Comment on acp-2021-948

# RC2 Anonymous Referee #2

The paper entitled "Seasonal characteristics of atmospheric peroxyacetyl nitrate (PAN) 1 in a coastal city 2 of Southeast China: Explanatory factors and photochemical effects" is generally well written, interesting and within the scope of ACP. However, the authors need to address a few minor comments, before the paper is suitable to be published in ACP.

**Response:** Thanks for your valuable comments. We have corrected this manuscript according to your suggestions.

Major comments:

I find the description of the VOC measurements a bit brief. Which classes of VOCs did the authors monitor with their GC and using which columns? It is not clear whether aldehydes and ketones were measured and if so which ones. Figure S5 doesn't discuss OVOCs as ozone precursors. While table S2 includes some OVOCs, it doesn't mention some important compounds like acetaldehyde, MVK, methylglyoxal and methacrolein. Where these compounds measured and if yes how? It appears to me that the authors are assuming that these are primarily secondary compounds that are derived from other primary emissions ethane, propene, isoprene and aromatics (line 57). If they are just model calculated and not measured then this could bias the results. While these compounds, no doubt have photochemical sources, there can also be substantial primary emissions particularly from various types of biomass burning (Pallavi et al. https://doi.org/10.5194/acp-19-15467-2019), industrial sources and, in the case of acetaldehyde also vegetation (Sarkar et al. 2017 https://doi.org/10.5194/acp-17-8129-2017)

**Response:** Thank you for your suggestions. We have added more details of the description of the VOC measurements in the manuscript, as follows. Meanwhile, the detailed compounds of 106 VOCs were shown in Table R1.

"A gas chromatography-mass spectrometer (GC-FID/MS, TH-300B, Wuhan, CN) was used for monitoring the atmospheric VOCs with a 1-hour time resolution. The instrument conducted sampling with a 30 L/min sampling rate, then samples were pre-concentrated by cooling to -160 °C in a cryogenic trap followed by heating to 100 °C, and subsequently transferred to the secondary trap by high-purity helium (He). The flame ionization detector (FID) detected the low-carbon (C2-C5) hydrocarbons by a PLOT (Al<sub>2</sub>O<sub>3</sub>/KCl) column (15  $m \times 0.32 mm \times 6.0 \mu m$ ); the other compounds were quantified using a DB-624 column (60 m × 0.25 mm × 1.4 µm). The instrument system can quantitatively analyze 106 VOCs in the ambient atmosphere, including 29 alkanes, 11 alkenes, one alkyne, 17 aromatics, 35 halogenated hydrocarbons, and 13 OVOCs. The singlepoint calibration was performed every day at 23:00 with the standard mixtures of PAMS and TO15, and multipoint calibration was performed one month. The detection limits of the measured VOCs were in the range of 0.02 ppbv to 0.30 ppbv, and the measurement precision was  $\leq 10\%$ ".

Because the states of calibration gases of aldehydes and ketones are very unstable, thereby it is difficult to

offer calibration gases during observation campaigns in our observation sites. Hence, the nine compounds can not be quantified (Acetaldehyde, Propanal, Crotonaldehyde, Methacrolein, n-butanal, Benzaldehyde, Valeraldehyde, m-Tolualdehyde, Hexanal). However, this phenomenon is common in many studies (Huang et al., 2021; Yang et al., 2019; Liu et al., 2021). Hence, the OVOCs that you mentioned can not be measured. All compounds of 106 VOCs detected by the gas chromatography-mass spectrometer are shown in Table R1. Meanwhile, the observation-based model has high requirements on the quality of the monitor data, and quantities of the pollutants inputting into the OBM model are limited, so VOCs with concentrations below the detection line were removed. Therefore, the VOCs in Table S2 were the all VOCs compounds that we used in the OBM model in our study.

The RIR values in Figure S5 were calculated by the observation data. As said above, some important aldehydes and ketones can not be considered, so the RIRs of OVOCs were underestimated. Hence, the discussion of RIRs of OVOCs is not included. In many relevant studies, their results of RIR also did not consider the OVOCs (Liu et al., 2021; Chen et al., 2020). In the future, we will calculate the RIR of OVOCs, because of our upgraded monitoring station with the OVOCs measurements.

Thank you for your suggestions. We agree with your opinions that the precursors of PAN of OVOCs have both primary and secondary sources (Sinha et al., 2019; Sarkar et al., 2017). When these OVOCs concentrations were not observed, the concentrations could be locally and reasonably calculated by the model. The three modules (initial value settings, observation data input, pre-ran for 2 days before running the OBM model) could constrain the unmeasured compounds reaching a steady-state and make the model localization. Some studies exploring the PAN formation mechanism based on OBM model also did not observe these OVOCs data (Chen et al., 2020, Liu et al., 2021; Xue et al., 2014). Meanwhile, we strongly agree with your idea and realized the importance of OVOCs, and some OVOCs of model calculation without measuring could bias the results. Hence, our team improved the monitoring of OVOCs in October 2021. A more optimized and complete monitoring system is also the future optimization goal of our model. Anymore, the index of agreement (IOA) judging the reliability of the model simulation results showed the performance of the OBM-MCM model was reasonably acceptable, and its detailed calculation is shown in the next question.

Compounds	Detector	Compounds	Detector
ethene	FID	p-ethyltoluene	MS
ethane	FID	1,2,4-trimethylbenzene	MS
propane	FID	1,2,3-trimethylbenzene	MS
iso-butane	FID	m-diethylbenzene	MS
n-butane	FID	p-diethylbenzene	MS
iso-pentane	FID	naphthalene	MS
n-pentane	FID	dichlorodifluoromethane	MS
cyclopentane	FID	chloromethane	MS
propene	FID	1,1,2,2-tetrachloro-Ethane	MS
1-butene	FID	vinylchloride	MS
cis-2-butene	FID	bromomethane	MS
trans-2-butene	FID	chloroethane	MS
1-pentene	FID	trichlorofluoromethane	MS
trans-2-pentene	FID	1,1,2-trichloro-1,2,2-trifluoroethane	MS
cis-2-pentene	FID	carbondisulfide	MS
acetylene	FID	dichloromethane	MS
isoprene	FID	cis-1,2-dichloroethene	MS

### Table R1 The detailed compounds of 106 VOCs and the detectors.

2,2-dimethylbutane	MS	1,1-dichloroethane	MS
2,3-dimethylbutane	MS	trans-1,2-dichloroethene	MS
2-methylpentane	MS	trichloromethane	MS
3-methylpentane	MS	1,1,1-trichloroethane	MS
n-hexane	MS	1,2-dichloroethane	MS
2,4-dimethylpentane	MS	carbon tetrachloride	MS
methylcyclopentane	MS	trichloroethene	MS
cyclohexane	MS	1,2-dichloropropane	MS
2-methylhexane	MS	dichlorobromomethane	MS
2,3-dimethylpentane	MS	cis-1,3-dichloropropene	MS
3-methylhexane	MS	trans-1,3-dichloropropene	MS
2,2,4-trimethylpentane	MS	1,1,2-trichloroethane	MS
n-heptane	MS	dibromochloromethane	MS
methylcyclohexane	MS	tetrachloroethene	MS
2,3,4-trimethylpentane	MS	1,2-ethylenedibromide	MS
2-methylheptane	MS	chlorobenzene	MS
3-methylheptane	MS	tribromomethane	MS
n-octane	MS	Ethane, 1,1,2,2-tetrachloro-	MS
n-nonane	MS	1,3-dichlorobenzene	MS
n-decane	MS	chlorotoluene	MS
n-undecane	MS	1,4-dichlorobenzene	MS
n-dodecane	MS	1,2-dichlorobenzene	MS
1-hexene	MS	1,2,4-trichlorobenzene	MS
1,3-butadiene	MS	1,1,2,3,4,4-hexachloro-1,3-butadiene	MS
1,1-dichloroethene	MS	acrolein	MS
benzene	MS	acetone	MS
toluene	MS	2-butanone	MS
m/p-xylene	MS	2-propanol	MS
ethylbenzene	MS	2-methoxy-2-methylpropane	MS
styrene	MS	vinylacetate	MS
o-xylene	MS	ethylacetate	MS
iso-propylbenzene	MS	tetrahydrofuran	MS
n-propylbenzene	MS	methyl methacrylate	MS
o-ethyltoluene	MS	1,4-dioxane	MS
m-ethyltoluene	MS	4-methyl-2-pentanone	MS
1,2,3-trimethylbenzene	MS	2-hexanone	MS

The authors should include a more detailed description of their measurements different VOC classes and if some VOCs where only calculated as secondary products in the observation-based model (OBM) then this should be mentioned and the corresponding uncertainties should be discussed. If they were directly measure, it would be better to include OVOCS in the analysis of Figure 6.

**Response:** Thank you for your suggestions. We have added more details of the description of the VOC measurements in the manuscript, as the above question shown. The instrument system can quantitatively analyze 106 VOCs in the ambient atmosphere as shown in Table R1, but the observation-based model has high requirements on the quality of monitor data, so VOCs with concentrations below the detection line were removed. Therefore, the VOCs in Table S2 were the all VOCs compounds that we used in the OBM model in our study, and these valid data contained key species used in most relevant studies, which could constrain the

model and realize the localization of the model well (Liu et al., 2021; Chen et al., 2020; Xue et al., 2014).

About Figure 6: Firstly, it is RIRs of first-generation precursors for PAN. Secondly, the RIRs of measured OVOCs were very minor, which could be ignored in Figure 6. And the detailed information of some VOCs was added in the manuscript, as follows:

"The second-generation precursors of PAN of CH<sub>3</sub>CHO and MGLY have both primary and secondary sources, and the other OVOCs are mainly oxidation products of hydrocarbons (Sinha et al., 2019; Sarkar et al., 2017). Consequently, the contribution and importance of first-generation precursors of PAN are necessary to identify to better control photochemical pollution, which will be discussed in the next section".

"During these simulations (except for NO and NO<sub>2</sub>), the model was not constrained by the OVOC measurements considering that these first-generation precursors contribute to PAN production through formation of OVOCs".

About the uncertainties of the model simulation results, the index of agreement (IOA) can be used to judge the reliability of the model simulation results, and the model validation results were added in the supplementary material of Text 1.

## **Text 1 Model validation**

The index of agreement (IOA) can be used to judge the reliability of the model simulation results, and its equation is (Liu et al., 2019):

$$IOA = 1 - \frac{\sum_{i=1}^{n} (O_i - S_i)^2}{\sum_{i=1}^{n} (|O_i - \bar{O}| - |S_i - \bar{O}|)^2}$$
(4)

where *Si* is simulated value, *Oi* represents observed value,  $\overline{O}$  is the average observed values, and n is the sample number. The IOA range is 0-1, and the higher the IOA value is, the better agreement between simulated and observed values is. In many studies, when IOA ranges from 0.68 to 0.90 (Wang et al., 2018), the simulation results are reasonable, and the IOA in our research is 0.88. Hence, the performance of the OBM-MCM model was reasonably acceptable.

Chemicals	Spring		Autumn		Chemicals	Spring		Autumn	
Chemicais	Mean	SD	Mean	SD	Chemicais	Mean	SD	Mean	SD
Alkanes	<b>9.41</b>	5.30	5.47	2.88	Alkyne	1.00	0.55	0.63	0.34
Ethane	2.39	1.02	1.31	0.43	Aromatics	2.71	2.33	1.62	1.15
Propane	2.31	1.29	1.19	0.58	Benzene	0.27	0.14	0.16	0.09
iso-Butane	0.87	0.57	0.52	0.37	Toluene	1.37	1.21	0.85	0.84
n-Butane	1.30	0.94	0.77	0.59	m/p-Xylene	0.53	0.63	0.39	0.32
iso-Pentane	1.15	1.27	0.52	0.44	Ethylbenzene	0.18	0.18	0.09	0.10
n-Pentane	0.44	0.42	0.24	0.21	Styrene	0.09	0.16	0.02	0.04
2,2-Dimethylbutane	0.02	0.02	0.02	0.01	o-Xylene	0.19	0.23	0.04	0.09
2,3-Dimethylbutane	0.05	0.06	0.05	0.05	m-Ethyltoluene	0.02	0.02	0.01	0.01
2-Methylpentane	0.08	0.09	0.05	0.04	1,3,5-Trimethylbenzene	0.01	0.01	0.01	0.01
3-Methylpentane	0.14	0.15	0.06	0.06	p-Ethyltoluene	0.01	0.01	0.01	0.005
n-Hexane	0.20	0.25	0.10	0.20	1,2,4-Trimethylbenzene	0.03	0.05	0.01	0.02
Cyclohexane	0.04	0.04	0.02	0.02	1,2,3-Trimethylbenzene	0.01	0.01	0.01	0.004
2-Methylhexane	0.05	0.06	0.04	0.05	Isoprene (BHC)	0.08	0.14	0.10	<b>0.17</b>
3-Methylhexane	0.08	0.09	0.05	0.08	Halocarbons	2.54	1.27	1.95	0.90
n-Heptane	0.07	0.08	0.05	0.06	Chloromethane	0.51	0.23	0.46	0.18
n-Octane	0.04	0.06	0.09	0.06	Bromomethane	0.06	0.03	0.04	0.02

Table S2. Descriptive statistics of measured VOCs mixing ratios (Units: ppbv).

n-Nonane	0.02	0.01	0.01	0.005	Dichloromethane	1.19	0.81	0.87	0.50
n-Decane	0.01	0.01	0.01	0.01	Trichloromethane	0.07	0.03	0.05	0.02
n-Undecane	0.02	0.02	0.03	0.03	1,2-Dichloroethane	0.51	0.34	0.36	0.22
n-Dodecane	0.12	0.29	0.36	0.84	Trichloroethene	0.02	0.02	0.02	0.01
Alkenes	1.30	0.89	0.85	0.48	1,2-Dichloropropane	0.12	0.13	0.10	0.08
Ethene	0.90	0.65	0.51	0.34	Tetrachloroethene	0.05	0.05	0.04	0.05
Propene	0.20	0.14	0.19	0.11	OVOCs	4.49	1.83	4.17	2.57
1-Butene	0.04	0.03	0.03	0.02	Acrolein	0.06	0.03	0.04	0.02
cis-2-Butene	0.05	0.06	0.03	0.03	Acetone	2.22	0.94	2.21	0.91
trans-2-Butene	0.03	0.06	0.03	0.02	2-Butanone	0.67	0.45	0.50	0.44
1-Pentene	0.02	0.02	0.01	0.01	2-Propanol	0.24	0.31	0.12	0.12
trans-2-Pentene	0.04	0.04	0.04	0.02	2-Methoxy-2-methylpropane	0.24	0.32	0.09	0.09
1,3-Butadiene	0.01	0.02	0.01	0.01	Ethylacetate	1.07	0.83	1.20	1.31

## Minor comments:

Line 28: I could not quite understand what the authors intend to say in the following sentence "Without considering the transformation of peroxyacetyl radical (PA) and PAN, acetaldehyde contributed to the dominant production of PA ( $46\pm4\%$ ), followed by methylglyoxal ( $28\pm3\%$ ) and radical cycling ( $19\pm3\%$ )."

**Response:** Thank you for your suggestions, and we are sorry for the confused expressions. The thermal decomposition of PAN can produce NO<sub>2</sub> and PA radicals (Eq. R1), and PA can quickly react with NO<sub>2</sub> producing PAN (Eq. R2). The ratio of reaction rates of K<sub>1</sub> and K<sub>2</sub> was close to 1, thus the contribution of PAN thermal decomposition to PA was minor. For better understanding, we revised the sentence as "Model simulations revealed that acetaldehyde oxidation (46±4%) contributed to the dominant formation pathway of PA (hence PAN), followed by methylglyoxal oxidation (28±3%) and radical cycling (19±3%)".

The reaction rate of K <sub>1</sub> :	$PAN \rightarrow PA+NO_2$	( <b>R</b> 1)
The reaction rate of K <sub>2</sub> :	$PA+NO_2 \rightarrow PAN$	(R2)

Line 30: Gramar needs to be improved, maybe the following will convey the intended meaning better: "The PAN formation was highly VOC-sensitive, as surplus NOx (compared with VOCs abundance) prevented NOx from being the limiting factor photochemical formation of secondary pollution."

**Response:** Thank you for your suggestions, we strongly agree with your descriptions, and we have revised this sentence as you said.

"The PAN formation was highly VOC-sensitive, as surplus NOx (compared with VOCs abundance) prevented NOx from being the limiting factor photochemical formation of secondary pollution".

Line 32: This sounds like a generic statement "PAN could promote or inhibit O<sub>3</sub> formation under high or low ROx levels, respectively.". It may be more appropriate to target this at the results of the present study "At our site, PAN promoted and inhibited O<sub>3</sub> formation under high and low ROx levels, respectively."

Response: Thank you for your suggestions, your expression was appropriate, and we revised the sentence

accordingly.

"At our site, PAN promoted and inhibited O3 formation under high and low ROx levels, respectively".

Line 36: The authors could be a bit more assertive and specific in highlighting the contribution of their study to the scientific understanding. "Might be helpful" doesn't sound very convincing to me and doesn't specify the main contribution.

**Response:** Thank you for your suggestions. Your suggestions are pretty good, and we have replaced some words and emphasized the main contribution.

"The analysis of PAN formation mechanism and its positive or negative effect on ozone in our study provided scientific insights into photochemical pollution mechanism under various pollution scenarios in coastal areas".

Line 52: Gramar: "is the only formation pathway" instead of "is solely formation pathway"

**Response:** Thank you for your suggestions, we revised the sentence accordingly.

Line 72 Language: "were the most significant contributors" instead of "offered the highest contribution"

**Response:** Thank you for your suggestions, we have revised the expressions accordingly.

Line 74: "Recently, negative and positive impacts of PAN photochemistry on  $O_3$  production were captured under the low and high NOx conditions, respectively." This statement should include a reference to the corresponding study.

**Response:** Thank you for your suggestions, we have added references to the corresponding study.

Zeng, L., Fan, G. J., Lyu, X., Guo, H., Wang, J. L., and Yao, D.: Atmospheric fate of peroxyacetyl nitrate in suburban Hong Kong and its impact on local ozone pollution, Environ Pollut, 10.1016/j.envpol.2019.06.004, 2019.

Liu, Y., Shen, H., Mu, J., Li, H., Chen, T., Yang, J., Jiang, Y., Zhu, Y., Meng, H., Dong, C., Wang, W., and Xue, L.: Formation of peroxyacetyl nitrate (PAN) and its impact on ozone production in the coastal atmosphere of Qingdao, North China, Sci Total Environ, 778, 146265, 10.1016/j.scitotenv.2021.146265, 2021.

Line 99: "was attributed to the downwind region of the downtown (Xiamen island) with densely population " can be simplified to "was downwind of the densely populated downtown region (Xiamen island)"

**Response:** Thank you for your suggestions, we have simplified the description.

Line 100 in my opinion field observation should be plural "field observations were" not singular

Response: Thank you for your suggestions, we agree with your opinion and changed its format into plural.

Line 122 I have never come across the term "ultrasonic atmospherium" before. I believe the correct name would be "weather station with 2D sonic anemometer".

**Response:** Thank you for your suggestions. We are sorry for the wrong usage of the term, and the correct name you mentioned was right. Hence, we changed the name to "weather station with sonic anemometer".

Line 209 please avoid colloquial language "The wind directions in late spring and early autumn were messy due to the season switch." More scientific "During the transition from spring to summer the wind direction fluctuated between ... and ... while during the transition from summer to autumn the wind direction fluctuated from ... to ..."

**Response:** Thank you for your suggestions. We have revised this description as "During the transition from spring to summer the wind direction fluctuated between northwest and southeast while during the transition from summer to autumn the wind direction fluctuated from southeast to northeast".

Line 210, respectively missing at the end of the sentence "The wind rose charts showed that the wind direction frequencies with relatively high wind speed in spring and autumn were southeast wind and northeast wind (Fig. S2), respectively." Also define "high" by inserting a number "(>... m/s)" in brackets.

**Response:** Thank you for your suggestions. We have revised this sentence as "The wind rose charts showed that the wind direction frequencies with relatively high wind speed (>3  $m \cdot s^{-1}$ ) in spring and autumn were southeast wind and northeast wind (Fig. S2), respectively".

In general, the manuscript should be run through a grammar check software before resubmission. It is better to avoid colloquial language and indirect phrases. It is OK to be direct and use simple sentences.

**Response:** We have performed the grammar corrections of our manuscript through grammar check software, and we also invited native speakers in related fields to polish the manuscript. Thanks for your suggestion, simple sentences, and direct phrases make my point clear.

### **Reference:**

Huang, Y., Gao, S., Wu, S., Che, X., Yang, Y., Gu, J., Tan, W., Ruan, D., Xiu, G., and Fu, Q.: Stationary monitoring and source apportionment of VOCs in a chemical industrial park by combining rapid direct-inlet MSs with a GC-FID/MS, Sci Total Environ, 795, 148639, 10.1016/j.scitotenv.2021.148639, 2021.

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