The Innovation, Volume 4

# Supplemental Information

# Antagonism between ambient ozone increase and urbanization-ori-

## ented population migration on Chinese cardiopulmonary mortality

Haitong Zhe Sun, Junchao Zhao, Xiang Liu, Minghao Qiu, Huizhong Shen, Serge Guillas, Chiara Giorio, Zosia Staniaszek, Pei Yu, Michelle W.L. Wan, Man Mei Chim, Kim Robin van Daalen, Yilin Li, Zhenze Liu, Mingtao Xia, Shengxian Ke, Haifan Zhao, Haikun Wang, Kebin He, Huan Liu, Yuming Guo, and Alexander T. Archibald

# **SUPPLEMENTARY MATERIALS**

# **Antagonism between ambient ozone increasing and urbanization-oriented population migration on Chinese cardiopulmonary mortality**

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*56 pages with 8 methodological notes, 17 tables, 20 figures, and 2 sections of listed contents for Supplementary Materials*

*Updated on 27 September 2023*

# **CONTENTS**

## **Supplementary Methods**



# **Supplementary Tables**



## **Supplementary Figures**





## **SUPPLEMENTARY METHODS**

### **Method S1 | Multi-model Fusion and Downscaling**

The initial version of ambient O<sub>3</sub> concentration dataset developed by space-time Bayesian neural network downscaler (BayNNDv1) followed two major steps: i) multi-model fusion<sup>1</sup>, and ii) urban-rural distinguished downscaling<sup>2</sup>. During multimodel fusion, a total of 10 CMIP6 historical simulations were selected as inputs for 1990–2014, and 6 SSP2-RCP4.5 scenario projections for 2015–20192. The imbalanced model numbers between the 2 phases (Phase 1: 1990–2014, Phase 2: 2015– 2019) introduced additional heterogeneities. The cross-scenario divergences were way lower than the cross-model discrepancies, and thus we replaced SSP2-RCP4.5 with SSP3-RCP7.0 to reach homogeneity with the maximum number of models between the two Phases. We fused 8 coupled earth system models with interactive chemistry as i) BCC-ESM1, ii) CESM2- WACCM, iii) EC-Earth3-AerChem, iv) GFDL-ESM4, v) GISS-E2-1, vi) MRI-ESM2-0, vii) UKESM1-0-LL, and viii) CCMI, an average of 2 earlier generation atmospheric models, CESM1-WACCM and CMAM<sup>3-10</sup>. All the involved CMIP6 model simulation outputs are downloaded from Earth System Grid Federation repository platform: https://esgf-node.llnl.gov/search/cmip6.

Following the established methodology with replacement of data sources and adding *in situ* observations during 2014– 2019 provided by China National Environmental Monitoring Centre (CNEMC), we improved the accuracy of BayNNDv1. The optimised product BayNNDv2 is of higher global overall accuracy *R*2=0.91, RMSE=4.5 ppb for urban, and *R*2=0.89, RMSE=5.2 ppb for rural sites.

#### **Method S2 | Phased Data Fusion**

As the base ambient  $O_3$  products were of different temporal coverage, time-period phased data fusion was conducted. For *Phase I* (Roman numerals were used here to avoid confusion with the aforementioned Phase 1) during 1990–2002, fusion with calibration were conducted on BayNNDv2 and M<sup>3</sup>-BME. Due to the lack of systematic observations in China during this period, we trained the supervised deep learning model merely based on the observation archives from Tropospheric Ozone Assessment Report (TOAR) project<sup>11</sup>, and predicted the ambient  $O_3$  for China assisted with geographic and sociodemographic features as a compromised choice. For *Phase II* of 2003–2012, BayNNDv2, M3-BME and CEML were blended after unification into 1/8°×1/8° spatial resolution. Still, no Chinese localised observations were involved, but satellite-based remote-sensing measurements were included to increase the reliability in capturing the spatiotemporal pattern. For *Phase III* of 2013–2019, we mixed all four base databases nested in China territory, supervised by *in situ* observations from China National Environmental Monitoring Centre (CNEMC). The urban-rural distinguishment was inherited from BayNNDv2, and data fusions were performed for urban and rural concentrations separately.

All ground-level site-based observations were aggregated into  $1/8^{\circ} \times 1/8^{\circ}$  as supervised training labels. The fusion processes can be expressed as follows:



where f stands for the trained elastic net linear regressor,  $s_1, s_2, s_3$  refer to the spatial geometric coordinates, and  $t_1, t_2, t_3$  are temporal periodical and sequential indicators as listed below<sup>12</sup>. Cross-validation test results and overall performance evaluations were summarised in Table S6.

$$
s_1 = \cos\left(2\pi \frac{\text{longitude}}{360}\right) \cos\left(2\pi \frac{\text{lattice}}{180}\right),
$$
  
\n
$$
s_2 = \cos\left(2\pi \frac{\text{longitude}}{360}\right) \sin\left(2\pi \frac{\text{lattice}}{180}\right),
$$
  
\n
$$
s_3 = \sin\left(2\pi \frac{\text{longitude}}{360}\right),
$$
  
\n
$$
t_1 = \cos\left(2\pi \frac{\text{month}}{12}\right),
$$
  
\n
$$
t_2 = \sin\left(2\pi \frac{\text{montth}}{12}\right),
$$
  
\n
$$
t_3 = \frac{\text{montth}}{360}.
$$

It should be furtherly noted that the BayNNDv2 urban-rural downscaled dataset was treated fully as a core basis dataset, and then 3 other well-developed datasets (M<sup>3</sup>-BME, CEML and TAP) were fused using elastic net regressor rather than being incorporated as auxiliary predictors for Bayesian neural network downscaler. We selected such design for the purpose of maintaining the temporal homogeneity, as the elastic net regressor would "*respect*" the source dataset *closest* to the labels for supervision (i.e. observations), and regard the other two datasets as a strategy of "*belt and braces* (*double insurance*)" in case the Bayesian neural network "*missed*" any information that had been captured by M3-BME, CEML or TAP. The elastic net regressor (instead of other base machine learners like random forest or gradient boosting decision tree) would not substantially destroy the spatiotemporal pattern of the very input dataset closet to the observations, and tune with the rest input datasets if necessary. It can effectively avoid causing "*fractures*" in the "*junction*" year of different datasets (e.g. CEML starts from 2003, and hence 2003 is a junction year that the temporal fracture will be inclined to occur). When calculating the importance features of *Phase III* (2013–2019), the core dataset BayNNDv2 occupied 96.8% and 94.1% weights for urban and rural predictions, respectively, justifying the necessity and credibility of long-term global-scale space-time integrated training.

#### **Method S3 | Detailed specification of Chinese administrative divisions**

We used 7-division scheme in this study. This scheme of Chinese Administrative Geographical Division considers geography, history, culture, and ethnicity into comprehensively. The municipalities directly under Chinese Central Government and Autonomous Regions are all of provincial executive level. *Northeast China* includes 3 provinces: Heilongjiang, Jilin, and Liaoning. *North China* includes 3 provinces: Hebei, Shanxi, Inner Mongolia Autonomous Region; and 2 direct-administered municipalities: Beijing and Tianjin. *East China* includes 7 provinces: Shandong, Jiangsu, Anhui, Zhejiang, Jiangxi, Fujian, and Taiwan; and a direct-administered municipality: Shanghai. *Central China* includes 3 provinces: Henan, Hubei, and Hunan. *South China* includes 3 provinces: Guangxi Zhuang Autonomous Region, Guangdong, and Hainan; and 2 Special Administrative Regions (SAR): Hong Kong SAR and Macao SAR. *Southwest China* includes 4 provinces: Tibet Autonomous Region, Yunnan, Sichuan, and Guizhou; and a direct-administered municipality: Chongqing. *Northwest China* includes 5 provinces: Xinjiang Uygur Autonomous Region, Qinghai, Gansu, Ningxia Hui Autonomous Region, and Shaanxi. *Jing-Jin-Ji* (JJJ) urban agglomeration consists of Beijing, Tianjin, 11 prefecture-level cities (Shijiazhuang, Baoding, Tangshan, Langfang, Qinhuangdao, Zhangjiakou, Chengde, Cangzhou, Hengshui, Xingtai, Handan) in Hebei Province, and Anyang in Henan Province. "Ji" ("冀", pronounced as jì) is the ancient name of Hebei Province. Some schools abbreviate the megalopolis as BTH (Beijing, Tianjin, and Hebei). *Cheng-Yu* (CY) urban agglomeration consists of Sichuan Province (excluding Liangshan, Panzhihua, Aba, Ganzi, Guangyuan, Bazhong) and Chongqing (excluding Qianjiang, Pengshui, Youyang, Xiushan, Chengkou, Wushan, Wuxi, Fengjie). The alternative historical name of Chongqing is "Yu" ("渝", pronounced as yú), and hence for the phonological harmony, Chengdu-Chongqing district is more commonly shorted as Cheng-Yu rather than Cheng-Chong. *Yangtze River Delta* (YRD) urban agglomeration consists of Jiangsu Province, Anhui Province, Zhejiang Province, and Shanghai. The China *Greater Bay Area* (GBA)

circumscribes Hong Kong SAR, Macao SAR, and 9 prefecture-level cities in Guangdong Province (Guangzhou, Shenzhen, Foshan, Dongguan, Zhongshan, Jiangmen, Huizhou, Zhuhai, Zhaoqing), which is alternatively entitled as the Guangdong– Hong Kong–Macao Greater Bay Area. The 9 cities in Guangdong Province are collectively named as Pearl River Delta (PRD) Economic Zone. The 7 Chinese administrative divisions and 4 megalopolises were mapped in Figure S1.

#### **Method S4 | Identification of mortality causes**

Meta-analyses were performed on the extracted cohort-based epidemiological evidences (e.g. hazard ratio, HR) relevant to long-term O<sub>3</sub> exposure from systematic review updated until October 2022. All reported mortality causes were included for meta-analysis extended from the latest relevant systematic reviews<sup>13,14</sup>, and the causes with pooled positive relative risks were considered for mortality estimation. Applying the Hunter-Schmidt estimator, 6 mortality causes (might not be mutually exclusive due to hierarchically overlapping) were identified to be of positive relative risks: non-accidental causes, chronic respiratory diseases, chronic obstructive pulmonary disease, cardiovascular diseases, ischaemic heart diseases, and congestive heart failure, as plotted in Supplementary Figures 3–7, and potential publication biases were tested by trim-and-fill method (Figure S8).

In terms of mortality estimation, the non-accidental cause mortalities were narrowed to mortalities of non-communicable diseases (NCDs), as it is reasonable to assume the non-accidental causes other than NCDs (e.g. communicable, maternal, neonatal, and nutritional diseases, injuries, suicide and homicide, etc.) are of no association with ambient  $O_3$  exposure. In addition, mortality estimations in this study did not include the congestive heart failure which was not listed as an individual mortality cause in the GBD 2019 Study<sup>15</sup>. Therefore, further explorations on the nonlinear exposure-response relationships and excess mortality estimations only involve i) NCDs, ii) chronic respiratory diseases (CRDs), iii) chronic obstructive pulmonary disease (COPD), iv) cardiovascular diseases (CVDs), and v) ischaemic heart disease (IHD).

#### **Method S5 | Construction of exposure-response curve**

As the exposure-response association strengths may not necessarily follow the linear pattern, curved trends were explored by meta-regression enhanced by exposure-range resampling for the sake of more accurate risk estimation<sup>14,16,17</sup>. Most of the pre-existing studies were conducted on the North American and European countries where ambient  $O_3$  pollution has been effectively constrained in the past decades, and thus the averaged exposure levels of the cohort participants were lower than the Chinese population. Under this circumstance, multi-cause mortality relative risks for Chinese residents estimated by conventional meta-regression method would rely on exposure extrapolation, leading to high uncertainties. To address this issue, exposure-range resampling would make full use of the literature-reported population exposure levels rather than the study-specific averaged exposure concentrations, so as to cover the exposure range as wide as possible and thus increase the estimation robustness.

The concentration-response curves were adopted in priority if reported in the literature. We queried the authors of the published studies providing the non-linear concentration-response relationships for the detailed values of the curves; and as for the studies we did not receive responses by October 2022, we recovered the values directly from the figure by mean of geometric measurement in Microsoft Visio. If the original studies did not explore the concentration-response trends, linear relative risk models were assumed across the reported exposure range, with the theoretical minimum risk exposure level (TMREL) presumed to be a random value uniformly distributed between the minimum and lowest  $5<sup>th</sup>$  percentile following a previous study<sup>18</sup>. The statistical approach to reproduce the lowest 5<sup>th</sup> percentile was described in a prior systematic review<sup>14</sup>,

and the resampled/imputed distribution statistics were listed in Table S15. The estimated concentration-response curves for mortality risks of NCDs, chronic respiratory diseases, COPD, cardiovascular diseases, and ischaemic heart disease were presented in Figure S9.

The exposure range resampling reproduced the exposure level (OSDMA8 in ppb) by every 1 ppb increment between the literature-reported minimum and maximum exposure level as  $x$ . In linear-model presumed relative risk recovering, for each resampled exposure concentration x, the corresponding effective exposure "dose"  $\Delta x$  is defined as

$$
\Delta x = ReLU\{x - TMREL\},\
$$

where ReLU is the rectified linear unit choosing the greater value between 0 and  $x - \textit{TMREL}$ . Given the reported risk association (i.e. HR) with 95% confidence interval (CI) as HR<sub>LB</sub> to HR<sub>UB</sub> by every  $\Delta y$  incremental exposure, the relative risk with 95% CI at exposure concentration  $x$  can be calculated as

$$
HR_x = e^{\ln HR \cdot \Delta x/\Delta y};
$$
  
\n
$$
HR_{LB,x} = e^{\ln HR_{LB} \cdot \Delta x/\Delta y};
$$
  
\n
$$
HR_{UB,x} = e^{\ln HR_{UB} \cdot \Delta x/\Delta y}.
$$

Following the procedures illustrated above leads to an exposure-response sequence for each study that did not report the concentration-response curve; the fully resampled sequences undergo MR-BRT multi-study pooling with the literaturereported exposure-response curves.

Several previous studies have provided estimations on O<sub>3</sub>-associated excess COPD mortality. Taking 2017 as an example, the GBD report estimated the COPD mortality as 113 (95% Uncertainty Interval, UI: 53-178) thousand<sup>17</sup>, which is lower than our results (183, 95% UI: 125-245 thousand), as GBD applied undersized RR values<sup>19</sup>. Yin et al. reported 178 (95% UI: 68–286) thousand COPD deaths attributable to  $O_3$  exposure<sup>20</sup>, which is more coherent with our result in terms of central estimate. This is because the RR value they used (RR=1.040, 95% CI: 1.013-1.067) from a single cohort study<sup>21</sup> is close to the multi-study pooled RR by our meta-analysis (RR=1.056, 95% CI: 1.029–1.084); but their result is still of greater estimation uncertainty. Contrarily, Malley et al. used oversized risk association strength (RR=1.12, 95% CI: 1.08–1.16) and reported 316 (95% UI: 230-403) thousand respiratory deaths for  $2010^{22}$ , which is substantially higher than our estimates (179, 95% UI: 122–241 thousand). The unneglectable cross-study divergences and great estimation uncertainties reveal the insufficiency of epidemiological evidences. Furthermore, leaving out cardiovascular mortality risks leads to dubious conservative overall estimations. We consider cardiopulmonary beyond respiratory mortality for the first time and thus provide an *aggressive* estimation to update the literature.

#### **Method S6 | Construction procedures of gridded population dataset**

The step-by-step procedures to construct the gridded Chinese population dataset are illustrated in the flowchart (Figure S17), in which the rounded rectangles indicate procedural semi-manufactures, rectangles refer to the initial input and final output datasets, and the number-marked arrows represent operations.

*Starting point: UN WPP-adjusted GPWv4.* The Gridded Population of the World with adjustment from United Nation World Population Prospects<sup>23</sup> (version 4.11) was set as the footstone, as it is the latest global population distribution product with the finest spatial resolution (30"×30") and densest temporal coverage (2000-2019).

*Step 1: Spatial re-gridding.* The spatial resolution of finally enhanced ambient O<sub>3</sub> concentration dataset with urban-rural distinguishment is 1/8°×1/8°, based on which the population exposure levels were assessed. By averaging the 15×15 adjacent grids ( $1/8^\circ$ =30"×15), the raw 30"×30" dataset was re-gridded into  $1/8^\circ \times 1/8^\circ$ .

*Step 2: Temporal extrapolation.* The GPWv4 dataset covers 20 studied years: 2000–2019. For each re-gridded 1/8°×1/8° cell, the restricted cubic spline regression model with 3 knots was applied to the cell-level population against year, so as to extrapolate the temporal coverage onto the complete study years: 1990–2019, following previous studies<sup>1,24</sup>.

*Step 3: China localisation.* The global longitude-latitude-based grids were geographically projected onto the map of China provided by the Ministry of Natural Resources of People's Republic of China, and all grids belonging to China territory were extracted for further processing. Geographical mapping and administrative division projection (i.e. country, provinces, and prefecture-level cities) were performed in QGIS (version 3.26.10).

*Step 4: Urban-rural distinguishment*. The Population Dynamics dataset (version 1.01), identifying urban and rural population counts for each  $1/8^\circ \times 1/8^\circ$  grid<sup>25</sup>, was extrapolated onto 30 consecutive years by mean of restricted cubic spline model2, based on which the urban and rural population fractions were calculated. The cell-specific fractions were then multiplied onto the 30-year extrapolated GPWv4 China gridded population dataset (i.e. procedural semi-manufactures of Step 3), to update the urban-rural distinguished population distribution. The consensus has been widely accepted that GPWv4 datasets reporting 20 consecutive years were more reliable than interpolated data products.

*Step 5: Urban-rural calibration.* The China Statistical Yearbook series reported the numbers of urban and rural residents for each year, with which the estimated values were linearly aligned. For an instance, if the predicted total count of urban residents of a certain province (Step 4) was  $Pop_{pred}$  while the factual count provided by the China Statistical Yearbook was Pop<sub>stat</sub>, the urban population count for each grid was then multiplied by a coefficient of Pop<sub>pred</sub>/Pop<sub>stat</sub>. Province-level calibrations were performed for 2005–2019 in accordance with the Yearbook precision, while nation-level calibrations were conducted for 1990–2004 as a compromise given the data unavailability.

*Step 6: Age group specification.* Fractions of population aged above 25 were retrieved from GBD Population Estimates 1950–2019,26 and the China Statistical Yearbook series 2004–201927. Values provided by the China Statistical Yearbook series were adopted in priority for 2004–2019, while for the earlier years 1990–2003 when the China Statistical Yearbook did not archive the population pyramid, the GBD Population Estimates were used as a compromise. The nation-level yearspecific fractions were multiplied onto each grid to identify the counts of population age ≥25. After this step, the enhanced gridded population age ≥25 differentiated with urban and rural residence was used as the capstone dataset for main analysis.

*Step 7: Gender group specification.* Genders were furtherly specified for sensitivity analysis. Province-level gender proportions for 2000–2019 and nation-level gender proportions for 1990–1999 were obtained from the China Statistical Yearbook series 1990–2019<sup>27</sup>. The province- or nation-level male and female proportions were uniformly applied onto each grid circumscribed inside the corresponding province or the whole country territory, respectively.

*Step 8: Dataset encapsulation.* After all the aforementioned data processing, the gridded population was structured into the meta-dataset:  $1/8° \times 1/8°$  spatial resolution; yearly resolved spanning 1990-2019; each grid encapsulating 4 population counts as: i) urban male age ≥25, ii) urban female age ≥25, iii) rural male age ≥25, and iv) rural female age ≥25.

#### **Method S7 | Sensitivity analyses**

Long-term ambient  $O_3$  tracking covers earlier years beyond the satellite-based remote sensing measurements or chemical reanalysis (i.e. 1990–2002), indicating predictions would merely relied on the CMIP6 numerical simulations for this period. We therefore extended a sensitivity analysis for the first-stage space-time Bayesian neural network-based data assimilation during 2003–2019 under two scenarios, as fusing eight CMIP6 models with (ScA) and without (ScB) a machine-learningcalibrated remote-sensing measurements and chemical reanalysis outputs<sup>28</sup>, assisted with over 40 auxiliary features<sup>2</sup>.

We then evaluated the accuracies of 10-fold cross-validation tests by random split (70% dataset matched with observations), external validation tests (the rest 30%), and overall fitting, as summarized in Table S16. We compared the developed ambient  $O_3$  datasets under the two scenarios by coefficient of variation (CoV): standard deviation divided by the arithmetic mean. We concluded that the deep-learning-based prediction accuracies by solely using CMIP6 simulations were as competitive as fusing additional measurements, and no substantial discrepancies were observed between ScA and ScB (CoV=1.0%, spatiotemporal 5-95<sup>th</sup>%ile: 0.1-2.8%).

We furtherly split the full dataset manually for cross-validation tests under ScB, maintaining the temporal coherence: i) 2003–2012 for training and 2013–2019 for testing; ii) 2003–2007 and 2015–2019 for training and 2008–2014 for testing; and iii) 2010–2019 for training and 2003–2009 for testing. All three temporally staged cross-validation tests had revealed good performances (*R*2=0.90, 0.92, 0.92; RMSE=2.86, 2.71, 2.70 ppb, respectively for the three tests)**.** The constrained crossscenario divergences and stable temporal generalizability verified the credibility of model-based ambient  $O_3$  tracking in the earlier years.

Parallel with the curved risk model, the *linear risk model* was adopted for attributable mortality estimation as reference, which assumed that relative risks change linearly with the exposure level  $x$  following

$$
RR_x = e^{lnRR \cdot \frac{\Delta x}{\Delta y}}
$$

where RR is the multi-study pooled value scaled in each  $\Delta y$  incremental exposure, and  $\Delta x$  is the effective dose above the TMREL.

We performed a series of further sensitivity analyses on the estimation for 2017 as an example. The exposure-response relationships might be the major source of estimation uncertainty, and thus we applied the multi-study pooled RRs onto the simplest log-linear model parallel to the curved model as presented in the main results. The threshold (also known as TMREL or low-concentration cut-off) for long-term  $O_3$  exposure-associated mortality risk was also contentious, and thence we tested several values as directed in literature: i) the global lowest  $5<sup>th</sup>$  percentile PWE in 2017 by BayNND, 42.6 ppb (Scenario 1, Sc1); ii) the 30-year global lowest 5<sup>th</sup> percentile PWE by BayNND, 40.8 ppb (Sc2); and iii) the maximum of literature-reported lowest 5<sup>th</sup> percentile exposure levels from studies included for meta-analysis, 44.0 ppb (Sc3). We used the grid-averaged ambient  $O_3$  concentrations to quantify population exposure, supposing the ambient  $O_3$  exposure levels were not distinguished for urban and rural environments, as Sc4. Gender-specified mortality metrics other than the gender-standardized estimates reported by IHME, were used as Sc5. Province-specific mortality metrics for 2017 provided by China CDC were applied as Sc6<sup>29</sup>. In Sc7, we replaced the O<sub>3</sub> tracking dataset with M<sup>3</sup>-BME solely, which was used in the GBD 2019 study. In Sc8, we adopted cardiovascular mortality risk association (RR=1.227, 95% CI: 1.108–1.359, *p*-*value*=0.79) pooled from 2 cohort studies on Chinese population reporting higher RRs<sup>30,31</sup>.

Estimations for excess deaths differentiating the designed schemes were summarized in Table S17. The cross-scheme discrepancies were constrained not to exceed 10%, and therefore sensitivity analyses validated the robustness of our mortality estimations, verified the coherence of the data sources, and justified the rationality of innovations in our study design.

#### **Method S8 | Cross-validation for spatiotemporal generalizability**

Since China lacked systematic ground-level measurements in earlier years before 2013, and the observation sites deployed in urban and rural environments were disproportional. We therefore decided to train the model at global scale with sufficient supervision by observations, and conducted strengthened rigorous cross-validation tests on the spatiotemporal extrapolation reliability to verify the generalizability of the deep learning downscaling algorithm. Besides the cross-validation and external validation tests by random split, we extended region-clustered cross-validation tests on spatial extrapolation capability (cvs<sub>1</sub>: training on North America, testing on Europe; cvs<sub>2</sub>: training on Europe, testing on North America; cvs<sub>3</sub>: training on North America and Europe, testing on Asia; and cvs4: training on locations outside China, testing on China), and staged cross-validation tests on global-scale temporal generalization (cvt<sub>1</sub>: training on 1990-2013, testing on 2014-2019; cvt<sub>2</sub>: training on 1990-2007 and 2014-2019, testing on 2008-2013; cvt<sub>3</sub>: training on 1990-2001 and 2008-2019, testing on 2002–2007; cvt<sub>4</sub>: training on 1990–1995 and 2002–2019, testing on 1996–2001; cvt<sub>5</sub>: training on 1996–2019, testing on 1990–1995) for the second-stage urban-rural differentiated downscaling. Spatiotemporal generalizability tests are summarized in Table S7.

# **Supplementary Tables**

## **Table S1 | Province-level average of ambient ozone concentrations in 1990 and 2019.**

Urban, rural and population-weighted exposure (PWE) concentrations are scaled as 6-month (April to September) ozone-season daily 8-hour maximum average (OSDMA8) in ppb for either year. Statistics include the regional median and spatial 5-95th percentile range. Hong Kong SAR and Macao SAR have realised full urbanisation before 1990, and thus rural concentrations are not considered.



### **Table S2 | Regional and nationwide 1990 mortality metrics associated with ozone exposure.**

Excess cardiopulmonary mortalities are defined as the total deaths caused from COPD and all-type cardiovascular diseases. Three mortality metrics are considered as i) number of excess deaths in thousand, ii) mortality rate per 100 000, and iii) years of life lost (YLLs) in million years. Estimates are summarised by median with 95% uncertainty intervals from 1000-time Monte Carlo bootstrap.



### **Table S3 | Historical 30-year regional and nationwide ozone-associated mortality trends.**

Longitudinal trends scaled in decadal average change rates are calculated by log-linear meta-regression maximum likelihood estimator from the annually resolved values with 95% confident intervals (CIs). When estimated trend approaches 0, an additional decimal place is reserved.



### **Table S4 | 30-year multi-cause cross-sectional baseline mortality rates of Chinese population.**

Mortality rates (per 100 000) of 5 causes (NCDs, non-communicable diseases; CRDs, chronic respiratory diseases; COPD, chronic obstructive pulmonary disease; CVDs, cardiovascular diseases; IHD, ischaemic heart disease) are retrieved from the IHME GBD 2019 result portal (https://vizhub.healthdata.org/gbd-results), with 95% uncertainty intervals.



#### **Table S5 | Associations between rural-urban ambient ozone difference and land cover features.**

The rural-urban differences are defined as localised (i.e. within a prescribed downscaled spatial grid) rural ambient  $O_3$  concentration minus the adjacent urban levels. Backward stepwise selection (*p*-value <0.20) is adopted to identify associated variables. Features with high collinearity is censored as appropriate (e.g. *emission rate of BC, aerosol optical depth at 550 nm, and surface PM2.5 concentrations are deleted due to collinearity with emission rate of OC*). Regression coefficient *β*<sup>s</sup> shows the standardised effect of each feature when controlling all the other considered factors, reported with Wald's *p*-value and 95% CI. The population-related features are obtained from aforementioned calibration. The emission rates of NO<sub>x</sub>, total NMVOC, organic carbon (OC), NH<sub>3</sub>, CO and SO<sub>2</sub> are retrieved from Emission Inventory developed by Peking University (PKU-Inventory)<sup>32-42</sup> and Multi-resolution Emission Inventory for China (MEIC)<sup>43-49</sup>, while the emission rates of biogenic NMVOC are modelled by CESM2-WACCM (accessed from the CMIP6 repository: https://esgf-node.llnl.gov/search/cmip6). Biomass features, vegetation, and urban land occupation fractions refer to the Land Use Harmonisation database (*historical* experiment for 1990–2014 and *ssp370* experiment for  $2015 - 2019)^{50,51}$ .



Interpretation: The research hypothesis to test is that "spatial pattern of the rural-urban ambient O<sub>3</sub> differences can be reflected by sociodemographic and geographical features in spatial statistics". Taking the variable "urban land occupation" as an example, the standardised coefficient is positive, as *β*<sup>s</sup> = 0.086, 95% CI: 0.036–0.136, which means summarising from all studied cells across the 30 years, **the greater the**  urban land occupation is, the larger the rural-urban ambient O<sub>3</sub> gap will be. This coincides with the fact that greater urban land occupations usually indicate higher emissions to form aerosols, and higher urban aerosols suppress the urban  $O_3$  formation, finally making the ruralurban gaps greater (urban ↓, rural–urban ↑). Relevant characteristics such as urban population fraction (*β*=0.144, 95% CI: 0.106–0.182), and organic carbon emission (*β*=1.379, 95% CI: 1.286–1.473) thus also show positive partial correlations. For another example, the coefficient of C3 annual and perennial crops is also positive as *β*<sup>s</sup> = 0.201, 95% CI: 0.057–0.345. This is a typical rural indicator, meaning that larger C3 crop vegetated land occupations usually indicate higher biogenic VOC emissions to form rural O3, finally making the rural–urban gaps greater (rural ↑, rural–urban ↑). The other studied features can be interpreted in similar way, that emission rate of CO (*β*=0.164, 95% CI: 0.133– 0.195), emission rate of biogenic non-methane VOCs (*β*=0.231, 95% CI: 0.193–0.270), and other vegetation coverage (e.g. cropland, pasture and rangeland), as rural indicators, also display positive associations with intensified rural  $O<sub>3</sub>$  pollution.

### **Table S6 | Performance evaluations of phased data fusion with urban-rural distinguishment.**

Algorithm performance assessments include 10-fold cross-validation tests and full-scale overall evaluations separately for urban and rural sites for phased data fusion. Full-scale refers to model training, prediction and evaluation using full dataset. Due to heterogeneity in input data, cross-validation tests for 30-year full-length evaluation are not applicable (NA).



### **Table S7 | Evaluation of spatial and temporal extrapolation accuracy by space-time Bayesian neural network downscaler with urban-rural differentiation.**

Different from classical cross-validation tests by randomly splitting the dataset, spatiotemporal generalisability validation tests manually divide the initial dataset by location or time period. Region-clustered spatial generalisability tests use observations in aggregated regions for algorithm training, and assign observations in other aggregated regions for testing, including four sub-experiments (cross-validation for spatial generalisability, cvs1: training on North America, testing on Europe; cvs2: training on Europe, testing on North America; cvs3: training on North America and Europe, testing on Asia; and cvs4: training on locations outside China, testing on China). Period-staged temporal generalisability tests treat six consecutive years as testing subset based on trainings from the rest 24-year global-scale dataset, including five subexperiments (cross-validation for temporal generalisability, cyt<sub>1</sub>: training on 1990–2013, testing on 2014–2019; cyt<sub>2</sub>: training on 1990–2007 and 2014–2019, testing on 2008–2013; cvt<sub>3</sub>: training on 1990–2001 and 2008–2019, testing on 2002–2007; cvt<sub>4</sub>: training on 1990–1995 and 2002–2019, testing on 1996–2001; cvt5: training on 1996–2019, testing on 1990–1995). Prediction evaluation statistics include crude *R*<sup>2</sup> and RMSE (in ppb) before 1:1 linear regression calibration, together with linear regression slope (*k*) and intercept (*b*).



### **Table S8 | Quality assessment tool for observational cohort and cross-sectional studies.**

A. Was the research question or objective in this paper clearly stated?

B. Was the study population clearly specified and defined?

C. Was the participation rate of eligible persons at least 50%?

D. Were all the subjects selected or recruited from the same or similar populations (including the same time period)? Were inclusion and exclusion criteria for being in the study prespecified and applied uniformly to all participants?

E. Was a sample size justification, power description, or variance and effect estimates provided?

F. For the analyses in this paper, were the exposure(s) of interest measured prior to the outcome(s) being measured?

G. Was the timeframe sufficient so that one could reasonably expect to see an association between exposure and outcome if it existed?

H. For exposures that can vary in amount or level, did the study examine different levels of the exposure as related to the outcome (e.g., categories of exposure, or exposure measured as continuous variable)?

I. Were the exposure measures (independent variables) clearly defined, valid, reliable, and implemented consistently across all study participants?

J. Was the exposure(s) assessed more than once over time?

K. Were the outcome measures (dependent variables) clearly defined, valid, reliable, and implemented consistently across all study participants?

L. Were the outcome assessors blinded to the exposure status of participants?

M. Was loss to follow-up after baseline 20% or less?

N. Were key potential confounding variables measured and adjusted statistically for their impact on the relationship between exposure(s) and outcome(s)?

Source: https://www.nhlbi.nih.gov/health-topics/study-quality-assessment-tools.

### **Table S9 | Quality assessment of 29 included cohort studies for meta-analysis.**

Study-specific quality assessments aim to examine the reliability of the epidemiological evidence and ensure the quality for meta-analysis. A total of 14 assessment items are considered according to the Quality Assessment Tool of Observational Cohort and Cross-Sectional Studies developed by the National Institute of Health (NIH) (Table S8), and assigned with one score for each, and the tallied scores are translated into a rating of quality. Studies scoring full marks, 14, are categorised as "Good," 10–13 as "Fair", and <10 as "Poor."



# **Table S10 | GRADE assessment for evidence of ozone-associated mortality risks of NCDs.**



# **Table S11 | GRADE assessment for evidence of ozone-associated mortality risks of CRDs.**



# **Table S12 | GRADE assessment for evidence of ozone-associated mortality risks of COPD.**



# **Table S13 | GRADE assessment for evidence of ozone-associated mortality risks of CVDs.**



# **Table S14 | GRADE assessment for evidence of ozone-associated mortality risks of IHD.**



#### **Table S15 | Statistically resampled distributions of ozone exposure levels for each study.**

The distribution features include arithmetic mean, standard deviation (SD), minimum,  $5^{th}$ ,  $25^{th}$ ,  $50^{th}$  (median),  $75^{th}$ , and  $95^{th}$  percentile, maximum, inter-quartile range (IQR), and full range, based on ozone exposure concentrations scaled by OSDMA8 metric in ppb. Values in **Bold** font represent the statistics reported by literature, while the rest indicate imputed values. Detailed resampling procedures and imputation accuracy evaluation can be found in a previous study $14$ .



Note: Jerrett et al. 2009 did not report the arithmetic mean and standard deviation directly. The values were derived by weighted averaging the centric concentrations of 4 exposure intervals on the populations given in Table 1 from the original literature. Zanobetti et al. 2011 did not provide the exposure distribution features directly. The quartiles were extracted from the legends in Fig. 1 of the original literature.

Methods: To reproduce the distribution, the arithmetic means and standard deviations (σ) were firstly extracted from literatures included for meta-analysis; if unavailable, the arithmetic means and standard deviations were estimated based on the reported descriptive statistics including median, first- and third-quartile, and all the other percentiles, to finally identify the parameters for presumed Gaussian normal distribution. Reported values were always treated as priority when divergences with estimations occurred. The centric level, arithmetic mean and median, were treated as exchangeable, but the arithmetic means were preferred. Theoretically, the minimum and maximum values of the distribution were not predictable, and thus 1<sup>st</sup> and 99<sup>th</sup> percentiles were used as proxies. Calculations for σ from key percentiles followed: 75th%ile = mean + 0.6745 σ, 95th%ile = mean + 1.6449 σ, and 99th%ile = mean + 2.3263 σ. If IQRs were stated, then IQR = 1.3490 σ; if the 5–95th percentile ranges were reported, then range5-95 = 3.2898 σ; if full minimum-maximum ranges were given, then range = 4.6527 σ. If more than one distribution features were provided, IQRs were more preferred for σ estimation due to higher robustness.

### **Table S16 | Evaluations of accuracies of deep-learning-based data assimilation with (ScA) and without (ScB) satellite-based remote-sensing measurements and chemical reanalysis outputs.**

Accuracy evaluations include coefficient of determination (R<sup>2</sup>) and root-mean-square error (RMSE, ppb) for 10-fold cross-validation tests using 70% observation-matched dataset by random split, external validation tests using 30% dataset, and overall model fitting for the two scenarios respectively. Given systematic *in situ* observations were unavailable in earlier years of China, and CNEMC sites were allocated in urban and rural environments disproportionally, model fitting and performance evaluations are conducted on global scale.



### **Table S17 | Multi-scenario sensitivity analysis.**

Sensitivity analyses are conducted on the estimation for 2017 as an example by multiple designed scenarios (Sc) beyond the main analysis. Cardiopulmonary mortality numbers are estimated for urban and rural population separately. Changes in total population mortalities (%) for different scenarios against the main analysis results are calculated. **Sc1:** Using log-linear risk model (rather than curved risk model in main analysis) with multi-study pooled RRs by random-effects meta-analysis, assuming threshold exposure level (also known as TMREL or lowconcentration cut-off) as the global lowest 5th percentile PWE in 2017 by BayNNDv2 dataset (see Method S1), 42.6 ppb. **Sc2:** Using loglinear risk model assuming threshold as the 30-year global lowest 5th percentile PWE by BayNNDv2 dataset, 40.8 ppb. **Sc3:** Using log-linear risk model assuming threshold as the maximum of literature-reported lowest 5<sup>th</sup> percentile exposure levels from studies included for metaanalysis, 44.0 ppb. Sc4: Using grid-averaged ambient ozone concentrations to quantify population exposure (following a previous study<sup>1</sup>), supposing the ambient ozone concentrations are not distinguished for urban and rural environments. **Sc5:** Using gender-specified other than the gender-standardised mortality metrics provided by IHME15 (GBD 2019 Study report). **Sc6:** Using province-specific mortality metrics for 2017 provided by China CDC<sup>29</sup>, as the cause-specific mortality rates are proportionally converted from the estimated DALY (disabilityadjusted life years) rates. **Sc7:** Using M<sup>3</sup>-BME ambient ozone tracking data product instead of the fused one. As M<sup>3</sup>-BME did not distinguish urban and rural ozone, urban and rural mortalities were not applicable (NA). **Sc8:** Using cardiovascular mortality linear risk association (RR=1.227, 95% CI: 1.108–1.359) pooled from two cohort studies exclusively on Chinese population73,74.



# **SUPPLEMENTARY FIGURES**



**Figure S1 | Mapping of 7 Chinese administrative divisions and 4 megalopolises.** 



### **Figure S2 | Nationwide and regional 30-year longitudinal trends of ambient ozone exposure.**

Population-weighted exposure (PWE) of total, rural- and urban-specified average exposure levels to ambient ozone are scaled in metric of OSDMA8. PWE levels are indicated by circles, based on which the rural–total (defined as rural-population average minus total PWE, similarly hereinafter) and total–urban differences are marked with directional bars. Upper apexes and lower vertexes represent nationwide or regional average ambient ozone exposure concentrations for rural and urban residents, respectively. Decadal average increasing rates (ppb per decade) are estimated by generalised linear model, as inserted in each subplot (T for total PWE; R for rural population exposure levels; U for urban population exposure levels). Longitudinal trends are summarised for nationwide, 7 geographical divisions, and 4 megalopolises (see Figure S1 for detailed definition).



#### **Figure S3 | Multi-study pooled mortality RR of NCDs associated with long-term ozone exposure.**

Risk strengths are defined as RRs per 10-ppb incremental exposure by OSDMA8 metric. The upper panel displays the meta-analysis results for all relevant cohort studies identified from systematic review, and the lower panel, censored meta-analysis, excludes i) studies conducted from the same cohort; ii) studies using over-smoothed metrics (e.g. 24-hour average) to quantify the individual-level exposure; iii) studies showing significant publication bias by trim-and-fill test (Figure S8); and iv) studies in which ozone hazards are mistakenly confounded by correlated or anticorrelated air pollutant species (e.g. NO2). For cohort duplication censoring, only one study covering the widest population is reserved in principle; unless different participant inclusion criteria are clearly stated (e.g. Di et al.64 conducted study on the whole Medicare cohort participants while Shi et al.71 focused on the low-exposure participants, thus both included for meta-analysis). Methodology of metric and unit unification has been illustrated in a previous review<sup>14</sup>. Supplementary Figs. 4-7 follow the same configuration.



**Figure S4 | Multi-study pooled mortality RR of CRDs associated with ozone exposure.**



**Figure S5 | Multi-study pooled mortality RR of COPD associated with ozone exposure.**



**Figure S6 | Multi-study pooled mortality RR of CVDs associated with ozone exposure.**





**Figure S7 | Multi-study pooled mortality RR of IHD and CHF associated with ozone exposure.**



**Figure S8 | Examination of publication biases by trim-and-fill method.** 

Scatter points are jittered appropriately to avoid excessive overlap.





The exposure-response (ER) curves are estimated for (**a**) ozone-associated mortality risks of non-communicable diseases (NCDs), (**b**) chronic respiratory diseases (CRDs), (**c**) chronic obstructive pulmonary disease (COPD), (**d**) cardiovascular diseases (CVDs), and (**e**) ischaemic heart disease (IHD) by mean of exposure range resampled meta-regression, Bayesian, regularised, and trimmed (MR-BRT). Exposures are quantified by 6-month (April–September) ozone-season 8-hour daily maximum average (OSDMA8) metric in ppb. Meta-regressions are performed on censored epidemiological evidence removing studies on duplicated cohort, unless the ER curved are clearly reported in the original literatures. Threshold exposure levels, also known as theoretical minimum risk exposure levels (TMREL), are indicated in each panel. The curved relative risks are used for mortality estimations as main analyses.



**Figure S10 | 30-year trend of hierarchical multi-cause mortality fractions.** 

Three hierarchical fraction values are calculated, as **a**) chronic obstructive pulmonary disease (COPD) excess deaths out of all chronic respiratory deaths, COPD/CRDs; **b**) ischaemic heart disease (IHD) excess deaths out of all cardiovascular deaths, IHD/CVDs; and **c**) total chronic respiratory and cardiovascular excess deaths out of deaths due to all non-communicable diseases, (CRDs+CVDs)/NCDs. The median values



## **Figure S11 | Gridded mapping of urban and rural cardiopulmonary premature deaths in 2019.**

The spatial resolution for grid-specific population ambient O<sub>3</sub> exposure assignment and associated mortality estimation with (a) urban and **(b)** rural differentiation is 1/8°×1/8° (approximately 10×10 *km*<sup>2</sup> ). Long-term ambient O3 exposure-associated excess cardiopulmonary premature deaths are defined as the total mortality cases caused from chronic obstructive pulmonary diseases (COPD) and all-type cardiovascular diseases. Intervals of colourbar are defined by Jenks natural breaks.



## **Figure S12 | Changes in population-weighted ozone exposure comparing 1990 with 2019.**

Panel **a** and **b** map population-weighted exposure (PWE) concentrations to ambient ozone (ppb) by OSDMA8 metric in year 1990 and 2019, respectively. Panel **c** presents the change of PWE (△PWE) from 1990 to 2019. Only 2 years of PWE are considered for comparison.

#### **Classical Downscaling a**



## **Figure S13 | Schematic diagram of (a) classical high-resolution downscaling and (b) urban-rural differentiated stacked downscaling.**

**a.** Classical downscaling requires predictions precise to target finer resolution (from 45 ppb to 47, 46, 26, … ppb for each finer cell, a total of 8×8=64 times of predictions), which however is frequently unfeasible in practice due to lack of high-resolution auxiliary datasets as predictors. Note in the diagram, spatial resolution and gridded values are manually faked, simply for illustration purpose. **b.** The left panel presents an 8×8 km<sup>2</sup> coarse cell of which the cell-level ambient O<sub>3</sub> concentrations (like 45 ppb) are sensible as an integrity (e.g. by remote-sensing measurement, model fusion calibrated by deep learning algorithms, etc.) to represent the average level of the whole cell. However, 8×8 *km*<sup>2</sup> is still a large domain with substantial intra-cell variability in term of ambient O3, as shown in the right part of panel **a**. Under the circumstance when it is unfeasible to realise higher-resolution downscaling (e.g. 1×1 *km<sup>2</sup>)* but there are multi-site urban- and rural-classified observations inside the studied cell, the urban and rural average ambient O<sub>3</sub> concentrations, 32 and 52 ppb, can be calculated and stacked to the cell, as shown in the right panel. The stacked downscaling only requires two times of predictions, from 45 to 32 ppb for urban concentration, and from 45 to 52 ppb for rural concentration. Note in the diagram, spatial resolution and gridded values are manually faked, aiming at illustrative presentations.



### **Figure S14 | Schematic diagram of Bayesian neural network multi-model fuser and downscaler.**

Right part demonstrates deep-learning-based multi-model fuser, and left part depicts urban-rural downscaler. The shaded elements refer to the external datasets not affected by neural network; the rectangle circumscribed elements indicate the input, processing and output variates inside the neural network; and non-rectangle circumscribed elements represent the final products. The schematic diagram is appropriately modified from a publication<sup>2</sup> with full consents from American Chemical Society Publications and involved authors.

**Abbreviations and denotations:** FC, *fully connected*; Sup., *supervised training*; DP, *dot product*; F, *multi-model fused output*; Obs, *observations*; ReLU, *rectified linear unit*; M, *calibrated CMIP6 models*; Softmax, *normalised exponential function*; tanh, *hyperbolic tangent function*.



#### **Figure S15 | Extrapolation validations on Chinese** *in situ* **observations with (a) urban, (b) rural, and (c) suburban differentiation by metric of monthly average of daily 8-hour maximum.**

Prediction-observation extrapolation evaluations span from May 2014 to December 2019, including statistics of coefficient of determination (*R*<sup>2</sup> ), root-mean-square error (RMSE, ppb), normalised mean bias (NMB, %, defined as difference that prediction minus observation proportion to observation), linear regression slope (*k*) and intercept (*b*). No Chinese *in situ* observations are included for Bayesian neural network framework training; predictions for urban and rural ambient O<sub>3</sub> in China are results of spatial extrapolation. Crude evaluations are performed on the observations and raw predictions by BayNND, and adjusted evaluations on the observations and 1:1-linearly calibrated predictions by BayNND. Adjusted evaluations are all of fixed NMB =0%, slope (*k*=1), and intercept (*b*=0). Panel (**b**) evaluates the coherence between "suburban"-labelled observations and rural O3 predictions, and (**c**) evaluates the consistency between "suburban"-labelled observations and urban  $O_3$  predictions. Data-based evidence reveals the "suburban"-labelled ambient  $O_3$  concentrations are closer to rural than urban pattern.



## **Figure S16 | Schematic diagram of urban-rural stacked gridded population upscaling.**

The left panel presents 1×1 *km*<sup>2</sup> higher-resolution population (in thousand) distribution in a target coarser 8×8 *km*<sup>2</sup> cell, in which urban and rural populations are defined based on population density. The upscaling process sums up the total finely gridded populations separately for urban and rural regions, and stacked the total urban population count 89,100 and rural population count 6,600 into the upscale coarse cell, as shown in the right panel. In further analyses, it will only be considered the upscaled cell-level total urban and rural populations (i.e. 89,100 and 6,600), rather than how the residents are spatially distributed (i.e. 2,100, 3,500, etc.). The populations scaled in coarse cell will be linked with ambient O<sub>3</sub> in same spatial resolution. Note in the diagram, spatial resolution and gridded values are manually faked, aiming at illustrative presentations.



### **Figure S17 | Flowchart of gridded population dataset construction and calibration.**

Rounded rectangles represent procedural data products; two rectangles refer to the initial input and final output datasets; number-marked arrows note manual operations for database development. Spatial resolution, space-time coverage, and population features are indicated in each dataset.



### **Figure S18 | Schematic diagram of cross-sectional population migration at cell-level definition.**

Panel (**a**) represents the initial population structure in an earlier year, when urban and rural populations are both 200,000. Panel (**b**) indicates a counterfactual scenario in a later year, that only population growth occurs without any urban-rural population structure change. The celllevel total population doubles from 400,000 to 800,000, among which urban and rural populations increase proportionally to 400,000. Panel (**c**) reflects the realistic population structure in the later year, when urban population is 700,000 and rural population is 100,000. Directly comparing the realistic situation (**a** and **c**), urban population expands by 500,000 and rural population shrinks by 100,000, which is affected both by population growth and migration. Adjusting the effect from population growth assuming urban and rural populations are of the same growing rate, the population migration flow can be equivalently perceived as 300,000 rural population inside the studied cell migrate to the urban environments in the same cell (comparing **b** and **c**), so that rural population can be perceived as 400,000–300,000=100,000, and urban population as 400,000+300,000=700,000.



### **Figure S19 | Schematic diagram of cell-level population exposure assignment in stacked context.**

The upper part presents upscaling of stacked urban-rural population, and the lower part shows the urban-rural differentiation of ambient O<sub>3</sub> concentrations. The right part demonstrates how urban (or rural) populations are linked to urban (or rural) ambient O3 exposure in the stacked concentrations. The right part demonstrates how urban (or rural) populations ar context, as 89,100 urban population are exposed to 32 ppb O<sub>3</sub> on average, and 6,600 rural population are exposed to 52 ppb O<sub>3</sub>.



#### **Figure S20 | External ozone prediction validations with literature reported observations.**

Enhanced external evaluations beyond CNEMC span from October 1993 to December 2019, including statistics of coefficient of determination (*R*<sup>2</sup> ), root-mean-square error (RMSE), normalised mean bias (NMB), linear regression slope (*k*) and intercept (*b*). Only point-to-point evaluations are performed, excluding literatures only reporting concentration ranges. All available metrics in monthly smoothed values are included with necessary cross-metric conversion. When multiple metrics are provided in literature, the daily 24-h average and diurnal maximum 8-h average are preferred. Crude evaluations are performed on the observations and raw predictions by BayNND, and adjusted evaluations on the observations and 1:1-linearly calibrated predictions by BayNND. Adjusted evaluations are all of fixed NMB = 0%, slope (*k* = 1), and intercept (*b* = 0). Full information can be found at Content S2**.** 

## **SUPPLEMENTARY CONTENTS**

### **Content S1 | Population density of "suburban"-labelled CNEMC observation stations in 2019.**

A total of 245 "suburban"-labelled CNEMC stations are projected to gridded population (see "*Population gridding and calibration*" section in *Methods*). Planar cell (approximated as rectangles) areas are calculated by planar meridional distance multiplied by planar parallel distance, where meridional (*m*) and parallel (*p*) distance follow the two formulae below, where *R* is the average Earth radius, 6378.137 *km*. Population where meridional (*m*) and parallel (*p*) distance follow the two formulae densities are calculated by cell-specific total population divided by cell area. Urban locations (U) are categorised by population density >1,500 people per *km*<sup>2</sup> (C1), and more conservatively, an additional urban categorisation by population density threshold >1,000 people per *km*<sup>2</sup> (C2) is provided as a sensitivity analysis. By C1, 242 out of 245 sites are classified as rural (R); by C2, 232 out sites are classified as rural, indicating "suburban"-labelled sites are more of rural sociodemographic characteristics.





Note:  $m = 2 \sin^{-1} \sqrt{\cos \left( \left( \frac{lat - \frac{1}{16} \right) \cdot \frac{\pi}{180}}{\cos \left( \left( \frac{lat + \frac{1}{16} \cdot \frac{\pi}{180}}{\sin \left( \frac{1}{16} \cdot \frac{\pi}{180} \right)} \right) \cdot \sin^2 \left( \frac{1}{16} \cdot \frac{\pi}{180} \right) \times R, \ p = 2 \sin^{-1} \sqrt{\sin^2 \left( \frac{1}{16} \cdot \frac{\pi}{180} \right) \times R}.$ 

#### **Content S2 | Literature-based external validations of urban-rural ambient ozone predictions.**

Accuracy evaluations on CNEMC observations are limited to the latest six years (2014–2019). To check the reliability of 30-yr deep-learningbased prediction, totally 68 peer-reviewed studies reporting *in situ* observations of ambient O3 are collected for enhanced model-observation comparison. The developed ambient O<sub>3</sub> database covers two metrics as i) monthly average of daily 24-h average, and ii) monthly average of daily maximum 8-h average. The metric, daily diurnal 7-h average, adopted in earlier literatures, are compared to daily maximum 8-h average as an alternative proxy. For prediction-observation comparisons on daily 1-h maximum metric, null-intercept linear conversion is applied to approximately project daily 8-h maximum average (DMA8h) concentrations onto daily 1-h maximum average (DMA1h) concentrations. The idea of null-intercept linear conversion was put forward by US EPA (Volume I, section 7.1.3.2)<sup>78</sup>, and the conversion coefficients have been updated by 30-yr historical observations archived in TOAR and CNEMC $^{14}$ . At multi-season or multi-year scale, the conversion follows: DMA1h = DMA8h × 1.213; in warm seasons (i.e. April to September), the conversion follows: DMA1h = DMA8h×1.202, where O<sub>3</sub> concentrations in DMA8h metric are obtained from Bayesian neural network downscaler. Observed and deep-learning-modelled ambient O<sub>3</sub> concentrations are both unified into ppb. IGAC (International Global Atmospheric Chemistry project) TOAR-II Working Group has doublechecked the external validation in August 2022, and recognised the credibility of the database for long-term population exposure tracking and risk assessment studies (https://igacproject.org/human-health-impacts-ozone-focus-working-group, accessed February 2023).







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