

We thank the reviewers for the comments that greatly improved the manuscript. Our responses to the reviewer's comments below are highlighted in blue.

### **General comments**

*The subject of the paper, studying the spatial and temporal variations of a priori HCHO profiles and their impact on AMF, is very relevant for current and future satellite retrievals. For their study, the authors used a regional model with a spatial resolution of 4x4km, at three different time of the day. The use of aircraft profiles and LP DOAS measurement to validate the model is giving to the paper an interesting added value to the paper, although their use is limited. However, while the title and the abstract promise to the reader for an evaluation of this resolution impact, the paper does not provide a quantitative answer. I would expect to get an estimate of the errors on AMF when the resolution is decreased in space or in time, with a distinction between both effects. What minimal model resolution is needed to capture the natural resolution of HCHO in the AMF (based on the model)?*

→ Both reviewers suggested to address the impact of spatial resolution in a quantitative manner. In the revised manuscript, we addressed this issue more in a systematic way. First, we compared the AMF from the SAO OMI HCHO retrieval (Gonzalez Abad et al., 2015) with the AMF in this study. In contrast to inhomogeneous AMF in this study, the AMF in the SAO OMI product does not vary much in the domain and is close to 1 (Figure R1 or Figure S3 in Supplementary Material). The average of AMF from the OMI SAO product for the domain (33.5N-34.5N, 117W-118.5W) is 1.12 while the same domain average of AMF from this study is 0.76. If AMF in this study is used, the HCHO column can increase by 47% on the domain-average (up to ~100% at a finer scale), compared with the OMI HCHO column. The vertical HCHO profile in the SAO OMI product is almost a constant in the domain while the model profile at 4 km x 4 km resolution varies substantially. This discussion is included in the revised manuscript P15, L9-P16, L4. As mentioned in the responses to the other reviewer's comments, the operational HCHO retrievals adopted global model results at roughly 1°-3° grid size as a priori profile, which are ~1000 times as large as the spatial resolution in our study (4 km x 4 km). Thus, we used "fine resolution" in the title. Second, we analyzed the effect of spatial resolution on capturing HCHO plumes

in the basin as the reviewer suggested. Figure 6 shows that AMF values are greatly reduced at HCHO mixing ratio of 2, 3 and 4 ppb. We examined the spatial resolutions at which the HCHO plumes of these critical levels of mixing ratio can be captured. The values for coarse grids (8 km – 300 km) are generated from the spatial averages of the original model results at 4 km resolution. Figure R2 and Table R1 (Figure 7 and Table 1 in the revised manuscript) indicate that the grid size  $\leq 12$  km can capture the plumes of HCHO VMR  $> 4$  ppb or 5 ppb by more than 70%. If the grid size is 8 km, the plumes of 1-5 ppb are detected by  $\sim 80\%$ . If the grid size is greater than 100 km, it does not capture the plume of VMR  $> 2$  ppb at this urban location. Thus, the AMF using the coarse resolution  $\geq 100$  km is about 1 because of low concentration that is less than 2 ppb. Currently typical spatial resolution of regional-scale models for the viewing domain of the geostationary satellites (e.g., air quality forecast models for the U.S.) is 12-30 km in each latitude and longitude direction. Our recommendation is to select the resolution as close as 4 km. Since the model simulation at 4 km resolution is computationally expensive for the current geostationary satellite viewing domain and all of high quality input data to the model are not readily available at this resolution (e.g., emission inventory), the model simulations at 8-12 km resolution are recommended to test and improve the model simulations and finally acquire *a priori* profile for next generation environmental geostationary satellite retrievals if computing resources are available. This is included in the revised manuscript P19, L12- P20, L16.

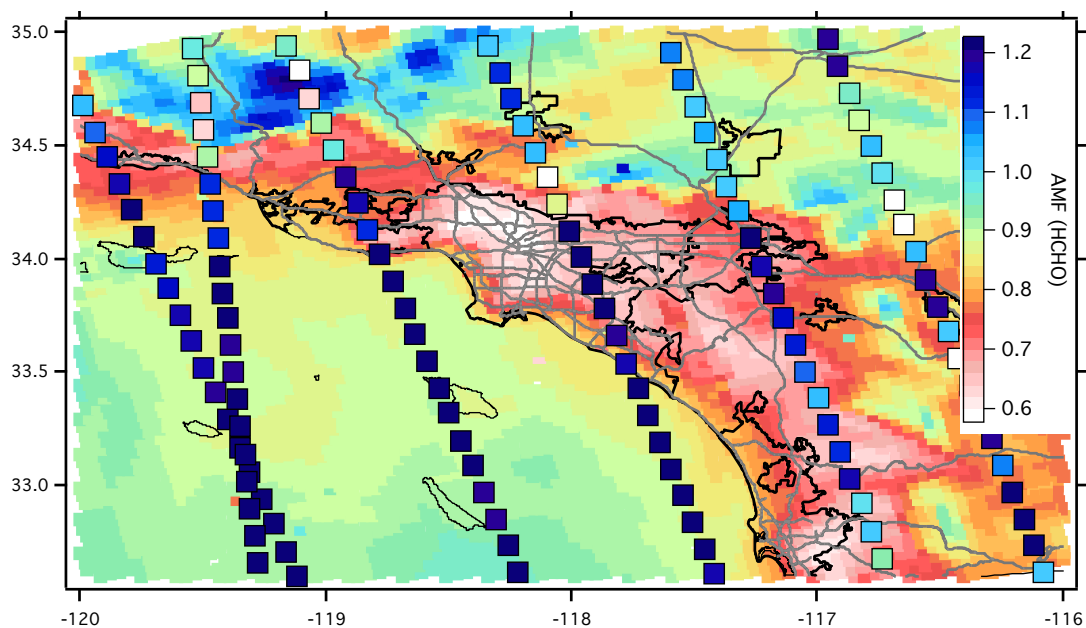


Figure R1. Comparison of the AMF in the OMI operational product (filled square at the center of the OMI swath) with the AMF from this study. An OMI pixel is 24 km x 13 km at nadir and the pixel size increases on either side of this point. The OMI AMF is about 1 on average (blue colors in the color scale used here).

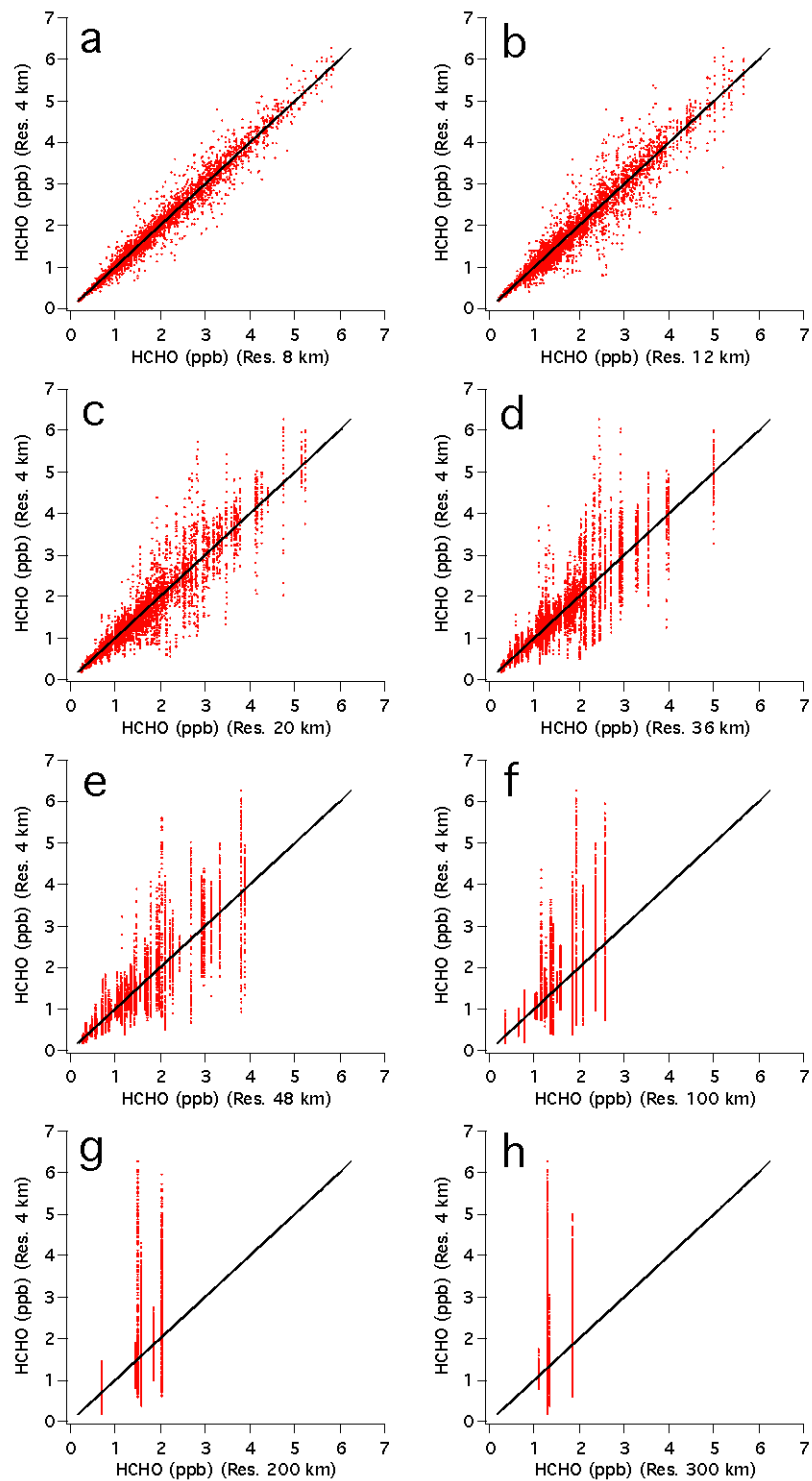


Figure R2. Comparison of HCHO mixing ratios at 4 km x 4 km resolution with mixing ratios at coarser resolutions of (a) 8 km x 8 km, (b) 12 km x 12 km, (c) 20 km x 20 km, (d) 36 km x 36 km, (e) 48 km x 48 km, (f) 100 km x 100 km, (g) 200 km x 200 km, and (h) 300 km x 300 km. The one-to-one line is shown in black.

Table R1. Percentage (%) of intense HCHO plumes retained as the spatial resolution is changed from 4 km. Each column shows the fraction of the plumes retained at coarser resolutions. Here the plume is defined by the area in which the HCHO mixing ratio is greater than the reference HCHO volume mixing ratio (VMR) (1-6 ppb) at 4 km resolution. For example, the second column shows how much area at 8-200 km resolution has a HCHO VMR > 1 ppb when compared with the area with VMR > 1 ppb at 4 km resolution. Similarly, the last column shows how often a model HCHO VMR is greater than 6 ppb at 8-200 km resolution compared with the same plume of VMR > 6 ppb at 4 km resolution; all coarser resolutions (8-200 km) fail to capture this most intense plume. Only model HCHO results at 200 m above ground level at 19 UTC (12 PDT) are used. The areas with HCHO VMRs greater than 1, 2, 3, 4, 5, or 6 ppb are 92800, 29136, 12832, 4256, 848, or 64 km<sup>2</sup>, respectively in the original simulations at 4 km resolution. The area of the domain is 143856 km<sup>2</sup>.

Spatial resolution (km)	Reference HCHO volume mixing ratio (ppb) at 4 km resolution					
	1	2	3	4	5	6
8	98 (%)	95	91	84	79	0
12	97	92	85	73	79	0
20	97	86	72	67	58	0
36	97	82	52	29	0	0
48	96	72	51	0	0	0
100	96	62	0	0	0	0
200	89	56	0	0	0	0
300	53	46	0	0	0	0

*A number of details are missing about how the AMFs are computed beside the a priori profiles?  
Angles, albedo, aerosols?*

→ The missing information is added in the revised manuscript. Solar zenith angles are 52.8°, 16.7°, and 28.8° at 16, 19, 22 UTC, respectively. Relative azimuth angles are 56.6°, 15.5°, 246.1° at 16, 19, 22 UTC, respectively. Viewing zenith angle in the VLIDORT is 46.5°. We assumed a constant surface reflectance of 0.05 across the domain. The AMF presented in the manuscript is selected at 340 nm similar to the current satellite retrieval. This information is included (Page 11, Line 9 – 19) in the revised manuscript.

*I think that the discussion about the shape factor introduce some confusion. I do not agree with the following sentence in the conclusion = “For similar profile shapes, the absolute magnitude of HCHO concentration is also an essential factor in determining the AMF”. The author should clarify the impact of a change at a given altitude, that will modify the shape factor, in opposition to a change at all altitudes (multiplicative factor) that will not modify the shape factor and therefore have no impact on the AMF. See also the detailed comments. I would rather conclude that the AMF are very sensitive to the absolute HCHO mixing ratio in the boundary layer.*

→ We agree. Thank you for your comments. We changed the sentence to “Our study reveals that **the AMF is very sensitive to the absolute HCHO mixing ratio (or number density) in the boundary layer.** Therefore, the absolute magnitude of HCHO concentration in the boundary layer is an essential factor in determining the AMF”.

I recommend publications after these comments have been addressed.

## Detailed comments

*P2, l15: please quantify the statement “can better capture”*

→ We added a quantitative analysis in the sentence. Now it reads “...can better capture the spatial distributions of the enhanced HCHO plumes in an urban area than the nearly constant AMFs used for current operational products **by increasing the columns by ~50% in the domain-average and up to 100% at a finer scale**”.

*P2, l16: This sentence is vague. Which operational product (reference?), what does “nearly constant AMF” mean?*

→ A reference on the SAO OMI HCHO product (Gonzalez Abad et al., 2015) is added. González Abad, G., Liu, X., Chance, K., Wang, H., Kurosu, T. P., and Suleiman, R. (2015), Updated Smithsonian Astrophysical Observatory Ozone Monitoring Instrument (SAO OMI) formaldehyde retrieval, *Atmos. Meas. Tech.*, 8, 19-32, <https://doi.org/10.5194/amt-8-19-2015>.

*P3, l12: please cite Jin, X., Fiore, A. M., Murray, L. T., Valin, L. C., Lamsal, L. N., Duncan, B., Folkert Boersma, K., De Smedt, I., Abad, G. G., Chance, K. and Tonnesen, G. S.: Evaluating a space-based indicator of surface ozone-NO<sub>x</sub>-VOC sensitivity over mid-latitude source regions and application to decadal trends, J. Geophys. Res. Atmos., 439–461, doi:10.1002/2017JD026720, 2017.*

→ Jin et al. (2017) is added in the revised manuscript.

*p4, l6-10: HCHO weak absorption in the UV has an impact on slant column uncertainties. AMF uncertainties do not result from the weak HCHO absorption in the UV. Please clarify.*

→ Agreed. We modified structures of sentences to make the meaning clear. Now it reads as “Because of its weak absorption in the ultraviolet (UV) spectral region, HCHO is regarded as one of the most difficult species to retrieve from satellite-based radiance observations in the UV-visible (UV-VIS) spectral region (e.g., GOME/GOME-2, SCIAMACHY, OMI, and OMPS; see Martin et al., 2003, Zhu et al., 2016 for references). **In addition**, the **large** uncertainties in satellite trace gas retrievals based on UV-VIS spectral measurements arise from the calculation

of the air mass factor (AMF), which converts the slant column density of a trace gas to its vertical column values by considering the vertical sensitivity of the observations (AMF = slant column/vertical column, Palmer et al., 2001; Boersma et al., 2004; Lorente et al., 2017).

**Therefore, it is important to identify factors affecting the accuracy of HCHO retrievals and to find a method to reduce these uncertainties.”**

*P4, l18: add reference to operational products.*

→ Gonzalez Abad et al. (2015) is added in the revised manuscript.

*P4, l16: ..., while the a priori profiles are generally derived from a 3D CTM.*

→ Corrected.

*P4, l18: which operational trace gas products? Please provide reference.*

→ References (Gonzalez Abad et al., 2015; De Smedt et al., 2017) are provided.

De Smedt, I., Theys, N., Yu, H., Danckaert, T., Lerot, C., Compernelle, S., Van Roozendael, M., Richter, A., Hilboll, A., Peters, E., Pedernana, M., Loyola, D., Beirle, S., Wagner, T., Eskes, H., van Geffen, J., Boersma, K. F., and Veeffkind, P.: Algorithm Theoretical Baseline for formaldehyde retrievals from S5P TROPOMI and from the QA4ECV project, Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2017-393>, in review, 2017.

*P5, l3-4: references are mixing satellite retrievals and inverse modelling papers.*

→ We modified the sentence. It now reads as “The HCHO retrievals from existing polar-orbiting satellites were investigated and **utilized** in previous studies...”.

*P5, l5-6: It is not clear what is meant by this sentence “these studies .... used the contrast between land and ocean”. Please add more explanations.*

→ This means that the detailed spatial variations in AMF in the US were not captured. We modified the sentence in the revised manuscript. Now it reads “these studies focused on regions with large biogenic sources or **showed large scale contrasts** between land and ocean.”



*P5, l10: Please provide a reference for TROPOMI.*

→ Veefkind et al. (2012) is added in the revised manuscript.

*P5, l15: Recent model provide a resolution of 1x1°, daily (TM5-MP, TROPOMI)*

→ Now it is changed to “horizontal grid resolutions of 1-3 degrees”.

*P13, l1: Please specify to what quantity 35 % refers to. Total AMF, AMF in a certain altitude range?*

→ It meant a change in total AMF. We clarified it. It is changed to “Global Ozone Monitoring Experiment (GOME) measurements that were ~35% less sensitive to the HCHO column (**or 35% smaller total AMF**) over Tennessee than over the North Pacific.”

*P13, l10: please provide a number (relative differences between cases a and b in figure A1) in order to estimate the “small” impact of surface pressure on AMF*

→ Quantitative analyses are shown in the revised manuscript. Please see our response to the other reviewer (Page 9–Page 11).

*P16, l15-16: I do not agree with this discussion. I completely agree that the AMF anti-correlates with the HCHO mixing ratio in the boundary layer. But if the absolute HCHO values changes in the boundary layer, and not at higher altitudes, this changes the profile shape quite strongly.*

→ Agreed. As suggested by the reviewer, we modified the sentences to “For UV-VIS retrievals, it is **well known** that the vertical profile shape affects the value of the AMF. Our study suggests a strong anti-correlation between the absolute concentration and the AMF: the AMF is low in the area of intense HCHO plumes. The changes in the absolute HCHO concentrations in the boundary layer (altitude AGL < 1-3 km) strongly modify profile shapes, which in turn affect AMF substantially.”

*P23, l 8-9: quantify the improvement*

→ In the revised manuscript, we added quantitative analyses in several places. Therefore, we did not change this general conclusion.

*P 23, l8-8: It would be an interesting conclusion to provide a minimum resolution in time and in space, to reduce the AMF uncertainty under a given threshold (ex 10%).*

→ We suggested minimum resolution based on new analysis in Figure 7 and Table 1 (Figure R2 and Table R1 above). Because the simulations at 4 km resolution for the full domain of geostationary environmental satellite are very expensive, it is recommended to use 8-12 km if computing resources are available. More detailed discussions are added in the revised manuscript. See the responses above.

*Figure 1: Specify the dates in the legend*

→ We specified the dates (May-June 2010) in the legend in Figure 1.

*Figure 3: Please improve the visibility of the colorbar and the inset text.*

→ The visibility of the color bar and the text is improved in the revised manuscript.

*Figure 4: The altitude above ground level is not shown in this figure.*

→ The altitude above ground level (or AGL) is noted wherever needed.

*Figure 7: Please do not use “slope” factor. It introduces confusion. You already use profile shape and shape factor.*

→ “slope” factor is changed to “shape” factor in the manuscript and the figure. It is Figure 8 in the revised manuscript.

*After the paper from Palmer et al. 2001, several papers highlighted the importance of the a priori profile shapes on satellite HCHO retrieval: Barkley et al., 2012; De Smedt et al., 2015; Lorente et al., 2017; Wang et al., 2017.*

*Barkley, M. P., Kurosu, T. P., Chance, K. V, De Smedt, I., Van Roozendael, M., Arneeth, A., Hagberg, D. and Guenther, A. B.: Assessing sources of uncertainty in formaldehyde air mass factors over tropical South America: Implications for top-down isoprene emission estimates, J. Geophys. Res., 117(D13), D13304, doi:10.1029/2011JD016827, 2012.*

De Smedt, I., Stavrou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., Theys, N., Lerot, C., Gielen, C., Vigouroux, C., Hermans, C., Fayt, C., Veeffkind, J. P., Müller, J.-F. and Van Roozendael, M.: Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from combined OMI and GOME-2 observations, *Atmos. Chem. Phys.*, 15(8), 12241–12300, doi:10.5194/acpd-15-12241-2015, 2015.

Lorente, A., Folkert Boersma, K., Yu, H., Dörner, S., Hilboll, A., Richter, A., Liu, M., Lamsal, L. N., Barkley, M., De Smedt, I., Van Roozendael, M., Wang, Y., Wagner, T., Beirle, S., Lin, J.-T., Krotkov, N., Stammes, P., Wang, P., Eskes, H. J., and Krol, M.: Structural uncertainty in air mass factor calculation for NO<sub>2</sub> and HCHO satellite retrievals, *Atmos. Meas. Tech.*, 10, 759-782, <https://doi.org/10.5194/amt-10-759-2017>, 2017.

Wang, Y., Beirle, S., Lampel, J., Koukouli, M., De Smedt, I., Theys, N., Li, A., Wu, D., Xie, P., Liu, C., Van Roozendael, M., Stavrou, T., Müller, J. F. and Wagner, T.: Validation of OMI, GOME-2A and GOME-2B tropospheric NO<sub>2</sub>, SO<sub>2</sub> and HCHO products using MAX-DOAS observations from 2011 to 2014 in Wuxi, China: Investigation of the effects of priori profiles and aerosols on the satellite products, *Atmos. Chem. Phys.*, 17(8), 5007–5033, doi:10.5194/acp-17-5007-2017, 2017

→ Thank you very much for excellent papers. We are glad to include these papers as reference. All of these papers above are referred in the revised manuscript.

### **Reference in the response and newly added in the revised manuscript**

Baidar, S., Oetjen, H., Coburn, S., Dix, B., Ortega, I., Sinreich, R., and Volkamer, R. (2013), The CU Airborne MAX-DOAS instrument: vertical profiling of aerosol extinction and trace gases, *Atmos. Meas. Tech.*, 6, 719-739, <https://doi.org/10.5194/amt-6-719-2013>.

Barkley, M. P., T. P. Kurosu, K. Chance, I. De Smedt, M. V. Roozendael, A. Arneth, D. Hagberg, and A. Guenther (2012), Assessing sources of uncertainty in formaldehyde air mass factors over tropical South America: Implications for top-down isoprene emission estimates, *J. Geophys. Res.-Atmos.*, 117, D13304, doi:10.1029/2011JD016827.

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De Smedt, I., Stavrou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., Theys, N., Lerot, C., Gielen, C., Vigouroux, C., Hermans, C., Fayt, C., Veeffkind, P., Müller, J.-F., and Van Roozendael, M. (2015), Diurnal, seasonal and long-term variations of global formaldehyde

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- De Smedt, I., Theys, N., Yu, H., Danckaert, T., Lerot, C., Compernelle, S., Van Roozendael, M., Richter, A., Hilboll, A., Peters, E., Pedernana, M., Loyola, D., Beirle, S., Wagner, T., Eskes, H., van Geffen, J., Boersma, K. F., and Veefkind, P. (2017), Algorithm Theoretical Baseline for formaldehyde retrievals from S5P TROPOMI and from the QA4ECV project, *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2017-393>, in review.
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- US EPA (2015a), Technical Support Document EPA's 2011 National-scale Air Toxics Assessment, 2011 NATA TSD, <https://www.epa.gov/sites/production/files/2015-12/documents/2011-nata-tsd.pdf>.
- Veefkind, J. P., et al. (2012), TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications. *Remote Sensing of Environment* 120, 70-83.
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