Author reply to Referee #2

Lisa K. Behrens et al.

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We thank Referee #2 for carefully reading our manuscript and for the helpful comments which will improve the quality of our manuscript. We will reply to the comments point by point.

Furthermore, we noticed a small mistake in the discussion manuscript. For the AMF calculation in the HCHO satellite retrieval, we accidentally used data from a different CTM (TM4-ECPL). However, for consistency, all retrievals in the manuscript should use the same CTM data as a-priori. Therefore, we have replaced the use of TM4-ECPL by the same MOZART-4 data as was also used in the MAX-DOAS and CHOCHO satellite retrievals. This introduces only small changes compared to the dataset shown in the discussion manuscript. In the revised manuscript, all HCHO and CHOCHO retrievals will use the same MOZART-4 data set.

Legend:
- referee comments
- authors comments
- text in manuscript
- **changed text in manuscript**

This manuscript describes the detection of HCHO and CHOCHO in African outflow during the COPMAR project in 2016. The authors find elevated HCHO and CHOCHO at higher altitudes, and suggest biomass burning and transport of long-lived precursor VOCs as the source. Overall, the manuscript presents an important set of observations and sufficient preliminary analysis and is suitable for ACP. The following comments should be addressed before publication:

Thank you very much for the positive comments.

Comments:

1.) Section 4.5 discusses MOZART outputs which show elevated HCHO and CHOCHO between 3000m and 6000m, while MAX-DOAS shows only elevated HCHO. FLEXPART is then used to investigate potential sources. Why not turn on/off biomass burning in MOZART and see the impact on modeled profiles, or look at MOZART outputs of other tracers?

Turning on/off biomass burning or other traces would be a good approach to investigate the differences between MOZART-4 and MAX-DOAS measurements. However, the MOZART-4 product which we use for the comparison and AMF calculation is an official product provided by NCAR through https://www.acom.ucar.edu/wrf-chem/mozart.shtml. Consequently, we did not run MOZART-4 by ourself and cannot run the model with different initial conditions. Furthermore, we suggested in the manuscript that the gases are trapped by aerosols and than transported. This mechanism cannot be shown by changes in the biomass burning initial conditions of MOZART-4.

In our study, we used FLEXPART simulations to gain further knowledge of the origin of the airmass which have been observed. FLEXPART can simulate the transport of such gases trapped by aerosols.

2.) If MOZART underestimates HCHO south of the equator, how does using incorrect MOZART profiles to calculate MAX-DOAS VCDs influence the retrieval in those regions?
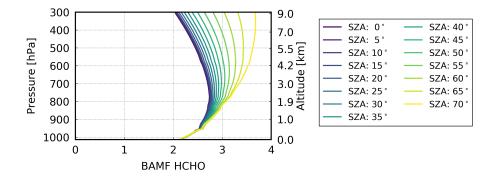

Figure 1: HCHO MAX-DOAS BAMF for 30° VZA and 90° RAA for different SZAs.

The influence of the RAA on the AMF is often smaller than the influence of the SZA, therefore, in the following only the SZA will be discussed.

First, MOZART-4 can differ from the real atmosphere in several ways. As shown in Fig. 1, the influence on the AMF depends on the SZA. For small SZAs, BAMF variation with altitude is less pronounced than for large SZAs. Figure 13 in the paper shows that MOZART-4 simulates slightly enhanced HCHO values between 600 and 700 hPa on the days with large differences between the datasets. If this layer is in reality higher than 600 hPa or lower than 900 hPa and the SZA is small, the AMF would decrease and lead to higher VCDs of the MAX-DOAS measurements. Between 700 and 900 hPa, variability of the BAMF are small. Changes in these layers would result in a decrease of the MAX-DOAS VCDs. For large SZAs, the BAMF show a constant increase above 600 hPa leading again to a decrease of the MAX-DOAS VCDs, whereas for HCHO below 700 hPa, the MAX-DOAS values would be enhanced.

Second, the absolute amount of HCHO could be not correctly simulated. If the model underestimates/overestimates the amount of HCHO, this would in good approximation not change the AMF. Consequently, the MAX-DOAS VCDs would not change.

Third, the model could miss an additional layer of HCHO. This would also lead to an underestimation/overestimation of HCHO which results in larger VCDs for the MAX-DOAS measurements.

Because Referee#1 made a similar comment, we will change the discussion in Sect. 4.2 as follows:

[...] The MAX-DOAS measurements are mostly higher than the satellite observations (Fig. 10 a) which results in a slope larger than one (1.57, OMI and 1.10, GOME-2B) and a large offset $(0.86 \times 10^{15} \, \text{molec cm}^{-2}$, OMI and $3.81 \times 10^{15} \, \text{molec cm}^{-2}$, GOME-2B) of the regression line (Table 2). These differences are clearly visible between 20° N and 32° N (09 October – 11 October) and between 10° S and 22° S (18 October – 21 October; Fig. 10 a), which have been measured in clean remote ocean areas with low pollution.

Compared to the MOZART-4 model values, the MAX-DOAS observations are often higher which can also be observed in a low slope of 0.68. Close to the Equator, the values show good agreement in the area of expected pollution outflow which leads to a high correlation coefficient between the two datasets of 0.72. However, the offset is also high between the two datasets ($4.82 \times 10^{15} \, \text{molec cm}^{-2}$).

Several reasons can contribute to an enhancement in MAX-DOAS data. Large

differences between the MAX-DOAS and the satellite or model data are found over regions with low air pollution north and south of the Equator. In these areas, high measurement uncertainties can be found in the satellite data due to the low columns which might influence the retrieved satellite values. It is also possible that the model and satellite results underestimate the VCDs, because of the potentially localised nature of the enhancements (see also Sect. 5). Increased uncertainties in the MAX-DOAS data in this region can be excluded as the DOAS fit RMS is nearly constant during the whole cruise (see Sect. 3.8.3). Furthermore, H_2O interferences might contribute to the differences. Then, the differences should be reduced/increased for higher/lower elevation angles, respectively. However, this does not seem to be the case, as a similar behaviour is visible for all viewing directions. Additionally, bad weather conditions can be excluded, because the affected days of the MAX-DOAS measurements had different weather conditions (see Table 1) and an intensity filter was used to exclude poor viewing conditions (see Sect. 3.1). Similar conditions were also used for satellite values. Here, only measurements with geometric cloud fraction smaller 0.3 were included.

The AMF could introduce the differences between the datasets. However, this seems unlikely as the MOZART-4 model is used for the AMF calculations for both MAX-DOAS and satellite measurements and this model does not show a similar behaviour as the MAX-DOAS measurements. For example, if the model underestimates/overestimates the amount of HCHO in the atmosphere, this would in good approximation not change AMFs, and therefore, the VCDs of both datasets are not influenced. Thus, no difference would be introduced between the satellite and MAX-DOAS data. Possibly, the differences between the datasets can be related to the HCHO model profile. The HCHO profile of the model could differ from the real atmospheric HCHO profile. On the one hand, the model could miss an additional layer which is present in the atmosphere. Such a difference between model and real atmosphere would result in larger VCDs for the MAX-DOAS and satellite measurements. On the other hand, the model could simulate the HCHO in the wrong atmospheric layer. This could lead to both an underestimation or an overestimation of the MAX-DOAS VCDs depending on the profile and the SZA or relative azimuth angle which is similar for satellite measurements. Additionally, aerosols could influence the AMFs leading to differences in satellite and MAX-DOAS VCDs observations.

3.) I echo reviewer 1's request to discuss the profile shapes used in satellite retrievals. Ideally, there would be consistency between these profiles and those used in the MAX-DOAS retrievals.

We agree and therefore have used the same profiles for the AMF calculations for satellite and MAX-DOAS data for CHOCHO. In the revised version of the manuscript, we will also use MOZART-4 for the AMF calculation of HCHO for the satellite data. Unfortunately, we used TM4 in the manuscript for the AMF of satellite HCHO which should not have been the case. However, this change introduces only small changes for the comparison of the datasets. Please see also the answer to the previous question. We will add a comment on this in Sect. 3.4:

The 4-D fields of HCHO and CHOCHO concentrations are needed as a priori information for the calculation of VCDs for both MAX-DOAS and satellite data (Sect. 3.7).

4.) The last two paragraphs of section 5 (comparison with other studies) are important, and it is difficult to interpret the authors' intent.

The authors first state that CHOCHO enhancements are "usually attributed to local production from CHOCHO precursors either originating from marine biota or from transported organic aerosol rich in dissolved organic carbon". They then state that "it seems more likely that on this days [sic] its source is related to transported precursors". Are they referring to their own observations only, or commenting also on previous authors' conclusions?

The next sentences are also problematic. The authors write "The aerosols trap the gases which are lifted and transported together. These stored gases are then re-released to the gas phase by reversible desorption after several days." There is no evidence of this in their work. It should be stated as a potential explanation or hypothesis rather than fact.

We will change the text as follows:

[...] Similarly to HCHO, also CHOCHO has been reported to be present at enhanced levels over remote ocean regions in satellite observations (Vrekoussis et al., 2009; Stavrakou et al., 2009; Lerot et al., 2010). These CHOCHO enhancements are mostly visible in regions of strong biogenic activity and biomass burning, and are usually attributed to local production from CHOCHO precursors either originating from marine biota or from transported organic aerosol rich in dissolved organic carbon (Vrekoussis et al., 2009).

Our MAX-DOAS measurements suggest that the observed HCHO **and** CHOCHO **enhancement is partly located in an elevated atmospheric layer which is in contrast to previous publications. It seems more likely that on the days where our measurements show enhanced values its source is related to transported precursors, which is also in line with the findings presented in Sect. 4.4. Possibly, the aerosols trap the gases which are lifted and transported together. These stored gases can then be re-released to the gas phase by reversible desorption after several days.** Similar results were found for a case study over continental area in Asia by Alvarado (2016). The presence of a combination of dust and biogenic aerosol, and thus, potential VOC sources in that region and during that season could be shown by Ridley et al. (2012) using Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIOP) and model data. Furthermore, Volkamer et al. (2015) found similar results for the Pacific Ocean. They found in the equatorial Pacific west of the American continent enhanced CHOCHO columns mostly in elevated layers, ruling out marine sources. [...]

5.) In general, there is not enough care given to instrument accuracy. A large portion of the manuscript is dedicated to describing the DOAS analysis. For that reason, uncertainty should be quantified if possible. AOD and Angstrom-exponent measurements are also discussed and compared without any comment on accuracy or precision.

For satellite measurements error analyses are discussed in the publications from Boersma et al. (2004) and Lorente et al. (2017) for NO_2 and HCHO. Furthermore, for the satellite products which we are using here the errors will be discussed in Alvarado et al. (2014, 2019). We will add the following sentences in the manuscript in Sect. 3.2:

The measurement accuracy for satellite data are described in Boersma et al. (2004) and Lorente et al. (2017) as well as in Alvarado et al. (2014, 2019). Generally, over remote ocean areas the uncertainty of tropospheric columns is dominated by errors in the stratospheric column (for NO_2**) and errors in the spectral fitting (Boersma et al., 2004). Boersma et al. (2004) found a relative uncertainty of up to 100 % for** NO_2 **in these areas.**

For the AOD and Ångström-exponent we used publicly available products which can be downloaded at: https://aeronet.gsfc.nasa.gov/new_web/maritime_aerosol_network.html. Details about

this product can be found in Smirnov et al. (2009). Measurements errors for this instrument are discussed in Ichoku et al. (2002), Morys et al. (2001), Porter et al. (2001) and Knobelspiesse et al. (2003, 2004). We will add the following sentence in the manuscript in Sect. 3.3:

Details of the Microtops handheld Sun photometers and their uncertainties can be found in Ichoku et al. (2002), Morys et al. (2001), Porter et al. (2001) and Knobelspiesse et al. (2003, 2004).

MAX-DOAS measurements are influence by systematic and random errors. The following paragraph will be added in Sect. 3.8.1:

Each MAX-DOAS measurement is influenced by a systematic and a random error (Irie et al., 2011; Takashima et al., 2012). The random error can be estimated from the residual of the fitted trace gases (Takashima et al., 2012). However, also systematic errors can be found in the residual such as not perfectly resolved cross sections. Furthermore, Boersma et al. (2004) and Takashima et al. (2012) showed that the temperature dependency of the cross section also leads to systematic errors in the retrieved SCD. For example for NO_2, it is in the order of $0.4\,\%\,K^{-1}$ (Takashima et al., 2012). For the vertical columns, the errors are in the order of 15 to $20\,\%$ (Takashima et al., 2012; Peters et al., 2012).

Minor comments:

1.) Page 2, line 11: "A global background concentration of HCHO exists of 0.3 – 2.0 parts per billion". This is a wide range and it is unclear what is meant by 'background'. For this manuscript, a useful number would be the concentration (in ppb or column density) over the remote ocean stemming from CH4 oxidation.

We will change the text as follows:

[...] Primary emissions are from biomass burning and the combustion of fossil fuels, but HCHO is also formed in the atmosphere from the oxidation of methane (CH_4) and non-methane hydrocarbons (Arlander et al., 1995). **Due to the oxidation of CH_4, HCHO is not only found close to its source regions, but a global background concentration of HCHO exists with surface levels of $0.2-1.0$ parts per billion in remote marine environments (ppb; Weller et al., 2000; Burkert et al., 2001; Singh et al., 2001).** Furthermore, it is an important indicator of the photochemical activity for a region (De Smedt et al., 2008; Vrekoussis et al., 2010). [...]

2.) Section 2.4: What is meant by 'the model data are linearly interpolated?' Interpolated in time? Sampled at the observation location?

Yes, in Section 3.4 a interpolation in time is meant. We will change the text as follows:

[...]**For the comparison, the model data are linearly interpolated in time on the cruise track.**

3.) Section 3.2: What versions of the satellite products are used? OMI-BIRA? OMI-SAO? The

only reference given is a Ph.D. thesis. Either the product should be described, or peer reviewed literature should be sited.

The satellite retrievals used in this study are in house products from IUP Bremen based on OMI level 1 data provided by NASA (Levelt et al., 2006) and GOME2 level 1 data from EUMETSAT (Callies et al., 2000; Munro et al., 2016). For NO$_2$, we used the fit settings presented in Richter et al. (2011). For HCHO and CHOCHO, the settings were presented in Alvarado et al. (2018) at EGU which will be published in Alvarado et al. (2019). The CHOCHO is an improved version of Alvarado et al. (2014). We will additionally include in the manuscript the reference Alvarado et al. (2018):

[...] **Additionally, satellite observations for** HCHO **and** CHOCHO **are taken into account which are described in Alvarado et al. (2018) and Alvarado et al. (2019).** [...]

4.) Section 3.6: Is FINN the inventory included in MOZART? If so, it should be in section 3.4.

No, MOZART uses the GFED-v2 inventory. We use FINN dataset in Sect. 4.5 to discuss potential sources of VOC precursors.

5.) Page 9 lines 26-27: "For the calculation of this limit, different approaches were used in previous studies, for example, Peters et al. (2012), Sinreich et al. (2010), and Platt et al. (1997). In this study, the detection limit is calculated for each trace gas individually with the method from Sinreich et al. (2010)." If these methods are different, the choice should be justified. If they are not significantly different, the other studies should not be discussed.

As suggested by the referee, we will remove the sentence: "For the calculation of this limit, different approaches were used in previous studies, for example, Peters et al. (2012), Sinreich et al. (2010), and Platt et al. (1997)." from the manuscript.

6.) Page 15 line 17: "Due to the primary emission sources of HCHO" sounds like direct HCHO emissions, which I do not believe is what the authors mean. Perhaps "photochemical production of HCHO from VOC precursors".

No, we meant primary emissions. The formulation might be misleading and we will change the manuscript as follows:

The latitudinal variation of daily mean MAX-DOAS HCHO dSCDs observed during the cruise shows enhanced values in an elevated layer at both $\sim 10°$ N and $\sim 5°$ S. This coincides with the area of expected outflow from the African continent as seen in satellite measurements (Fig. 1 a). **Typically, the** HCHO **concentration in polluted areas is expected to be highest close to the surface (De Smedt et al., 2008; Heckel et al., 2005) due to the primary emission sources of** HCHO **and rapid photochemical production from VOC precursors. Thus, also the highest dSCDs should be observed at low elevation angles near the sources.** Figure 9 illustrates the daily mean HCHO dSCDs during the cruise at different elevation angles. [...]

7.) Page 27 line 1: Clarify what is meant by aerosol 'type'.

We will change the sentence as follows:

[...] **The observed aerosol differs between Africa north and south of the Equator (having different AOD and Ångström-exponent) and therefore, the outflow in both hemispheres seems to be from different sources.** [...]

8.) Page 27 line 5: "The present study is the first to confirm the enhanced levels of HCHO and CHOCHO frequently observed from satellites over the Atlantic Ocean using ship-based measurements." Is this true (could be), or is it the first ship-based MAX-DOAS measurements over the Atlantic?

This were not the first MAX-DOAS measurements over the Atlantic Ocean, but the first where both, HCHO and CHOCHO, were analysed and connected to outflow from the African continent. Mahajan et al. (2014) analysed CHOCHO MAX-DOAS measurements over the Atlantic Ocean. To our knowledge, HCHO has previously only been analysed from other measurements types.

9.) Throughout: A correlation coefficient of 0.55 or 0.56 is 'moderate' (not 'good') agreement.

Changed as suggested.

Technical comments:

1.) Page 9 line 14: "In this study, the trace gases are expected to be in elevated layers and from satellite measurements". Please clarify.

"In this study, the trace gases are expected to be in elevated layers and from satellite measurements. Furthermore, it is expected that they have a latitudinal dependency (see Fig. 1)."
We will change the manuscript as follows:

[...]**In this study, the trace gases are expected to be in elevated layers. Furthermore, satellite measurements indicate that they depend on latitude (see Fig. 1).**[...]

2.) Page 9 line 25: "ground-based" should be "ship-based".

Changed as suggested.

3.) Page 26, line 18: It is unclear what is meant by "On 3 respectively 2 days".

We will change the text as follows:

[...]**For HCHO on 3 and for CHOCHO on 2 days, our measurements show clearly enhanced levels of these trace gases.** [...]

Additionally as suggested by Referee #1, we will change three main points in the revised manuscript:

1. We clarified the discussion of HCHO for the days 13, 14, and 17 October.

2. We added an altitude axis to Fig. 13 and Fig. 14.

3. We added the distances to the continent/hot spot area for the days with observed outflow events.

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