

1                                    **Supplementary Information of**  
2                                    **High-resolution simulation of link-level vehicle emissions and**  
3                                    **concentrations for air pollutants in a traffic-populated East Asian city**  
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1 **Supplementary Tables**

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3 **Table S1.** Estimated traffic activity by vehicle category in Macao during a typical weekday of 2010

Vehicle category	Traffic activity (veh km d <sup>-1</sup> )	Vehicle category	Traffic activity (veh km d <sup>-1</sup> )
LDPV	1.72×10 <sup>6</sup>	Taxi	6.01×10 <sup>5</sup>
MDPV	1.08×10 <sup>5</sup>	Motorcycle	1.23×10 <sup>6</sup>
HDPV	8.44×10 <sup>4</sup>	LDT	1.37×10 <sup>5</sup>
Public bus	1.42×10 <sup>5</sup>	HDT	2.12×10 <sup>4</sup>

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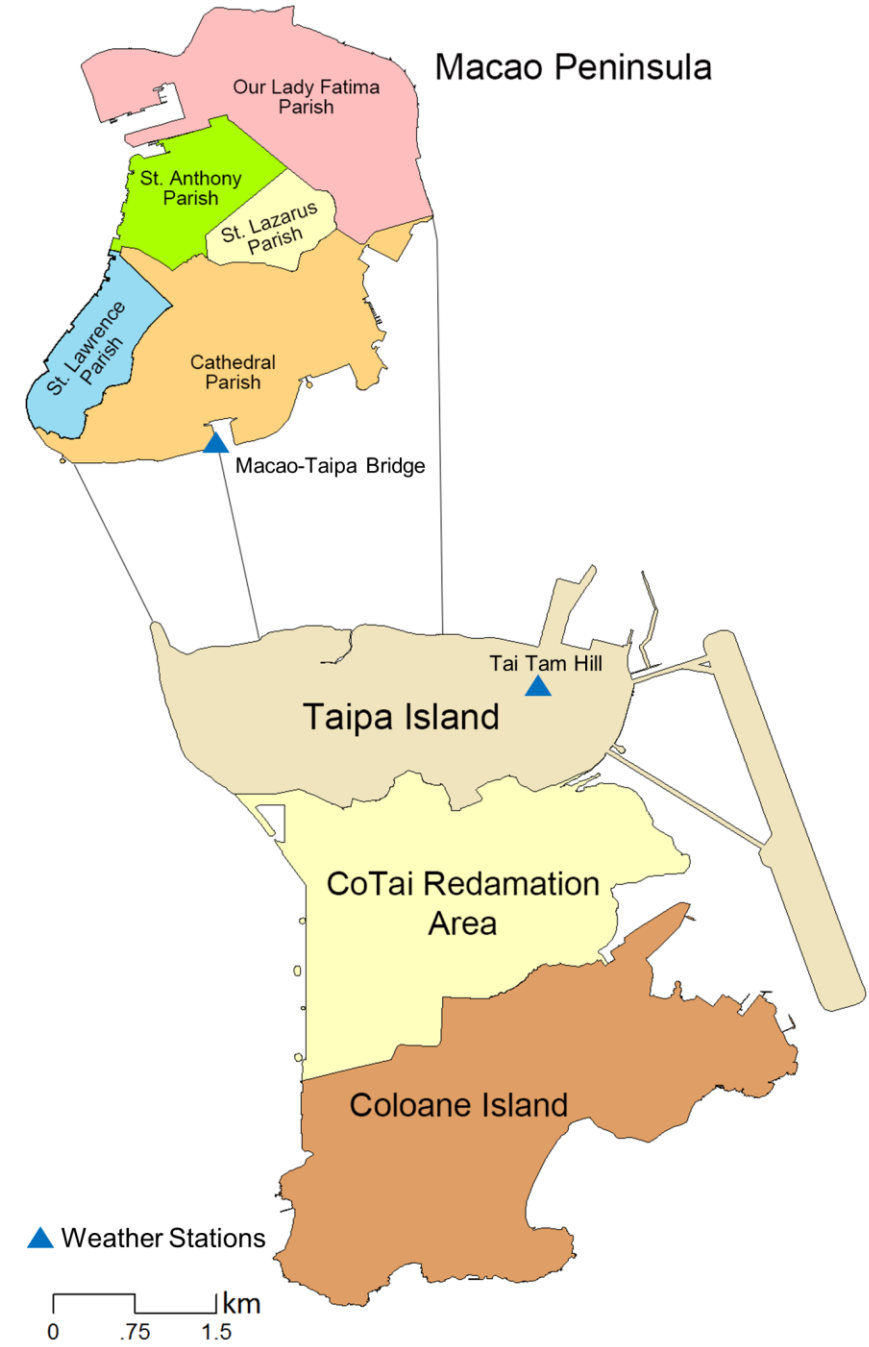
1 **Table S2.** Spatial allocation of vehicle emissions in Macao during a typical weekday of 2010

Region / Parish	Allocation of vehicle emissions				
	CO	THC	NO <sub>x</sub>	PM <sub>2.5</sub>	CO <sub>2</sub>
Macao Peninsula	76%	78%	58%	52%	59%
St. Lazarus Parish	12%	12%	9%	8%	9%
St. Lawrence Parish	8%	8%	6%	5%	6%
Our Lady Fatima Parish	23%	25%	17%	15%	18%
St. Anthony Parish	16%	17%	12%	10%	13%
Cathedral Parish	16%	16%	14%	13%	13%
Taipa	12%	10%	21%	23%	19%
CoTai Reclamation Area	5%	5%	9%	10%	8%
Coloane	2%	2%	2%	2%	3%
Others (three cross-sea bridges)	6%	5%	10%	13%	10%

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1 **Supplementary Figures**



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4 Fig. S1. Map of the Special Administrative Region of Macao, China  
5 Two weather stations are marked in the figure.  
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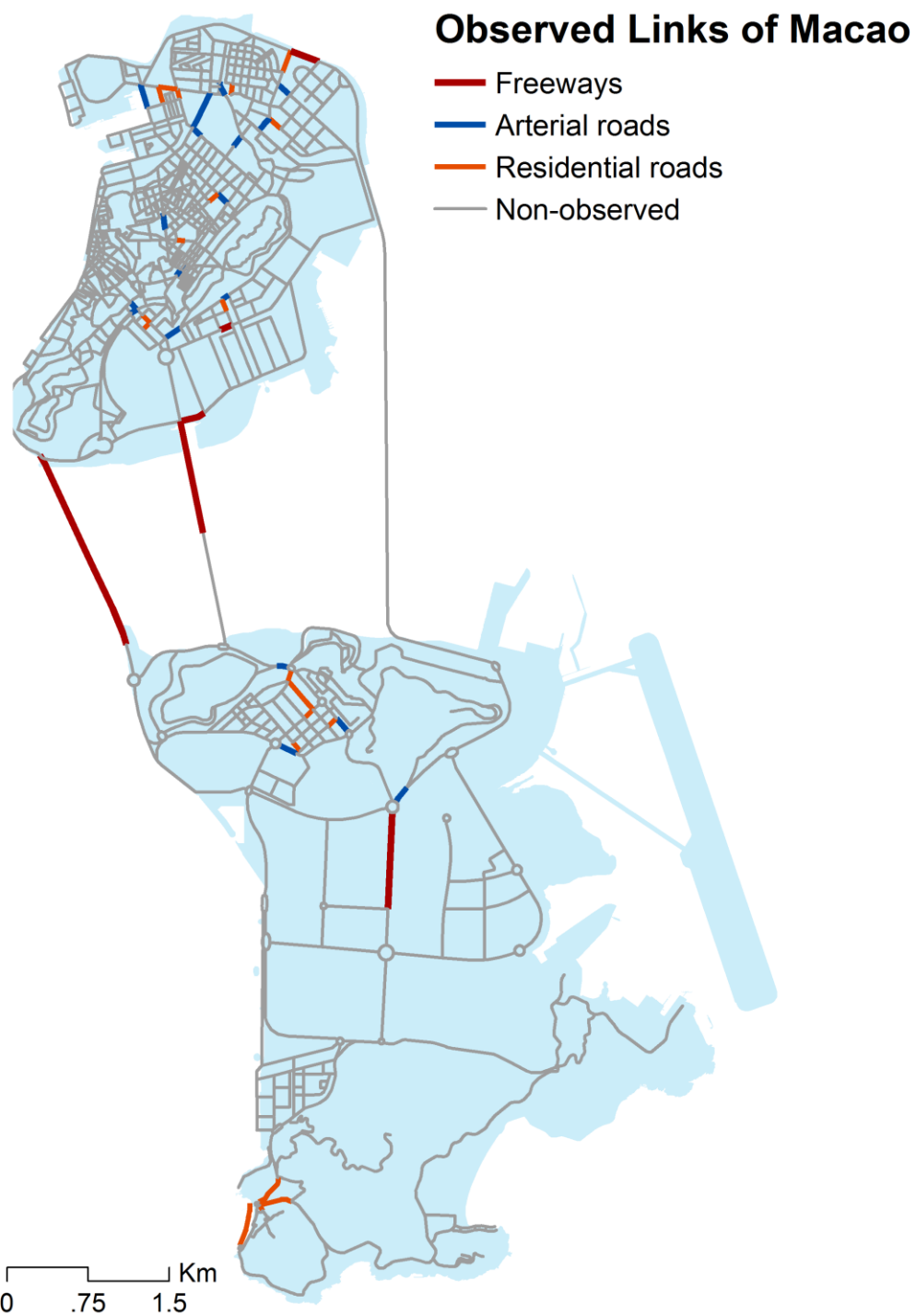


Fig. S2. Map of the road links with observed traffic volume data

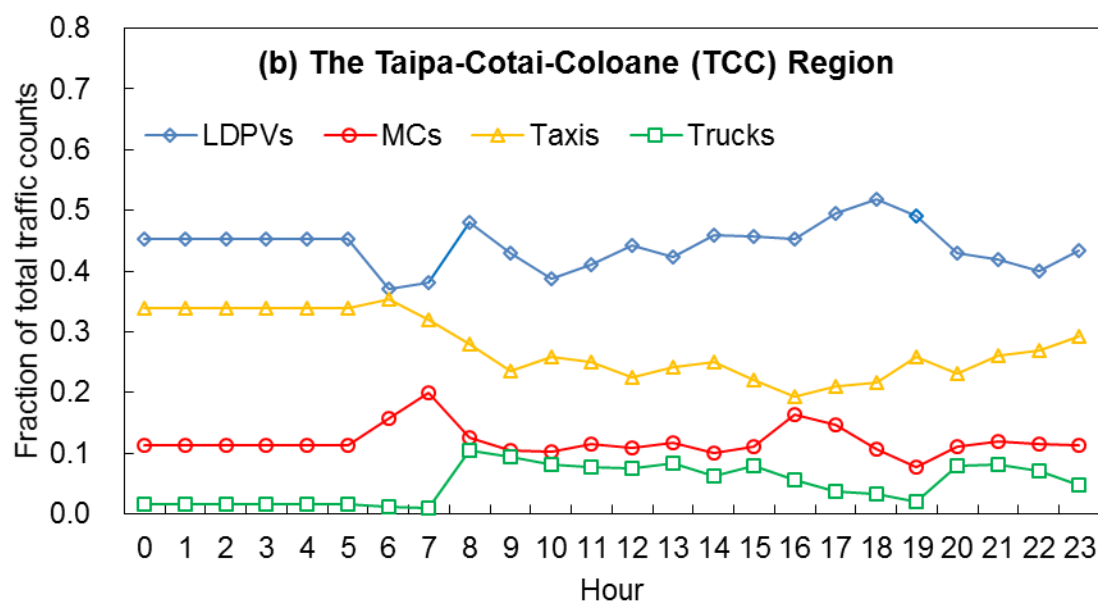
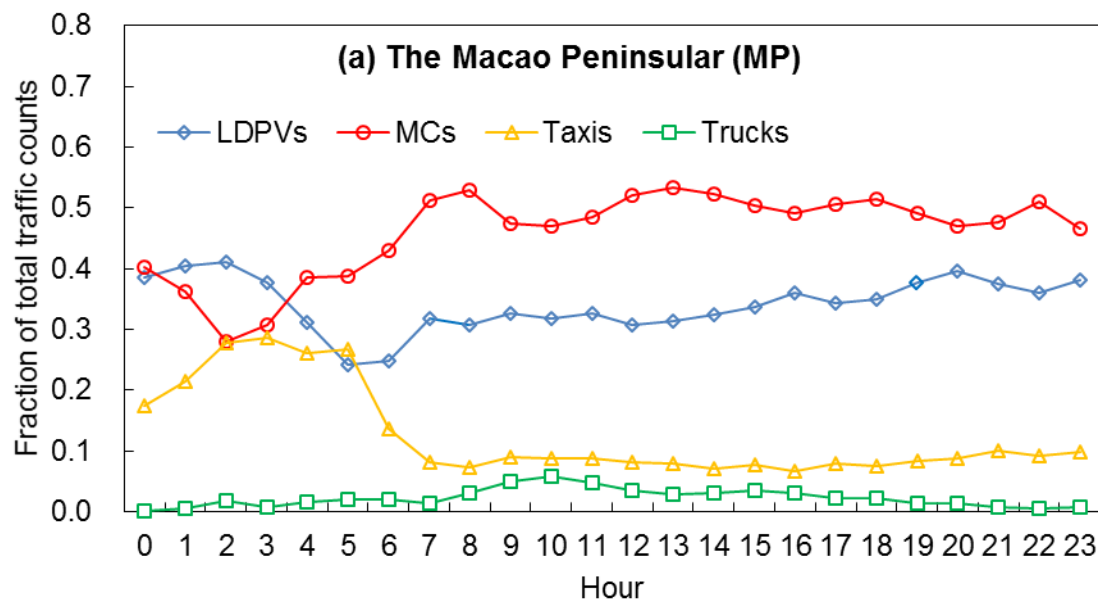


Fig. S3. Hourly allocations of traffic volume for LDPVs, motorcycles, taxis and trucks on the arterial roads in (a) the Macao Peninsula and (b) the Taipa-Cotai-Coloane Region.  
Note: Hourly truck volume data in this figure is the combined results for LDTs and HDTs.

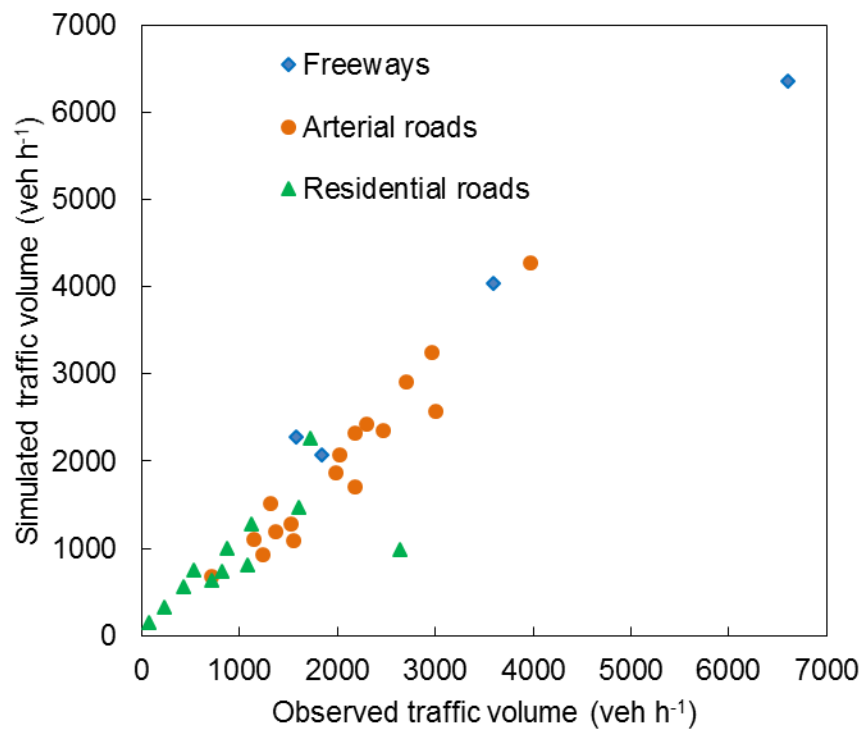


Fig. S4. Comparison of observed traffic volume and simulated results with the TransCAD model for 33 roads during 6 p.m. hour of typical weekdays

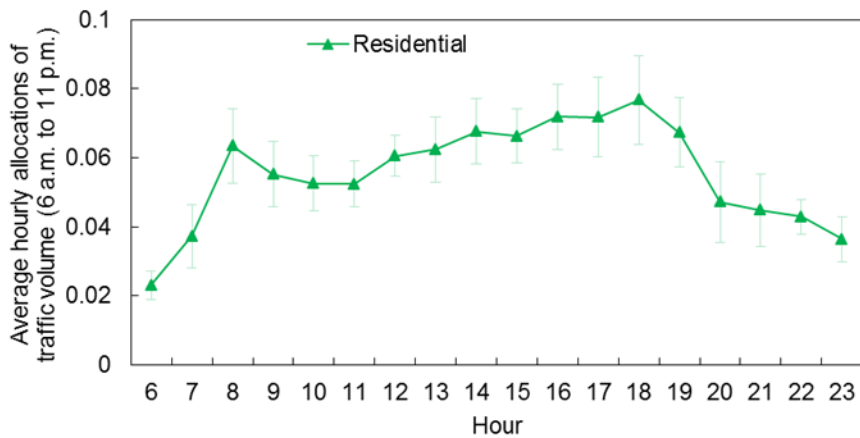
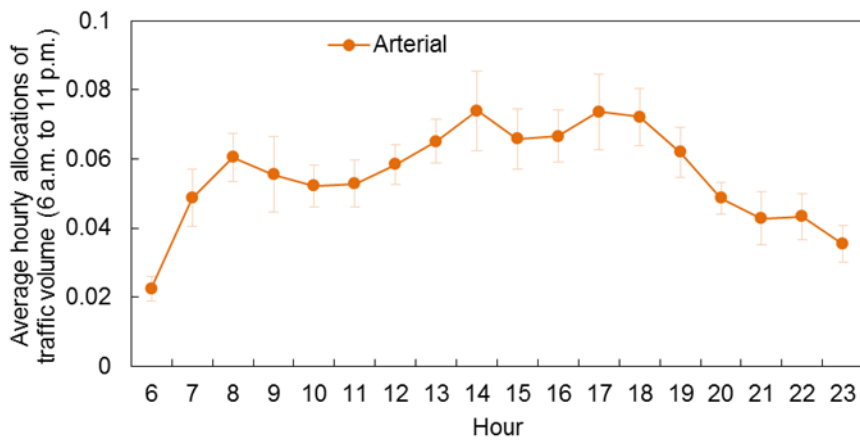
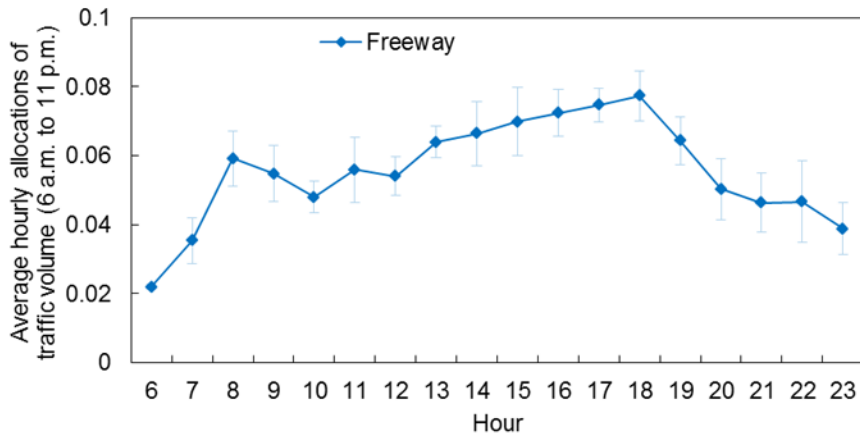


Fig. S5. Average allocations of hourly traffic volume in the total traffic volume from 6 a.m. to 11 p.m. Only roads with observed traffic volume data are included.



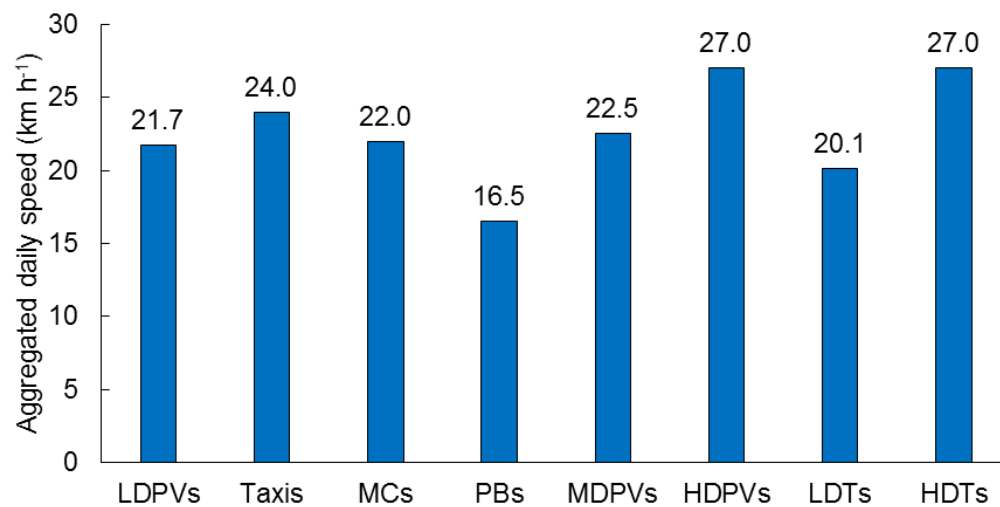
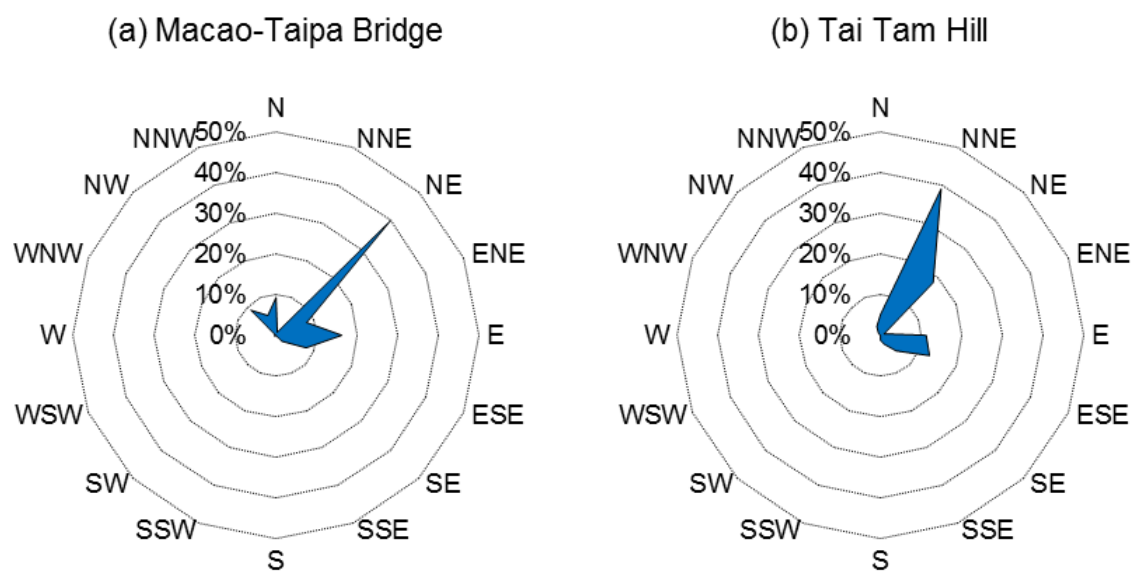


Fig. S6. Fleet-average speed of each vehicle category in Macao, during a typical weekday of 2010.

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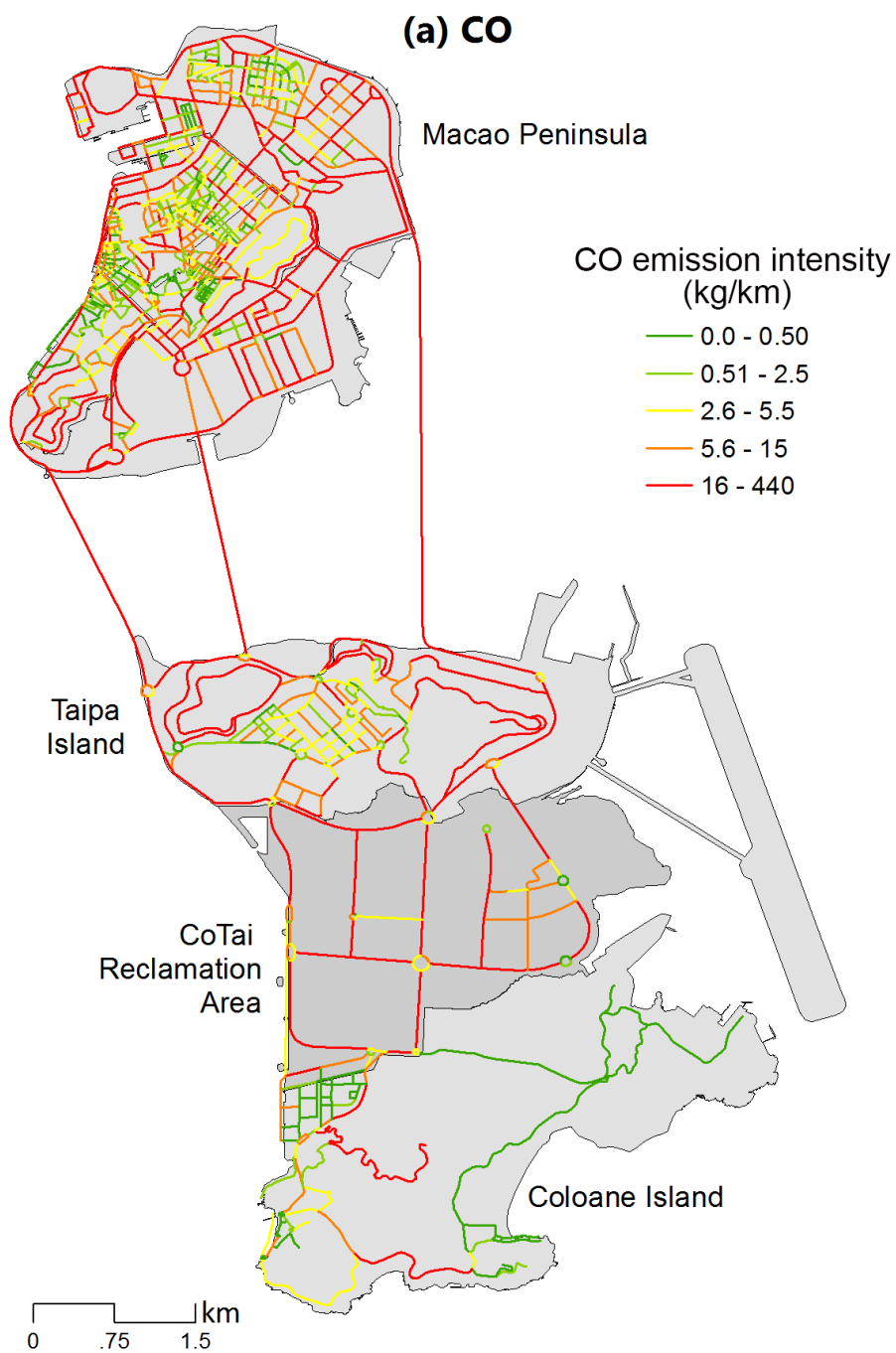
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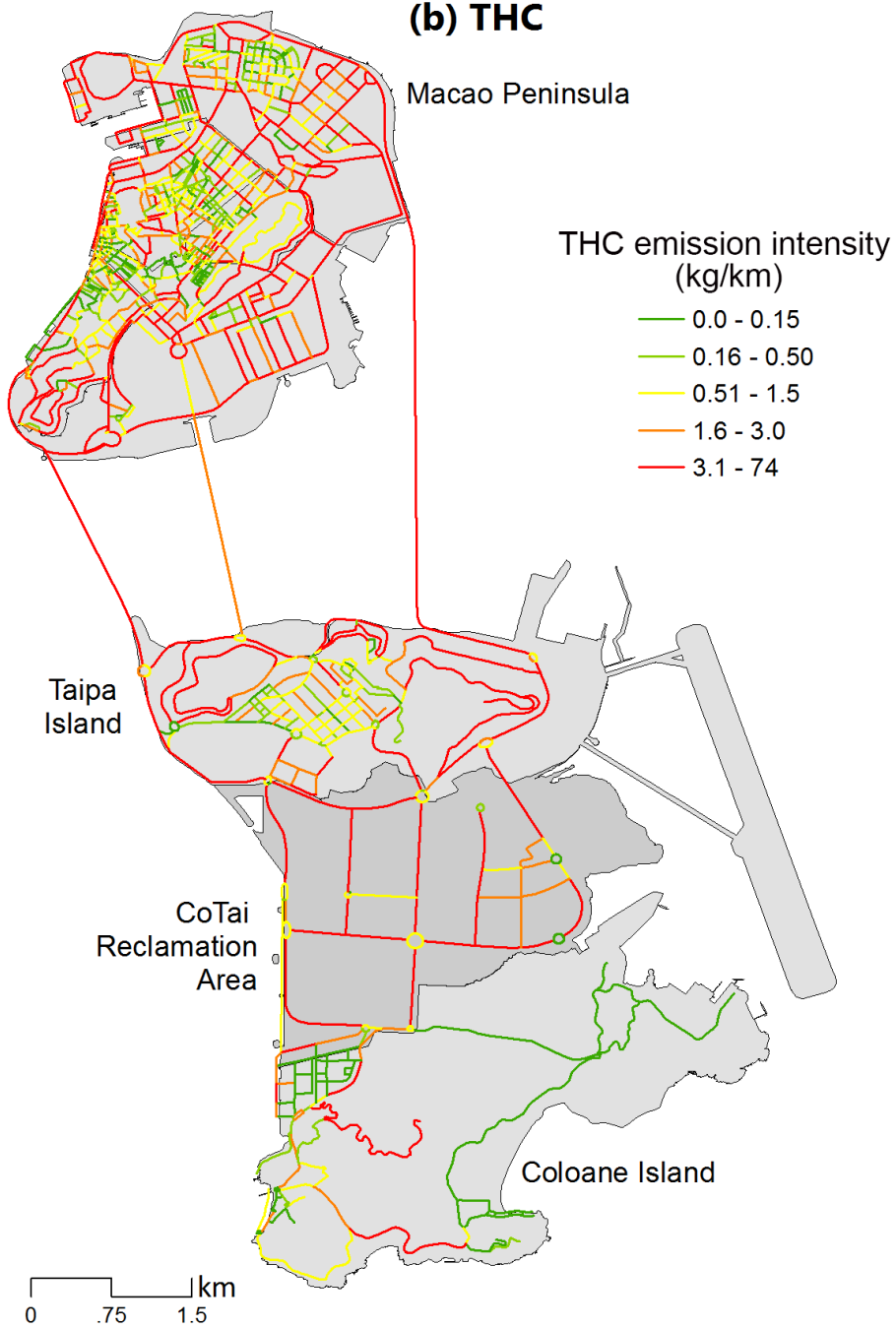
4 Fig. S7. Wind rose map of two weather stations in Macao during weekdays of November 2010.

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## (b) THC



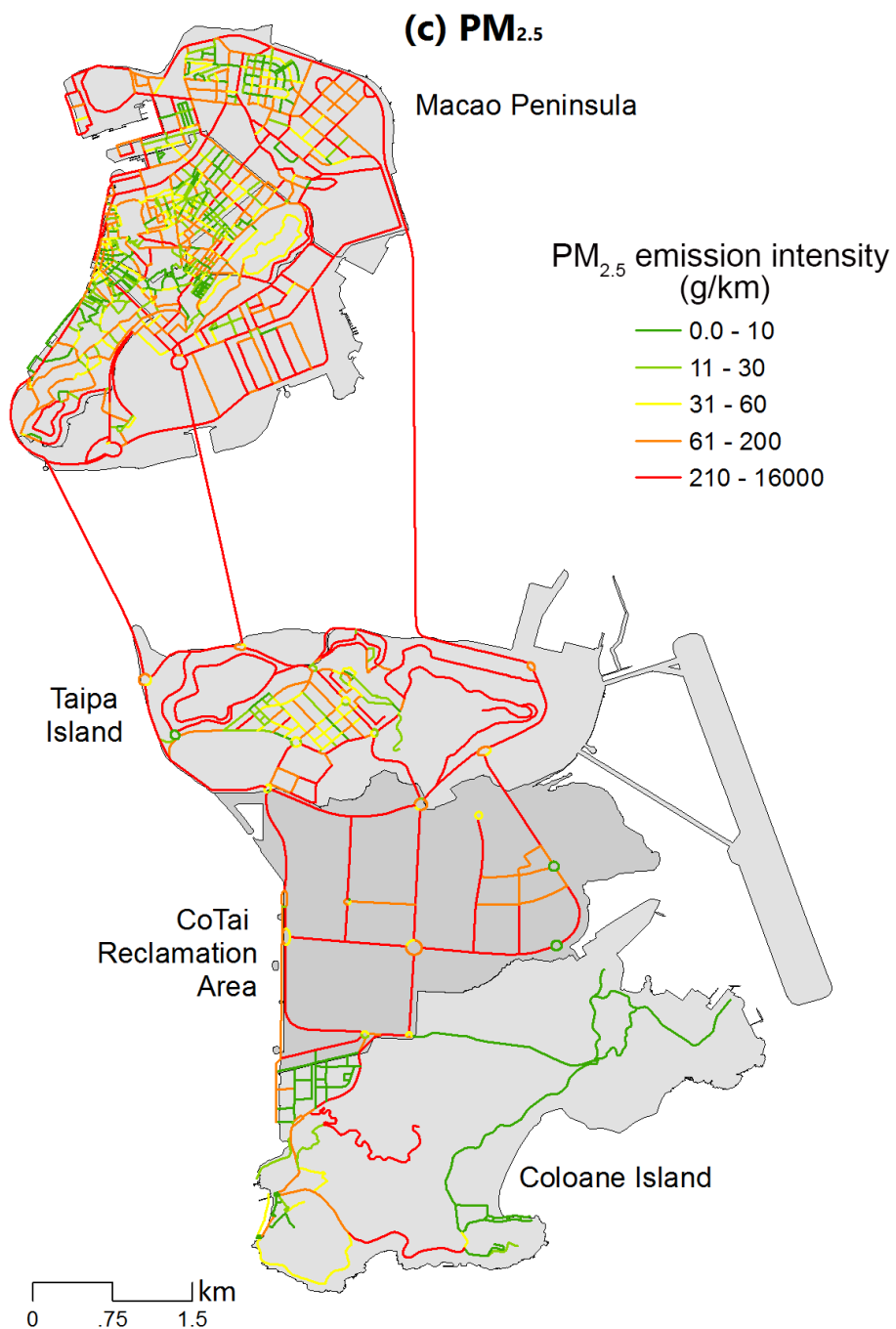


Fig. S8. The spatial distribution of vehicle emissions for (a) CO, (b) THC, (c) PM<sub>2.5</sub> and (d) CO<sub>2</sub> in Macao during a typical weekday of 2010

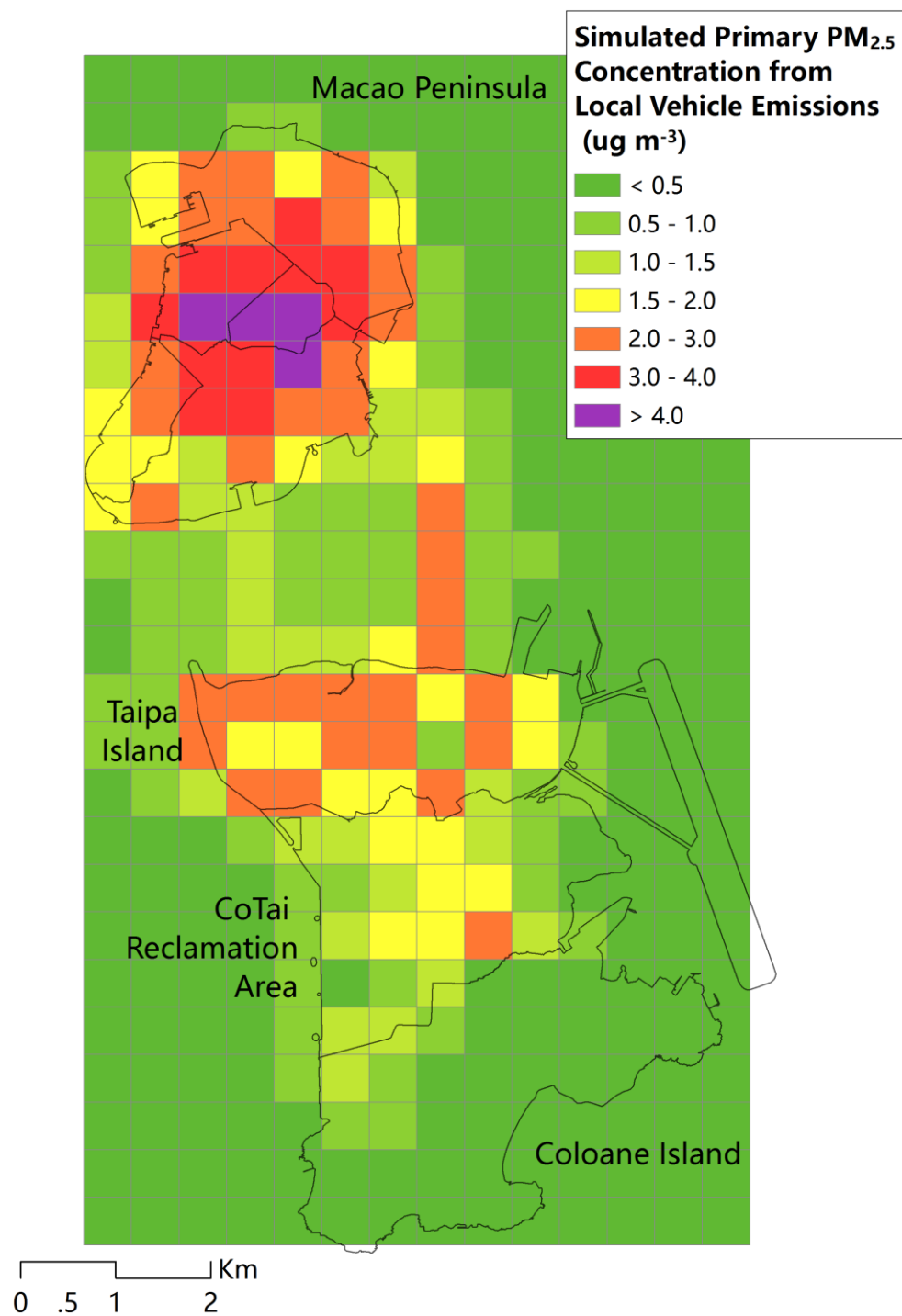


Fig. S9. Simulated vehicle-contributed concentrations of PM<sub>2.5</sub> in Macao during weekdays of November, 2010

## Air Quality Modeling with the CMAQ model

In this study, the Weather Research Forecasting model (WRF, version 3.3) and Community Multiscale Air Quality model (CMAQ, version 4.7.1) were employed to investigate impacts from regional background, cross-boundary transport and other area sources. National Centers for Environmental Prediction (NCEP) and the automated data processing (ADP) data, with a resolution of  $1^{\circ} \times 1^{\circ}$  were used as initial guess fields of WRF. Four-dimensional data assimilation (FDDA) is applied for objective analysis. Detailed physical parameterizations include the Kain-Fritsch 2 cumulus scheme, the Pleim-Xiu PBL scheme and land surface model, the mixed phase explicit moisture scheme for cloud microphysics, the cloud-radiation shortwave radiation scheme and the Rapid Radiative Transfer Model (RRTM) longwave radiation scheme. The Meteorology Chemistry Interface Processor (MCIP) version 3.6 was applied to process the meteorological data to the data format for CMAQ. CB-05 gas-phase chemical mechanism with AERO5 aerosol module were used. The aerosol thermodynamic equilibrium model is ISORROPIA.

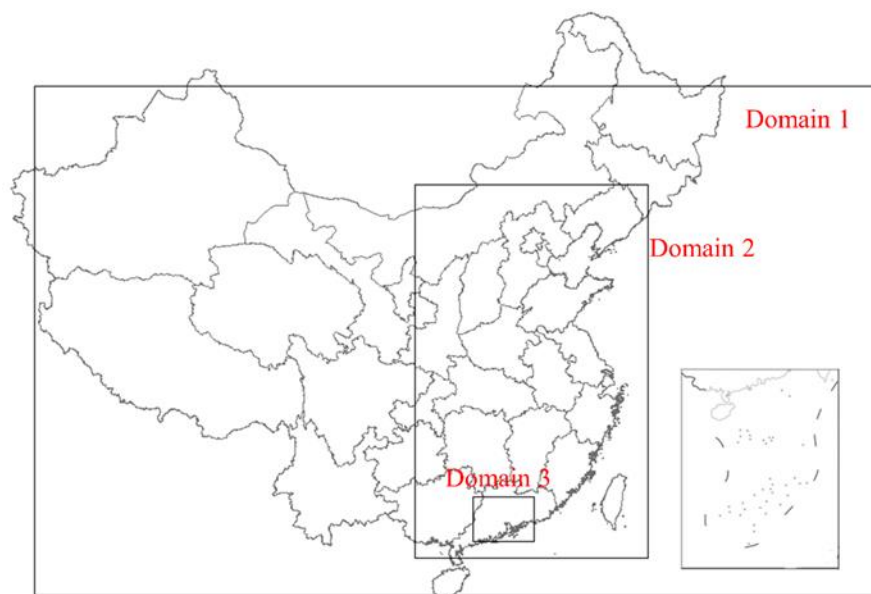


Fig. S10. Triple-nested simulation domain of CMAQ at a horizontal grid resolution of 36 km, 12 km and 4 km.

A triple-nested simulation domain, shown in Figure S10, was used in this study. Domain 1 covers most of China of  $36 \text{ km} \times 36 \text{ km}$  horizontal resolution. Domain 2 covers East of China with  $12 \text{ km} \times 12 \text{ km}$  horizontal resolution. Domain 3 covers Pearl River Delta (PRD) with  $4 \text{ km} \times 4 \text{ km}$  horizontal resolution. 23 vertical layers ( $\sigma$ : 1.000、0.995、0.988、0.980、0.970、0.956、0.938、0.916、0.893、0.868、0.839、

0.808、0.777、0.744、0.702、0.648、0.582、0.500、0.400、0.300、0.200、0.120、0.052 and 0.000) were included ranging from surface to 100 mbar. The configurations of chemical initial conditions and boundary conditions, and regional emission inventory are consistent with our previous papers (Zhao et al 2013a, 2013b). The local emissions for other sectors (e.g., residential, power, and industrial sectors) in Macao were provided by the Macao Environmental Protection Bureau, together with the vehicle emissions estimated by this study. The simulated results of regional air quality are presented in Fig. S11.

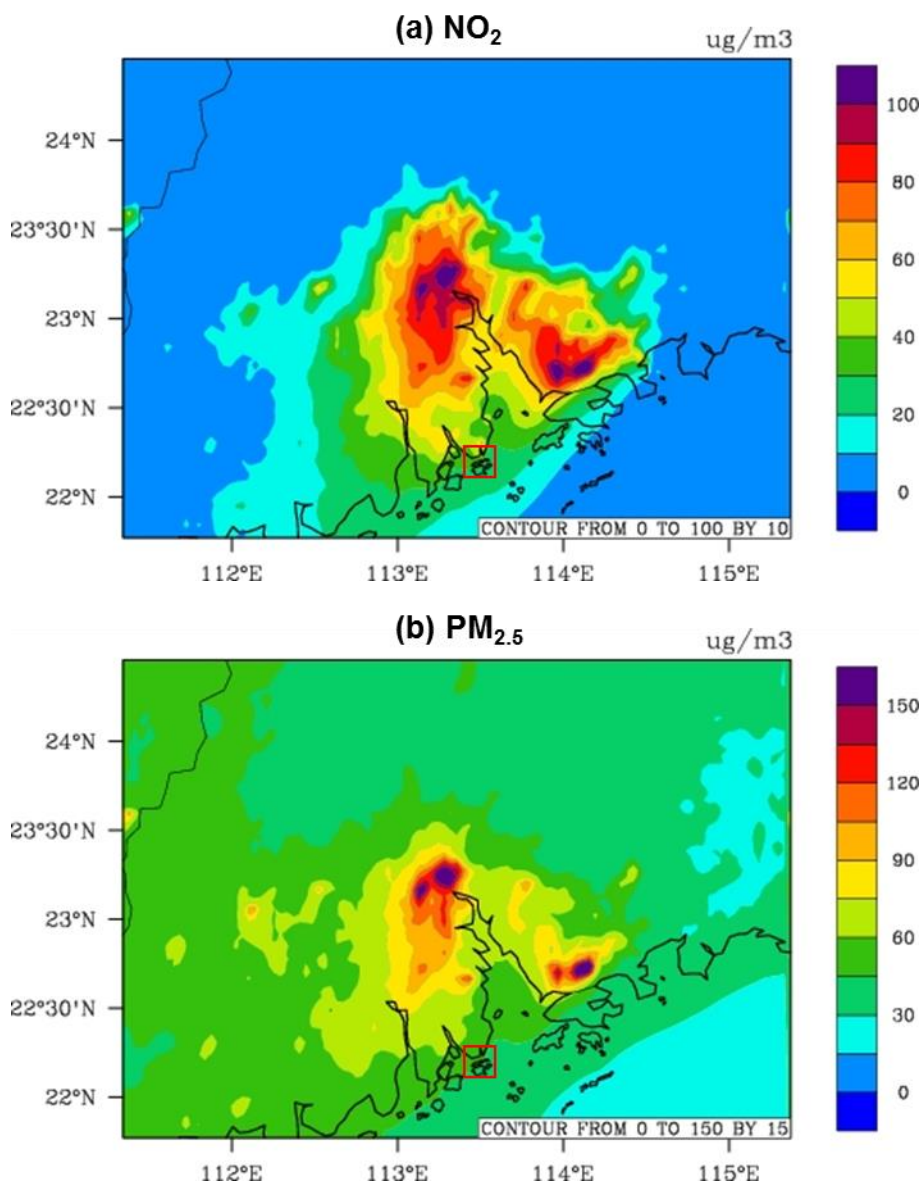


Fig. S11. Monthly average of simulated concentrations for  $\text{NO}_2$  and  $\text{PM}_{2.5}$  in the Pearl River Delta Region (i.e., Domain 3), November 2010



Simulated monthly average concentrations of NO<sub>2</sub> and PM<sub>2.5</sub> (November 2010) of the 4 km × 4 km cells are presented in Table S3, within which the air quality stations are located. We also include the observed NO<sub>2</sub> and PM<sub>10</sub> concentrations during November 2010 for comparison. It is noted that the PM<sub>2.5</sub> monitoring was not conducted in Macao until July 2012. The one-year monitoring results in 2013 indicate that the mass fraction of PM<sub>2.5</sub> in PM<sub>10</sub> was approximately 60%. The CMAQ model is not capable of the high NO<sub>2</sub> concentrations in Macao with normalized mean bias of -37% to 55%. On the other hand, although the AERMOD model may yield higher NO<sub>2</sub> concentrations in traffic populated areas, however, the model limitations would bring in considerable uncertainty (e.g., diurnal fluctuations). Thus, we suggest that future efforts are required to develop more advanced air quality model to enhance spatial heterogeneity and chemical transport at the same time.

**Table S3.** Comparison of simulated and observed air pollutant concentrations, monthly average of November 2010 (μg m<sup>-3</sup>, mean ± standard deviation)

Monitoring sites	Observed NO <sub>2</sub>	Simulated NO <sub>2</sub>	NMB	Observed PM <sub>10</sub>	Simulated PM <sub>2.5</sub>	NMB <sup>a</sup>
MP 1	68.0±12.7	37.8±16.7	-44%	99.1±28.6	41.2±18.3	-31%
MP 2	64.9±13.1	40.8±14.3	-37%	96.6±30.3	41.8±17.1	-28%
Taipa 1	52.2±10.4	33.1±16.8	-37%	84.8±23.1	37.0±16.7	-27%
Taipa 2	55.1±14.8	29.3±15.9	-47%	86.5±14.8	33.2±16.6	-36%
Coloane	65.0±17.3	29.8±16.5	-54%	86.1±24.9	34.8±17.8	-33%

Note: <sup>a</sup> We assume that the mass fraction of PM<sub>2.5</sub> in PM<sub>10</sub> was 0.6.

#### References

- Zhao, B.; Wang, S. X.; Dong, X. Y.; Wang, J. D.; Duan, L.; Fu, X.; Hao, J. M.; Fu, J., Environmental effects of the recent emission changes in China: implications for particulate matter pollution and soil acidification. *Environmental Research Letters* **2013a**, 8, (2).
- Zhao, B.; Wang, S.; Wang, J.; Fu, J. S.; Liu, T.; Xu, J.; Fu, X.; Hao, J., Impact of national NO<sub>x</sub> and SO<sub>2</sub> control policies on particulate matter pollution in China. *Atmospheric Environment* **2013b**, 77, (0), 453-463.