

Contributions of Nordic anthropogenic emissions on air pollution and premature mortality over the Nordic region and the Arctic

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Abstract

This modelling study presents the sectoral contributions of anthropogenic emissions in the four Nordic countries; Denmark, Finland, Norway and Sweden, on air pollution levels and the associated health impacts and costs over the Nordic and the Arctic region for the year 2015. The Danish Eulerian Hemispheric Model (DEHM) has been used on a 50 km resolution over Europe in tagged mode in order to calculate the response of a 30% reduction of each emission sector in each Nordic country individually. The emission sectors considered in the study were energy production, non-industrial/commercial heating, industry, traffic, off-road mobile sources, and waste management/agriculture. In total, 28 simulations were carried out. Following the air pollution modelling, the Economic Valuation of Air Pollution (EVA) model has been used to calculate the associated premature mortality and their costs. Results showed that more than 80% of the PM_{2.5} concentration was attributed to transport from outside these four countries, implying an effort outside the Nordic region in order to decrease the pollutant levels over the area. The leading emission sector in each country was found to be non-industrial combustion (contributing by more than 60% to the total PM_{2.5} mass coming from the country itself), except for Sweden, where industry contributed to PM_{2.5} with a comparable amount as non-industrial combustion. In addition to non-industrial combustion, the next most important source categories were industry, agriculture and traffic. The main chemical constituent of PM_{2.5} concentrations that comes from the country itself is calculated to be organic carbon in all countries, which suggested that non-industrial wood burning was the dominant national source of pollution in the Nordic countries. We have estimated the total number of premature mortality cases due to air pollution to be around 4 000 in Denmark and Sweden and around 2 000 in Finland and Norway. These premature mortality cases led to a total cost of 7 billion Euros in the selected Nordic countries. The assessment of the related premature mortality and associated cost estimates suggested that non-industrial combustion, together with industry and traffic, will be the main sectors to be targeted in emission mitigation strategies in the future.

49 Introduction

50

51 Air pollution is the world's largest single environmental health risk (WHO, 2014), estimated to be
52 responsible for 3.7 million premature deaths in 2012 from urban and rural sources worldwide. In
53 Europe, recent results (Andersson et al., 2009; Brandt et al., 2013a; 2013b; Geels et al., 2015; Im et
54 al., 2018a; Liang et al., 2018; Solazzo et al., 2018) show that outdoor air pollution causes ~500 000
55 premature deaths in Europe. Brandt et al. (2013a) calculated that due to exposure to ambient air
56 pollution, there were around 3.500 premature deaths in 2011 in Denmark alone. Lehtomäki et al.
57 (2018) have recently evaluated that ambient air pollution caused approximately 2000 premature
58 deaths in Finland in 2015. Other studies have made assessments for some of the Nordic countries
59 (Denmark, Sweden and Finland) with estimates ranging from 6500 to 9500 for the year 2000 (Geels
60 et al., 2014; Watkiss et al., 2005, Karvosenoja et al., 2010, respectively). Kukkonen et al. (2018)
61 and Forsberg et al. (2015) have concluded that long-range transported fine particulate matter
62 dominates the health effects in the Nordic countries, with largest contribution to long-term effects in
63 Sweden originating from south-western Europe, while the largest contribution to short-term
64 exposure originates from south-eastern Europe (Jönsson et al. 2013).

65

66 Air pollution is a transboundary problem covering global, regional, national and local sources,
67 leading to large spatial variability and therefore to large differences in the geographical distribution
68 of human exposure to air pollution (Im et al., 2018a,b). In the Nordic countries, there are large
69 spatial differences in air pollution levels because of long-range transported and polluted air masses
70 especially from the south and east as well as due to the degree of urbanization. There are also local
71 differences depending on wind direction and distance from local emission sources such as road
72 transport, power plants and industry (Brandt et al., 2013a). Furthermore, the widespread use of
73 domestic wood stoves in the Nordic countries represents a special challenge for exposure to air
74 pollution (Kukkonen et al., 2019) , where e.g. more than a third of the health impacts from Danish
75 emissions are due to smoke from wood stoves. International ship traffic is also a significant source
76 of air pollution and health impacts in highly trafficked areas of the Baltic and North Seas (Brandt et
77 al., 2013b; Jalkanen et al., 2016, Johansson et al., 2017). Based on simulations for the period 1997-
78 2003, Andersson et al. (2009) calculated that Sweden contributed to 1.4% of the European Primary
79 PM_{2.5} (PPM_{2.5}) mass concentrations while Denmark, Finland and Norway were responsible for 4%
80 of European PPM_{2.5}. Contribution to secondary inorganic aerosol (SIA) levels were much smaller
81 (0.5% from Sweden and 1.4% from Denmark, Finland and Norway). They also calculated a death
82 rate increase of 2 and 3% due to exposure to PPM_{2.5} and SIA, respectively, in Europe due to
83 emissions from Denmark, Finland, Norway and Sweden.

84

85 The external (or indirect) costs to society related to health impacts from air pollution are substantial.
86 In the whole of Europe, the total external costs have been estimated to be approx. 800 billion Euros
87 per year and in Denmark alone the external costs are nearly 4 billion Euro per year (Brandt et al.,
88 2013a). In a more recent study, Im et al. (2018a), using a multi-model ensemble of 14 chemistry
89 transport models (CTM), estimated that ambient air pollution in Europe in 2010 was responsible for
90 414 000±100 000 premature deaths, leading to a cost of 300 billion Euros. The study also showed
91 that a 20% decrease of anthropogenic emissions in Europe source could avoid 47 000 premature
92 deaths in Europe, while a similar reduction in the U.S. would avoid around 1 000 premature deaths
93 in Europe due to long-range transport.

94

95 The Nordic countries are generally characterized among the EU countries with low air pollution
96 levels (EEA, 2018). PM_{2.5} levels are below the EU legislated limit value of 25 µg m⁻³ as well as the

97 WHO limit value of $10 \mu\text{g m}^{-3}$ (EEA, 2018). However, there are still large impacts of air pollution
98 on human health and climate in the region itself (Arctic Council, 2011; Brandt et al., 2013a;
99 Forsberg et al., 2015), as well as over the Arctic (Sand et al., 2015). The Task Force on Short Lived
100 Climate Forcers of the Arctic Council reported that measures aimed at decreasing Nordic emissions
101 will have positive health effects for communities exposed to air pollution. In a recent study, Sand et
102 al. (2015) showed that although the largest Arctic warming source is from Asian emissions, the
103 Arctic is most sensitive, per unit mass emitted, to Short Lived Climate Forcers (SLCF) emissions
104 from a small number of activities within the Arctic nations themselves.

105
106 The aim of the study is to quantify the contributions of the main emission sectors in each of the
107 Nordic countries to air pollutant levels and their impacts on premature mortality and associated
108 costs in the Nordic region and the Arctic. This will help us identify the emission sectors in these
109 Nordic countries that should be targeted for mitigation to decrease the air pollution and exposure
110 levels in the Nordic countries, that are originated within the region. In addition, we also aim to give
111 a first estimate of the impact of transported air pollution on the Arctic population. In order to
112 achieve this, we have coupled the Danish Eulerian Hemispheric Model (DEHM) to the Economic
113 Valuation of Air Pollution (EVA) model and conducted a number of perturbation simulations
114 targeting different emission sectors in the four Nordic countries; Denmark, Finland, Norway and
115 Sweden, for the year 2015. Year 2015 is selected to be in agreement with the ongoing Coupled
116 Model Intercomparison Project Phase 6 (CMIP6: Eyring et al., 2016), where the current year is
117 2015. As the present study will also look at the impacts in the future using baseline scenarios from
118 the CMIP6, we have selected the present year to be 2015 for consistency. The models and
119 perturbation simulations are described in Section 2, the model evaluation against surface
120 measurements in the Nordic countries are presented in Section 3.1, the contributions of sectoral
121 emissions on the air pollution levels in the Nordic region and the Arctic are presented in Section
122 3.2., and the health impacts and associated costs are presented in Section 3.3. Conclusions are given
123 in Section 4.

124 125 1. Materials and methods

126 127 2.1. Danish Eulerian Hemispheric Model (DEHM)

128
129 The DEHM model was originally developed mainly to study the transport of SO_2 and SO_4 to the
130 Arctic (Christensen 1997), but has been extended to different applications during the last decades. It
131 has been documented extensively in Brandt et al. (2012) and evaluated in several intercomparison
132 studies (e.g. Solazzo et al., 2012 a,b; Solazzo et al., 2017; Im et al., 2018a,b) and recently joined the
133 suit of operational models in the Copernicus Atmospheric Monitoring System (CAMS) to provide
134 regional forecasts of air pollution over Europe. The DEHM model uses a $150 \text{ km} \times 150 \text{ km}$ spatial
135 resolution over the Northern Hemisphere, then nests to $50 \text{ km} \times 50 \text{ km}$ resolution over Europe,
136 extending up to 100 hPa through 29 vertical levels, with the first layer height of approximately 20
137 m. The meteorological fields were simulated by the Weather Research and Forecast Model (WRF,
138 Skamarock et al., 2008) setup with identical domains and resolution. The time resolution of the
139 DEHM model is one hour. The gas-phase chemistry module includes 58 chemical species, 9
140 primary particles, including natural particles such as sea-salt and 122 chemical reactions (Brandt et
141 al., 2012). The model also describes atmospheric transport and chemistry of lead, mercury, CO_2 , as
142 well as POPs. Secondary organic aerosols (SOA) are calculated using the Volatility Base System
143 (VBS: Bergstrom et al., 2012). In addition to the anthropogenic PM and SOA due to biogenic
144 emissions, DEHM model also calculates sea-salt emissions and their transport and interactions with

145 other pollutants. The current version of the DEHM model does not include wind-blown or re-
146 suspended dust emissions. DEHM model does not output a PM_{2.5} or PM₁₀ diagnostic, however
147 these are calculated off-line, using all anthropogenic and natural components of PM, in order to be
148 used in the health impact assessment described in Section 2.2.

149

150 In the current study, the DEHM model used anthropogenic emissions from the EDGAR-HTAP
151 database and biogenic emissions are calculated online based on the MEGAN model. The total
152 emission per country for the different pollutants are presented in Table 1. The sectoral distributions
153 of emissions in each country are presented in Figure 1. As seen in the Table 2, most SNAP
154 (Selected Nomenclature for Air Pollutants; CEIP, 2019) sectors are considered individually, while
155 some are merged in order to reduce the computational costs. All sectors in relation to industrial
156 activities (combustion, processes, solvent use and extraction and transport of fossil fuels) are
157 merged into an “Industry” source sector, while waste management and agriculture sectors were
158 lumped into one source sector.

159

160 As seen in Figure 1, non-industrial combustion (orange bars), where non-industrial combustion
161 dominates, stands out as a major source contributing to CO and PM emissions while industry (grey
162 bars) (Table 2) is the largest source of NMVOCs, NO_x and SO_x. Traffic (yellow bars) also
163 contributes significantly to CO and NO_x. The largest source of NH₃ is from agriculture in
164 particular, as well as waste management (green bars) (Table 2).

165

166 Table 1. Total pollutant emissions in the Nordic countries (in Gg) in 2015.

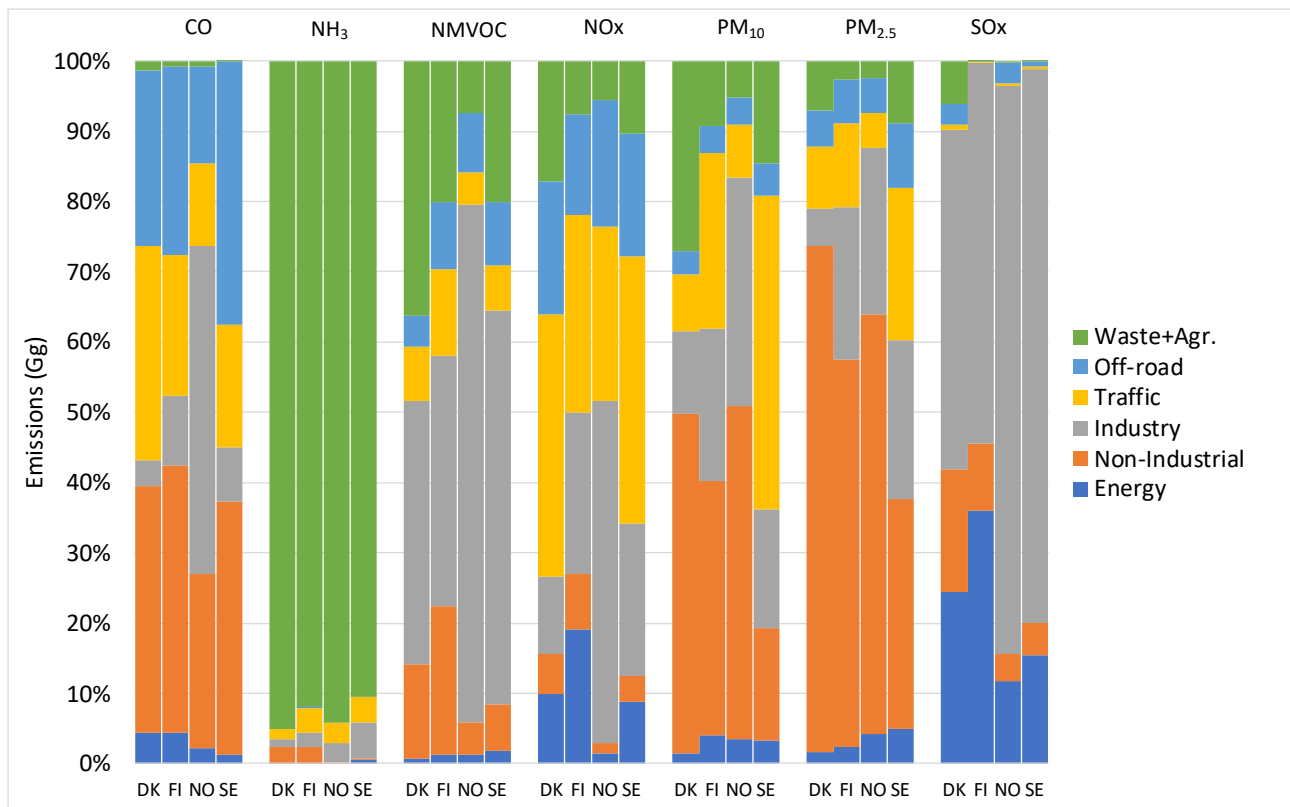
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	CO	NH ₃	NMVOC	NO _x	SO _x	PM ₁₀	PM _{2.5}
DK	251	75	106	102	9	31	20
FI	302	31	85	128	41	31	19
NO	378	28	155	133	16	35	27
SE	413	54	159	129	18	37	18

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174 Figure 1. Relative distributions (%) of sectoral emissions of major air pollutants in the Nordic
175 countries.

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177 2.1.1. Tagging Method

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179 The tagging method keeps track of contributions to the concentration field from a particular
180 emission source or sector, as explained in detail in Brandt et al. (2013a). Tagging involves
181 modelling the background concentrations and the δ -concentrations (the contributions from a
182 specific emission source or sector to the overall air pollution levels) in parallel (as two different
183 runs under the same run), where special treatment is required for the non-linear process of
184 atmospheric chemistry, since the δ -concentrations are strongly influenced by the background
185 concentrations in such processes. Although this treatment involves taking the difference of two
186 concentration fields, it does not magnify the spurious oscillations (the Gibbs phenomenon), which
187 are primarily generated in the advection step. The non-linear effects can be accounted for in the δ -
188 concentrations without losing track of the contributions arising from the specific emission source or
189 sector.

190

191 2.1.2. Model evaluation

192

193 Surface concentrations modelled by the DEHM model were evaluated against data at selected urban
194 background and regional or global monitoring stations in each Nordic country. The statistical
195 comparisons included using correlation coefficient (r), mean bias (MB) and normalized mean bias
196 (NMB) and root mean square error ($RMSE$). The station information is provided in Table S1, along
197 with the descriptions of the monitoring network in each country.

198

199

200 2.2. Economic Valuation of Air Pollution (EVA) System

201

202 The EVA system (Brandt et al., 2013a,b; Geels et al., 2015; Im et al., 2018a) is based on the
203 impact-pathway chain method (Friedrich and Bickel, 2001). The EVA system can estimate acute
204 (short-term) and chronic (long-term) mortality, related to acute exposure to O₃, and SO₂, and
205 chronic exposure to PM_{2.5}, and the associated external costs. The EVA system requires gridded
206 concentrations along with gridded population data, exposure-response functions (ERFs) for health
207 impacts, which are recommended by the WHO (2013), and economic valuation functions of the
208 impacts from air pollution. In addition, EVA uses population densities over fixed age intervals,
209 corresponding to babies (under one year), children (under 15), adults (above 15 and above 30), and
210 elderlies (above 65). The impacts of short-term exposure to O₃, and SO₂, and the long-term
211 exposure to PM_{2.5} are well established. EVA uses the annual mean concentrations of SO₂, and
212 PM_{2.5}, while for O₃, it uses the SOMO35 metric that is defined as the annual sum of the daily
213 maximum of 8-hour running average over 35 ppb, following WHO (2013) and EEA (2017).
214

215 The health impacts are calculated using an ERF of the following form:

$$216 R = \alpha \times \delta_c \times P$$

217 where R is the response of the mortality rate or the years of life lost (in cases or days), δ_c denotes
218 the pollutant concentration, P denotes the affected share of the population, and α an empirically
219 determined constant for the particular health outcome. EVA uses ERFs that are modelled as a linear
220 function, which is a reasonable approximation for the region of interest in the present study, as
221 showed in several studies (e.g. Pope et al., 2000; the joint World Health Organization/UNECE Task
222 Force on Health (EU, 2004; Watkiss et al., 2005)). However, some studies showed non-linear
223 relationships, being steeper at lower than at higher concentrations (e.g. Samoli et al., 2005).
224 Therefore, linear relationships may lead to overestimated health impacts over highly polluted areas.
225 Exposure response functions (ERF) for all-cause chronic mortality due to PM_{2.5} are based on Pope
226 et al., 2002; Krewski et al., 2009), which are also recommended by the WHO (2013). These are the
227 most extensive and up-to-date data, although there are ongoing studies in Europe, and in particular
228 in the Nordic region to develop regional-specific ERFs (e.g. the Nordic WelfAir project:
229 <https://projects.au.dk/nordicwelfare/>). The current version of the EVA system used in the present
230 study does not include impacts due to exposure to NO₂. However, a new version is currently under
231 development under the NordicWelfAir project.

232

233 EVA calculates the number of lost life years for a Danish population cohort with normal age
234 distribution, when applying the ERF of Pope et al. (2002) for all-cause mortality (relative risk, RR=
235 1.062 (1.040-1.083) on 95% confidence interval). The latency period sums to 1138 year of life lost
236 (YOLL) per 100 000 individuals for an annual PM_{2.5} increase of 10 µg m⁻³ (Andersen et al, 2008).
237 The YOLL is then converted to number of cases by dividing by 10.6 following Watkiss et al.
238 (2005). The counterfactual PM_{2.5} concentration is assumed to be 0 µg m⁻³ following the EEA
239 methodology, meaning that the impacts have been estimated for the simulated total (anthropogenic
240 and natural) PM_{2.5} mass. Applying a low counterfactual concentration can underestimate health
241 impacts at low concentrations if the relationship is linear or close to linear (Anenberg et al., 2016).
242 However, it is important to note that uncertainty in the health impact results may increase at low
243 concentrations due to sparse epidemiological data. Assuming linearity at very low concentrations

244 may distort the true health impacts of air pollution in relatively clean atmospheres (Anenberg et al.,
245 2016).

246 Regarding short-term exposure to O₃, EVA uses the ERF recommended by the CAFE Programme
247 (Hurley et al., 2005) and WHO (2013) that uses the daily maximum of 8-hour mean O₃
248 concentrations. There are also studies showing that SO₂ is associated with acute mortality, and EVA
249 adopts the ERF identified in the APHENA study – Air Pollution and Health: A European Approach
250 (Katsouyanni et al., 1997). Some recent studies also report the chronic effects from O₃ (e.g. Turner,
251 2016), however the current version of the EVA model does not include these effects. The ERFs
252 used in EVA to calculate mortality are presented in Table 2.

253
254 Table 2. Exposure-response functions (ERF) used in EVA to calculate premature mortality.
255

Health effects (compounds)	Exposure-response coefficient	Valuation, €
	(α)	(EU27)
Acute mortality ^{2,3} (SO ₂)	7.85E-6 cases/ μgm^{-3}	1,532,099 per case
Acute mortality ^{2,3} (O ₃)	3.27E-6*SOMO35 cases/ μgm^{-3}	57,510 per YOLL
Chronic mortality ^{1,4} , YOLL (PM)	1.138E-3 YOLL/ μgm^{-3} (>30 years)	2,298,148 per case
Infant mortality ⁵ , IM (PM)	6.68E-6 cases/ μgm^{-3} (< 9 months)	

268
269 ¹ Pope et al. (2002), ² Anderson (1996), ³ Touloumi (1996), ⁴ Pope et al. (1995), ⁵ Woodruff et al. (1997).

270
271 For the valuation of the health impacts, a value of EUR 1.5 million was applied for preventing an
272 acute death, following expert panel advice (EC, 2001), while for the valuation of a life year, a value
273 of EUR 57 500 per year of life lost (YOLL) were applied (Alberini et al., 2006). More details can
274 be found in Im et al. (2018a).

275 276 2.3. Scenarios (response and contribution)

277
278 We have applied a 30% reduction on land-based anthropogenic emissions from each of the
279 continental Nordic countries, which include Denmark, Finland, Norway and Sweden. Each
280 simulation perturbed a SNAP sector from an individual Nordic country, which are listed in Table 3.
281 Industry is perturbed as the combination of SNAP 3,4,5 and 6, while agriculture (SNAP9) and
282 waste management (SNAP 10) are perturbed as one combined sector.

283
284 DEHM model has been run on “tagged” mode, explained in section 2.1., so each simulation
285 included a “perturbed” and “non-perturbed” concentration, which we used to calculate the response
286 to the 30% reduction in the particular country and sector. These responses are then converted to
287 population-weighted contributions using the gridded population densities and by assuming a linear
288 extrapolation to 100%.

289
290

291 Table 3. Source sectors used in the perturbation scenarios.
 292

Source Sectors	SNAP Code
Combustion in energy and transformation industries	1
Non-industrial Combustion	2
Industry	3,4,5,6
Road transport	7
Other mobile sources and machinery	8
Waste and agriculture	9,10

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295 3. Results and Discussion

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297 3.1. Evaluation

299 Surface ozone and PM_{2.5} concentrations calculated by the DEHM model have been evaluated using
 300 surface observations from the urban background and regional background monitoring stations in the
 301 Nordic countries. The comparison of the mean of all observed concentrations in each country and
 302 the corresponding modelled concentrations are presented in Table 4 while Figs. 2 and 3 present
 303 Taylor diagrams for each station in each Nordic country, giving insight to the spatial distribution of
 304 model performance. As seen in Table 3, temporal variation of O₃ levels are well reproduced by the
 305 DEHM model over all countries ($r > 0.6$), however with an overestimation of ~10% over Denmark,
 306 Finland and Sweden, and ~30% over Norway. The daily variations of PM_{2.5} levels, averaged over
 307 all stations in each Nordic country are well reproduced for Denmark ($r > \sim 0.7$), moderately over
 308 Norway and Sweden ($r > 0.4$), and poorly ($r \sim 0$) over Finland (Table 3). PM_{2.5} concentrations are
 309 underestimated by up to 35% over Denmark, Finland and Norway, and overestimated by 8% over
 310 Sweden.

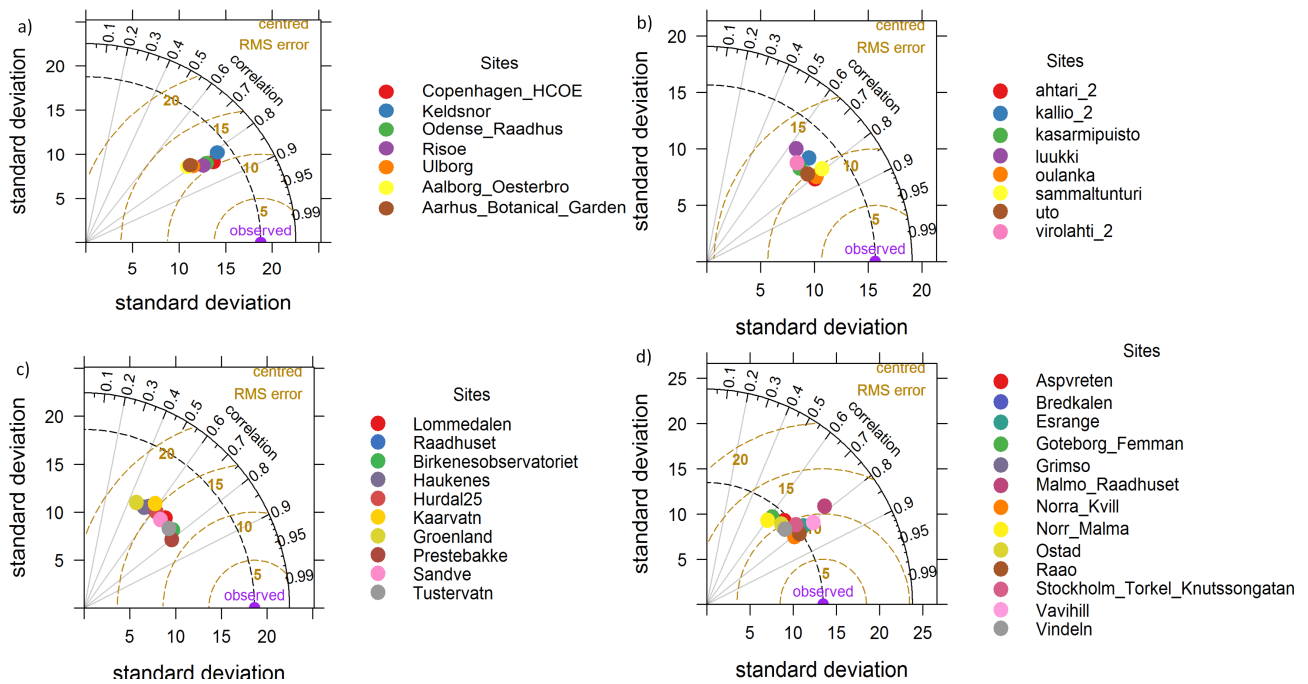
311
 312 In all countries, lower *NMB* values are calculated for O₃ over the regional background stations
 313 compared to urban background stations, where values are overestimated. Regarding PM_{2.5}, no such
 314 conclusions can be drawn due to very limited number of regional background stations in Denmark
 315 and Norway. In Finland, lower *NMB* values for PM_{2.5} are calculated for the regional background
 316 stations, while in Sweden, much lower *NMB* values are calculated for the urban stations. These
 317 differences reflect the underestimations in emissions as well as the coarse model resolution, as well
 318 as missing sources, in particular for PM, such as wind-blown and resuspended dust in the DEHM
 319 model. It should also be mentioned that the modelled PM does not contain residual water. Table S2
 320 shows the same comparisons for NO₂ and SO₂. The underestimations in the modelled PM_{2.5} levels
 321 imply an underestimated exposure to PM_{2.5} levels, given the dominance of PM_{2.5} in premature
 322 mortality. Similarly, the overestimations in O₃ levels can be attributed to the underestimated NO-
 323 titration (Table S2).

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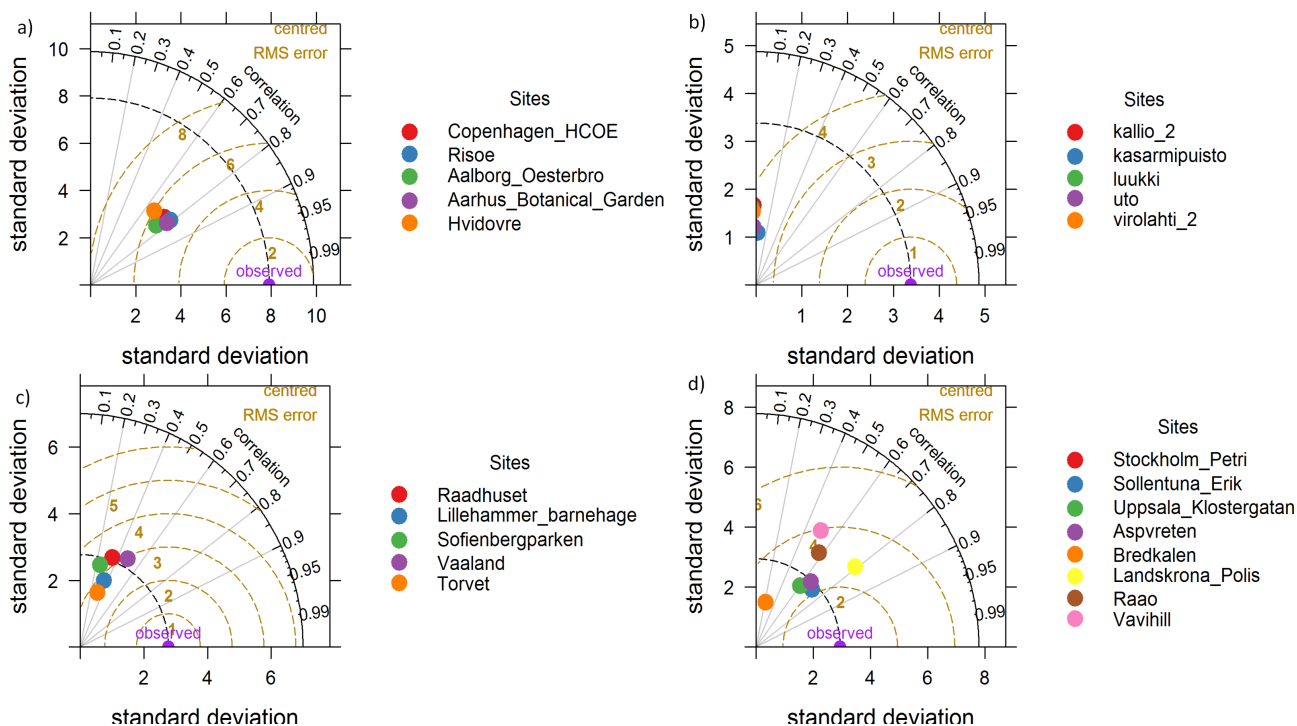
326 Table 4. Model evaluation for the daily mean concentrations of O₃ and PM_{2.5} for all the selected
 327 stations in the Nordic countries.
 328

	O ₃					PM _{2.5}				
	<i>r</i>	<i>Obs.</i> ($\mu\text{g m}^{-3}$)	<i>NMB</i> (%)	<i>NME</i> (%)	<i>RMSE</i> ($\mu\text{g m}^{-3}$)	<i>r</i>	<i>Obs.</i> ($\mu\text{g m}^{-3}$)	<i>NMB</i> (%)	<i>NME</i> (%)	<i>RMSE</i> ($\mu\text{g m}^{-3}$)
Denmark	0.91	59.59	0.10	0.11	7.65	0.85	10.77	-0.31	0.31	3.78
Finland	0.85	55.20	0.10	0.15	9.24	0.02	5.05	-0.16	0.24	1.56
Norway	0.73	54.65	0.27	0.29	14.78	0.66	6.85	-0.36	0.36	2.76
Sweden	0.86	57.88	0.13	0.15	9.49	0.35	5.00	0.08	0.30	1.62

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331
 332 Figure 2. Taylor diagrams for daily mean O₃ for all stations in a) Denmark, b) Finland, c) Norway
 333 and d) Sweden.
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339 Figure 3. Taylor diagrams for daily mean PM_{2.5} for all stations in a) Denmark, b) Finland
340 and d) Sweden.

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342 3.2. Sectoral contributions to surface concentrations

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344 3.2.1. Nordic countries

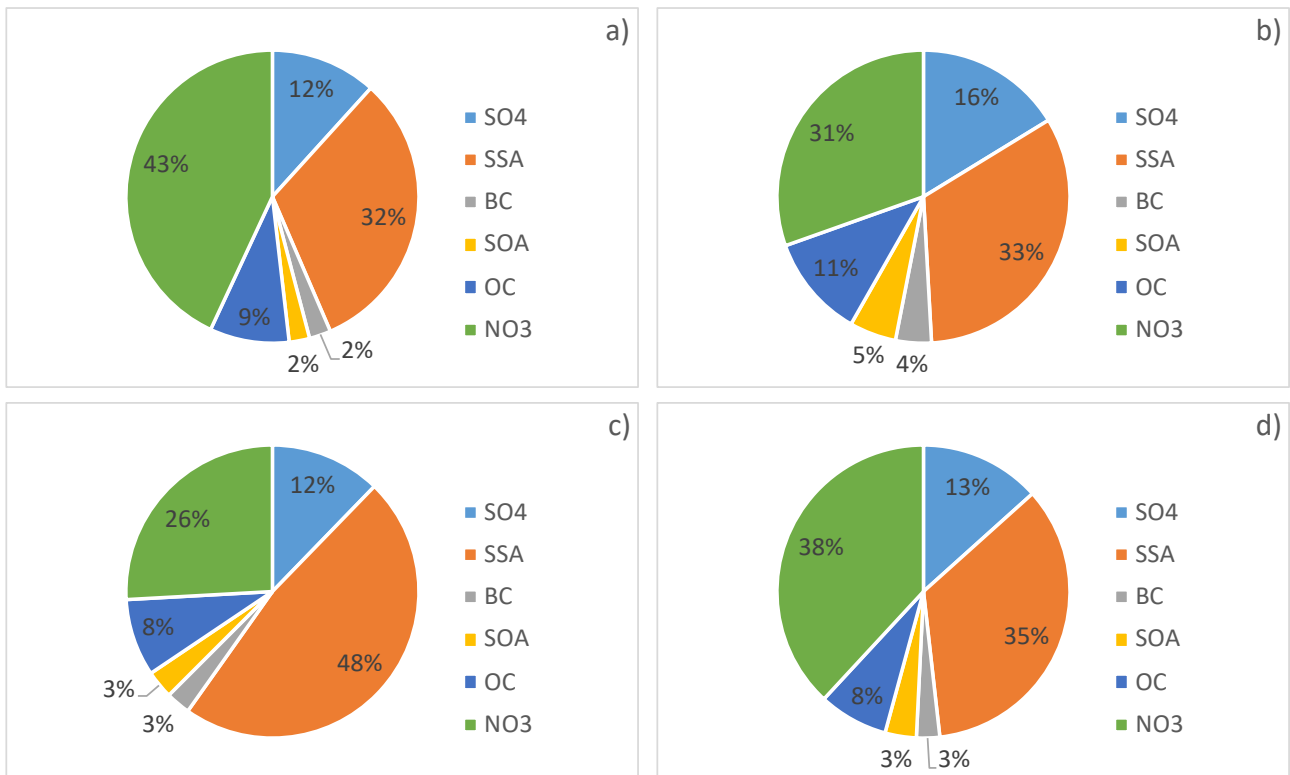
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346 In general, the long-term transport of air pollutants from one country to another is dependent on the
347 global and regional atmospheric circulation and on the relative geographic positions of the
348 countries. Nordic countries are influenced by substantial long-range transported contributions of air
349 pollution especially from the central, western and central eastern parts of Europe. In the region
350 containing the continental Nordic countries, the prevailing atmospheric flow directions near the
351 ground surface are from the west, south-west and south. Based on the prevailing atmospheric
352 circulation patterns, it is therefore to be expected that, e.g., the emissions in Denmark will have a
353 relatively larger influence on the pollution levels in the other Nordic countries than those in
354 Finland.

355

356 Our simulations show that PM_{2.5} mass concentrations over the Nordic countries are dominated by
357 nitrate aerosols (30% - 45 %) and sea-salt (30% - 50%). SO₄ aerosols contribute 10 to 15% of PM_{2.5}
358 concentrations while OC contributes by 8-11%, and BC by 2-4% of the PM_{2.5} mass. As SO₄ and
359 NO₃ aerosols include NH₄ in DEHM, results suggest that NH₄ aerosols contribute by more than half
360 of the PM_{2.5} mass over the Nordic countries. The annual mean surface PM_{2.5} concentrations for
361 Denmark, Finland, Norway and Sweden are calculated to be 9.1 μg m⁻³, 4.4 μg m⁻³, 4.8 μg m⁻³ and
362 5.8 μg m⁻³, respectively. These values are in agreement with those reported by the EEA (2017),
363 however underestimating by 12% (Denmark) up to 30% (Norway).

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Figure 4. Simulated surface PM_{2.5} chemical composition over a) Denmark, b) Finland, c) Norway, and d) Sweden.

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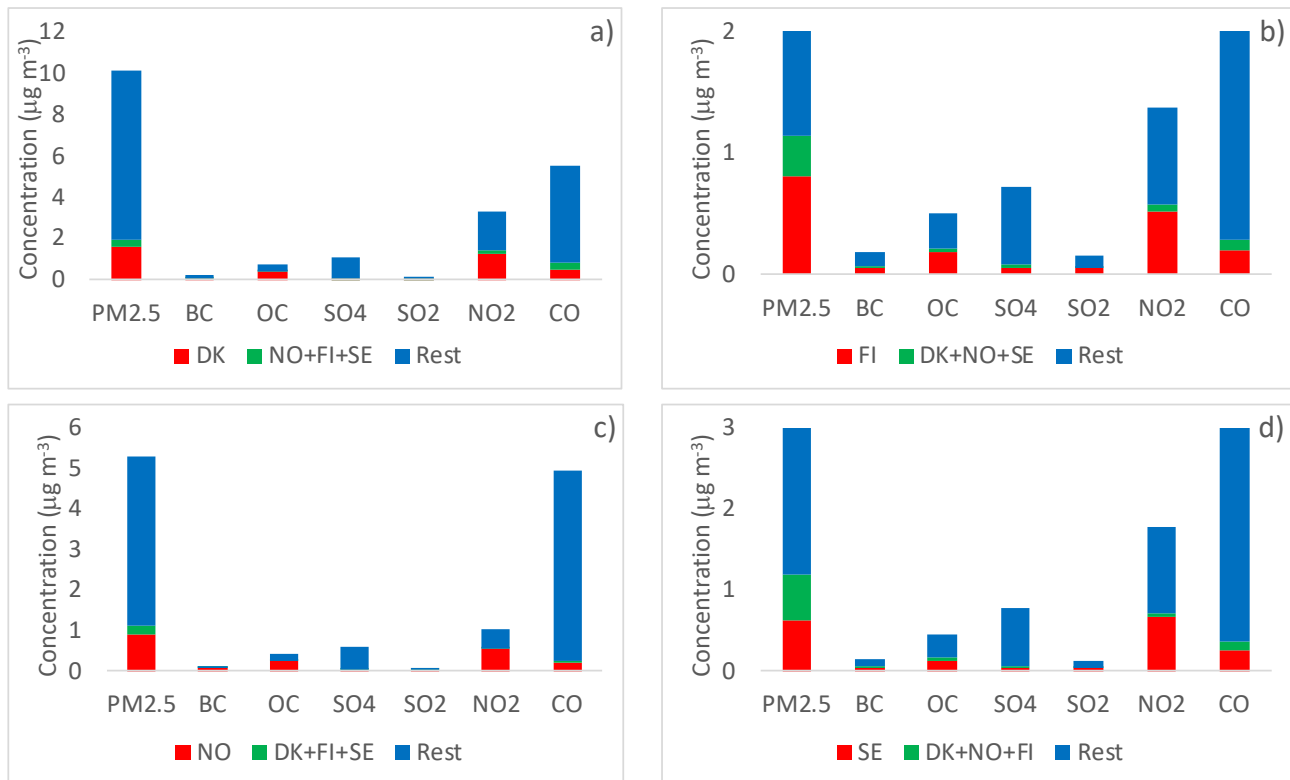
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Figure 5 compares the contribution of the total contributions anthropogenic sectors of each Nordic country on the surface concentrations over the country itself, with contributions from the anthropogenic sources in rest of the Nordic countries and rest of the world. Therefore, PM_{2.5} in the figure does not contain the natural components that cannot be regulated, such as sea-salt. Figure 5 clearly shows that over 80% or more of PM_{2.5} surface levels are transported outside the Nordic region, pointing that the Nordic countries are responsible for less than 20% of the particulate pollution in the region. This suggests significant decreases in the PM_{2.5} levels in the region can only be possible by reductions in the emissions upwind. Similar high contributions for other species including CO also shows that Nordic countries are exposed to airmasses coming from rest of the world while local pollution is low. The figure also shows that PM_{2.5} levels are generally low in the Nordic countries, with annual means lower than 10 $\mu\text{g m}^{-3}$ (highest in Denmark and lowest in Finland). Similar to PM_{2.5}, annual mean surface O₃ levels are also low ($\sim 30 \mu\text{g m}^{-3}$). Similar analyses done for O₃ (not shown) show that O₃ levels are controlled largely regional, where the local sources in the Nordic countries lead to small sink of O₃ due to NO-titration. This is also in agreement with Im et al. (2018b) reporting high Response to Extra-Regional Emission Reductions (RERER) values (>0.8) suggesting that O₃ is a regional background pollutant in Europe.



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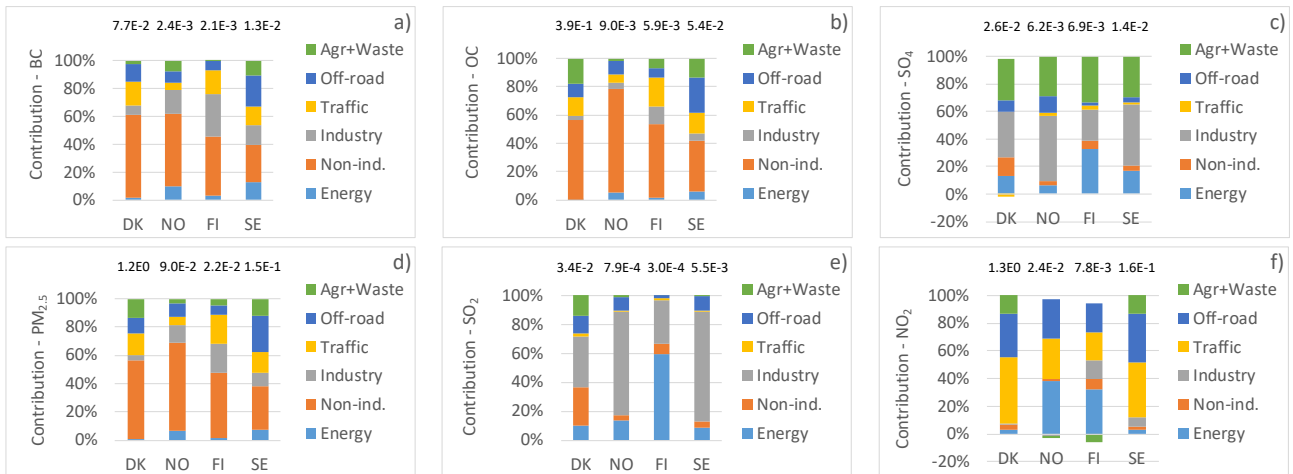
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Figure 5. Absolute contributions of national, Scandinavian and other sources on the surface levels of major air pollutants over a) Denmark, b) Finland, c) Norway and d) Sweden. Note that CO concentrations are divided by 20 to scale with other pollutants.

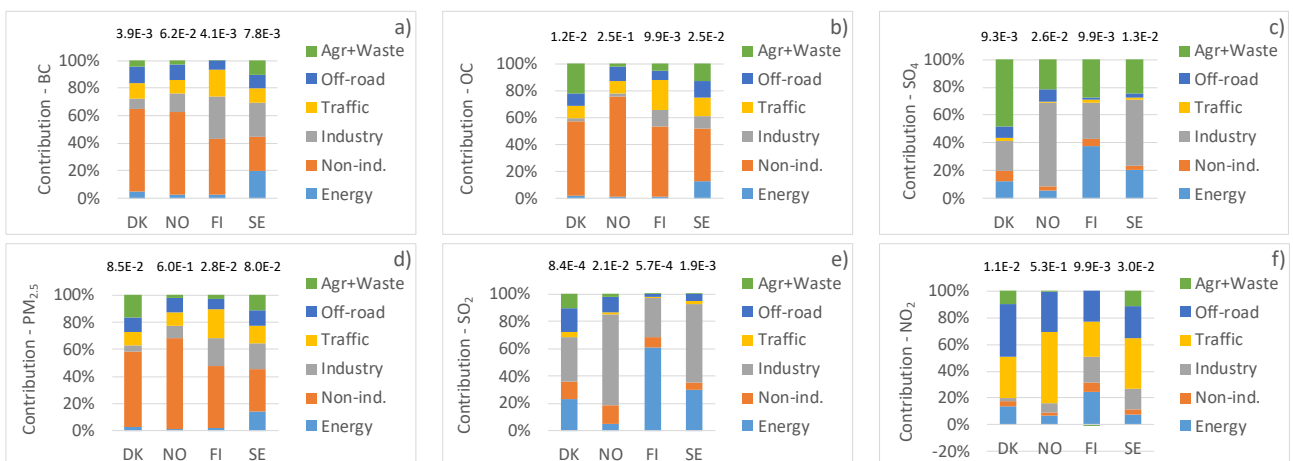
Danish emissions contribute to only $1.14 \mu\text{g m}^{-3}$ (13%) of the surface PM_{2.5} concentrations over Denmark ($9.1 \mu\text{g m}^{-3}$), while contributions to other Nordic countries are about 3% (Figure 6). Non-industrial combustion (SNAP2), which is dominated by non-industrial combustion, is responsible for $0.36 \mu\text{g m}^{-3}$ (60%) of the Danish contribution to surface PM_{2.5} concentrations over Denmark. Non-industrial combustion contributes to $0.22 \mu\text{g m}^{-3}$ (56%) of the Danish contribution to surface organic carbon (OC) concentrations over the country, suggesting the importance of non-industrial wood burning for heating. Industry contributes to $0.01 \mu\text{g m}^{-3}$ (35%) of the Danish contribution to the surface SO₂ concentrations over Denmark, while on-road and off-road transport contributes equally to the Danish share of the in surface NO₂ concentrations by $1.02 \mu\text{g m}^{-3}$ (~79% together). Agriculture and waste handling are important sources for surface SO₄ levels over Denmark as well as over the other Nordic countries, via the formation of ammonium sulfate ((NH₄)₂SO₄) due to the large ammonia (NH₃) emissions from these sectors. $0.26 \mu\text{g m}^{-3}$ of PM_{2.5} over Denmark comes the other Nordic countries, with $0.03 \mu\text{g m}^{-3}$ coming from non-industrial combustion only.



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Figure 6. Population-weighted sectoral contributions of Danish emissions on surface a) BC, b) OC, c) SO₄, d) PM_{2.5}, e) SO₂ and f) NO₂ over the Nordic countries. The labels above the bars show the absolute total contribution in $\mu\text{g m}^{-3}$ from all the sectors in Denmark.

Contributions of the Norwegian emissions over the Nordic countries are presented in Figure 7. Similar to the Danish emissions, Norwegian emissions contribute to 0.6 $\mu\text{g m}^{-3}$ (13%) of the surface PM_{2.5} concentrations over Norway, while contributions to other Nordic countries are below 1%, except for NO₂, where on-road transport emissions from Norway contributes to almost 0.02 $\mu\text{g m}^{-3}$ (42%) of the surface NO₂ levels over Finland. Non-industrial combustion is the main source of pollutant levels, in particular for OC, where Norwegian emissions are responsible for 0.18 $\mu\text{g m}^{-3}$ (74%) of local contribution to the surface OC levels over Norway. Industry is a major source of surface SO₂ levels over Norway, contributing to 0.02 $\mu\text{g m}^{-3}$ (66%) of the local contribution. 0.2 $\mu\text{g m}^{-3}$ of PM_{2.5} levels over Norway comes from the other Nordic countries, 0.02 $\mu\text{g m}^{-3}$ being from non-residential combustion.

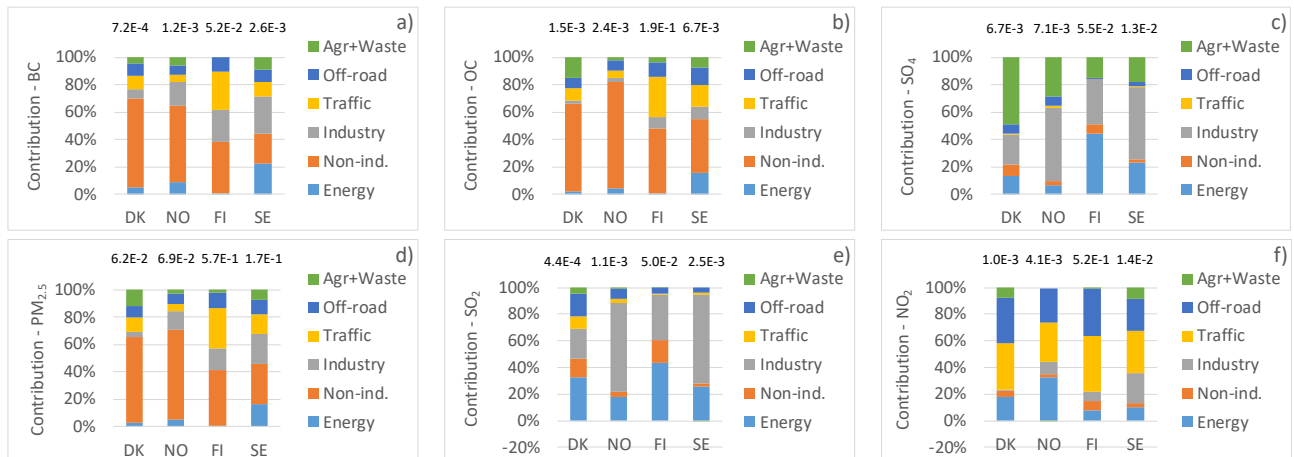


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Figure 7. Population-weighted sectoral contributions of Norwegian emissions on surface a) BC, b) OC, c) SO₄, d) PM_{2.5}, e) SO₂ and f) NO₂ over the Nordic countries. The labels above the bars show the absolute total contribution in $\mu\text{g m}^{-3}$ from all the sectors in Norway.

Figure 8 shows the contributions of Finnish emissions on the pollutant levels over the Nordic countries. Similar to Denmark and Norway, non-industrial combustion is the major source of

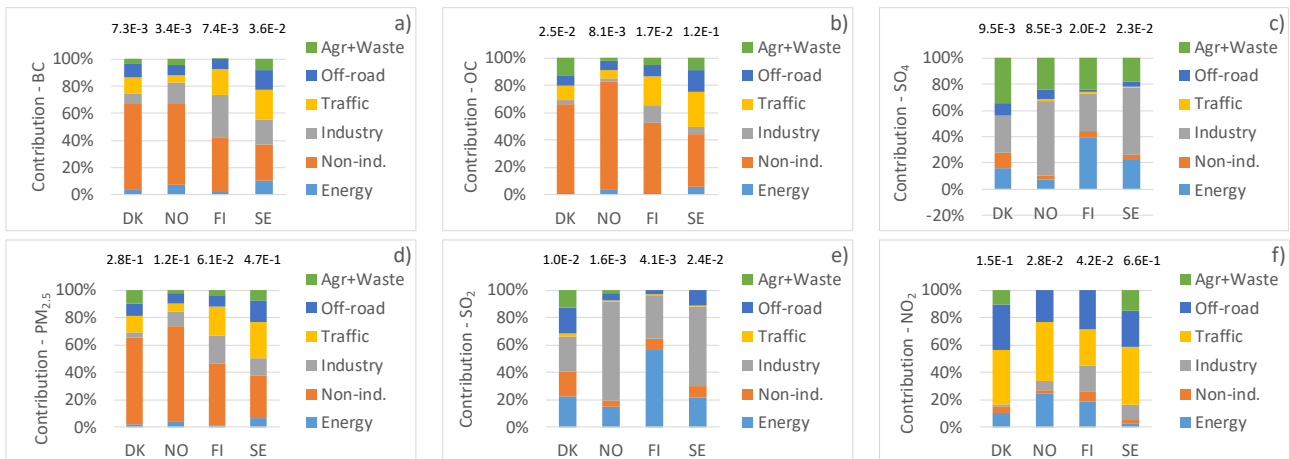
431 pollution over Finland, although contributions are lower compared to Denmark and Norway (0.19
 432 $\mu\text{g m}^{-3}$ (41%) of $\text{PM}_{2.5}$ and 0.11 $\mu\text{g m}^{-3}$ (48%) of OC). Another noticeable difference is that energy
 433 production is also an important contributor to surface SO_2 (0.01 $\mu\text{g m}^{-3}$: 44%) and SO_4 (0.03 $\mu\text{g m}^{-3}$:
 434 44%) levels over Finland. 0.3 $\mu\text{g m}^{-3}$ of $\text{PM}_{2.5}$ levels over Finland come from the other Nordic
 435 countries, 0.2 $\mu\text{g m}^{-3}$ being from non-residential combustion. Finnish emissions, in particular
 436 industrial combustion, contribute largest to the air pollution over Sweden.
 437



438
 439 Figure 8. Population-weighted sectoral contributions of Finnish emissions on surface a) BC, b) OC,
 440 c) SO_4 , d) $\text{PM}_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the
 441 absolute total contribution in $\mu\text{g m}^{-3}$ from all the sectors in Finland.
 442

443 Contributions from the Swedish emission sources to surface pollutant levels over the Nordic
 444 countries are presented in Figure 9. Unlike other Nordic countries, Swedish emissions have larger
 445 contributions to pollution levels over the other Nordic countries, in particular over Norway. The
 446 figure also shows that Sweden does not experience as dominant contribution from non-industrial
 447 combustion (32%) like the other Nordic countries show. Swedish emissions from SNAP2 are much
 448 lower than for the rest of the Nordic countries (official emissions reported to the CLRTAP), most
 449 probably due to lower emission factors. Non-industrial combustion and industry contribute
 450 similarly to the surface $\text{PM}_{2.5}$ levels. Industry also has an important contribution to surface SO_4
 451 levels (0.01 $\mu\text{g m}^{-3}$: 51%), as well to SO_2 (0.01 $\mu\text{g m}^{-3}$: 58%) and BC (0.006 $\mu\text{g m}^{-3}$: 18%). 0.5 μg
 452 m^{-3} of surface $\text{PM}_{2.5}$ levels over Sweden comes from the other Nordic countries, of which, 0.1 μg
 453 m^{-3} comes from non-residential combustion.
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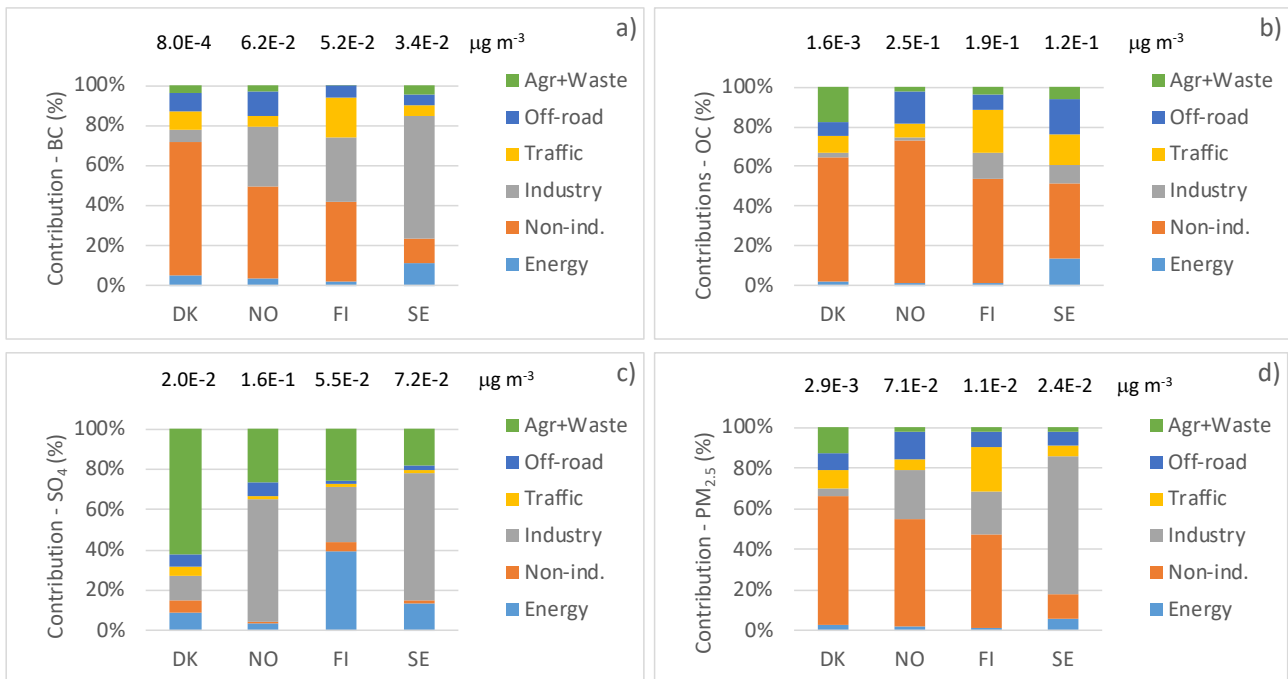
458 Figure 9. Population-weighted sectoral contributions of Swedish emissions on surface a) BC, b)
459 SO₄, c) OC, d) PM_{2.5}, e) SO₂ and f) NO₂ over the Nordic countries. The labels above the bars show
460 the absolute total contribution in $\mu\text{g m}^{-3}$ from all the sectors in Sweden.
461

462 3.2.2. Arctic

463

464 The contributions of the emission sources in the different Nordic countries on the surface aerosol
465 concentrations over the Arctic region (defined as the area north of 67 °N latitude) are presented in
466 Figure 10. Results show that overall, Norway has the largest contribution to surface aerosol levels
467 over the Arctic, while Denmark has the lowest contribution, although contributions are only a few
468 percent. Norwegian emissions, in particular non-industrial combustion, contributes to about 2% of
469 the surface BC levels over the Arctic. Non-industrial combustion in the Nordic countries is also the
470 largest contributor to Arctic BC levels, except for Sweden, where industry plays a more important
471 role. Non-industrial combustion is also the dominant contributor to OC levels over the Arctic.
472 Sulfate levels are largely influenced by the contributed from the agriculture and waste treatment
473 facilities over the Nordic countries. Contributions to Arctic PM_{2.5} levels are similar to the
474 contributions to the BC levels.
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479 Figure 10. Population-weighted sectoral contributions from a) Denmark, b) Norway, c) Finland and
480 d) Sweden to the surface aerosol levels over the Arctic (north of 67°N). The labels above the bars
481 show the absolute total contribution in µg m⁻³ from all the sectors in each source country.
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3.2.3. Spatial distributions of contributions

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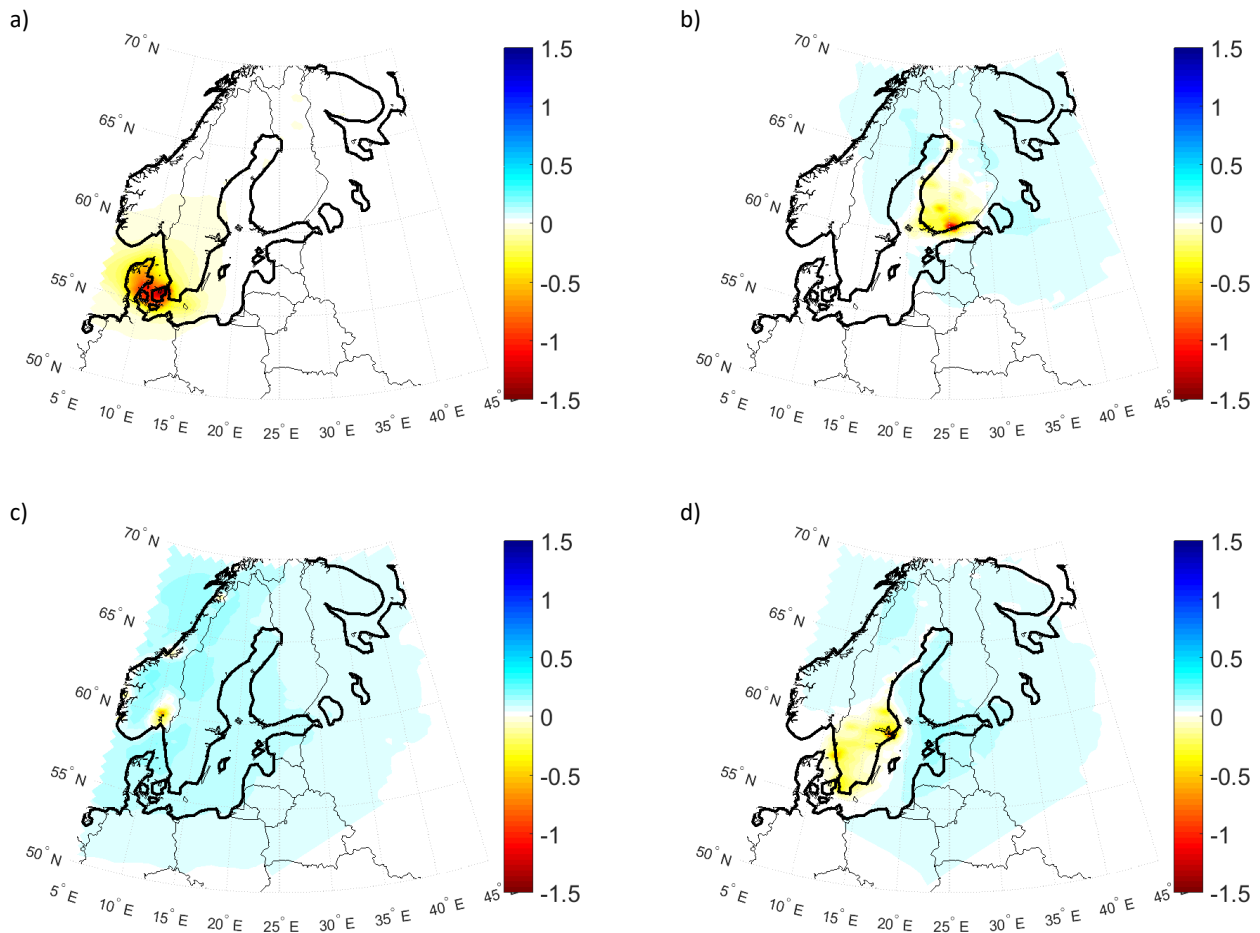
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The geographical distributions of total anthropogenic emissions from each Nordic country to surface PM_{2.5} and O₃ levels are calculated to investigate the extent of contributions from each Nordic country to its neighbours and to the Arctic. Figure 11 shows the annual-mean absolute contributions (%) of total land-based anthropogenic emissions to surface O₃ levels in the Nordic region from each country. The annual-mean contributions are very low, (up to 1.5 µg m⁻³: 5%). Largest contributions in each country are calculated in the source region in the particular country, implying the impact of O₃ titration by local fresh NO emissions. Danish anthropogenic emissions (Figure 11a) lead to a titration of up to 1.5 µg m⁻³ (around 4-5%), particularly over capital region. The largest impact of Finnish emissions is around the Helsinki area, responsible for up to 1 µg m⁻³ (5%) of surface O₃ destruction over the area (Figure 11b). Finnish emissions also lead to an increase of surface O₃ levels by up to 0.5 µg m⁻³ (1%) over the downwind regions to the southeast and northwest. Impact of Norwegian emissions to surface O₃ levels (Figure 11c) are largest (up to 1 µg m⁻³: 2%) over the Oslo area and the impact extents over the northern part of Oslo with a slightly larger spatial contribution to O₃ levels compared to Denmark and Finland. The Swedish emissions have a larger geographical impact on the surface O₃ levels (Figure 11d) over the country itself compared to the other Nordic countries but the magnitude is similar to the impact from the Norwegian emissions.



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Figure 11. Spatial distributions of annual population-weighted mean absolute contributions ($\mu\text{g m}^{-3}$) of total emissions from a) Denmark, b) Finland, c) Norway, and d) Sweden to surface O_3 levels in the Nordic region.

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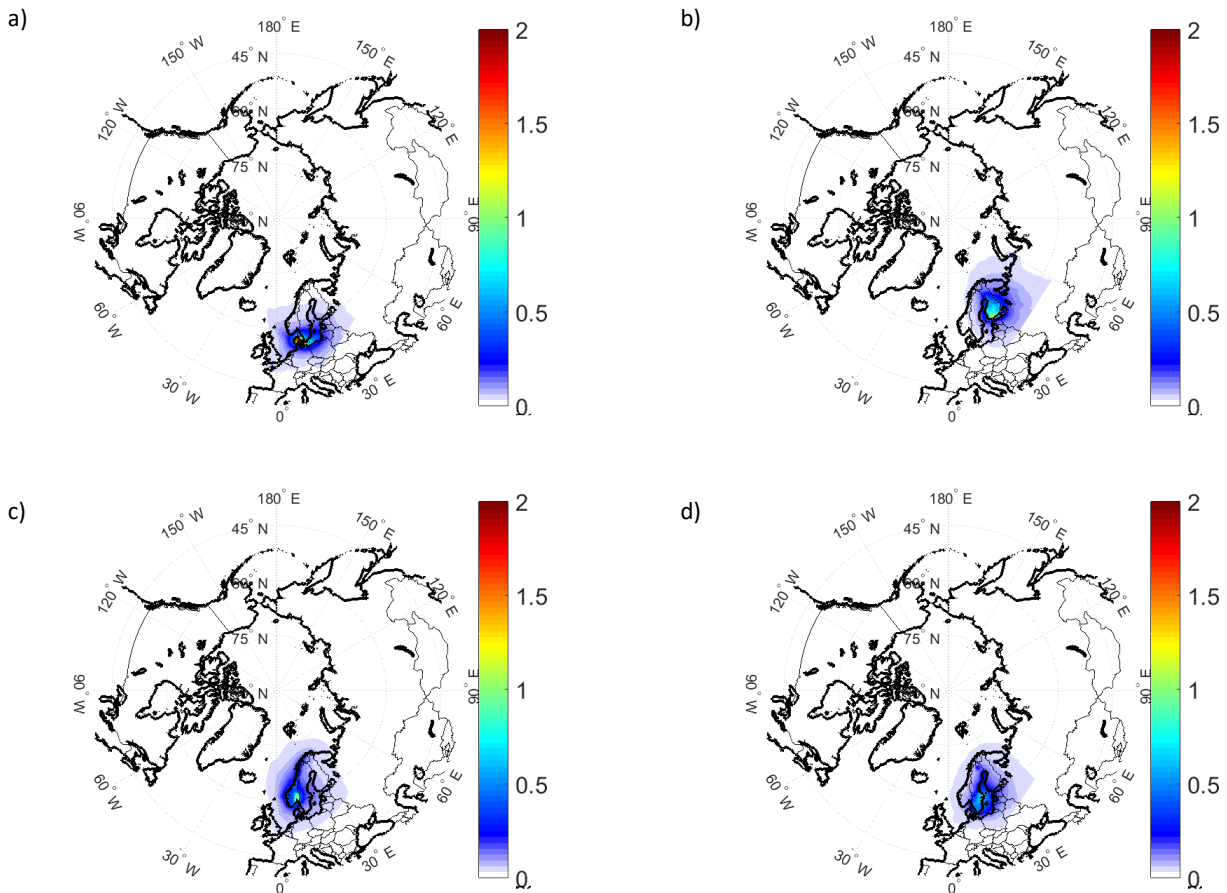
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Figure 12 shows the annual-mean absolute contributions of each Nordic country on the surface $\text{PM}_{2.5}$ levels in the entire model domain. Danish anthropogenic emissions are responsible for up to 20% of surface $\text{PM}_{2.5}$ levels over Denmark, with largest contributions over the capital region (Greater Copenhagen area) (Figure 12a). Danish land emissions also impact the surface $\text{PM}_{2.5}$ levels over the southern part of Sweden and Norway, by around 4% and 2%, respectively. The Finnish anthropogenic emissions have the largest impact on surface $\text{PM}_{2.5}$ levels over the southern part of the country, around the capital region by up to 30% (Figure 12b). Finnish emissions also have a small impact, lower than 3%, on the central part of Sweden and northern parts of Norway. Norwegian anthropogenic emissions have largest contributions to surface $\text{PM}_{2.5}$ level around the capital region by up to 30%, while there is also a significant impact on surface $\text{PM}_{2.5}$ levels over Sweden by around 7% (Figure 12c). Finally, Swedish anthropogenic emissions have large contribution to surface $\text{PM}_{2.5}$ levels over the Stockholm area by around 15% and also contributes to $\text{PM}_{2.5}$ levels over Finland, in particular over the southwestern parts of Finland, by up to 5% (Figure 12d).

Figure 12 also shows the impact of anthropogenic emissions from each Nordic country to the surface $\text{PM}_{2.5}$ over the Arctic. Overall, the impacts are very small, around a few per cent, as seen in

526 the figure. The Danish emissions (Figure 12a) have a more local contribution compared to other
 527 Nordic countries and the impact does not reach above roughly 70 °N. The outflow from Finland,
 528 Norway and Sweden can reach to the central Arctic ocean over to the northern parts of Greenland,
 529 however contributions are around 1-2% (Figs. 12b-d).
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532
 533 Figure 12. Spatial distributions of annual population-weighted mean absolute contributions ($\mu\text{g m}^{-3}$)
 534 of total emissions from a) Denmark, b) Finland, c) Norway, and d) Sweden to surface $\text{PM}_{2.5}$ levels
 535 over the Nordic and the Arctic regions (north of 67°N).
 536

537 3.3. Contribution to premature mortality and costs

538
 539 The number of acute and chronic premature mortality in the four selected Nordic countries and the
 540 Arctic region (north of 67°N), along with the associated costs are presented in Table 5. The 95%
 541 confidence intervals provided in the brackets are calculated by scaling the calculated health
 542 outcomes by the confidence intervals of relative risk (RR) presented in section 2.2 (RR= 1.062
 543 [1.040-1.083]). As seen in the Table, chronic mortality due to $\text{PM}_{2.5}$ is the major source for
 544 premature mortality, as EVA calculates chronic mortality only due to exposure to $\text{PM}_{2.5}$ (see Table
 545 2). The highest number of cases is calculated for Sweden ($\sim 4\,200$ cases), followed by Denmark
 546 ($\sim 3\,500$ cases), Finland ($\sim 1\,800$) and Norway ($\sim 1\,700$). Results also show that SO_2 is responsible
 547 for almost all acute mortalities in the region, which is consistent with earlier studies (e.g. Brandt et
 548 al., 2013). This is due to the decrease of O_3 in the region by fresh NO emissions, leading to low

549 mortality due to O₃-exposure. These numbers lead to an associated cost of more than 2 billion Euros
 550 in Sweden and Denmark and ~ 1 billion Euros in Finland and Norway. The number of premature
 551 death cases are comparable with existing literature (e.g. Brandt et al., 2013a for Denmark; Solazzo
 552 et al., 2018 for all four Nordic countries; EEA, 2017 for all four Nordic countries). In the Arctic
 553 region, the total number of premature mortality cases is calculated to be 94, 93 of which are due to
 554 exposure to PM_{2.5} (chronic), leading to a cost of 58 million Euros.
 555

556 Table 5. Acute and chronic premature death cases in the Nordic countries and the Arctic region
 557 (north of 67°N) in 2015 and the associated costs. The brackets show the 95% confidence interval.
 558

	Denmark	Finland	Norway	Sweden	Arctic
Premature Mortality (number of cases)					
Acute	19 [19 20]	18 [18 18]	6 [6 6]	25 [24 25]	1 [1 1]
Chronic	3 332 [3 263 3 398]	1 707 [1 671 1 740]	1 596 [1 563 1 628]	4 091 [4 006 4 172]	93 [91 95]
Total	3 351 [3 282 3 417]	1 725 [1 689 1 759]	1 602 [1 569 1 634]	4 115 [4 030 4 197]	94 [92 96]
Cost (million Euros)					
Acute	30 [29 30]	28 [27 28]	9 [9 10]	38 [37 38]	1 [1 1]
Chronic	2 031 [1 989 2 071]	1 040 [1 019 1 061]	973 [953 992]	2 494 [2 442 2 543]	57 [56 58]
Total	2 061 [2 018 2 102]	1 068 [1 046 1 089]	982 [962 1 002]	2 531 [2 479 2 582]	58 [57 59]

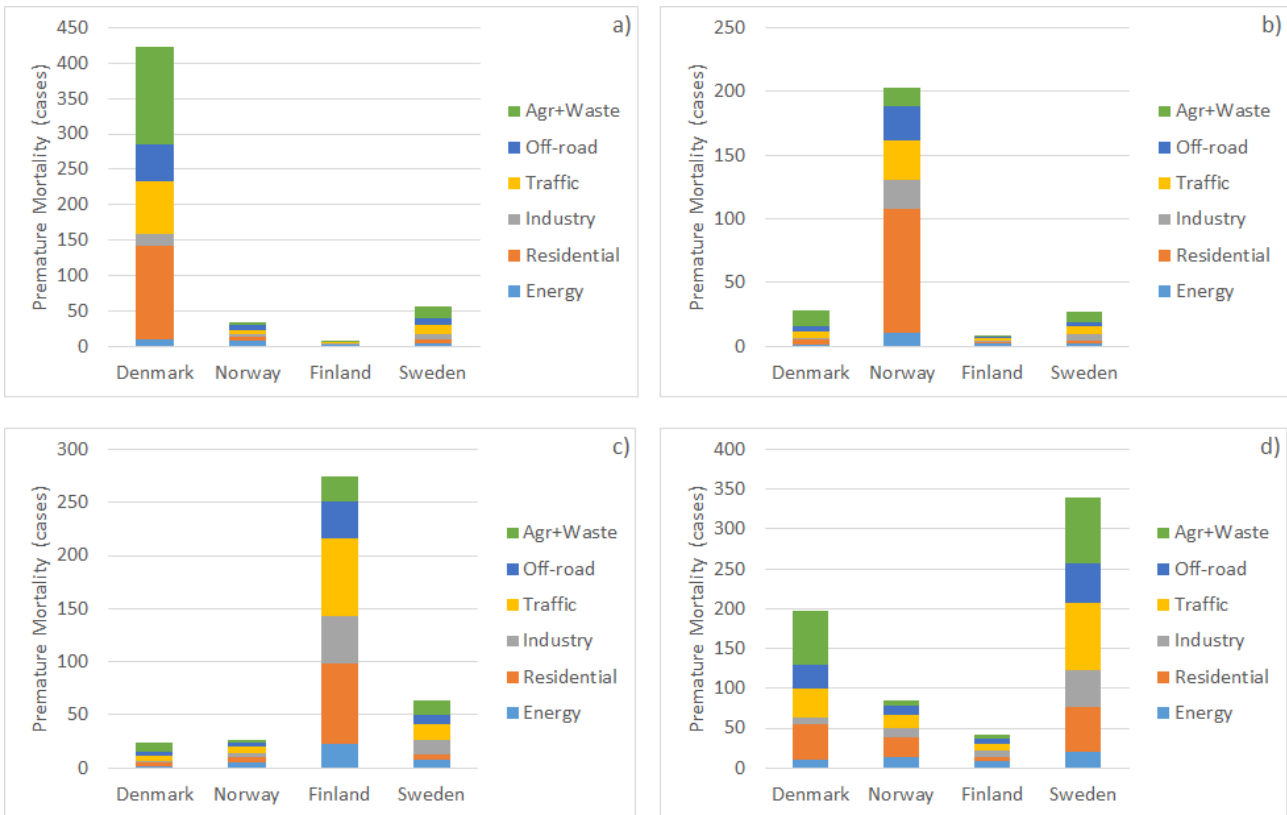
559
 560 The EVA model has been used to calculate the contributions of Nordic emissions to the total
 561 premature mortality (acute + chronic) in the Nordic countries for the year 2015. Table 6 presents a
 562 source/receptor matrix of the contributions to premature mortality on the Nordic countries. Danish
 563 emissions contribute to ~400 premature deaths in Denmark, dominated by agriculture (33%), non-
 564 industrial combustion (31%) and traffic (18%). In Norway, the dominating sector contributing is
 565 non-industrial combustion, responsible for 48% of the ~200 premature deaths in Norway. In
 566 Finland, the total number of premature deaths in 2015 is calculated to be ~270, where non-industrial
 567 combustion and traffic are responsible for more than half. Finally, in Sweden, traffic and waste
 568 management/agriculture are responsible for 50% of the total premature death in Sweden (~330).
 569

570 Table 6. Source/Receptor relationships of the contributions of anthropogenic emissions from the
 571 Nordic countries to the premature mortality in the Nordic area. The brackets show the 95%
 572 confidence interval.
 573

Source/Receptor	Denmark	Finland	Norway	Sweden
Denmark	422 [414 431]	24 [23 24]	29 [28 29]	198 [194 202]
Finland	8 [8 8]	274 [269 280]	9 [9 9]	42 [41 43]
Norway	33 [33 34]	26 [26 27]	203 [199 207]	86 [84 87]
Sweden	57 [55 58]	64 [63 65]	27 [26 28]	340 [333 346]

574
 575
 576 Figure 13 shows the contributions of sectoral emissions from each Nordic country to the total
 577 premature death cases in 2015 in the different Nordic countries. Overall, Nordic countries
 578 contribute to low premature death cases in their Nordic neighbours (≤50). As seen in the figure,
 579 agriculture and waste management sectors together can have significant share in the premature
 580 mortality (e.g. Denmark) due to the dominant contribution of NH₄ aerosols in the region (Figure 4).

581 The largest transboundary contribution is calculated for the Danish emissions, dominated by
 582 agriculture, non-industrial combustion and traffic, contributing to ~200 premature death cases in
 583 Sweden.
 584



585
 586
 587 Figure 13. Source contributions from the anthropogenic emissions of a) Denmark, b) Norway, c)
 588 Finland, and d) Sweden to total premature mortality (acute+chronic) in the Nordic countries.
 589

590 Table 7 shows the cost of air pollution on human health in each of the Nordic countries in the
 591 source country and the neighbouring Nordic countries. Among the four Nordic countries, Denmark
 592 has the largest external costs due to air pollution, followed by Sweden, Finland and Norway,
 593 respectively. Following the mortality rates, Denmark, Finland and Norway have the largest cost
 594 contribution to Sweden, while Sweden contributes largest to Denmark.
 595
 596
 597

598 Table 7. Contribution of costs (million €) of air pollution impacts on human health in the Nordic
 599 countries. The brackets show the 95% confidence interval.
 600

Source	Receptors			
	Denmark	Finland	Norway	Sweden
Denmark	261 [256 266]	14 [14 15]	17 [17 18]	122 [119 124]
Finland	5 [5 5]	172 [169 176]	6 [5 6]	26 [26 27]
Norway	20 [20 21]	16 [16 16]	126 [123 128]	53 [51 54]
Sweden	36 [35 36]	39 [39 40]	17 [16 17]	212 [207 216]

601
 602 Regarding the costs attributed to each of the source sectors, Figure S1 summarizes the contributions
 603 per country. For Denmark, results suggest that non-industrial combustion and agriculture/waste
 604 management are the main sectors to be targeted to reduce the negative impacts of air pollution. In
 605 Norway, reduction of non-industrial combustion emissions alone can substantially reduce the costs
 606 of air pollution. In Finland, similar to Denmark and Norway, non-industrial combustion should be
 607 targeted for developing emission reduction strategies, along with the traffic emissions, which
 608 contribute as large as the non-industrial combustion. Finally, in Sweden, traffic and
 609 agriculture/waste management sectors should be targeted to reduce the adverse impacts of air
 610 pollution and their associated costs. However, as the local contributions to air pollutants are
 611 generally low in the region, it should be noted that significant reductions can only be achieved by
 612 reducing the emissions upwind, which would require a coordinated effort in Europe.

613
 614 4. Conclusions
 615

616 The sectoral contributions of land-based anthropogenic emission sources in the four Nordic
 617 countries; Denmark, Finland, Norway and Sweden, on air pollution levels and premature mortality
 618 in these countries and over the Arctic have been estimated using the DEHM/EVA impact
 619 assessment system for the year 2015. The chemistry and transport model, DEHM, was run with
 620 tagging mode in order to calculate inline the sectoral contributions based on 30% reductions of each
 621 sector separately. Using the modelled surface concentrations of O₃, SO₂ and PM_{2.5}, the EVA model
 622 calculated the acute (O₃ and SO₂) and chronic (PM_{2.5}) premature mortality due to exposure to these
 623 pollutants.
 624

625 Results show that the Nordic countries are responsible for 5-10% of the regional background
 626 surface PM_{2.5} concentrations in the countries itself. The non-industrial combustion (SNAP2), which
 627 is dominated by the non-industrial wood combustion, is responsible for 50% to 80% of the
 628 contribution to surface PM_{2.5} in the Nordic countries. In Denmark, Finland and Norway, non-
 629 industrial combustion contributes largely to surface OC (by 60% - 80%). In Sweden, SNAP2 is
 630 responsible for 43% of the contribution to surface OC, while 43% comes from industrial activities.
 631 Similar to OC, BC is also dominated by non-industrial combustion (by 50%-65%), except for
 632 Sweden, where 25% originates from non-industrial combustion and 31% from industrial activities.
 633 The dominant source for surface SO₄ and SO₂ in all four Nordic countries is calculated to be
 634 industrial activities. In Norway and Sweden, around 70% of SO₂ are coming from industrial
 635 activities, while in Denmark and Finland, industrial activities are responsible for around 30% of
 636 SO₂. Off-road traffic is responsible for 21% of SO₂, while energy production is responsible for 50%
 637 of SO₂ in Finland. Industrial activities are also responsible for 60% of SO₄ in Norway and Sweden
 638 and 30% in Denmark and Finland. The dominant source for NO₂ is calculated to mobile sources,

639 and the share between on-road and off-road traffic varies depending on the country. Almost 35% of
640 NO₂ comes from on-road traffic in all four Nordic countries while off-road traffic contributes by
641 25% to 35%.

642
643 Norway has the largest contribution to aerosol levels over the Arctic, while Denmark has the lowest
644 contribution, although contributions are only a few percent. Non-industrial combustion in the
645 Nordic countries is also the largest contributor to Arctic OC and BC levels, except for Sweden,
646 where industry plays a more important role in relation to the Arctic levels. Agriculture and waste
647 treatment facilities over the Nordic countries are responsible contribute to the sulfate levels over the
648 Arctic.

649
650 Anthropogenic emissions lead to a titration of around 4-5%, particularly over the source countries
651 and lead to a very small surface O₃ increase (>1%) in the downwind regions. The largest impacts
652 are calculated to be around the capital regions. Danish emissions also impact the surface PM_{2.5}
653 levels over the southern part of Sweden and Norway, by around 3%. Finnish emissions also have a
654 small impact, lower than 3%, on the central part of Sweden and northern parts of Norway.
655 Norwegian anthropogenic emissions impacts PM_{2.5} levels over Sweden by around 7% while
656 Swedish anthropogenic emissions contribute to PM_{2.5} levels over the southwestern parts of Finland,
657 by up to 5%. It should be noted that these results are calculated for a specific year, 2015, therefore
658 transport from one country to others can significantly vary in different years due to meteorology, in
659 particular wind speed and direction.

660
661 The total number of premature mortality cases due to air pollution are calculated to be ~4 000 in
662 Denmark and Sweden and ~2 000 in Finland and Norway, leading to a total cost of 7 billion Euros
663 in the selected Nordic countries. The contributions of emission sectors to premature mortality in
664 each of the Nordic countries vary. Danish agriculture and industrial emissions contribute similarly
665 (by 33%) to ~400 premature mortality cases in Denmark, that are due to the Danish emissions. In
666 Norway, non-industrial combustion, dominated by non-industrial wood combustion, is responsible
667 for 48% of the ~200 premature deaths in Norway due to the exposure to pollution from the Nordic
668 sources. In Finland, non-industrial combustion and traffic are responsible for more than half of the
669 ~270 premature deaths in 2015, caused by the sources within the region. Finally, in Sweden, traffic
670 and waste management/agriculture are responsible for 50% of the total premature death in Sweden
671 (~330), caused by the emissions in the Nordic region. In Denmark, Finland and Norway, non-
672 industrial combustion is the main sectors to be targeted to reduce the negative impacts of air
673 pollution, while in Sweden, traffic and agriculture/waste management sectors should be targeted to
674 reduce the adverse impacts of air pollution and their associated costs. Overall, Nordic countries
675 contribute to low premature death cases in their Nordic neighbours (≤50). Among the four Nordic
676 countries, Denmark has the largest external costs due to air pollution, followed by Sweden, Finland
677 and Norway, respectively. Following the mortality rates, Denmark, Finland and Norway have the
678 largest cost contribution to Sweden, while Sweden contributes largest to Denmark.

679
680 Overall, results from the estimates of pollution export, premature mortality and associated costs
681 suggest that in the Nordic countries, non-industrial combustion, which is dominated by non-
682 industrial wood combustion, together with industry and traffic are the main sectors to be targeted
683 for emission mitigation strategies. The contributions of emissions from Nordic countries to each
684 other are small (≤10%), and to the Arctic (up to 2%), meaning that large reductions can be achieved
685 only by coordinated efforts to decrease emissions in the upwind countries.
686

687 **Author Contribution**

688

689 UI and JHC conducted the model simulations. JHC and OKN worked with the emissions input. MS
690 and RM contributed to the experimental design of the model simulations. UI, JK, CA and SL-A
691 extracted measurement data from Denmark, Finland, Sweden and Norway, respectively. CG and JB
692 contributed to premature mortality and cost calculations. All co-authors contributed to the
693 manuscript.

694

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696

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703

704

705 **REFERENCES**

706

707 Aas W., Markus F., Solberg S., and Yttri, K.E.: Monitoring of long-range transported air pollutants
708 in Norway, Annual Report 2017. NILU report 10/2018, NILU, Kjeller, 2018.

709

710 Alberini, A., Hunt, A., and Markandya, A.: Willingness to pay to reduce mortality risks: Evidence
711 from a three-country contingent valuation study, *Environ. Resour. Econ.*, 33, 251–264, 2006.

712

713 Andersson, C., Bergström, R. and Johansson, C.: Population exposure and mortality due to regional
714 background PM in Europe – long-term simulations of source-region and shipping contributions,
715 *Atmospheric Environment* 43, 3614-3620, 2009.

716

717 Andersen, M. S., Frohn, L. M., Nielsen, J. S., Nielsen, M., Jensen, S. S., Christensen, J. H., and
718 Brandt, J.: A Non-linear Eulerian Ap- proach for Assessment of Health-cost Externalities of Air
719 Pollu- tion, Proceedings of the European Association of Environmental and Resource Economists
16th Annual Conference, Gothenburg, Sweden, 25–28 June 2008, 23 pp., 2008

720

721 Anderson, H.R., Ponce de Leon, A., Bland, J.M., Bower, J.S. and Strachan, D.P. Air Pollution and
722 daily mortality in London: 1987-92. *British Medical Journal*, 312, 665-669, 1996.

722

723 Arctic Council: Arctic Council Task Force on Short-Lived Climate Forcers - An Assessment of
724 Emissions and Mitigation Options for Black Carbon for the Arctic Council, available at:
725 <https://oaarchive.arctic-council.org/handle/11374/926> (last access: 26 February 2019), 2011.

726

727 Brandt, J., Silver, J. D., Christensen, J. H., Andersen, M. S., Bønløkke, J. H., Sigsgaard, T., Geels,
728 C., Gross, A., Hansen, A. B., Hansen, K. M., Hedegaard, G. B., Kaas, E., and Frohn, L. M.:
729 Contribution from the ten major emission sectors in Europe and Denmark to the health-cost
730 externalities of air pollution using the EVA model system – an integrated modelling approach,
731 *Atmos. Chem. Phys.*, 13, 7725–7746, <https://doi.org/10.5194/acp-13-7725-2013>, 2013a.

732

733 Brandt, J., Silver, J. D., Christensen, J. H., Andersen, M. S., Bønløkke, J. H., Sigsgaard, T., Geels,
734 C., Gross, A., Hansen, A. B., Hansen, K. M., Hedegaard, G. B., Kaas, E., and Frohn, L. M.:
735 Assessment of past, present and future health-cost externalities of air pollution in Europe and the
736 contribution from international ship traffic using the EVA model system, *Atmos. Chem. Phys.*, 13,
737 7747–7764, <https://doi.org/10.5194/acp-13-7747-2013>, 2013b.
738

739 Brandt, J., Silver, J. D., Frohn, L. M., Geels, C., Gross, A., Hansen, A. B., Hansen, K. M.,
740 Hedegaard, G. B., Skjøth, C. A., Villadsen, H., Zare, A., and Christensen, J. H.: An integrated
741 model study for Europe and North America using the Danish Eulerian Hemispheric Model with
742 focus on intercontinental transport, *Atmos. Environ.*, 53, 156–176, 2012.
743

744 CEIP: Centre for Emission Inventories and Projections: Reported Emissions by
745 Parties under the Convention for Long-range Transboundary Air Pollution.
746 <http://www.ceip.at/webdab-emission-atabase/officially-reported-emissiondata/>
747 (last access: 26 February 2019), 2019.
748

749 EEA: Air quality in Europe, Technical report 13/2017, Copenhagen, European Environment
750 Agency, ISSN 1977-8449, 2017.
751

752 Ellermann, T., Nygaard, J., Nøjgaard, J.K., Nordstrøm, C., Brandt, J., Christensen, J., Ketzel, M.,
753 Massling, A., and Jensen, S.S.: The Danish Air Quality Monitoring Programme. Annual Summary
754 for 2015. Aarhus University, DCE – Danish Centre for Environment and Energy, 65 pp. Scientific
755 Report from DCE – Danish Centre for Environment and Energy No. 201, 2016.
756

757 European Commission (EC): Recommended interim values for the value of preventing a fatality in
758 DG Environment Cost Benefit analysis, Bruxelles, available at:
759 http://ec.europa.eu/environment/enveco/others/pdf/recommended_interim_values.pdf (last access:
760 14 March 2019), 2001.
761

762 EU Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on
763 ambient air quality and cleaner air for Europe. *Off. J. Eur. Com.*, L 141, 11/06/2008, 1-44, 2008.
764

765 Forsberg, B., Hansson, H.C., Johansson, C., Aureskou, H., Persson, K., and Järholm, B.:
766 Comparative health impact assessment of local and regional particulate air pollutants in
767 Scandinavia. *Ambio*, 34, 11-19, 2005.
768

769 Friedrich, R. and Bickel, P.: *Environmental External Costs of Transport*, Springer, München, 2001.
770

771 Geels, C., Andersson, C., Hänninen, O., Lansø, A. S., Schwarze, P., and Brandt, J.: Future
772 Premature Mortality due to Air Pollution in Europe – Sensitivity to Changes in Climate,
773 Anthropogenic Emissions, Population and Building stock, *Int. J. Env. Res. Pub. He.*, 12, 2837–
774 2869, 2015.
775

776 Im, U., Christensen, J. H., Geels, C., Hansen, K. M., Brandt, J., Solazzo, E., Alyuz, U., Balzarini, A.,
777 Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A., Curci, G., Farrow, A., Flemming, J.,
778 Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liu, P., Nopmongkol, U., Palacios-Peña, L.,
779 Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A., Vivanco, M. G.,
780 Yarwood, G., Hogrefe, C., and Galmarini, S.: Influence of anthropogenic emissions and boundary

781 conditions on multi-model simulations of major air pollutants over Europe and North America in the
782 framework of AQMEII3, *Atmos. Chem. Phys.*, 18, 8929-8952, [https://doi.org/10.5194/acp-18-8929-](https://doi.org/10.5194/acp-18-8929-2018)
783 2018, 2018b.

784

785 Im, U., Brandt, J., Geels, C., Hansen, K. M., Christensen, J. H., Andersen, M. S., Solazzo, E.,
786 Kioutsioukis, I., Alyuz, U., Balzarini, A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A.,
787 Curci, G., Farrow, A., Flemming, J., Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liang, C.-K.,
788 Nopmongcol, U., Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A.,
789 Vivanco, M. G., West, J., Yarwood, G., Hogrefe, C., and Galmarini, S.: Assessment and economic
790 valuation of air pollution impacts on human health over Europe and the United States as calculated
791 by a multi-model ensemble in the framework of AQMEII3, *Atmos. Chem. Phys.*, 18, 5967-5989,
792 <https://doi.org/10.5194/acp-18-5967-2018>, 2018a.

793

794 Jalkanen, J.-P., Johansson, L., and Kukkonen, J.: A comprehensive inventory of ship traffic exhaust
795 emissions in the European sea areas in 2011. *Atmos. Chem. Phys.*, 16, 71–84, 2016.

796

797 Johansson, L., Jalkanen, J.-K., Kukkonen, J.: Global assessment of shipping emissions in 2015 on a
798 high spatial and temporal resolution. *Atmospheric Environment*, Volume 167, October 2017, Pages
799 403-415, 2017.

800

801 Jönsson, O., Andersson, C., Forsberg, B. and Johansson, C.: Health impacts and air pollution
802 episodes in Stockholm regional background air due to European source regions, *Boreal*
803 *Environment Research* 18, 280-302, 2013.

804

805 Hurley, F., Hunt, A., Cowie, H., Holland, Miller, B., Pye, S. and Watkiss, P. Development of
806 Methodology for the CBA of the Clean Air For Europe (CAFE) Programme, Volume 2: Health
807 Impact Assessment, Report for European Commission DG Environment, 2005.

808

809 Im, U., Christensen, J. H., Geels, C., Hansen, K. M., Brandt, J., Solazzo, E., Alyuz, U., Balzarini,
810 A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A., Curci, G., Farrow, A., Flemming, J.,
811 Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liu, P., Nopmongcol, U., Palacios-Peña, L.,
812 Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A., Vivanco, M. G.,
813 Yarwood, G., Hogrefe, C., and Galmarini, S.: Influence of anthropogenic emissions and boundary
814 conditions on multi-model simulations of major air pollutants over Europe and North America in
815 the framework of AQMEII3, *Atmos. Chem. Phys.*, 18, 8929-8952, [https://doi.org/10.5194/acp-18-](https://doi.org/10.5194/acp-18-8929-2018)
816 8929-2018, 2018b.

817

818 Im, U., Brandt, J., Geels, C., Hansen, K. M., Christensen, J. H., Andersen, M. S., Solazzo, E.,
819 Kioutsioukis, I., Alyuz, U., Balzarini, A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette,
820 A., Curci, G., Farrow, A., Flemming, J., Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liang, C.-
821 K., Nopmongcol, U., Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal,
822 A., Vivanco, M. G., West, J., Yarwood, G., Hogrefe, C., and Galmarini, S.: Assessment and
823 economic valuation of air pollution impacts on human health over Europe and the United States as
824 calculated by a multi-model ensemble in the framework of AQMEII3, *Atmos. Chem. Phys.*, 18,
825 5967-5989, <https://doi.org/10.5194/acp-18-5967-2018>, 2018b.

826

827 Karvosenoja, N., Kangas, L., Kupiainen, K., Kukkonen, J., Karppinen, A., Sofiev, M., Tainio, M.,
828 Paunu, V.-V., Ahtoniemi, P., Tuomisto, J.T. and Porvari, P.: Integrated modeling assessments of

829 the population exposure in Finland to primary PM_{2.5} from traffic and domestic wood combustion
830 on the resolutions of 1 and 10 km, *Air Qual. Atmos. Health*, 4, 3–4, 179–188, 2011.
831

832 Krewski, D., Jerrett, M., Burnett, R.T., Ma, R., Hughes, E., Shi, Y., Turner, M.C., Arden Pope III,
833 C., Thurston, G., Calle, E.E. and Thun, M.J. Extended Follow-Up and Spatial Analysis of the
834 American Cancer Society Study Linking Particulate Air Pollution and Mortality. *Health Effects*
835 *Institute Research Report*, 140, 1–154, 2009.
836

837 Kukkonen, J., López-Aparicio, S., Segersson, D., Geels, C., Kangas, L., Kauhaniemi, M.,
838 Maragkidou, A., Jensen, A., Assmuth, T., Karppinen, A., Sofiev, M., Hellen, H., Riikonen, K.,
839 Nikmo, J., Kousa, A., Niemi, J. V., Karvosenoja, N., Santos, G. S., Sundvor, I., Im, U., Christensen,
840 J. H., Nielsen, O.-K., Plejdrup, M. S., Nøjgaard, J. K., Omstedt, G., Andersson, C., Forsberg, B., and
841 Brandt, J.: The influence of residential wood combustion on the concentrations of PM_{2.5} in four
842 Nordic cities, *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2019-564>, in review, 2019.
843

844 Kukkonen, J., Kangas, L., Kauhaniemi, M., Sofiev, M., Aarnio, M., Jaakkola, J.J.K., Kousa, A., and
845 Karppinen, A.: Modelling of the urban concentrations of PM_{2.5} for a period of 35 years, for the
846 assessment of lifetime exposure and health effects. *Atmos. Chem. Phys.*, 18, 8041–8064, 2018.
847

848 Lehtomäki, H., Korhonen, A., Asikainen, A., Karvosenoja, N., Kupiainen, K., Paunu, V.V.,
849 Savolahti, M., Sofiev, M., Palamarchuck, Y., Karppinen, A., Kukkonen, J., and Hänninen, O.: Health
850 Impacts of Ambient Air Pollution in Finland. *Int. J. Environ. Res. and Public Health*. 15, 736, 2018.
851

852 Liang, C.-K., West, J. J., Silva, R. A., Bian, H., Chin, M., Davila, Y., Dentener, F. J., Emmons, L.,
853 Flemming, J., Folberth, G., Henze, D., Im, U., Jonson, J. E., Keating, T. J., Kucsera, T., Lenzen, A.,
854 Lin, M., Lund, M. T., Pan, X., Park, R. J., Pierce, R. B., Sekiya, T., Sudo, K., and Takemura, T.:
855 HTAP2 multi-model estimates of premature human mortality due to intercontinental transport of air
856 pollution and emission sectors, *Atmos. Chem. Phys.*, 18, 10497-10520, <https://doi.org/10.5194/acp-18-10497-2018>, 2018.
857
858

859 Pope, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K. and Thurston, G.D. Lung
860 cancer, cardiopulmonary mortality and long-term exposure to fine particulate air pollution. *Journal*
861 *of American Medical Association*, 287 (9), 1132-1141, 2002.
862

863 Pope, C.A. Thun, M.J., Namboodiri, M.M., Dockery, D.W., Evans, J.S., Speizer, F.E. and Heath Jr,
864 C.W. Particulate air pollution as a predictor of mortality in a prospective study of US adults.
865 *American Journal of Respiratory and Critical Care Medicine*, 151, 669-674, 1995.
866

867 Sand, M., Berntsen, T.K., von Salzen, K., Flanner, M.G. and Viktor, D.G.: Response of Arctic
868 temperature to changes in emissions of short-lived climate forcers, *Nature Climate Change*, 6, 286-
869 289, 2015.
870

871 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Wang, W., and Powers, J. G.:
872 A description of the Advanced Research WRF Version 3, Technical report, NCAR, nCAR Tech
873 Notes-468+STR, 2008.
874

875 Solazzo, E., Riccio, A., Van Dingenen, R., and Galmarini, S.: Evaluation and uncertainty estimation
876 of the impact of air quality modelling on crop yields and premature deaths using a multi-model
877 ensemble, *Sci. Total Environ.*, 663, 1437–1452, 2018.
878

879 Touloumi, G., Samoli, E. and Katsuyanni, K. Daily mortality and "winter type" air pollution in
880 Athens, Greece - a time series analysis within the APHEA project. *Journal of Epidemiology and*
881 *Community Health*, 50 (suppl 1), S47 - S51, 1996.
882

883 Turner, M.: Long-Term Ozone Exposure and Mortality in a Large Prospective Study. *Am. J.*
884 *Respir. Crit. Care Med.* 193:1134–1142; doi: 10.1164/rccm.201508-1633OC, 2016.
885

886 Watkiss, P., Pye, S., and Holland, M.: Cafe CBA: Baseline Analysis 2000 to 2020. Service Contract
887 for Carrying out Cost-Benefit Analysis of Air Quality Related Issues, in Particular in the Clean Air
888 for Europe (Cafe) Programme, available at:
889 http://ec.europa.eu/environment/archives/cafe/activities/pdf/cba_baseline_results2000_2020.pdf
890 (last access: 29 February 2019), 2005.
891

892 WHO: 7 million premature deaths annually linked to air pollution, News release, World Health
893 Organization, available at: <http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/>
894 (last access: 26 February 2019), 2014.
895

896 WHO: Health risks of air pollution in Europe – HRAPIE: Recommendations of concentration-
897 response functions for cost-benefit analysis of particulate matter, ozone and nitrogen dioxide,
898 World Health Organization, available at:
899 [http://www.euro.who.int/__data/assets/pdf_file/0006/238956/Health_risks_air_pollution_HRAPIE_](http://www.euro.who.int/__data/assets/pdf_file/0006/238956/Health_risks_air_pollution_HRAPIE_project.pdf?ua=1)
900 [project.pdf?ua=1](http://www.euro.who.int/__data/assets/pdf_file/0006/238956/Health_risks_air_pollution_HRAPIE_project.pdf?ua=1) (last access: 14 March 2019), 2013.
901

902 Woodruff, T.J., Grillo, J. and Schoendorf, K.C. The relationship between selected causes of
903 postneonatal infant mortality and particulate air pollution in the United States. *Environmental*
904 *Health Perspectives*, 105, 608-612, 1997.
905
906