Supporting Information

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SI Text

We present supporting material describing the meteorological analysis, the global aerosol model, the representation of the volcanic emissions in the model, the calculation of the mean effect performing a meta-analysis of four short-term exposure studies, and the calculation of excess mortality assuming exposure of the European population to a baseline concentration of particulate matter with diameters smaller than 2.5 μm (PM2.5) typical of urban conditions.

Meteorological Analysis. We used six-hourly wind fields and geopotential height from the European Centre for Medium-Range Weather Forecasts (ECMWF) (00, 06, 12, and 18 Coordinated Universal Time analyses) from 1958 to 2010 (1, 2). Wind fields were examined for the 300, 500, and 700 hPa levels of constant pressure, corresponding to heights of approximately 9, 5.5, and 3 km, respectively. An index of the flow between Iceland and the United Kingdom was calculated. The index is designed to be positive for predominantly northwesterly flow. The index is

$$|\nu| \times \begin{cases} \cos(2\theta) & \text{if } |\theta| < \frac{\pi}{2} \\ -1 & \text{if } |\theta| \geq \frac{\pi}{2} \end{cases},$$

where $|\nu|$ is the wind speed in meters per second and θ is the angular deviation of the flow direction from the northwest. The index is evaluated at 10 locations (20°W, 65°N; 15°W, 65°N; 20°W, 62.5°N; 15°W, 62.5°N; 10°W, 62.5°N; 15°W, 60°N; 10°W, 60°N; 5°W, 60°N; 10°W, 57.5°N; 5°W, 57.5°N) and then averaged with a cos(latitude) weighting to give a single value every 6 h for each of the three pressure levels. Fig. 1 in the main text shows the annual percentage of positive values at 500 hPa for the years 1958–2010.

For our model analysis of air quality over Europe, we have simulated a year with a low frequency of northwesterly flow (2003) and a year with a high frequency of northwesterly flow (2005). Reconstruction of the meteorological situation in 1783 (see Fig. S1) suggests the Laki eruption itself occurred in a year with low northwesterly flow (3).

The 3D Global Chemistry-Aerosol Microphysics Model. We used the Global Model of Aerosol Processes (GLOMAP) to simulate sulfate, sea salt, elemental carbon, organic aerosol, and dust from natural and anthropogenic sources. The model is an extension of the TOMCAT 3D global chemical transport model (4). We used the modal version of the model (GLOMAP mode) where the aerosol size distribution is treated using a two-moment modal scheme (5). In these simulations, we included seven modes: hygroscopic nucleation, Aitken, accumulation and coarse modes plus nonhygroscopic Aitken, accumulation, and coarse modes. The simulations were conducted using a $2.8° \times 2.8°$ resolution, and reanalyzed ECMWF meteorology (1, 2). The control runs used a present-day atmospheric setting with the following natural and anthropogenic emissions: anthropogenic sulfur dioxide (SO_2) (6), volcanic SO_2 following recommendations of Dentener et al. (7), oceanic dimethyl sulfide (8, 9), sea spray (10), primary organic carbon (OC) and black carbon (BC) from biofuel and fossil fuel (11), biomass burning SO_2, and BC/OC (12) as well as dust (7). The simulations were conducted using the sulfur chemistry scheme described in Breider et al. (13) coupled to the full background TOMCAT chemistry (resolving Ox-NOy-HOx, C_1–C_3 nonmethane hydrocarbons and isoprene reactions).

The model has been evaluated against a wide range of aerosol measurements around the world (5). To supplement that evaluation, we show in Fig. S2 the comparison of modeled PM2.5 data against 399 multiannual measurements at European ground stations (14). We obtained a Pearson's correlation coefficient of 0.7 and a normalized mean bias of −0.14 (i.e., the model is 14% biased low on average).

Representing the Volcanic Emissions in the Model. To represent emissions from the Laki vent system in our model, we emitted a total of 94.3 Tg of SO_2 into model grid boxes above Iceland (17°W, 64°N) between 9 and 13 km as 10 discrete SO_2 mass loadings representing the 10 eruption episodes (15, 16) each injected within one 6-h period. In addition, a total of 27.6 Tg of SO_2 was continuously emitted into grid boxes between the surface and 1 km, representing passive degassing from the Laki lava flows (17).

Exposed Population. Data on the exposed population were obtained from the History Database of the Global Environment (HYDE 3.1) (18) for the year 2004 (see Fig. S3). The HYDE 3.1 data were provided on a $0.083° \times 0.083°$ grid and have been regridded to our model resolution ($2.8^{\circ} \times 2.8°$).

Meta-Analysis Using Existing Short-Term Exposure Studies. In order to obtain a combined effect for all-cause mortality due to short-term exposure to PM2.5, we used the four studies listed in Table S1 and calculated a mean effect using the standard technique of inverse-variance weighting (which assigns weight to each study based on the inverse of the variance).

Long-Term Excess Mortality Using Urban-Typical Baseline PM2.5 Concentrations. In order to address the uncertainty arising from the baseline PM2.5 exposure, we performed one additional calculation using the extreme assumption that the entire European population is exposed to PM2.5 concentrations that are typically observed in urban environments in Europe. First, we used the European air quality database (AIRBASE) observations (14) in order to obtain a European mean urban-to-rural PM2.5 ratio of 1.6. Second, we applied this urban-to-rural ratio to the gridded 12-mo mean PM2.5 concentrations in our control simulations as follows:

$$X_{\mathrm{Cy_{urban}}} = X_{\mathrm{Cy}} \times 1.6$$

and to the perturbed simulation as follows:

$$X_{\mathrm{Py_{urban}}} = X_{\mathrm{Cy}} \times 1.6 + (X_{\mathrm{Py}} - X_{\mathrm{Cy}}).$$

Third, we have calculated excess mortality due to long-term exposure to PM2.5 as described in the main text (see Eq. **1** in the main text and the equations in *Materials and Methods*).

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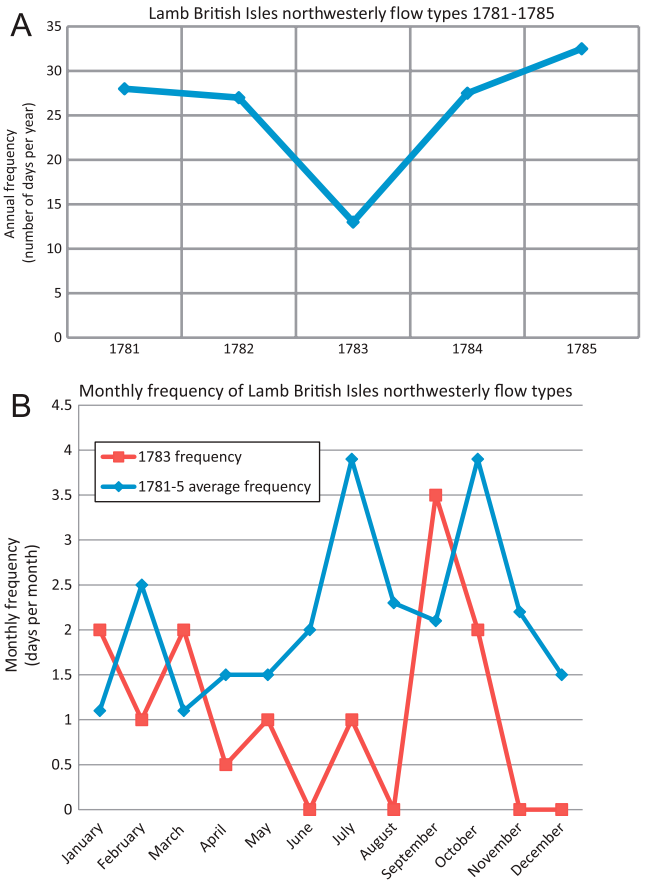

Fig. S1. (*A*) Annual frequency (number of days per year) of Lamb British Isles northwesterly flow types for the years 1781–1785. (*B*) Monthly frequency (number of days per month) of Lamb British Isles northwesterly flow types for the year 1783 (red line) and monthly mean frequency for the period 1781–1785 (blue line). Based on data from Kington (3).

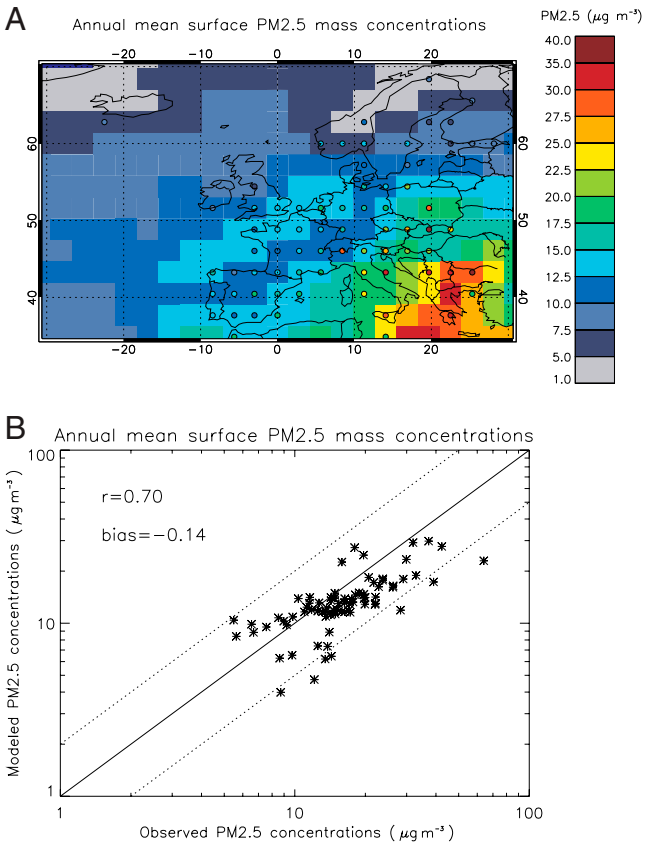

Fig. S2. Comparison of modeled annual mean surface PM2.5 mass concentrations against multiannual measurements at European ground stations (14) (overplotted circles represent mean of measurements in each respective grid box) in (*A*), and Pearson's correlation coefficient and normalized mean bias stated in (*B*).

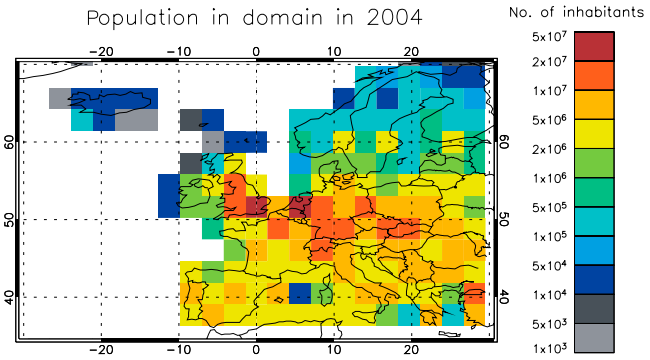

Fig. S3. Number of inhabitants in each grid box of the European model domain using 2004 population data from the HYDE 3.1 database (18).

Table S1. Meta-analysis using existing short-term exposure studies

Study	ER	ER low	ER high
Klemm and Mason (1)	1.2	0.8	1.6
Ostro et al. (2)	0.6	0.2	1.0
Franklin et al. (3)	1.2	0.3	2.1
Zanobetti and Schwartz (4)	0.98	0.75	1.22
Meta estimate ($\overline{ER}$)	0.96	0.79	1.13

Excess risks (ER) from the four studies considered (1–4), the inverse-variance weighted mean excess risk ($\overline{ER}$), and lower and upper 95% confidence intervals are given as percent increase in mortality per 10 $\mu g/m^3$ of PM2.5. Note that in order to obtain γ one divides the weighted mean excess risk by 1,000 in order to move from percent change per 10 $\mu g/m^3$ to excess risk per micron.

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3 Franklin M, Zeka A, Schwartz J (2007) Association between PM2.5 and all-cause and specific-cause mortality in 27 US communities. *J Expo Sci Environ Epidemiol* 17:279–287.

4 Zanobetti A, Schwartz J (2009) The effect of fine and coarse particulate air pollution on mortality: A national analysis. *Environ Health Perspect* 117:898–903.