1 2	Implications of Mitigating the Ozone and Fine Particulate Matter Pollution in the				
3	Greater Bay Area Using a Regional-to-Local Coupling Model				
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14	Key Points:				
15	• A novel regional-to-local coupled model was set up to explore the likely impact of				
16	emissions reductions and pollution mitigation pathways.				
17	• Substantial NO <sub>x</sub> controls in the traffic sector worsened the urban O <sub>3</sub> concentration,				
18	revealing the VOC-limited formation regime.				
19	• Probing into frequent summer O <sub>3</sub> episodes emphasized the value of more stringent				
20	VOC controls, in particular for the industrial sector.				
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## 41 Abstract

- 42 Ultrahigh-resolution air quality models resolving sharp concentration gradients benefit health
- 43 calculations. Mitigating fine particular matter (PM<sub>2.5</sub>) in the past decade triggered ozone (O<sub>3</sub>)
- 44 deterioration in China. Effectively controlling both pollutants is still understudied from an
- 45 ultrahigh-resolution perspective. This study proposes a coupled regional-to-local-scale
- 46 modeling system suitable for quantitatively mitigating pollution pathways at various
- resolutions. Sensitivity scenarios for controlling nitrogen oxide (NO<sub>x</sub>) and volatile organic
   compounds (VOC) emissions, concentrating on traffic and industry sectors, are explored. The
- results show that concurrent controls on both sectors lead to an overall 17%, 5%, and 47%
- emission reduction in NO<sub>x</sub>,  $PM_{2.5}$  and VOC, respectively. The 50% less traffic scenario leads
- to reduced NO<sub>2</sub> and PM<sub>2.5</sub>, but increased  $O_3$  concentrations in urban areas, revealing a VOC-
- 52 limited regime. The reduced industrial VOC emission scenario leads to reduced  $O_3$
- 53 concentrations throughout the mitigation domain. The maximum decrease in the median of
- hourly NO<sub>2</sub> is over  $11 \mu g/m^3$ , while the maximum increase in the median of maximum 8-hour
- rolling  $O_3$  is over 10  $\mu$ g/m<sup>3</sup> for the reduced traffic scenario. When both the traffic and
- industrial controls are applied, the impact on  $O_3$  reduces to an increase of less than 7  $\mu$ g/m<sup>3</sup>.
- 57 The daily-averaged PM<sub>2.5</sub> decreases by less than  $2 \mu g/m^3$  for the reduced traffic scenario and
- varies little for the reduced VOC scenario. An  $O_3$  episode analysis for both controls scenarios
- <sup>59</sup> leads to O<sub>3</sub> decreases of up to 15  $\mu$ g/m<sup>3</sup> (8-h metric) and 25  $\mu$ g/m<sup>3</sup> (1-h metric) in a rural area
- 60 to the northeast of the mitigation domain.
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## 62 Plain Language Summary

- 63 Spatial concentration maps of air pollutants showing variations over small distances are
- useful for assessing human health in metropolitan regions. The combined control of fine
- 65 particulate matter and ozone has not yet been fully understood at high resolution. This study
- 66 implements a regional-to-local urban modeling system for quantitatively assessing
- 67 possibilities for pollution reduction. Typical air pollutants are explored, concentrating on
- emissions from traffic and industry source sectors. We find that combined control on both
- 69 sectors leads to considerable emission reductions. The scenario analysis reveals the most
- <sup>70</sup> substantial factors affecting ozone pollution in various locations. The contributions of air
- 71 pollution from both sectors are assessed. The research findings would improve the awareness
- 72 of air quality management strategies and benefit multi-level governments for joint control of
- regional air pollution problems.
- 74

# 75 Index Terms

- 76 3355 Regional modeling (4316); 0345 Pollution: urban and regional (0305, 0478, 4251,
- 4325); 0545 Modeling (1952, 4255, 4316); 3329 Mesoscale meteorology; 6620 Science
- 78 policy (0485, 4338).
- 79
- 80 Keywords
- 81 street-scale; air dispersion model; CMAQ-ADMS-Urban; sensitivity analysis; ozone; Greater
- 82 Bay Area.
- 83

#### 84 **1 Introduction**

Air pollution has generated significant interest in recent years due to its adverse 85 effects on human health (Che et al., 2020; Conibear et al., 2021; Wu et al., 2019) and 86 mitigation of climate change (Li et al., 2019c; Qin et al., 2017). It has become an urgent and 87 intractable issue since Chinese president Xi announced a bold pledge that Chinese carbon 88 emissions would peak before 2030 and China would become carbon neutral by 2060 (Cheng 89 et al., 2021; Cui et al., 2021). The Chinese government has devoted tremendous efforts and 90 released a series of emissions control policies to address this challenge (Cai et al., 2018; Jiang 91 92 et al., 2015; Wu et al., 2019; Zhang et al., 2020). Zhang et al. (2020) revealed a holistic emissions control picture by utilizing a chemical transport model method to assess the 93 implemented emissions control policies covering various sectors during the 13<sup>th</sup> Five-Year-94 Plan in China. In response to the stringent national controls, ambient fine particulate matter 95 (PM<sub>2.5</sub>) has decreased substantially while ozone (O<sub>3</sub>) pollution levels are becoming severe (Li 96 97 et al., 2019b; Zhao et al., 2021). The combined health effect has been improved during the 98 past decade (Zhang et al., 2021a), although high O<sub>3</sub> concentrations have an adverse effect on habitats (Yli-Pelkonen et al., 2017), so they must, in the long-term, also be controlled. 99 Strategies for effectively controlling the absolute concentrations of O<sub>3</sub> and PM<sub>2.5</sub> 100 simultaneously in urban regions, where people spend the majority of their time, are of 101 increasing interest and importance. 102

A growing number of studies have devoted great efforts to the mechanism of coupled 103 O<sub>3</sub> and PM<sub>2.5</sub> pollution levels in various regions in China. Li et al. (2019a) firstly found the 104 major reason for worsening O<sub>3</sub> was due to the significantly reduced PM<sub>2.5</sub> that led to fewer 105 hydroperoxy radicals for O<sub>3</sub> formation since 2013 in the North China Plain (NCP). Zhao et al. 106 107 (2021) confirmed the interactions between the two pollutants and called for the concurrent control of both pollutants following analysis of four-year observational data in China. Li et al. 108 (2019b) proposed to reduce the nitrogen oxide  $(NO_x)$  and volatile organic compound (VOC)109 110 emissions aggressively to control both pollutants in the NCP by analyzing summer O<sub>3</sub> surface 111 data from 2013-2018. Gong et al. (2021) utilized the Community Multiscale Air Quality (CMAQ) model at 12 km resolution to trace the precursors of PM<sub>2.5</sub> and O<sub>3</sub>; the cross-112 113 boundary transport effect on the two-pollutant interaction was found in the Yangtze River Delta (YRD). In addition, Li et al. (2021a) concluded that the industrial and traffic emission 114 source sectors dominated contributions to both pollutants in YRD, also through CMAQ at 12 115 km resolution. Thus recent literature motivates studies on the impact of NO<sub>x</sub> and VOC 116 emissions from traffic and industry in China. 117

Other studies have focused efforts on quantifying the effect of model resolutions on 118 air pollution simulations and health calculations. A previous study in the US proved that finer 119 resolution (comparing 4 km and 12 km grid spacing) of a CMAQ model was better for 120 calculating health effects in urban areas; the results were comparable in rural areas (Jiang and 121 Yoo, 2018). Tao et al. (2020) found that the finer-resolution modeling could better capture 122 and reproduce the temporal trends and magnitudes of meteorological conditions and air 123 quality in Beijing. On the contrary, a local study indicated that grid resolution has little effect 124 on the simulation of PM<sub>2.5</sub> and O<sub>3</sub> in YRD (Wang et al., 2021). Liu et al. (2020) demonstrated 125 that model resolution did not significantly improve predictions for PM<sub>2.5</sub> and daily maximum 126 8-hour O<sub>3</sub> in Nanjing. However, the spatial distributions of both pollutants were better 127 captured with a finer resolution model, leading to in excess of 20% difference in premature 128 mortality due to exposure to O<sub>3</sub>. The impact of model resolutions on pollutant simulations 129 and health calculations varied in different cities or regions (urban or rural) within an 130 agglomeration. Consequently, it is of great interest to explore whether higher resolution 131 modeling techniques benefit model simulations for highly urbanized cities. 132

Previous studies have, for the most part, used a coarse resolution (>1 km) model and 133 an observations-oriented method to explore pollution sources or calculate health effects for 134 the coupled two-pollutant system (Li et al., 2021b; Silver et al., 2020). Although a previous 135 study (Silveira et al., 2019) summarized a great number of coupled regional-to-local models 136 which have been applied in worldwide urban regions, the model calculation algorithms and 137 assumptions varied widely between studies. The presented regional-to-local scale coupling 138 system follows the approach introduced in (Hood et al., 2018; Stocker et al., 2012). In this 139 system, double-counting of local emissions is avoided by deducting local modelling of all 140 emissions represented as grid sources from the regional modelling of all emissions before 141 adding street-scale local modelling with explicit and gridded sources. The local modelling of 142 road sources in ADMS-Urban can include street canyon effects, which affect the predicted 143 concentrations both inside and outside the canyon (Hood et al., 2021). The ADMS-Urban 144 street canyon module was designed to account for street canyons with higher H/W ratios than 145 the popular Operational Street Pollution Model (OSPM) model, which was developed for 146 H/W ratio around 1. Very few studies have applied coupled regional and very high-resolution 147 (street-level) modeling techniques to investigate the traffic and industrial contribution to the 148 complex coupled two-pollutant problem through emission scenario testing. A street-scale 149 model that provides a detailed representation of the spatial variation of pollutant gradients has 150 clear advantages in terms of calculating health exposure, compared with previous studies 151 applying regional or global models. 152

Although urban-scale models have investigated air pollution interactions in 153 megacities, very few coupled model systems were applied based on an Urban Atmospheric 154 Dispersion Modelling System (ADMS-Urban) and CMAQ model for the metropolitan region 155 of the Greater Bay Area (GBA). Zhang et al. (2015) integrated CMAQ and the CALifornia 156 PUFF (CALPUFF) model to simulate the contribution of SO<sub>2</sub> concentration due to local 157 emissions in GBA region. A regional European Monitoring and Evaluation Program Unified 158 Model for the UK (EMEP4UK) was coupled with ADMS-Urban to perform street-level air 159 pollutant simulations in London (Hood et al., 2018). Although ADMS-Urban was applied 160 within the sixth-ring in Beijing for simulating various pollutants (Biggart et al., 2020), the 161 background concentration level was adapted directly from measurement data and was 162 assumed uniformly distributed across the model domain, instead of coupling with a regional 163 model. As a result, the current study is the first localized coupled regional-to-local scale 164 model system consisting of ADMS-Urban and CMAQ to assess the sensitivity of two 165 pollutants to emissions from traffic and industrial sectors in the GBA. The motivation for 166 targeting the two sectors relates to the importance of the O<sub>3</sub> precursors, VOC and NO<sub>x</sub>, from 167 the anthropogenic industrial and traffic sectors in GBA, respectively. Consequently, assessing 168 the impact of changes to emitted NO<sub>x</sub> (and hence NO<sub>2</sub>) and VOCs is of interest. Section 2 of 169 this article describes the regional and local street-scale model configuration and the 170 sensitivity scenarios. Regional and local model simulation results and the model performance 171 of selected monitoring stations are discussed in section 3. Section 4 presents a discussion of 172 173 research findings, followed by conclusions in section 5.

## 174 **2 Research Methods**

175 2.1 Model configuration

The street-scale resolution ADMS-Urban dispersion model has been coupled with the CMAQ regional model configuration to investigate O<sub>3</sub> and PM<sub>2.5</sub> concentrations and the sensitivity of both pollutants to emissions from the traffic and industrial sectors. The regional CMAQ model has been widely used in assessing 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. In terms of the regional model 181 configuration, the detailed setting is given in the aforementioned publications, and only key 182 points have been listed below. The Sparse Matrix Operations Kernels Emissions (SMOKE) 183 model has been used to process the localized bottom-up emission inventory, including 184 industrial sources, mobile sources, power plants, residential sources, and marine sources for 185 the GBA. The marine emissions have been split into Ocean Going Vessels (OGV), Local 186 Vessels (LV), and River Vessels (RV) and were calculated using Automatic Identification 187 System (AIS) data. The emission inventory for outside GBA has been adapted from the 188 Multi-resolution Emission (MEIC) data (Tong et al., 2020). As shown in Figure S1, four 189 190 nested domains with resolutions of 27 km (D1), 9 km (D2), 3 km (D3), and 1 km (D4) were utilized for the regional CMAQ model. The boundary conditions of the outermost D1 domain 191 were obtained from a global chemical transport model (GEOS-chem) (Lam and Fu, 2010); 192 boundary conditions for the nested domains D2-D4 were obtained from the respective mother 193 domains. Outputs from the SMOKE and CMAQ models were used to drive the ADMS-194 Urban model. 195

ADMS-Urban is a street-scale resolution, quasi-Gaussian plume dispersion model 196 from the Atmospheric Dispersion Modeling System (ADMS) family, which has already been 197 widely applied across the world to assess the environmental impact, control policies, and 198 pollution concentration forecasts (Biggart et al., 2020; Carruthers et al., 1994; Hood et al., 199 2018; Lao and Teixidó, 2011). The model simulates the dispersion of pollutant emissions in 200 urban areas by: representing sources at high spatial resolution (primarily traffic and industry); 201 202 modeling the influence of urban morphology on dispersion processes (street canyons, building density, tunnels, road elevation); and applying simplified, near-field chemical 203 schemes. Sharp concentration gradients resulting from emissions released from sources such 204 205 as traffic can be resolved in the model calculations and captured for output using the irregularly-spaced receptor grid generated by the model. Spatially varying meteorological 206 parameters from the Weather Research and Forecast (WRF) model, such as wind and surface 207 208 sensible heat flux, have been used as input into the ADMS-Urban model to drive pollutant dispersion (Hood et al., 2018). 'Background' pollutant concentrations, representing long-209 range pollutant transport, have been derived from the hourly simulation data of the CMAQ 210 model. Due to a lack of representative source parameters (stack heights, efflux parameters), 211 industrial and power plant sources have been coarsely represented in the regional model 212 using appropriate factors to disaggregate emissions vertically, while an explicit road network 213 was applied to distribute the ground-level traffic emissions within the ADMS-Urban model. 214 Different weighting factors were given to different types of roads, but the same factors were 215 assigned to different species. The road network data were obtained from the OpenStreetMap 216 source (http://openstreetmap.org/), with some minor roads removed to reduce the 217 computational expense. A more detailed description of the coupling methodology between a 218 regional model and ADMS-Urban can be found in our previous publication (Hood et al., 219 2018). 220

## 221 2.2 Sensitivity scenario design

The control measures affecting emissions from traffic and industrial sectors are 222 applied in the regional CMAQ model over the Pearl River Delta Economic Zone (PRD EZ), 223 while explicit traffic emissions scenarios are applied to the road traffic network modelled in 224 225 ADMS-Urban. Four potential sensitivity scenarios are designed. As shown in Table 1, the Base case is a business as usual (BAU) scenario for both the regional CMAQ model and the 226 local ADMS-Urban model. Evaluation of the system is performed for a historical period 227 where measurement data are readily available. Meteorological conditions influence the 228 229 likelihood of O<sub>3</sub> episodes. Since higher concentrations are recorded in the spring and autumn of 2019, O<sub>3</sub> episodes in April and May 2019 are chosen for the control measures scenario

study. In table 1, three control scenarios are listed. Since  $NO_x$  and VOC are important precursors for  $O_3$  formation, the non-linear relationship between the precursors and  $O_3$  is of

precursors for  $O_3$  formation, the non-linear relationship between the precursors and  $O_3$  is of great importance. Due to the short lifetime of  $NO_x$ , which is emitted mainly from the traffic

sector, the Half-traffic case considers a 50% reduction in traffic emissions for all the standard

pollutants in the coupled modeling system. Since the majority of anthropogenic VOC

emissions are from the industrial sector, the Half-industry case considers the 50% reduction

in industrial VOC emissions only in the regional model but with BAU in the local model. The

Both control case integrates the control measures in both the Half-traffic and Half-industry

scenarios.

Scenarios	I. Base case	II. Half Traffic case	III. Half Industry	IV. Both Control case
			VOC case	
Scenario description	Business As Usual (BAU)	50% reduction in traffic emissions	50% reduction in industrial VOC emissions	Scenarios II & III
Regional 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 model emissions	I BAU	50% reduction in emissions from explicitly defined road traffic sources	BAU	50% reduction in emissions from explicitly defined road traffic sources

Table 1. Scenario design for the coupling system

## 241 2.3 Scenario emissions comparison

It is important to put the '50% reduction' control measures in the context of the total 242 emissions. A summary of total annual anthropogenic NO<sub>x</sub>, VOC, and PM<sub>2.5</sub> emissions for the 243 regional model domain covering central GBA is presented in Figure S2. Note that both the 244 'point' and 'area' emissions categories are considered to represent primarily industrial 245 activities, with VOC emissions affected by the 'half industry' control. Relative to total 246 emissions, the maximum reduction for  $NO_x$  is approximately 17% and for  $PM_{2.5}$  is only 5%. 247 For VOCs, which are impacted by both control measures, emissions are reduced by a much 248 larger amount, 47%, However, it is important to note that total VOC emissions also have a 249 large contribution from biogenic sources (around 50%), so in real terms, the maximum VOC 250 reduction due to control measures is closer to 25%. In terms of traffic emissions reductions, 251 252 the scenario modelled corresponds to a reduction in vehicle numbers and/or distances driven, rather than improvements in vehicle technologies. Whilst technological improvements 253 (including the introduction of electric vehicles) can reduce tailpipe emissions to zero, non-254 exhaust particulate emissions such as brake and tyre wear are a direct result of vehicle 255 activity. Consequently, non-exhaust vehicle emissions are not mitigated by improvements to 256 engine technology, although there may be associated technological improvements in relation 257 to non-exhaust emissions, such as regenerative braking. 258

Some example emissions plots for the 1 km domain are shown in Figure S3 to Figure 259 S8. All emissions presented are given as 'daily column' values, i.e., the values correspond to 260 daily average emissions summed over all vertical levels included in the modeled 3D 261 emissions grids. In the regional model, VOCs are a complex mixture of different components. 262 For the purpose of the emissions plots shown here, paraffin (PAR) emissions have been used 263 as a species representative of total VOCs. Figure S3 compares the total PAR emissions for 264 the BAU case and the three scenarios. Visually, the reduction in PAR emissions on the roads 265 can be seen by comparing Figures S3a and S3b, i.e., the signature of the road sources is 266

- reduced; comparing Figures S3a and S3c highlights the reduction in industrial source
- emissions; and Figure S3d shows the result of both reductions. Figure S4 presents spatial
- 269 plots of differences which emphasize the change in emissions. Figure S4a clearly indicates 270 reductions in the road network in the difference between the base case and the reduced traffic
- scenario. At the same time, Figure S4b shows large reductions at locations where there is
- intensive industrial activity (Shenzhen, Dongguan, and Guangzhou). Similar patterns of  $NO_x$
- and  $PM_{2.5}$  could be found in Figure S5 and S7, respectively. The reduction in VOC has no
- effects on NO<sub>x</sub> and PM<sub>2.5</sub> emissions, as shown in Figures S6b and S8b.

## 275 **3 Results**

The regional model has been configured and run for April – May 2019. The coupled system results have been generated for the same period for the urban sub-domains developed as a demonstration area for this study, i.e., a 6 km x 6 km area in central Guangzhou. Periodaverage concentrations have been calculated. Both urban and rural monitors have been selected to illustrate the pollution variation.

281 3.1 Regional model period-average air quality maps

Figures 1a and 1b show period-average NO<sub>2</sub> concentrations for major PRD EZ for the base case and the half traffic case respectively, at 1 km grid resolution. As expected, there is a clear reduction in Guangzhou and Shenzhen NO<sub>2</sub> concentrations due to the half traffic emissions scenario. NO<sub>2</sub> concentrations are significantly higher in the HKSAR (to the south of Shenzhen), in industrial areas towards Guangzhou, and also along shipping lanes than in the urban area. Figure 1c quantifies the reduction in NO<sub>2</sub> concentrations for this period; concentrations are reduced by up to 5 ppb in central Guangzhou and Shenzhen.



113°E 113.2°E113.4°E113.6°E113.8°E 114°E 114.2°E114.4°E114.6°E

Figure 1. Simulated spatial maps of period-average NO<sub>2</sub> concentrations for (a) Base case, (b) Half
 Traffic case, and (c) Difference plot: Both controls – Base case (ppb)

- Figures 2a and 2b compare period-average O<sub>3</sub> concentrations for major PRD EZ for 291
- the base case and both controls together respectively. Due to the reduction in NO<sub>x</sub> 292
- concentrations leading to a reduction in O<sub>3</sub> titration by NO<sub>x</sub>, O<sub>3</sub> concentrations increase in 293
- Shenzhen as a result of the emissions controls. Conversely, in the rural areas to the northeast 294
- of the domain, downwind of the highly polluting areas, O3 concentrations decrease due to the 295 controls because, overall, less oxidant (the sum of  $NO_2$  and  $O_3$ ) is present in the atmosphere.
- 296



- 297 **Figure 2.** Simulated spatial maps of period-averaged  $O_3$  for (a) Base case and (b) Both controls (ppb); 298 white star indicates the location for further quantification of regional model concentrations (Figure 3)
  - **(b)** (a) 23.1°N 23.1°N 23°] 23°N 22.9 22.9°N 22.8°N 22.8°N 22.7°N 22.7°N 22.6°N 22.6°N 22.5°N 22.5°N 22.4°N-22.4 22.3°N-22.3°N-22.2°N 22.2°N-22.1°N 22.1°N ~ 0:3 0:0 113°E 113.2°E113.4°E113.6°E113.8°E 114°E 114.2°E114.4°E114.6°E 113°E 113.2°E113.4°E113.6°E113.8°E 114°E 114.2°E114.4°E114.6°E (c) 23.1°N 2.3° 22.9 22.8°N 22.7°N 22.6°N 22.5°N 22.4°N 22.3°N 22.2°N 22.1°N

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Figure 3. Simulated spatial maps of period-averaged O<sub>3</sub> concentrations - Difference plots for (a) Half 300 Traffic – Base case; (b) Half Industry VOC – Base case, and (c) Both controls – Base case (ppb). 301

Figures 3a to 3c quantify the changes in O<sub>3</sub> concentrations for this period, for each of 302 the controls and both controls together. Figure 3a relates to the half traffic case; 303

- 304 concentrations increase in the urban areas and decrease downwind in the rural areas due to
- reduced NO<sub>x</sub> titration and lower oxidant emissions, as discussed above. Figure 3b relates to
- the reduced VOC case; here,  $O_3$  concentrations are reduced throughout the domain because
- 307 lower concentrations of VOCs correspond to lower levels of reactive species in the 308 atmosphere, resulting in less oxidant (in this case, O<sub>3</sub>) being generated. When both controls
- antiosphere, resulting in less oxidant (in this case,  $O_3$ ) being generated. When both controls are considered together, increased  $O_3$  concentrations are still seen in the urban areas. Still, the
- magnitude is less than in Figure 3a due to the influence of reduced VOC concentrations. In
- the majority of locations away from the urban areas, O<sub>3</sub> concentrations are reduced, albeit by
- a relatively small amount (few ppb) when this average  $O_3$  metric is considered.
- Figures 4a and 4b show period-average  $PM_{2.5}$  concentrations for the PRD EZ for the base case and the half traffic case respectively, at 1 km grid resolution. Although inspection of Figure 4c would suggest that emissions reductions are minimal, in fact, considerable  $PM_{2.5}$ concentration differences result from the traffic restrictions imposed in the urban areas. Figure 4c quantifies the reduction in  $PM_{2.5}$  concentrations for this period; concentrations are reduced by up to 3 µg/m<sup>3</sup> in central Shenzhen.



- Figure 4. Simulated spatial maps of period-average  $PM_{2.5}$  concentrations for (a) Base case, (b) Half Traffic case, and (c) Difference plot: Both controls – Base case ( $\mu g/m^3$ ).
- 321 3.2 Coupled system period-average air quality maps.

Concentrations within the Guangzhou coupled system domains are calculated at high
 temporal and spatial resolution. Therefore, rather than presenting results as average
 concentrations over the two months as for the regional model domain, hourly concentrations
 are presented. For some pollutants, these detailed, hourly air quality maps relate to the
 metrics included in the Chinese air quality standards (specifically, the 200 µg/m<sup>3</sup> and

- 327 160  $\mu$ g/m<sup>3</sup> standards for NO<sub>2</sub> and O<sub>3</sub> respectively, which are applicable in urban areas).
- 328 Figure 5 shows a case for the Guangzhou domain where NO<sub>2</sub> concentrations exceed the

- 329 hourly limit value in the middle of the day during May; concentrations are higher during the
- morning rush hour. However, Figures 5b and 5d show that the area of exceedance of the limit
- value of 200  $\mu$ g/m<sup>3</sup> is significantly reduced when the traffic emissions are halved; local NO<sub>2</sub>
- concentrations are not seen to change with variations in the VOC emissions (Figures 5c).



- Figure 5. Simulated high-resolution spatial maps of NO<sub>2</sub> (Guangzhou domain) at 14:00, 10<sup>th</sup> May
   2019 for a) Base case, b) Half Traffic case, c) Half Industry VOC case, d) Both Control case (µg/m<sup>3</sup>).
- Figure 6 presents O<sub>3</sub> concentrations in the Guangzhou domain during the same 335 pollution episode (14:00 on 10<sup>th</sup> May 2019). As indicated earlier in relation to the regional 336 model results, reducing traffic emissions increases the spatial extent of the O<sub>3</sub> exceedances in 337 urban areas due to reduced NOx titration of O3 (compare Figures 6a and 6b). Conversely, 338 reducing VOCs leads to a reduction in the area of O<sub>3</sub> exceedance within this local domain 339 (compare Figures 6a and 6c). When both controls are applied in the local area (comparing 340 Figures 6a and 6d), the net effect is a slight increase in near-road O<sub>3</sub> concentrations, but a 341 decrease in concentrations elsewhere. This is an interesting result that again demonstrates the 342
- importance of accounting for both regional and local dispersion and chemistry.



Figure 6. Simulated high-resolution spatial maps of  $O_3$  (Guangzhou domain) at 14:00, 10<sup>th</sup> May 2019 for (a) Base case, (b) Half-Traffic case, (c) Half Industry VOC case, (d) Both-Control case ( $\mu$ g/m<sup>3</sup>)

Modelled concentrations for a different time are presented for PM<sub>2.5</sub>, as the 346 atmospheric conditions associated with PM<sub>2.5</sub> pollution episodes differ from those associated 347 with O<sub>3</sub> and NO<sub>2</sub> episodes. PM<sub>2.5</sub> concentrations for 11:00 on the 23<sup>rd</sup> April 2019 are shown 348 for all four scenarios in Figure 7. Although there is a very small relative reduction in PM<sub>2.5</sub> 349 emissions (Figure S2)), the impact in urban areas is significant during this episode (compare 350 Figures 7a and 7b), as this emissions reduction relates to near-ground traffic sources. The 351 change in VOC emissions has a negligible effect on PM<sub>2.5</sub> concentrations at this scale, so 352 353 there is no difference between the base case and the half VOC emissions case (Figures 7a and 354 7c). 355

356



Figure 7. Simulated high-resolution spatial maps of  $PM_{2.5}$  (Guangzhou domain) at 11:00, 23<sup>rd</sup> April 2019 for (a) Base case, (b) Half Traffic case, (c) Half Industry VOC case, (d) Both-Control case ( $\mu$ g/m<sup>3</sup>)

360 3.3 Modelled concentrations at selected urban and rural locations

Pointwise concentrations are discussed in this section. Where possible, the locations 361 considered relate to air quality measurement sites within the domain. Figure S9 shows the 362 363 location of the three reference monitors located within the Guangzhou coupled system urban model domain; in addition to other pollutants, NO<sub>2</sub>, O<sub>3</sub>, and PM<sub>2.5</sub> concentrations are 364 365 recorded at these sites. Figure 8 compares modelled concentrations to the measurements recorded at these three locations, for the base case and three coupled model scenarios in 366 addition to the base case regional model. Here box plots of the short-term pollutant metrics 367 are shown, i.e., daily maximum of the hourly NO<sub>2</sub>, daily maximum of the 8-hour rolling O<sub>3</sub>, 368 and daily mean PM<sub>2.5</sub>. As this is the first time the regional model concentrations have been 369 presented alongside the coupled model concentrations, it is worth noting the difference in the 370

- 371 concentrations for the two modelling approaches. Specifically, for NO<sub>2</sub> and PM<sub>2.5</sub> at the
- majority of sites, the coupled system predicts higher concentrations than the relatively coarse resolution regional model, and for  $O_3$  the coupled system predicts lower concentrations.
- These differences are expected at the monitoring locations identified, which are strongly
- influenced by local road traffic source increments. The respective concentration changes of
- the various sensitivity scenarios in the selected monitoring stations are similar to the trend
- 377 illustrated in the comparisons of the spatial concentration map. Figure 8a shows that the NO<sub>2</sub>
- 378 concentrations are dominantly contributed from the traffic sector. The NOx titration effects
- on the  $O_3$  concentration in Figure 8b drive up the  $O_3$  concentration, while cutting the industry
- 380 VOC emission sources is more effective for the O<sub>3</sub> control, revealing a VOC-limited regime
- in this region.



**Figure 8.** Box plots comparing measured concentrations (pale blue) and regional model

- 383 concentrations (red) to the four high-resolution coupled system model scenarios: Base case (orange),
- Half Traffic case (light green), Half Industry VOC case (darker green), and both controls (bright blue)
- for (a) daily maximum hourly NO<sub>2</sub>, (b) daily maximum 8-hour rolling O<sub>3</sub>, and (c) daily average PM<sub>2.5</sub>. Unit is in  $\mu$ g/m<sup>3</sup>.

In terms of the differences in modelled concentrations for the three scenarios: Over all sites, the maximum decrease in the median NO<sub>2</sub> hourly metric due to emission controls is over 11  $\mu$ g/m<sup>3</sup> at the roadside site CN\_1352A, which corresponds to the implementation of the traffic controls. In terms of O<sub>3</sub>, the maximum increase in the median value is over 10  $\mu$ g/m<sup>3</sup> for the reduced traffic scenario. However, this increase is reduced to under 7  $\mu$ g/m<sup>3</sup> when both controls are applied together. Decreases in median PM<sub>2.5</sub> are less than 2  $\mu$ g/m<sup>3</sup> for the low traffic scenario.



Figure 9. Box plots comparing the regional model concentrations (red) to the four high-resolution
 coupled system model scenarios at a rural site: 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<sub>2</sub>, (b) daily maximum 8-hour rolling O<sub>3</sub>, and (c) daily average PM<sub>2.5</sub>. Unit is in µg/m<sup>3</sup>.

It is of interest to quantify the decrease in  $O_3$  concentrations shown in Figures 2 and 3, 398 to the northeast of the model domain. Unfortunately, measurements were unavailable at this 399 rural location. Furthermore, the coupled system has only been configured for the example 400 urban sub-domains in Guangzhou. Consequently, the only comparison to be made at this 401 location is between concentrations calculated by the regional model. Concentration data for 402 403 the location indicated by the white star in Figure 2 are presented in Figure 9; the metrics calculated for NO<sub>2</sub>, O<sub>3</sub>, and PM<sub>2.5</sub> are the same as those presented in Figure 8. At this rural 404 location, the different emissions mitigation options only significantly alter the NO<sub>2</sub> 405 concentrations, out of the three pollutants modelled. This is unsurprising because Figure S2 406 shows that traffic emissions make up a large proportion of NO<sub>x</sub> emissions over the whole 407 domain, so changes to NO<sub>x</sub> emissions are likely to impact NO<sub>2</sub> concentrations in the rural as 408 409 well as urban areas. Conversely, traffic makes up a relatively small proportion of primary PM<sub>2.5</sub> emissions; ambient PM<sub>2.5</sub> levels in rural areas are more influenced by industrial point 410

and area source emissions in addition to the formation of secondary organic and inorganicparticulates (Wu and Xie, 2018).

In terms of O<sub>3</sub>, there is a relatively minimal reduction in terms of the median of 413 maximum 8-hourly average concentrations resulting from the reduced VOC emissions 414 scenario. This is perhaps surprising when looking back to Figure 6, as, for the corresponding 415 scenarios, decreases of tens of  $\mu g/m^3$  are shown throughout the urban model domain. To 416 understand this, it is helpful to look at a time series of modelled O<sub>3</sub> concentrations during an 417 episode (Figure 10a). Here we see that while there is very little difference in concentrations 418 419 for the majority of the time, the mitigation scenarios have a substantial impact in this rural location when  $O_3$  levels are at their highest: up to 15  $\mu$ g/m<sup>3</sup> for the 8-hour rolling average 420 metric and hourly concentration differences are greater, up to 25 µg/m<sup>3</sup> during the same 421 422 period,







424 average  $O_3$  during an episode in April 2019 at a location to the north-east of the model domain: Base 425 case (red), Half Traffic case (light green), Half Industry VOC case (darker green), and both controls 426 (bright blue). Unit is in  $\mu g/m^3$ .

## 427 4 Discussion

428 A regional-to-local scale coupled air quality modeling system consisting of the 429 regional CMAQ model (Zhang et al., 2020) and the street-scale ADMS-Urban model

430 (Biggart et al., 2020) has been configured to explore mitigation of NO<sub>2</sub>, O<sub>3</sub>, and PM<sub>2.5</sub> by

431 controlling  $NO_x$  and VOCs in traffic and industrial sectors for the GBA, at varied resolutions.

432 Separately, the regional and street-scale models account for pollutant dispersion and

433 atmospheric chemistry at different temporal and spatial scales, and to varying degrees of

434 complexity; all dominant processes are accounted for when the models are linked via the

435 coupled system. An operational forecasting system has been developed, with street-scale
 436 resolution output at an example urban location, a 6 km x 6 km area in central Guangzhou.

Figures showing the spatial variation of emissions changes corresponding to these 437 mitigation scenarios for NO<sub>x</sub>, VOCs, and PM<sub>2.5</sub> have been presented. Graphs showing the 438 impact of these mitigations on total emissions indicate that the combination of both controls 439 leads to 17, 5, and 47% emissions reductions in terms of anthropogenic NO<sub>x</sub>, PM<sub>2.5</sub>, and 440 VOC, respectively; of note here is that anthropogenic emissions account for a considerable 441 part of total VOC emissions, with the remainder being from biogenic sources, which will be 442 larger under the climate change impact (Li et al., 2018). A selection of period-average 443 concentration plots showing the impact of the different mitigation scenarios has been 444 presented. The regional modeling results illustrate that the half-traffic scenario leads to 445 reductions of NO<sub>2</sub> and PM<sub>2.5</sub>. Still, increases in O<sub>3</sub> concentrations in urban areas due to a 446 titration effect, revealing a VOC-limited O<sub>3</sub> formation regime in GBA urban areas, which is 447 consistent with previous studies (Wu et al., 2021; Zhang et al., 2021a). Conversely, this 448 449 reduced traffic scenario leads to O<sub>3</sub> decreases in rural areas downwind. He et al. (2019) identified a VOC-limited O<sub>3</sub> formation regime near a rural monitoring site in GBA using a 450 box model with the master chemistry mechanism in the Autumn of 2014. This highlights that 451 the complex O<sub>3</sub> formation regime varies at different times of the year and suggests that the O<sub>3</sub> 452 formation regime may transfer from VOC-limited to mixing regimes in the GBA (Wang et 453 al., 2019a). More sensitivity analysis work needs to be done for the O<sub>3</sub> formation mechanism 454 of the downwind rural areas, occurring substantial VOC emissions. The reduced industrial 455 VOC emissions scenario leads to reduced O<sub>3</sub> concentrations throughout the mitigation 456 domain but has a negligible impact on NO<sub>2</sub> and PM<sub>2.5</sub> concentrations. This sheds light on the 457 importance of stringent VOC control measures applied to the industrial sector in the near 458 459 future.

The impacts of the different mitigation options at the street scale have also been 460 presented. For these examples, pollution maps representative of pollution levels for single 461 hours are shown for the Guangzhou domain. These plots show the large variation in pollutant 462 concentrations over the areas modelled: a consequence of both the regional model variations 463 (at 1 km resolution) in addition to the local model component resolving the sharp 464 concentration gradients in the vicinity of road sources. Similar to the regional modeling 465 results but with more details near the roadside, the half-traffic scenario leads to large 466 reductions of NO<sub>2</sub> and PM<sub>2.5</sub> in the vicinity of road sources; and when both controls are 467 applied together, there is a slight increase in near-road O<sub>3</sub> concentrations, but a decrease in O<sub>3</sub> 468 elsewhere. This highlights that synergistic controls of NO<sub>x</sub> and VOCs would be a promising 469 way to alleviate the PM<sub>2.5</sub> and O<sub>3</sub> simultaneously (Wu et al., 2021). 470

Specific locations have been selected for further analysis. Relative to the baseline, the 471 urban air quality receptors show that the maximum decrease in the median of hourly NO<sub>2</sub> 472 concentrations is over  $11 \,\mu\text{g/m}^3$ , and the maximum increase in the median of maximum 8-473 hour rolling  $O_3$  concentrations is over 10  $\mu$ g/m<sup>3</sup> for the reduced traffic scenario. The  $O_3$ 474 increase is reduced to under 7  $\mu$ g/m<sup>3</sup> when both controls are considered, indicating the 475 effectiveness of the synergistic control measures of NO<sub>x</sub> and VOC in both sectors. The 476 decreases in the daily average PM<sub>2.5</sub> metrics are less than  $2 \mu g/m^3$  for the reduced traffic 477 scenario, confirming that the PM<sub>2.5</sub> primary emissions from traffic sources have a relatively 478 low contribution to overall concentrations (Figure S2). The magnitude of these relative 479 changes should be taken in the context of the metric considered: hourly values (i.e., NO<sub>2</sub>) 480 demonstrate the greatest variations because the maximum differences at peak traffic times are 481 quantified; conversely, for daily average values (i.e., for PM2.5), the impact of peak values is 482 smoothed out by the inclusion of hours where pollutant concentrations may be dominated by 483

regional rather than local air pollution. O<sub>3</sub> episodes are of particular current interest to
government officials and stakeholders. The modelling work has demonstrated that whilst O<sub>3</sub>
concentrations increase in the urban areas as a result of the mitigation options considered, O<sub>3</sub>
decreases downwind. Inspection of modelled pollutant concentrations at a rural location to
the northeast of the modelling domain during an O<sub>3</sub> episode shows that concentrations are

reduced by up to  $15 \ \mu g/m^3$  for the 8-hour metric and up to  $25 \ \mu g/m^3$  for the 1-hour metric, which emphasizes the value of more stringent VOC controls, applied to the industrial sector.

## 491 **5 Conclusion**

To address the challenges of controlling  $PM_{2.5}$  and  $O_3$  concentrations simultaneously 492 using an ultrahigh spatial resolution approach, this study presents the regional air quality 493 model CMAQ coupled to the street-scale model ADMS-Urban. The coupled system allows a 494 thorough assessment of the impact that NO<sub>x</sub> and VOC emissions from traffic and industry 495 have on ambient O<sub>3</sub> and PM<sub>2.5</sub>, drawing a holistic pollution mitigation picture at a range of 496 spatial resolutions, as well as highlighting the temporal relationship between emissions, 497 meteorological conditions and O<sub>3</sub> concentrations. The regional modeling results show the 498 499 half-traffic scenario leads to reductions of NO<sub>2</sub> and PM<sub>2.5</sub>, but increases in O<sub>3</sub> concentrations in urban areas (and decreases in rural areas downwind), revealing a VOC-limited O<sub>3</sub> 500 formation regime. The reduced industrial VOC emissions scenario leads to reduced O<sub>3</sub> 501 concentrations throughout the mitigation domain; it has a negligible impact on NO<sub>2</sub> and 502 PM<sub>2.5</sub> concentrations. This finding suggests more stringent VOC control measures in the 503 industrial sector will substantially alleviate the increasing O<sub>3</sub> pollution. 504

With coupling, the street-scale ADMS-Urban model resolves the sharp concentration 505 gradients in the vicinity of road sources, and the half-traffic scenario leads to large reductions 506 of NO<sub>2</sub> and PM<sub>2.5</sub> in those locations. When both controls are applied together, there is a slight 507 increase in near-road O<sub>3</sub> concentrations, but a decrease in O<sub>3</sub> elsewhere. Examples of urban 508 and rural monitoring sites in central Guangzhou are used to better interpret findings. Relative 509 to the base case, the maximum decrease in the median hourly NO<sub>2</sub> metric is over  $11 \, \mu g/m^3$ 510 for the reduced traffic scenario; the maximum increase in the median maximum 8-hour 511 rolling  $O_3$  metric is over 10  $\mu$ g/m<sup>3</sup> for the reduced traffic scenario, although this increase is 512 reduced to under 7  $\mu$ g/m<sup>3</sup> when both controls are considered; and decreases in the daily 513 514 average PM<sub>2.5</sub> metrics are less than 2  $\mu$ g/m<sup>3</sup> for the reduced traffic scenario.

Although the detailed mitigation pathways modeled here support the second phase of 515 the Air Pollution Prevention and Control Action Plan-the Three-Year Action Plan for Clean 516 Air—released by the State Council of China in 2018, further refinements are required as part 517 of future studies. Subsequent studies would benefit from: analysis using a more 518 comprehensive observational pollutant concentrations dataset; application of the model over 519 larger urban areas in the region; and application of the coupled street-scale air quality 520 modeling system to similar urban cities. In addition, a more advanced emission preparation 521 methodology (Lam et al., 2021) must be applied in order to minimize the uncertainties 522 associated with the emission inventory, and more elaborate emission sources could be 523 modelled explicitly in the ADMS-Urban model, e.g., industrial stacks (Hood et al., 2018). 524 Since the meteorological fields such as the wind are of great importance to the coupled model 525 simulations (Wang et al., 2019b), improving the representation of urban morphological data 526 in the model could improve the baseline model biases. Finally, assessing the reduction radios 527 of NO<sub>x</sub> and VOC in various areas of a city or different cities should be cautiously assessed 528 for efficient complex co-photochemical controls. 529

## 530 Acknowledgements

We sincerely thank the Hong Kong Environmental Protection Department (EPD) for providing the bottom-up emission inventory. We appreciate the Guangdong Environmental Monitoring Centre, Macao EPD, and HK EPD for offering the observational data to validate the model runs. All of the data used in this paper are cited and referred to in the reference list. The authors would like to acknowledge funding from the Newton Fund through Innovate UK as part of the project titled Air Pollution Monitoring and Very High Resolution Early Warning Platform for Guangdong (project number 104313), Science and Technology Planning Project of Guangdong Province (2018A050501004), and HSBC 150th Anniversary Charity Programme through the PRAISE-HK project. 

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