Comments on acp-2022-691

We thank the reviewers for their constructive comments, which we believe have improved our manuscript. To facilitate the review process, our replies are highlighted in blue color font with *blue italicized font* representing text excerpts and ***blue bold italicized font*** representing new additions to the text. Please see our replies to each comment below.

Anonymous Referee #1

General comments This manuscript presented a studying using CMAQ model with HDDM and PMF methods together to identify the contributions of VOCs to O3 at south part of Taiwan. The authors presented their efforts to conduct model simulations at 1km grid resolution to quantify the impacts of each VOC species from 8 different emission sectors, and also defined their own index to indicate the importance of each factor. Air pollution is always an critical issue for densely populated areas with intensive industrial facilities, and it's important to find the most economic-effective control strategy. This study made a contribution by providing indepth investigation of O3 response to emissions changes, which could be an applicable suggestion for air quality management policy makers. The manuscript is well organized with solid method and data, and the results are presented and discussed thoroughly. Therefore I would recommend it to be published with minor revisions. My comments are listed as below.

We thank the reviewer for recognizing an important finding of our work.

Major Comment#1: The objective of this study was to identify the key VOC species and emission sectors to ozone abatement as the manuscript mentioned. So for the simulation period over a specific pollution event, is it able to represent typical ozone pollution conditions over southern Taiwan? Sec2.1 suggested the synoptic weather in fall usually favors ozone pollution due to less precipitation and weak vertical dispersion, but rest of the manuscript were mostly focused on impacts of emission. Therefore it is necessary to clarify if the conclusions of this study were applicable for fall ozone management only, as the driving factors and chemistry may differ in other seasons too. In addition, a brief description of ozone seasonality at southern Taiwan would be helpful to demonstrate how severe it is in fall season.

Response Major Comment #1

The selected case can reasonably represent the typical ozone pollution episodes in southern Taiwan because the synoptic weather pattern of the event features a weak intrusion of Asian continental anticyclone system with no apparent influence from Pacific subtropical high-pressure system (see Figure S1). According to Hsu & Cheng (2019), this synoptic weather pattern is conducive to ozone pollution episodes, typically occurs during the seasonal transition period and has the highest occurrence in October. The ozone seasonality in southern Taiwan is also briefly described. We addressed and clarified accordingly in the revised manuscript on line 114-129.

Line 114-129: ***"The selected case in October 2018 is the seasonal transition period when the summer season is in transition to the winter. The case can reasonably represent the typical ozone pollution conditions during seasonal transition period in Taiwan because the synoptic weather pattern of the event features a weak intrusion of Asian continental anticyclone system which slowly propagated eastward causing the prevailing wind in Taiwan dominated by weak northeasterly (NE) flows due to continental high-pressure peripheral circulation (see Figure S1). Hsu & Cheng (2019) identified six synoptic weather patterns in Taiwan and studied the characteristics of corresponding air pollutants in each pattern using 6 years (2013-2018) daily averaged wind fields and sea-level pressure observed at surface weather stations in Taiwan. Among the six patterns (C1-C6), C4 has the highest mean O_3 concentrations and occurs predominantly in October. It features a weak anticyclone over the Asian continent and the Pacific subtropical high-pressure system does not have an apparent influence in Taiwan, which is similar to our selected case in October 2018. Although the photochemistry is strong in summer season, the seasonal O_3 variation in Taiwan shows that the monthly O_3 concentration is relatively higher during the seasonal transition months (i.e. October) compared to other seasons (Hsu and Cheng, 2019; Chen et al., 2021; Cheng et al., 2015). This is because during the seasonal transition months, when the photochemical reaction is still strong compared to that of the winter months together with the reduced ventilation capability, the O_3 concentration can accumulate (Yen and Chen, 2000; Tsai et al., 2008). (See Figure S2 for ozone seasonality in Taiwan)"***

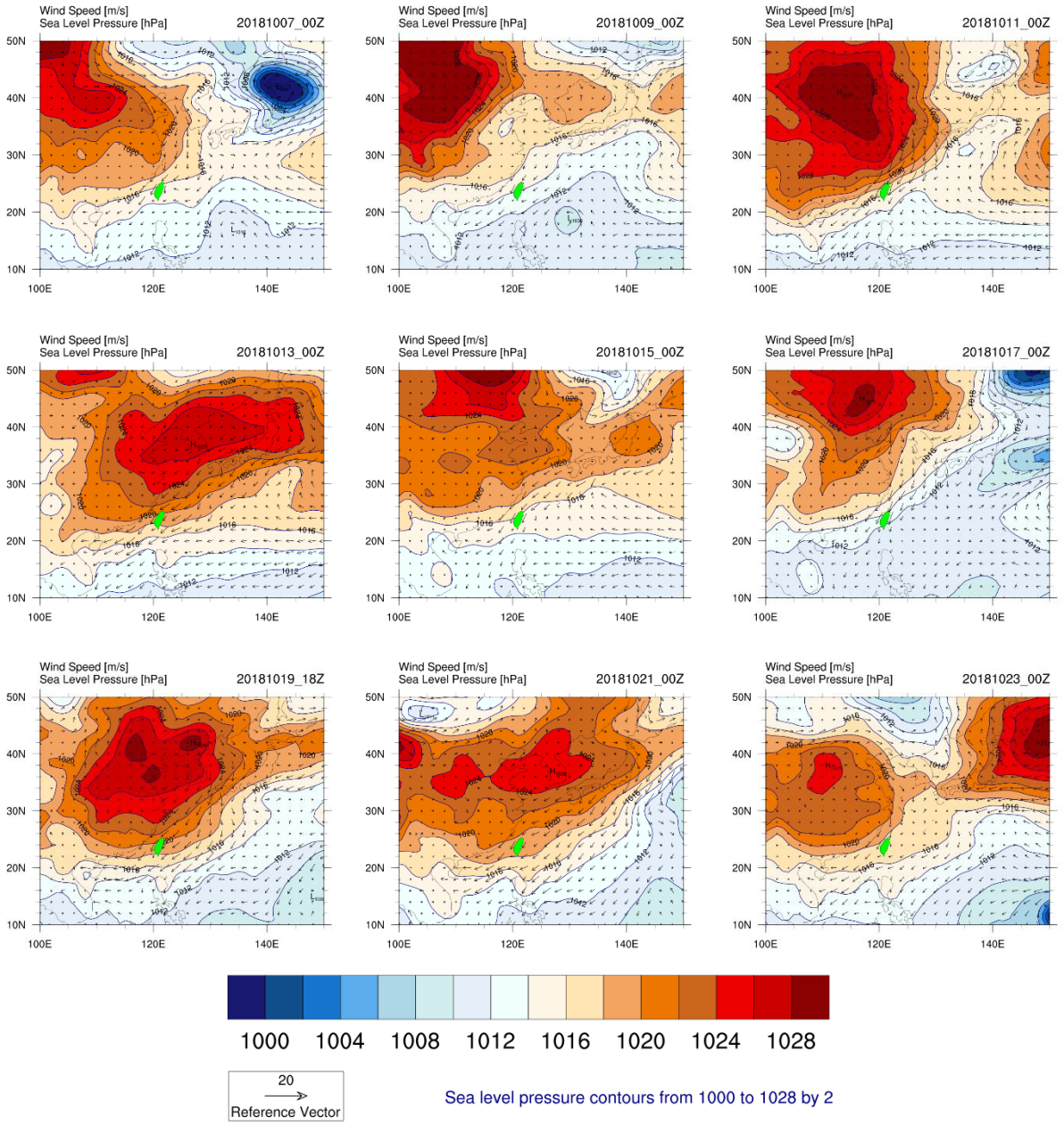

Figure S1: Synoptic weather pattern retrieved from NCEP-FNL reanalysis data valid at 00 UTC from 07 October 2018 to 23 October 2018 showing 850 hPa winds in vector referenced at 20 m s^{-1} and sea level pressure in color contoured from 980 to 1020 hPa by 2 hPa. Taiwan is highlighted with green color.

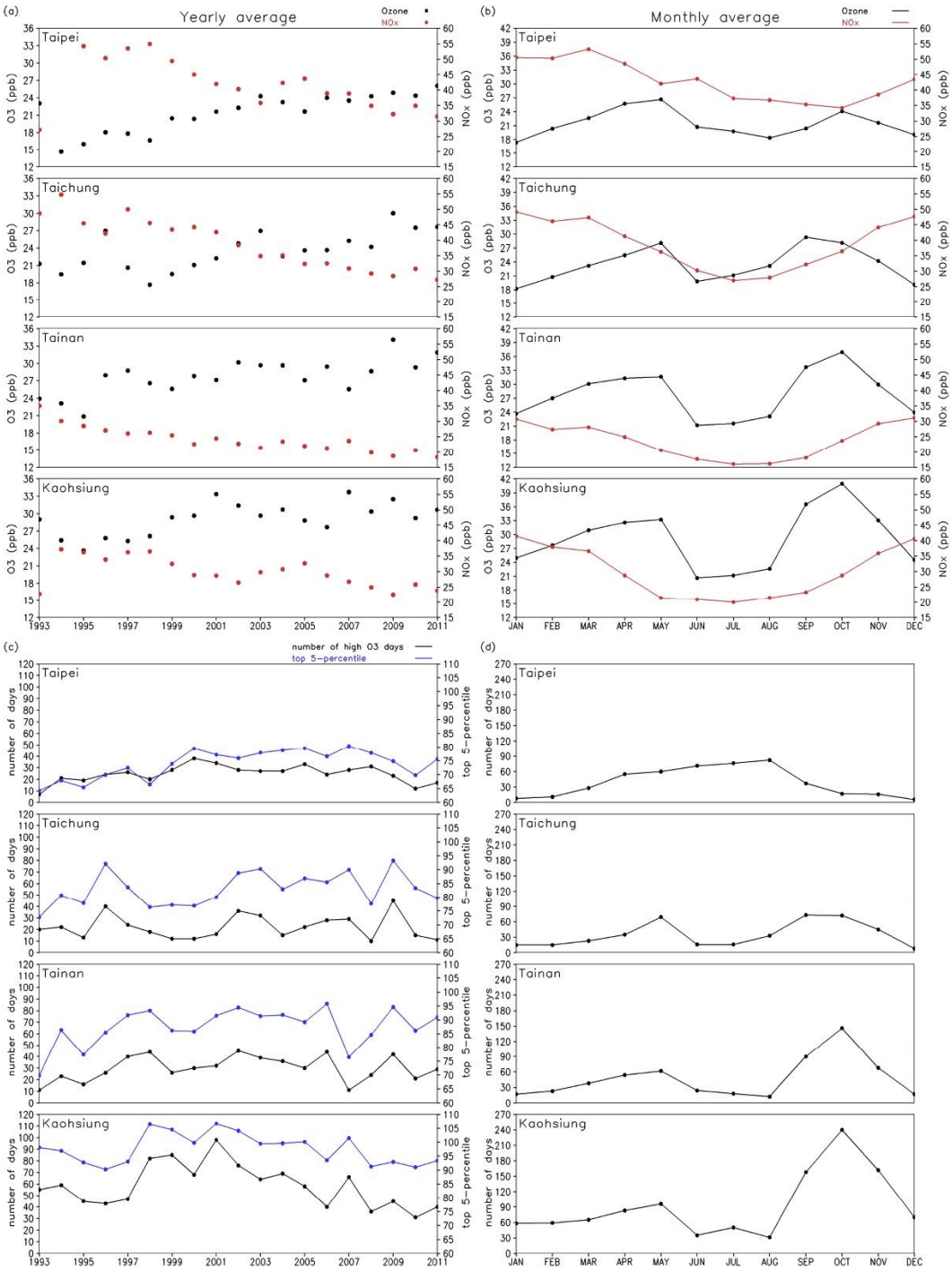

Figure S2. (a) Annual averages of O_3 and NO_x over a 19-year period at the Taipei, Taichung, Tainan and Kaohsiung sites. (b) Same observed data sets as (a) but with O_3 and NO_x presented as monthly variations. (c) Number of high O_3 days (daily maximum O_3 > 100 ppbv) and the average of the top fifth-percentile O_3 concentration in each year. (d) Similar to (c) but presenting the occurrence of days with O_3 > 100 ppbv as monthly variations. Unit for concentrations is in ppbv. Source: (Cheng et al., 2015)

Major Comment#2: There was no description regarding chlorine emission in the manuscript, which could be a very important factor that may change the conclusion of this study, considering a vast part of simulation domain was over ocean. CMAQ was able to handle chlorine-related chemistry since earlier versions but usually it was applied over inland areas without too much attention devoted to oceanic chlorine emission. But for this study the emission from ocean may play an important role in ozone pollution over coastal cities in southern Taiwan.

Response Major Comment #2

Thank you for the comment. The oceanic chlorine emission in our simulation is handled by online CMAQ module which is calculated based on an OCEAN file, whereas the anthropogenic chlorine emission is obtained from TEDS v10 emission inventory. We clarified accordingly in the revised manuscript on line 174-182 and 214-216.

Line 174-182: ***"In our work, the oceanic chlorine emission is calculated online by CMAQ as a function of meteorology using an OCEAN file which specifies the fraction of each grid cell that is open ocean (OPEN) or surf zone (SURF). Figure 2a-c presents the spatial distribution of CMAQ calculated sea-salt aerosol cations (ASEACAT - Na^+, K^+, Ca^{2+} and Mg^{2+}), fine-mode chlorine and coarse-mode chlorine emission rates averaged during the entire simulation period. The sea-spray emissions were higher in the surf zone area and highest emission rates were found along the eastern offshore of southern Taiwan. This is because of the enhanced formation of sea-spray aerosols associated with higher relative humidity and greater offshore winds along the eastern offshore of southern Taiwan that is open to the Western Pacific Ocean. Besides, the anthropogenic chlorine emissions (PCL) are obtained from TEDS v10 emissions, and they are concentrated over the heavily industrialized urban areas of southern Taiwan (see Figure 2d)."***

Line 214-216: ***"The halogen chemistry in CB6 considers chlorine-related reactions such as $ClNO_2$, HCl and HNO_3 production from heterogeneous uptake of N_2O_5 on the aerosol surface, which are important to ozone pollution over coastal cities in southern Taiwan."***

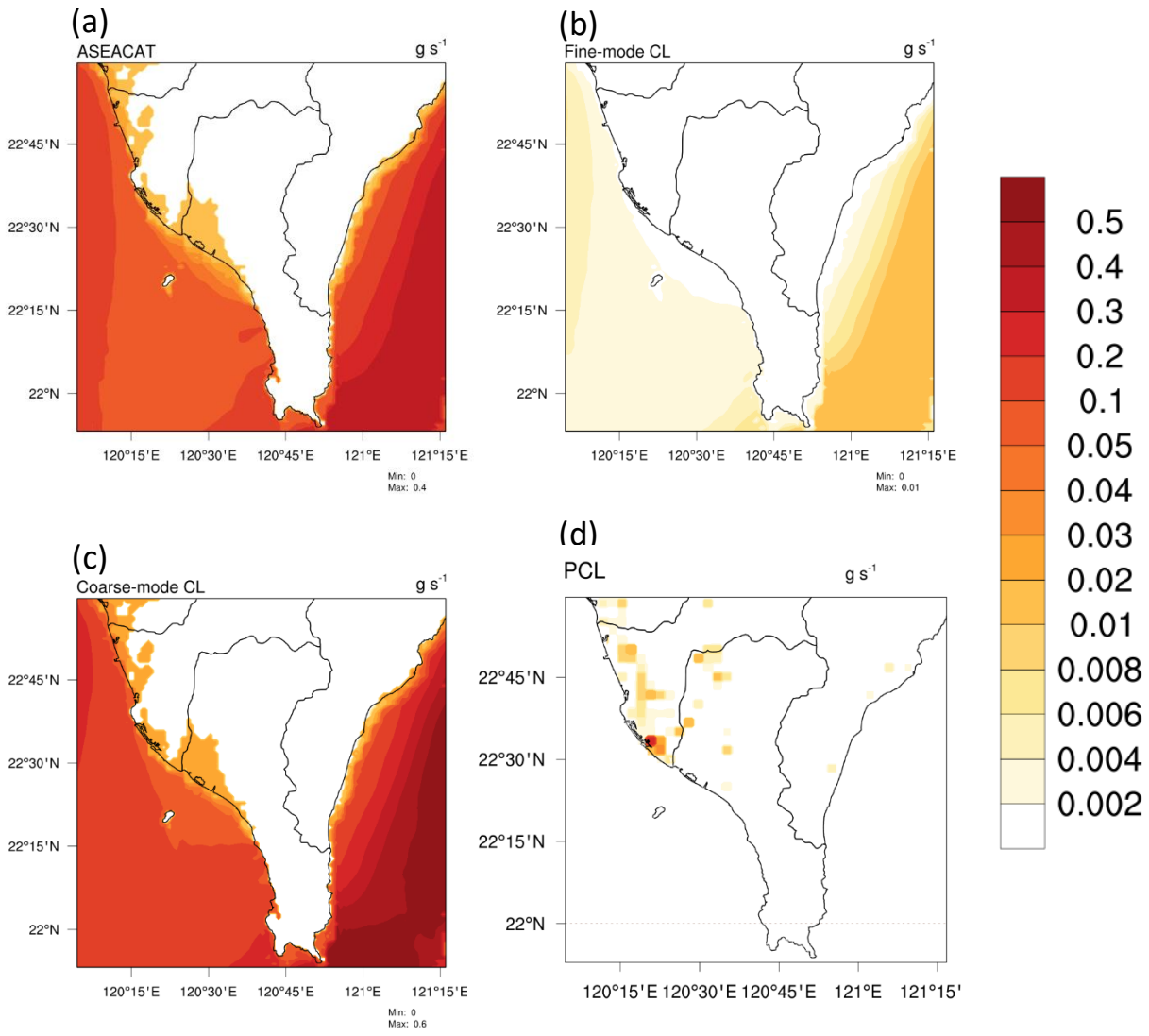

Figure 2: CMAQ calculated (a) sea-salt aerosol cations (ASEACAT) emissions (Na^+, K^+, Ca^{2+} and Mg^{2+}), (b) fine-mode chlorine SSA emissions, (c) coarse-mode chlorine SSA emissions and (d) TEDS v10 anthropogenic chlorine (PCL) emissions averaged during the entire simulation period.

Detailed comments:

(1). Did domain D03 simulation also applied HDDM and PMF? It would be interesting to reveal the differences between D03 and D04 due to grid resolution. 1km simulation is obviously too expensive for large domain, thus it would be helpful to see how well the 3km simulation can capture key factors to ozone formation.

Response Detailed Comment #1

Thank you for the comment. We acknowledge that the differences between D03 and D04 are interesting to reveal, unfortunately HDDM is turned off for domain D01-D03 to save the expensive computation cost. Besides, we also think that this issue deserves a standalone topic to be explored in the future. Therefore, we carefully address this issue as future work in the conclusion.

Line 698-703: ***"In the current work, WRF and CMAQ are resolved at high resolution 1 km to best represent the features of local circulations (i.e land-sea breeze, urban heat island effect) at urban scale. However, simulation at 1 km is obviously too expensive for large domain or regional modelling. The differences in HDDM and PMF analysis between D03 (3 km) and D04 (1 km) due to grid resolution remain an open question which deserves future in-depth investigations."***

(2). line145: Using 2016 TEDS emission is acceptable for 2018 simulation as there shall be no significant difference, but why not simulating a pollution in 2016 instead?

Response Detailed Comment #2

We selected the case 2018 for 2016 TEDS v10 emission because TEDS emission inventory is updated every 3 years, in which the next update is 2019 TEDS v11 emission. At the time of preparing the work, 2016 TEDS v10 emission is the latest emission inventory readily available in gridded format for CMAQ simulation. Therefore, we selected the case 2018 to represent the latest year useable for 2016 TEDS v10 emission. In addition, 2018 case is also more representative of Taiwan local's emissions with less influence from LRT of China's emissions.

(3). Fig.10: Diurnal patterns of factor contribution are very different between sectors, for example, biogenic and motorcycle exhausts. Please provide a brief discussion.

Response Detailed Comment #3

Thank you for the comment. Please see line 580-588 for the additional brief discussion.

Line 580-588: *"**The diurnal pattern of factor distribution of biogenic sector displays a clear diurnal cycle peak at the noontime 10-12 LST in all three stations. Meanwhile, the motor exhaust and vehicle emissions sector peak during the morning and evening traffic rush hours. The aged airmass sector is only identified in CZ and its hourly factor contribution mostly existed at more stable levels when compared to other factors except for an obvious peak at 12 LST due to the sea-breeze penetration that pushes the urban polluted aged air mass towards inland area. The diurnal pattern related to industrial activity exhibits different peak hours depending on sector and station. For instance, the hourly factor contribution of solvent usage in XG has a clear bimodal peak at 10 and 16 LST; plastic industry in QT and CZ has a clear diurnal cycle peak in the noon time at 11 and 13 LST; mixed industry in QT and XG has a clear bimodal peak at 07 and 18 LST.**"*

(4). Was nudging turned on for WRF? Please clarify it in the manuscript.

Response Detailed Comment #4

Yes, grid nudging was turned on for WRF in the coarse domain D01 and D02 while observation nudging was turned on in the fine domain D03 and D04. Details of the nudging technique are clarified in the revised manuscript on line 198-207. Please see below revision.

Line 198-207: *"**To obtain more accurate dynamical downscaling, grid nudging was applied in the coarse domain D01 and D02; observation nudging was applied in the fine domain D03 and D04. Grid nudging is applied to the horizontal wind components, potential temperature, and water vapor mixing ratio; it is only applied above the PBL. The observational data for observation nudging include hourly surface observations such as atmospheric pressure,**

air temperature, relative humidity, wind speed and wind direction from 36 surface meteorological stations (https://www.epa.gov.tw/), and the twice-daily at 00:00 and 12:00 UTC sounding data such as potential height, temperature, dew point temperature, RH, wind direction, wind speed at each specified isobaric level from 2 radiosonde observation stations in Taiwan. The nudging coefficients, which determine the strength of the assimilation tendency term were set to be 6×10^{-4} for observation nudging and 3×10^{-4} for grid nudging. These values of coefficients were recommended by the WRF user guide and tested to be appropriate in previous studies (Li et al., 2022; Borge et al., 2008)."

(5). "DDM 3D" on line 47, "GCE" and "RRTM" on line 173 need clarifications.

Response Detailed Comment #5

Revised.

Line 47: "…***Decoupled Direct Method in Three Dimensions*** *(DDM-3D)…*"

Line 227-228: *"Also used were the* ***Goddard Cumulus Ensemble (GCE)*** *microphysics scheme (Tao et al., 2003),* ***Rapid Radiative Transfer Model (RRTM)*** *longwave radiation scheme (Gallus and Bresch, 2006) …"*

(6). line 114-115 needs references.

Response Detailed Comment #6

Revised.

Line 130: *"During the seasonal transition period in autumn, southern Taiwan often suffers from high O3 episodes* ***(Hsu and Cheng, 2019; Chen et al., 2004, 2021)****."*

(7). line112 "Oct" should be "Oct.".

Response Detailed Comment #7

Revised and all other occurrences are also revised accordingly.

Line 113: *"In this study, we selected the period from 07-20* ***Oct.*** *2018…"*

Line 137: *"A 5-day spin-up period (02-06* ***Oct.*** *2018) was discarded…"*

(8). line266: Please clarify how was the threshold 0.5 determined

Response Detailed Comment #8

This threshold value 0.5 was determined according to the EPA PMF v5.0 user guide. Please see the clarification below.

Line 330-332: "***This threshold value 0.5 was determined according to the EPA PMF v5.0 user guide and also recommended by other PMF studies (Rajput et al., 2016; Reff et al., 2007)".***

(9). Please using consistent denotes for models. Line134 suggested using CMAQv5.2.1, while line219 indicated using v5.2.

Response Detailed Comment #9

Revised.

Line 287: *"In this study, CMAQ-HDDM-3D* ***v5.2.1*** *is used to..."*

(10). Was HDDM approach only used in the experiments S3~5 when considering higher order sensitivity, while DDM approach is used for the rest experiments and in conjunction with PMF analysis? If so, please clarify these details in the manuscript.

Response Detailed Comment #10

Yes, we clarified accordingly in the revised manuscript on line 294-296.

Line 294-296: *"**Noted that HDDM approach was only used in experiment S3-S5 which involves calculation of higher order sensitivity, while DDM approach was used in all other experiments and in conjunction with PMF analysis**."*

(11). The spatial contribution of the O3 sensitivity to each VOCs component is related to the spatial pattern of each VOCs emissions as discussed on 413-416. Please include the several plots to show the spatial pattern of VOC emissions in the supporting information

Response Detailed Comment #11

Thank you for the comment. We agree that the spatial distribution of O_3 sensitivity is related to the spatial distribution of each VOCs emissions. We included several plots of the spatial distribution of some highly sensitive VOC component emissions (i.e. XYL, OLE, PAR, ETH, TOL, IOL) in the supplementary material.

Line 483-486: ***"To render a clearer picture on the spatial distribution of O_3 sensitivity to each VOCs component, the spatial distribution of some highly sensitive VOC component emissions (i.e. XYL, OLE, PAR, ETH, TOL, IOL) are provided in the supplementary material (see Figure S7)."***

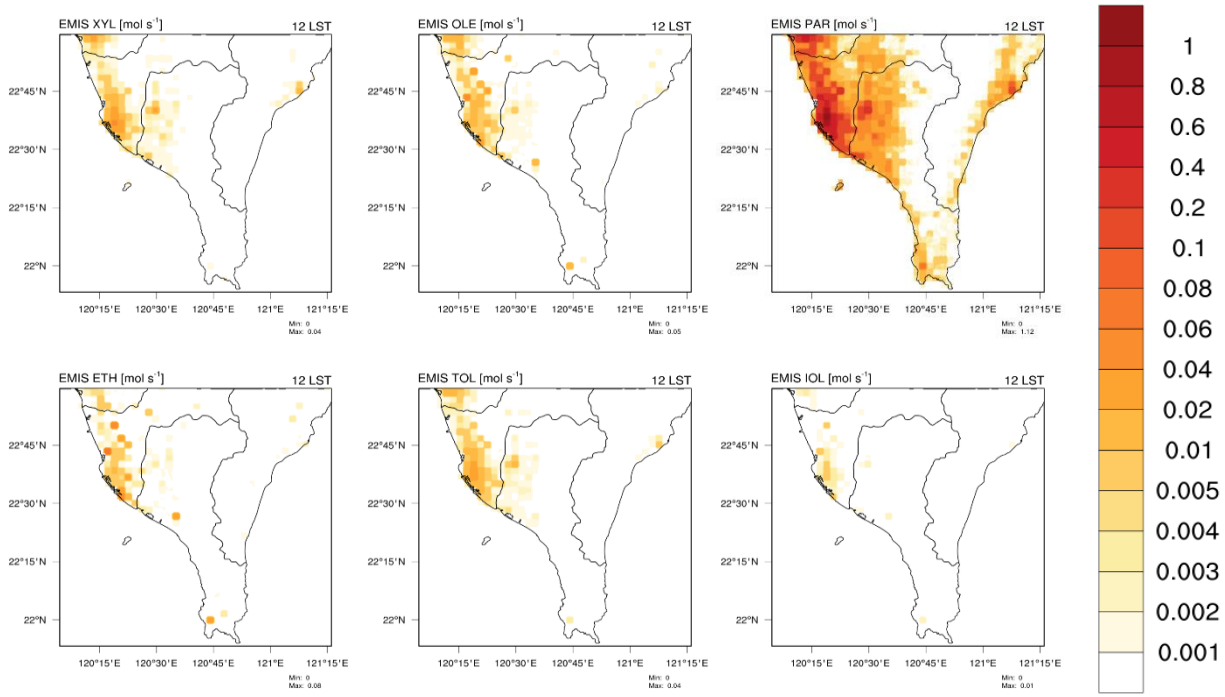

Figure S7: Spatial distribution of some highly sensitive VOC component emissions (i.e. XYL, OLE, PAR, ETH, TOL, IOL) at 12 LST.

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