Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-1104-RC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterisation of Central-African aerosol and trace-gas emissions based on MAX-DOAS measurements and model simulations over Bujumbura, Burundi" *by* Clio Gielen et al.

Anonymous Referee #1

Received and published: 13 February 2017

This paper presents a unique multi-year data set obtained over central Africa. Because there are almost no similar data records over this part of the globe, the data set presented here is of great value to obtain insights in tropical atmospheric chemistry and for validation of air quality models and satellite observations in this region.

The paper is very well written and presents many interesting and relevant findings in informative figures and tables. I recommend the paper for publication in ACP after the following comments have been addressed.

I have one main comment [C1], and four other comments [C2-C5]

[C1] Figures 9 and 13 show diurnal evolution of column amounts and profiles respectively. There are some aspects of both figures that raise questions:

First: in figure 9 a strong increase in AOD is seen at the end of the diurnal cycle for the MAX-DOAS product at 360 and 477nm, whereas a similar increase is not seen in the AERONET data.

Second: in figure 13 a strong variability is seen in the aerosol extinction profiles between morning and afternoon. This effect is not in line with the weak diurnal pattern in aerosol profile shapes (figure 13, right column) nor with the diurnal pattern in AERONET AOD. Even though the MAX-DOAS AOD shows a relatively weak diurnal variability in AOD when compared to the strong feature in fig. 13 upper left panel.

A possible explanation for this discrepancy could be that the MAX-DOAS results shown in these figures are affected by inhomogeneous temporal sampling over the seasons. This view is supported by the green numbers in figure 9. It is mentioned by the authors (p.9,l.27-28) that the diurnal patterns are not affected by the fact that less values are used to determine the afternoon percentiles. However, I am not convinced because it is quite probable that the results after 1PM, and especially the second half of the afternoon are dominated by measurements that are not representative for the entire year. This effect may also affect the diurnal cycles for NO2 and HCHO columns: although the column retrieval is generally quite robust compared to the profile retrieval, quality control (cloud filtering) may lead to an apparent diurnal cycle when - for instance - some seasons have considerably more cloudy afternoons than others. Perhaps it would make sense to be more strict in the selection of data. For instance: include only data from days where at least 3 out of 6 data points in the morning AND 3 out of 6 data points in the afternoon pass the quality control criteria. This ensures a more representative temporal sampling over the day and avoids the possibility that some hourly bins are dominated by one or two seasons, whereas others are representative for all seasons. Furthermore the temporal sampling should, in my view, be made consistent for the figures 9 and 13 and transparent in the sense that the reader should be able

to see how many data points from each season contribute to each bin of the diurnal cycle. Another possibility is to compute monthly averaged diurnal cycles and to give these equal weight (1/12) when determining the diurnal cycle averaged over one year. Please reconsider all interpretation throughout the manuscript of the patterns seen in these figures after the figures have been made again according to the suggestions above (or similarly).

Further comments:

[C2] Fig. 4. Please report relevant AOD in the caption of this figure.

[C3] Fig. 9 shows descending NO2 and HCHO columns after 8-9AM. It is argued that this diurnal variation is - at least for NO2 - due to peak emissions during the rush hour (p.10, l.4-10). It is indeed remarkable that the same is observed for HCHO, as noted by the authors, because of the different origins of this gas. The question therefore is: what are the possible explanations for the diurnal variability found for the HCHO columns. Before investigating further this mechanism in terms of emissions, transport and atmospheric photo-chemistry, alternative explanations - related to retrieval accuracy and further data analysis - should be considered. The issue of temporal sampling was already mentioned above: this could possibly affect this figure and should be checked first to my opinion. Secondly, I am not fully convinced that this diurnal cycle for HCHO is not related to variability in shielding strength by aerosols to trace gases above a certain altitude: for high AOD the MAX-DOAS measurements become at some point more and more insensitive to trace gases above a height of (say) 500-1000 meters (this altitude depends on the AOD). The fraction of the column for which this is the case may depend not only on the AOD, but also on the relative HCHO and aerosol profile shape and therefore vary throughout the day. In theory this effect should be taken into account by the MAX-DOAS profile retrieval method, but the effectiveness of this algorithm is doubtful, given the flat curves of Figure 13 (right column). I would like to know the authors view on this, and in case they share this concern ask them to add a few sentences on this topic to the discussion.

[C4] The aerosol profile shapes retrieved at 477 nm show in general little variablity throughout the seasons, but are most variable in months with low AOD (fig. 8). This could be an indication of several things: profile shapes are indeed more variable for conditions with low AOD, for instance because more sunlight reaches the earth surface leading to convection (a physical explanation); profile shapes just happen to be more variable in those months (i.e. coincidence of furthermore uncorrelated events); the profile retrieval is ineffective for conditions with high AOD (methodological limitation). I think this should be mentioned to the reader who is not familiar with the subtleties of MAX-DOAS profile retrieval and its interpretation.

[C5] Sect. 4.2.3 Although I think much effort is done to compare the satellite and the MAX-DOAS NO2 and HCHO columns, I think there ares still methodological limitations in addition to the point mentioned by the authors on p. 13, l.31-33. For instance: clear sky averaging kernels are used in relation to the satellite products, but these do not account for the presence of aerosols. This may be quite relevant in this region (depending on the season). In a sense, the comparison is only partially 'consistent' between satellite and MAX-DOAS (referring to the use of profile information to make a more valid comparison). Being aware of the practical challenges to add this element, I do not request to quantify this aspect in the study, but I would like to see this point mentioned in the discussion.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1104, 2017.