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36 Data collection

37 We collected station-observed particulate matter with diameters $\leq 2.5 \mu\text{m}$ (PM_{2.5}) from
38 5446 monitoring stations in 2000-2019. Information regarding the distribution of
39 included monitoring stations and the sources of data can be found in the previous study.¹
40 To ensure the comparability of health risks estimated from observed and estimated
41 PM_{2.5} sources, our study was limited to Multi-City Multi-Country (MCC) cities where
42 both station-observed PM_{2.5} and corresponding model-estimated values were available
43 during the study period. We disregarded the data from monitoring stations located
44 beyond the boundaries of the MCC cities or those whose data fell outside the recording
45 period. Finally, 1710 stations were involved in 347 MCC cities. We excluded the
46 extreme outliers above 99.9th quantiles of the collected PM_{2.5} monitoring dataset. In
47 addition, we gathered data on the daily mean temperature and daily mean relative
48 humidity in the 347 MCC cities from the EAR5 dataset (the fifth-generation European
49 Centre for Medium-Range Weather Forecasts Reanalysis).² For the grid-based model-
50 estimated PM_{2.5} exposure, comprehensive details regarding the validation of the global
51 daily PM_{2.5} concentrations can be found in our previous study¹.

52 Sensitivity analysis method

53 Several sensitivity analyses were conducted to test the robustness of the exposure-
54 response (E-R) relationship estimations. We examined potential non-linearity in the E-
55 R associations between short-term PM_{2.5} exposure and mortality from both observed
56 and estimated PM_{2.5} concentrations. Specifically, a B-spline function with two knots of
57 mean PM_{2.5} concentration was used for all 347 MCC cities, and the overall non-linear
58 E-R relationship curves were pooled and compared. A multivariate meta-regression
59 method was employed to examine the statistical differences in the E-R relationships
60 from observed and estimated daily PM_{2.5} data. Furthermore, we used different knots
61 and degree of freedom for temperature and PM_{2.5} concentrations, as well as extending
62 the maximum lag days from two to five days to examine the robustness of our
63 estimations. To examine the applicability of the model-estimated PM_{2.5} in health risk
64 assessment outside the 5446 monitoring stations, we examine the disparities in model-
65 estimated PM_{2.5} for mortality risk assessment by comparing them with another external
66 MCC monitor station data sources. Specifically, we selected MCC cities where the
67 monitoring stations were not included in the Deep Ensemble Machine Learning model
68 training. We then compared the disparities in E-R estimates derived from both model-
69 estimated exposure and station-observed PM_{2.5} data. Ultimately, our validation analysis
70 encompasses 81 MCC cities across 11 countries, with a cumulative total of over 4.4
71 million all-cause deaths. The details regarding the included MCC cities and countries
72 can be found in Table S6.

73 All statistical analyses were performed using R software (version 4.0.1). The Spearman
74 correlation analysis was conducted to test the correlation between monitoring station-
75 observed and model-estimated daily PM_{2.5} concentrations at city and country levels.

76 Meta-regression method

77 We used the multivariate meta-regression method to examine the statistical differences

78 in the E-R relationships from station-observed and model-estimated daily PM_{2.5} data.

79 Here, we outlined a procedure for conducting the meta-regression approach. Initially,

80 we computed the effect size (Relative Risk Increase, RRI) and its corresponding

81 standard error for each city. Subsequently, a binary indicator (x) was introduced, where

82 '1' signifies RRI based on station-observed PM_{2.5} data and '0' indicates those relying on

83 model-estimated data. The 'mixmeta' R function was employed to develop a meta-

84 regression model to examine if the effect size varies depending on whether PM_{2.5} data

85 is observed or estimated. Finally, by examining the coefficient of 'x' and its associated

86 *P*-value from the meta-regression output, we ascertained the statistical differences in

87 the E-R relationships from observed and estimated daily PM_{2.5} exposure. We provided

88 an example data (see Table S1) and the R code to illustrate how to conduct meta-

89 regression model to examine the statistical differences in the E-R relationships from

90 station-observed and model-estimated daily PM_{2.5} data.

91

92 **Table S1.** Example data for meta regression

City	ISO3	coef	vcov	type
Winston	USA	0.025689	2.89E-05	model
Winston	USA	0.012943	1.82E-05	station
Worcester	USA	0.029642	1.53E-05	model
Worcester	USA	0.020381	1.03E-05	station
Warrington	GBR	0.002961	3.29E-05	model
Warrington	GBR	0.013875	2.36E-05	station

WDC	USA	0.034593	1.18E-05	model
WDC	USA	0.02491	8.39E-06	station
Washington	USA	0.003676	3.50E-05	model
Washington	USA	0.006518	2.11E-05	station
West Midlands	GBR	-0.00591	1.89E-06	model
West Midlands	GBR	-0.00476	1.55E-06	station
West Yorkshire	GBR	-0.00016	2.90E-06	model
West Yorkshire	GBR	-0.0006	2.15E-06	station
Youngstown	USA	0.010351	1.57E-05	model
Youngstown	USA	0.005055	1.06E-05	station
Yorktown	GBR	-0.01519	4.00E-05	model
Yorktown	GBR	-0.00467	3.03E-05	station
york	USA	0.004386	1.86E-05	model
york	USA	-0.01022	9.18E-06	station

93 Notes: The coef and vcov are the coefficient and variance-covariance matrix for each
94 city obtained by the first stage city-specific daily fine particulate matter (PM_{2.5}) E-R
95 associations using a quasi-Poisson regression.
96

97 R code for meta-regression

98 `model_test_example <- mixmeta(coef~ 1+type, vcov, data=df, method="ml")`

99 `p <- summary(model_test_example)$coefficients[,4][2]`

100

101 Study strengths

102 This study has several strengths. To the best of our knowledge, our study is the first to
103 estimate the mortality effect of short-term PM_{2.5} exposure from both station-observed
104 and model-estimated daily PM_{2.5} observations from 347 cities worldwide. We used a
105 large dataset of over 15.8 million deaths over a long period of approximately 20 years
106 to estimate the mortality associations with PM_{2.5} short-term daily exposure. Moreover,
107 we examined the global daily PM_{2.5} model estimations with a high spatial resolution of
108 0.1°×0.1° to evaluate the association between mortality risks and short-term PM_{2.5}
109 exposure. The E-R estimates using model-estimated data were consistent with that from
110 station-observed data. These comparative analyses offer compelling evidence for the
111 use of the estimated global PM_{2.5} data in future epidemiological health risk assessments.
112 Furthermore, we employed external MCC monitoring stations to further validate our
113 model-estimated exposure-mortality risk assessment, yielding robust results. It should
114 be noted that utilizing model-estimated PM_{2.5} in conjunction with data from more
115 monitoring stations globally could potentially lead to even more unbiased estimates,
116 making it a promising area for future exploration.

117

118 Supplementary results

119

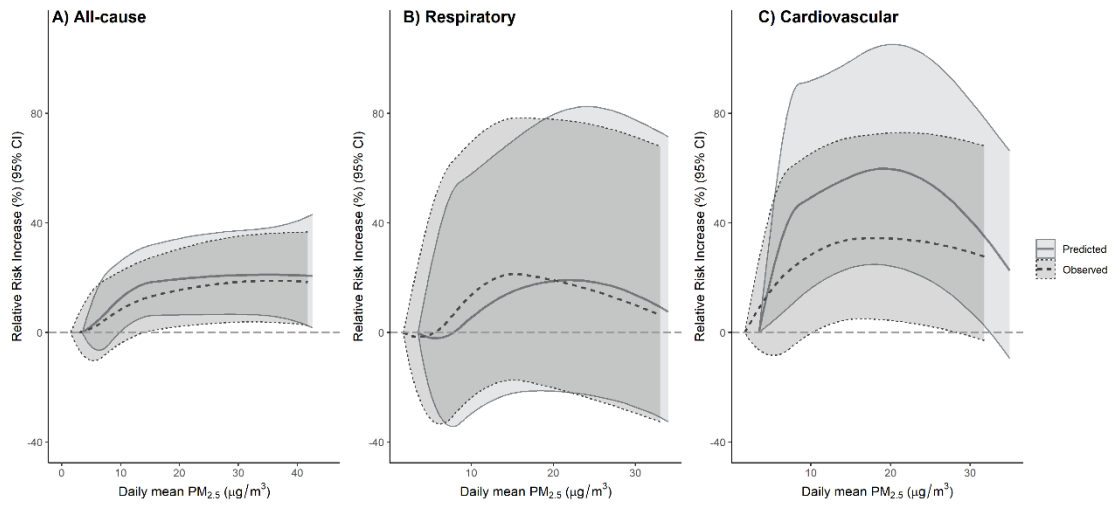


120

121 **Figure S1.** The correlation between station-observed and model-estimated daily PM_{2.5}
122 in the study period in 2000-2019

123 Notes: PM_{2.5}: particulate matter with a diameter of < 2.5 μm; Correlation coefficient
124 was based on Spearman correlation analysis.

125



126

127 **Figure S2.** The non-linear exposure-response relationships of station-observed and
 128 model-predicted PM_{2.5} with all-cause, respiratory, and cardiovascular mortality

129 Notes: PM_{2.5}: particulate matter with a diameter of < 2.5 µm.

130

131 **Table S2.** The summary of the included cities and the consistency between station-
 132 observed and model-estimated daily fine particulate matter (PM_{2.5}) in the study period

Country & region	No. of cities	Start date	End date	Study period (year)	R ²	RMSE
Australia	40	1/01/2009	31/12/2017	9	0.53	3.26
China	3	27/11/2013	31/12/2015	2.1	0.98	6.74
Czechia	4	1/01/2004	31/12/2015	12	0.76	9.98
Finland	1	1/01/2001	31/12/2014	14	0.74	2.80
France	19	1/01/2011	31/12/2015	5	0.82	4.94
Germany	8	1/01/2003	31/12/2015	13	0.85	4.39
Italy	3	1/01/2013	31/12/2015	2.7	0.64	8.70
Netherlands	5	1/01/2013	31/12/2016	4	0.96	1.86
Norway	1	1/01/2003	31/12/2018	16	0.79	2.80
Portugal	2	9/03/2002	31/12/2018	16.8	0.66	7.40
Spain	16	1/01/2004	31/12/2014	11	0.64	4.63
Sweden	3	8/01/2002	31/12/2016	15	0.74	3.32
Taiwan	6	21/12/2016	31/12/2018	2	0.84	4.88
United Kingdom	30	12/01/2000	31/12/2016	17	0.82	3.88
USA	206	1/01/2000	31/12/2006	7	0.79	3.89
Total	347	1/01/2000	31/12/2018	8.3	0.82	4.31

133 Notes: R²: coefficients of determination; RMSE: Root Mean Square Error.

134

135 **Table S3.** The comparison of pooled RRI (%) for all-cause, respiratory, and cardiovascular mortality associated with a 10 µg/m³ increase in
 136 station-observed and model-estimated daily mean fine particulate matter (PM_{2.5}) by country

Country & Region	All-cause		Respiratory		Cardiovascular	
	Observed PM _{2.5}	Estimated PM _{2.5}	Observed PM _{2.5}	Estimated PM _{2.5}	Observed PM _{2.5}	Estimated PM _{2.5}
Australia	1.17 (-1.36, 3.77)	-0.72 (-6.27, 5.15)	4.28 (-4.66, 14.05)	3.10 (-18.50, 30.44)	3.31 (-3.55, 10.65)	4.22 (-7.37, 17.26)
China	0.39 (0.04, 0.74)	0.42 (0.06, 0.78)	0.61 (-0.26, 1.49)	0.71 (-0.30, 1.73)	0.23 (-0.09, 0.56)	0.25 (-0.10, 0.59)
Czechia	0.18 (-0.01, 0.37)	-0.02 (-0.21, 0.18)	0.51 (-0.20, 1.23)	0.53 (-1.04, 2.13)	-0.08 (-0.38, 0.22)	-0.42 (-0.85, 0.01)
Finland	0.98 (0.48, 1.48)	0.93 (0.37, 1.50)	1.33 (-0.79, 3.51)	1.40 (-1.04, 3.89)	0.89 (0.08, 1.70)	0.35 (-0.57, 1.27)
France	0.59 (-0.07, 1.25)	0.45 (-0.38, 1.29)	0.86 (-0.54, 2.27)	0.68 (-1.23, 2.63)	NA	NA
Germany	0.22 (-0.05, 0.48)	0.24 (0.06, 0.43)	NA	NA	NA	NA
Italy	2.07 (-0.38, 4.58)	1.59 (0.37, 2.83)	NA	NA	NA	NA
Netherlands	-0.08 (-0.81, 0.65)	-0.32 (-1.23, 0.60)	NA	NA	NA	NA
Norway	-1.06 (-1.61, -0.52)	-1.16 (-1.88, -0.43)	-1.57 (-3.35, 0.24)	-3.90 (-6.36, -1.38)	-0.78 (-1.79, 0.25)	-1.05 (-2.50, 0.42)
Portugal	0.11 (-0.07, 0.30)	0.53 (0.23, 0.83)	-0.15 (-0.83, 0.54)	-0.59 (-1.09, -0.08)	0.06 (-0.13, 0.24)	0.26 (-0.03, 0.55)
Spain	-0.97 (-2.55, 0.64)	-0.48 (-1.84, 0.89)	-0.19 (-4.26, 4.05)	-0.94 (-4.97, 3.25)	-2.29 (-4.94, 0.43)	-0.84 (-3.59, 1.99)
Sweden	-0.05 (-1.27, 1.19)	0.42 (0.02, 0.83)	-0.22 (-4.56, 4.33)	-1.23 (-5.10, 2.80)	0.45 (-0.23, 1.13)	0.49 (-0.18, 1.16)
Taiwan	0.11 (-0.75, 0.99)	-0.27 (-1.50, 0.98)	0.10 (-3.19, 3.51)	0.38 (-3.57, 4.50)	-1.39 (-2.99, 0.24)	-1.68 (-4.08, 0.78)
United Kingdom	0.49 (0.12, 0.86)	0.57 (0.07, 1.07)	0.47 (-0.63, 1.59)	0.51 (-0.91, 1.96)	0.75 (-0.02, 1.53)	0.78 (-0.21, 1.77)
USA	0.86 (0.68, 1.04)	1.17 (0.92, 1.43)	0.68 (-0.03, 1.40)	1.08 (0.11, 2.06)	0.56 (0.23, 0.89)	0.89 (0.45, 1.33)

137 Notes: PM_{2.5}: particulate matter with a diameter of < 2.5 µm; RRI: relative risk increase; Values in brackets are 95% confidence interval.

138

139 **Table S4.** The pooled RRIs (%) for all-cause mortality associated with a 10 $\mu\text{g}/\text{m}^3$
 140 increase in station-observed and model-predicted daily mean fine particulate matter
 141 ($\text{PM}_{2.5}$) with different degree of freedom for temperature and relative humidity

df	$\text{PM}_{2.5}$ sources	RRI (%)	95% CI
3	Model-predicted	0.86	0.65 to 1.07
3	Station-observed	0.67	0.51 to 0.83
4	Model-predicted	0.87	0.66 to 1.08
4	Station-observed	0.68	0.52 to 0.83
5	Model-predicted	0.86	0.66 to 1.07
5	Station-observed	0.68	0.52 to 0.83

142 Notes: $\text{PM}_{2.5}$: particulate matter with a diameter of $< 2.5 \mu\text{m}$; df: degree of freedom;
 143 RRI: relative risk increase; 95% CI: 95% confidence interval.

144

145 **Table S5.** The pooled RRIs (%) for all-cause mortality associated with a 10 $\mu\text{g}/\text{m}^3$
 146 increase in station-observed and model-predicted daily mean fine particulate matter
 147 ($\text{PM}_{2.5}$) with different sets of knots for spline functions for temperature and relative
 148 humidity

Knot	$\text{PM}_{2.5}$ sources	RRI (%)	95% CI
Type1	Model-predicted	0.86	0.65 to 1.07
Type1	Station-observed	0.67	0.51 to 0.82
Type2	Model-predicted	0.86	0.66 to 1.07
Type2	Station-observed	0.67	0.52 to 0.83
Type3	Model-predicted	0.87	0.66 to 1.08
Type3	Station-observed	0.67	0.52 to 0.83

149 Notes: $\text{PM}_{2.5}$: particulate matter with a diameter of $< 2.5 \mu\text{m}$; Type1: setting knot of
 150 the 25th, 50th, and 75th percentiles of temperature for its spline function; Type2:
 151 setting knot of the 10th and 90th percentiles of temperature for its spline function;
 152 Type3: setting knot of the 25th and 75th percentiles of temperature for its spline
 153 function; RRI: relative risk increase; 95% CI: 95% confidence interval.

154

155 **Table S6.** The pooled RRIs (%) for all-cause mortality associated with a 10 $\mu\text{g}/\text{m}^3$
 156 increase in station-observed and model-predicted daily mean fine particulate matter
 157 ($\text{PM}_{2.5}$) by using different lag days

Lag days	$\text{PM}_{2.5}$ sources	RRI (%)	95% CI
2	Station-observed	0.02	-0.20 to 0.24
2	Model-predicted	0.02	-0.25 to 0.30
3	Station-observed	-0.25	-0.46 to -0.04
3	Model-predicted	-0.38	-0.66 to -0.09
4	Station-observed	-0.15	-0.34 to 0.04
4	Model-predicted	-0.28	-0.53 to -0.04
5	Station-observed	-0.14	-0.33 to 0.06
5	Model-predicted	-0.27	-0.54 to 0.00

158 Notes: $\text{PM}_{2.5}$: particulate matter with a diameter of $< 2.5 \mu\text{m}$; RRI: relative risk
 159 increase; 95% CI: 95% confidence interval.

160

161 **Table S7.** The pooled RRIs (%) for all-cause mortality associated with a 10 µg/m³ increase in station-observed and model-predicted daily mean
 162 fine particulate matter (PM_{2.5}) using cities with monitoring stations not included in model training

Country	No. of cities	Mortality counts in 2000-2018			RRI (%) for all-cause deaths	
		Start date	End date	All-cause	Observed PM _{2.5}	Estimated PM _{2.5}
Canada	19	1/01/2000	31/12/2015	1282938	1.21 (0.65 to 1.78)	0.28 (0.04 to 0.52)
Ecuador	1	1/01/2014	31/12/2018	41204	1.43 (0.72 to 2.15)	0.16 (-2.89 to 3.30)
Estonia	3	19/08/2008	31/12/2018	25376	1.87 (1.08 to 2.66)	0.29 (-3.38 to 4.10)
Greece	1	1/01/2007	31/12/2010	115466	1.12 (0.88 to 1.36)	0.66 (0.44 to 0.89)
Iran	2	13/04/2013	31/12/2015	84894	0.11 (-0.07 to 0.29)	-0.64 (-1.05 to -0.22)
Japan	38	1/01/2011	31/12/2015	1324405	1.00 (0.60 to 1.39)	0.60 (0.25 to 0.96)
Mexico	4	7/05/2003	31/12/2012	1016016	1.42 (0.96 to 1.89)	0.70 (0.04 to 1.37)
Portugal	2	1/01/2004	31/12/2018	57483	-0.86 (-1.93 to 0.22)	0.19 (-2.86 to 3.34)
South Africa	5	31/07/2004	31/12/2013	404505	0.56 (0.28 to 0.83)	1.18 (0.44 to 1.92)
United Kingdom	2	5/11/2008	31/12/2016	58931	-0.33 (-0.67 to 0.02)	-0.08 (-0.82 to 0.67)
USA	4	1/01/2000	31/12/2006	21959	1.50 (-0.18 to 3.21)	-0.56 (-3.12 to 2.06)
Total	81	1/01/2000	31/12/2018	4433177	0.98 (0.72 to 1.25)	0.46 (0.19 to 0.73)

163 Notes: PM_{2.5}: particulate matter with a diameter of < 2.5 µm; RRI: relative risk increase; Values in brackets are 95% confidence interval.

164

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