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Supplementary appendix 1

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Supplementary Appendix

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1. Methods

1.1 Estimation of wildfire-related O_3

Daily concentration of wildfire-related O_3 was firstly estimated globally using the GEOS-Chem at a spatial resolution of 2° × 2·5°, and then was adjusted and downscaled for study areas at a spatial resolution of 0·25° × 0·25° using ground-level measurements of O_3 and other predictors.

GEOS-Chem Model development

In the 3-D GEOS-Chem model, biomass burning emissions from Global Fire Emissions Database version 4·1 **(**GFED v4·1) was used,[1-4] which considers six land cover types: temperate forests, peat, savanna, deforestation, boreal forest, and agricultural waste (https://daac.ornl.gov/VEGETATION/guides/fire_emissions_v4_R1.html). For each land type, fire-induced emissions are estimated as the product of dry matter and species-specific emission factors.[5] By default, GFED v4·1 provides monthly fire emissions. From the year 2003, daily fire emissions become available by multiplying daily scale factors onto the monthly data.[6] Following the same protocols as previous studies,[7] daily enhancement of O_3 by fires in 2000-2016 was estimated as the differences between simulations with and without fire emissions.

Adjustment and downscaling of estimated wildfire-related O_3

We have improved the exposure assessment for wildfire-related O_3 by further adjustment and downscaling method, as following steps:

Step 1: The all-source O_3 were further adjusted using a random forest (RF) model and data on ground measured O_3 and other spatial and temporal predictors (e.g., meteorological data). Particularly, global meteorological data on temperature, precipitation, barometric pressure, and wind speed have a spatial resolution of 0·25° (≈28km). The following random forest model was developed using ground measured O_3 data from 6,882 sites in 61 countries (where daily O_3 was monitored, Figure S1), GEOS-Chem derived all-source O_3, and meteorological data (0·25° spatial resolution).

$$O_{3\_station} = f\,(O_{3\_chem\_total}, T_{mean}, T_{max}, T_{min}, TV, RH, Wind\_u, Wind\_v, Precip, Pressure, UV, Year, Month, DOW, DOY, Lon, Lat)$$

where $O_{3\_station}$ is the daily ground measured O_3 at a monitoring station. $O_{3\_chem\_total}$ is the estimated all-source O_3 by GEOS-Chem. T_{mean}, T_{min} and T_{max} *are* daily mean/minimum/maximum ambient temperature. TV is the standard deviations of 24 hourly temperatures. Other meteorological variables includes daily mean eastward component of 10m wind (Wind_u), daily mean northward component of 10m wind (Wind_v), daily total precipitation (Precip), daily mean surface air pressure (Pressure), and daily mean downward ultraviolet radiation at the surface (UV). *DOW* and *DOY* are day of the week and the year. *Lon* and *Lat* are longitude and latitude. The predictive ability of this daily RF model was examined using a 10-fold cross-validation (CV) method. It showed that the CV R^2 and Root Mean Square Error (RMSE) were 80% and 19·6 μg/m^3 (Figure S2). The all-source O_3 was predicted for all grid cells of a 0·25°grid across the world using above random forest model and predictors.

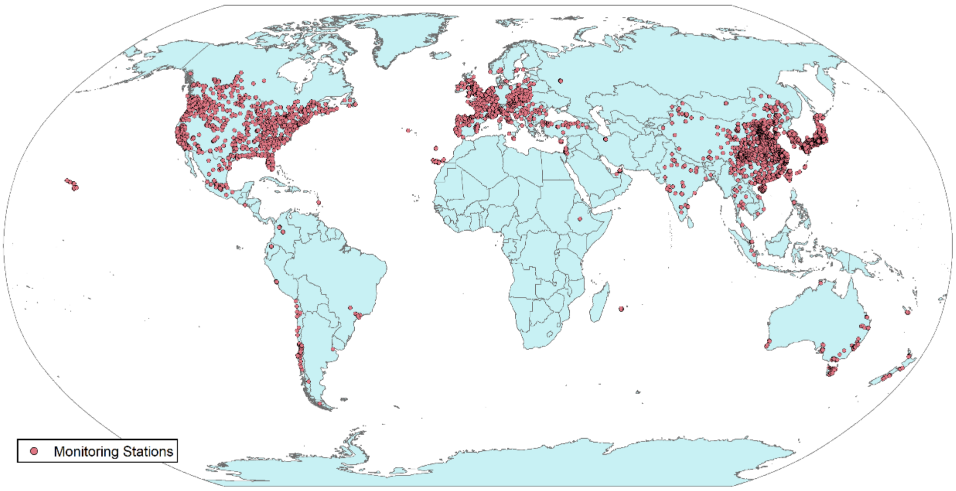

Figure S1. Locations of 6,882 ground monitoring sites used for data validation and adjustment.

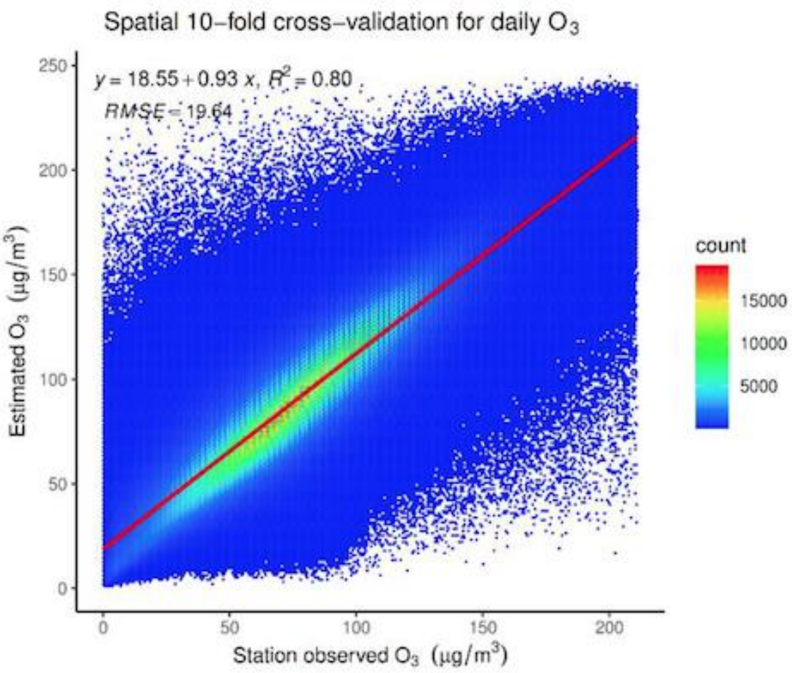

Figure S2. Density scatterplot of 10-fold cross-validation for the daily random forest model.

Note: RMSE, root mean squared prediction error ($\mu g/m^3$)

Step 2: The daily all-source O_3 and wildfire-related O_3 derived from GEOS-Chem were downscaled to a global 0·25°-grid on each day, using the inverse distance weighted (IDW) method. Then, the ratio of downscaled wildfire-related O_3 to all-source O_3 derived from GEOS-Chem was calculated for each grid cell on each day.

Step 3: The adjusted wildfire-related O_3 in each grid cell was derived by multiplying adjusted all-source O_3 (from step 1) with the ratio of GEOS-Chem derived wildfire-related to all-source O_3 (from step 2). The level of estimated wildfire-related O_3 in each city was assigned as the average of all the cell values fell in that city, which was used in the final analyses. The high-resolution data on wildfire-related O_3 showed a CV R^2 of 80% and had a high spatial resolution of 0·25° (≈28km).

Table S1. Global and continent-specific results for 10-fold cross-validation of all-source O_3 estimation

Continents	Stations (n)	Observations (n)	CV R^2	RMSE
Africa	165	315284	0.43	25.29
Asia	1831	2865348	0.77	23.35
Europe	2393	10043651	0.79	18.71
Northern America	2247	7459422	0.82	19.20
Oceania	98	263885	0.70	14.19
South America	117	150245	0.63	20.14
Overall	6851	21097835	0.80	19.64

CV: Cross-validation; RMSE: Root mean squared error.

1.2 Calculation of excess mortality fraction

Excess mortality fraction in each city was calculated using the following formulas:

$$AN_i = n * B\_AF_i$$

$$B\_AF_i = 1 - \exp\left(-\sum_{l=0}^{L} \beta_{i-l}\right)$$

where: ‘i’ is the day when deaths occur; AN_i is the number of deaths attributable to acute wildfire-related O_3 on day ‘i’; n is the reported number of deaths; B_AF_i is the attributable fraction due to cumulative effects of O_3 on day ‘$i - l$’, with backward approach; ‘l’ is the lag time; L is the maximum lag time; β_{i-l} is the pooled effect estimate associated with level of wildfire-related O_3 on day ‘$i - l$’. In addition, the upper value and lower value of 95%CI of pooled effect estimates were used to calculate the 95%CI of excess mortality fraction using the above formulas. We chose to estimate excess annual deaths using the global pooled risk estimate rather than location-specific BLUP-based estimates due to several reasons. Firstly, employing the global pooled risk estimate offers a comprehensive overview that encompasses a wider spectrum of data, thereby not only ensuring a broader representation but also enhancing the credibility and validity of the obtained results. Secondly, it allows for better comparability across different geographical regions, facilitating a more standardized analysis. Annual excess deaths for all-cause, cardiovascular, and respiratory causes were first calculated for each location separately, considering both the current-day and lag effects of wildfire-related O_3. Then, the sum of reported deaths in all locations was divided by the sum of excess deaths to derive the excess mortality fraction at the country, regional, and global levels.

Table S2. The quasi-likelihood version of the Akaike information criterion for different lag times of exposure to wildfire-related O_3

Lag (days)	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	qAIC	qAIC	qAIC
1	-2808	-1609	-1260
2	-2817	-1652	-1433
3	-2794	-1577	-1339
5	-2671	-1419	-1178
7	-2433	-1326	-1077
10	-2086	-1147	-904

Table S3. The quasi-likelihood version of the Akaike information criterion for degrees of freedom for natural cubic splines of time

Degrees of freedom/year	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	qAIC	qAIC	qAIC
6	-2794	-1564	-1070
7	-2823	-1716	-1262
8	-2870	-1727	-1368
9	2812	-1616	-991

Table S4. The quasi-likelihood version of the Akaike information criterion for different lag times of meteorological variables in the model

Lag (days)	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	qAIC	qAIC	qAIC
1	-2798	-1564	-1368
2	-2802	-1578	-1385
3	-2793	-1549	-1357
5	-2794	-1577	-1339
7	-2798	-1600	-1314
10	-2758	-1637	-1264

2. Results

Table S5. A summary of study areas, periods, and number of deaths in 43 countries/regions included in this study.

Country/region	UN Regions	Study period	Total Mortality		Cardiovascular mortality		Respiratory mortality	
			City (n)	Deaths (n)	City (n)	Deaths (n)	City (n)	Deaths (n)
Argentina	South America	2005-2015	3	686,333	NA	NA	NA	NA
Australia	Australia	2000-2009	3	513,527	NA	NA	NA	NA
Brazil	South America	2000-2011	18	2,778,330	NA	NA	NA	NA
Canada	Northern America	2000-2015	26	2,116,195	26	642,418	26	177,751
Chile	South America	2004-2014	4	325,462	NA	NA	NA	NA
China	Eastern Asia	2000-2015	15	1,081,700	14	433,839	14	140,017
Colombia	South America	2000-2013	5	843,633	5	237,346	5	88,819
Costa Rica	South America	2000-2016	1	29,120	1	8,783	1	2,467
Czech Republic	Central Europe	2000-2015	4	505,932	4	246,331	4	29,860
Ecuador	South America	2014-2016	2	64,351	2	18,473	2	7,574
Estonia	North Europe	2000-2015	5	127,135	NA	NA	NA	NA
Finland	North Europe	2000-2014	1	110,385	1	39,840	1	6,152
France	Central Europe	2000-2014	18	1,639,262	NA	NA	18	101,731
Germany	Central Europe	2000-2015	12	2,120,825	NA	NA	NA	NA
Greece	South Europe	2001-2010	1	287,969	1	136,194	1	28,771
Guatemala	Central America	2009-2016	1	62,715	NA	NA	NA	NA
Iran	Western Asia	2004-2013	1	121,585	1	40,704	1	6,745
Ireland	North Europe	2000-2007	6	333,088	6	91,232	6	50,077
Italy	South Europe	2006-2015	18	804,278	NA	NA	NA	NA
Japan	Eastern Asia	2000-2015	47	18,008,670	47	5,233,495	47	2,750,685
Kuwait	Western Asia	2000-2016	1	73,748	1	35,285	1	5,715
Mexico	Central America	2000-2014	10	2,682,202	10	691,353	10	253,922

(continued)

Table S5. A summary of study areas, periods, and number of deaths in 43 countries/regions included in this study (continued).

Country/ region	UN Regions	Study period	Total Mortality		Cardiovascular mortality		Respiratory mortality	
			City (n)	Deaths (n)	City (n)	Deaths (n)	City (n)	Deaths (n)
Moldova	Central Europe	2001-2010	4	59,906	NA	NA	NA	NA
Netherlands	North Europe	2000-2016	5	338,448	NA	NA	NA	NA
Norway	North Europe	2000-2016	1	76,577	1	24,518	1	7,548
Panama	Central America	2013-2016	1	11,457	1	3,862	1	971
Paraguay	South America	2004-2016	1	39,713	1	12,791	1	3,544
Peru	South America	2008-2014	18	633,137	NA	NA	NA	NA
Philippines	Southeastern Asia	2006-2010	4	274,516	4	87,401	4	31,190
Portugal	South Europe	2000-2016	5	779,638	5	250,047	5	88,472
Puerto Rico	Central America	2009-2016	1	26,564	NA	NA	NA	NA
Romania	Central Europe	2000-2016	8	697,505	NA	NA	NA	NA
South Africa	Southern Africa	2000-2013	52	7,443,918	52	1,110,304	52	943,671
South Korea	Eastern Asia	2000-2016	36	2,362,545	36	542,146	36	166,046
Spain	South Europe	2000-2014	52	1,859,279	52	600,992	52	221,208
Sweden	North Europe	2000-2016	3	452,463	3	181,068	3	32,440
Switzerland	Central Europe	2000-2013	8	173,519	8	62,428	8	11,201
Taiwan	Southeastern Asia	2000-2014	3	907,141	3	199,305	3	93,464
Thailand	Southeastern Asia	2000-2008	62	1,666,292	62	299,721	62	205,900
UK	North Europe	2000-2016	70	3,642,897	70	1,183,375	70	528,080
USA	Northern America	2000-2006	210	8,594,149	210	2,672,728	210	849,506
Uruguay	South America	2012-2016	1	153,554	NA	NA	NA	NA
Vietnam	Southeastern Asia	2009-2013	2	108,173	2	24,433	2	8,970
In total	-	2000-2016	749	65,617,836	629	15,110,412	647	6,842,497

*160 out of 749 with non-external cause mortality data

Table S6. A summary of maximum daily levels (μg/m³) of wildfire-related ozone in study locations by hemisphere.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Locations in the Northern Hemisphere (n=647)												
Mean	3·17	3·17	3·46	3·27	3·37	2·52	2·61	2·61	2·52	1·90	1·15	1·93
Median	0·78	0·72	1·44	1·29	2·59	2·48	2·26	2·31	2·31	1·60	1·01	0·75
IQR	0·84	0·50	0·82	4·09	1·94	1·15	2·03	1·10	1·12	0·48	0·40	0·36
Locations in the Southern Hemisphere (n=102)												
Mean	4·54	4·13	3·12	2·91	4·19	5·81	8·50	17·56	19·34	13·60	6·45	3·67
Median	3·29	2·88	2·46	2·56	4·15	5·16	8·04	17·87	19·87	12·69	5·61	2·55
IQR	3·82	2·71	1·53	1·02	3·58	4·90	4·33	7·72	10·81	8·42	4·57	1·59

IQR: Interquartile range.

Table S7. Excess mortality fraction of annual mortality due to exposure to wildfire-related O_3 during lag 0-2 days by different subregions and GDP levels

Region/GDP	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	%, 95%CI	%, 95%CI	%, 95%CI
Subregion			
Australia	0·80 (0·43, 1·16)	NA	NA
Central America	1·02 (0·55, 1·48)	0·29 (-0·07, 0·64)	0·47 (0·10, 0·84)
Central Europe	0·39 (0·21, 0·57)	0·39 (-0·09, 0·87)	0·58 (0·12, 1·03)
Eastern Asia	0·32 (0·17, 0·47)	0·41 (-0·10, 0·90)	0·73 (0·16, 1·29)
Western Asia	0·40 (0·22, 0·59)	0·25 (-0·06, 0·55)	0·55 (0·12, 0·99)
Northern America	0·35 (0·19, 0·51)	0·34 (-0·08, 0·75)	0·70 (0·15, 1·24)
North Europe	0·37 (0·20, 0·55)	0·34 (-0·08, 0·75)	0·53 (0·11, 0·94)
Southern Africa	1·22 (0·66, 1·78)	0·79 (-0·19, 1·72)	1·83 (0·40, 3·19)
South America	1·21 (0·65, 1·76)	0·69 (-0·16, 1·52)	1·18 (0·25, 2·08)
South Europe	0·32 (0·17, 0·46)	0·58 (-0·14, 1·29)	1·32 (0·28, 2·34)
Southeastern Asia	0·98 (0·53, 1·41)	0·42 (-0·10, 0·94)	0·58 (0·12, 1·03)
GDP level (US$)			
<10000	1·10 (0·60, 1·60)	0·66 (-0·16, 1·46)	1·43 (0·31, 2·50)
10000-19999	0·36 (0·19, 0·52)	0·44 (-0·11, 0·98)	0·99 (0·21, 1·75)
20000-29999	0·42 (0·22, 0·61)	0·35 (-0·08, 0·79)	0·52 (0·11, 0·94)
≥30000	0·33 (0·18, 0·49)	0·32 (-0·08, 0·72)	0·60 (0·13, 1·07)

Table S8. Excess annual deaths due exposure to wildfire-related O_3 during lag 0-2 days in 43 countries/regions

Country/Region	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	n, 95%CI)	n, 95%CI	n, 95%CI
Argentina	460 (248, 672)	NA	NA
Australia	436 (235, 634)	NA	NA
Brazil	2568 (1386, 3737)	NA	NA
Canada	399 (214, 584)	98 (-23, 219)	49 (10, 87)
Chile	207 (111, 302)	NA	NA
China	922 (495, 1346)	725 (-173, 1591)	549 (119, 956)
Colombia	1244 (674, 1802)	148 (-35, 326)	94 (20, 166)
Costa Rica	12 (6, 17)	2 (0, 3)	1 (0, 1)
Czech Republic	126 (68, 184)	40 (-9, 89)	9 (2, 16)
Ecuador	479 (260, 693)	18 (-4, 39)	13 (3, 24)
Estonia	29 (16, 43)	NA	NA
Finland	27 (14, 39)	8 (-2, 19)	2 (1, 4)
France	396 (213, 580)	NA	38 (8, 67)
Germany	546 (293, 798)	NA	NA
Greece	120 (65, 176)	47 (-11, 104)	18 (4, 31)
Guatemala	98 (53, 142)	NA	NA
Iran	52 (28, 75)	10 (-2, 23)	4 (1, 7)
Ireland	132 (71, 194)	36 (-8, 80)	40 (8, 71)

(continued)

Table S8. Excess annual deaths due exposure to wildfire-related O_3 during lag 0-2 days in 43 countries/regions (continued)

Country/Region	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	n, 95%CI)	n, 95%CI	n, 95%CI
Italy	244 (131, 357)	NA	NA
Japan	3289 (1766, 4807)	973 (-229, 2165)	915 (195, 1624)
Kuwait	15 (8, 22)	5 (-1, 11)	2 (0, 3)
Mexico	1834 (990, 2665)	133 (-31, 295)	80 (17, 142)
Moldova	27 (15, 40)	NA	NA
Netherland	89 (48, 130)	NA	NA
Norway	15 (8, 22)	4 (-1, 8)	2 (1, 4)
Panama	20 (11, 29)	3 (-1, 8)	1 (0, 2)
Paraguay	66 (36, 95)	3 (-1, 6)	1 (0, 3)
Peru	1401 (760, 2029)	NA	NA
Philippines	244 (131, 356)	50 (-12, 111)	31 (7, 55)
Portugal	138 (74, 201)	40 (-9, 90)	56 (12, 99)
Puerto Rico	9 (5, 14)	NA	NA
Romania	166 (89, 243)	NA	NA
South Africa	6508 (3513, 9465)	624 (-150, 1367)	1235 (268, 2151)
South Korea	502 (269, 733)	79 (-19, 175)	48 (10, 86)
Spain	397 (213, 581)	313 (-74, 689)	228 (49, 403)
Sweden	95 (51, 139)	88 (-21, 194)	10 (2, 18)
Switzerland	44 (24, 64)	38 (-9, 83)	8 (2, 15)
Taiwan	163 (88, 239)	42 (-10, 93)	34 (7, 61)
Thailand	2588 (1405, 3743)	183 (-43, 405)	142 (30, 254)
UK	814 (437, 1190)	189 (-44, 421)	157 (33, 279)
USA	4324 (2323, 6320)	1330 (-314, 2953)	880 (188, 1559)
Uruguay	162 (87, 237)	NA	NA
Vietnam	199 (107, 289)	23 (-5, 51)	10 (2, 18)

Note: Attributable numbers of deaths were calculated using the pooled global-level risk estimates.

Table S9. Excess annual deaths due to exposure to wildfire-related O_3 during lag 0-2 days by different Subregions and GDP levels

Region/GDP	All-cause mortality	Cardiovascular mortality	Respiratory mortality
	AN (n, 95%CI)	AN (n, 95%CI)	AN (n, 95%CI)
Subregion			
Australia	436 (235, 634)	NA	NA
Central America	1960 (1059, 2849)	136 (-32, 303)	81 (17, 145)
Central Europe	1306 (701, 1909)	78 (-18, 172)	55 (12, 97)
Eastern Asia	4712 (2531, 6886)	1776 (-421, 3931)	1511 (324, 2666)
Western Asia	67 (36, 97)	15 (-4, 34)	6 (1, 10)
Northern America	4724 (2537, 6904)	1428 (-337, 3172)	929 (198, 1645)
North Europe	1202 (645, 1757)	325 (-77, 723)	212 (45, 377)
Southern Africa	6508 (3513, 9465)	624 (-150, 1367)	1235 (268, 2151)
South America	6599 (3568, 9583)	170 (-41, 375)	110 (24, 193)
South Europe	899 (483, 1314)	400 (-95, 883)	302 (65, 533)
Southeastern Asia	3193 (1730, 4627)	297 (-70, 659)	217 (46, 388)
GDP level (US$)			
<10000	18866 (10195, 27408)	1917 (-458, 4217)	2160 (467, 3777)
10000-19999	1744 (937, 2549)	516 (-122, 1143)	377 (80, 666)
20000-29999	1183 (636, 1726)	269 (-63, 598)	116 (25, 207)
≥30000	9813 (5270, 14343)	2546 (-601, 5661)	2004 (427, 3555)

Note: Excess deaths were calculated using the pooled global-level risk estimates.

Table S10. *P* values for two-sample tests comparing effects estimates of main models with alternative models.

Models	All-causes mortality	Cardiovascular mortality	Respiratory mortality
Main model + fire_$PM_{2.5}$	0.61	0.53	0.94
Main model + other_O_3	0.11	0.99	0.95
Main model + all_$PM_{2.5}$	1.00	0.98	0.80
Main model + all_$PM_{2.5}$ + other_O_3	0.14	0.66	0.87

*Other pollutants were fitted similarly to wildfire-related O3, indicating their mean concentrations during the lag 0-2 days.

Table S11. Pooled percentage change in mortality associated with per 1 μg/m^3 increase in wildfire-related O_3 during lag 0-2 days by using different degrees of freedom for meteorological variables.

Df	All-cause mortality		Cardiovascular mortality		Respiratory mortality	
	Pooled estimates (%, 95%)	P for difference*	Pooled estimates (%, 95%)	P for difference	Pooled estimates (%, 95%)	P for difference*
3	0·55 (0·29, 0·80)	-	0·44 (-0·10, 0·99)	-	0·82 (0·18, 1·47)	-
4	0·51 (0·26, 0·76)	0.84	0·39 (-0·16, 0·95)	0.90	0·86 (0·21, 1·52)	0.93
5	0·51 (0·26, 0·76)	0.84	0·40 (-0·14, 0·96)	0.92	0·83 (0·17, 1·49)	0.99
6	0·51 (0·26, 0·76)	0.83	0·41 (-0·14, 0·96)	0.93	0·83 (0·17, 1·49)	0.99

**P* value for difference was calculated using a 2-sample test. The models used for sensitivity analyses were identical to the main model, except for the degrees of freedom for ambient temperature.

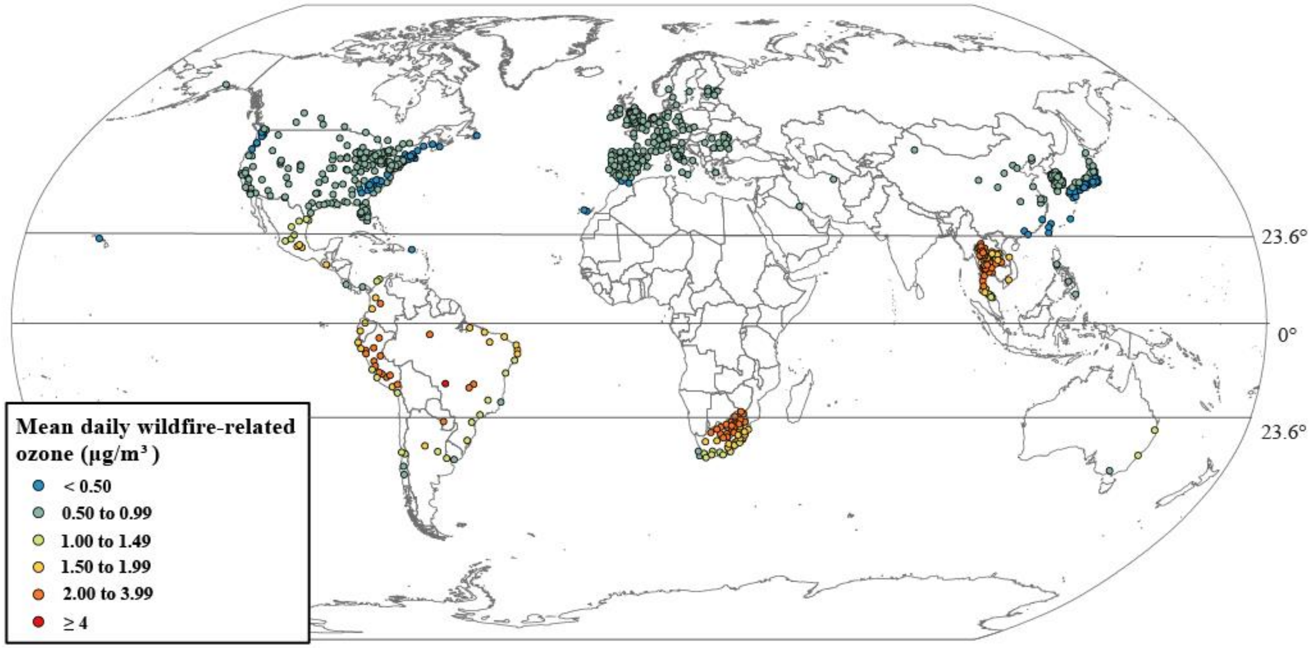

Figure S3. Mean levels of estimated daily wildfire-related O_3 in study locations during 2000–2016

Table S12. Pooled percentage change in mortality associated with per 1 μg/m^3 in wildfire-related O_3 during lag 0-2 days by controlling temperature and humidity.

Model	All-cause mortality		Cardiovascular mortality		Respiratory mortality	
	Pooled estimates (%, 95%)	P for difference*	Pooled estimates (%, 95%)	P for difference	Pooled estimates (%, 95%)	P for difference*
TEMP only	0·55 (0·29, 0·80)	-	0·44 (-0·10, 0·99)	-	0·82 (0·18, 1·47)	-
TEMP+RH	0·40 (0·14, 0·66)	0.42	0·32 (-0·24, 0·89)	0.76	0·73 (0·0.07, 1·39)	0.85

*P value for difference was calculated using a 2-sample test. Based on the main model, the "TEMP+ RH" model further included the mean relative humidity during lag 0-2 days fitted with a natural cubic spline (with 3 degrees of freedom).

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