Responses to Reviewer Comments

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Title: Implications of Mitigating the Ozone and Fine Particulate Matter Pollution in the Greater Bay Area Using a Regional-to-Local Coupling Model
Authors: Xuguo Zhang, Jenny Stocker, Kate Johnson, Yik Him Fung, Teng Yao, Christina Hood, David Carruthers, and Jimmy C. H. Fung*

Dear Editor and Reviewers:

Thank you for your letter and for the reviewers' comments concerning our manuscript entitled "Implications of Mitigating the Ozone and Fine Particulate Matter Pollution in the Greater Bay Area Using a Regional-to-Local Coupling Model" (Manuscript ID: 2021GH000506R). Those comments are all valuable, constructive, and very helpful for revising and improving our manuscript and the important guiding significance to our research. We have studied comments carefully and have made corrections, which we hope to meet with approval. The main corrections in the manuscript and the responses to the reviewer's comments are as follows. We hope the revised manuscript will be deemed suitable for publication in GeoHealth Journal.

Reviewer #1 Evaluations:
Recommendation: Return to author for major revisions
Significant: The paper has some unclear or incomplete reasoning but will likely be a significant contribution with revision and clarification.
Supported: Mostly yes, but some further information and/or data are needed.
Referencing: Yes
Quality: The organization of the manuscript and presentation of the data and results need some improvement.
Data: No
Accurate Key Points: Yes

Reviewer #1 (Formal Review for Authors (shown to authors)):
General comments:
This manuscript describes a coupled modeling system including the CMAQ regional model and the ADMS-Urban street-level dispersion model. The coupled system is used to assess emission control scenarios in the PRD area. The rationale is that finer resolution modeling will better represent the emission impacts on air quality in urban areas.
While this study uses a reasonable approach, there needs to be more explanation of the modeling. For example, the description of the model coupling is unclear. If the emissions used in the ADMS-U model are removed from the CMAQ model, then that could alter the non-linear chemistry in CMAQ. Better to really use the method described by Hood et al 2018. The spatial domains of the ADMS-U application are also unclear. Should show a map showing where the ADMS-U is applied. Also, spatial references are confusing: PRD EZ, GBA, Guangzhou coupled system domains, Guangzhou domain. How are these defined. More detail on the traffic emissions and building data are needed. What are the limitations of the approaches used? Is there any evaluation of the ADMS-U for street level concentrations?
There is a lot of repetition in the Results, Discussion, and Conclusion sections. The effects of the controls on NO2, O3 and PM25 are repeated 4 times! I think the Discussion section could be removed. Needs through editing for proper grammar.

Response: *We sincerely appreciate the reviewer for the careful and detailed comments. The point-by-point responses to the comments are given below. Responses are in blue and italic. The underlined and bold line numbers in the square brackets are according to the revised manuscript. Please see the following itemized responses.*

(1) *In terms of the model coupling, we add the research framework (Figure S1) and also amend the related contents as follows.*

> *Figure S1 shows a research framework for the CMAQ–ADMS-Urban air quality modeling system. The street-scale resolution ADMS-Urban dispersion model was coupled with the regional CMAQ model using the ADMS-Urban Regional Model Link (ADMS-Urban RML) to investigate O_3 and $PM_{2.5}$ concentrations and the sensitivity of both pollutants to emissions from the traffic and industrial sectors. The ADMS-Urban RML is used to automatically prepare nested data from the regional model (CMAQ) and the meso-scale meteorological model (WRF) for the street-level ADMS-Urban model.* **[Line 187-193]**

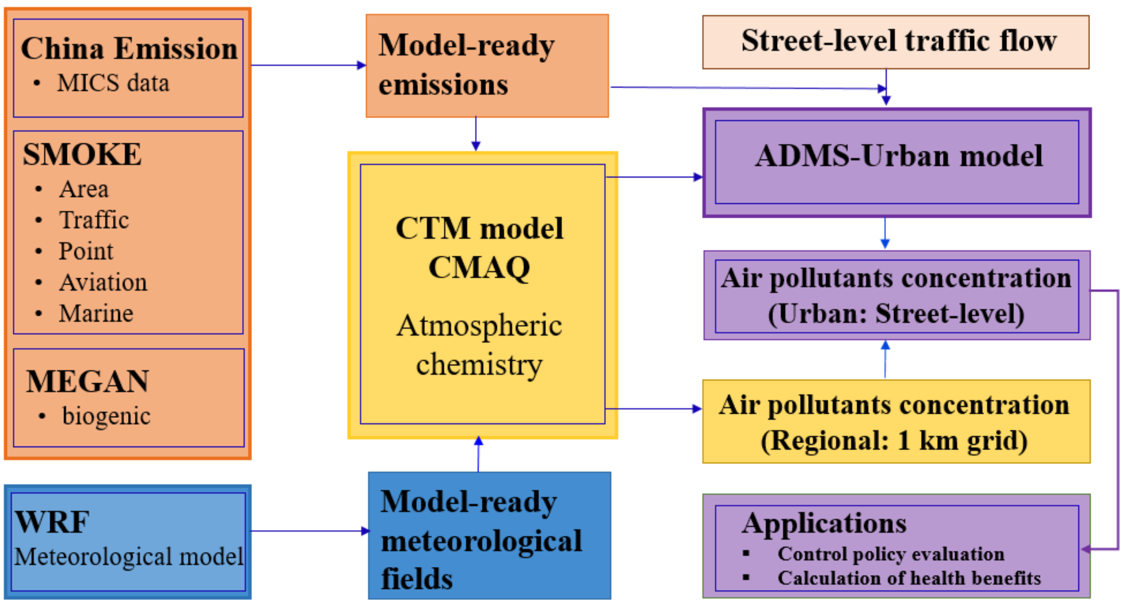

Figure S1. *Research framework for the CMAQ–ADMS-Urban modeling system.*

(2) *In terms of the emissions of ADMS-Urban model, we do not remove any emissions from the CMAQ model. Instead, the imitative simulated concentration of ADMS-Urban applying for the grid emissions (evenly distributed) of CMAQ model are reduced from the prepared background CMAQ concentrations. In our initial submission, we clarified the dealing method to avoid double-counting emissions as follows.*

> *In this system, double-counting of local emissions is avoided by deducting local urban-scale modelling of all emissions represented as grid sources from the regional modelling of all emissions before adding street-scale local modelling with explicit and gridded sources.* **[Line 142-145]**

In order to make it clearer, we add the following content:

> *We balance the additional high-resolution emissions in the local urban-scale model by using a corresponding negative amount of emissions at the same gridding as the regional CMAQ model to emulate the effects of removing such emissions from the regional model grid. The coupled modeling system redistributes emissions within each of the regional grids for the purpose of running the local urban-scale model.* **[Line 145-149]**

(3) In terms of the spatial domains, we amend Figure S2 and also add the geographic city locations in Figure S2 as follows:

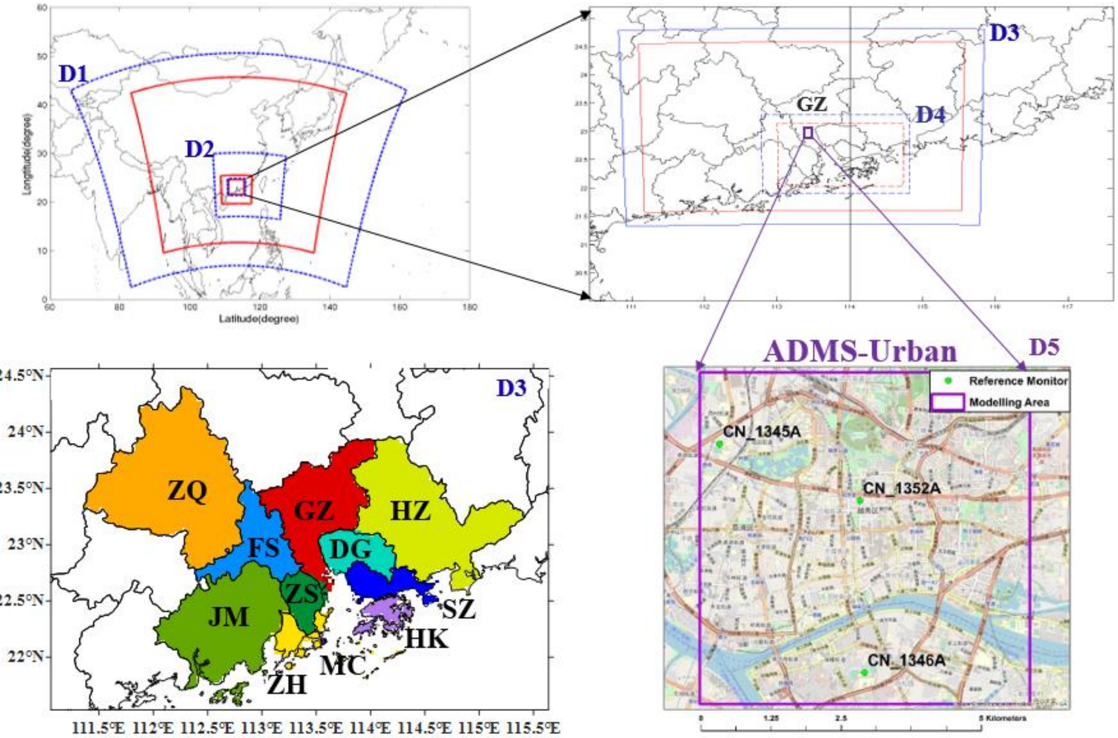

Figure S2. *Geographic domain setting of the CMAQ–ADMS-Urban modeling system: meso-scale meteorological model WRF (blue lines) and regional chemical transport model CMAQ (red lines) for D1 (27 km), D2 (9 km), D3 (3 km), D4 (1 km) and street-level ADMS-Urban model (purple lines) for D5 (6 km × 6 km). The Greater Bay Area (GBA) includes the PRD Economic Zone (Guangzhou (GZ), Shenzhen (SZ), Foshan (FS), Dongguan (DG), Zhuhai (ZH), Zhongshan (ZS), Jiangmen (JM), Huizhou (HZ), Zhaoqing (ZQ)), Hong Kong (HK), and Macau (MC).*

We also add the following clarification for the domain setting.

> *…… and Domain 5 (6 km × 6 km), in an urban area of Guangzhou City, was chosen to drive the street-level ADMS-Urban model. The GBA includes Hong Kong (HK), Macau (MC), and the Pearl River Delta Economic Zone (PRD EZ), which includes nine cities; i.e., Guangzhou (GZ), Shenzhen (SZ), Foshan (FS), Dongguan (DG), Zhuhai (ZH), Zhongshan (ZS), Jiangmen (JM), Huizhou (HZ), and Zhaoqing (ZQ). The WRF domains are larger than the CMAQ domain by at least 3–5 grids to remove the boundary effects of the WRF model on the CMAQ model.* **[Line 216-222]**

(4) In terms of the traffic emissions and building data, we clarify more on the redistribution of the traffic emissions. However, urban morphology data such as street canyon and building data are lacking in the Guangzhou region. Therefore, these can be considered in the coupled model in the future if such data become available. We add the contents as follows:

> *Two sets of explicit traffic emissions were prepared: one set emulated the CMAQ grid concentrations that were distributed evenly across the traffic emissions model grid and extracted from the CMAQ model grids; the second set redistributed the CMAQ grid traffic emissions into explicit high-resolution traffic emissions within the facilitated road network. The emulated ADMS-Urban concentrations using the evenly distributed grid emissions were reduced from the ultimate ADMS-Urban concentration calculations to avoid double-counting of emissions. The allocation*

formula for redistributing the traffic emissions is detailed in Biggart et al. (2020). The road length was determined and calculated on the basis of the CMAQ grids. **[Line 245-253]**

Urban morphology data, such as street canyon and building data, are lacking in the Guangzhou region; these could be considered in the coupled model in the future if such data become available.

[Line 257-259]

(5) *In terms of the limitations of the approaches, we add the following content in the discussion part.*

Although the implemented coupled CMAQ–ADMS-Urban modeling system is capable of resolving the fine concentration gradient near road networks in this study, several limitations remain to be further investigated in future studies. First, more complete emission sectors, such as point, industry, or residential sources, should be included to construct holistic, high-resolution concentration maps. Second, the urban domain should be further expanded to cover the whole GBA to obtain more complete measurements for model validation and exploration of photochemical mechanisms. Finally, the street canyon module and more detailed building data will most certainly benefit accurate calculations of the dispersion of air pollutants. **[Line 558-566]**

(6) *In terms of evaluation of the ADMS-Urban model system, we add the content as follows:*

Statistical parameter performances (Table 2) and time series plots for typical monitoring stations (Figures S10–S12) in the GBA were analyzed to validate the base case of the regional CMAQ model at a 1-km resolution. Table 2 clearly shows that the CMAQ model obtains an acceptable level of accuracy for $PM_{2.5}$ simulations (with mean fractional bias $\leq \pm 0.6$ and mean fractional error ≤ 0.75) according to the criteria proposed by Hu et al. (2016). The averaged O_3 observation is 28.6 ppb, and the mean O_3 simulation is 28.9 ppb, with an Index of Agreement (IOA) of 0.63. Although the CMAQ model underestimates NO_2 concentrations by 2.5 ppb, the IOA of NO_2 is up to 0.57, and the root mean square error is around 10. Overall, the CMAQ model simulation is considered an acceptable input to drive the ADMS-Urban model. In addition to the capability of the CMAQ model to capture the main trend in the time series plots during the modeling period, Figures S13–S15 show the time series comparisons of the base case for both the CMAQ and ADMS-Urban models. Substantial improvement was observed during specific pollution episodes, which illustrates the advantages of coupled urban dispersion models. **[Line 328-341]**

Table 2. Statistical performance of the CMAQ base scenario in regional model domain 4, at a resolution of 1 km. The units of NO_2 and O_3 are ppb, the unit of $PM_{2.5}$ is μg/m^3.

	OBS	Model	IOA	RMSE	MNB	MNE	MFB	MFE
NO_2	15.3	12.8	0.57	10.37	0.26	0.81	-0.19	0.61
O_3	28.6	28.9	0.63	18.51	1.32	1.63	0.18	0.61
$PM_{2.5}$	18.2	13.92	0.49	12.03	0.07	0.65	-0.25	0.57

(7) *In terms of the repetition comments, we think that the reviewer has such an impression mainly because three air pollutants are discussed at the same time.*

The reviewer comments: 'The effects of the controls on NO2, O3 and PM25 are repeated 4 times!', but without indicating the line numbers. The reviewer comments: 'Needs through editing for proper grammar.', but still without indicating the line numbers with grammar errors. Therefore, it's hard for us to correct those accordingly.

In responding to the reviewer's comments, we rewrite the discussion part instead of simply removing it. We also double-check the manuscript to make sure no grammar errors left.

Specific comments:

1. Lines 105-106: This sentence does not make sense. O3 is worsening due to fewer hydroperoxy radicals for O3 formation?

Response: *Thanks for pointing out this. We change the wording as follows:*

> *Li et al. (2019a) found that the main cause of increasing O_3 concentrations in the North China Plain (NCP) after 2013 was a significant reduction in $PM_{2.5}$ concentrations, which slowed hydroperoxy radical consumption and increased the rate of O_3 formation.* **[Line 104-107]**

2. Ln 112-113: What "cross-boundary transport"?

Response: *Thanks for the question. The "cross-boundary transport" means the regional effects of pollution transport between cities in the Yangtze River Delta (YRD) region. In order to avoid confusing the readers, we change the wording as follows:*

> *…… exploring the regional effects of pollution transport on the interaction among the $PM_{2.5}$ and O_3 from cities in the Yangtze River Delta (YRD) region.* **[Line 113-115]**

3. Ln 139-142: Please re-write this sentence for more clarity. It is too confusing. It seems that this study does not follow Hood et al. 2018. See my comment for ln 218-220

Response: *Thanks for your comments. We add the content as follows to make it clearer.*

> *We balance the additional high-resolution emissions in the local urban-scale model by using a corresponding negative amount of emissions at the same gridding as the regional CMAQ model to emulate the effects of removing such emissions from the regional model grid. The coupled modeling system redistributes emissions within each of the regional grids for the purpose of running the local urban-scale model.* **[Line 145-149]**
>
> *Two sets of explicit traffic emissions were prepared: one set emulated the CMAQ grid concentrations that were distributed evenly across the traffic emissions model grid and extracted from the CMAQ model grids; the second set redistributed the CMAQ grid traffic emissions into explicit high-resolution traffic emissions within the facilitated road network. The emulated ADMS-Urban concentrations using the evenly distributed grid emissions were reduced from the ultimate ADMS-Urban concentration calculations to avoid double-counting of emissions.*
>
> **[Line 245-251]**

4. Ln 207-209: Was an urban scheme which accounts for buildings and street canyons used in WRF? Since the WRF data is used to drive the ADMS-Urban, it seems important to include the effects of building on wind fields, turbulence, and temperatures.

Response: *Thanks for the question. We did not include any urban schemes (buildings and street canyons) in the running WRF model. The major reason is that there is no such data in the GBA at the moment. We agree with the reviewer that these factors are important. However, the major focus of this study is to use the regional model (CMAQ) simulation to drive the street-scale model (ADMS-Urban) with resolved traffic emissions. We may consider the effects of buildings and street canyons on the meteorological fields in WRF if such data are available in the future.*

5. Ln 218-220: It seems that the coupling described here is different from that described by Hood et al 2018. They do not remove emissions from the regional scale grid model but rather subtract concentration in the plumes up to a mixing time (equation 1). Why was this technique not used for the current study? This needs to be explained rather than just referring to Hood et al.

Response: *Thanks for the question. We use the same coupling method described by Hood et al. (2018). As answered in Question 3 above, we add more content in Line 145-149 and Line 245-251 to make it clearer.*

6. Ln 259-274: This section is very hard to follow especially when all the figures are in the supplement. In general, if a figure is discussed in the main text the figure should be in the main text. Since the authors probably don't want to add 6 more multi-panel figure to the main text, I suggest moving this section to the supplement with a brief referral to it in the main text.
Response: *Thanks for the suggestion. We have followed the suggestion and adjusted the related contents.*

> *Daily column emissions comparisons for NO_x, VOC, and $PM_{2.5}$ are provided in the supporting information (Figures S3–S9).* **[Line 314-315]**

7. Figures S4 and S6: Why no differences in the HKSAR?
Response: *Thanks for the question. We assume no emission control actions in HK since we want to evaluate how the air pollution of the PRD EZ region change corresponding to the local controls. All the scenario runs are based on the sensitivity of the PRD EZ region since the Guangdong government is more interested in the air pollution changes of the PRD EZ region when only local controls are applied. In responding to the reviewer question, we add the content as follows:*

> *We assumed no emissions control activities in HK, as this study focuses on evaluating how air pollution in the PRD EZ changes in response to local controls.* **[Line 315-317]**

8. Ln 285 287: Confusing! "NO2 concentrations are significantly higher" than what? Surely not higher in the half traffic case?
Response: *Thanks for the question. That sentence shows the spatial distribution of Figures 1a and b. In order to make it clearer, we add the content as follows.*

> *In terms of the spatial distributions illustrated in Figures 1a and b, NO_2 concentrations are markedly higher in HK (south of Shenzhen), in industrial areas towards Guangzhou, and along shipping lanes than in the urban area in the GBA.* **[Line 354-357]**

9. Figure 1c: again, why no difference in HKSAR? Were the emission control scenarios not applied in the HKSAR? If not, why not?
Response: *Thanks for the question. As explained in above question 7, we assume no emission control measures in the HK region. Therefore, the difference map (Figure 1c) has no difference in the HK region. The reason we do not apply the emission controls is because the Guangdong government is more interested in the air pollution changes of the PRD EZ when only local controls are applied.*

10. In Figure S9 there is a purple box labeled modelling domain. Please explain!
Response: *Thanks for the question. The purple box (6 km × 6 km) is the model domain for the ADMS-Urban, which covers an urban area of Guangzhou city. In the revision, we integrate the ADMS-Urban model domain setting (figure S9) with Figure S2 to make the domain setting clearer.*

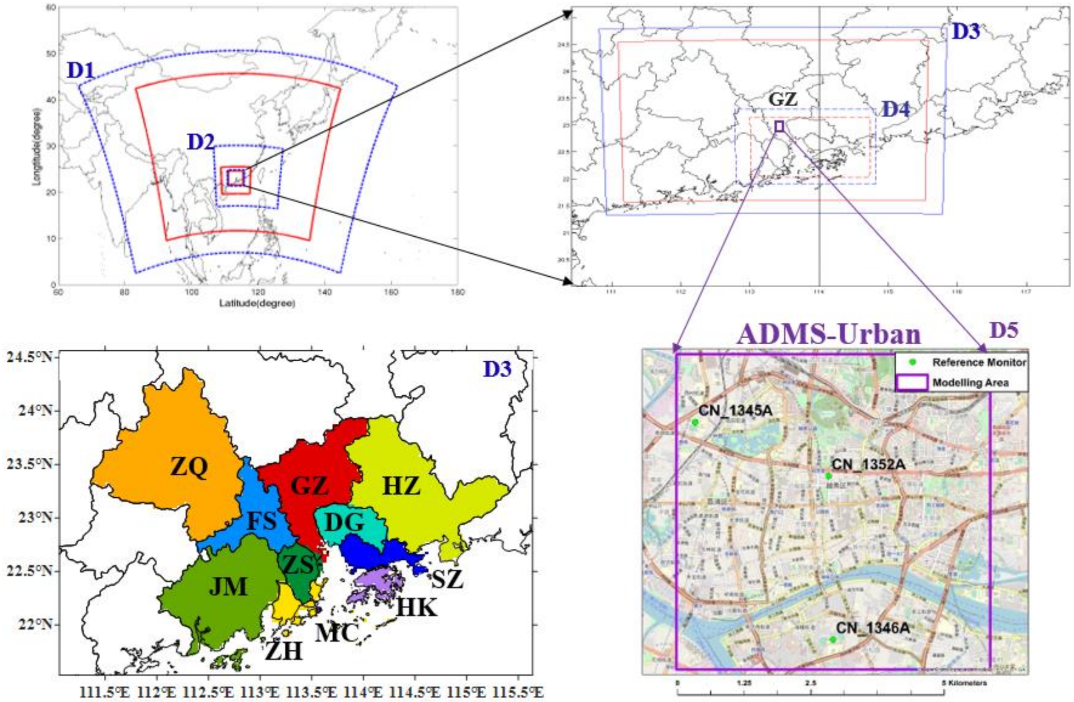

Figure S2. *Geographic domain setting of the CMAQ–ADMS-Urban modeling system: meso-scale meteorological model WRF (blue lines) and regional chemical transport model CMAQ (red lines) for D1 (27 km), D2 (9 km), D3 (3 km), D4 (1 km) and street-level ADMS-Urban model (purple lines) for D5 (6 km × 6 km). The Greater Bay Area (GBA) includes the PRD Economic Zone (Guangzhou (GZ), Shenzhen (SZ), Foshan (FS), Dongguan (DG), Zhuhai (ZH), Zhongshan (ZS), Jiangmen (JM), Huizhou (HZ), Zhaoqing (ZQ)), Hong Kong (HK), and Macau (MC).*

11. Ln 369-371: For the regional model results is the regional model run by itself (no ADMS-U)? If not, are all emissions included in the regional run?
Response: *Thanks for the question. The regional model results are obtained purely from the regional model CMAQ.*

12. Figure 9: The caption says that the regional model (red) is compared to the 4 high-res scenarios. Base is also Red. It seems that only the 4 high-res scenarios are shown and not the regional model. Please explain.

Response: *Thanks for pointing out this. Figure 9 (Now Figure 8) is a plot of regional model simulations. The four colors mean the CMAQ model base case and the three sensitivity scenarios (from the CMAQ model). We adjust the caption to avoid misunderstanding.*

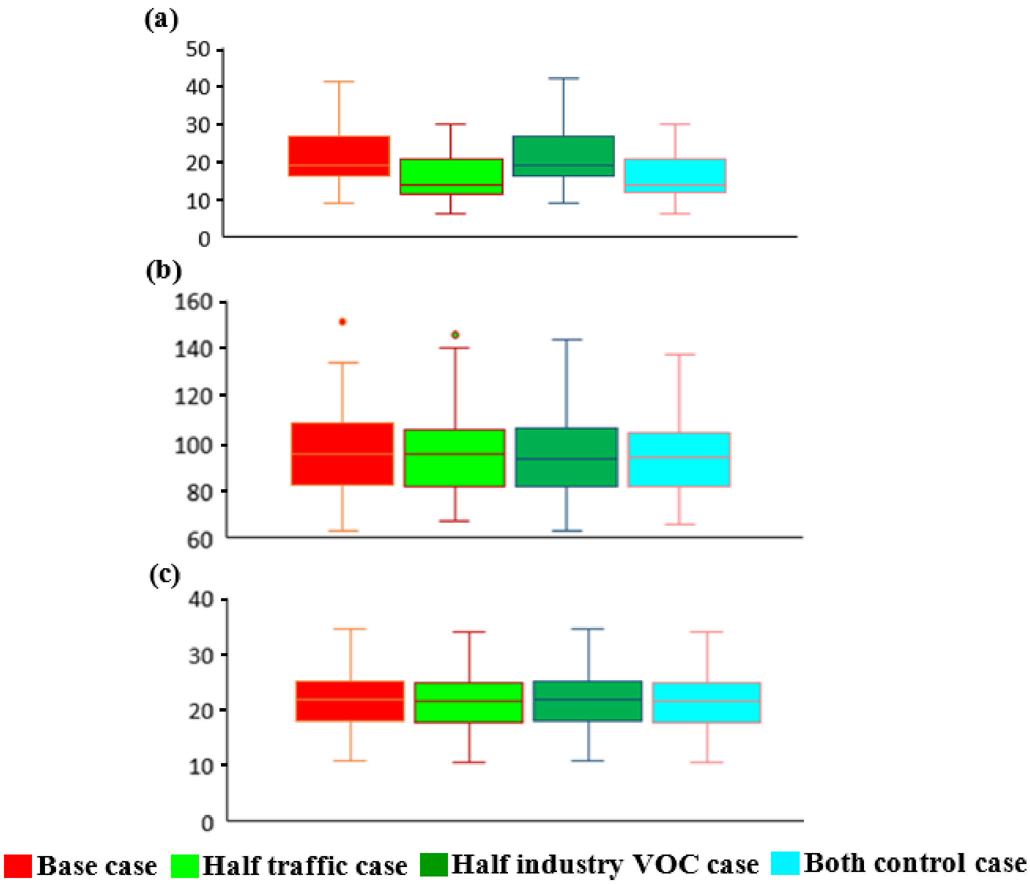

Figure 8. *Box plots comparing the regional CMAQ model concentrations at a rural location (white star in Figure 2): Base case (red), Half Traffic case (light green), Half Industry VOC case (darker green), and Both Controls (bright blue) for: (a) daily maximum hourly NO_2, (b) daily maximum 8-hour rolling O_3, and (c) daily average $PM_{2.5}$. Unit is in μg/m³.*

13. Ln 400-402: This sentence relates to the confusion about the CMAQ-ADMS-U coupling. If the coupling is done according to Hood et al (2018), then the effects of the high-resolution ADMS-U modeling in the urban parts should have effect on the regional results downwind.
Response: *Thanks for the question. We amend the content related to the coupling of the CMAQ–ADMS-Urban system and also add a figure of our research framework to make the coupling of the regional CMAQ model and local urban model clearer. In this study, the street-scale resolution ADMS-Urban dispersion model has been coupled with the regional CMAQ model using the ADMS-Urban Regional Model Link (ADMS-Urban RML), which is used to automatically prepare the nested data from the regional model (CMAQ) and the meso-scale meteorological model (WRF) for the street-level ADMS-Urban.*

> *Figure S1 shows a research framework for the CMAQ–ADMS-Urban air quality modeling system. The street-scale resolution ADMS-Urban dispersion model was coupled with the regional CMAQ model using the ADMS-Urban Regional Model Link (ADMS-Urban RML) to investigate O_3 and $PM_{2.5}$ concentrations and the sensitivity of both pollutants to emissions from the traffic and industrial sectors. The ADMS-Urban RML is used to automatically prepare nested data from the regional model (CMAQ) and the meso-scale meteorological model (WRF) for the street-level ADMS-Urban model.* **[Line 187-193]**

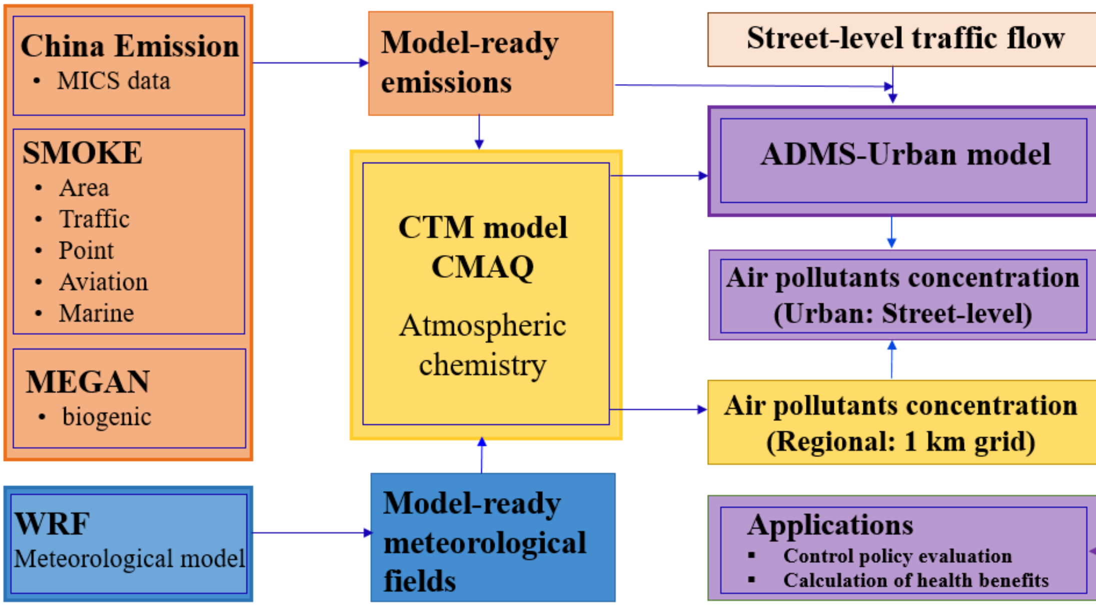

Figure S1. *Research framework for the CMAQ–ADMS-Urban modeling system.*

14. Ln 436: is this the only area where ADMS-U is applied?
Response: *Thanks for the question. Guangzhou urban area is the current applied ADMS-Urban domain.*

15. Ln 451-455: These sentences do not make sense. Please rewrite.
Response: *Thanks for the question. We rewrite the discussion part based on other reviewers' comments. We decided to delete the mentioned sentences in order to avoid confusion.*

16. Ln 527: "reduction radios"?
Response: *Thanks for pointing out this. We change the wording as follows:*

> *……Finally, assessing reduction in the NO_x/VOC radio in various areas of a city or in different cities should be cautiously assessed ……* **[Line 593-594]**

Reviewer #2 Evaluations:
Recommendation: Return to author for major revisions
Significant: Yes, the paper is a significant contribution and worthy of prompt publication.
Supported: Yes
Referencing: Yes
Quality: The organization of the manuscript and presentation of the data and results need some improvement.
Data: Yes
Accurate Key Points: Please Select

Reviewer #2 (Formal Review for Authors (shown to authors)):
In this paper, a regional-to-local coupled model is used to study the spatial variation characteristics of NOX, O3 and PM2.5 in the Greater Bay Area and Guangzhou, with the different emissions scenarios from traffic sources and industrial sources. The topic of this study is very interesting, and the methodology is sound. I recommend the publication by addressing the concerns below:

The major concern includes
17. The theoretical significance of halving the precursor emissions from traffic and industrial sources in this study, or any justification of the emission reduction? The author should explain why there was no change in Hong Kong and Macao under different control scenarios in Figure 1, 3 and 4.

Response: *Thanks for the questions. The main reason for halving the precursor emissions is to assess the sensitivity of the air pollutant concentrations on the corresponding emission changes. The brute-force method used in the sensitivity analysis will cause an accuracy issue if small emission changes are applied (Clappier et al., 2017; Yarwood et al., 2017). A previous study (Tsimpidi et al., 2008) utilized the same strategy to assess the fine particulate matter changes corresponding to the halved emissions in NOx and VOC in the US. However, only $PM_{2.5}$ concentrations have been explored for different regions in the US, and no more detailed analysis on the specific sectors was shown. Therefore, halving the precursor emissions in sensitivity analysis of air quality modeling is a typical and effective way to evaluate the sectoral concentration responses.*

In terms of the no emission changes in Hong Kong and Macao under different control scenarios (Figure 1, 3, 4), as we answered in Question 7 above, the provincial government in mainland China cares more about how the air pollutants concentrations change if the control measures are implemented in the PRD Economic Zone (PRD EZ) area. Therefore, we assume there are no emission changes in HK and Macau. The current scenario setting is designed to investigate the concentration responses of the local control measures.

In order to avoid confusing the audience, we add the contents as follows:

> *The main reason for halving the precursor emissions was to assess the sensitivity of the air pollutant concentrations to corresponding changes in emission. The brute-force method used in the sensitivity analysis would cause accuracy issues if small emission changes were applied (Clappier et al., 2017 and Yarwood et al., 2017). A previous study (Tsimpidi et al., 2008) utilized the same strategy to assess fine particulate matter changes corresponding to halved NO_x and VOC emissions in the United States. However, although $PM_{2.5}$ concentrations were investigated in different regions, no analyses were conducted for specific sectors. Therefore, halving the precursor*

emissions in sensitivity analyses of air quality modeling is a typical and effective way to evaluate the sectoral concentration responses. **[Line 286-295]**

……for the Guangzhou urban area. The provincial government in mainland China focuses largely on how the concentrations of air pollutants change if control measures are implemented in the PRD EZ; therefore, we assumed that there were no changes in emissions for HK and MC. The aim of the scenario was to investigate the concentration responses of local control measures. **[Line 265-269]**

Clappier, A., Belis, C. A., Pernigotti, D., & Thunis, P. (2017). Source apportionment and sensitivity analysis: two methodologies with two different purposes. Geoscientific Model Development, 10(11), 4245-4256.

Yarwood, G., Morris, R. E., & Wilson, G. M. (2007). Particulate matter source apportionment technology (PSAT) in the CAMx photochemical grid model. In Air Pollution Modeling and Its Application XVII (pp. 478-492). Springer, Boston, MA.

Tsimpidi, A. P., Karydis, V. A., & Pandis, S. N. (2008). Response of fine particulate matter to emission changes of oxides of nitrogen and anthropogenic volatile organic compounds in the Eastern United States. Journal of the Air & Waste Management Association, 58(11), 1463-1473.

18. One of the key points of this study is to evaluate the impact of NOX and VOC on O3 and PM2.5, but the conclusion is that the industrial VOC emissions have no significant impact on PM2.5, which seems to be different from previous studies. Any comparison with those literature? What is the guiding significance for coordinated emission reduction?

Response: *Thanks for the questions. In our scenario, the negative impacts on $PM_{2.5}$ on reduced industrial VOC emissions is mainly observed in the background regional CMAQ model, as the ADMS-Urban model has no explicit industrial sources in the reduced industrial VOC scenario. Reducing industrial VOC emissions will directly impact oxidant levels, thus impacting the formation of nitrate, sulfate, and secondary organic aerosols, which are important components of $PM_{2.5}$. When VOC emissions are reduced by 50%, the level of oxidants in summer will increase, leading to increased sulfate or nitrate formation. However, organic matter will be reduced, as more secondary organic aerosol will be generated by increased VOC emissions. Therefore, the net change in $PM_{2.5}$ would be small. This result is consistent with a previous study (Tsimpidi et al., 2008), indicating that controlling industrial VOC emissions may not be an efficient method of controlling $PM_{2.5}$. The simultaneous control of $PM_{2.5}$ and O_3 is a complex issue, and mitigation strategies will vary between areas with different formation regimes (i.e., VOC-limited, NOx limited, or NH_3-rich/poor) (Xing et al., 2019). NH_3 emissions need to be considered to further mitigate $PM_{2.5}$ concentrations in the PRD EZ, as NH_3 has also been detected in eastern China, as well (Geng et al., 2019).*

In responding to the reviewer's questions, we change the wording as follows:

The change in VOC emissions has little effect (Figure 6a and 6c) on $PM_{2.5}$ concentrations. In our scenario, the negative impacts on $PM_{2.5}$ on reduced industrial VOC emissions are mainly observed in the background regional CMAQ model, as the ADMS-Urban model has no explicit industrial sources in the reduced industrial VOC scenario. Reducing industrial VOC emissions will directly impact oxidant levels, thus impacting the formation of nitrate, sulfate, and secondary organic aerosols, which are important components of $PM_{2.5}$. When VOC emissions are reduced by 50%, the level of oxidants in summer will increase, leading to increased sulfate or nitrate formation. However, organic matter will be reduced, as more secondary organic aerosol will be generated by increased VOC emissions. Therefore, the net change in $PM_{2.5}$ would be small. This result is consistent with a previous study (Tsimpidi et al., 2008), indicating that controlling industrial VOC emissions may not be an *efficient method of controlling $PM_{2.5}$. The simultaneous control of $PM_{2.5}$ and O_3 is a complex issue, and mitigation strategies will vary between areas with different formation regimes (i.e., VOC-limited, NOx limited, or NH_3-rich/poor) (Xing et al., 2019). NH_3 emissions need to be considered to further mitigate $PM_{2.5}$ concentrations in the PRD EZ, as NH_3 has also been detected in eastern China, as well (Geng et al. 2019).*

[Line 447-462]

Tsimpidi, A. P., Karydis, V. A., & Pandis, S. N. (2008). Response of fine particulate matter to emission changes of oxides of nitrogen and anthropogenic volatile organic compounds in the Eastern United States. Journal of the Air & Waste Management Association, 58(11), 1463-1473.

Geng, G., Xiao, Q., Zheng, Y., Tong, D., Zhang, Y., Zhang, X., ... & Liu, Y. (2019). Impact of China's air pollution prevention and control action plan on PM 2.5 chemical composition over eastern China. Science China Earth Sciences, 62(12), 1872-1884.

Xing, J., Ding, D., Wang, S., Dong, Z., Kelly, J. T., Jang, C., ... & Hao, J. (2019). Development and application of observable response indicators for design of an effective ozone and fine-particle pollution control strategy in China. Atmospheric chemistry and physics, 19(21), 13627-13646.

Minor comments:

19. Line128 " leading to in excess of 20% difference in premature mortality due to exposure to O3 ...", please add references to this conclusion.

Response: *Thanks for pointing out this. We add the reference.*

20. Line 244 " ...'point' and 'area' emissions categories are considered to represent primarily industrial, with VOC emissions affected by the 'half industry' control. ". However, when the authors consider the half emission of industry, it seems only "area" emissions was reduced by 50% based on Fig.2.

Response: *Thanks for the comments and questions. We are a little bit confused with the reviewer's question since the original Fig 2 is the simulated spatial distribution of O_3 concentrations instead of emissions. In terms of half industrial VOC case, we only cut VOC by 50% in the industry sector instead of cutting all emissions for all the pollutants. Fig. S3 clearly shows that the VOC emissions in Fig. S3b are reduced by around 50% for point (grey color) and area (blue color) sectors in the half industry VOC case. We consider the 50% reduction in both area and point sources sectors.*

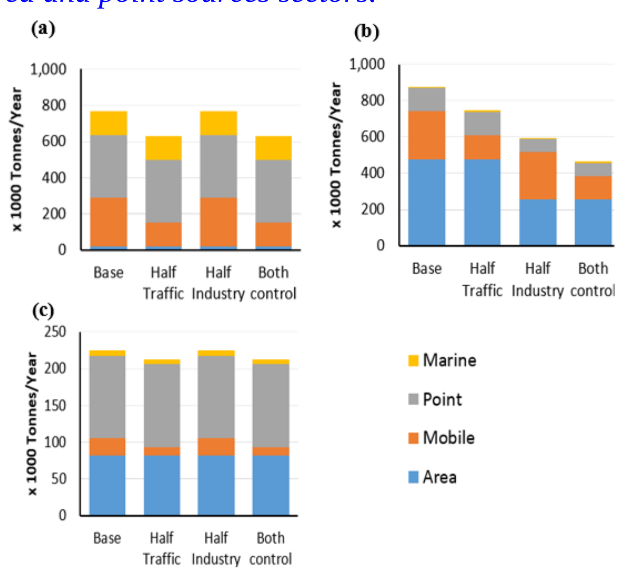

Figure S3. Emissions for the inner regional model domain covering central GBA for (a) NO_x, (b) VOC, and (c) $PM_{2.5}$. Unit: thousand tonnes per year.

21. Line 249 "However, it is important to note that total VOC emissions also have a large contribution from biogenic sources (around 50%)...". Does this statement mean the study area or in general sense? Please specify.

Response: *Thanks for the question. This statement means the accounted percentage of the VOC emissions amount. Although the proportions of biogenic and anthropogenic VOC may vary in different seasons and environmental conditions (e.g., temperature, humidity), a previous study estimated that biogenic emissions represent nearly 50% of China's total VOC emissions (Cao et al., 2018). Therefore, biogenic emissions are likely to contribute substantially to VOC emissions in the GBA. We put the statement here to illustrate the magnitude of the overall VOC reduction in this region. We adjust the content as follows to make it clearer.*

> *Although the proportions of biogenic and anthropogenic VOC may vary in different seasons and environmental conditions (e.g., temperature, humidity), a previous study estimated that biogenic emissions represent nearly 50% of China's total VOC emissions (Cao et al., 2018). Therefore, biogenic emissions are likely to contribute substantially to VOC emissions in the GBA.*

[Line 301-305]

Cao, H., Fu, T.M., Zhang, L., Henze, D.K., Miller, C.C., Lerot, C., Abad, G.G., Smedt, I.D., Zhang, Q., Roozendael, M.V. and Hendrick, F., 2018. Adjoint inversion of Chinese non-methane volatile organic compound emissions using space-based observations of formaldehyde and glyoxal. Atmospheric Chemistry and Physics, 18(20), pp.15017-15046.

22. The authors could possibly move Figure 2 to SI, as little discussions were involved in the main text.

Response: *Thanks for the suggestion. We remove the original Figure 2 and use the spatial distribution maps of difference for period-averaged O_3 concentrations (current Figure 2) to illustrate the results. We also rewrite the discussions of the main results for the current Figure 2 from Line 359 to 396.*

23. Please improve the resolution of Figure 5-7, and add the longitude and latitude.

Response: *Thanks for the suggestion. We replace Figure 5-7 (Now Figure 4-6) with the vector-based figures, which have high resolutions with detailed longitudes and latitudes.*

24. Line 304 Please indicate the predominant wind direction.

Response: Thanks for pointing out this. What we mean by "downwind" here is the downwind of the severe air pollution area, which is the north-east of Guangdong province. We re-write the description of Figure 2, so this sentence is removed. We also added a wind rose diagram in Figure 2 to make it clearer.

25. Please add the boundary in Figure S7(b).

Response: *Thanks for pointing out this. We have replaced Figure S7b (Now Figure S8b), adding the boundary.*

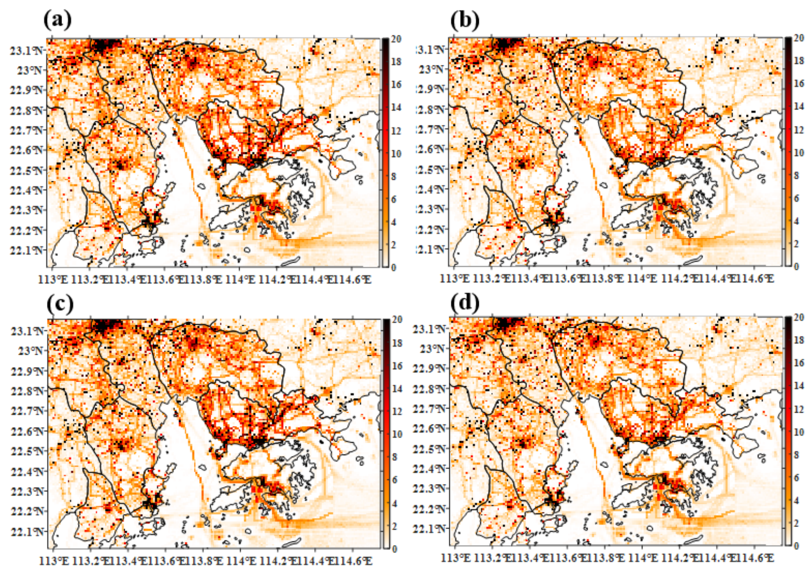

Figure S8. Daily column emission comparison of anthropogenic $PM_{2.5}$ for (a) Base case, (b) Half Traffic case, (c) Half Industry VOC case, and (d) Both control case. Unit: g/s.

26. Line 422 "," the comma should be changed to the period.
Response: Thanks for pointing out this. We have changed the "," to ".".

27. Please add scales in Figure 9 - 10.
Response: Thanks for pointing out this. We add scales for the two figures.

Reviewer #3 Evaluations:
Recommendation: Return to author for major revisions
Significant: The paper has some unclear or incomplete reasoning but will likely be a significant contribution with revision and clarification.
Supported: Yes
Referencing: Yes
Quality: The organization of the manuscript and presentation of the data and results need some improvement.
Data: Yes
Accurate Key Points: Yes

Reviewer #3 (Formal Review for Authors (shown to authors)):

Review comments on a manuscript entitled "Implications of Mitigating the Ozone and Fine Particulate Matter Pollution in the Great Bay Area Using a Regional-to-Local Coupling Model" authored by Zhang etc.

General comments: Anthropogenic emissions and model resolutions are two important factors driving ambient air pollutant predictions. In this study, the authors proposed to use a coupled regional-to-street-scale air quality modeling system (i.e., CMAQ-ADMS) rather than a regional model to assess the impact of anthropogenic emissions on surface O3 and PM2.5 predictions. They suggested that a more stringent control strategy of VOC emissions, especially from industrial sector has the largest mitigation effect on ambient levels of O3 and frequency of O3 episodes in the Great Bay Area in China. Overall, the manuscript is well written and organized. However, more details of the model description and in-depth discussion are required. Please see the comments below.

Specific comments.
28. The authors claimed that a coupled regional-to-local-scale modeling system was used in this study (Lines 45-46). Meanwhile, they pointed out that the CMAQ outputs were used to drive the ADMS-Urban dispersion model (L194-195). Thus, the term "coupled" is a little bit confused. I am not sure that CMAQ and ADMS are really coupled as an integrated modeling system or the only linkage between both is that the CMAQ outputs are used to drive the dispersion model runs. Some details on the "coupling" are necessary. Especially, a flow chart of the CMAQ-ADMS coupling system is helpful.
Response: *Thanks for the question and suggestion. In terms of the model coupling, we add the research framework (Figure S1) and also amend the related contents as follows.*

> *Figure S1 shows a research framework for the CMAQ–ADMS-Urban air quality modeling system. The street-scale resolution ADMS-Urban dispersion model was coupled with the regional CMAQ model using the ADMS-Urban Regional Model Link (ADMS-Urban RML) to investigate O_3 and $PM_{2.5}$ concentrations and the sensitivity of both pollutants to emissions from the traffic and industrial sectors. The ADMS-Urban RML is used to automatically prepare nested data from the regional model (CMAQ) and the meso-scale meteorological model (WRF) for the street-level ADMS-Urban model.* **[Line 187-193]**

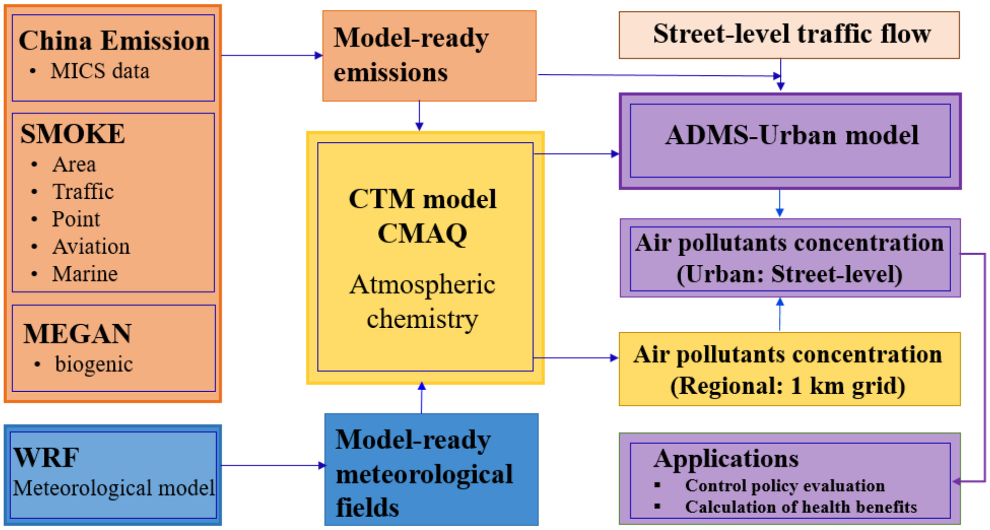

Figure S1. *Research framework for the CMAQ–ADMS-Urban modeling system.*

29. It is not clear how the ADMS-Urban dispersion model calculates the concentrations of gas-phase chemical species like O3 and NO2, and aerosol species like PM2.5. Does it include a gas-phase chemical mechanism for gas-phase species and an aerosol module for aerosol species predictions as CMAQ does? Or the ADMS model treats individual species as a tracer no matter it is gas or aerosol species? It will be helpful to provide a brief description on how the dispersion model predicts O3 since O3 is a secondary air pollutant and does not have emission sources.

Response: *Thanks for the questions. ADMS-Urban inherits the concentration outputs from the CMAQ model as background concentrations at each modeled hour. Most of the slow reactions are considered by the CMAQ chemical reaction scheme. The ADMS-Urban model is specialized in capturing the fine concentration gradient and rapid chemical reactions when emissions are released from pollution sources, such as in traffic settings. Over time, the concentration gradient lowers; the CMAQ model is then able to simulate regional transport and the associated chemical reactions. In addition to the CMAQ output, the following reaction sets are calculated by the ADMS-Urban model. For NO_x–O_3, the Generic Reaction Set (Malkin et al., 2016) is used with an extra reaction introduced, i.e., $2NO + O_2 => 2NO_2$. For sulfates, sulfur dioxide is oxidized to particulates via the reactions $2SO_2 + O_2 => 2SO_3$, $SO_3 + H_2O =>H_2SO_4$, and $H_2SO_4 + 2NH_3 => (NH_4)_2SO_4$. In order to make it clearer, we add the content as follows:*

> *ADMS-Urban inherits the concentration outputs from the CMAQ model as background concentrations at each modeled hour. Most of the slow reactions are considered by the CMAQ chemical reaction scheme. The ADMS-Urban model is specialized in capturing the fine concentration gradient and rapid chemical reactions when emissions are released from pollution sources, such as in traffic settings. Over time, the concentration gradient lowers; the CMAQ model can simulate regional transport and the associated chemical reactions. In addition to the CMAQ output, the following reaction sets are calculated by the ADMS-Urban model. For NO_x–O_3, the Generic Reaction Set (Malkin et al., 2016) is used with an extra reaction introduced, i.e., $2NO + O_2 => 2NO_2$. For sulfates, sulfur dioxide is oxidized to particulates via the reactions $2SO_2 + O_2 => 2SO_3$, $SO_3 + H_2O =>H_2SO_4$, and $H_2SO_4 + 2NH_3 => (NH_4)_2SO_4$.* **[Line 193-203]**

Malkin, T.L., Heard, D.E., Hood, C., Stocker, J., Carruthers, D., MacKenzie, I.A., Doherty, R.M., Vieno, M., Lee, J., Kleffmann, J. and Laufs, S., 2016. Assessing chemistry schemes and constraints in air quality models used to predict ozone in London against the detailed Master Chemical Mechanism. Faraday discussions, 189, pp.589-616.

30. Section 3.1: Why do the authors show the period-average prediction instead of daily maximum of 8-hr average O3 and 24-hr average PM2.5 during the simulation period? Does "period" represent two-month period? Is it possible to add a time series comparison rather than spatial patterns only between Base case and three sensitivities runs for the inner most domain or the regions rather than a location (Fig.10) only? The time series will help us to better understand how O3, PM2.5 and NO2 response to different emission control strategies during daytime, daytime, and on the days with and without episode cases.

Response: *Thanks for the questions. The period-average prediction is the hourly average value for the whole modeling running period, which lasts from April 1 to May 31 in 2019. The reason we choose this period is that frequent O_3 episodes happened during this period and the local government has greater interests in exploring the interesting mechanism in Guangzhou urban region. One-week spinning up time has been excluded. We choose to use the episode analysis instead of the time series for the whole running period mainly because the ADMS-Urban model is specialized in capturing the concentration gradient and fast chemistry reactions when emission is released from the pollution sources, such as the roadside environment. The ADMS-Urban inherits the concentration outputs from the CMAQ model as background concentration at each modelled hour. Most of the slow reactions are considered by the CMAQ chemical reaction scheme. Over time, the concentration gradient lowers, and the CMAQ model is then able to simulate the regional transport and the associated chemical reactions.*

In responding to the reviewer's questions, we add the following content.

> *The period-averaged prediction is the hourly average value for the entire modeling period (April 1–May 31, 2019). This period was chosen owing to the occurrence of frequent O_3 episodes, and the local government is particularly interested in exploring the mechanisms of occurrence in the Guangzhou urban area. A 1-week spin-up period was used.* **[Line 323-327]**

We also add the time series plots of typical stations for the base case for the regional model (Figure S10-S12) in Domain 4 and the street-level ADMS-Urban model (Figure S13-S15) in domain 5. In terms of the sensitivity scenarios, we choose to use the spatial plots to illustrate the results since the overall holistic impacts of the domain will be shown in this way. We choose Figure 9 to explain the reason for decreased O_3 concentration in a rural region (white star) in Figure 2.

31. Figure 1: Why did the authors only show simulated spatial patterns of NO2 for Base case and Half Traffic case but not for Half Industry VOC case? In addition, for the difference map, the authors only showed the between the case with Both Controls and Base case? I would suggest showing spatial pattern of simulations for the Base case and the difference maps between three sensitivity runs and the Base case individually.

Response: *Thanks for the questions. Showing different scenarios depends on different dominant factors. Figure 1 illustrates the spatial concentration plots of NO_2, in which the half-traffic case dominants at a substantial magnitude. Therefore, only traffic sector-related scenarios were shown here. Evaluating the impact of the VOC emissions on the NO_2 concentration is not a focus of the major targets. Instead, the half industrial VOC case was shown to illustrate its impacts on O_3 concentration in Figure 2. In responding to the reviewer's questions, we add the content as follows to make it clearer.*

Figure 1 shows the simulated spatial concentration maps of period-averaged NO_2 concentrations from the regional CMAQ model, in which the half traffic case dominants by a substantial margin. Therefore, the traffic sector-related scenarios are selected to demonstrate.

[Line 347-349]

32. Figures 2-3: It is suggested to combine both figures together. It will be better by showing spatial distribution map for the Base case, and difference maps between individual cases and Base case, like what you did for NO2 for consistency. The same suggestion should be suitable for PM2.5 (i.e., Figure 4).

Response: *Thanks for the suggestions. Since one reviewer suggested we remove Figure 2 (Question 22), a second reviewer suggested we add the wind direction for Figure 2 (Question 24). Finally, we remove original Figure 2 and use the spatial distribution maps of differences of period-averaged O_3 concentrations (current Figure 2 in the revised submission) to illustrate the main results. We also added a wind rose diagram based on the reviewer's suggestion. We re-write the description and discussion of Figure 2 from Line 359 to Line 396 to give ,more precise explanations.*

As we explained in question 31, Showing different scenarios depends on different dominant factors. Figure 1 illustrates the spatial concentration plots of NO_2, in which the half-traffic case dominants at a substantial magnitude. Therefore, only traffic sector-related scenarios were shown here. Evaluating the impact of the VOC emissions on the NO_2 concentration is not a focus of the major targets. A similar conclusion holds for $PM_{2.5}$. Therefore, we keep the original format for NO_2 (Figure 1) and $PM_{2.5}$ (Figure 3).

33. It is surprised that no changes in O3 predictions are found over the Hong Kong region since all the sensitivity studies applied the emission reductions for the whole CMAQ domain 4 (see Fig.3). Any explanations on that?

Response: *Thanks for the question. In our initial submission, Figure S7-S9 in the supporting information show there are no emission changes for the HK region. As answered in reviewers' Question 7 and 9, we assume no emission control actions in HK since we want to evaluate how the air pollution of the PRD EZ change corresponds to the local controls. All the scenario runs are based on the sensitivity of the PRD EZ since the Guangdong government is more interested in the air pollution changes of the PRD EZ when only local controls are applied.*

34. Figures 5-7: The figures' quality is not high. Please use English street name for the background map in all the figures. Please specify that all the replots were generated from the CMAQ or dispersion model simulations. It is better to add the locations of those three sites shown in Fig. 5 rather than Fig. S9 which can be deleted.

Response: *Thanks for the question. We replace all three plots with vector-based spatial concentration maps. We adjust all the titles to specify all the figures clearer. We deleted Figure S9 and recompiled Figure S2 to show the domains and monitoring stations clearer, also based on other reviewer's suggestion.*

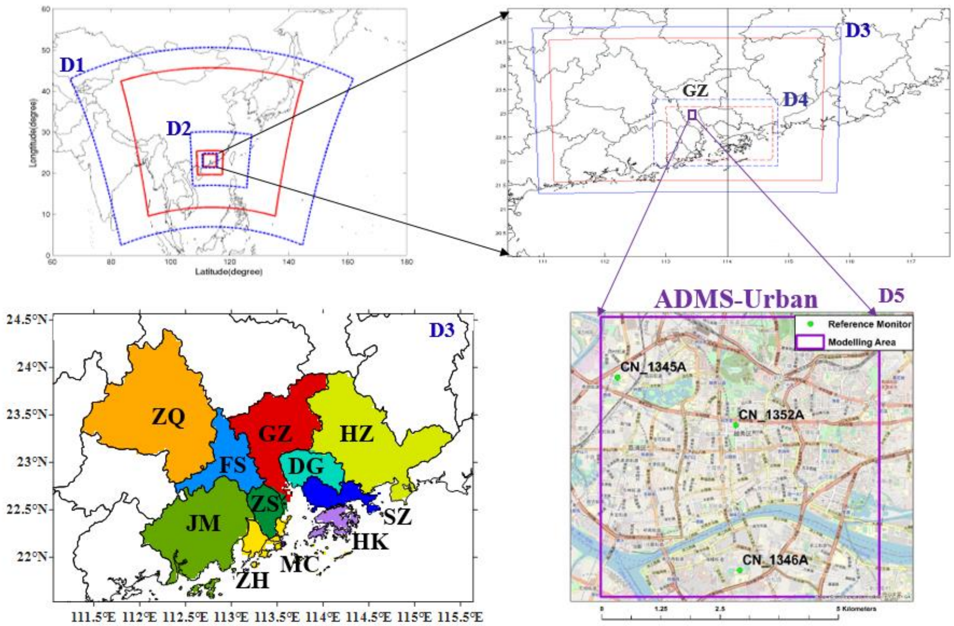

Figure S2. *Geographic domain setting of the CMAQ–ADMS-Urban modeling system: meso-scale meteorological model WRF (blue lines) and regional chemical transport model CMAQ (red lines) for D1 (27 km), D2 (9 km), D3 (3 km), D4 (1 km) and street-level ADMS-Urban model (purple lines) for D5 (6 km × 6 km). The Greater Bay Area (GBA) includes the PRD Economic Zone (Guangzhou (GZ), Shenzhen (SZ), Foshan (FS), Dongguan (DG), Zhuhai (ZH), Zhongshan (ZS), Jiangmen (JM), Huizhou (HZ), Zhaoqing (ZQ)), Hong Kong (HK), and Macau (MC).*

35. Section 3.3: Again, is it possible to compare their time series or period-average or monthly mean of diurnal patterns for a comparison?

Response: *Thanks for the question. We add the CMAQ model validation of the base case using both the statistical parameters and time series plots. Considering the limited observational stations in the urban domain 5 (6 km * 6 km) and aiming to show the advantages of the ADMS-Urban performance, we add the time series comparison of the base case for both the CMAQ model and the ADMS-Urban model at the three monitoring stations. We add the content as follows:*

> *Statistical parameter performances (Table 2) and time series plots for typical monitoring stations (Figures S10–S12) in the GBA were analyzed to validate the base case of the regional CMAQ model at a 1-km resolution. Table 2 clearly shows that the CMAQ model obtains an acceptable level of accuracy for $PM_{2.5}$ simulations (with mean fractional bias ≤ ±0.6 and mean fractional error ≤ 0.75) according to the criteria proposed by Hu et al. (2016). The averaged O_3 observation is 28.6 ppb, and the mean O_3 simulation is 28.9 ppb, with an Index of Agreement (IOA) of 0.63. Although the CMAQ model underestimates NO_2 concentrations by 2.5 ppb, the IOA of NO_2 is up to 0.57, and the root mean square error is around 10. Overall, the CMAQ model simulation is considered an acceptable input to drive the ADMS-Urban model. In addition to the capability of the CMAQ model to capture the main trend in the time series plots during the modeling period, Figures S13–S15 show the time series comparisons of the base case for both the CMAQ and ADMS-Urban models. Substantial improvement was observed during specific pollution episodes, which illustrates the advantages of coupled urban dispersion models.*

[Line 328-341]

36. Figure 8 shows a comparison of CMAQ- and ADMS-predicted daily maximum hourly NO2, daily maximum 8-hr average O3 and daily average PM2.5 at the three selected urban sites in Guangzhou. Please have a double check the location of the three sites and make sure they are in the inner most domain D4 (Figure S2) since the inner most domain (D4) of CMAQ only cover southern part of Guangzhou.
Response: *Thanks for the question. We make sure that the three sites are in the Domain 4. The adjusted Figure S2 could show it clearly.*

37. L17-18 and L50-52: It is not surprised, or it well known that ozone formation in an urban area is dominated by the VOC-limited regime. I do not think this is a new finding.
Response: *Thanks for the question. We understand larger-scale air quality modeling or observation-based researches have concluded such findings. However, the model resolution is a key factor impacting the accuracy of the modeling results. This conclusion was held when our regional-to-local scale air quality modeling system CMAQ–ADMS-Urban was implemented successfully in urban Guangzhou. In responding to the reviewer's comments, we change the wording in Line 17-18.*

> *The O_3 formation regime in Guangzhou is VOC-limited, and the traffic sector is of paramount importance for controlling NO_x and O_3.* **[Line 17-18]**

> *The reduced traffic scenario leads to reduced NO_2 and $PM_{2.5}$ but increased O_3 concentrations in urban areas. Guangzhou is located in a VOC-limited O_3 formation regime, and traffic is a key factor in controlling NO_x and O_3.* **[Line 51-53]**

38. L142-144: If ADMS-Urban model considers street canyon effects that may improve air pollutants' (e.g., O3 and PM2.5) predictions. I do not recall that the authors used this augment to interpret the model results and the comparisons with the regional model predictions.
Response: *Thanks for the question. The ADMS-Urban model has the capability of capturing a more fine concentration gradient and rapid chemical reactions when emission is released from the pollution sources, such as the roadside environment. We do not consider the street canyon effects in the current study. This will be included in our future studies.*

39. L160-164: What I learned from here is, the authors used a regional model's output with temporal and spatial variation rather than a constant measurement to drive the ADMS-Urban dispersion model. This can be considered as a major contribution of this study in terms of methodology effort(s). Can the authors further illustrate the improvement of ADMS-Urban predictions on street-level resolution by using a regional model output as the model inputs as compared to the case by using measurements as the model inputs?
Response: *Thanks for the question. Biggart et al. (2020) claimed in the paper that their model is over-estimating the NO_2, which may be due to the assumption of using constant measurements data. We admit this comparison will be interesting. However, it is hard to make such a comparison mainly because the observations data in the current model domain (6 km * 6 km) is too sparse. There are insufficient measurements data to carry out the test scenario. The ADMS-Urban model needs to capture a background concentration at each time step from the upper wind direction of the targeting points, which are lacking at the moment.*

Biggart, M., Stocker, J., Doherty, R.M., Wild, O., Hollaway, M., Carruthers, D., Li, J., Zhang, Q., Wu, R., Kotthaus, S., 2020. Street-scale air quality modelling for Beijing during a winter 2016 measurement campaign. Atmospheric Chemistry and Physics 20, 2755-2780.

40. L263-265: I am curious why you did not present total VOCs and anthropogenic VOCs?
Response: *Thanks for the question. Total VOC emissions include the biogenic and anthropogenic VOCs. We claimed the annual total comparison of the anthropogenic VOCs in our initial submission in Figure S3.*

> *A summary of total annual anthropogenic NO_x, VOC, and $PM_{2.5}$ emissions for the regional model domain covering central GBA is presented in Figure S3.* **[Line 295-296]**

We also add content about the proportions of the biogenic VOCs and anthropogenic VOC in Line 301-305. However, we do not consider the biogenic VOC in this study. The current plot of Figure S4 is a typical species of the CMAQ model-ready daily column emission total comparison, which are produced based on the SMOKE model output.

41. L308 and L308: The statement of "lower oxidant emissions" (L308) has a conflict with "in less oxidant (in this case, O3)" since O3 does not have emissions.
Response: *Thanks for the question. We delete the "emissions" to make it consistent.*

42. L291-318: The authors seem to simply repeat the discussion of Figure 1 for that of Figures 2-4. Some in-depth analyses are required.
Response: *Thanks for the questions. The reviewer has such an impression owing that the three pollutants are discussed for the same sensitivity scenarios. In responding to the reviewer's question, we adjusted the previous content and added a more in-depth analysis from the chemical perspective in section 3.1 from Line 343 to 408.*

43. Figures 8, 9 and 10: Can you use statical parameters to verify the model performance and then quantify the differences between the Base case and sensitivity runs?

Response: *Thanks for the question. We agree that using the statistical parameters is a good way to verify the model performance for the regional CMAQ model. However, the statistical parameters cannot be used to quantify the differences between base case and sensitivity runs since there are no observations for the sensitivity scenarios. The statistical parameters are less efficient to validate the ADMS-Urban model since the current designed domain (6 km * 6 km) of urban Guangzhou covers only three monitoring stations (an urban station in the north, an urban station in the south, and another roadside station), which have been shown in Figure 2.*

In responding to the reviewer's question, we add the statistical performance of the CMAQ model (table 2) compared with the monitoring data in domain 4 (1 km resolution). We also add the time series comparison of typical monitoring stations for the CMAQ base case in Figure S10-S12, at 1 km resolution. In order to better illustrate the ADMS-Urban model shows improved performance, we add the time series comparison of the base case for both the CMAQ model and ADMS-Urban model in Figure S13-15. We add the content as follows:

> *Statistical parameter performances (Table 2) and time series plots for typical monitoring stations (Figures S10–S12) in the GBA were analyzed to validate the base case of the regional CMAQ model at a 1-km resolution. Table 2 clearly shows that the CMAQ model obtains an acceptable level of accuracy for $PM_{2.5}$ simulations (with mean fractional bias $\leq \pm 0.6$ and mean fractional error ≤ 0.75) according to the criteria proposed by Hu et al. (2016). The averaged O_3 observation is 28.6 ppb, and the mean O_3 simulation is 28.9 ppb, with an Index of Agreement (IOA) of 0.63. Although the CMAQ model underestimates NO_2 concentrations by 2.5 ppb, the IOA of NO_2 is up to 0.57, and the root mean square error is around 10. Overall, the CMAQ model simulation is considered an acceptable input to drive the ADMS-Urban model. In addition to the capability of the CMAQ model to capture the main trend in the time series plots during the modeling period, Figures S13–S15 show the time series comparisons of the base case for both the CMAQ and*

ADMS-Urban models. Substantial improvement was observed during specific pollution episodes, which illustrates the advantages of coupled urban dispersion models. **[Line 328-341]**

In terms of Figure 9 and Figure 10 (currently named Figure 8 and 9), both figures are used to explore the concentration changes for a rural location (white star in Figure 2) from the regional CMAQ model, instead of showing the advantages of ADMS-Urban. The rural location is out of the urban domain 5 of the ADMS-Urban model. Therefore, we choose to keep both of the figures.

Hu, J., Chen, J., Ying, Q. and Zhang, H., 2016. One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system. Atmospheric Chemistry and Physics, 16(16), pp.10333-10350.

44. Figure 10: Is it possible to include the dispersion model predictions in the time series for a comparison? That may further illustrate the advantage of the high-resolution or street-level scale model results as compared to the regional model predictions.
Response: *Thanks for the question. Figure 10 (currently named Figure 9), which shows the comparison of the rural location of the while star (In Figure 2), is out of the ADMS-Urban model domain (see Figure S2). Therefore, we cannot add the dispersion model predictions.*

45. L450: It is strange that a VOC-limited regime was identified near a rural area downwind. Do you have any idea on that? Or what is the indication to your study?
Responses:
Response: *Thanks for the question. This is a measurement work combined with a Box model done by another research group in the PRD region (He et al., 2019). It shows the VOC-limited O_3 formation regime in a rural monitoring site (Heshan station) during Oct and Nov 2014. Since this study is only focused on one-month measurements and for a specific period (autumn), More work related to the VOC/NOx ratio and O_3 formation regime needs to be done in the future. We remove this sentence when we re-write the discussion part, suggested in Question 46.*

He, Z., Wang, X., Ling, Z., Zhao, J., Guo, H., Shao, M., Wang, Z., 2019. Contributions of different anthropogenic volatile organic compound sources to ozone formation at a receptor site in the Pearl River Delta region and its policy implications. Atmospheric Chemistry and Physics 19, 8801-8816.

46. I want to know what the difference between "Discussion" and "Conclusion" parts is. To me, the former is just a summary of this study, I do not get additional points or indication from this discussion.
Response: *Thanks for the question. We amend the discussion part by adding more discussions compared with the current literature and also add the limitation of the current study.*

47. L505-506: It seems that the street-scale ADMS0-Urban model shows an improvement in capturing a sharp concentration gradient near traffic streams spatially. What about improvement in terms of temporal variation? Did the authors get a chance to do such a temporal comparison?
Response: *Thanks for the questions. The idea of coupling the CMAQ model with the ADMS-Urban model is to resolve the concentration gradients adjacent to the road emission sources. ADMS-Urban model is responsible for capturing the rapid chemical reactions near the road in this study instead of exploring the temporal improvement, which is out of scope. We may consider it in future studies.*

48. Figures S2: Why do you present emission comparisons using the unit of KT/year while other figures (Fig.S3-S8) use different units with g or mole per second? Is it possible to make them be consistent?

Response: *Thanks for the questions. The emission total comparison is based on the raw annual emission data, which is in tonnes or KT per year. Figs. S3-S8 are the plots of the SMOKE model output, which describes the emission intensity. Therefore, the units are different.*

Minor comments or technique issues

49. Lines 87-90: I do not think this sentence has a direct implication on this study.

Response: *Thanks for the comments. We change the wording as follows:*

> …… greater efforts have been made to alleviate air pollution …… **[Line 89]**

50. L127-129: Any reference(s) for this?

Response: *Thanks for the question. We add the reference.*

51. L145: Please define the H/W ratio(s)?

Response: Thanks for the question. We add the following content to define the H/W ratio.

> *In the ADMS model, the H/W ratio is considered in street canyons, where a street is flanked by buildings on both sides to form a canyon-like environment. The H/W ratio is defined as the average building height on both sides of the street canyon divided by the distance between the two sides.* **[Line 152-155]**

52. L155: ADMS should be defined at the first time when it was used.

Response: *Thanks for the question. We define ADMS as follows:*

> *The local modelling of road sources in an Urban Atmospheric Dispersion Modelling System (ADMS-Urban) can include street canyon effects……* **[Line 149-151]**

53. L178-182: You should move them to reference review in Introduction Section.

Response: *Thanks for the suggestion. The three references listed here were published by the first author of this manuscript. We listed those three references here to illustrate the configuration of the regional model CMAQ is reliable since it has been applied in different studies of our modeling group. In order to avoid confusing readers, we adjusted it as follows instead of moving them to the reference review in the introduction section.*

> *The regional CMAQ model applied in this study is the same as that used to assess holistic emission control policies (Zhang et al., 2020), combined health effects (Zhang et al., 2021a), and data assimilation of model bias corrections (Zhang et al., 2021b) in our previous publications.*
>
> **[Line 204-207]**

54. L272: I cannot find the locations of Shenzhen, Dongguan, and Guangzhou in any of the figures. This will be difficult to the readers who are not familiar with those cities.

Response: *Thanks for the comments. In order to make it clearer, we combine Figure S2 (regional model domains) and Figure S9 (street-scale model domain) and then add the geographic locations of all the cities in the Greater Bayer Area.*

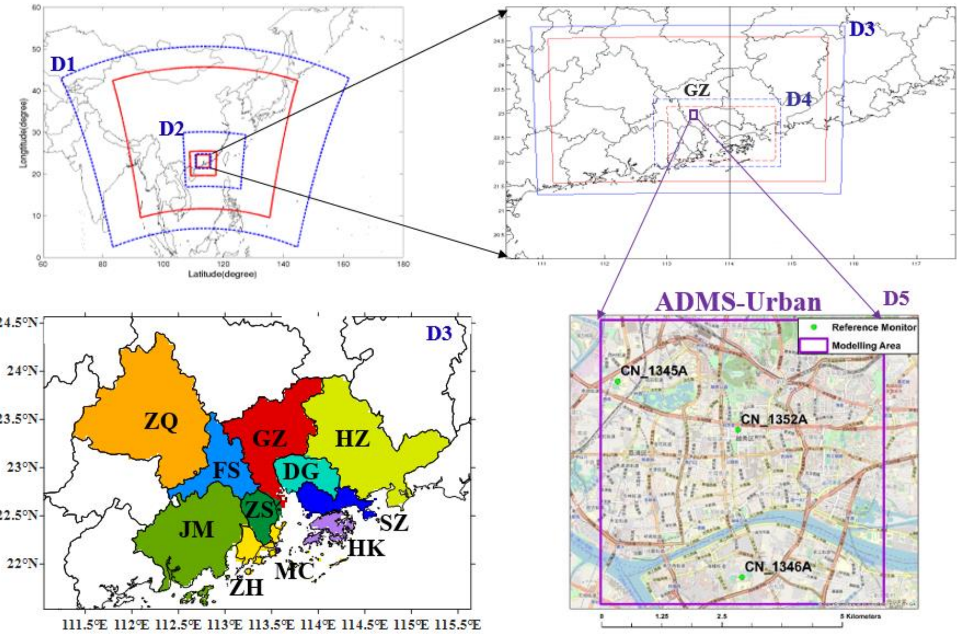

Figure S2. *Geographic domain setting of the CMAQ–ADMS-Urban modeling system: meso-scale meteorological model WRF (blue lines) and regional chemical transport model CMAQ (red lines) for D1 (27 km), D2 (9 km), D3 (3 km), D4 (1 km) and street-level ADMS-Urban model (purple lines) for D5 (6 km × 6 km). The Greater Bay Area (GBA) includes the PRD Economic Zone (Guangzhou (GZ), Shenzhen (SZ), Foshan (FS), Dongguan (DG), Zhuhai (ZH), Zhongshan (ZS), Jiangmen (JM), Huizhou (HZ), Zhaoqing (ZQ)), Hong Kong (HK), and Macau (MC).*

55. L273-274: The statement of "The reduction in VOC has no effects on NOx and PM2.5 emissions" is meaningless.
Response: *Thanks for pointing out this. We delete this statement.*

56. Figure 9 and L395: where is "a rural site"? Can you add the location of the rural site in Figure 1?
Response: *Thanks for the question. It is a rural location instead of a rural site, which was marked as a white star in Figure 2 in our initial submission. We change the wording in the title of Figure 9 (currently named Figure 8). The reason we marked it in Figure 2 is that the O_3 concentration decreases because of NOx reduction. We choose a typical rural location to illustrate the pollution changes of O_3, $PM_{2.5}$, and NO_2 for the respective sensitivity scenarios.*

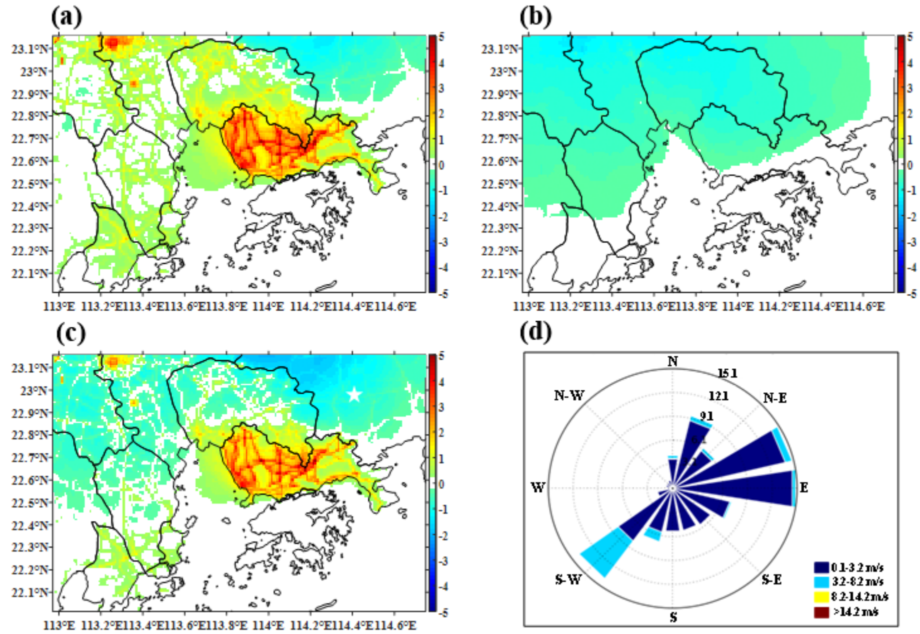

Figure 2. Simulated spatial maps of period-averaged O_3 concentrations - Difference plots from the CMAQ model for (a) Half Traffic – Base case; (b) Half Industry VOC – Base case, and (c) Both controls – Base case (ppb); (d) Wind Rose diagram showing regional wind directions.

57. Figure 10 and L424: Where is "a location to the north-east of the model main"? Can you add it to Figure 1?

Response: *Thanks for the question. Figure 10 (currently named Figure 9) discusses the rural location region, which was marked as a star in Figure 2. We change the wording of the title of Figure 10 to make it clearer.*

Reviewer #4 Evaluations:
Recommendation: Return to author for minor revisions
Significant: Yes, the science is at the forefront of the discipline.
Supported: Yes
Referencing: Yes
Quality: Yes, it is well-written, logically organized, and the figures and tables are appropriate.
Data: Yes
Accurate Key Points: Yes

Reviewer #4 (Formal Review for Authors (shown to authors)):

This manuscript implemented a new coupled regional-to-local scale air quality modeling system (CMAQ-ADMS-Urban) to explore the sensitivity of the emission controls on the ozone and fine particulate matter pollution, covering the traffic and industrial sectors. The VOC-limited O3 formation regime was found and the further industrial VOC controls were called to be strengthened using this coupled regional-to-local scale air quality modeling system. Higher-resolution coupled modeling techniques are of great importance for large urban clusters such as Greater Bay Area and Hong Kong with high-rise tall buildings. It is also very important for refining the health exposure calculations in urban clusters. Very few applications of ADMS-Urban coupled with regional CMAQ model were applied in urban cities in China. With the direct coupling application of the ADMS-Urban development & maintenance team's work, the research findings focusing on the GBA will give more hints and implications in future governmental policymaking and applications of the high-resolution modeling. The modeled scenarios are clear and reasonably designed. The manuscript has been well structured and written. Therefore, I recommend to accept the manuscript (minor revision) after clarifying the following minor questions and typos.

Response*: Thanks a lot for the positive comments. We carefully read the proposed questions and answered them as follows.*

58. One question is about the computing time of the coupled model system (CMAQ and ADMS-Urban) at a very high resolution. How much extra computing power have been increased when coupling the ADMS-Urban model, comparing with running the regional CMAQ model alone? Would the computing power be acceptable and doable when expanding the current ADMS model domain to a larger one such as the whole GBA?

Response*: Thanks for the questions. For the domain mentioned, it takes 1-2% more computational time to complete coupled model than the CMAQ model itself. Our current system has been set up for the entire city of Hong Kong (Che et al., 2020), with acceptable computing time for forecast purposes. Some tests were also done on some parts of Shenzhen, and it is believed that the computational time should not be a major concern if expanding to the entire GBA.*

Che, W., Frey, H.C., Fung, J.C., Ning, Z., Qu, H., Lo, H.K., Chen, L., Wong, T.-W., Wong, M.K., Lee, O.C., 2020. PRAISE-HK: A personalized real-time air quality informatics system for citizen participation in exposure and health risk management. Sustainable Cities and Society 54, 101986.

59. Line 140: The "double-counting" of the emissions in local model seems confusing, please explain more about that.

***Response**: Thanks for the comments. Our system does not simply provide emission data twice to the system, once to the regional model, and another once to the local model. We balanced the additional high-resolution emissions in the local urban-scale model by giving the same magnitude but negative amount of emissions in a gridded form on the same gridding of the regional CMAQ model to emulate the effect of removing such emissions of pollutants from the grid of the regional model. The coupled modeling system is redistributing emissions within each of the regional model grids for the purpose of running the local urban-scale model. In order to make it clearer, we add the following clarification:*

> *We balance the additional high-resolution emissions in the local urban-scale model by using a corresponding negative amount of emissions at the same gridding as the regional CMAQ model to emulate the effects of removing such emissions from the regional model grid. The coupled modeling system redistributes emissions within each of the regional grids for the purpose of running the local urban-scale model.* **[Line 145-149]**
>
> *Two sets of explicit traffic emissions were prepared: one set emulated the CMAQ grid concentrations that were distributed evenly across the traffic emissions model grid and extracted from the CMAQ model grids; the second set redistributed the CMAQ grid traffic emissions into explicit high-resolution traffic emissions within the facilitated road network. The emulated ADMS-Urban concentrations using the evenly distributed grid emissions were reduced from the ultimate ADMS-Urban concentration calculations to avoid double-counting of emissions. The allocation formula for redistributing the traffic emissions is detailed in Biggart et al. (2020). The road length was determined and calculated on the basis of the CMAQ grids.* **[Line 245-253]**

60. Table 1: what does the regional model and local model mean here? Please indicate it clearly in the table title.
Thank

***Response**: Thanks for the questions. We adjust the table title as follows to make it clearer.*

Table 1. Scenario design for the CMAQ–ADMS-Urban coupling system integrating the regional CMAQ model and local street-level ADMS-Urban model.

Scenarios	I. Base case	II. Half Traffic case	III. Half Industry VOC case	IV. Both Control case
Scenario description	Business As Usual (BAU)	50% reduction in traffic emissions	50% reduction in industrial VOC emissions	Scenarios II & III
Regional CMAQ model emissions	BAU	50% emission reduction in Mobile sector (all pollutants)	50% emission reduction in VOC from Industrial sector	50% emission reduction in a) mobile sector (all pollutants) and b) VOC emissions from the industrial sector
Local street-level model emissions	BAU	50% reduction in emissions from explicitly defined road traffic sources	BAU	50% reduction in emissions from explicitly defined road traffic sources

61. Line 135 and 149: Please indicate what the "two-pollutant" mean?

***Response**: Thanks for the question. The "Two-pollutant" means the O_3 and $PM_{2.5}$. We adjust the content as follows in order to make it clearer.*

> *……explore pollution sources or estimate health impacts for O_3 and $PM_{2.5}$ in the coupled systems……* **[Line 136-137]**

……investigate the traffic and industrial contributions to complex coupled O_3 and $PM_{2.5}$ issues through testing of emissions scenarios. **[Line 159-161]**

62. Figure 9 and 10: Please rename the "Scenario 2, 3, 4" to the scenarios defined in table 1.
***Response**: Thanks for the question. We correct those labels.*

63. Line 59, 620: The page number and volume number are missing in these two references.
***Response**: Thanks for pointing it out. We add the page number and volume number to the references.*

Cheng, J., Tong, D., Zhang, Q., Liu, Y., Lei, Y., Yan, G., Yan, L., Yu, S., Cui, R.Y., Clarke, L., 2021. Pathways of China's $PM_{2.5}$ air quality 2015–2060 in the context of carbon neutrality. National Science Review. 0, nwab078.

Lam, Y.F., Cheung, C.C., Zhang, X., Fu, J.S., Fung, J.C.H., 2021. Development of New Emission Reallocation Method for Industrial Nonpoint Source in China. Atmospheric Chemistry and Physics, 21, 12895–12908.

64. Line 673: Capital character of the Journal name. Please check all the reference.
***Response**: Thanks for pointing it out. We correct the journal name and also check it for all the references.*

65. Line 538: The project number of the PRAISE-HK is missing?
***Response**: Thanks for pointing it out. There is no specific project number for the PRAISE-HK project since it is a charity program funded by the HSBC 150th Anniversary Charity Program. The project webpage could be found at: http://praise.ust.hk/*

Other adjustments by authors:

1. We add one more project (Project number: T24/504/17), which provides parts of the resources.
2. We add the "Data Availability Statement" to show the sources of the supporting data in this manuscript.