

Response to Reviewers

Comment on acp-2022-292

Referee #1: Ye, Chunxiang

General comments:

This manuscript reported relatively comprehensive measurements of HCHO, HONO, PAN, NO_x, GC-FID/MS VOCs etc in city of Xiamen in Southeast China and presented source appointment analysis of HCHO by PMF model and simulation of oxidative capacity of the atmosphere by chemical model. First of all, I personally for now can not accept the PMF model source appointment of a relative short-lived species of multiple sources, like HCHO. This point surely needs further discussion. For example, the author can provide justification of the method, which is not available in current manuscript. Secondly, scientific motivation and measurement validation, or at least uncertainty analysis are two lost parts in discussion of the chemical model simulation. Otherwise, the result might appear to be of local interests or not robust, which is out of the scope of ACP journal.

Response: We appreciate Professor Ye for the positive comments and helpful suggestions. In the revised manuscript, we have addressed all of the comments, and particularly adopted the suggestion to synthesize our results and compare against existing findings of previous studies. The manuscript has been significantly revised and improved based on these suggestions. For clarity, the reviewer's comments are listed below in black, while our responses and changes in the manuscript are shown in blue and red respectively. We answer your questions about the (1) PMF model and (2) OBM-MCM model.

(1)PMF model

We agree with your idea that PMF model analysis of a relatively short-lived species requires caution, and we are afraid of the brief model introduction may cause confusion. Previous studies showed that photochemical processes could lead to the deviation between the primary and secondary sources of HCHO by the PMF model (Yuan et al., 2012b; Chen et al., 2014). Yuan et al. (2012) proved the capacity of the PMF approach in identifying the role of chemical aging for better understanding the PMF factors. The VOCs emission ratios derived from the PMF fresh factors agreed well with the ones calculated based on photochemical ages, indicating that the PMF approach could identify the contributions from primary emissions reasonably. Additionally, the abundances of NMHCs in the PMF-aged factors could be reproduced by the photochemical aging of fresh factors. In this study, we run PMF models and discussed the apportionment results to investigate whether the PMF approach can separate carbonyl sources well.

To further evaluate the performance of PMF simulation, the relationship between the contribution of one factor to each species and its chemical reactivity (K_{OH}) was analyzed to separate factors that are associated with fresh and aged emissions (Chen et al., 2014; Yuan et al., 2012). If the factor was related to fresh emissions, the distribution of each species would not correlate with its chemical reactivity, the HCHO distribution profile would be similar to that of each species in that factor. As the air mass from a source was aging, the NMHCs underwent photochemical reactions, and the more reactive species would be more largely consumed, while the HCHO distribution profile would be higher than each species in the aged factor due to the secondary production. As a result, a negative correlation could be seen between the aged factor contributions to each NMHC species and their K_{OH} value. Figure 5 showed relationships between the factor contributions to each species and K_{OH} values (representing chemical activities) for species. Indeed, the contributions of HCHO were higher than that of other species in factor 1 of secondary formation (the aged factor in Fig. 5a), but lower than or similar to other species in factor 2, 3, and 5 (the fresh factors in Fig. 5b, c, e). The factor 4 of biogenic source were thought as the aged factor because of the isoprene precursor for HCHO, thus the contributions of HCHO were relatively higher than other species (except isoprene) (Fig. 5d), furtherly confirming that PMF could reasonably identify the contributions of primary and secondary sources of HCHO (Chen et al., 2014). Anymore, the good correlations between the PMF model predicted and observed concentrations of each species were shown in Table S6 in Supplement. The correlation coefficients (R^2) were 0.76 for HCHO, 0.92 for O_3 , 0.89 for 1,2-dichloroethane, and in the range of 0.60–0.99 for NMHC species. The PMF model is a multivariate factor analysis tool that also has been repeatedly applied to the source identification of carbonyls (Zeng et al., 2019; Chen et al., 2014; Guo et al., 2013; Ling et al., 2017; Yuan et al., 2012).

The relevant contents of the justification of the PMF method were added to our manuscript, as follows:

“Yuan et al. (2012) proved the capacity of the PMF approach in identifying the role of chemical aging, and the abundances of NMHCs in the PMF-aged factors could be reproduced by the photochemical aging of fresh factors. Considering the photochemical process impacts of the PMF factors, the relationship between the contribution of one factor to each species and its chemical reactivity (K_{OH}) was analyzed to separate factors that are associated with fresh and aged emissions (Chen et al., 2014; Yuan et al., 2012). The distribution of each species in the fresh factor would not correlate with its chemical reactivity, thus the HCHO distribution profile would be similar to that of each species in that factor. In the aged factors, the NMHCs with more chemical activities would be more largely consumed by photochemical reactions, while the HCHO distribution profile would be higher than each species due to the secondary production. Figure 5 showed relationships between the factor contributions to each species and K_{OH} values (representing chemical activities) for species. Indeed, the contributions of HCHO were higher than that of other species in factor 1 of secondary formation (the aged factor in Fig. 5a), but lower than or similar to other species in factor 2, 3, and 5 (the fresh factors

in Fig. 5b,c, e). The factor 4 of biogenic source were thought as the aged factor because of the isoprene precursor for HCHO, thus the contributions of HCHO were relatively higher than other species (except isoprene) (Fig. 5d), further confirming that PMF reasonably identified the contributions of primary and secondary sources of HCHO (Chen et al., 2014). In this study, we run PMF models and discussed the apportionment results indicating the PMF approach can separate HCHO sources well.”

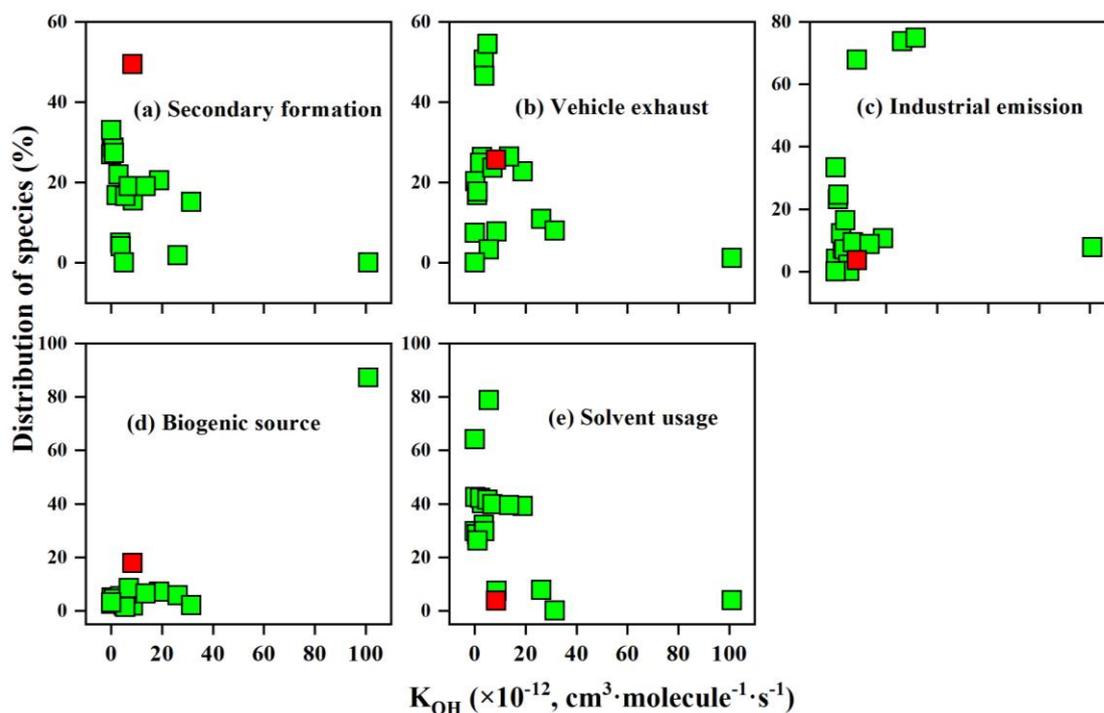


Figure 5. Relationship between the factor contributions to each species and K_{OH} values (representing chemical activities) of the species. Each square represents one species, while HCHO is represented as a square in red.

Table S6. Correlation coefficients (R^2) between observed and simulated concentrations of each species

Species	R^2
O ₃	0.92
ethene	0.90
acetylene	0.84
ethane	0.93
propene	0.86
propane	0.77
iso-butane	0.84
1-butene	0.87
n-butane	0.88
iso-pentane	0.78
n-pentane	0.93
isoprene	0.99
3-methylpentane	0.81
benzene	0.85
toluene	0.86
m/p-xylene	0.60
ethylbenzene	0.69
o-xylene	0.63
formaldehyde	0.76
1,2-dichloroethane	0.89

(2) the chemical model of OBM-MCM

We agree with your idea that measurement validation and model performance evaluation are essential. Figure S1 shows the simulated and observed HCHO at the study site. According to previous studies, the inconsistency between simulated and observed HCHO could be caused by the uncertainties in the treatment of dry deposition, faster vertical transport, uptake of HCHO, atmospheric diffusion/dilution meteorological conditions, and fresh emission of precursor VOCs (Li et al., 2014; Zhang et al., 2021). The differences between the modeled HCHO concentrations and observed concentrations frequently is used to judge the rationality of the model results, and the methods were named the index of agreement (IOA), which were calculated by the equation (Zhang et al., 2021):

$$IOA = 1 - \frac{\sum_{i=1}^n (O_i - S_i)^2}{\sum_{i=1}^n (|O_i - \bar{O}| + |S_i - \bar{O}|)^2} \quad (1)$$

Where, S_i is modeled HCHO value, O_i represents observed HCHO concentration, \bar{O} is the average observed HCHO value, and n is the sample number. The IOA range is 0-1, and the higher the IOA value is, the better agreement between modeled and observed values is. In many studies, IOA ranges from 0.68 to 0.89 (Wang et al., 2018), and the

modeled results are reasonable. The IOAs in spring and autumn in our research are 0.83 and 0.80, respectively. Hence, the modeled and observed HCHO during the observation periods at the study site matched the true situation, and the performance of the OBM-MCM model was reasonably acceptable. Although there is a certain discrepancy, the model could generally reflect the atmospheric chemical processes, and these results still provide valuable information on secondary formation of HCHO at our study site. And this part of the model validation of IOA also has been added to the supplement materials of Text 1.

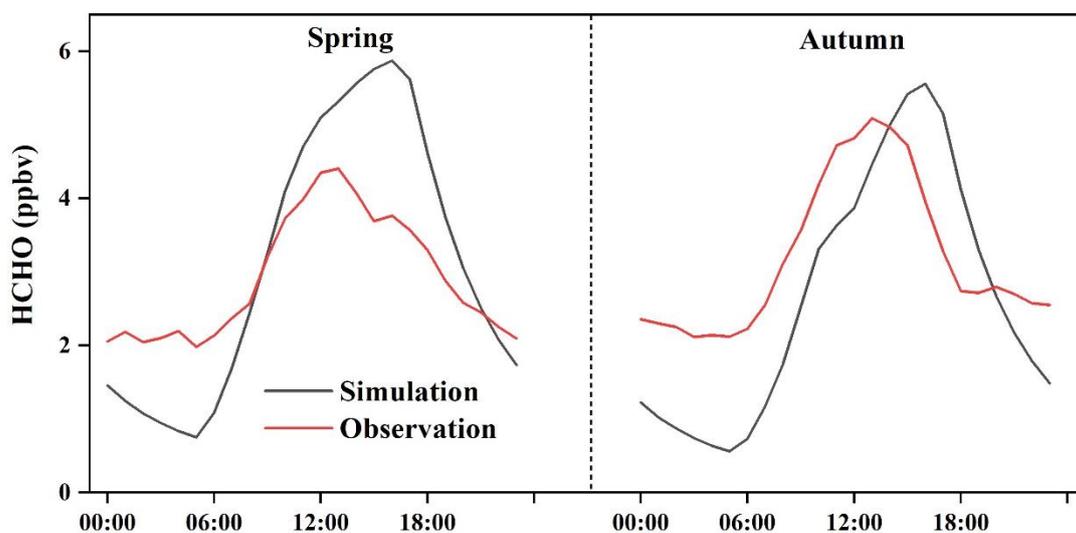


Figure S1. The simulated and observed HCHO at the study site.

Specific comments

I found some puzzled statement. For example,

In line 191-194 The author emphasis experiment design in a coastal area. However, what is special for this area? Any implication for general coastal area from this dataset?

Response: Xiamen is located in the coastal region of Southeast China under the East Asian monsoon control, belonging to the subtropical marine climate (Liu et al., 2020a, b). In spring, north cold airflow and south warm airflow formed the quasi-stationary front causing atmospheric stagnation. In autumn, under the control of the West Pacific subtropical high (WPSH), favorable meteorological conditions enhanced the formation and accumulation of photochemical pollutants (Wu et al., 2020). Meanwhile, the southeastern coastal region is influenced by the East Asian monsoon and acts as an important transport path between the Yangtze River Delta (YRD) and the Pearl River Delta (PRD) (T. Liu et al., 2020a, b). In Xiamen, the concentrations of O₃ precursors were higher than those in remote sites and backgrounds but lower than those in most urban and suburban areas, even lower than those in rural regions (Table R1). Any more,

our previous study showed that atmospheric particles were low in Xiamen (Hu et al., 2022; Deng et al., 2020). Despite the few pollution sources, Xiamen frequently appeared O₃ pollution events in spring and autumn, when the meteorological conditions were governed by weather systems such as the quasi-stationary front and the west pacific subtropical high (Liu et al., 2022a; Wu et al., 2019). The favorable photochemical reaction conditions (including high air temperature, low relative humidity, intense solar radiation, and stagnant atmosphere) provided a good ‘laboratory’ to further explore the HCHO formation mechanism and its impact on O₃ formation.

Table R1. Comparison of NO, NO₂, and total VOCs levels in cities between China and other countries (Units: ppbv).

Location	NO ₂	NO	VOCs	Site category	Observation periods	Reference
Xiamen	9.8	3.7	23.9	Urban	May.-Jun. 2021	This study
Xiamen	8.6	2.2	17.4	Urban	Sep. 2021	This study
Beijing	16.8	2.1	44.2	Urban		
Wuhan	17.5	3.2	30.2	Urban	Summer 2018 (episode)	Liu et al., 2021b
Lanzhou	15.8	2.9	45.3	Urban		
Shanghai	14.2	3.38	25.3	Urban	Jun. 2019 (episode)	Zhu et al., 2020
Chengdu	39.0	3.6	36.0	Urban	Jul. 2017 (episode)	Yang et al., 2020
Los Angeles	-	-	41.3	Urban	May.–Jun. 2010	Warneke et al., 2012
London	-	-	22.1	Urban	1998–2008	Von Sch. et al., 2010
Tokyo	-	-	43.4	Urban	2003–2005	Hoshi et al., 2008
Beijing	11.5	4.8	28.1	Suburban	Aug. 2018	Yang et al., 2021
Hong Kong	25.0	14.0	26.9	Suburban	Aug. to Nov. 2013	Wang et al., 2018
Chengdu	11.4	8.0	28.0	Suburban	Summer 2019	Yang et al., 2021a
Qingdao	16.7	1.6	7.6	Rural	Oct.–Nov. 2019	Liu et al., 2021a
The Pearl River Delta	39.9	4.2	38.0	Rural	Octo.–Nov. 2014	He et al., 2019
Hong Kong	12.2	1.9	10.9	Regional background	Aug.–Dec. 2012	Li et al., 2018
Mt. Wuyi	-	-	4.7	Background	Dec. 2016	Hong et al., 2019
Mt. Tai	-	-	8.8	Background	Jun. 2006	Suthawaree et al., 2010
Mt. Waliguan	-	-	2.6	Remote region	Jul.–Aug. 2003	Xue et al., 2013

Note: “-” means that the data was not mentioned in the relevant studies.

The AOC levels depend on solar radiation, concentrations, and types of air pollutants in different sites. Table R2 shows the precursor concentration, AOC, and JNO₂ levels in Xiamen, Shanghai, and Hong Kong. Due to the characteristics of our study site, although the levels of O₃ precursors in these urban sites were higher than those in Xiamen, the photolysis rates in these cities were lower than those in Xiamen. Hence, the recalculated AOC level in this study was comparable to those at urban sites in Hongkong and Shanghai we mentioned.

Table R2. Comparison of NO, NO₂, total VOCs (ppbv), AOC (molecules cm⁻³ s⁻¹), and J(NO₂) levels in Xiamen, Shanghai, and Hong Kong.

Location	NO ₂	NO	VOCs	Site category	AOC	Maximum AOC	JNO ₂ (10 ⁻³ s ⁻¹)	Maximum JNO ₂	Reference
Xiamen	8.6	2.2	17.4	Urban	6.7×10 ⁷	1.5×10 ⁸	3.4	11.5	This study
Shanghai	14.2	3.4	25.3	Urban	3.9×10 ⁷	1.0×10 ⁸	2.8	8.0	Liu et al., 2020
Hong Kong	-	-	32.7	Urban	6.3×10 ⁷	1.3×10 ⁸	-	6.0	Xue et al., 2016
Hong Kong	12.2	1.9	10.9	Regional background	1.6 × 10 ⁷	6.2 × 10 ⁷	2.3	9.3	Li et al., 2018

Note: “-” means that the data was not mentioned in the relevant studies.

Moreover, the favorable photochemical reaction conditions (including high air temperature, low relative humidity, intense solar radiation, and stagnant atmosphere) in our study site promoted the photochemical reaction of HCHO. Hence, HCHO photochemical rates were higher than those in some urban/suburban sites with similar precursor levels, and comparable to those in some polluted regions, but the daytime average HCHO loss rate was lower than that in these sites (Table R3 and Figure R1). The intensive solar radiation in our study accelerated HCHO loss rate, but relatively low precursor levels limited HCHO secondary formation (Zhang et al., 2021a; Yang et al., 2020). The net HCHO production rate during the daytime (06:00-17:00 LT) in autumn (-0.4 ± 0.7 ppbv h⁻¹) was lower than that in spring (0.1 ± 0.4 ppbv h⁻¹), and a negative net HCHO production indicates that strong photochemical reactions can constrain high HCHO levels in certain situations.

The findings of this study provide significant guidance for emission reduction and regional collaboration for future photochemical pollution control in the relatively clean coastal cities of China and similar countries. We have also revised relevant contents in the manuscript to highlight the characteristics of our findings.

Table R3. Comparison of HCHO (ppbv) and HCHO production and loss rates (ppbv·s⁻¹) in Xiamen, Wuhan, Shanghai, and Hong Kong.

Location	HCHO	Site category	HCHO production rate	HCHO loss rate	Reference
Xiamen	2.9	Urban-spring	1.92	1.88	This study (Fig. R1 a)
	3.2	Urban-autumn	2.01	2.48	
Shanghai	6.7	Suburban	3.3	2.6	Zhang et al., 2021 (Fig. R1 b)
	2.2	Suburban (WJS)	1.2	1.02	
Wuhan	2.1	Urban (ZY)	0.8	0.6	Zeng et al., 2019 (Fig. R1 c)
	3.4	Roadside (HK)	0.33	0.15	
Hong Kong	2.0	Regional background	0.75	0.63	Yang et al., 2020 (Fig. R1 d)

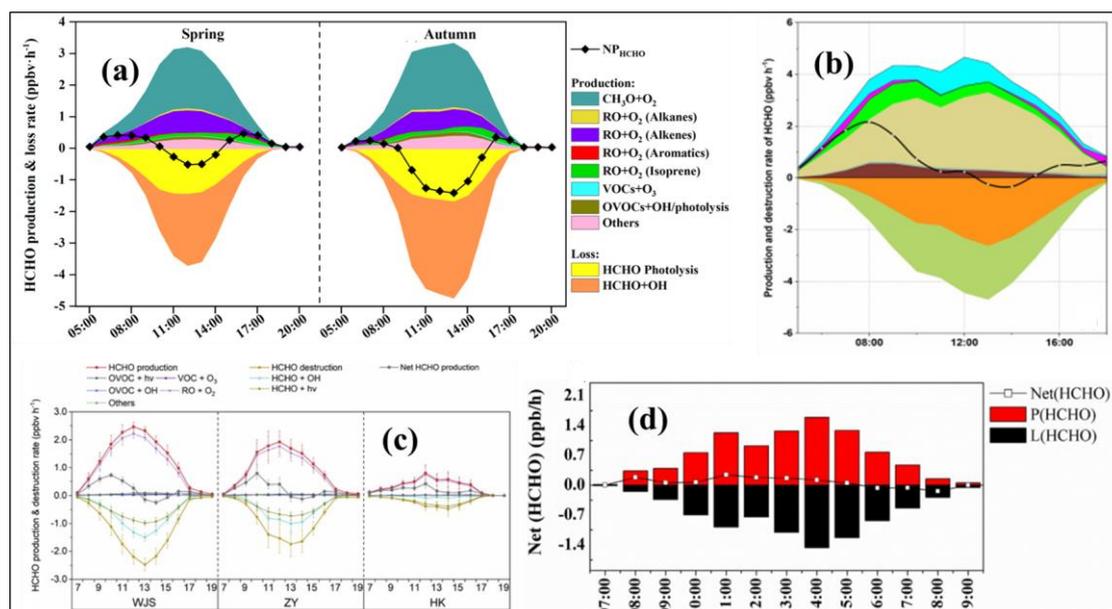


Figure R1. Average production and destruction pathways of HCHO at (a) Xiamen, (b) Shanghai, (c)Wuhan of sites in WJS, ZY, and HK, and (d) Hong Kong. Wujiashan (WJS) (30.64° N, 114.21° E) is a suburban site settled on the top floor (~12m above ground level (a.g.l)) of a high school and surrounded by residential apartments. ZY (30.55° N, 114.30° E) is a typical urban site located in a mixed commercial and residential area in the center of Wuhan. HK (30.60° N, 114.27° E) is a roadside site.

In line 18-19: whether measurements of HCHO is scarce or not?

Response: We are sorry for the confusion caused by the unclear description. Numerous studies have reported on the serious HCHO pollution in China, most of which mainly focused on either its pollution levels or potential sources in the polluted regions of China (Li et al., 2010; Ling et al., 2017; Yang et al., 2017; Yang et al., 2019; Cheng et al., 2014; Guo, 2009; Louie et al., 2013). In recent, limited studies have focused on the formation mechanisms of HCHO in the megacities and regions with rapid economic development or with severe pollution in China, such as Beijing (Liu et al., 2015; Yang et al., 2018) and Pearl River Delta (PRD) (Li et al., 2014; Ling et al., 2017). Yet, to the best of our knowledge, the HCHO budget in the Xiamen region remains unclear currently. In recent years, few studies quantified the HCHO photochemical reactions and the impacts of HCHO on atmospheric photochemistry (Zhang et al, 2021a; Zeng et al, 2019), and the relevant researches are still scarce in southeast coastal areas of China. Different types and sources for HCHO precursors, as well as the influences of weather and meteorological conditions, lead to the complicated secondary formation mechanisms in various regions, thus the exploration of HCHO sources and photochemical effects are very necessary for ozone pollution mitigation by efficient control strategies. To avoid misunderstanding, we have revised the relevant expressions in the manuscript, as follows:

“Yet, the current studies on quantification of HCHO impacts on atmospheric photochemistry in southeast coastal areas remain scarce and unclear,”

I would say the role of HCHO in radical chemistry is relative well accepted. If the author suggest otherwise, what is new in the understanding of this subject from your data?

Response: We thank the reviewer very much for the critical comments which would definitely help us to improve our work.

Firstly, our study is based on a full suite of field observations, then an observation-based photochemical model was adopted as the main research tool. Our observations of multi-parameters were based on the Atmospheric Environment Observation Supersite (AEOS, 24.61° N, 118.06° E), which is an atmospheric environment observation platform with multi-parameter and high temporal resolution, especially the analyzers of HCHO (FMS-100, Focused Photonics Inc., Hangzhou, China), PAN (PANs-1000, Focused Photonics Inc., Hangzhou, China) and HONO (MARGA, ADI 2080, Applikon Analytical B.V., the Netherlands), to find out the distribution law and the characteristics of the important active species that affect the photochemical pollution mechanism of the atmosphere in the study area significantly. Meanwhile, the photolysis frequencies are the driving force and main controlling factor of atmospheric photochemical process. In our study, eight key photolysis frequencies (i.e. J_{HCHO} , $J_{\text{O}^1\text{D}}$, J_{NO_2} , J_{HONO} , $J_{\text{H}_2\text{O}_2}$, and J_{NO_3}) are monitored by a photolysis spectrometer (PFS-100, Focused Photonics Inc., Hangzhou, China) in real-time. Because of lacking systematic field measurement, many previous researchers only used basic data (VOCs, criteria air pollutants of O_3 , NO_x , and CO) to run the model, and the missing active species did not constrain the model or were substituted with parameters (such as HONO was fixed to 0.02 of the observed NO_2 levels), while the photolysis frequencies would be estimated using the Tropospheric Ultraviolet and Visible Radiation (TUV) model (<http://cprm.acom.ucar.edu/Models/TUV/>) (Tan et al., 2019; Jiang et al., 2020). The lack of monitoring values of these important parameters will greatly increase the uncertainty of the model. Hence, the relatively systematic observed data can well optimize and localize the model, which is the advantage of field observation.

Secondly, we just would like to state the rationale of this study, which is to quantify the reaction pathways of pollution mechanism and the contribution of HCHO to photochemistry based on the ‘Known Chemistry’ as well as comprehensive measurements of related species/parameters. We think these results should be helpful for better understanding the atmospheric oxidation chemistry in the relatively clean coastal city of Xiamen. As the answer to your first specific question shown, the southeastern coastal area has a long duration of sunshine throughout the year, intense solar radiation, and fast photochemical conversion rates. The local formation, regional transport, and offshore complex meteorological conditions were necessary conditions for photochemical pollution in our study site with relatively few pollution sources (Liu et al., 2022). In this study, the regional characteristics brought the differences in

photochemistry compared with other regions (such as the AOC and HCHO mechanism of the answer to your second specific question). Overall, these results should be useful for the community to understand the atmospheric chemistry in different metropolitan areas of the world. Xiamen, a typical city with an upward trend in ozone pollution, was selected as the study area to research the distribution of photochemical active species of HCHO and its impact on atmospheric chemistry, which is the advantage of regional characteristics.

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