

**Supplement for:**

## **Trends in air pollution in Europe, 2000-2019**

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Table S1. Cont.

Code	O <sub>3</sub>	SO <sub>2</sub>	SO <sub>4a</sub>	SO <sub>4wd</sub>	NO <sub>2</sub>	sNO <sub>3</sub>	NO <sub>3a</sub>	HNO <sub>3</sub>	NO <sub>3wd</sub>	sNH <sub>4</sub>	NH <sub>4a</sub>	NH <sub>3</sub>	NH <sub>4wd</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	EC/OC
GB0013R	x	b	b	x	x	-	-	b	-	b	b	b	-	-	-	-
GB0014R	x	-	b	b	x	b	-	b	x	b	b	b	x	-	-	-
GB0015R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
GB0031R	x	-	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0033R	x	-	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0036R	-	x	-	-	x	-	-	-	-	-	-	-	-	x	x	-
GB0037R	x	x	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0038R	x	x	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0039R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
GB0043R	x	x	-	-	x	-	-	-	-	-	-	-	-	x	-	-
GB0045R	x	-	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0048R	-	y	x	x	-	-	y	y	x	-	y	y	x	y	x	-
GB0049R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
GB0050R	x	-	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0051R	-	-	-	-	x	-	-	-	-	-	-	-	-	-	-	-
GB0053R	-	-	-	-	y	-	-	-	-	-	-	-	-	-	-	-
GR0002R	-	-	-	-	-	-	-	-	-	-	-	-	-	b	-	-
HR0002R	-	-	-	c	-	-	-	-	c	-	-	-	-	-	-	-
HU0002R	x	x	x	x	x	b	x	x	x	b	x	x	x	y	c	-
IE0001R	x	x	x	x	ie	x	-	-	-	x	-	-	x	-	-	-
IE0005R	-	-	x	y	-	-	x	-	y	-	x	-	-	-	-	-
IE0006R	-	-	x	-	-	-	x	-	-	-	x	-	-	-	-	-
IE0008R	-	-	-	-	-	-	x	-	-	-	x	-	-	-	-	-
IE0009R	-	-	-	x	ie	-	-	-	x	-	-	-	x	-	-	-
IE0031R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
IS0091R	-	-	-	-	-	-	x	-	-	-	-	-	-	-	-	-
IT0001R	-	b	b	-	x	-	b	-	x	-	b	-	-	x	-	-
IT0004R	x	x	x	x	-	-	x	-	x	-	x	-	x	-	x	c
LT0015R	x	x	x	x	x	x	-	-	x	x	-	-	x	-	-	-
LV0010R	x	x	x	x	x	x	-	-	-	x	x	-	-	y	y	-
LV0016R	-	b	-	-	b	b	-	-	-	b	b	-	-	-	-	-
MD0013R	-	c	-	-	-	-	-	-	-	-	-	-	-	-	-	-
NL0007R	-	y	-	-	y	-	-	-	-	-	-	-	-	x	-	-
NL0009R	x	x	b	-	x	-	-	-	-	-	b	-	-	x	y	-
NL0010R	x	b	b	-	x	-	-	-	-	-	b	-	-	x	y	-
NL0091R	-	y	-	c	x	-	-	-	c	-	b	x	-	y	y	-
NL0644R	-	c	-	-	c	-	-	-	-	-	-	-	-	c	-	-
NO0001R	-	-	-	x	-	-	-	-	x	-	-	-	x	-	-	-
NO0002R	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	c
NO0002R;NO0001R	x	x	x	-	x	x	x	x	-	x	x	x	-	x	x	-
NO0015R	x	x	x	x	x	-	-	-	x	-	-	-	x	-	-	-
NO0039R	x	x	x	x	x	-	-	-	x	-	-	-	x	y	y	c
NO0043R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
NO0052R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
NO0055R	-	b	b	-	-	-	-	-	-	-	-	-	-	-	-	-
NO0056R	x	x	x	x	x	x	x	x	x	x	x	-	x	y	y	c
PL0002R	x	x	x	x	x	x	x	-	x	x	x	-	x	-	-	-
PL0004R	x	x	x	c	x	x	x	-	c	x	x	-	-	-	-	-
PL0005R	x	x	x	x	x	-	-	-	x	x	y	y	x	x	y	c
PT0003R	-	-	-	b	-	-	-	-	-	-	-	-	-	-	-	-
PT0004R	-	-	-	b	-	-	-	-	-	-	-	-	-	-	-	-
RU0001R	-	b	-	-	-	-	-	-	-	-	-	-	-	-	-	-
RU0018R	-	-	-	x	-	-	-	-	x	-	x	-	x	-	-	-
RU0020R	-	c	-	-	-	-	-	-	x	-	c	-	x	-	-	-
SE0002R	-	-	-	-	-	-	-	-	-	-	-	-	x	-	-	-
SE0005R	x	x	x	x	x	x	x	x	x	x	x	x	-	c	c	-
SE0008R	-	b	b	-	b	-	-	-	-	-	-	-	-	-	-	-
SE0011R	-	-	-	-	-	-	-	-	-	-	-	-	-	x	x	-
SE0012R	x	c	c	c	c	c	c	c	c	c	c	c	-	x	x	-
SE0013R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
SE0014R	-	-	-	-	-	-	-	-	-	-	-	-	-	y	y	-
SE0014R;SE0002R	x	x	x	x	x	x	x	x	x	x	x	x	-	-	-	-
SE0020R;SE0011R	x	x	x	x	x	x	x	x	x	x	x	x	-	-	-	-
SE0032R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
SE0035R	x	-	-	-	-	-	-	-	-	-	-	-	-	b	-	-
SE0039R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
SI0008R	x	x	x	x	x	x	-	-	x	x	-	-	x	x	x	c
SI0031R	x	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
SK0004R	x	b	b	-	-	-	b	-	-	-	-	-	x	x	-	-
SK0005R	-	b	b	b	b	-	b	-	b	-	-	-	-	-	-	-
SK0006R	x	x	x	x	x	-	-	-	x	-	y	-	x	y	-	-
SK0007R	x	-	-	x	-	-	-	-	x	-	-	-	x	y	-	-

**Table S2.** Annual total emissions (Tg) in the EU27+UK+EFTA countries. Numbers from EMEP status report 1/2021.

Annual total emissions							
year	SOx	NOx	NH <sub>3</sub>	NMVOC	PM <sub>2.5</sub>	PM10	PMcoarse
2000	10047	13696	4369	12672	1924	2878	954
2001	9629	13493	4330	12181	1874	2841	967
2002	9123	13194	4264	11644	1761	2712	951
2003	8690	13050	4233	11245	1807	2761	954
2004	8161	12860	4192	10844	1753	2712	959
2005	7740	12673	4139	10536	1737	2666	929
2006	7482	12352	4108	10263	1696	2626	931
2007	7074	11991	4129	9871	1673	2578	906
2008	5456	11116	4016	9449	1650	2538	888
2009	4546	10206	3941	8789	1579	2402	822
2010	4242	9981	3911	8704	1601	2407	805
2011	4117	9590	3878	8293	1484	2274	790
2012	3731	9292	3865	8117	1487	2233	746
2013	3230	8867	3862	7866	1463	2204	740
2014	2954	8525	3896	7646	1339	2067	728
2015	2786	8343	3941	7606	1350	2078	729
2016	2332	8027	3943	7540	1326	2046	720
2017	2287	7838	3957	7607	1327	2053	726
2018	2134	7535	3923	7482	1285	2012	728
2019	1901	7160	3839	7377	1245	1967	722

  

Emission reduction							
Periods	SOx	NOx	NH <sub>3</sub>	NMVOC	PM <sub>2.5</sub>	PM10	PMcoarse
2000-2019 total	81%	48%	12%	42%	35%	32%	24%
pr year	4.3 %	2.5 %	0.6 %	2.2 %	1.9 %	1.7 %	1.3 %
2000-2010 total	58%	27%	10%	31%	17%	16%	16%
pr year	5.8 %	2.7 %	1.0 %	3.1 %	1.7 %	1.6 %	1.6 %
2010-2019 total	55%	28%	2%	15%	22%	18%	10%
pr year	5.5 %	2.8 %	0.2 %	1.5 %	2.2 %	1.8 %	1.0 %
2005-2019 total	75%	44%	7%	30%	28%	26%	22%
pr year	5.4 %	3.1 %	0.5 %	2.1 %	2.0 %	1.9 %	1.6 %

**Table S3.** Absolute and relative change and corresponding 95% confidence intervals in observed and modelled annual and seasonal aggregated  $SO_2$  concentrations for the different time periods. The number of sites with a significant outcome is provided.

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	49	47	49	-0.067	(-0.084, -0.05)	-0.087	(-0.107, -0.066)	-3.87	(-4.21, -3.53)	-5.15	(-5.46, -4.85)
	autumn	49	37	48	-0.050	(-0.065, -0.035)	-0.085	(-0.106, -0.064)	-3.53	(-3.92, -3.13)	-5.08	(-5.37, -4.79)
	spring	49	46	49	-0.069	(-0.085, -0.052)	-0.080	(-0.099, -0.060)	-4.13	(-4.43, -3.83)	-5.13	(-5.39, -4.87)
	summer	49	34	49	-0.043	(-0.057, -0.029)	-0.059	(-0.077, -0.041)	-3.02	(-3.58, -2.45)	-5.00	(-5.36, -4.63)
	winter	51	45	50	-0.100	(-0.125, -0.075)	-0.123	(-0.151, -0.094)	-4.06	(-4.34, -3.78)	-4.99	(-5.29, -4.7)
2000-2010	all	67	31	59	-0.078	(-0.102, -0.055)	-0.116	(-0.141, -0.091)	-3.55	(-4.34, -2.77)	-5.42	(-5.96, -4.88)
2005-2019	all	58	41	55	-0.054	(-0.067, -0.041)	-0.066	(-0.082, -0.049)	-4.24	(-4.68, -3.8)	-5.29	(-5.62, -4.95)
2010-2019	all	61	37	43	-0.049	(-0.061, -0.036)	-0.055	(-0.071, -0.04)	-4.60	(-5.78, -3.43)	-6.37	(-7.12, -5.62)

**Table S4.** As Table S3, but for  $SO_4^{2-}$  concentrations in aerosols.

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	39	38	39	-0.074	(-0.087, -0.062)	-0.065	(-0.076, -0.053)	-3.20	(-3.46, -2.93)	-3.81	(-4.04, -3.58)
	autumn	37	34	37	-0.062	(-0.073, -0.05)	-0.057	(-0.067, -0.046)	-3.08	(-3.4, -2.76)	-3.59	(-3.81, -3.38)
	spring	38	36	38	-0.085	(-0.098, -0.072)	-0.065	(-0.075, -0.055)	-3.46	(-3.72, -3.19)	-4.03	(-4.21, -3.84)
	summer	38	36	38	-0.082	(-0.102, -0.063)	-0.078	(-0.097, -0.059)	-3.15	(-3.46, -2.85)	-4.20	(-4.5, -3.91)
	winter	40	29	34	-0.064	(-0.08, -0.049)	-0.056	(-0.066, -0.046)	-2.75	(-3.22, -2.28)	-3.21	(-3.44, -2.97)
2000-2010	all	54	15	43	-0.068	(-0.085, -0.051)	-0.094	(-0.109, -0.079)	-2.51	(-2.95, -2.07)	-4.19	(-4.52, -3.86)
2005-2019	all	43	35	42	-0.067	(-0.079, -0.055)	-0.054	(-0.063, -0.046)	-3.48	(-3.88, -3.09)	-4.01	(-4.25, -3.77)
2010-2019	all	46	20	32	-0.053	(-0.068, -0.038)	-0.044	(-0.055, -0.034)	-3.43	(-4.11, -2.75)	-4.26	(-4.91, -3.62)

**Table S5.** As Table S35, but for wet deposition of  $SO_4^{2-}$ .

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^2 \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	49	40	49	-29.5	(-34.8, -24.2)	-48.1	(-56.9, -39.4)	-3.14	(-3.46, -2.82)	-4.27	(-4.5, -4.04)
	autumn	46	27	42	-26.7	(-32.3, -21.1)	-45	(-54.6, -35.5)	-3.00	(-3.49, -2.51)	-4.25	(-4.52, -3.99)
	spring	45	30	45	-32.7	(-39.1, -26.2)	-49.2	(-59.7, -38.6)	-3.23	(-3.6, -2.86)	-4.19	(-4.41, -3.98)
	summer	47	31	45	-36.5	(-44.5, -28.5)	-52.6	(-63.0, -42.2)	-2.95	(-3.35, -2.54)	-4.20	(-4.46, -3.94)
	winter	47	25	44	-19.5	(-24.5, -14.5)	-41.4	(-49.5, -33.3)	-2.87	(-3.34, -2.4)	-3.97	(-4.21, -3.73)
2000-2010	all	54	28	44	-46.9	(-56.6, -37.1)	-68.5	(-81.4, -55.7)	-4.44	(-5.15, -3.74)	-5.29	(-5.68, -4.9)
2005-2019	all	53	30	50	-22.9	(-27.9, -17.9)	-36.3	(-42.4, -30.2)	-2.87	(-3.51, -2.22)	-4.53	(-4.79, -4.27)
2010-2019	all	60	15	40	-20.9	(-27.1, -14.6)	-34.0	(-40.6, -27.3)	-2.86	(-3.78, -1.93)	-5.08	(-5.56, -4.59)

**Table S6.** As Table S5, but for NO<sub>2</sub> concentrations in air

Period	Seasons	Number of sites			Absolute change per year (ug m <sup>-3</sup> yr <sup>-1</sup> )				Relative change per year (% yr <sup>-1</sup> )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	59	44	58	-0.138	(-0.167, -0.109)	-0.193	(-0.238, -0.149)	-1.67	(-2.05, -1.29)	-2.24	(-2.42, -2.06)
	autumn	58	40	57	-0.156	(-0.191, -0.121)	-0.191	(-0.239, -0.144)	-1.75	(-2.19, -1.31)	-2.20	(-2.38, -2.02)
	spring	58	36	54	-0.125	(-0.154, -0.096)	-0.161	(-0.204, -0.118)	-1.43	(-2.02, -0.84)	-2.07	(-2.27, -1.88)
	summer	58	38	54	-0.097	(-0.121, -0.073)	-0.134	(-0.177, -0.091)	-1.59	(-1.96, -1.23)	-1.89	(-2.13, -1.64)
	winter	60	29	57	-0.158	(-0.197, -0.120)	-0.259	(-0.306, -0.212)	-1.55	(-1.92, -1.18)	-2.35	(-2.52, -2.18)
2000-2010	all	63	10	41	-0.098	(-0.146, -0.049)	-0.183	(-0.234, -0.132)	-1.23	(-1.83, -0.64)	-1.97	(-2.35, -1.6)
2005-2019	all	63	48	62	-0.167	(-0.200, -0.134)	-0.197	(-0.24, -0.154)	-2.13	(-2.60, -1.66)	-2.58	(-2.80, -2.36)
2010-2019	all	65	36	52	-0.193	(-0.232, -0.154)	-0.176	(-0.215, -0.138)	-2.86	(-3.48, -2.24)	-2.65	(-2.92, -2.38)

**Table S7.** As Table S3, but for sum HNO<sub>3</sub>+NO<sub>3</sub><sup>-</sup> concentrations in air and aerosols.

Period	Seasons	Number of sites			Absolute change per year (ug m <sup>-3</sup> yr <sup>-1</sup> )				Relative change per year (% yr <sup>-1</sup> )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	25	17	25	-0.008	(-0.011, -0.005)	-0.013	(-0.016, -0.01)	-1.60	(-2.0, -1.2)	-2.13	(-2.33, -1.94)
	autumn	24	16	16	-0.009	(-0.013, -0.006)	-0.014	(-0.019, -0.01)	-1.89	(-2.43, -1.35)	-2.20	(-2.48, -1.93)
	spring	25	13	21	-0.01	(-0.013, -0.007)	-0.015	(-0.018, -0.011)	-1.69	(-2.08, -1.29)	-2.24	(-2.46, -2.02)
	summer	25	17	23	-0.005	(-0.007, -0.003)	-0.012	(-0.015, -0.009)	-1.43	(-1.86, -1.0)	-2.27	(-2.43, -2.1)
	winter	27	7	16	-0.009	(-0.014, -0.003)	-0.014	(-0.018, -0.01)	-1.21	(-1.75, -0.66)	-1.93	(-2.26, -1.6)
2000-2010	all	33	7	11	-0.006	(-0.011, -0.001)	-0.017	(-0.022, -0.012)	-0.54	(-1.55, 0.47)	-2.12	(-2.66, -1.57)
2005-2019	all	31	18	26	-0.011	(-0.014, -0.008)	-0.014	(-0.017, -0.011)	-2.29	(-2.74, -1.84)	-2.51	(-2.8, -2.22)
2010-2019	all	29	16	9	-0.015	(-0.019, -0.010)	-0.01	(-0.014, -0.007)	-3.38	(-4.26, -2.5)	-2.16	(-2.58, -1.75)

**Table S8.** As Table S3, but for HNO<sub>3</sub> concentrations in air.

Period	Seasons	Number of sites			Absolute change per year (ug m <sup>-3</sup> yr <sup>-1</sup> )				Relative change per year (% yr <sup>-1</sup> )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	6	4	6	-0.002	(-0.004, -0.001)	-0.003	(-0.005, -0.002)	-1.94	(-2.77, -1.11)	-2.35	(-2.64, -2.07)
	autumn	6	3	6	-0.002	(-0.003, -0.000)	-0.003	(-0.004, -0.001)	-2.09	(-3.22, -0.97)	-2.49	(-2.99, -1.99)
	spring	6	4	6	-0.002	(-0.004, -0.001)	-0.003	(-0.005, -0.002)	-2.05	(-3.15, -0.95)	-2.68	(-3.17, -2.19)
	summer	6	3	6	-0.002	(-0.004, -0.001)	-0.004	(-0.006, -0.002)	-1.83	(-2.97, -0.7)	-1.85	(-2.06, -1.63)
	winter	6	3	4	-0.003	(-0.005, -0.001)	-0.002	(-0.003, -0.001)	-2.15	(-2.92, -1.37)	-2.80	(-3.71, -1.89)
2000-2010	all	10	3	4	-0.006	(-0.012, 0.000)	-0.005	(-0.007, -0.003)	-1.63	(-3.86, 0.61)	-3.33	(-4.17, -2.5)
2005-2019	all	12	7	9	-0.004	(-0.005, -0.003)	-0.003	(-0.004, -0.001)	-2.48	(-3.16, -1.8)	-2.26	(-2.85, -1.67)
2010-2019	all	16	4	3	-0.005	(-0.007, -0.003)	-0.002	(-0.003, -0.0)	-4.25	(-5.65, -2.86)	-0.67	(-2.31, 0.97)

**Table S9.** As Table S3, but for  $\text{NO}_3^-$  concentrations in aerosols.

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	21	13	15	-0.009	(-0.012, -0.006)	-0.014	(-0.018, -0.009)	-2.01	(-2.49, -1.53)	-2.53	(-2.86, -2.2)
	autumn	20	11	9	-0.01	(-0.013, -0.007)	-0.014	(-0.021, -0.007)	-2.41	(-2.8, -2.01)	-2.42	(-2.83, -2.01)
	spring	20	12	15	-0.011	(-0.017, -0.005)	-0.012	(-0.018, -0.007)	-1.89	(-2.69, -1.09)	-2.21	(-2.97, -1.45)
	summer	20	8	13	-0.004	(-0.006, -0.002)	-0.01	(-0.014, -0.006)	-1.55	(-2.17, -0.94)	-2.7	(-2.98, -2.43)
	winter	20	8	5	-0.012	(-0.018, -0.005)	-0.012	(-0.016, -0.008)	-1.85	(-2.77, -0.93)	-2.01	(-2.55, -1.47)
2000-2010	all	20	3	9	-0.006	(-0.013, 0.001)	-0.02	(-0.028, -0.012)	-1.54	(-2.8, -0.27)	-3.01	(-3.99, -2.03)
2005-2019	all	26	11	16	-0.011	(-0.015, -0.006)	-0.014	(-0.018, -0.009)	-2.32	(-2.84, -1.81)	-2.77	(-3.21, -2.33)
2010-2019	all	32	3	6	-0.010	(-0.015, -0.004)	-0.009	(-0.013, -0.005)	-1.98	(-2.85, -1.11)	-1.46	(-2.23, -0.68)

**Table S10.** As Table S3, but for wet deposition of  $\text{NO}_3^-$ .

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^2 \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	46	21	44	-12.1	(-15.7, -8.5)	-26.4	(-31.3, -21.4)	-1.36	(-1.74, -0.98)	-2.35	(-2.49, -2.21)
	autumn	45	12	28	-12.8	(-16.6, -9.0)	-25.6	(-31.6, -19.7)	-1.53	(-2.01, -1.05)	-2.36	(-2.59, -2.13)
	spring	43	17	33	-14.2	(-20.5, -7.9)	-27.8	(-35.2, -20.3)	-1.11	(-1.74, -0.48)	-2.33	(-2.54, -2.12)
	summer	45	10	33	-11.3	(-16.3, -6.2)	-30.4	(-37.4, -23.3)	-0.02	(-1.83, 1.78)	-2.17	(-2.44, -1.9)
	winter	45	6	25	-8.3	(-12.4, -4.3)	-19.9	(-23.5, -16.4)	-1.16	(-1.69, -0.63)	-2.23	(-2.46, -2.0)
2000-2010	all	45	8	15	-13.2	(-20.8, -5.5)	-26.4	(-34.3, -18.5)	-1.12	(-2.22, -0.03)	-2.24	(-2.65, -1.83)
2005-2019	all	50	15	40	-10.0	(-14.8, -5.2)	-24.0	(-27.7, -20.3)	-0.53	(-2.04, 0.98)	-2.62	(-2.8, -2.44)
2010-2019	all	58	7	19	-13.3	(-17.6, -8.9)	-23.6	(-28.3, -18.9)	-1.54	(-2.23, -0.86)	-2.60	(-2.98, -2.22)

**Table S11.** As Table S3, but for sum  $\text{NH}_3 + \text{NH}_4^+$  concentrations in air and aerosols.

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	25	17	18	-0.016	(-0.024, -0.007)	-0.019	(-0.027, -0.012)	-1.45	(-1.99, -0.91)	-1.36	(-1.58, -1.14)
	autumn	24	11	7	-0.015	(-0.025, -0.006)	-0.016	(-0.023, -0.009)	-1.67	(-2.27, -1.08)	-1.32	(-1.61, -1.03)
	spring	25	10	16	-0.017	(-0.029, -0.006)	-0.023	(-0.032, -0.015)	-1.48	(-2.21, -0.75)	-1.46	(-1.71, -1.2)
	summer	25	10	20	-0.009	(-0.018, -0.001)	-0.017	(-0.025, -0.009)	-1.02	(-1.61, -0.44)	-1.44	(-1.71, -1.18)
	winter	28	9	10	-0.018	(-0.029, -0.008)	-0.023	(-0.033, -0.013)	-1.18	(-1.72, -0.64)	-1.14	(-1.47, -0.81)
2000-2010	all	37	6	13	-0.022	(-0.034, -0.01)	-0.024	(-0.033, -0.016)	-1.36	(-2.11, -0.62)	-1.2	(-1.82, -0.58)
2005-2019	all	27	12	12	-0.019	(-0.029, -0.009)	-0.02	(-0.029, -0.011)	-1.90	(-2.55, -1.24)	-1.53	(-1.88, -1.18)
2010-2019	all	27	6	7	-0.017	(-0.026, -0.008)	-0.012	(-0.021, -0.004)	-2.31	(-3.25, -1.36)	-1.12	(-1.75, -0.5)

**Table S12.** As Table S3, but for  $\text{NH}_3$  concentrations in air.

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	8	2	6	0.01	(-0.005, 0.024)	0.006	(-0.006, 0.017)	1.55	(0.22, 2.88)	1.54	(0.42, 2.66)
	autumn	8	1	4	0.008	(-0.005, 0.02)	0.007	(-0.001, 0.015)	0.94	(-0.36, 2.24)	1.49	(0.65, 2.33)
	spring	8	3	3	0.011	(-0.007, 0.03)	0.003	(-0.005, 0.011)	1.76	(-0.32, 3.83)	1.46	(0.09, 2.84)
	summer	8	1	6	0.013	(-0.009, 0.035)	0.009	(-0.002, 0.02)	2.53	(-0.3, 5.36)	2.27	(0.88, 3.65)
	winter	8	1	3	0.007	(-0.001, 0.016)	0.002	(-0.007, 0.011)	4.03	(0.61, 7.44)	1.47	(0.1, 2.84)
2000-2010	all	12	3	4	0.003	(-0.021, 0.028)	0.012	(0.0, 0.025)	3.14	(0.5, 5.78)	1.99	(0.4, 3.57)
2005-2019	all	18	3	7	0.005	(-0.002, 0.012)	0	(-0.009, 0.01)	0.57	(-0.26, 1.39)	0.84	(0.1, 1.59)
2010-2019	all	22	4	5	0.012	(-0.009, 0.032)	0.001	(-0.017, 0.019)	2.8	(-1.35, 6.95)	2.58	(0.67, 4.49)

**Table S13.** As Table S3, but for  $\text{NH}_4^+$  concentrations in aerosols.

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	21	15	20	-0.024	(-0.033, -0.016)	-0.021	(-0.028, -0.014)	-2.61	(-3.01, -2.22)	-2.61	(-2.79, -2.43)
	autumn	19	11	11	-0.023	(-0.031, -0.014)	-0.022	(-0.032, -0.012)	-2.75	(-3.65, -1.85)	-2.58	(-2.8, -2.35)
	spring	20	17	16	-0.032	(-0.043, -0.021)	-0.022	(-0.03, -0.015)	-3.09	(-3.64, -2.55)	-2.74	(-3.17, -2.31)
	summer	20	14	20	-0.017	(-0.023, -0.012)	-0.018	(-0.023, -0.013)	-2.78	(-3.22, -2.35)	-3.39	(-3.62, -3.16)
	winter	22	10	9	-0.019	(-0.035, -0.002)	-0.02	(-0.028, -0.013)	-1.47	(-2.49, -0.45)	-1.94	(-2.43, -1.45)
2000-2010	all	23	9	12	-0.034	(-0.048, -0.02)	-0.036	(-0.047, -0.024)	-3.43	(-4.6, -2.27)	-3.18	(-3.63, -2.73)
2005-2019	all	27	16	23	-0.025	(-0.033, -0.017)	-0.022	(-0.028, -0.016)	-2.9	(-3.41, -2.38)	-3.01	(-3.2, -2.83)
2010-2019	all	31	16	17	-0.029	(-0.041, -0.017)	-0.019	(-0.027, -0.012)	-3.48	(-4.7, -2.26)	-3.22	(-3.9, -2.54)

**Table S14.** As Table S3, but for wet deposition of  $\text{NH}_4^+$ .

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-2} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	44	13	9	-7.4	(-11.8, -3.0)	-4.8	(-8.1, -1.5)	-0.31	(-0.99, 0.37)	-0.40	(-0.71, -0.09)
	autumn	43	10	4	-7.5	(-12.8, -2.3)	-5.2	(-9.3, -1.0)	-0.45	(-1.37, 0.47)	-0.35	(-0.8, 0.11)
	spring	42	5	4	-12.2	(-19.4, -4.9)	-4.7	(-11.5, 2.1)	-0.39	(-1.1, 0.31)	-0.27	(-0.64, 0.11)
	summer	43	9	6	-10.4	(-16.7, -4.1)	-8.0	(-11.6, -4.4)	-0.12	(-1.17, 0.93)	-0.48	(-0.84, -0.12)
	winter	44	5	3	-5.3	(-9.4, -1.3)	-1.7	(-3.8, 0.4)	-0.25	(-0.94, 0.43)	-0.39	(-0.72, -0.06)
2000-2010	all	44	4	3	-9.3	(-17.8, -0.8)	-4.0	(-12.2, 4.2)	-0.36	(-1.24, 0.52)	-0.09	(-0.91, 0.73)
2005-2019	all	52	8	4	-4.8	(-10.7, 1.0)	-3.4	(-6.4, -0.3)	-0.12	(-0.95, 0.71)	-0.43	(-0.78, -0.08)
2010-2019	all	62	3	4	-2.7	(-8.6, 3.1)	-5.8	(-11.1, -0.4)	0.90	(-0.95, 2.75)	-0.47	(-1.04, 0.11)



**Table S15.** As Table S3, but for  $PM_{10}$  concentrations

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	37	29	36	-0.36	(-0.43, -0.29)	-0.29	(-0.32, -0.25)	-1.83	(-2.09, -1.57)	-1.95	(-2.11, -1.79)
	autumn	35	22	29	-0.34	(-0.42, -0.26)	-0.30	(-0.34, -0.26)	-1.81	(-2.1, -1.51)	-1.91	(-2.09, -1.74)
	spring	34	24	32	-0.39	(-0.47, -0.31)	-0.30	(-0.35, -0.25)	-1.84	(-2.11, -1.56)	-2.10	(-2.29, -1.9)
	summer	36	27	31	-0.34	(-0.42, -0.25)	-0.32	(-0.37, -0.26)	-1.72	(-2.09, -1.35)	-2.25	(-2.47, -2.03)
	winter	34	18	20	-0.36	(-0.47, -0.25)	-0.26	(-0.31, -0.21)	-1.73	(-2.07, -1.4)	-1.64	(-1.83, -1.45)
2000-2010	all	36	14	24	-0.46	(-0.56, -0.36)	-0.35	(-0.46, -0.24)	-2.36	(-2.77, -1.95)	-2.40	(-2.97, -1.83)
2005-2019	all	54	29	38	-0.33	(-0.41, -0.25)	-0.23	(-0.27, -0.19)	-1.82	(-2.23, -1.41)	-1.79	(-2.07, -1.51)
2010-2019	all	56	17	23	-0.32	(-0.42, -0.21)	-0.16	(-0.21, -0.11)	-1.84	(-2.45, -1.23)	-1.38	(-1.81, -0.95)

**Table S16.** As Table S3, but for  $PM_{2.5}$  concentrations

Period	Seasons	Number of sites			Absolute change per year ( $\mu\text{g m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
2000-2019	all	19	16	19	-0.29	(-0.39, -0.2)	-0.26	(-0.31, -0.22)	-2.42	(-2.84, -2.0)	-2.52	(-2.71, -2.32)
	autumn	18	12	16	-0.26	(-0.36, -0.15)	-0.26	(-0.33, -0.18)	-2.22	(-2.71, -1.74)	-2.29	(-2.5, -2.07)
	spring	19	14	19	-0.32	(-0.41, -0.24)	-0.27	(-0.31, -0.22)	-2.47	(-2.91, -2.03)	-2.69	(-2.88, -2.51)
	summer	18	14	17	-0.26	(-0.33, -0.19)	-0.30	(-0.36, -0.23)	-2.36	(-2.91, -1.82)	-2.81	(-3.16, -2.47)
	winter	18	12	12	-0.36	(-0.54, -0.18)	-0.21	(-0.25, -0.17)	-2.29	(-2.74, -1.85)	-2.08	(-2.35, -1.82)
2000-2010	all	20	7	12	-0.36	(-0.5, -0.21)	-0.36	(-0.46, -0.26)	-2.65	(-3.56, -1.73)	-2.93	(-3.58, -2.27)
2005-2019	all	36	23	28	-0.33	(-0.42, -0.23)	-0.21	(-0.25, -0.17)	-2.66	(-3.16, -2.16)	-2.28	(-2.55, -2.01)
2010-2019	all	43	20	19	-0.34	(-0.43, -0.25)	-0.18	(-0.23, -0.13)	-3.14	(-3.7, -2.57)	-2.21	(-2.67, -1.74)

**Table S18.** As Table S3, but for EC in  $PM_{2.5}$  concentrations and period 2010-2019

Seasons	Number of sites			Absolute change per year ( $\mu\text{gC m}^{-3} \text{yr}^{-1}$ )				Relative change per year ( $\% \text{yr}^{-1}$ )			
	total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
all	15	11	12	-0.019	(-0.031, -0.008)	-0.014	(-0.021, -0.007)	-4.49	(-5.25, -3.73)	-3.81	(-4.59, -3.03)
Winter	15	6	4	-0.029	(-0.053, -0.006)	-0.019	(-0.032, -0.006)	-4.27	(-5.49, -3.06)	-3.10	(-4.21, -1.98)
Spring	15	5	8	-0.012	(-0.018, -0.006)	-0.013	(-0.018, -0.009)	-3.88	(-4.67, -3.09)	-3.86	(-4.92, -2.8)
Summer	15	8	10	-0.011	(-0.015, -0.007)	-0.008	(-0.013, -0.004)	-4.73	(-5.71, -3.74)	-3.71	(-4.59, -2.82)
Autumn	15	8	11	-0.023	(-0.036, -0.01)	-0.018	(-0.027, -0.009)	-4.96	(-6.03, -3.9)	-4.25	(-5.15, -3.35)

**Table S19.** As Table S3, but for OC in PM<sub>2.5</sub> concentrations and period 2010-2019

Seasons	Number of sites			Absolute change per year (ugC m <sup>-3</sup> yr <sup>-1</sup> )				Relative change per year (% yr <sup>-1</sup> )			
	total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
all	15	2	1	-0.077	(-0.132, -0.022)	-0.009	(-0.019, 0.0)	-2.40	(-3.23, -1.57)	-0.59	(-1.21, 0.03)
Winter	15	6	1	-0.148	(-0.249, -0.047)	-0.018	(-0.032, -0.004)	-4.19	(-5.16, -3.22)	-1.09	(-2.03, -0.15)
Spring	15	1	0	-0.067	(-0.104, -0.03)	-0.013	(-0.023, -0.003)	-2.18	(-3.68, -0.67)	-0.74	(-1.94, 0.47)
Summer	15	0	1	0	(-0.024, 0.023)	0.019	(-0.003, 0.041)	-0.03	(-1.19, 1.12)	2.13	(0.44, 3.83)
Autumn	15	1	0	-0.098	(-0.16, -0.037)	-0.020	(-0.041, 0.0)	-3.03	(-4.43, -1.62)	-1.16	(-2.47, 0.15)

**Table S20.** Absolute and relative change and the corresponding 95% confidence intervals in observed and modelled annual aggregated OC/PM<sub>2.5</sub> and EC/PM<sub>2.5</sub> ratios at 15 sites across Europe for 2010–2019. The number of sites with a significant outcome is provided.

EC/PM <sub>2.5</sub>						
	Number of sites		Absolute change per year (ugC m <sup>-3</sup> yr <sup>-1</sup> )		Relative change per year (% yr <sup>-1</sup> )	
	total	sign.	abs	Conf. interv.	%yr <sup>-1</sup>	Conf. interv.
Model	15	5	-0.0006	(-0.0009, -0.0004)	-1.71	(-2.32, -1.1)
Obs.	15	4	-0.0009	(-0.0012, -0.0006)	-2.41	(-2.96, -1.85)

  

OC/PM <sub>2.5</sub>						
	Number of sites		Absolute change per year (ugC m <sup>-3</sup> yr <sup>-1</sup> )		Relative change per year (% yr <sup>-1</sup> )	
	total	sign.	abs	Conf. interv.	% yr <sup>-1</sup>	Conf. interv.
Model	15	7	0.0029	(0.0018, 0.004)	2.35	(1.3, 3.4)
Obs.	15	2	0.0001	(-0.003, 0.0032)	0.19	(-1.24, 1.62)

**Table S21.** As Table S3, but for O<sub>3</sub> metrics based on stations north and south of 49°N for the period 2000-2019. Results are given for the 75th and 99th percentile of daily maximum ozone, as well as for SOMO35 and AOT40 for crops (AOT40c) forests (AOT40f). Note that here we use the EU definition of AOT40.

		Number of sites			Absolute change per year (ppb yr <sup>-1</sup> )				Relative change per year (% yr <sup>-1</sup> )			
		total	sign.(obs.)	sign.(mod.)	obs.	Conf. Interval	mod.	Conf. Interval	obs.	Conf. Interval	mod.	Conf. Interval
N of 49N	75perc.	52	19	30	-0.15	(-0.19, -0.10)	-0.13	(-0.14, -0.10)	-0.31	(-0.40, -0.21)	-0.26	(-0.30, -0.22)
	99perc.	52	13	39	-0.41	(-0.48, -0.33)	-0.34	(-0.37, -0.30)	-0.58	(-0.68, -0.47)	-0.54	(-0.59, -0.49)
	SOMO35	55	9	33	-18.3	(-26.3, -11.2)	-17.92	(-21.1, -14.7)	-0.54	(-1.41, 0.021)	-0.85	(-1.02, -0.70)
	AOT40c	55	12	42	-71.6	(-91.2, -52.4)	-153	(-175, -132)	-1.35	(-2.03, -0.73)	-2.10	(-2.32, -1.88)
	AOT40f	55	9	43	-121	(-156., -85.5)	-220	(-250, -191)	-1.19	(-1.86, -0.47)	-1.75	(-1.92, -1.56)
S of 49N	75perc	40	16	38	-0.20	(-0.27, -0.13)	-0.26	(-0.27, -0.23)	-0.35	(-0.46, -0.21)	-0.46	(-0.49, -0.43)
	99perc	40	18	37	-0.48	(-0.59, -0.36)	-0.51	(-0.54, -0.46)	-0.57	(-0.71, -0.43)	-0.70	(-0.74, -0.65)
	SOMO35	35	14	31	-36.7	(-54.9, -18.6)	-40.6	(-45.3, -36.2)	-0.77	(-1.27, -0.28)	-1.12	(-1.23, -1.01)
	AOT40c	35	13	33	-190	(-247., -128.)	-286	(-313., -260.)	-1.49	(-1.97, -1.02)	-1.98	(-2.17, -1.80)
	AOT40f	35	16	31	-312	(-422., -196.)	-480	(-537., -425.)	-1.36	(-1.87, -0.85)	-1.74	(-1.94, -1.55)