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Supplemental Material

Long-Term Exposure to Fine Particle Elemental Components and Natural and Cause-Specific Mortality—a Pooled Analysis of Eight European Cohorts within the ELAPSE Project

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Area-level socio-economic status (SES) variable harmonization

In the ELAPSE study manual (version 2, 31-10-2016) we identified that area-level SES variables were needed as potential confounders in the epidemiological analysis of the pooled cohort and the administrative cohorts. The Area-level SES Workgroup identified that harmonized SES data were not available from European databases for the neighborhood-scale. Therefore all local partners were asked to obtain the area-level SES data. We specified what data should be obtained and linked to the cohort data.

The area-level SES variables we aimed to obtain included composite score (combining different dimensions in one overall score), mean household income, low household income rate, income support rate, unemployment rate, low education rate, high education rate and ethnicity.

All variables were collected for a small area (neighborhood) and large area (region) to allow for confounding at commonly used spatial scales in previous cohort studies, realizing that not all data are necessary.

A **neighborhood** is a part of a city, with about 1,000 – 10,000 people. Ideally, we use a standard definition, referring to externally defined areas. If this is not available, postal codes were used, if they refer to the approximate number of people defined above. Examples include “buurt” and “wijk” in the Netherlands including on average 1,400 and 6,000 subjects; parish or census district (~4,300 subjects) in Denmark. Quite a few of the ESCAPE cohorts have used municipality (or local administrative unit 2 (LAU2, former NUTS5)) in the ESCAPE project. We now aimed at a finer spatial scale, given that many of the included ESCAPE cohorts include a large metropolitan area with surrounding smaller towns. For smaller towns (e.g. below 10,000 subjects), the town (community) level was deemed to be fine. In some countries, data are available for multiple scales within the specified range. Availability of type of data (in multiple years) and comparability with other cohorts were criteria to select the scale. A very fine scale e.g. below 1,000 subjects is problematic for computational reasons (random effect models), particularly if the outcome is relatively rare.

The **region** is important for national cohorts. Each cohort defined this locally and judged whether a region scale is needed. When both a neighborhood and region scale is used, neighborhood should be nested within region.

SES has multiple dimensions, including income, education, occupation and employment. We use national composite scores that combine the different dimensions and in addition the main individual components as the association with air pollution and health may differ between dimensions. SES scores at regional scale were calculated by aggregating the raw variables to region level and then calculate the SES score.

Table S1. Characteristics of the Cardiovascular Effects of Air Pollution and Noise in Stockholm (CEANS) cohort

All participants resided in Stockholm County, Sweden. The cohort is comprised of four sub-cohorts: The Stockholm Diabetes Preventive Program (SDPP) is a population-based prospective study of 7,949 subjects aged 35–54 years. The Stockholm Cohort of 60-year-olds (SIXTY) sub-cohort consists of a random population sample of one-third of all men and women living in Stockholm County turning 60 years between August 1997 and March 1999. The Screening Across the Lifespan Twin Study (SALT) sampled 7,043 individuals from the Swedish Twin Register born 1958 and earlier, who lived in Stockholm County. Lastly, The Swedish National Study of Aging and Care in Kungsholmen (SNAC-K) randomly sampled individuals 60+ years of age from a central area in Stockholm.

		CEANS, sub-cohorts			
Variable		SDPP	SIXTY	SALT	SNAC-K
Baseline year, range		1992–1998	1997–1999	1998–2003	2001–2004
Enrolled, N[a]		7,835	4,180	6,724	3,248
Included in mortality analyses		7,716[b]	3,965[c]	6,174[d]	2,830[e]
Deaths, N (%)					
	Natural cause	337 (4.4)	593 (15.0)	891 (14.4)	960 (33.9)
	Cardiovascular diseases	74 (1.0)	163 (4.1)	326 (5.3)	416 (14.7)
	Respiratory diseases	12 (0.2)	38 (1.0)	57 (0.9)	61 (2.2)
	Lung Cancer	39 (0.5)	46 (1.2)	55 (0.9)	26 (0.9)
Age at baseline, yrs (mean ± SD)		47.1 ± 4.9	60.0 ± 0.0	57.8 ± 10.6	72.9 ± 10.4
Women, N (%)		4,721 (61.2)	2,065 (52.1)	3,416 (55.3)	1,767 (62.4)
Employed, N (%)		7,005 (90.8)	2,684 (67.7)	3,976 (64.4)	656 (23.2)
Marital status, N (%)					
	Single	1,271 (16.5)	182 (4.6)	863 (14.0)	459 (16.2)
	Married	6,445 (83.5)	2,930 (73.9)	4,179 (67.7)	1,300 (45.9)
	Divorced	-	649 (16.4)	693 (11.2)	388 (13.7)
	Widowed	-	204 (5.1)	439 (7.1)	683 (24.1)
Smoking status, N (%)					
	Current	2,035 (26.4)	839 (21.2)	1,311 (21.2)	404 (14.3)
	Previous	2,811 (36.4)	1,520 (38.3)	2,058 (33.3)	1,079 (38.1)
	Never	2,870 (37.2)	1,606 (40.5)	2,805 (45.4)	1,347 (47.6)
Smoking intensity[af], g/d (mean ± SD)		13.5 ± 7.4	13.4 ± 7.6	12.7 ± 8.0	11.7 ± 8.2
Smoking duration[af], yrs (mean ± SD)		27.9 ± 8.6	36.3 ± 9.9	37.9 ± 9.3	43.3 ± 13.6
BMI, kg/m^2, N (%)					
	< 18.5	54 (0.7)	26 (0.7)	94 (1.5)	78 (2.8)
	18.5–24.9	3,688 (47.8)	1,397 (35.2)	3,622 (58.7)	1,251 (44.2)
	25.0–29.9	3,007 (39.0)	1,767 (44.6)	2,054 (33.3)	1,134 (40.1)
	30.0+	967 (12.5)	775 (19.5)	404 (6.5)	367 (13.0)
Neighborhood income[g] (mean ± SD)		24.3 ± 4.2	24.7 ± 6.9	25.3 ± 6.6	28.7 ± 2.2

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to missing exposure (11), smoking status (6), smoking duration (23), smoking intensity (7), BMI (25), marital status (34), employment status (29), neighborhood income (4).

[c]Subjects were excluded due to missing exposure (4), smoking status (124), smoking duration (171), smoking intensity (124), marital status (122), employment status (158).

[d]Subjects were excluded due to missing exposure (2), smoking status (170), smoking duration (447), smoking intensity (170), BMI (253), marital status (103), employment status (29).

[e]Subjects were excluded due to fail in logistical checks (1), missing smoking status (88), smoking duration (183), smoking intensity (139), BMI (290), marital status (7), employment status (73), neighborhood income (4).
[f]For current smokers
[g]EUR per 1,000, year 2001

References:
(Eriksson et al., 2008; Lagergren et al., 2004; Lichtenstein et al., 2006; Wandell et al., 2007)

Table S2. Characteristics of the Diet, Cancer and Health (DCH) cohort

Participants were recruited among persons aged 50–64 years from the areas of greater Copenhagen and Aarhus, Denmark, who were born in Denmark and free of cancer at baseline.

Variable		**DCH**
Baseline year, range		1993–1997
Enrolled, N[a]		56,308
Included in mortality analyses[b]		52,779
Deaths, N (%)		
	Natural cause	10,490 (19.9)
	Cardiovascular diseases	2,143 (4.1)
	Respiratory diseases	861 (1.6)
	Lung Cancer	1,282 (2.4)
Age at baseline, yrs (mean ± SD)		56.7 ± 4.4
Women, N (%)		27,709 (52.5)
Employed, N (%)		41,313 (78.3)
Marital status, N (%)		
	Single	3,220 (6.1)
	Married	37,665 (71.4)
	Divorced	8,980 (17.0)
	Widowed	2,914 (5.5)
Smoking status, N (%)		
	Current	19,175 (36.3)
	Previous	14,685 (27.8)
	Never	18,919 (35.8)
Smoking intensity[c], g/d (mean ± SD)		16.5 ± 9.0
Smoking duration[c], yrs (mean ± SD)		36.3 ± 7.7
BMI, kg/m^2, N (%)		
	< 18.5	414 (0.8)
	18.5–24.9	22,781 (43.2)
	25.0–29.9	21,941 (41.6)
	30.0+	7,643 (14.5)
Neighborhood income[d] (mean ± SD)		20.1 ± 3.4

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to missing exposure (907), smoking status (75), smoking duration (433), smoking intensity (1062), BMI (42), marital status (461), employment status (309), neighborhood income (930).

[c]For current smokers

[d]EUR per 1,000, year 2001

Reference: (Tjonneland et al., 2007)

Table S3. Characteristics of the Danish Nurse Cohort (DNC)

The cohort was sampled among members of The Danish Nurse Organization (DNO) including both working and retired nurses. Questionnaires were mailed in 1993 to members aged 45+ years and again in 1999 with the inclusion of new members (45+ years).

		DNC, sub-cohorts	
Variable		DNC-1993	DNC-1999
Baseline year		1993	1999
Enrolled, N[a]		19,664	8,769
Included in mortality analyses		17,017[b]	8,117[c]
Deaths, N (%)			
	Natural cause	3,997 (23.5)	309 (3.8)
	Cardiovascular diseases	937 (5.5)	50 (0.6)
	Respiratory diseases	359 (2.1)	14 (0.2)
	Lung Cancer	351 (2.1)	30 (0.4)
Age at baseline, yrs (mean ± SD)		56.2 ± 8.4	47.9 ± 4.2
Women, N (%)		17,017 (100.0)	8,117 (100.0)
Employed, N (%)		11,907 (70.0)	7,693 (94.8)
Marital status, N (%)			
	Single	1,799 (10.6)	757 (9.3)
	Married	11,511 (67.6)	6,154 (75.8)
	Divorced	2,111 (12.4)	1,043 (12.8)
	Widowed	1,596 (9.4)	163 (2.0)
Smoking status, N (%)			
	Current	6,373 (37.5)	2,320 (28.6)
	Previous	4,864 (28.6)	2,646 (32.6)
	Never	5,780 (34.0)	3,151 (38.8)
Smoking intensity[d], g/d (mean ± SD)		13.9 ± 8.2	13.3 ± 7.3
Smoking duration[d], yrs (mean ± SD)		31.6 ± 9.9	27.1 ± 7.1
BMI, kg/m^2, N (%)			
	< 18.5	499 (2.9)	142 (1.7)
	18.5–24.9	11,742 (69.0)	5,539 (68.2)
	25.0–29.9	3,893 (22.9)	1,897 (23.4)
	30.0+	883 (5.2)	539 (6.6)
Neighborhood income[e] (mean ± SD)		19.2 ± 2.6	19.0 ± 2.4

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to missing exposure (32), smoking status (922), smoking duration (1641), smoking intensity (1314), BMI (156), marital status (201), employment status (590), neighborhood income (81).

[c]Subjects were excluded due to missing exposure (11), smoking status (30), smoking duration (165), smoking intensity (190), BMI (40), marital status (42), employment status (310).

[d]For current smokers

[e]EUR per 1,000, year 2001

Reference: (Hundrup et al., 2012)

Table S4. Characteristics of the European Prospective Investigation into Cancer and Nutrition, the Netherlands (EPIC-NL)

The EPIC-NL combines two Dutch EPIC-cohorts: The Monitoring Project on Risk Factors and chronic diseases in the Netherlands (MORGEN) cohort which consists of a general population sample aged 20–59 years from three Dutch towns (Amsterdam, Doetinchem and Maastricht). Prospect is a prospective cohort study among women aged 49–70, residing in the city of Utrecht or its vicinity, who participated in the nation-wide Dutch breast cancer screening programme between 1993 and 1997.

		EPIC-NL, sub-cohorts	
Variable		MORGEN	PROSPECT
Baseline year		1993–1997	1993–1997
Enrolled, N[a]		20,711	16,194
Included in mortality analyses		18,292[b]	14,570[c]
Deaths, N (%)			
	Natural cause	1,180 (6.5)	1,997 (13.7)
	Cardiovascular diseases	261 (1.4)	459 (3.2)
	Respiratory diseases	63 (0.3)	126 (0.9)
	Lung Cancer	160 (0.9)	168 (1.2)
Age at baseline, yrs (mean ± SD)		42.9 ± 11.3	57.7 ± 6.1
Women, N (%)		10,051 (54.9)	14,570 (100.0)
Employed, N (%)		12,571 (68.7)	7,402 (50.8)
Marital status, N (%)			
	Single	4629 (25.3)	836 (5.7)
	Married	11,916 (65.1)	11,179 (76.7)
	Divorced	1,380 (7.5)	1,172 (8.0)
	Widowed	367 (2.0)	1,383 (9.5)
Smoking status, N (%)			
	Current	6,357 (34.8)	3,335 (22.9)
	Previous	5,153 (28.2)	4,795 (32.9)
	Never	6,782 (37.1)	6,440 (44.2)
Smoking intensity[d], g/d (mean ± SD)		15.7 ± 8.6	13.7 ± 8.7
Smoking duration[d], yrs (mean ± SD)		24.8 ± 10.6	36.8 ± 7.6
BMI, kg/m^2, N (%)			
	< 18.5	188 (1.0)	87 (0.6)
	18.5–24.9	9,122 (49.9)	6,505 (44.6)
	25.0–29.9	6,869 (37.6)	5,790 (39.7)
	30.0+	2,113 (11.6)	2,188 (15.0)
Neighborhood income[e] (mean ± SD)		12.2 ± 1.6	13.1 ± 1.4

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to fail in logistical checks (28), missing exposure (11), smoking status (34), smoking duration (326), smoking intensity (1517), BMI (5), marital status (75), employment status (830), neighborhood income (2).

[c]Subjects were excluded due to fail in logistical checks (1), missing smoking status (100), smoking duration (374), smoking intensity (716), BMI (20), marital status (101), employment status (104), neighborhood income (883).

[d]For current smokers

[e]EUR per 1,000, year 2001

Reference: (Beulens et al., 2010)

Table S5. Characteristics of the Heinz Nixdorf Recall study (HNR)

The cohort consists of randomly sampled persons aged 45 to 75 years from the Ruhr area, Germany primarily in the three adjacent large cities Bochum, Essen, and Mülheim.

Variable		**HNR**
Baseline year, range		2000–2003
Enrolled, N[a]		4,809
Included in mortality analyses[b]		4,733
Deaths, N (%)		
	Natural cause	694 (14.7)
	Cardiovascular diseases	190 (4.0)
	Respiratory diseases	44 (0.9)
	Lung Cancer	63 (1.3)
Age at baseline, yrs (mean ± SD)		59.7 ± 7.8
Women, N (%)		2,382 (50.3)
Employed, N (%)		1,895 (40.0)
Marital status, N (%)		
	Single	274 (5.8)
	Married	3,538 (74.8)
	Divorced	472 (10.0)
	Widowed	449 (9.5)
Smoking status, N (%)		
	Current	1,113 (23.5)
	Previous	1,619 (34.2)
	Never	2,001 (42.3)
Smoking intensity[c], g/d (mean ± SD)		18.6 ± 12.0
Smoking duration[c], yrs (mean ± SD)		34.5 ± 9.4
BMI, kg/m^2, N (%)		
	< 18.5	16 (0.3)
	18.5–24.9	1,237 (26.1)
	25.0–29.9	2,171 (45.9)
	30.0+	1,309 (27.7)
Neighborhood income[d] (mean ± SD)		25.2 ± 8.2

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort
[b]Subjects were excluded due to missing smoking status (10), smoking duration (49), smoking intensity (14), BMI (29), marital status (12), employment status (15).
[c]For current smokers
[d]EUR per 1,000, year 2001

Reference: (Schmermund et al., 2002)

Table S6. Characteristics of the Etude Epidémiologique auprès de femmes de la Mutuelle Générale de l'Education Nationale (E3N)

The cohort was selected among French women aged 40 to 65 years who were insured through a national health system that primarily covered teachers. The cohort is nation-wide.

Variable		**E3N**
Baseline year, range		1989–1991
Enrolled, N[a]		53,521
Included in mortality analyses[b]		38,537
Deaths, N (%)		
	Natural cause	1,941 (5.0)
	Cardiovascular diseases	266 (0.7)
	Respiratory diseases	59 (0.2)
	Lung Cancer	132 (0.3)
Age at baseline, yrs (mean ± SD)		53.0 ± 6.8
Women, N (%)		38,537 (100.0)
Employed, N (%)		26,158 (67.9)
Marital status, N (%)		
	Single	6,436 (16.7)
	Married	32,101 (83.3)
	Divorced	-
	Widowed	-
Smoking status, N (%)		
	Current	4,988 (12.9)
	Previous	7,411 (19.2)
	Never	26,138 (67.8)
Smoking intensity[c], g/d (mean ± SD)		11.3 (9.2)
Smoking duration[c], yrs (mean ± SD)		28.5 (7.6)
BMI, kg/m^2, N (%)		
	< 18.5	1,386 (3.6)
	18.5–24.9	29,205 (75.8)
	25.0–29.9	6,574 (17.1)
	30.0+	1,372 (3.6)
Neighborhood income[d] (mean ± SD)		11.2 ± 3.0

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to fail in logistical checks (14), missing exposure (629), smoking duration (8211), smoking intensity (9729), BMI (2644), marital status (1989), neighborhood income (222).

[c]For current smokers

[d]EUR per 1,000, year 2001

Reference: (Clavel-Chapelon and Group, 2015)

Table S7. Characteristics of the Cooperative Health Research in the Region of Augsburg (KORA)

Two cross-sectional population-representative surveys were conducted in 1994-1995 (survey S3) and 1999-2001 (survey S4) in the city of Augsburg and two adjacent rural counties including inhabitants of German nationality aged 25 to 74.

		KORA, sub-cohorts	
Variable		**S3**	**S4**
Baseline year, range		1994–1995	1999–2001
Enrolled, N[a]		4,566	4,257
Included in mortality analyses		2,572[b]	2,281[c]
Deaths, N (%)			
	Natural cause	391 (15.2)	215 (9.4)
	Cardiovascular diseases	159 (6.2)	72 (3.2)
	Respiratory diseases	33 (1.3)	20 (0.9)
	Lung Cancer	25 (1.0)	19 (0.8)
Age at baseline, yrs (mean ± SD)		49.4 ± 13.9	49.3 ± 13.8
Women, N (%)		1,308 (50.9)	1,173 (51.4)
Employed, N (%)		1,423 (55.3)	1,356 (59.4)
Marital status, N (%)			
	Single	227 (8.8)	184 (8.1)
	Married	2,060 (80.1)	1,807 (79.2)
	Divorced	108 (4.2)	151 (6.6)
	Widowed	177 (6.9)	139 (6.1)
Smoking status, N (%)			
	Current	519 (20.2)	523 (22.9)
	Previous	740 (28.8)	720 (31.6)
	Never	1,313 (51.0)	1,038 (45.5)
Smoking intensity[d], g/d (mean ± SD)		16.5 ± 9.5	15.7 ± 9.5
Smoking duration[d], yrs (mean ± SD)		25.2 ± 12.1	24.3 ± 11.6
BMI, kg/m^2, N (%)			
	< 18.5	13 (0.5)	8 (0.4)
	18.5–24.9	837 (32.5)	710 (31.1)
	25.0–29.9	1,116 (43.4)	996 (43.7)
	30.0+	606 (23.6)	567 (24.9)
Neighborhood income[e] (mean ± SD)		36.7 ± 4.4	38.0 ± 7.3

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to fail in logistical checks (10), missing smoking duration (84), smoking intensity (129), BMI (52), neighborhood income (1825).

[c]Subjects were excluded due to missing smoking status (5), smoking duration (84), smoking intensity (18), BMI (37), marital status (5), employment status (6), neighborhood income (1892).

[d]For current smokers

[e]EUR per 1,000, year 2001

Reference: (Holle et al., 2005)

Table S8. Characteristics of the Vorarlberg Health Monitoring and Prevention Programme (VHM&PP)

The VHM&PP is a population-based cohort recruited among all adults of the province of Vorarlberg, Austria. Vorarlberg is the western-most province of Austria consisting of towns and villages (30,000 inhabitants and smaller) and significant altitude differences.

Variable		**VHM&PP**
Baseline year, range		1985–2005
Enrolled, N[a]		170,250
Included in mortality analyses[b]		144,199
Deaths, N (%)		
	Natural cause	22,645 (15.7)
	Cardiovascular diseases	9,976 (6.9)
	Respiratory diseases	1,099 (0.8)
	Lung Cancer	1,380 (1.0)
Age at baseline, yrs (mean ± SD)		42.1 ± 15.0
Women, N (%)		81,017 (56.2)
Employed, N (%)		100,585 (69.8)
Marital status, N (%)		
	Single	24,832 (17.2)
	Married	99,400 (68.9)
	Divorced	9,762 (6.8)
	Widowed	10,205 (7.1)
Smoking status, N (%)		
	Current	28,871 (20.0)
	Previous	8,995 (6.2)
	Never	106,333 (73.7)
Smoking intensity[c], g/d (mean ± SD)		15.6 ± 8.9
Smoking duration[c], yrs (mean ± SD)		13.4 ± 8.3
BMI, kg/m^2, N (%)		
	< 18.5	4,450 (3.1)
	18.5–24.9	78,575 (54.5)
	25.0–29.9	45,533 (31.6)
	30.0+	15,641 (10.8)
Neighborhood income[d] (mean ± SD)		22.9 ± 1.7

[a]The number of subjects for which information was transferred to Utrecht University for construction of the pooled cohort

[b]Subjects were excluded due to missing exposure (396), smoking duration (5789), smoking intensity (6376), BMI (11), marital status (9970), employment status (15056), neighborhood income (1895).

[c]For current smokers

[d]EUR per 1,000, year 2001

Reference: (Ulmer et al., 2007)

Table S9. Performance of Europe-wide $PM_{2.5}$ composition models in five-fold hold-out validation[a]

	Cu	Fe	K	Ni	S	Si	V	Zn
Performance of $PM_{2.5}$ composition models over Europe: r^2								
SLR	0.48	0.48	0.59	0.56	0.79	0.46	0.63	0.41
RF	0.59	0.61	0.80	0.76	0.90	0.62	0.86	0.71
Performance of $PM_{2.5}$ composition models to assess within-area variation: average within-area r^2								
SLR	0.35	0.36	0.07	0.17	0.14	0.20	0.19	0.19
RF	0.29	0.29	0.07	0.21	0.23	0.17	0.29	0.25

[a] Values extracted from Chen et al. (2020)

r^2 = squared Pearson correlation, SLR = Supervised Linear Regression model, RF = Random Forest model

Table S10. Truncation frequency (Truncation performed for model 3 population, N=323,782)

Pollutant	Exposure model	N below zero (%)	N above maximum (%)
$PM_{2.5}$ Cu	SLR	36,683 (11.3)[a]	2 (0)[b]
	RF	0 (0)	0 (0)
$PM_{2.5}$ Fe	SLR	1,645 (0.5)[c]	0 (0)
	RF	0 (0)	0 (0)
$PM_{2.5}$ K	SLR	0 (0)	0 (0)
	RF	0 (0)	0 (0)
$PM_{2.5}$ Ni	SLR	37,470 (11.6)[d]	24 (0)[e]
	RF	0 (0)	0 (0)
$PM_{2.5}$ S	SLR	0 (0)	0 (0)
	RF	0 (0)	0 (0)
$PM_{2.5}$ Si	SLR	0 (0)	0 (0)
	RF	0 (0)	0 (0)
$PM_{2.5}$ V	SLR	46,243 (14.3)[f]	0 (0)
	RF	0 (0)	0 (0)
$PM_{2.5}$ Zn	SLR	8,154 (2.5)[g]	240 (0.1)[h]
	RF	0 (0)	0 (0)

N = Number of observations; SLR = Supervised Linear Regression model, RF = Random Forest model

[a] 7464 (97%) in CEANS-SDPP, 1754 (44%) in CEANS-SIXTY, 2348 (38%) in CEANS-SALT, 2 (<1%) in CEANS-SNACK, 3158 (6%) in DCH, 7237 (43%) in DNC-1993, 3564 (44%) in DNC-1999, 1 (<1%) in EPIC-NL-Morgen, 110 (<1%) in E3N, 11045 (8%) in VHM&PP

[b] 2 (<1%) in E3N

[c] 563 (7%) in CEANS-SDPP, 30 (1%) in CEANS-SIXTY, 53 (1%) in CEANS-SALT, 58 (<1%) in DCH, 640 (4%) in DNC-1993, 301 (4%) in DNC-1999,

[d] 51 (1%) in CEANS-SDPP, 2 (<1%) in CEANS-SIXTY, 3 (<1%) in CEANS-SALT, 3 (<1%) in DCH, 94 (1%) in DNC-1993, 32 (<1%) in DNC-1999, 438 (1%) in E3N, 382 (15%) in KORA-S3, 250 (11%) in KORA-S4, 36215 (25%) in VHM&PP

[e] 4 (<1%) in DCH, 3 (<1%) in DNC-1993, 2 (<1%) in EPIC-NL-Prospect, 10 (<1%) in E3N, 5 (<1%) in VHM&PP

[f] 37 (1%) in CEANS-SIXTY, 68 (1%) in CEANS-SALT, 183 (6%) in CEANS- SNACK, 80 (<1%) in DCH, 12 (<1%) in DNC-1993, 5 (<1%) in DNC-1999, 582 (2%) in E3N, 1102 (43%) in KORA-S3, 1078 (47%) in KORA-S4, 43096 (30%) in VHM&PP

[g] 127 (<1%) in E3N, 8027 (6%) in VHM&PP

[h] 1 (<1%) in DNC-1993, 16 (<1%) in EPIC-NL-Morgen, 3 (<1%) in HNR, 220 (1%) in E3N

Table S11. Exposure distribution of $PM_{2.5}$ composition in the pooled cohort

Exposure	Exposure model	Mean	SD	IQR	Min	P5	P25	Median	P75	P95	Max
$PM_{2.5}$ Cu	SLR	3.5	2.6	3.7	0.0	0.0	1.4	3.5	5.1	7.3	42.4
	RF	3.9	1.6	1.9	0.9	1.9	2.7	3.9	4.6	6.7	19.2
$PM_{2.5}$ Fe	SLR	87.0	45.5	55.9	0.0	20.9	56.3	84.3	112.2	158.3	453.9
	RF	83.8	33.5	34.2	21.0	43.9	62.5	75.0	96.8	154.1	311.8
$PM_{2.5}$ K	SLR	166.7	52.4	82.2	31.8	89.8	123.1	165.0	205.3	255.6	321.4
	RF	212.3	101.6	200.1	74.4	89.7	112.0	210.7	312.1	371.0	480.6
$PM_{2.5}$ Ni	SLR	0.8	0.7	0.8	0.0	0.0	0.3	0.6	1.1	2.2	12.7
	RF	0.8	0.6	0.9	0.1	0.2	0.3	0.8	1.2	1.9	3.8
$PM_{2.5}$ S	SLR	658.4	139.9	212.9	299.0	438.3	554.1	649.7	766.9	884.2	1251.9
	RF	688.9	132.5	123.2	484.2	528.2	613.2	641.3	736.4	926.8	1314.1
$PM_{2.5}$ Si	SLR	96.2	20.6	23.9	37.5	68.7	82.4	93.7	106.3	133.7	255.3
	RF	85.9	25.0	23.3	38.1	60.6	71.0	78.7	94.2	132.7	299.7
$PM_{2.5}$ V	SLR	1.3	1.4	1.7	0.0	0.0	0.4	0.8	2.1	4.1	17.8
	RF	1.3	1.1	1.6	0.3	0.3	0.3	1.3	1.9	3.5	7.3
$PM_{2.5}$ Zn	SLR	16.8	11.0	10.6	0.0	3.8	10.9	15.3	21.5	31.0	145.4
	RF	19.6	7.4	9.7	9.5	11.2	13.5	20.2	23.2	30.6	73.9

SLR = Supervised Linear Regression model, RF = Random Forest model, SD = standard deviation, IQR = interquartile range, P5 to P95 are percentiles.
Unit for pollutants: ng/m^3

Table S12. Spearman correlation coefficient between $PM_{2.5}$ composition and $PM_{2.5}$ mass (N=323,782)

Sub-cohort	$PM_{2.5}$ Cu		$PM_{2.5}$ Fe		$PM_{2.5}$ K		$PM_{2.5}$ Ni		$PM_{2.5}$ S		$PM_{2.5}$ Si		$PM_{2.5}$ V		$PM_{2.5}$ Zn	
	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF
Average[a]	**0.44**	**0.49**	**0.47**	**0.45**	**0.21**	**0.42**	**0.25**	**0.18**	**0.48**	**0.41**	**0.38**	**0.22**	**0.13**	**0.13**	**0.42**	**0.43**
CEANS-SDPP	0.19	0.60	0.31	0.36	-0.07	0.39	-0.23	0.07	0.55	0.48	0.21	-0.19	-0.23	0.32	-0.18	0.44
CEANS-SIXTY	0.49	0.60	0.54	0.52	0.06	0.54	0.31	0.34	0.48	0.34	0.48	0.37	-0.03	0.24	0.45	0.43
CEANS-SALT	0.49	0.57	0.51	0.52	0.06	0.53	0.29	0.35	0.47	0.31	0.47	0.41	-0.02	0.28	0.41	0.42
CEANS-SNACK	0.50	0.64	0.53	0.65	0.55	0.55	0.19	0.51	0.46	0.25	0.45	0.31	0.43	0.30	0.39	0.57
DCH	0.74	0.72	0.76	0.66	-0.19	0.54	0.52	0.64	0.66	0.65	0.65	0.41	0.32	0.60	0.68	0.67
DNC-1993	0.42	0.40	0.38	0.37	0.17	0.42	0.22	0.27	0.42	0.50	0.36	0.22	0.11	0.28	0.48	0.34
DNC-1999	0.30	0.29	0.27	0.23	0.11	0.36	0.12	0.19	0.30	0.38	0.20	0.12	0.00	0.22	0.36	0.21
EPIC-NL-Morgen	0.22	0.24	0.28	0.24	0.50	0.49	0.14	0.06	0.46	0.61	0.28	0.46	-0.16	-0.52	0.55	0.55
EPIC-NL-Prospect	0.40	0.32	0.37	0.33	0.44	0.43	0.11	0.41	0.63	0.54	0.30	0.23	0.23	0.22	0.34	0.29
HNR	0.39	0.44	0.44	0.63	-0.34	0.54	0.28	0.52	0.48	0.27	0.35	0.29	0.30	0.41	0.46	0.59
E3N	0.62	0.63	0.67	0.56	0.31	-0.12	0.41	0.20	0.43	0.50	0.51	0.08	0.17	0.24	0.64	0.66
KORA-S3	0.28	0.28	0.41	0.33	0.26	0.36	0.22	-0.32	0.27	0.19	0.26	0.19	0.06	-0.36	0.22	0.16
KORA-S4	0.34	0.48	0.46	0.48	0.28	0.25	0.41	-0.35	0.33	0.19	0.44	0.43	0.00	-0.36	0.37	0.18
VHM&PP	0.72	0.63	0.62	0.38	0.76	0.59	0.54	-0.31	0.79	0.56	0.38	-0.21	0.65	-0.04	0.68	0.44

[a]Average of cohort-specific correlation coefficients

Table S13. Spearman correlation coefficients between $PM_{2.5}$ composition and NO_2 (N=323,782)

Sub-cohort	$PM_{2.5}$ Cu		$PM_{2.5}$ Fe		$PM_{2.5}$ K		$PM_{2.5}$ Ni		$PM_{2.5}$ S		$PM_{2.5}$ Si		$PM_{2.5}$ V		$PM_{2.5}$ Zn	
	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF	SLR	RF
Average[a]	**0.71**	**0.77**	**0.78**	**0.74**	**0.09**	**0.38**	**0.46**	**0.33**	**0.46**	**0.33**	**0.68**	**0.37**	**0.27**	**0.22**	**0.51**	**0.54**
CEANS-SDPP	0.29	0.81	0.84	0.70	0.07	0.79	0.16	0.30	0.31	-0.06	0.73	-0.20	0.04	0.33	0.39	0.62
CEANS-SIXTY	0.83	0.89	0.90	0.86	-0.24	0.69	0.53	0.58	0.46	0.32	0.82	0.65	-0.13	0.21	0.58	0.70
CEANS-SALT	0.85	0.89	0.90	0.87	-0.28	0.68	0.52	0.62	0.44	0.33	0.82	0.71	-0.11	0.25	0.56	0.69
CEANS-SNACK	0.74	0.84	0.77	0.88	0.72	0.72	0.27	0.66	0.47	0.36	0.68	0.56	0.63	0.30	0.54	0.70
DCH	0.90	0.88	0.88	0.79	-0.24	0.63	0.55	0.62	0.57	0.58	0.63	0.31	0.36	0.61	0.62	0.68
DNC-1993	0.57	0.61	0.59	0.51	-0.12	0.42	0.27	0.25	0.33	0.28	0.50	0.17	0.02	0.27	0.39	0.51
DNC-1999	0.35	0.36	0.35	0.28	-0.13	0.28	0.13	0.13	0.19	0.18	0.26	0.08	-0.07	0.17	0.23	0.29
EPIC-NL-Morgen	0.88	0.90	0.88	0.88	-0.39	-0.45	0.72	0.71	0.27	-0.30	0.76	0.51	0.65	0.46	0.27	-0.35
EPIC-NL-Prospect	0.86	0.89	0.89	0.88	-0.12	0.71	0.57	0.09	0.41	0.56	0.85	0.77	0.32	-0.21	0.66	0.73
HNR	0.69	0.71	0.71	0.72	-0.16	0.34	0.17	0.63	0.41	0.32	0.52	0.33	0.33	0.42	0.26	0.50
E3N	0.81	0.82	0.86	0.81	0.28	-0.16	0.58	0.36	0.58	0.59	0.76	0.31	0.36	0.43	0.73	0.72
KORA-S3	0.72	0.69	0.72	0.72	0.58	0.14	0.60	0.05	0.63	0.42	0.67	0.58	0.32	-0.23	0.6	0.52
KORA-S4	0.68	0.69	0.73	0.74	0.56	-0.09	0.60	0.14	0.63	0.39	0.67	0.60	0.32	0.00	0.61	0.57
VHM&PP	0.83	0.79	0.88	0.76	0.71	0.67	0.72	-0.52	0.72	0.63	0.80	-0.24	0.75	0.10	0.76	0.65

[a]Average of cohort-specific correlation coefficients

Table S14. Associations of $PM_{2.5}$ composition with natural mortality in single pollutant and two-pollutant models

Exposure	Exposure model	Single pollutant HR	Two-pollutant model adjusting for $PM_{2.5}$		Two-pollutant model adjusting for NO_2	
			component	$PM_{2.5}$[a]	component	NO_2[a]
$PM_{2.5}$ Cu	SLR	1.120 (1.094, 1.147)	1.043 (1.011, 1.076)	1.105 (1.073, 1.137)	1.023 (0.983, 1.065)	1.074 (1.047, 1.102)
	RF	1.154 (1.111, 1.198)	1.035 (0.989, 1.083)	1.122 (1.093, 1.151)	0.943 (0.887, 1.002)	1.107 (1.081, 1.134)
$PM_{2.5}$ Fe	SLR	1.139 (1.110, 1.169)	1.065 (1.031, 1.100)	1.099 (1.070, 1.129)	1.024 (0.974, 1.076)	1.075 (1.044, 1.106)
	RF	1.132 (1.090, 1.176)	1.055 (1.013, 1.099)	1.122 (1.096, 1.148)	0.921 (0.869, 0.976)	1.114 (1.089, 1.139)
$PM_{2.5}$ K	SLR	1.049 (1.035, 1.064)	0.998 (0.981, 1.015)	1.136 (1.105, 1.169)	1.027 (1.012, 1.041)	1.077 (1.060, 1.094)
	RF	1.056 (1.042, 1.070)	1.021 (1.006, 1.037)	1.114 (1.086, 1.143)	1.031 (1.017, 1.046)	1.072 (1.055, 1.090)
$PM_{2.5}$ Ni	SLR	1.084 (1.063, 1.106)	1.043 (1.020, 1.066)	1.114 (1.087, 1.140)	1.030 (1.006, 1.055)	1.074 (1.055, 1.093)
	RF	1.011 (0.971, 1.053)	0.993 (0.953, 1.034)	1.134 (1.110, 1.159)	0.949 (0.909, 0.990)	1.093 (1.076, 1.110)
$PM_{2.5}$ S	SLR	1.142 (1.113, 1.173)	1.049 (1.009, 1.090)	1.102 (1.068, 1.137)	1.074 (1.039, 1.109)	1.061 (1.042, 1.081)
	RF	1.127 (1.079, 1.177)	0.999 (0.951, 1.051)	1.134 (1.106, 1.162)	1.013 (0.964, 1.064)	1.085 (1.067, 1.103)
$PM_{2.5}$ Si	SLR	1.268 (1.205, 1.336)	1.151 (1.087, 1.217)	1.108 (1.082, 1.134)	1.071 (0.995, 1.152)	1.072 (1.050, 1.095)
	RF	0.967 (0.921, 1.014)	0.969 (0.924, 1.017)	1.134 (1.109, 1.159)	0.906 (0.863, 0.952)	1.095 (1.078, 1.112)
$PM_{2.5}$ V	SLR	1.061 (1.044, 1.079)	1.033 (1.015, 1.052)	1.120 (1.094, 1.145)	1.026 (1.007, 1.045)	1.077 (1.060, 1.095)
	RF	1.092 (1.050, 1.135)	1.056 (1.015, 1.099)	1.128 (1.104, 1.153)	1.026 (0.985, 1.069)	1.084 (1.067, 1.101)
$PM_{2.5}$ Zn	SLR	1.051 (1.039, 1.064)	1.015 (0.999, 1.031)	1.118 (1.089, 1.147)	1.021 (1.006, 1.036)	1.074 (1.055, 1.093)
	RF	1.062 (1.036, 1.089)	0.992 (0.964, 1.021)	1.137 (1.110, 1.165)	1.002 (0.974, 1.030)	1.087 (1.069, 1.105)

Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from natural mortality = 46,640.

HR = Hazard Ratio, SLR = Supervised Linear Regression model, RF = Random Forest model

HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3; main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income

[a]Single pollutant HRs are 1.134, 95% CI: 1.109, 1.159 per 5 µg/m^3 increase in $PM_{2.5}$ mass and 1.087, 95% CI 1.071, 1.103 per 10 µg/m^3 in NO_2. $PM_{2.5}$ mass and NO_2 exposure were estimated using SLR only.

Table S15. Associations of $PM_{2.5}$ composition with natural mortality with increasing control for covariates

Exposure	Exposure model	Model 1 HR N = 323,782[a]	Model 2 HR N = 323,782[a]	Model 3 HR N = 323,782[a]	Model 1 HR N = 378,979[b]	Model 2 HR N = 330,667[c]
$PM_{2.5}$ Cu	SLR	1.175 (1.148, 1.202)	1.103 (1.078, 1.128)	1.120 (1.094, 1.147)	1.168 (1.144, 1.193)	1.099 (1.075, 1.125)
	RF	1.257 (1.211, 1.305)	1.130 (1.088, 1.173)	1.154 (1.111, 1.198)	1.239 (1.198, 1.280)	1.119 (1.079, 1.160)
$PM_{2.5}$ Fe	SLR	1.216 (1.186, 1.248)	1.121 (1.092, 1.150)	1.139 (1.110, 1.169)	1.202 (1.175, 1.231)	1.117 (1.089, 1.145)
	RF	1.258 (1.212, 1.306)	1.119 (1.078, 1.162)	1.132 (1.090, 1.176)	1.233 (1.192, 1.275)	1.107 (1.067, 1.148)
$PM_{2.5}$ K	SLR	1.049 (1.035, 1.063)	1.043 (1.029, 1.056)	1.049 (1.035, 1.064)	1.053 (1.040, 1.066)	1.042 (1.028, 1.055)
	RF	1.059 (1.046, 1.073)	1.048 (1.034, 1.061)	1.056 (1.042, 1.070)	1.062 (1.049, 1.075)	1.050 (1.036, 1.063)
$PM_{2.5}$ Ni	SLR	1.163 (1.142, 1.185)	1.087 (1.066, 1.109)	1.084 (1.063, 1.106)	1.164 (1.145, 1.184)	1.087 (1.066, 1.108)
	RF	1.122 (1.078, 1.168)	1.027 (0.986, 1.069)	1.011 (0.971, 1.053)	1.108 (1.068, 1.149)	1.018 (0.978, 1.060)
$PM_{2.5}$ S	SLR	1.204 (1.173, 1.236)	1.135 (1.106, 1.165)	1.142 (1.113, 1.173)	1.182 (1.155, 1.210)	1.119 (1.091, 1.148)
	RF	1.219 (1.167, 1.274)	1.120 (1.072, 1.170)	1.127 (1.079, 1.177)	1.141 (1.101, 1.182)	1.076 (1.034, 1.119)
$PM_{2.5}$ Si	SLR	1.534 (1.458, 1.614)	1.264 (1.200, 1.330)	1.268 (1.205, 1.336)	1.477 (1.411, 1.546)	1.243 (1.183, 1.307)
	RF	1.055 (1.007, 1.105)	0.986 (0.941, 1.034)	0.967 (0.921, 1.014)	1.033 (0.990, 1.078)	0.978 (0.934, 1.024)
$PM_{2.5}$ V	SLR	1.129 (1.111, 1.148)	1.066 (1.048, 1.084)	1.061 (1.044, 1.079)	1.135 (1.118, 1.152)	1.066 (1.048, 1.084)
	RF	1.205 (1.159, 1.252)	1.099 (1.058, 1.143)	1.092 (1.050, 1.135)	1.204 (1.161, 1.248)	1.094 (1.052, 1.137)
$PM_{2.5}$ Zn	SLR	1.074 (1.062, 1.087)	1.045 (1.033, 1.058)	1.051 (1.039, 1.064)	1.072 (1.060, 1.083)	1.044 (1.031, 1.056)
	RF	1.084 (1.058, 1.111)	1.050 (1.024, 1.076)	1.062 (1.036, 1.089)	1.084 (1.060, 1.108)	1.047 (1.022, 1.073)

[a] Model 3 population = 323,782, person-years at risk = 6,317,235; number of death from natural mortality = 46,640.

[b] Model 1 population = 378,979, person-years at risk = 7,291,866; number of death from natural mortality = 52,849. We excluded 54 (<0.1%) subjects after logistical checks and 2,003 (0.5%) subjects due to missing exposure. Missing data for each cohort are shown in Tables S1 to S8.

[c] Model 2 population = 330,667, person-years at risk = 6,433,069; number of death from natural mortality = 47,524. We further excluded 48312 (12.7%) subjects from Model 1 population due to missing individual level covariates. Missing data for each cohort are shown in Tables S1 to S8.

HR = Hazard Ratio, SLR = Supervised Linear Regression model, RF = Random Forest model

HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3; Model 1 adjusted for age, sub-cohort id, sex and year of enrollment; Model 2 further adjusted for smoking (status, duration, intensity, intensity^2), BMI categories, marital status and employment status; Model 3 further adjusted for 2001 neighborhood-level mean income

Table S16. Associations of $PM_{2.5}$ composition with cardiovascular mortality in single pollutant and two-pollutant models

Exposure	Exposure model	Single pollutant HR	Two-pollutant model adjusting for $PM_{2.5}$		Two-pollutant model adjusting for NO_2	
			component	$PM_{2.5}$[a]	component	NO_2[a]
$PM_{2.5}$ Cu	SLR	1.126 (1.080, 1.173)	1.033 (0.977, 1.092)	1.116 (1.064, 1.171)	1.045 (0.974, 1.122)	1.064 (1.014, 1.115)
	RF	1.168 (1.086, 1.257)	1.011 (0.926, 1.105)	1.134 (1.086, 1.183)	0.941 (0.835, 1.060)	1.110 (1.061, 1.161)
$PM_{2.5}$ Fe	SLR	1.147 (1.094, 1.204)	1.055 (0.994, 1.119)	1.111 (1.064, 1.161)	1.035 (0.940, 1.139)	1.071 (1.014, 1.132)
	RF	1.129 (1.048, 1.217)	1.042 (0.963, 1.127)	1.130 (1.089, 1.173)	0.887 (0.792, 0.994)	1.127 (1.081, 1.176)
$PM_{2.5}$ K	SLR	1.054 (1.032, 1.077)	0.997 (0.967, 1.027)	1.141 (1.086, 1.199)	1.030 (1.006, 1.054)	1.072 (1.039, 1.105)
	RF	1.075 (1.052, 1.098)	1.042 (1.015, 1.070)	1.092 (1.045, 1.142)	1.054 (1.029, 1.080)	1.054 (1.021, 1.088)
$PM_{2.5}$ Ni	SLR	1.080 (1.038, 1.124)	1.023 (0.978, 1.070)	1.129 (1.086, 1.173)	1.012 (0.964, 1.063)	1.085 (1.050, 1.121)
	RF	0.943 (0.866, 1.026)	0.947 (0.870, 1.031)	1.137 (1.097, 1.178)	0.903 (0.829, 0.984)	1.096 (1.065, 1.127)
$PM_{2.5}$ S	SLR	1.149 (1.098, 1.201)	1.036 (0.967, 1.110)	1.113 (1.054, 1.176)	1.090 (1.029, 1.154)	1.054 (1.017, 1.092)
	RF	1.090 (1.005, 1.181)	0.925 (0.844, 1.015)	1.156 (1.110, 1.204)	0.952 (0.867, 1.044)	1.099 (1.064, 1.135)
$PM_{2.5}$ Si	SLR	1.267 (1.148, 1.399)	1.128 (1.015, 1.255)	1.119 (1.077, 1.163)	1.033 (0.895, 1.192)	1.083 (1.040, 1.127)
	RF	0.915 (0.835, 1.003)	0.936 (0.854, 1.025)	1.135 (1.095, 1.177)	0.868 (0.791, 0.953)	1.098 (1.067, 1.129)
$PM_{2.5}$ V	SLR	1.066 (1.028, 1.104)	1.027 (0.989, 1.066)	1.128 (1.087, 1.171)	1.022 (0.982, 1.063)	1.082 (1.050, 1.116)
	RF	1.091 (1.002, 1.187)	1.056 (0.969, 1.150)	1.134 (1.094, 1.176)	1.022 (0.936, 1.116)	1.088 (1.057, 1.120)
$PM_{2.5}$ Zn	SLR	1.054 (1.030, 1.079)	1.003 (0.973, 1.033)	1.134 (1.084, 1.186)	1.018 (0.990, 1.048)	1.077 (1.041, 1.114)
	RF	1.074 (1.026, 1.125)	0.982 (0.930, 1.038)	1.145 (1.099, 1.192)	1.002 (0.950, 1.058)	1.089 (1.055, 1.124)

Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from cardiovascular mortality = 15,492.

HR = Hazard Ratio, SLR = Supervised Linear Regression model, RF = Random Forest model

HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3; main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income

[a]Single pollutant HRs are 1.137, 95% CI: 1.097, 1.178 per 5 µg/m^3 increase in $PM_{2.5}$ mass and 1.090, 95% CI 1.060, 1.120 per 10 µg/m^3 in NO_2. $PM_{2.5}$ mass and NO_2 exposure were estimated using SLR only.

Table S17. Associations of $PM_{2.5}$ composition with non-malignant respiratory mortality in single pollutant and two-pollutant models

Exposure	Exposure model	Single pollutant HR	Two-pollutant model adjusting for $PM_{2.5}$		Two-pollutant model adjusting for NO_2	
			component	$PM_{2.5}$[a]	component	NO_2[a]
$PM_{2.5}$ Cu	SLR	1.085 (0.984, 1.197)	1.088 (0.955, 1.239)	0.996 (0.881, 1.127)	0.861 (0.728, 1.018)	1.189 (1.073, 1.317)
	RF	1.124 (0.961, 1.315)	1.108 (0.921, 1.333)	1.016 (0.911, 1.134)	0.791 (0.611, 1.023)	1.184 (1.075, 1.304)
$PM_{2.5}$ Fe	SLR	1.148 (1.031, 1.277)	1.173 (1.027, 1.340)	0.968 (0.863, 1.086)	0.952 (0.778, 1.166)	1.130 (1.010, 1.263)
	RF	1.184 (1.018, 1.378)	1.175 (0.999, 1.383)	1.013 (0.917, 1.119)	0.955 (0.759, 1.201)	1.119 (1.023, 1.224)
$PM_{2.5}$ K	SLR	0.973 (0.918, 1.032)	0.929 (0.863, 1.001)	1.125 (1.002, 1.263)	0.944 (0.888, 1.004)	1.121 (1.054, 1.192)
	RF	1.020 (0.961, 1.082)	1.003 (0.933, 1.078)	1.047 (0.936, 1.171)	0.975 (0.914, 1.041)	1.115 (1.045, 1.190)
$PM_{2.5}$ Ni	SLR	1.142 (1.063, 1.227)	1.150 (1.064, 1.243)	0.978 (0.884, 1.081)	1.099 (1.007, 1.199)	1.059 (0.986, 1.137)
	RF	1.209 (1.036, 1.411)	1.200 (1.026, 1.402)	1.033 (0.941, 1.134)	1.124 (0.953, 1.325)	1.087 (1.021, 1.158)
$PM_{2.5}$ S	SLR	1.113 (0.995, 1.244)	1.147 (0.975, 1.350)	0.966 (0.843, 1.106)	0.996 (0.867, 1.143)	1.105 (1.027, 1.190)
	RF	1.063 (0.879, 1.287)	1.016 (0.816, 1.265)	1.046 (0.940, 1.163)	0.909 (0.735, 1.123)	1.119 (1.047, 1.195)
$PM_{2.5}$ Si	SLR	1.252 (1.016, 1.544)	1.241 (0.987, 1.562)	1.009 (0.912, 1.117)	0.972 (0.728, 1.298)	1.110 (1.023, 1.205)
	RF	0.982 (0.812, 1.186)	0.980 (0.811, 1.184)	1.050 (0.957, 1.152)	0.907 (0.746, 1.103)	1.112 (1.046, 1.182)
$PM_{2.5}$ V	SLR	1.082 (1.019, 1.148)	1.079 (1.014, 1.149)	1.012 (0.918, 1.115)	1.046 (0.980, 1.117)	1.084 (1.015, 1.157)
	RF	1.110 (0.963, 1.278)	1.097 (0.949, 1.268)	1.037 (0.943, 1.139)	1.028 (0.884, 1.196)	1.100 (1.032, 1.171)
$PM_{2.5}$ Zn	SLR	1.031 (0.975, 1.091)	1.020 (0.951, 1.095)	1.029 (0.917, 1.155)	0.971 (0.905, 1.041)	1.123 (1.045, 1.206)
	RF	1.006 (0.898, 1.127)	0.972 (0.853, 1.107)	1.061 (0.955, 1.179)	0.913 (0.802, 1.039)	1.126 (1.054, 1.203)

Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from non-malignant respiratory mortality = 2,846.
HR = Hazard Ratio, SLR = Supervised Linear Regression model, RF = Random Forest model
HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3; main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income
[a]Single pollutant HRs are 1.050, 95% CI: 0.957, 1.152 per 5 µg/m^3 increase in $PM_{2.5}$ mass and 1.104, 95% CI 1.040, 1.172 per 10 µg/m^3 in NO_2. $PM_{2.5}$ mass and NO_2 exposure were estimated using SLR only.

Table S18. Associations of $PM_{2.5}$ composition with lung cancer mortality in single pollutant and two-pollutant models

Exposure	Exposure model	Single pollutant HR	Two-pollutant model adjusting for $PM_{2.5}$		Two-pollutant model adjusting for NO_2	
			component	$PM_{2.5}$[a]	component	NO_2[a]
$PM_{2.5}$ Cu	SLR	1.114 (1.024, 1.211)	0.983 (0.879, 1.100)	1.199 (1.076, 1.337)	1.021 (0.884, 1.180)	1.068 (0.978, 1.166)
	RF	1.127 (0.989, 1.285)	0.957 (0.817, 1.121)	1.204 (1.092, 1.327)	0.914 (0.737, 1.134)	1.109 (1.020, 1.205)
$PM_{2.5}$ Fe	SLR	1.125 (1.027, 1.233)	1.003 (0.894, 1.125)	1.185 (1.070, 1.312)	1.012 (0.852, 1.202)	1.073 (0.974, 1.181)
	RF	1.113 (0.978, 1.266)	1.004 (0.872, 1.156)	1.185 (1.085, 1.295)	0.918 (0.754, 1.118)	1.106 (1.024, 1.196)
$PM_{2.5}$ K	SLR	1.076 (1.022, 1.133)	1.014 (0.951, 1.081)	1.171 (1.059, 1.295)	1.060 (1.005, 1.118)	1.064 (1.009, 1.122)
	RF	1.061 (1.008, 1.117)	1.005 (0.947, 1.067)	1.182 (1.075, 1.298)	1.037 (0.982, 1.096)	1.065 (1.008, 1.125)
$PM_{2.5}$ Ni	SLR	1.091 (1.028, 1.159)	1.043 (0.975, 1.116)	1.160 (1.061, 1.268)	1.056 (0.981, 1.136)	1.052 (0.990, 1.119)
	RF	1.007 (0.885, 1.146)	0.962 (0.844, 1.098)	1.191 (1.096, 1.295)	0.932 (0.811, 1.071)	1.089 (1.031, 1.151)
$PM_{2.5}$ S	SLR	1.265 (1.148, 1.395)	1.201 (1.044, 1.382)	1.063 (0.945, 1.196)	1.258 (1.116, 1.419)	1.005 (0.943, 1.070)
	RF	1.203 (1.024, 1.414)	1.029 (0.855, 1.237)	1.178 (1.074, 1.294)	1.112 (0.932, 1.327)	1.064 (1.006, 1.125)
$PM_{2.5}$ Si	SLR	1.292 (1.082, 1.544)	1.124 (0.923, 1.369)	1.160 (1.060, 1.270)	1.153 (0.901, 1.475)	1.049 (0.977, 1.126)
	RF	1.034 (0.878, 1.218)	1.022 (0.867, 1.204)	1.186 (1.093, 1.287)	0.974 (0.823, 1.154)	1.081 (1.025, 1.139)
$PM_{2.5}$ V	SLR	1.071 (1.019, 1.126)	1.040 (0.986, 1.096)	1.163 (1.067, 1.268)	1.047 (0.990, 1.106)	1.058 (1.000, 1.119)
	RF	1.133 (1.009, 1.273)	1.082 (0.960, 1.219)	1.173 (1.079, 1.275)	1.077 (0.951, 1.220)	1.066 (1.010, 1.126)
$PM_{2.5}$ Zn	SLR	1.053 (1.010, 1.098)	1.004 (0.952, 1.059)	1.182 (1.072, 1.303)	1.029 (0.980, 1.081)	1.061 (1.002, 1.125)
	RF	1.070 (0.988, 1.159)	0.997 (0.911, 1.091)	1.188 (1.086, 1.299)	1.032 (0.947, 1.124)	1.071 (1.015, 1.131)

Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from lung cancer mortality = 3,776.

HR = Hazard Ratio, SLR = Supervised Linear Regression model, RF = Random Forest model

HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3; main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income

[a] Single pollutant HRs are 1.187, 95% CI: 1.093, 1.288 per 5 µg/m^3 increase in $PM_{2.5}$ mass and 1.079, 95% CI 1.025, 1.135 per 10 µg/m^3 in NO_2. $PM_{2.5}$ mass and NO_2 exposure were estimated using SLR only.

Table S19. Sensitivity analysis: Associations of $PM_{2.5}$ composition with natural mortality with restricted follow-up period

Exposure	Exposure model	Hazard Ratio (95% confidence interval)			
		Full follow-up[a]	After 2000[b]	After 2005[c]	After 2008[d]
$PM_{2.5}$ Cu	SLR	1.120 (1.094, 1.147)	1.117 (1.089, 1.146)	1.099 (1.067, 1.132)	1.080 (1.043, 1.118)
	RF	1.154 (1.111, 1.198)	1.143 (1.098, 1.190)	1.132 (1.080, 1.186)	1.116 (1.056, 1.180)
$PM_{2.5}$ Fe	SLR	1.139 (1.110, 1.169)	1.135 (1.104, 1.167)	1.119 (1.083, 1.156)	1.106 (1.064, 1.149)
	RF	1.132 (1.090, 1.176)	1.126 (1.081, 1.172)	1.117 (1.067, 1.171)	1.117 (1.057, 1.180)
$PM_{2.5}$ K	SLR	1.049 (1.035, 1.064)	1.040 (1.025, 1.056)	1.025 (1.007, 1.043)	1.009 (0.989, 1.030)
	RF	1.056 (1.042, 1.070)	1.048 (1.033, 1.064)	1.039 (1.022, 1.057)	1.028 (1.007, 1.048)
$PM_{2.5}$ Ni	SLR	1.084 (1.063, 1.106)	1.085 (1.062, 1.107)	1.079 (1.053, 1.105)	1.078 (1.048, 1.108)
	RF	1.011 (0.971, 1.053)	1.047 (1.003, 1.092)	1.049 (0.999, 1.101)	1.060 (1.001, 1.122)
$PM_{2.5}$ S	SLR	1.142 (1.113, 1.173)	1.130 (1.098, 1.163)	1.095 (1.060, 1.132)	1.076 (1.035, 1.119)
	RF	1.127 (1.079, 1.177)	1.118 (1.066, 1.172)	1.110 (1.051, 1.173)	1.089 (1.021, 1.162)
$PM_{2.5}$ Si	SLR	1.268 (1.205, 1.336)	1.237 (1.170, 1.306)	1.218 (1.143, 1.298)	1.220 (1.132, 1.315)
	RF	0.967 (0.921, 1.014)	0.982 (0.934, 1.032)	0.975 (0.921, 1.033)	0.987 (0.922, 1.056)
$PM_{2.5}$ V	SLR	1.061 (1.044, 1.079)	1.061 (1.042, 1.080)	1.053 (1.032, 1.075)	1.051 (1.026, 1.076)
	RF	1.092 (1.050, 1.135)	1.112 (1.068, 1.159)	1.096 (1.045, 1.149)	1.091 (1.033, 1.153)
$PM_{2.5}$ Zn	SLR	1.051 (1.039, 1.064)	1.044 (1.030, 1.058)	1.041 (1.026, 1.057)	1.032 (1.013, 1.051)
	RF	1.062 (1.036, 1.089)	1.051 (1.023, 1.079)	1.049 (1.017, 1.081)	1.031 (0.994, 1.070)

[a]Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from natural mortality = 46,640.

[b]Follow-up period restricted to after year 2000, number of observations = 315,197 (97%); person-years at risk = 4,380,951 (69%); number of death from natural mortality = 39,321 (84%)

[c]Follow-up period restricted to after year 2005, number of observations = 303,022 (94%); person-years at risk = 2,905,753 (46%); number of death from natural mortality = 29,629 (64%)

[d]Follow-up period restricted to after year 2008, number of observations = 293,717 (91%); person-years at risk = 2,011,584 (32%); number of death from natural mortality = 21,785 (47%)

SLR = Supervised Linear Regression model, RF = Random Forest model. HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3; main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income

Table S20. Sensitivity analysis: Associations between $PM_{2.5}$ composition and natural mortality with additional adjustment for individual level socio-economic status

Exposure	Exposure model	Full population[a]	w/o VHM (N = 178,387)		w/o DCH (N = 271,003)		w/o DCH, E3N, EPIC_NL, HNR (N = 181,082)	
			Main model[b]	Main model + education	Main model	Main model + Occupational Status	Main model	Main model + Collar Blue
$PM_{2.5}$ Cu	SLR	1.120 (1.094, 1.147)	1.076 (1.039, 1.114)	1.078 (1.041, 1.116)	1.083 (1.055, 1.112)	1.082 (1.054, 1.111)	1.111 (1.077, 1.146)	1.117 (1.083, 1.153)
	RF	1.154 (1.111, 1.198)	1.086 (1.037, 1.138)	1.090 (1.041, 1.141)	1.095 (1.047, 1.145)	1.094 (1.046, 1.144)	1.168 (1.098, 1.242)	1.184 (1.113, 1.259)
$PM_{2.5}$ Fe	SLR	1.139 (1.110, 1.169)	1.081 (1.045, 1.119)	1.082 (1.046, 1.120)	1.095 (1.062, 1.128)	1.094 (1.062, 1.128)	1.139 (1.097, 1.183)	1.151 (1.108, 1.195)
	RF	1.132 (1.090, 1.176)	1.073 (1.028, 1.121)	1.076 (1.030, 1.123)	1.079 (1.030, 1.129)	1.077 (1.029, 1.128)	1.148 (1.079, 1.221)	1.162 (1.092, 1.236)
$PM_{2.5}$ K	SLR	1.049 (1.035, 1.064)	0.980 (0.947, 1.015)	0.980 (0.947, 1.015)	1.050 (1.035, 1.064)	1.050 (1.035, 1.064)	1.058 (1.042, 1.074)	1.060 (1.045, 1.077)
	RF	1.056 (1.042, 1.070)	1.003 (0.973, 1.034)	1.005 (0.975, 1.036)	1.049 (1.035, 1.063)	1.049 (1.035, 1.063)	1.066 (1.049, 1.082)	1.069 (1.053, 1.085)
$PM_{2.5}$ Ni	SLR	1.084 (1.063, 1.106)	1.048 (1.024, 1.072)	1.050 (1.026, 1.075)	1.018 (0.987, 1.049)	1.017 (0.986, 1.048)	1.051 (1.010, 1.093)	1.056 (1.015, 1.098)
	RF	1.011 (0.971, 1.053)	1.053 (1.009, 1.099)	1.059 (1.014, 1.105)	0.873 (0.827, 0.920)	0.870 (0.825, 0.917)	0.776 (0.718, 0.838)	0.758 (0.702, 0.820)
$PM_{2.5}$ S	SLR	1.142 (1.113, 1.173)	1.087 (1.040, 1.136)	1.086 (1.039, 1.135)	1.112 (1.080, 1.145)	1.112 (1.080, 1.144)	1.134 (1.097, 1.172)	1.139 (1.102, 1.177)
	RF	1.127 (1.079, 1.177)	1.065 (1.003, 1.129)	1.065 (1.003, 1.130)	1.077 (1.029, 1.127)	1.076 (1.029, 1.126)	1.108 (1.045, 1.176)	1.124 (1.060, 1.193)
$PM_{2.5}$ Si	SLR	1.268 (1.205, 1.336)	1.135 (1.067, 1.207)	1.135 (1.066, 1.207)	1.177 (1.107, 1.252)	1.176 (1.106, 1.251)	1.311 (1.210, 1.420)	1.348 (1.244, 1.460)
	RF	0.967 (0.921, 1.014)	1.039 (0.986, 1.094)	1.039 (0.986, 1.095)	0.918 (0.867, 0.973)	0.918 (0.866, 0.972)	0.903 (0.839, 0.971)	0.896 (0.833, 0.964)
$PM_{2.5}$ V	SLR	1.061 (1.044, 1.079)	1.029 (1.010, 1.049)	1.031 (1.012, 1.050)	0.995 (0.963, 1.028)	0.994 (0.962, 1.027)	1.029 (0.987, 1.073)	1.034 (0.992, 1.078)
	RF	1.092 (1.050, 1.135)	1.067 (1.025, 1.110)	1.072 (1.030, 1.116)	0.956 (0.902, 1.013)	0.953 (0.899, 1.010)	0.934 (0.863, 1.010)	0.934 (0.863, 1.010)
$PM_{2.5}$ Zn	SLR	1.051 (1.039, 1.064)	1.017 (1.000, 1.034)	1.017 (1.000, 1.034)	1.030 (1.016, 1.043)	1.029 (1.016, 1.043)	1.078 (1.056, 1.100)	1.083 (1.062, 1.105)
	RF	1.062 (1.036, 1.089)	1.017 (0.985, 1.050)	1.016 (0.984, 1.049)	1.046 (1.020, 1.073)	1.045 (1.019, 1.072)	1.111 (1.065, 1.158)	1.124 (1.079, 1.172)

[a]Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from natural mortality = 46,640.

[b]Main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income

SLR = Supervised Linear Regression model, RF = Random Forest model. HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3.

Table S21. Sensitivity analysis: Associations between $PM_{2.5}$ composition and natural mortality with additional adjustment for neighborhood-level socio-economic status[a]

Exposure	Exposure model	w/o CEANS (N = 302,968)		w/o EPIC_NL, HNR, KORA (N = 281,333)		w/o EPIC_NL, HNR, KORA (N = 281,333)		w/o CEANS, HNR, KORA (N = 293,510)	
		Main model[b]	Main model + unemployment rate	Main model	Main model + low educational level rate	Main model	Main model + high educational level rate	Main model	Main model + Ethnicity
$PM_{2.5}$ Cu	SLR	1.121 (1.094, 1.148)	1.118 (1.091, 1.145)	1.127 (1.100, 1.154)	1.129 (1.102, 1.157)	1.127 (1.100, 1.154)	1.132 (1.104, 1.159)	1.123 (1.096, 1.150)	1.109 (1.081, 1.138)
	RF	1.157 (1.113, 1.203)	1.151 (1.107, 1.197)	1.171 (1.125, 1.219)	1.177 (1.130, 1.226)	1.171 (1.125, 1.219)	1.185 (1.137, 1.234)	1.159 (1.114, 1.206)	1.129 (1.081, 1.178)
$PM_{2.5}$ Fe	SLR	1.143 (1.113, 1.174)	1.139 (1.109, 1.171)	1.149 (1.119, 1.181)	1.154 (1.123, 1.186)	1.149 (1.119, 1.181)	1.160 (1.128, 1.192)	1.145 (1.115, 1.177)	1.131 (1.098, 1.164)
	RF	1.141 (1.096, 1.188)	1.136 (1.091, 1.182)	1.152 (1.106, 1.201)	1.160 (1.112, 1.209)	1.152 (1.106, 1.201)	1.168 (1.120, 1.218)	1.139 (1.093, 1.186)	1.105 (1.058, 1.155)
$PM_{2.5}$ K	SLR	1.048 (1.034, 1.062)	1.046 (1.032, 1.060)	1.051 (1.037, 1.066)	1.051 (1.036, 1.065)	1.051 (1.037, 1.066)	1.050 (1.036, 1.065)	1.049 (1.035, 1.064)	1.050 (1.036, 1.065)
	RF	1.054 (1.040, 1.068)	1.053 (1.040, 1.067)	1.056 (1.043, 1.070)	1.057 (1.043, 1.071)	1.056 (1.043, 1.070)	1.056 (1.042, 1.070)	1.055 (1.042, 1.069)	1.053 (1.039, 1.067)
$PM_{2.5}$ Ni	SLR	1.086 (1.065, 1.108)	1.084 (1.063, 1.106)	1.099 (1.077, 1.122)	1.112 (1.088, 1.135)	1.099 (1.077, 1.122)	1.114 (1.091, 1.138)	1.087 (1.066, 1.109)	1.074 (1.051, 1.097)
	RF	1.023 (0.980, 1.067)	1.024 (0.982, 1.068)	1.018 (0.974, 1.064)	1.023 (0.978, 1.070)	1.018 (0.974, 1.064)	1.028 (0.983, 1.076)	1.017 (0.974, 1.062)	0.976 (0.932, 1.021)
$PM_{2.5}$ S	SLR	1.138 (1.108, 1.169)	1.135 (1.105, 1.166)	1.146 (1.115, 1.177)	1.146 (1.116, 1.177)	1.146 (1.115, 1.177)	1.146 (1.115, 1.177)	1.139 (1.109, 1.170)	1.127 (1.097, 1.158)
	RF	1.123 (1.074, 1.174)	1.114 (1.065, 1.166)	1.132 (1.082, 1.185)	1.132 (1.082, 1.185)	1.132 (1.082, 1.185)	1.131 (1.081, 1.184)	1.128 (1.078, 1.181)	1.111 (1.061, 1.163)
$PM_{2.5}$ Si	SLR	1.287 (1.219, 1.358)	1.279 (1.211, 1.352)	1.301 (1.232, 1.373)	1.313 (1.243, 1.388)	1.301 (1.232, 1.373)	1.324 (1.252, 1.399)	1.292 (1.223, 1.365)	1.256 (1.184, 1.331)
	RF	0.951 (0.902, 1.004)	0.945 (0.896, 0.998)	0.964 (0.916, 1.014)	0.963 (0.915, 1.013)	0.964 (0.916, 1.014)	0.964 (0.916, 1.014)	0.947 (0.896, 1.000)	0.924 (0.874, 0.976)
$PM_{2.5}$ V	SLR	1.065 (1.047, 1.083)	1.063 (1.045, 1.081)	1.070 (1.051, 1.089)	1.077 (1.058, 1.097)	1.070 (1.051, 1.089)	1.079 (1.060, 1.099)	1.066 (1.048, 1.084)	1.055 (1.036, 1.074)
	RF	1.095 (1.053, 1.139)	1.095 (1.053, 1.139)	1.107 (1.062, 1.154)	1.111 (1.066, 1.159)	1.107 (1.062, 1.154)	1.113 (1.067, 1.160)	1.096 (1.053, 1.140)	1.061 (1.017, 1.106)
$PM_{2.5}$ Zn	SLR	1.050 (1.038, 1.063)	1.049 (1.036, 1.062)	1.074 (1.059, 1.088)	1.074 (1.060, 1.089)	1.074 (1.059, 1.088)	1.074 (1.060, 1.089)	1.056 (1.043, 1.070)	1.051 (1.038, 1.065)
	RF	1.058 (1.033, 1.085)	1.055 (1.029, 1.082)	1.113 (1.078, 1.149)	1.113 (1.078, 1.149)	1.113 (1.078, 1.149)	1.112 (1.078, 1.148)	1.058 (1.032, 1.085)	1.055 (1.029, 1.082)

[a]See Table S20 for effect estimates in the full population. Total number of observations = 323,782; person-years at risk = 6,317,235; number of death from natural mortality = 46,640.

[b]Main model adjusted for sub-cohort id, age, sex, year of enrollment, smoking (status, duration, intensity, intensity^2), BMI categories, marital status, employment status and 2001 neighborhood-level mean income

SLR = Supervised Linear Regression model, RF = Random Forest model. HR (95% confidence interval) presented for the following increments: $PM_{2.5}$ Cu – 5 ng/m^3, $PM_{2.5}$ Fe – 100 ng/m^3, $PM_{2.5}$ K – 50 ng/m^3, $PM_{2.5}$ Ni – 1 ng/m^3, $PM_{2.5}$ S – 200 ng/m^3, $PM_{2.5}$ Si – 100 ng/m^3, $PM_{2.5}$ V – 2 ng/m^3, $PM_{2.5}$ Zn – 10 ng/m^3, $PM_{2.5}$ mass – 5 µg/m^3, NO_2 – 10 µg/m^3.

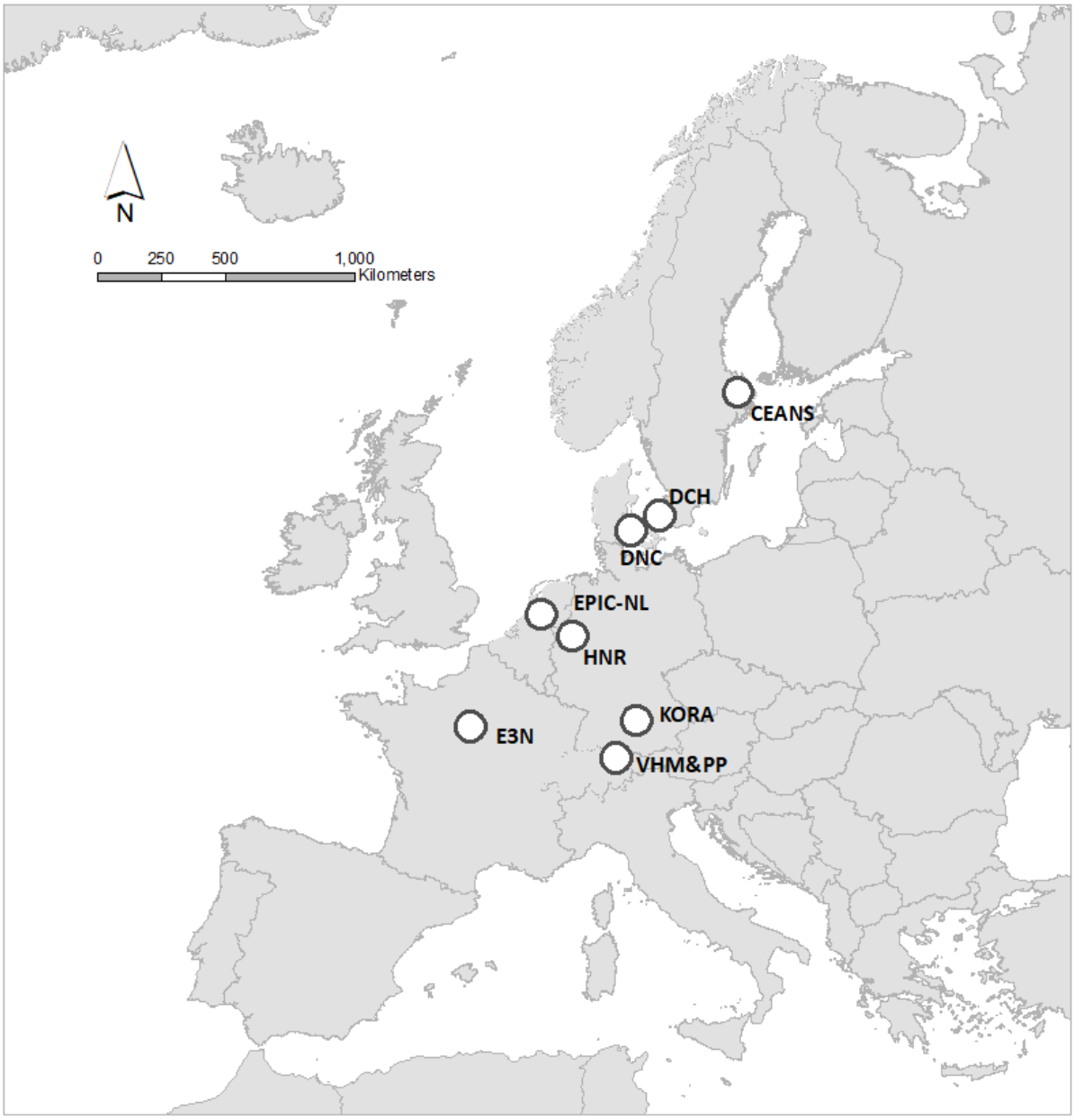


Figure S1. Map of study areas

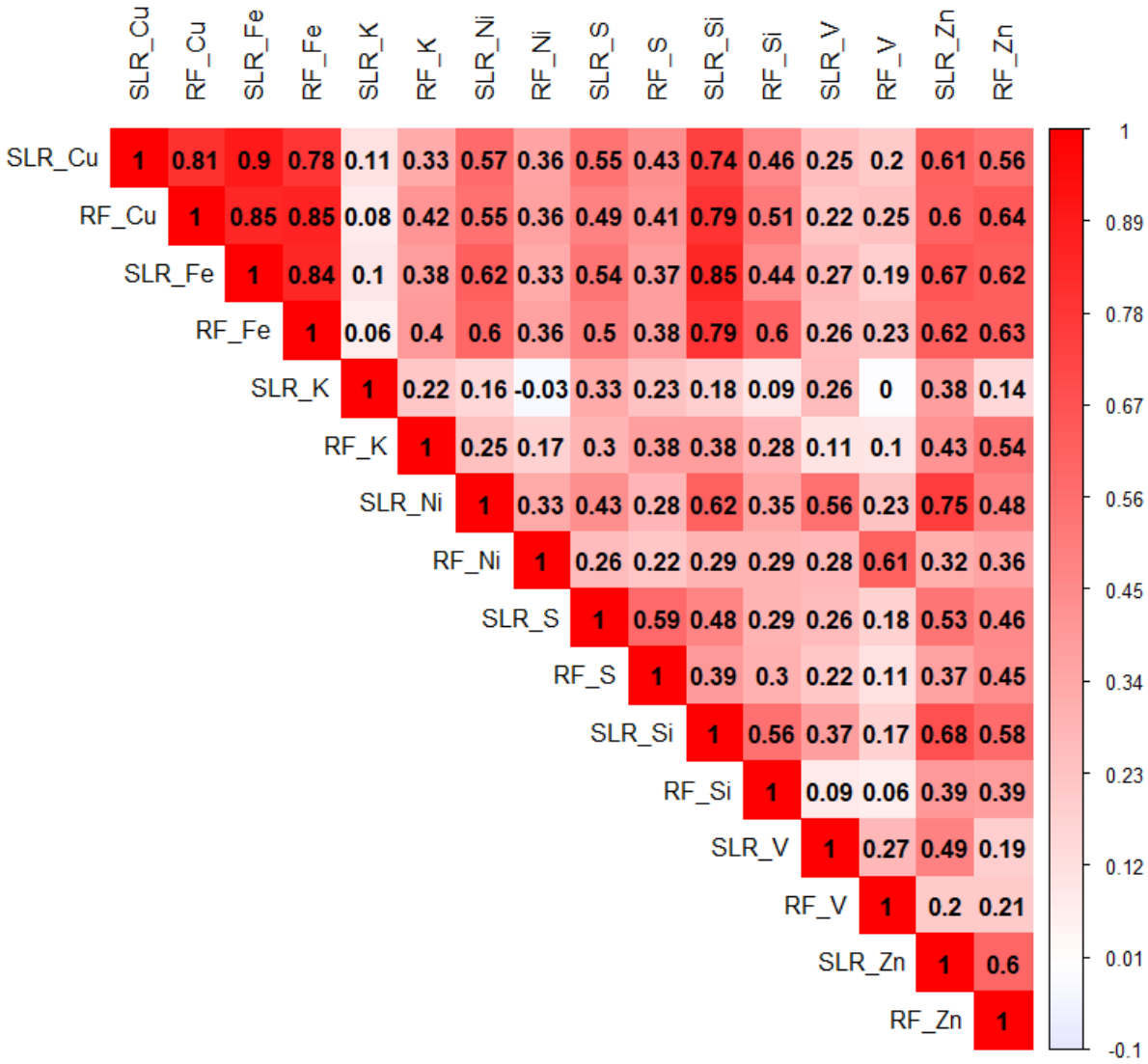
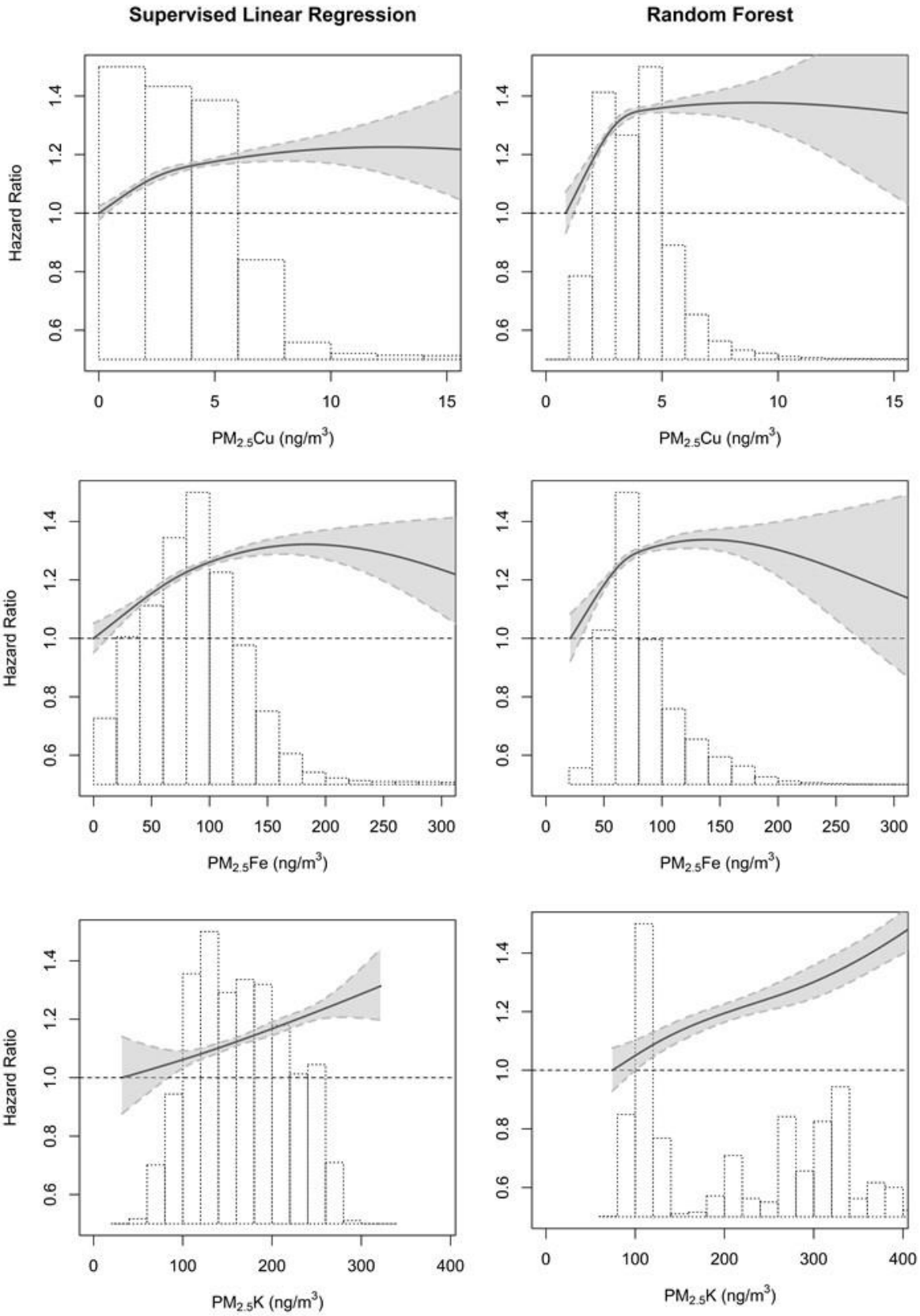
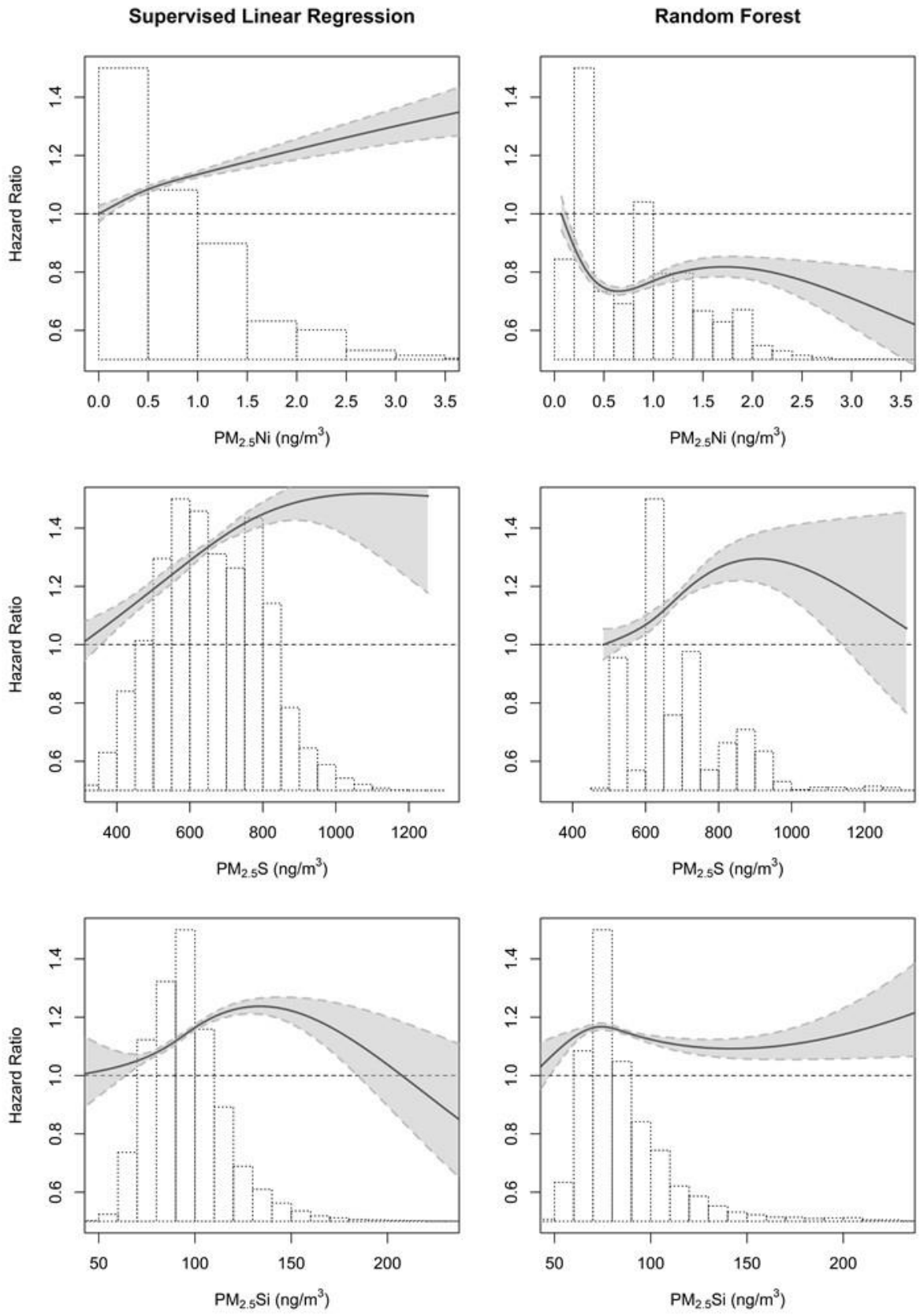


Figure S2. Average of cohort-specific Spearman correlation coefficients between $PM_{2.5}$ composition


Supervised Linear Regression
Random Forest
Hazard Ratio
0.6
0.8
1.0
1.2
1.4
0
5
10
15
PM2.5Cu (ng/m3)
0
50
100
150
200
250
300
PM2.5Fe (ng/m3)
0
100
200
300
400
PM2.5K (ng/m3)


Supervised Linear Regression
Random Forest
Hazard Ratio
0.6
0.8
1.0
1.2
1.4
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
PM2.5Ni (ng/m3)
400
600
800
1000
1200
PM2.5S (ng/m3)
50
100
150
200
PM2.5Si (ng/m3)

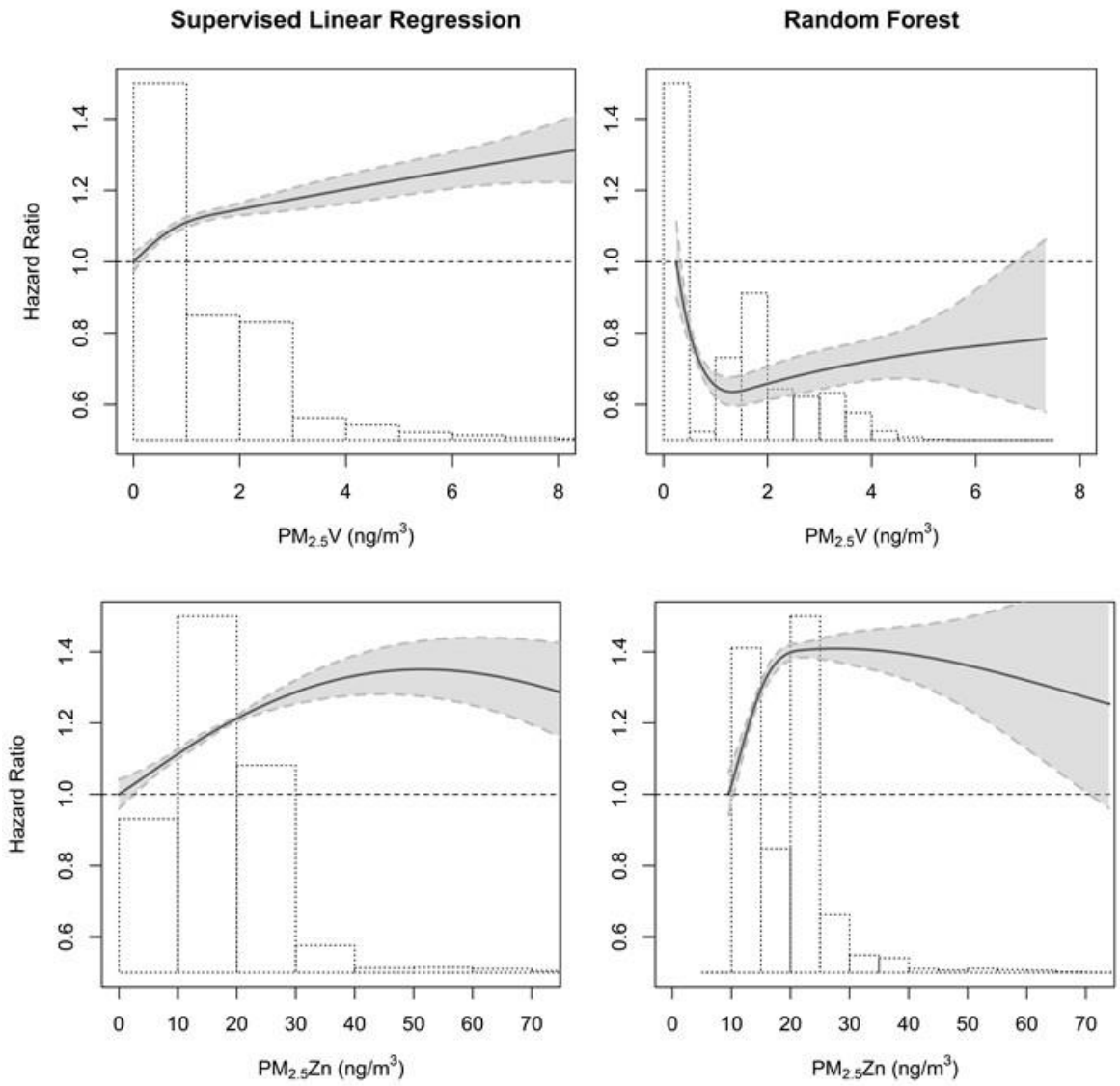


Figure S3. Natural spline (3 degree of freedom) for associations between $PM_{2.5}$ composition and natural cause mortality

Histogram shows the exposure distribution (exposure distribution for each individual cohort is shown in Figure 1); Shaded: 95% confidence intervals; Y-axis truncated at 0.5 and 1.5 for all components; Hazard ratios are expressed relative to minimum exposure.

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