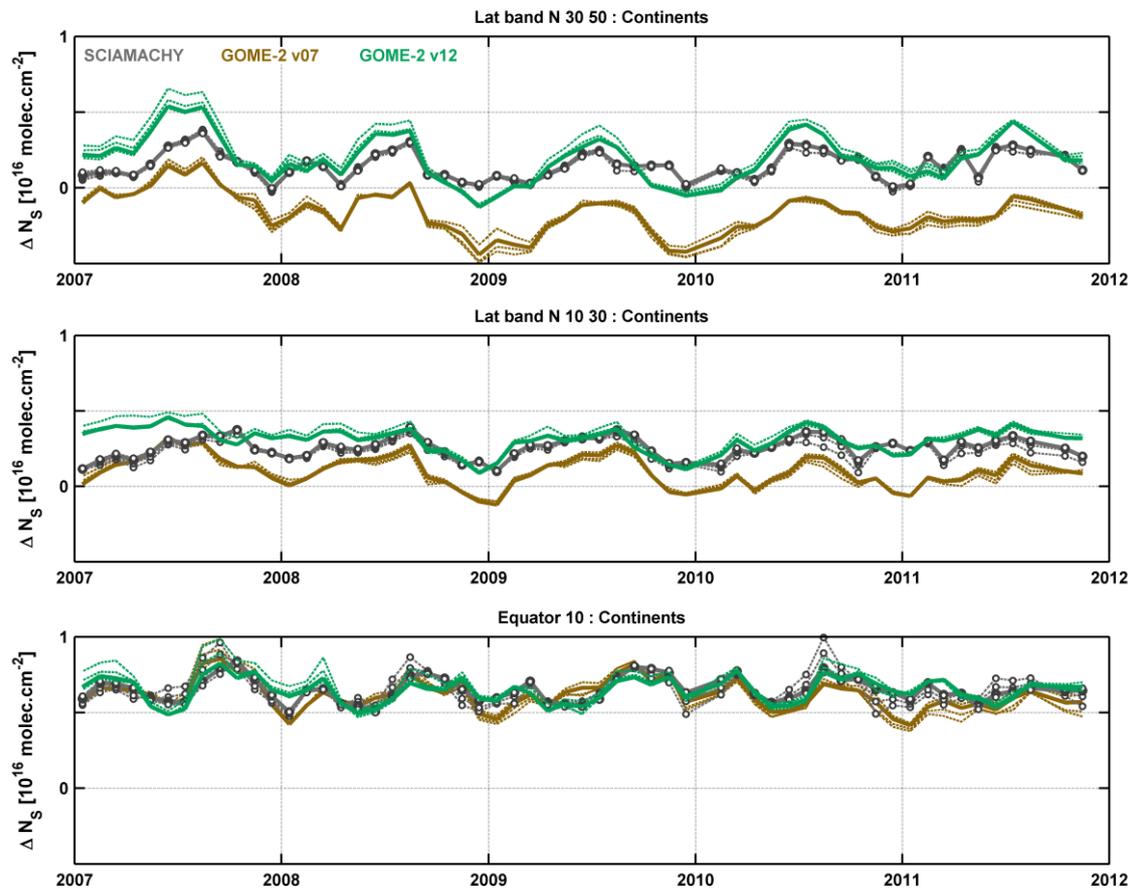


Figure 1: Monthly H₂CO differential slant columns averaged in 3 latitude bands over continents, as retrieved from SCIAMACHY and GOME-2 measurements. GOME-2 observations have been restricted to viewing angles lower than 32° and to coincident observations with SCIAMACHY. The maroon line shows the GOME-2 results obtained with the initial retrieval settings (v07), while the green line shows the results obtained with the improved settings (v12). The dotted lines show different cloud filtering, from 10% to 60%.

Section 3.2 *I'm not familiar with the approach of Pukite et al. (2010). Can you please provide more details on this correction, especially as it has a significant impact on the retrieval.*

The paper of Pukite et al. (2010) addresses the application of the DOAS method to limb measurements in the UV. In the case of limb view, the light paths are significantly longer than in nadir, and consequently the ozone absorption is not optically thin as it is required to apply classical DOAS. I copy here a part of the abstract of Pukite et al. (2010): "However, the DOAS method can be extended to cases with medium to strong absorptions and for broader wavelength intervals by the so called air mass factor modified (or extended) DOAS and the weighting function modified DOAS. These approaches take into account the wavelength dependency of the slant column densities (SCDs), but also require a priori knowledge for the air mass factor or the weighting function from radiative transfer modelling. We describe an approach that considers the fitting results obtained from DOAS, the SCDs, as a function of wavelength and vertical optical depth and expands this function into a Taylor series of both

quantities. The Taylor coefficients are then applied as additional fitting parameters in the DOAS analysis. Thus the variability of the SCD in the fit window is determined by the retrieval itself. This new approach provides a description of the SCD the exactness of which depends on the order of the Taylor expansion, and is independent from any assumptions or a priori knowledge of the considered absorbers.”

By extension, their method can also be applied to nadir observations, to cope with O₃ absorption at large solar zenith angles, when the optical paths are longer in the stratosphere and the optically thin atmosphere assumption of DOAS is no longer valid. As stated in our paper, in practice, we use the Taylor series they propose (that describes the O₃ slant column as function of wavelength and of vertical optical depth over the fitting window) to the first order, and we add two cross-sections to the fit: $\lambda\sigma_{O_3}$ and $\sigma_{O_3}^2$ (equation 11 in Puķīte et al. (2010)). It allows a much better treatment of optically thick ozone absorption in the retrieval, while keeping the simplicity and speed of DOAS.

We believe it is unnecessary to further develop this method in our paper, since it has been fully documented in the paper by Puķīte et al. (2010)).

Section 4: Which version of MEGAN was used in the IMAGES model?

In the MEGAN-ECMWF model presented in Müller et al. (2008), the global emissions of isoprene are calculated based on the MEGAN (Model of Emissions of Gases and Aerosols from Nature) version 2 model (Guenther et al., 2006) and a detailed multi-layer canopy environment model for the calculation of leaf temperature and visible radiation fluxes. The calculation is driven by meteorological fields provided by analyses of the European Centre for Medium-Range Weather Forecasts (ECMWF).

For this work, a new version of isoprene emissions is derived based on ERA-Interim meteorological fields (e.g. temperature at 2 m above the ground, soil moisture, etc.) and leaf area index from collection 5 8-day MODIS composite from Aqua and Terra. The emissions are estimated at 393 Tg in 2007, 373 Tg in 2008, 396 in 2009, 419 in 2010, and 395 in 2011. They are available at <http://tropo.aeronomie.be/models/isoprene.htm>.

Figure 2: Can the y-scale be stretched (e.g., from -0.5 to 1.0), to help distinguish the different lines?

Done.

Figure 6, RMS Panel: It is unclear which FWHM is being plotted. Is it the pre-flight or asymmetric slit function? Please clarify.

It is the fitted FWHM of the asymmetric slit function. It has been clarified in the legend. The pre-flight FWHM is by definition constant.

Fig. 6. (a) GOME-2 slit function width (FWHM) and asymmetry factor (AF) fitted during the calibration procedure of the DOAS analysis. (b) Mean residuals of the GOME-2 solar spectrum calibration between 330 and 360 nm, using the pre-flight slit function or fitting a Gaussian

asymmetric slit function. The FWHM of the fitted Gaussian asymmetric slit function is shown as second y-axis.