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 #2

The manuscript utilizes CMAQ model with HDDM and PMF methods to study the contributions of VOCs and O3 in Southern Taiwan. The High-resolution HDDM with indepth analysis of VOCs and NOx relationship on ozone formation has been presented. The article is well-written. I would recommend it with minor revisions. See my comments below.

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

Comments#1 Some discussion on the contribution of long-range transport or from a non-Taiwan background contribution should be included to allow readers to better understand the air quality situation in Taiwan. As the selected case is in October, a large influence may be exhibited from non-Taiwan sources (e.g., Chinese emissions). Moreover, the recent reduction in China may contribute more to Taiwan than the local reduction. Therefore, it is worth including some discussion.

Response Comment #1

Thank you for the comment. We acknowledge the substantial contribution of long-range transport to air quality in northern and central Taiwan especially under strong northeasterly wind conditions. However, the contribution of long-range transport is relatively smaller in southern Taiwan due to the geographic blocking. For clarity, we included a brief discussion pertaining to this issue in the revised manuscript.

Line 164-172: ***"Other than local anthropogenic emissions, the contribution of long-range transport (LRT) from East Asia (e.g. Chinese emissions) is also substantial to Taiwan's air quality especially under strong northeasterly winds condition (Wu and Huang, 2021; Chang et al., 2022a). Lin et al. (2004) identified three types of common LRT events in Taiwan: (1) dust storm (DS), LRT with pollutants (frontal pollution; FP), and (3) LRT of background airmasses (BG). When there is no frontal system, local pollution (LP) dominates the air quality in Taiwan. During wintertime and springtime, the occurrence of LP cases were 70% and about 30% were LRT cases (Lin et al., 2004). Lin et al. (2005) estimated that the long-range transport of pollutants contributes to about 30 μg m^{-3}, 230 ppb and 0.5 ppb to the PM_{10}, CO, and SO_2 concentrations, respectively, in northern and eastern Taiwan. Meanwhile a smaller contribution is estimated in southern Taiwan due to the geographic (Lin et al., 2005)".***

Comments#2 The definition of "urban" and "inland rural area" was not clearly defined. The location or boundary was not clear. The general meteorological pattern (i.e., wind direction) for the event and topology (terrain location) were unclear. Although it has been presented in tables, it is hard for readers to understand the general transport pattern between urban and inland rural areas (e.g., the Influence of sea breeze, or terrain on wind speed and direction). They are important transport mechanisms influencing O3 formation locally. The authors used abbreviations (e.g., XG, CZ etc.), but it wasn't labelled in the figures. It makes it difficult to understand. It is suggested to mark it directly in the figures (e.g., Fig 3, 4 and 6, *S4, S10*).

Response Comment #2

We apologized for the confusion on the definition of "urban" and "inland" area, which was not clearly defined in the previous manuscript. We clarified accordingly in Section 2.2 WRF-CMAQ Model Configuration. For the general meteorological pattern of the event, we addressed accordingly in an additional paragraph in Section 2.1 Study Period & Area on line 238-248. The paragraph also briefly describes the transport mechanism of urban O_3 by local circulations (i.e. land-sea breeze, wind speed and wind direction) with an additional Figure 3. The topography of the innermost domain is also shown in additional Figure 2. After revision, the abbreviations (e.g., XG, CZ, and QT) are now labelled in Fig 5, 6 and 8, S6, S13. Please see the revised figures below.

Line 238-248: ***"The "urban" and "inland" grid cells are defined according to the USGS-24 Land Use Category. "Urban" area is represented by Class 1 - Urban and Built-up Land, which we further classified into Class 31, 32, 33 (see Figure 1b) for WRF Single-Layer Urban Canopy Scheme (SLUCM) simulation. We refer the readers to our previous work for detailed discussion on the land use classification and SLUCM implementation in Chang et al. (2022b). "Inland" area is represented by Class 6 – Cropland/Woodland Mosaic (see Figure 1b). The general meteorological pattern of the event features a weak intrusion of Asian continental anticyclone system which slowly propagated eastward causing the prevailing wind at synoptic scale in Taiwan dominated by weak northeasterly (NE) flows due to continental high-pressure peripheral circulation (see Figure S1). At local scale in southern Taiwan, the steering of weak NE flows by the orographic effect of the Central Mountain Range (CMR) enhanced the local circulations (i.e. land-sea breeze), and eventually pushed the locally produced urban O_3 as well as its precursors NO_x and NMHC towards the inland areas (see Figure 3)."***

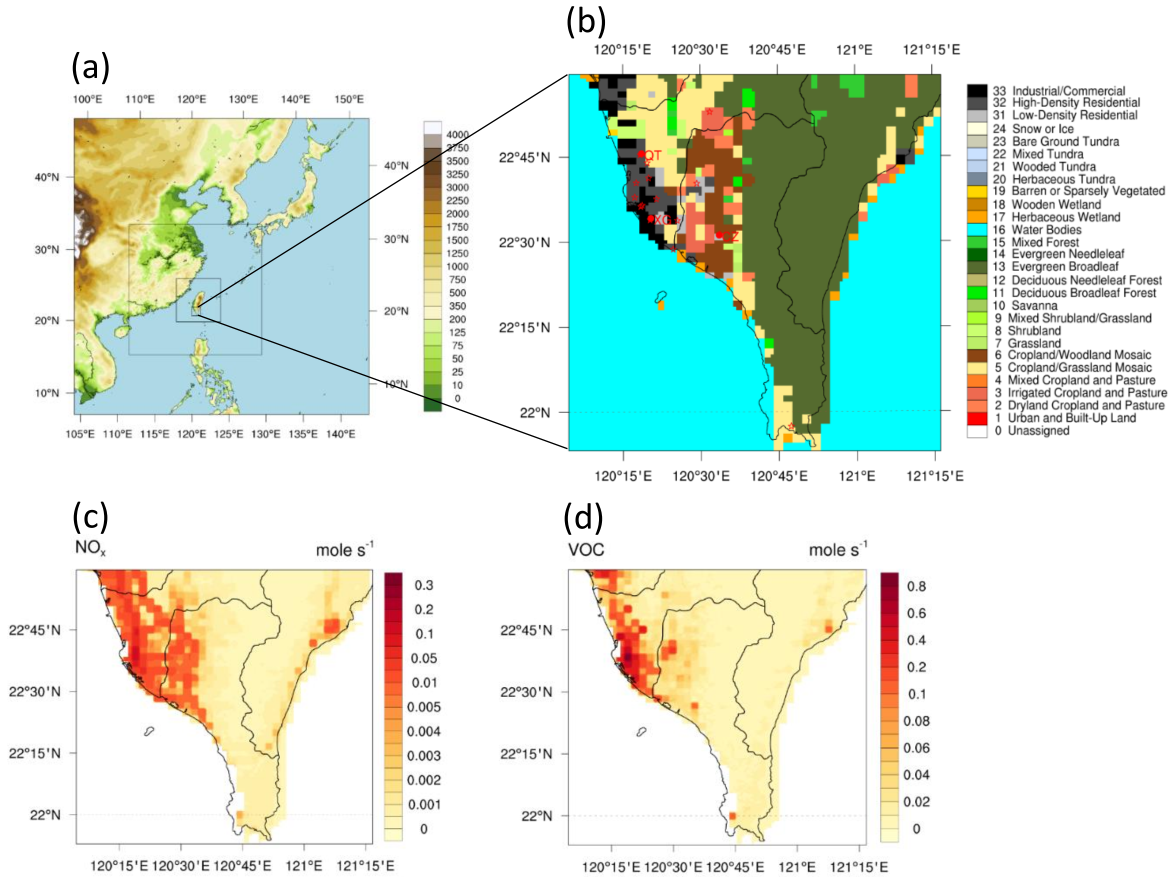

Figure 1: (a) Domain configuration of four-nested grid system, (b) land use of the innermost domain; ***"urban" and "inland" areas are represented by Class 31, 32, 33 and Class 6, respectively****, (c,d) monthly averaged NO_x and VOC emissions in the innermost domain obtained from 2016 TEDS-10 emission inventory. The location of each TEPA air quality stations (red stars) and PAMS stations (red dots with label) used in the current study are displayed in the innermost domain. Refer Figure S1 and Table S3 for details of each TEPA and PAMS station.*

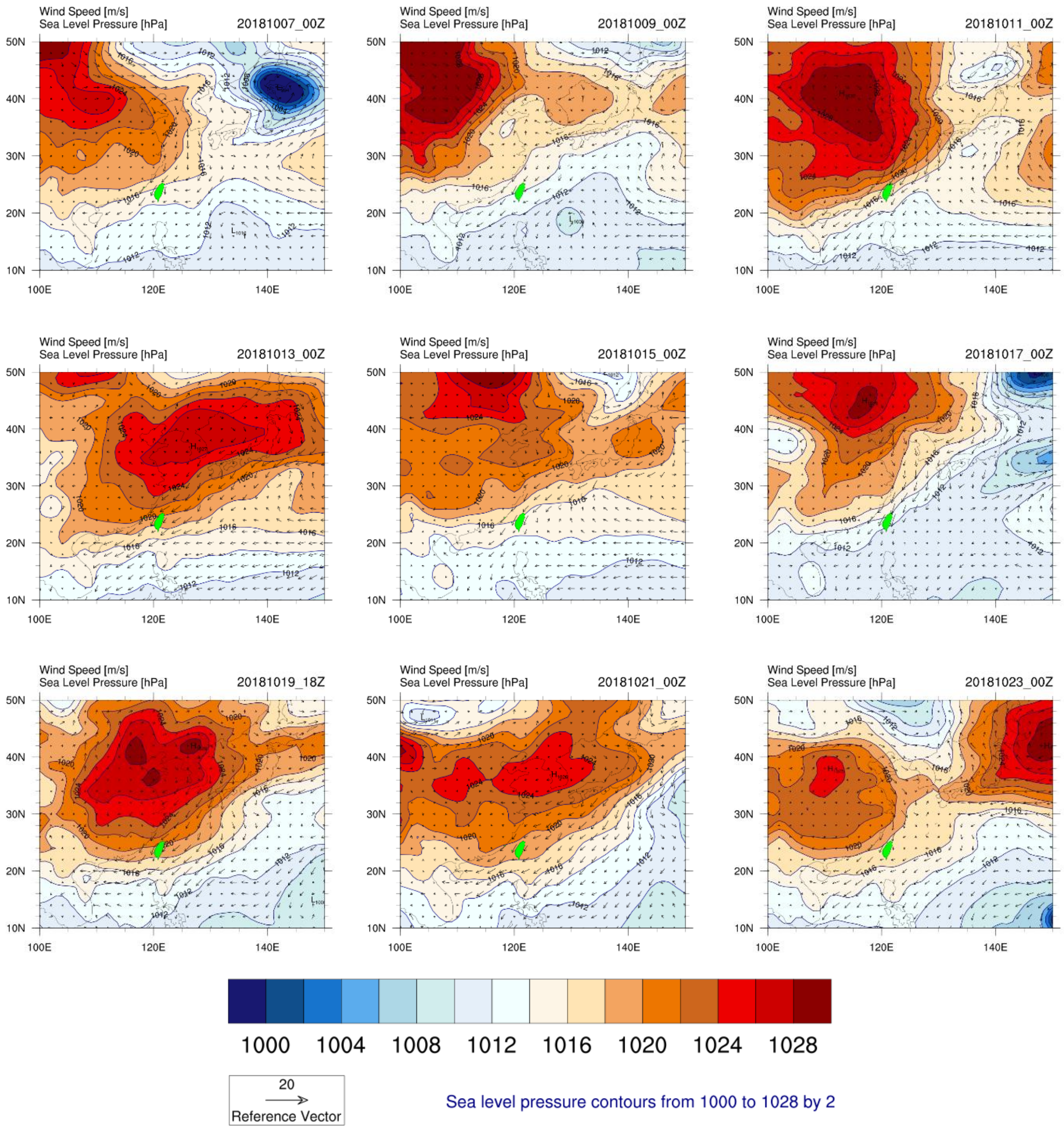

Figure S1: Synoptic weather pattern retrieved from NCEP-FNL reanalysis data valid at 00 UTC from 07 October 2018 to 23 October 2018 showing 850 hPa winds in vector referenced at 20 m s^{-1} and sea level pressure in color contoured from 980 to 1020 hPa by 2 hPa. Taiwan is highlighted with green color.

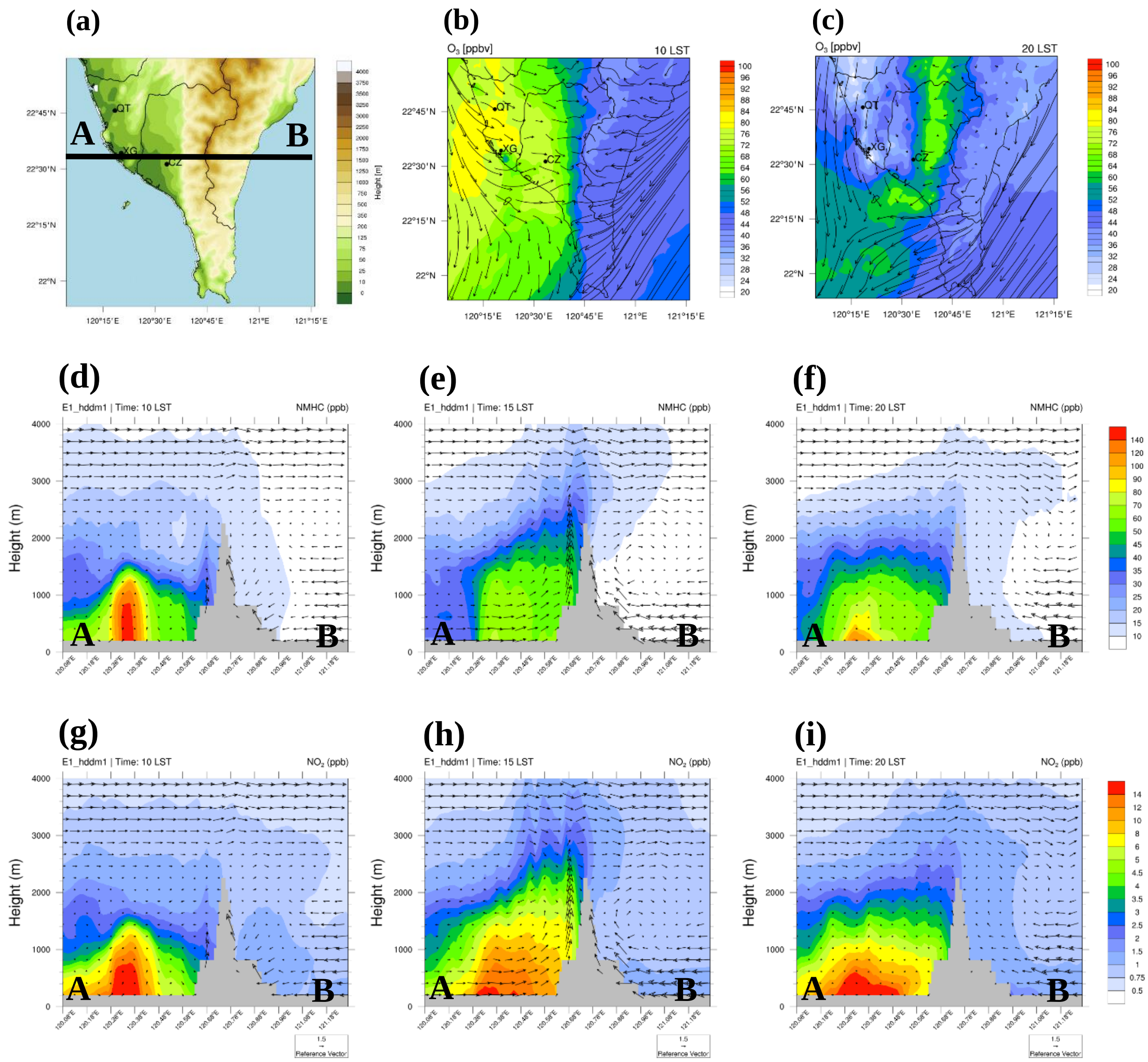

Figure 3: (a) Topography of the innermost domain. (b, c) Spatial distribution of O_3 concentration averaged during the entire simulation at daytime 10 LST and nighttime 20 LST, respectively. (d-f) Vertical profile of NMHC concentration cross sectioned at AB (see Figure 2a) averaged during the entire simulation at 10 LST, 15 LST, and 20 LST, respectively. (h-i) Same as d-f but for NO_2 concentration.

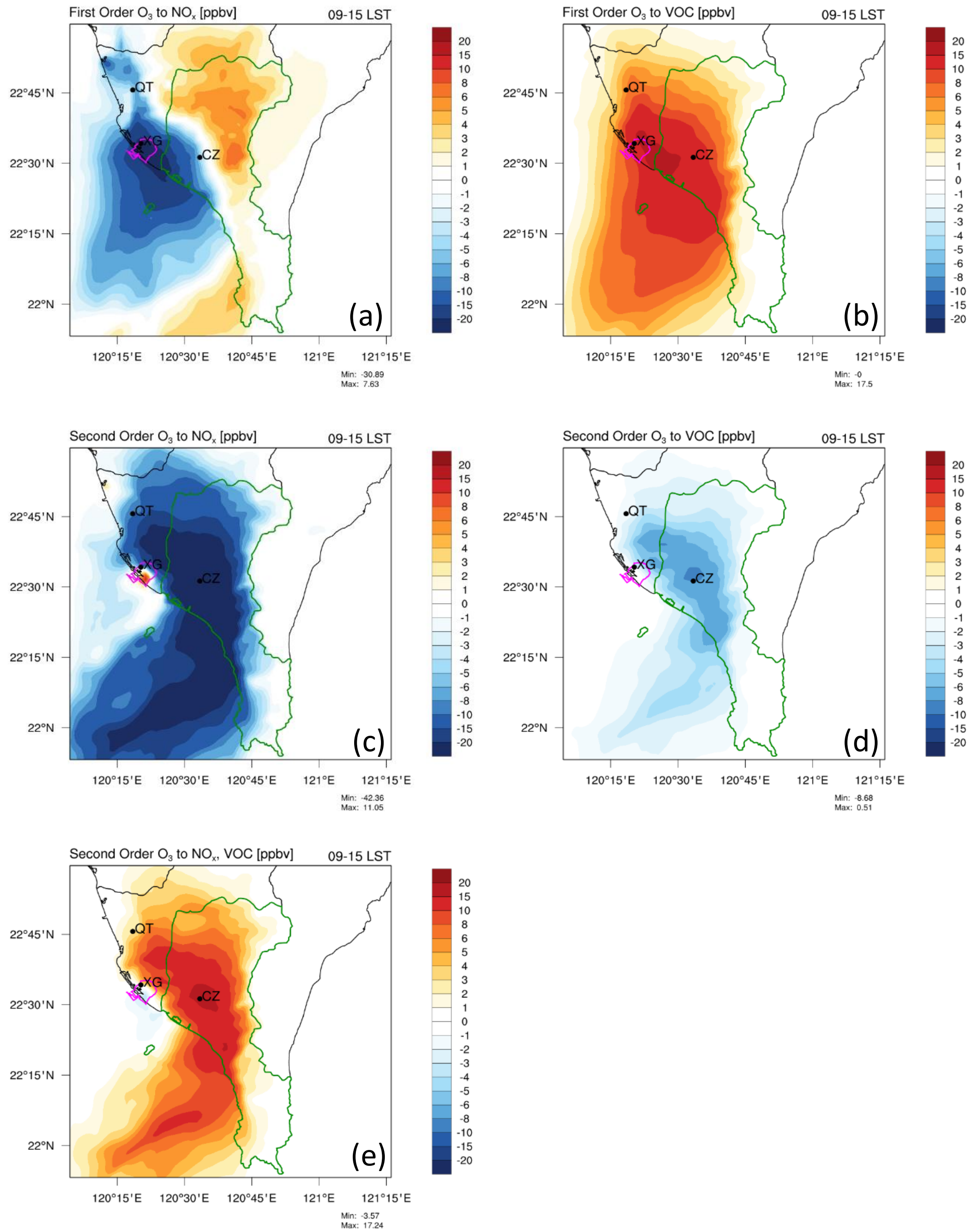

Figure 5: CMAQ-HDDM first-order sensitivity coefficient of O_3 to (a) NO_x emissions, (b) VOC emissions, second-order sensitivity coefficient of O_3 to (c) NO_x emissions, (d) VOC emissions, (e) second-order cross sensitivity coefficients of O_3 to NO_x, VOC emissions, at daytime 09-15 LST averaged during the entire simulation period. Magenta and green highlighted borderline represents Xiaogang District and Pingtung region, respectively.

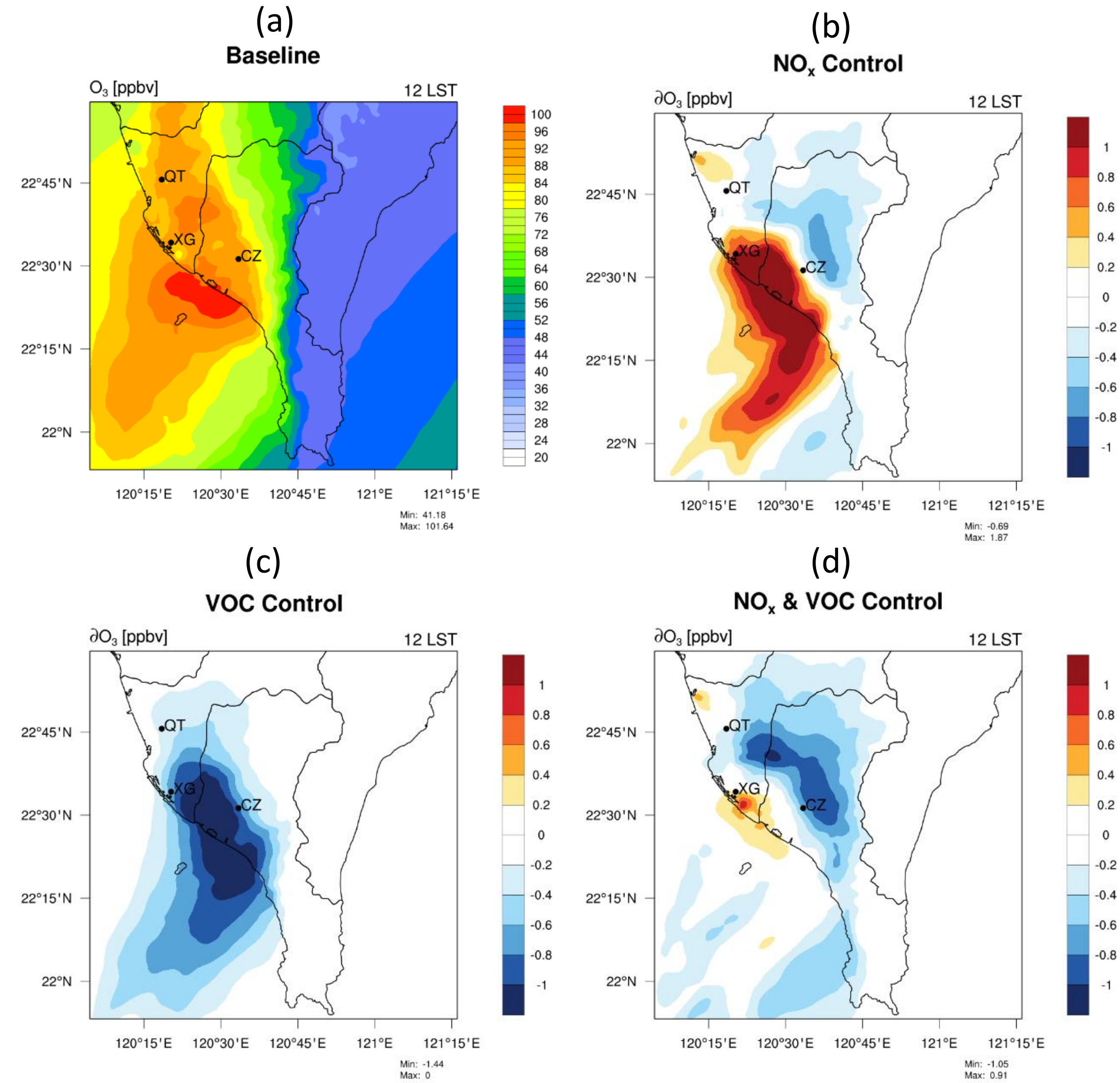

Figure 6: Spatial distribution of O_3 concentration in (a) baseline with no perturbations in NO_x and VOC emissions, and changes in O_3 concentration under (b) NO_x control scenario, (c) VOC control scenario, and (d) NO_x & VOC control scenario at daytime 12 LST. All scenarios reduced the targeted emissions by 5% except for the baseline.

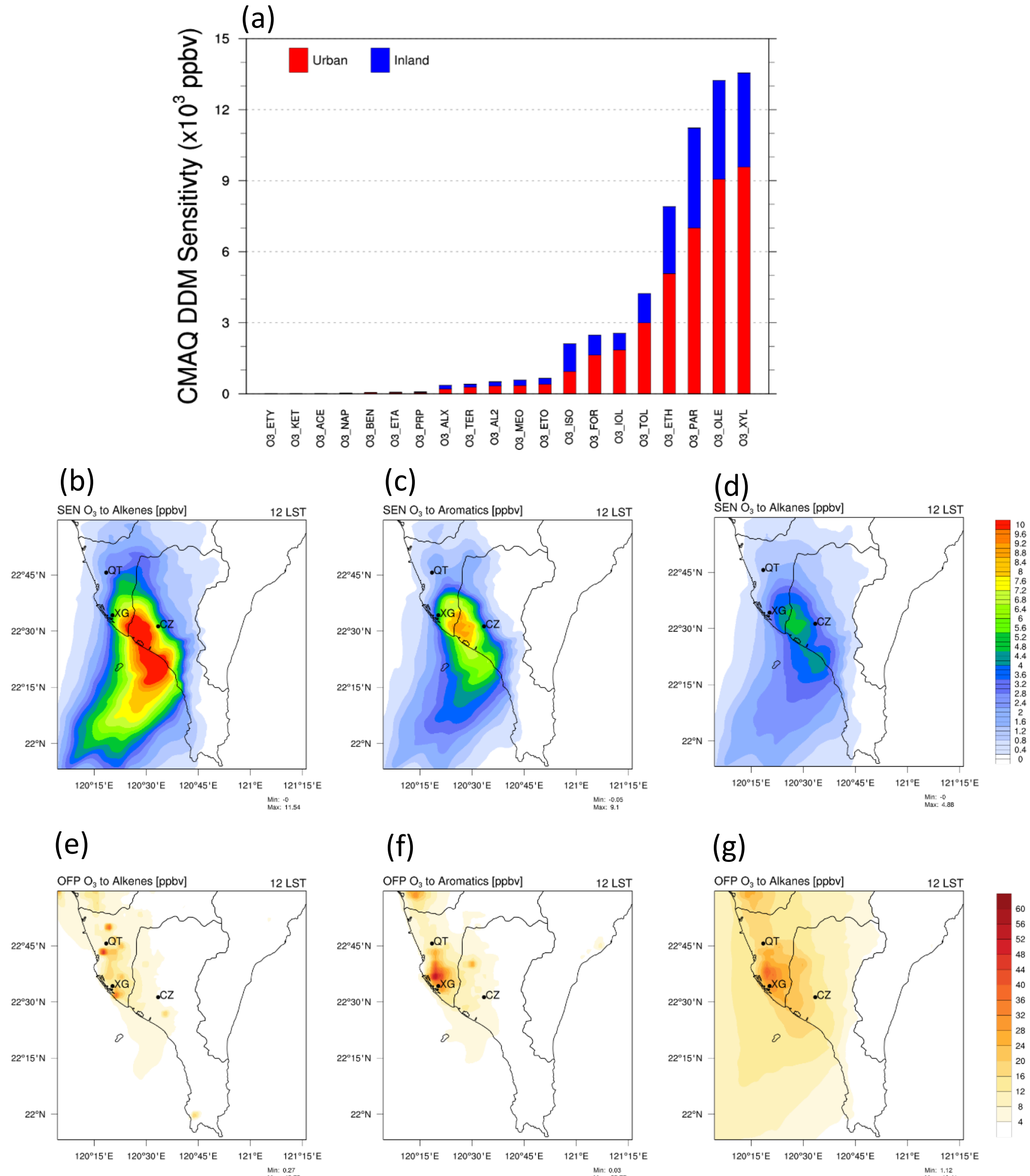

Figure 8: (a) Daily averaged CMAQ-DDM first-order sensitivity coefficient of O_3 concentrations calculated per number of grids to each modelled VOC species arranged in ascending order for urban and inland area. Sensitivity of O_3 and ozone formation potential (OFP) to (b,e) alkenes emissions (OLE + ETH + IOL), (c,f) aromatics emissions (XYL + TOL), and (d,g) alkanes emissions (PAR) at 12 LST.

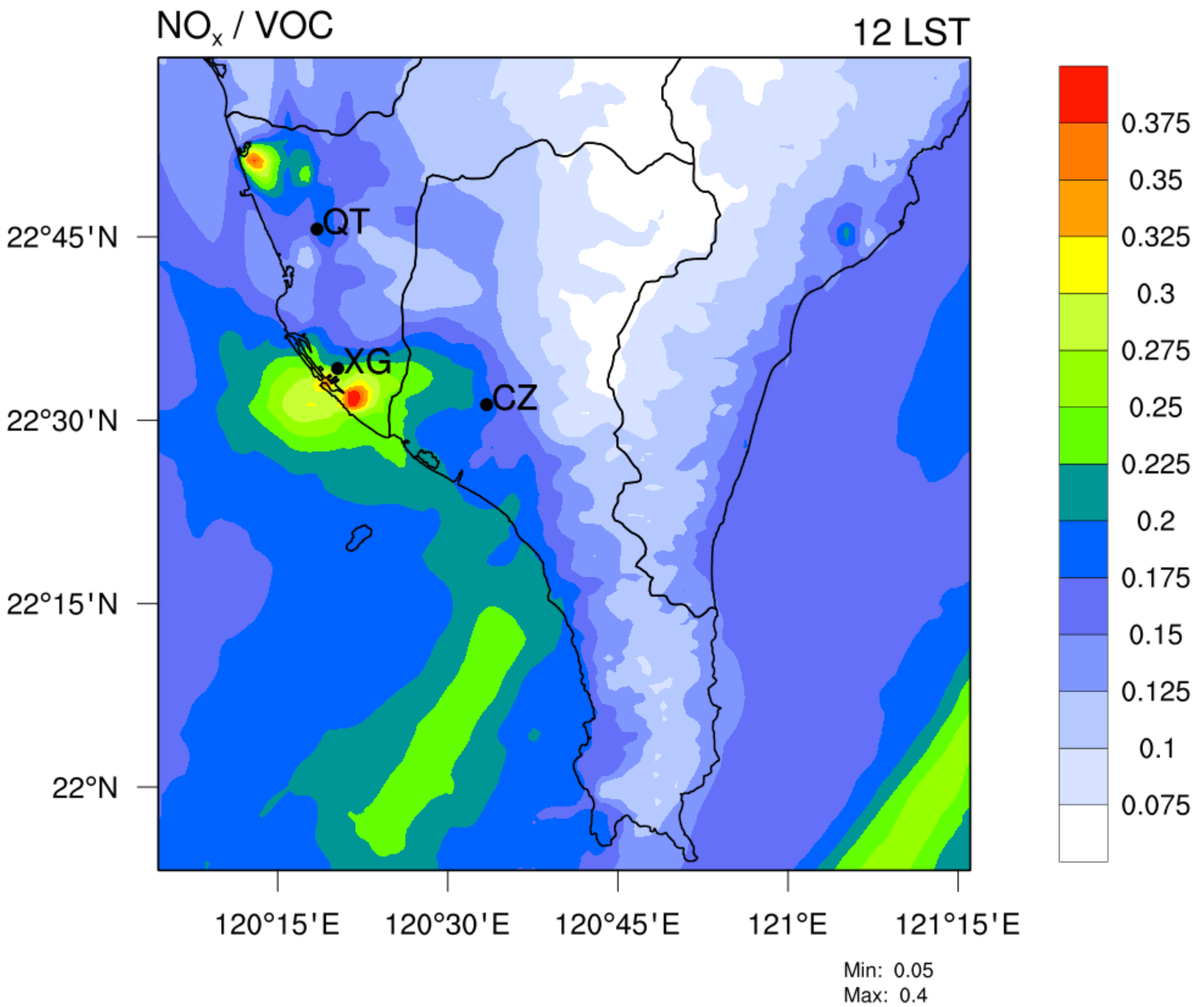

Figure S6: Spatial distribution of ratio NO_x / VOC averaged at 12:00 LST during the entire simulation period.

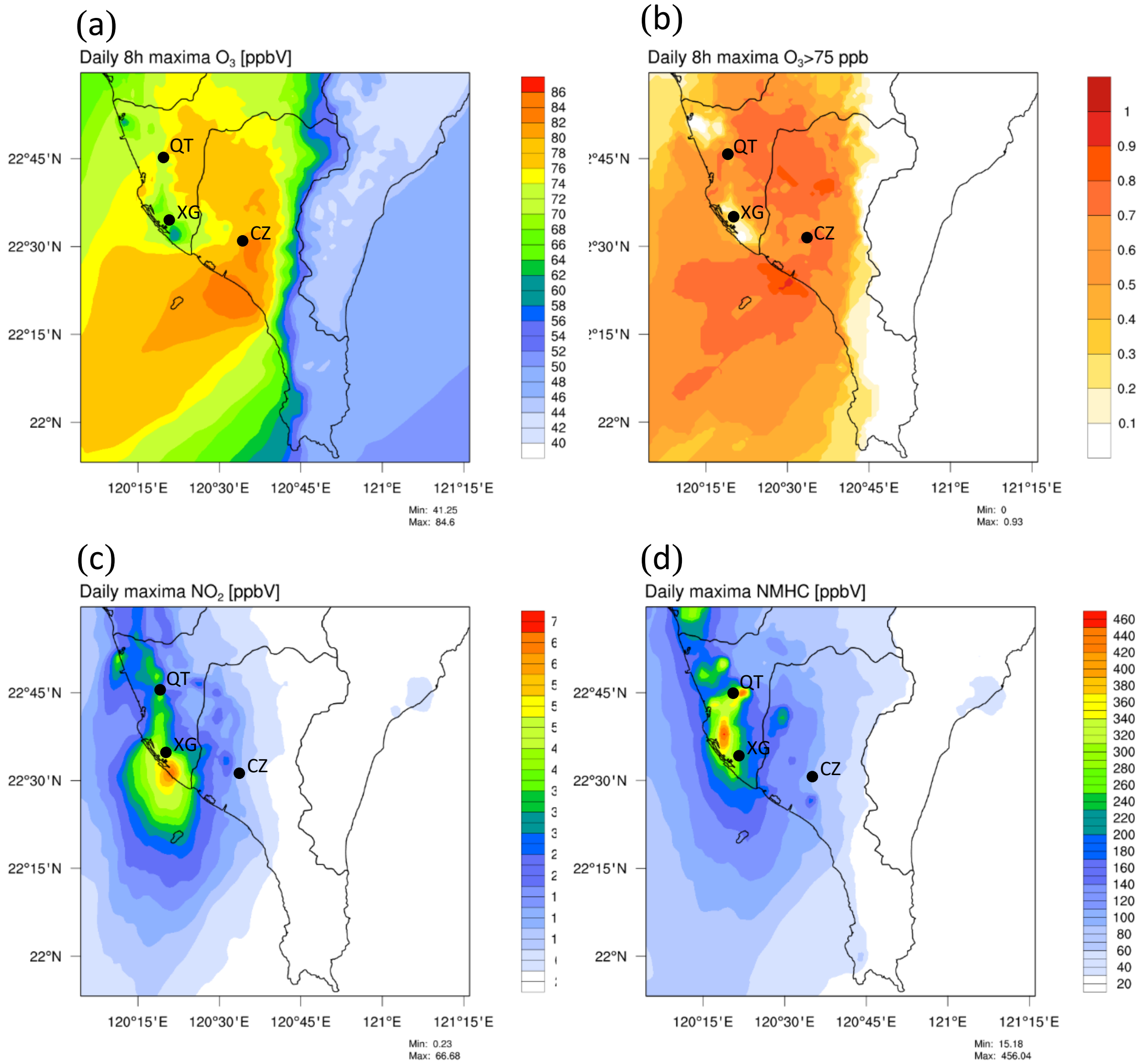

Figure S13: Spatial distribution of (a) daily 8h maxima O_3, (b) occurrence of daily 8h maxima O_3 >75 ppb (c) daily maxima NO_2, (d) daily maxima VOC, averaged during the entire simulation period at the lowest model level in the innermost domain.

Comments#3 What will be the impact on the O3 and VOCs analysis under high underestimation in temperature and overestimation in wind speed in urban shown in Fig S8? It is recommended to have more discussion on the base-case model performance. Will the overestimated NO2 create VOC limited situation in Urban (Fig S9), instead of emissions? How much error will it create? What will be the impact of overestimated NO2 on HDDM and PMF analysis?

Response Comment #3

Thank you for the comment. The discussion on the base-case model performance is provided in supplementary material (see Supplementary Material – Model Evaluations). We addressed the possible impact on the O_3 and VOC analysis under notable differences in temperature and wind speed on line 702-707. For the overestimated NO_2 in urban areas, we believe that such small error (MB = 5.5 ppb, MNB = 28%) should have minimal impact on HDDM and PMF analysis and is unlikely to create VOC-limited condition, instead of emissions. We showed in the Taylor-series approximation analysis when we hypothetically reduce NO_x emission at arbitrary -50% and -25% scenario, which should expect an improvement in the overestimation of NO_2, urban O_3 at XG remains in a VOC-limited condition. Therefore, it is certain that the VOC-limited condition at urban areas of southern Taiwan is mainly due to the high anthropogenic NO_x emissions. We addressed this issue accordingly in the revised manuscript on line 439-442.

Line 702-707***: "Although the performance of the simulated meteorological parameters (T2, WS, and WD) at both urban and rural stations are acceptable in the benchmark recommended by USEPA, notable differences in temperature (underestimation) and wind speed (overestimation) are still observable in our simulation work. These biases could be susceptible to underestimation in photochemical ozone production due to the fictitious cold bias and enhanced dispersion. Therefore, careful treatment on the urban-scale data assimilation in temperature, wind field and relative humidity are recommended in future to improve the model prediction."***

Line 439-442***: "We showed in the Taylor-series approximation analysis when we hypothetically reduce*** NO_x ***emission at arbitrary -50% and -25% scenario, which should expect an improvement in the overestimation of*** NO_2***, urban*** O_3 ***at XG remains in a VOC-limited condition. Therefore, it is certain that the VOC-limited condition at urban areas of southern Taiwan is mainly due to the high anthropogenic*** NO_x ***emissions."***

Comments#4 There are many colour labels in Fig S1. However, they were not defined. Please also add (QT, CZ, and XG – abbreviation) into Fig S1.

Response Comment #4

After revision, the color labels are defined and the abbreviations (QT, CZ, XG) are added in Figure S3.

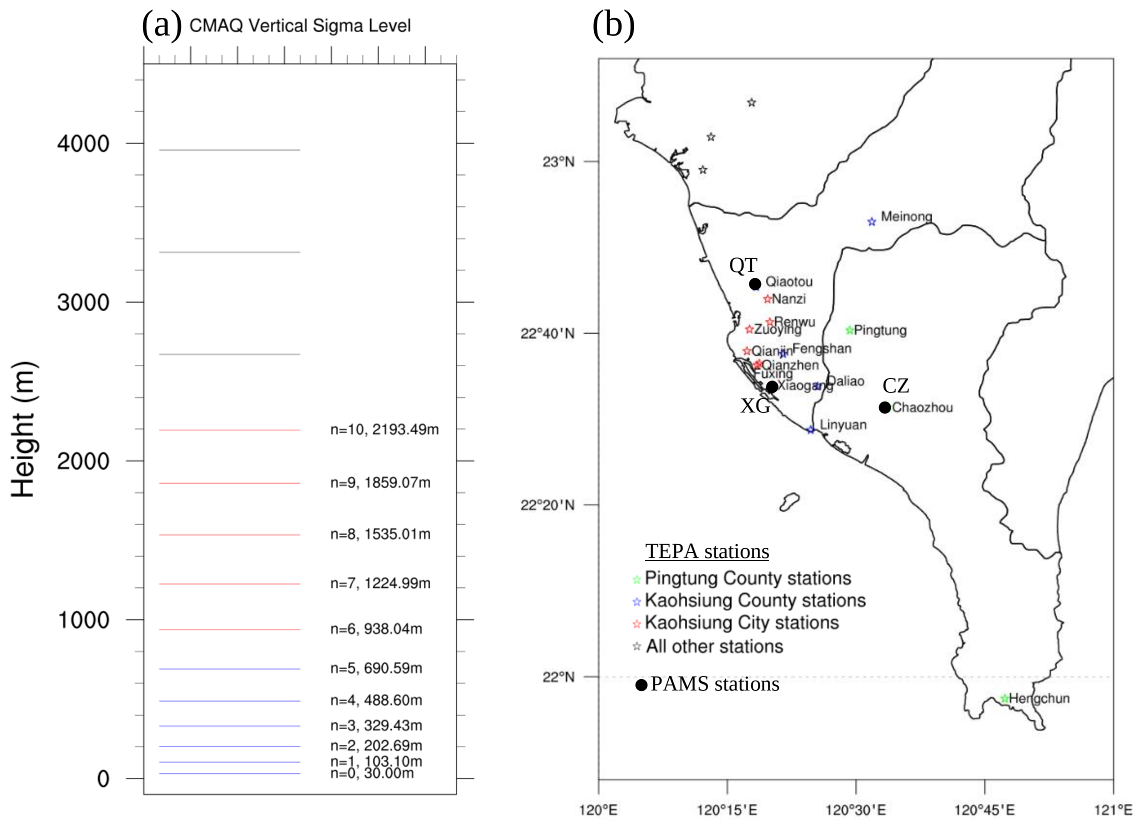

Figure S3: (a) CMAQ vertical layer distribution in eta sigma level. The lowest model level n=0 is approximately 30.0 m above the ground. (b) Distribution of TEPA air quality stations over southern Taiwan.

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