Interactive comment on "Sensitivity of formaldehyde (HCHO) column measurements from a geostationary satellite to aerosol temporal variation in East Asia" by H.-A. Kwon *et al.*

General comments

This manuscript summarises the results of an OSSE study with the goal to evaluate the sensitivity of HCHO retrievals from a Geostationary sensor (the Korean GEMS mission) to aerosol variability. The OSSE is performed as a best-case scenario, *i.e.* without noise, and tries to identify the best strategy when taking into account explicit aerosol correction into the computation of AMF: either by considering hourly of monthly aerosol variability. This is an important question since trace gas retrievals from a geostationary sensor will deliver an unprecedented high number of observations with a high temporal frequency. This is an interesting study, focusing on a very important topic. In general, this manuscript is well written, but several explanations or sentences should be further explained (please see details in "Technical corrections").

However, I recommend some major revisions to be addressed by the authors before publishing the manuscript. Indeed, in my personal opinion, several elements are either critically missing or not correctly addressed. I would like to emphasize, in particular, the point 1) in "Specific comments" which, I believe, is the most crucial one that needs to be carefully clarified in order to better understand / evaluate the importance of the outcome of this study. As this may cause quite some works, I am open to simple exercises suggested by the authors to answer the questions below.

My main remarks about this manuscript are summarised below, but please see details in "Specific comments":

- The importance of the temporal variability of the aerosol altitude (or vertical distribution), in addition to the optical properties (AOT and SSA) is not specifically addressed and analysed. Some lines in the manuscript mention it but in a too ambiguous way to be able to understand how this parameter was considered in this study. By reading the manuscript, I had the feeling that either the aerosol altitude was considered as negligible, and/or it was just ignored for unknown reasons. By experience, aerosol vertical profile is one of the key parameter when retrieving trace gases from space (if not the most crucial one) and its impact should not be neglected or minimized compared to AOT and/or SSA.
- No analyses focussing on observation times that are only accessible by a geostationary instrument are shown here (*i.e.* outside of the range 11:00-12:00-13:00, typical overpass times of present day polar orbiting satellites). It is a bit a pity since this is one of the original element provided by a geostationary platform.
- The definition of "monthly averaged AMF" and the methodology of computing the averaged profiles are a bit misleading, in particular for readers outside of the HCHO retrieval community.
- The assumed aerosol profile in the OMI HCHO exercise (Section 5 of the manuscript) is not discussed. In addition, it would be nice to compare hourly and monthly HCHO AMF for the Dust storm event of 23-29 March 2009. Only the AMFs with and without aerosols are here compared.

Specific comments

1) Aerosol altitude and/or profile?

I struggle to understand the assumptions made by the authors about the shape of vertical distribution of aerosols from hour to hour, day-to-day and month-to-month and how they explicitly impact the computed HCHO AMF depending on the considered methodology (either hourly variability or monthly averages). So far, in my understanding, the authors only considered the impact of assuming constant AOD and SSA properties:

- Line33 P10, "each one of the HCHO profiles and aerosol optical properties is allowed to vary hourly"
- Line19, P11, "we compare hourly AOD and SSA at 300 nm with monthly values"
- Figures 6 and 7 only focus on AOD and SSA variability (which are of course of importance) but do not show the aerosol altitude changes.

These statements and figures, and many others, seem to suggest that the variability of the vertical aerosol profile itself was not explicitly considered, independently and/or combined with their optical property variability. Moreover, the authors mentioned on P.9 that "the peak altitude of aerosols increases from the surface to 2 km". I don't think that such a general statement is always true. Is it a general conclusion supported by referent observations studies over the considered area, or what is seen in the GEOS-Chem model? I would expect to see quite some variations about the height of the peak of the aerosols as it should be strongly driven by 1) the injection height (either in the boundary layer or in the free troposphere), 2) how well the boundary layer (season and synoptic variability) is developed, and 3) specific chemistry processes associated with aerosol particles that may vary depending on their type and the seasons. For example, [Castellanos *et al.*, 2015] demonstrated that biomass burning aerosols extend to high altitudes (about 2 km). But dust particles that are transported over long distance can be found sometimes higher than 2 km. Similarly, sulphate and nitrate particles which result from precursor trace gases may be confined close to the surface where the sources are present.

P9, it is said "Increasing AOD for scattering aerosols (SSA = 0.92) results in an increase of AMF whereas the absorbing aerosols (SSA = 0.82) result in a decrease of AMF". I tend to disagree with such a general statement because:

- Aerosols with SSA=0.92 are still in my view absorbing (although less than with SSA = 0.89). And therefore, I am not sure they can be named "scattering";
- The balance between enhancement or shielding effect strongly depends on 1) the shape of aerosol vertical profile, 2) the shape of trace gas (here HCHO) vertical profile, and thus the relative altitude between the 2 components. Many studies emphasized the importance of the relative vertical distributions of both aerosols and trace gases (such as NO_2) on the satellite AMFs [Boersma *et al.*, 2004; Chimot *et al.*, 2016; Shaiganfar *et al.*, 2011; Ma *et al.*, 2013; Kanaya *et al.*, 2014; Wang *et al.*, 2016]. The magnitude then, of the shielding or enhancement effects, relies on the AOD and SSA associated with particles present in the observed scene. Increasing AOD may not always lead to a decrease of AMF, depending on the aerosol altitude and also the surface albedo. For instance, if very scattering particles are located far from the surface and above the tropospheric HCHO bulk, then we should expect to see an increase of enhancement effect with increasing AOD...
- Absorbing aerosols mostly reduce the sensitivity to HCHO concentration [De Smedt *et al.*, 2008] which can result either in a stronger shield effect or a lower enhancement effect compared to scattering particles, depending again on their relative altitude to the HCHO tropospheric bulk.

The authors should give clarifications how much the vertical distribution of aerosols, based on full GEMS-Chem simulations, varies and how the relative altitudes with respect to HCHO vary as well. I trust this information should be available. Is there a dependency from day-to-day or on the seasons?

Furthermore, how the vertical profile of the particles was considered in the present work: was a full vertical profile simulated every hour by GEMS? Or did the authors only consider 1 finite and homogeneous aerosol layer with variable mid-level of pressure / altitude? Of course, I understand that finding a good that finding a good aerosol profile shape estimate is a complex task, but any assumption made about this should be clarified here.

Did the authors average the vertical profiles as well or did they keep them constant hour-to-hour and day-to-day? All these elements are at least as important as hourly AOD, and much more than hourly SSA (as considered in Figure 6 and so), and should have crucial impacts on the variability of HCHO AMFs. I suggest that, in addition of monthly averages of SSA and AOD, the authors indicate us how monthly averages of the vertical profile shape and/or the effective aerosol altitude impact as well the accuracy of the results.

Finally, could the authors clarify and support with figures or references the statement on P. 9, lines 25-28 "This indicates that the aerosol height may not be a significant factor for GEMS HCHO measurements with a fully developed planetary boundary layer height during the afternoon, but could be an important consideration with a shallow boundary layer, a residual aerosol layer above, and long range transport aerosols"? I do not either understand the message of the authors here...

I realize that my demands, here, may cause quite a lot of work for the authors. If they cannot fully be addressed by coupling the transport-chemistry model for aerosol profile shape estimates, I would like the authors to propose then simple aerosol profile shape sensitivity exercises with academic scenarios (*e.g.* low, intermediate and high aerosol profile), to compute the AMF for these scenarios and address the conclusions. If not, then I think that the limitations of this study (*i.e.* one important parameter not considered in the temporal aerosol variability) should be explicitly written in the title, abstract and other places of the manuscript.

2) Notion of "monthly averaged AMF" is ambiguous

The notion of monthly averaged AMF is a little ambiguous. [De Smedt *et al.*, 2008] & [Gonzalez Abad *et al.*, 2015] do not apply a monthly averaged AMF to GOME single pixels but a specific AMF deduced for each observation pixel, based, among other elements:

- A climatology surface albedo [Koelemeijer *et al.*, 2003] which provides monthly Lambert-equivalent reflectivity at 335 nm;
- And monthly vertical profiles of HCHO distribution from a global chemical transport model (GEOS-CHEM or IMAGES).

The other parameters such as effective clouds, angles, surface altitude / pressure are not averaged at the monthly scale but used on a daily basis. Therefore, the mentioned references in this paper did not strictly use a monthly averaged AMF as stated by the author.

Same about the monthly average AMF of the author here: are only aerosols and HCHO profiles averaged or also other parameters? Following point 1) above, what was averaged regarding the aerosols: AOD and SSA only? Or the vertical profile as well? Or was this last element kept constant? I suggest the author to clearly define the monthly average AMF at the beginning of the manuscript.

3) Clarification of monthly average definition?

Following point 2) above, could the authors precise the period over which the averages were computed? Were they performed over all times of all days in 1 month, or were the averages computed over all days at 12:00 only? Are all the times, or only some of them, considered for the monthly averages?

4) Typical geostationary observation times

Why in Section 4 and on figures 3-5 do the authors only show the impact of the different AMFs at 11:00-12:00-13:00? These times are typically encountered by LEO instruments. But with a geostationary sensor, it could be interested to evaluate the impacts outside of this time range such as early in the morning (9:00-11:00) and close to the end of the afternoons (15:00-17:00).

5) OMI HCHO exercise

Following the discussions above, could the authors:

- Detail which altitude and vertical profile they considered when computing the OMI HCHO AMF? Does it come from GEOS-Chem simulations? In my knowledge, the OMI aerosol product from [Torres *et al.*, 2013] includes AOD and SSA but no vertical profiles.
- Regarding the dust storm event of March 2006 from 23 to 29, could the authors show as well the ratio of hourly *vs.* monthly AMF? Only the ratio of AMF without vs. with aerosols is here shown.

6) HCHO aerosol correction AMF

The author mentioned in Section 3 that "previous algorithms used in sun-synchroneous satellites to retrieve HCHO have not accounted for aerosol effects on AMF calculations".

This is not correct. They corrected for aerosol effects but in an implicit way: *i.e.* the effective cloud parameters are used to partially correct these effects since the cloud retrieval algorithm is perturbed over cloud-free scenes but dominated by aerosol particles. These parameters are either derived from the O_2-band and/or the O_2-O_2 band. The authors [De Smedt *et al.*, 2008] and [Gonzales *et al.*, 2015] clearly said "the presence of aerosols is not explicitly accounted for".

Similarly to the other trace gas retrievals from UV-Vis air quality satellite measurements, the use of a simple Lambertian cloud-scheme, although allows to mitigate their impacts, does not apply a comprehensive correction. See [Boersma *et al.*, 2004, 2011; Chimot *et al.*, 2016; Castellanos et al., 2015] who explained this mechanism in case of tropospheric NO_2 AMF calculations.

Here, the author considers an explicit aerosol correction scheme on the HCHO AMF computation. The relevant question here is then, what would be the best strategy if an explicit aerosol correction is assumed: monthly average or hourly aerosol profile and properties?

Assuming that the author would not have enough explicit information about aerosol properties and vertical distribution, would the use of daily effective cloud parameters, derived for each single observation pixel, be enough to compensate of temporal variability of aerosol effects?

Technical corrections

Abstract:

- 29: Please see my general comments about scattering and absorbing aerosols and correct your general statement accordingly.
- P2, 2: Please precise that you are talking about the impact of aerosol variability, not the aerosols in general.

P2, 30: "frequencies of 1 to 6 days". I suggest to replace by "between 1 and 6 days".

P3, 16: Please add references about Sentinel-4.

P3, 31: "pre-calculated monthly averaged AMF": please precise following point 2) above.

P4, 2-4: these lines are more appropriate in the conclusion section, not in the introduction, since they summarise your results of this manuscript.

P5, 12-15: please reformulate. Computed radiances cannot "become" synthetic radiances…

P5, 21: Were H_2O and O_2-O_2 included as well?

P6, 30 and equation 1: I do not fully understand how this equation has been derived and did not manage to find it in other references. Could you please provide with 1-2 details about it and any references supporting it? What are the limits of the integrals?

P.9, title of section 4: the sensitivity of the HCHO retrieval to the HCHO profile was investigated too (to be added in the title).

P.9, 4-8: Please add references supporting these statements here (*e.g.* Eck *et al.*, 2005; Jethva *et al.*, 2014)

P.9, 17-29: Please see my major remarks in point 1) above (cf. Details about aerosol altitude and vertical profile), and update this sub-section accordingly.

P.9, 21: "Our AMF calculation is consistent with the previous study". Which study are you referring to? In which sense your AMF is consistent? In terms of precision or employed methodology? Please clarify.

P.9, 30-31: this statement is hard to understand, since the previous lines somehow said that aerosol profiles are not important....Please clarify or reformulate.

P.30, 7-8: Which figure are you referring to?

P.10-11, 30-1: Following point 1) above, please clarify if you kept constant or made vary the aerosol profile? How was this parameter considered here and how did it impact your results?

P.11, 23-25: "In other words, absorbing aerosols [...] cause the increase of AMF": How can you deduce that? Is it always true or should not it depend on the aerosol / HCHO altitude?

P.12, last sub-section of section 4: Not sure if this is necessary here to repeat the explanations about "best case scenario'.

P.13 29-30: "aerosol layer height is also important to determine AMF". I agree but since no analysis w.r.t this parameter are given before, it is quite hard to understand why the authors write this here...Please clarify.

P. 14, 1-11: Please check what is really useful for the conclusion, and not redundant with the general part also present in the introduction. For example, it is not necessary here to repeat the nature of HCHO, why sun-synchroneous satellites are limited etc...
"constellation of geostationary": first time this notion is introduced. Could you please precise it?

P.14, 19: Would the ratio of hourly AMF to monthly AMF not be more useful (than the ratio of monthly to hourly) to illustrate the variability into HCHO VCDs?

P.14, 32-33: "Our test with the OMI products indicated a possibility that simultaneously measured aerosol products can be used to calculate AMF considering aerosol".
This was illustrated based on the OMI AOT and SSA in the UV, but not about the aerosol layer height. Any future expectations regarding this last variable?

P14, 8-10: The authors mentioned the importance of aerosol height in the boundary layer and to use simultaneous measurements. But no measurements about aerosols in the boundary layer are shown and used here. Where could it come from? Are such measurements available somewhere?

P21, Figure 1: Did you compute and use the vertical averaging kernel to convert the GEOS-Chen trace gas profile into vertical column densities in order to validate your retrievals? How do you compute them and where should they be present in your OSSE diagram?

P23, Figure 3: Could you please also times that are available from geostationary observations but not from sensors like OMI (*i.e.* early in the morning, late in the afternoon)?

P24, Figure 4: please indicate for which time(s) of the day are plotted these retrievals.

P25, Figure 5: The sign of the absolute and relative differences are opposite, and thus the colours are reversed between the columns (i.e. what is red on the left, in absolute, becomes blue on the right in relative...). Please correct this.

P28, Figure 8: The ratio of the 2 AMFs is not strictly equal to the ratio of the 2 VDCs, since these last variables include artefacts due to the spectral fit when deriving the slant column densities. However, it represents the part of AMF computation errors included in the VDC products at the end. Please correct your second statement, in the caption, accordingly.

Additional bibliography recommended

Boersma, K. F., Eskes, H. J., and Brinksma, E. J.: Error analysis for tropospheric NO_2 retrieval from space, J. Geophys. Res., 109, D04311, doi:10.1029/2003JD003962, 2004.

Castellanos, P., Boersma, K. F., Torres, O., and de Haan, J. F.: OMI tropospheric NO_2 air mass factors over South America: effects of biomass burning aerosols, Atmospheric Measurement Techniques, 8, 3831–3849, doi:10.5194/amt-8-3831-2015, http://www.atmos-meas-tech.net/8/3831/2015/, 2015.

Chimot, J., Vlemmix, T., Veefkind, J. P., de Haan, J. F., and Levelt, P. F.: Impact of aerosols on the OMI tropospheric NO_2 retrievals over industrialized regions: how accurate is the aerosol correction of cloud-free scenes *via* a simple cloud model?, Atmospheric Measurement Techniques, 9, 359–382, doi:10.5194/amt-9-359-2016, http://www.atmos-meas-tech.net/9/359/2016/, 2016.

de Smedt, I., Müller, J.-F., Stavrakou, T., van der A, R., Eskes, H., and Van Roozendael, M.: Twelve years of global observations of formaldehyde in the troposphere using GOME and SCIAMACHY sensors, Atmospheric Chemistry and Physics, 8, 4947–4963, doi:10.5194/acp-8-4947-2008, http://www.atmos-chem-phys.net/8/4947/2008/, 2008.

Ma, J. Z., Beirle, S., Jin, J. L., Shaiganfar, R., Yan, P., and Wagner, T.: Tropospheric NO_2 vertical column densities over Beijing: results of the first three years of ground-based MAX-DOAS measurements (2008 and 2011) and satellite validation, Atmospheric Chemistry and Physics, 13, 1547–1567, doi:10.5194/acp-13-1547-2013, http://www.atmos-chem-phys.net/13/1547/2013/, 2013.

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Wang, Y., Beirle, S., lampel, J., Koukouli, M., De Smedt, I., Theys, N., Li, A., Wu, D., Xie, P., Liu, C., Van Roozendael, M., and Wagner, T.: Validation of OMI, GOME-2A and GOME-2B tropospheric NO2, SO2 and HCHO products using MAX-DOAS observations from 2011 to 2014 in Wuxi, China, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-735, in review, 2016.