Responses to Referee's Comments

Thanks again for raising an important issue, to which we wrote our responses in blue and the revised manuscript in red.

Review 2

The authors have substantially modified and improved the original manuscript addressing all my questions and comments. In particular, I am satisfied now to see a better distinction between aerosol optical properties and their vertical distribution impacts on HCHO AMF variability. I thank them for these works.

I have some remaining more minor comments or questions listed below. They mostly concern some needs, on my side, to clarify news statements or analyses written in the updated manuscript.

Main comments:

1) P10 & 11: The analyses now clearly separate and investigate each important parameter affecting the variability of the HCHO AMF: HCHO profile, aerosol profile and aerosol optical properties. However, I feel that a few clarifications should be more emphasized or added to clarify the key messages here. These messages are somehow there but a bit complex to extract or properly summarize. Please, find my own deduced conclusions here below and see whether you (more or less) agree with them and how you can more emphasize in your respective section:

- As you somehow mentioned on p10 l31, impacts of HCHO and aerosol profile are quite correlated. This makes sense as overall, we are looking at the resulting enhancement or shielding effect. What really matters at the end is the relative altitude between HCHO and aerosols, more than the absolute altitude of HCHO or aerosols themselves. Therefore, to properly take into account the variability of aerosols, not only their vertical distribution and optical properties have to be included but also the HCHO profile variability. This is confirmed by the numbers that somehow present same order of magnitude when looking at the differences between the AMFs. This should be properly emphasized.

We partly agree with you that the relative vertical distribution of HCHO and aerosol is a key factor for AMF calculations, but our analysis indicates that aerosol optical properties are also important. We emphasize our argument in the revised manuscript as follows:

In order to examine the factors for a shielding effect ($AMF_h < AMF_m$) and an enhancement effect ($AMF_h > AMF_m$) as shown in blue and red boxes in Fig. 5(a), we plot mean profiles of aerosol and HCHO averaged over the two boxes as shown in Fig. 6. First of all, we find that aerosol profiles considerably differ between monthly and hourly values especially for its peak height, whereas relatively insignificant changes exist for HCHO profiles. The shielding effect appears to be associated with the aerosol layer higher than that of HCHO (Fig. 6(a)) and the enhancement effect is due to the opposite vertical distributions of the two (Fig. 6(b)), which is consistent with the previous studies by Leitão et al. (2010) and Chimot et al. (2016).

Our analysis further reveals the importance of aerosol optical properties especially for the shielding effect shown in the blue box of Fig. 5(a). If the relative vertical distributions of aerosol and HCHO is a single crucial factor for the shielding effect, we should expect a similar magnitude of AMF_h decreases relative to AMF_m for the AMF sensitivity test to aerosol vertical distributions (Fig. 5(d)). In the sensitivity test, we used the same vertical profiles of aerosol (solid) and HCHO (dotted) shown in Fig. 6(a), but the resulting changes of AMF_h in Fig. 5(d) are much smaller relative to the values shown in Fig. 5(c) from the sensitivity test to aerosol optical properties. This is because the sensitivity results shown in Fig. 5(d) were obtained using the monthly mean aerosol SSA (=0.95), which is higher than hourly aerosol SSA (=0.87). In other words, the shielding effect is more pronounced with an absorbing aerosol layer rather than a scattering aerosol layer aloft, which might diminish the shielding effect by increasing a photon path length within or below the aerosol layer by the multiple light

scattering (Dickerson et al., 1997).

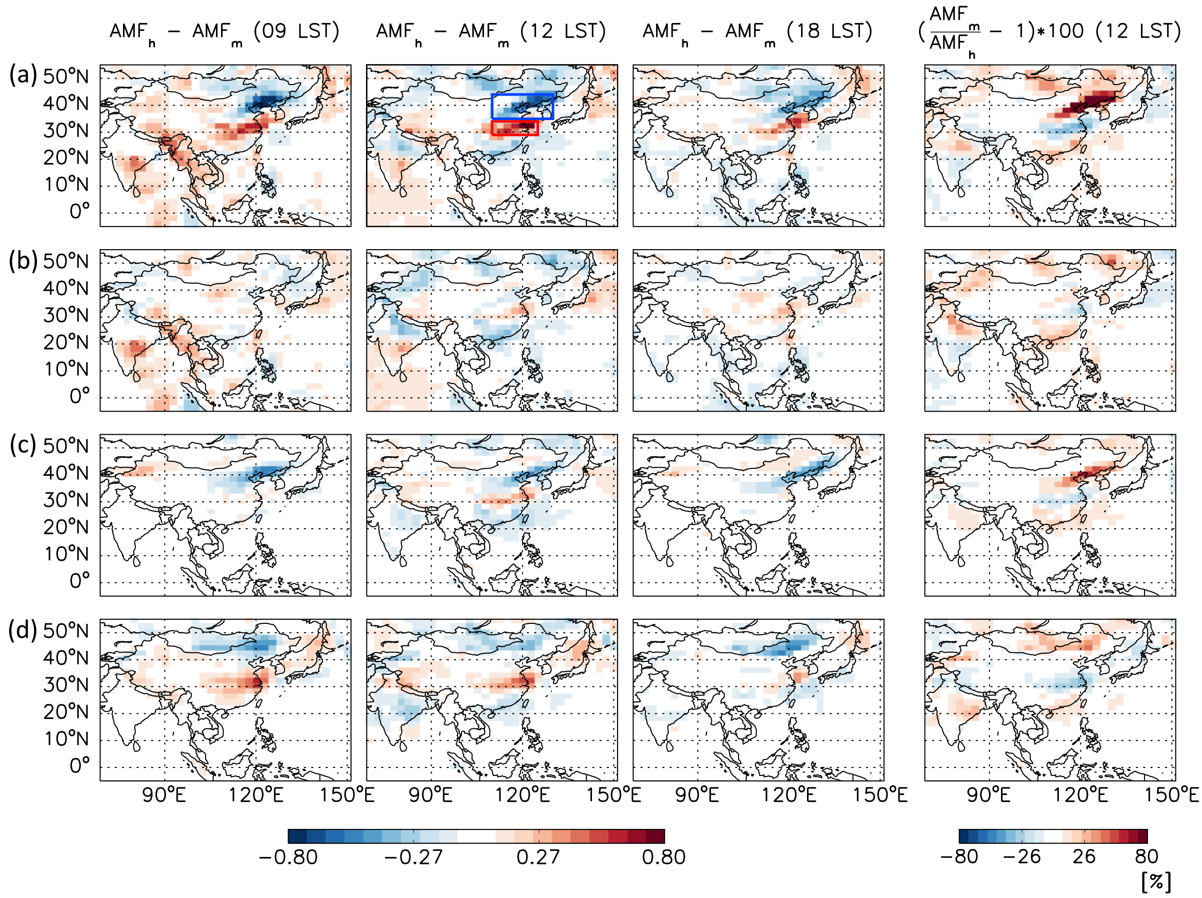

Figure 5. (a) Differences between AMF_h and AMF_m values and relative contributions to them by the temporal changes of (b) HCHO profiles, (c) aerosol optical properties, and (d) aerosol vertical distributions. The first to third columns are results at 9, 12, and 18 LST at Seoul on 21 June 2009. The fourth column gives percentage differences for the ratio of AMF_m to AMF_h indicating changes of HCHO VCDs with AMF_h relative to those with AMF_m at 12 LST. Blue and red boxes denote regions of shielding and enhancement effects.

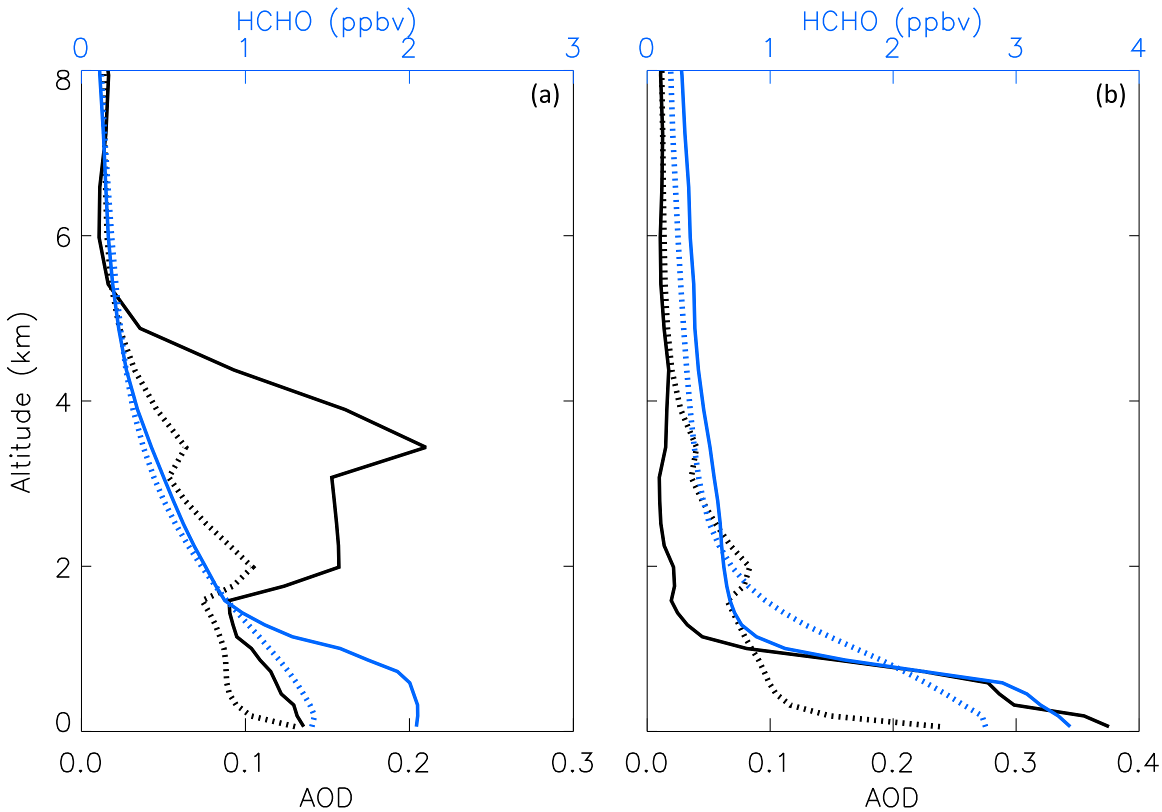

Figure 6. (a) Mean profiles of AOD (black) and HCHO (blue) over a region with decreased AMF_h relative to AMF_m (blue box in Fig. 5(a)). (b) Same as in (a) but for values over a region with increased AMF_h relative to AMF_m (red box in Fig. 5(a)). Solid and dotted lines denote hourly and monthly values, respectively.

- I liked the Figure S1 that you show in your answer (AOD profiles over Eastern China). I think it is really important. Why isn't it in your manuscript? It should be there I believe. Furthermore, following my previous remark, could you add (in black for example) the vertical HCHO profile given by your model? We need to know where HCHO tropospheric bulk is located and the importance of its variability.

Following your suggestion, we added a figure showing vertical profiles of HCHO and aerosols over regions with pronounced AMF_h changes. Corresponding discussion for the figure is shown in our response above.

2) P14, l5-6: I am a bit confused here. Perhaps I am wrong but you said that the ratio of AMF without aerosol to AMF with aerosols increases. But on Fig9 3rd row, we clearly see that most of the area is blue (thus values below 1). Thus, this ratio looks like decreasing for me, not increasing. Am I right? Moreover, Figure 8 shows that the difference AMF aerosol – AMF no aerosol is higher than 0 (red colour). So that confirms that AMF with aerosol is larger. So why do you state or interpret the contrary? By the way, what is the added value here of this Fig8 compared to the Fig9? They both depict the same messages no? I think it is better to keep only the figures focusing on your selected case study (i.e. the days of dust storm).

Yes. You are right. The ratio of AMF without aerosol to AMF with aerosol decreases over most regions of the domain. However, the ratio increases over some regions, where AOD is high and SSA is relatively low, indicating absorbing dust aerosols.
We clarified this in the revised sentence as follows:

The ratio of AMF_{no} to AMF_{a} is less than one over most regions but higher than one over regions with dust aerosols (high AOD and relatively low SSA). The increased AMF_{a} relative to AMF_{no} is a consequence of shielding effects caused by the absorbing dust aerosols.

Figure 9 shows a difference between AMF_{a} and AMF_{no}, and Fig. 10 shows the ratio of AMF_{no} to AMF_{a}, so the results shown in the two figures are consistent. In order to clarify this, we re-plotted Figure 9(a) showing the ratio of AMF_{no} to AMF_{a}.

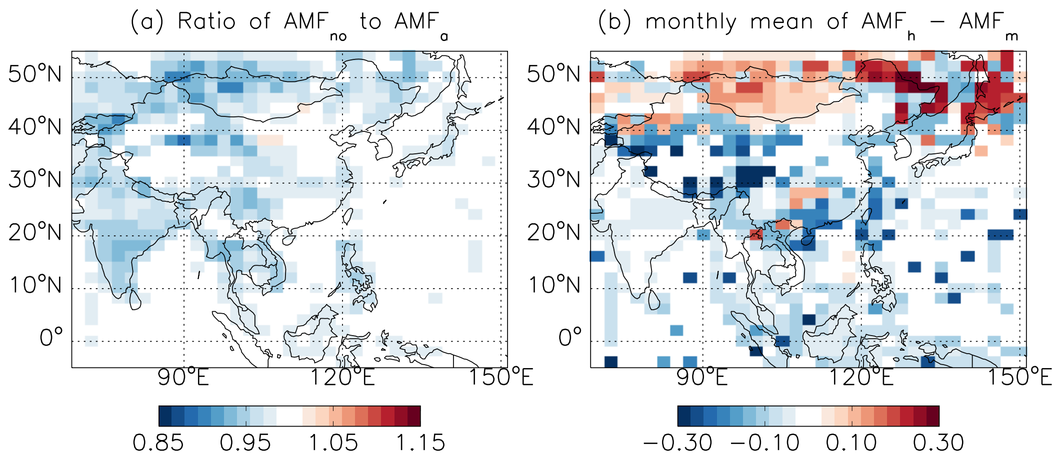

Figure 9. (a) Ratio of AMF without aerosols (AMF_{no}) to AMF with aerosols (AMF_a). (b) Differences of the monthly mean of AMF_h versus AMF_m. AMF_h denotes a value using AOD and SSA at each measurement time, and AMF_m is a value using monthly mean AOD and SSA. Aerosol optical properties used in the calculation are from OMI observations (OMAERUV) for March 2006.

We would like to keep Figure 9 in the revised manuscript because it shows the effects of aerosol on OMI HCHO retrieval, indicating that the present OMI HCHO column concentrations might be biased high on a monthly mean basis. We think that this bias is caused by neglecting the effect of scattering aerosols on the OMI AMF calculation in East Asia. However, in an episodic case of dust storm outbreaks shown in Fig. 10, absorbing dust aerosols have an opposite effect on AMF. Therefore, we would like to keep both figures in the manuscript to make this point.

P14, l10-11: I do not understand how you can see that absorbing aerosols increase VCDs while scattering aerosols decrease VCDs. Which figure does show this? Such a conclusion is not that clearly visible for me on Fig.9 Moreover, what would be the reason from physics point of view. As discussed in my former comments, and as shown in your analyses in the previous section, the key factor that determines shielding or enhancement effect is the relative altitude of HCHO – aerosols. Whether particles are more scattering or more absorbing will mostly drive the magnitude of this shielding or enhancement effect.

The ratio of AMF_{no} to AMF_a indicates change of HCHO VCD because HCHO VCD is inversely proportional to AMF. From the ratio in Fig. 10, we showed that absorbing

aerosols increase VCDs vice versa. We clarified related sentences as follows:

The ratio indicates the change of HCHO VCDs which are inversely proportional to AMF. Therefore, the aerosol effects on AMF make HCHO VCDs increased by 32% due to absorbing aerosols and decreased by 25% due to scattering aerosols compared to those using AMF without aerosols.

We discussed physical reasons related with aerosol optical properties and AMF in our responses above. Also, previous studies (Martin et al., 2003; Lee et al., 2009) referred to the effects of SSA on AMF.

Also, I think you should keep in mind that your computed AMF are based on your GEOS-Chem (average or hourly) aerosol profiles. If you take aerosol profiles from another model or from observations, they may differ and therefore change our AMF values, and perhaps even transform a shielding effect into an enhancement effect (or reciprocally)... Or do you mean, that, on average, scattering particles are usually elevated while absorbing aerosols are more located close to the surface (and thus below HCHO bulk)?
Please clarify here your statement, or provide elements supporting such a principle.

We think that AMF changes do not result from only relative distribution. Aerosol optical properties can lead to shielding and enhancement effects. We discussed related physics above.

Technical comments

1) p2 l4: It should be precised that "aerosol hourly / daily variability uncertainty" cannot be neglected for Geostationary, not simply aerosol variability...

We revised the sentence as follows:

The impact of aerosol temporal variability cannot be neglected for future geostationary observations.

2) p4 l1-2: please check sentence, it is not clear here what you exactly mean (used words are not appropriate I believe).

We revised the sentence as follows:

Here we examine the necessity of temporal AMF for geostationary satellite observations.

p4, l4: "such as HCHO": you mean HCHO profile right? Please clarify
Moreover, you should clearly mention which input parameters you investigate in terms of temporal variations (aerosols profile, optical properties, HCHO profile). So then no ambiguities are left.

We revised the sentence as follows:

We analyze the retrieval sensitivity to AMF calculated with different temporal variations of input parameters such as aerosol optical properties and vertical distributions of HCHO and aerosol.

3) At several places, "achived" should be changed in "achieved". Please correct it thorough the manuscript.

We cannot find the word "achived".

4) p9, l13: "AMFmh changes hourly" => "AMFmh changes every hour"

We corrected it.

p9, l14: "to retrieved HCHO SCDs": Please change "retrieved" into "derived" (or something similar) to avoid to duplicate the word "retrieved" already further written in the same sentence...

We corrected it.

5) p11 l12-14: Chimot et al., (2016) did not specifically investigate the impact of aerosols on HCHO but on NO2. However, the findings there are, I believe, similar to any trace gas in UV-Vis. Quantitatively, numbers may vary of course. Please correct then accordingly.

Thanks. We missed typo. We corrected it as follows:

Chimot et al. (2016) suggested the enhancement (albedo) effect associated with the relative vertical distribution between an absorbing gas and aerosol.

6) p11, l15: "by aerosol backscatter": Please be more specific like for instance "by additional scattering effects, and thus more photons sampling the upper atmospheric layers, due to the presence of aerosols in the observed scene".

We revised the sentence as follows:

HCHO absorptions increase within and above aerosol layers because of an increased photon path length caused by additional aerosol scattering effects, which is referred to as an enhancement (albedo) effect (Chimot et al., 2016).

7) p13 l6: "AMF look-up table is not a function of aerosol layer heights": Perhaps to avoid any confusion for the reader, you should clearly say something like "aerosol layer heights is not an explicit input parameter of the LUT as the HCHO AMF values are based on average aerosol profile given by the GEOS-Chem simulation". On the previous page, you just mention "monthly mean data" but I believe you should explicitly refer to the aerosol profiles.

Thanks for your comments. We clarified the sentence as follows:

However, the information is not yet available from the satellites with ultraviolet and visible channels. Thus, aerosol layer heights are not an explicit input parameter of our AMF look-up table as AMF values are based on monthly averaged aerosol profiles given by the GEOS-Chem simulation.