Second review of “Contributions of Nordic anthropogenic emissions on 1 air pollution and premature mortality over the Nordic region and the Arctic” by Im et al.

The authors have adequately addressed most issues raised in the first review, however there are still a number of inaccuracies and errors to be corrected.

Response: We thank the reviewer for the comments. We have tried to answer all the issues raised by the reviewer now and hope that the manuscript is now suitable for publication.

Major issue:

Comment: Fig. 4 shows clearly the contribution of seasalt as a major PM2.5 component. Obviously, this contribution is not part of the considered anthropogenic sectors and cannot be regulated. It is not clear from the manuscript if the large ‘Rest’ contribution in Fig. 5 includes seasalt, or if only anthropogenic sectors are considered. This should be made clear in the text, and if seasalt is indeed included in Fig. 5, it sheds a different light on the ‘external’ contribution to PM2.5 and on the potential of Nordic versus external air quality control measures.

Response: Figure 5 does not include sea-salt. We have now updated the text accordingly (Lines 370-373).

Minor comments:

Comment: L63: originating from south-western Europe

Response: Modified.

Comment: L. 96 – 97: The EU limit level for annual mean PM2.5 is 25µg/mÑ (not 40). The WHO limit value is 10µg/mÑ (not 20).

Response: Corrected.

Comment: Section 2.1: Please mention whether residual water is included in the modelled PM2.5 mass or if it is dry PM2.5 (monitoring stations determine the mass at a standard RH of 50% which in theory would dry pure ammonium salts, however in reality the more complex chemical mixture retains some water).

Response: We have updated the text accordingly (Line 319).

Comment: Is there a difference between SO2 (table 1) and SOx (Figure 1)?

Response: We have corrected Table 1 accordingly.

Section 2.2 (health impacts):

Comment: L217: c should be δc

Response: Modified accordingly.

Comment: L236: (Andersen, 2008) is missing in References

Response: Reference is corrected and added to the list.

Comment: R in the equation (L216) should be specified as mortality rate or life years lost (per population); I don't see how it could represent 'days' or 'episodes'. The alpha values in Table 2 unit is cases/µgm-3/population (maybe it's more clear if the values are multiplied with a scaling factor 1E6 and expressed as cases/µgm-3/million population. I think SOMO35 should not be in the alpha formula in Table 2 (in the case of O3, SOMO35 = δc in the R equation).

Response: EVA model calculates also morbidity as well as mortality, including number of restricted days. However the present paper only considers mortality. We have modified the text accordingly (Line 217). We do keep the table as it is to be in agreement with previous publications.

Comment: Infant mortality: should be < 9 months instead of > 9 months in Table 2.

Response: This is corrected.

Comment: In this section, long-term health impact from PM is expressed in YOLL, however in Table 5 they are presented as number of premature deaths. Please make this consistent.

Response: We have now updated the text accordingly (Lines 237-238).

Comment: Section 3.1 (Evaluation): the authors did not address the request to include as well mean concentrations for O3 and PM2.5 in Table 4. Some values are given in the text in L357 – 359. This should be moved to the validation section and preferably introduced in the table. Same for model results for O3 (and NO2 and SO2 in the SI).

Response: We have now added the observed concentrations in the tables.

Comment: L311: where the overestimations are higher => where values are overestimated

Response: Modified accordingly.

Comment: L312: die to => due to

Response: Corrected.

Comment: L 316-317 "Differences ... can be attributed to ... " : basically repeats what was written in L315.

Response: We have now modified this part (Lines 316-319).

Section 3.2:

Comment: L371: downwind => upwind (also in L602)

Response: Corrected.

Comment: L485: leads => lead

Response: Corrected.

Figure 11: please include country borders

Response: Figures are updated.

Comment: L537: is almost responsible for => is responsible for almost

Response: Corrected.

Comment: Table 5 and 6: What are the values in brackets? Describe in the caption. Presumably confidence intervals? Based on what? It's hard to believe that the model can predict mortalities with a 2% accuracy!

Response: We have now updated the captions and the text accordingly (Lines 540-543).

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

Ulas Im[1,2], Jesper H. Christensen[1,2], Ole-Kenneth Nielsen[1,2], Maria Sand[3], Risto Makkonen[4,5], Camilla Geels[1,2], Camilla Anderson[6], Jaakko Kukkonen[4], Susana Lopez-Aparicio[7], Jørgen Brandt[1,2]

1 Aarhus University, Department of Environmental Science, Atmospheric Modelling Section, Frederiksborgvej 399, Roskilde, Denmark.
2 Interdisciplinary Center for Climate Change (iCLIMATE), Frederiksborgvej 399, Roskilde, Denmark.
3 Center for International Climate Research, Postboks 1129 Blindern, 0318 Oslo, Norway.
4 Finnish Meteorological Institute, Erik Palmenin aukio 1, P.O.Box 503, FI-00101, Helsinki, Finland.
5 University of Helsinki, Institute for Atmospheric and Earth System Research, P.O. Box 64, 00014, Helsinki, Finland.
6 Swedish Meteorological and Hydrological Institute, SE-60176 Norrköping, Sweden.
7 NILU - Norwegian Institute for Air Research, Instituttveien 18, P.O. Box 100, 2027 Kjeller, Norway.

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.

Introduction

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

Deleted: es

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

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

The Nordic countries are generally characterized among the EU countries with low air pollution levels (EEA, 2018). PM$_{2.5}$ levels are below the EU legislated limit value of 25 µg m^{-3} as well as the

Deleted: 40

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

Deleted: 2

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

1. Materials and methods

2.1. Danish Eulerian Hemispheric Model (DEHM)

The DEHM model was originally developed mainly to study the transport of SO2 and SO4 to the Arctic (Christensen 1997), but has been extended to different applications during the last decades. It has been documented extensively in Brandt et al. (2012) and evaluated in several intercomparison studies (e.g. Solazzo et al., 2012 a,b; Solazzo et al., 2017; Im et al., 2018a,b) and recently joined the suit of operational models in the Copernicus Atmospheric Monitoring System (CAMS) to provide regional forecasts of air pollution over Europe. The DEHM model uses a 150 km×150 km spatial resolution over the Northern Hemisphere, then nests to 50 km×50 km resolution over Europe, extending up to 100 hPa through 29 vertical levels, with the first layer height of approximately 20 m. The meteorological fields were simulated by the Weather Research and Forecast Model (WRF, Skamarock et al., 2008) setup with identical domains and resolution. The time resolution of the DEHM model is one hour. The gas-phase chemistry module includes 58 chemical species, 9 primary particles, including natural particles such as sea-salt and 122 chemical reactions (Brandt et al., 2012). The model also describes atmospheric transport and chemistry of lead, mercury, CO$_2$, as well as POPs. Secondary organic aerosols (SOA) are calculated using the Volatility Base System (VBS: Bergstrom et al., 2012). In addition to the anthropogenic PM and SOA due to biogenic emissions, DEHM model also calculates sea-salt emissions and their transport and interactions with other pollutants. The current version of the DEHM model does not include wind-blown or re-suspended dust emissions. DEHM model does not output a $PM_{2.5}$ or PM_{10} diagnostic, however these are calculated off-line, using all anthropogenic and natural components of PM, in order to be used in the health impact assessment described in Section 2.2.

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

As seen in Figure 1, non-industrial combustion (orange bars), where non-industrial combustion dominates, stands out as a major source contributing to CO and PM emissions while industry (grey bars) (Table 2) is the largest source of NMVOCs, NOx and SOx. Traffic (yellow bars) also contributes significantly to CO and NOx. The largest source of NH_3 is from agriculture in particular, as well as waste management (green bars) (Table 2).

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

	CO	NH_3	NMVOC	NOx	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

Deleted: 2

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

2.1.1. Tagging Method

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

2.1.2. Model evaluation

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

2.2. Economic Valuation of Air Pollution (EVA) System

The EVA system (Brandt et al., 2013a,b; Geels et al., 2015; Im et al., 2018a) is based on the impact-pathway chain method (Friedrich and Bickel, 2001). The EVA system can estimate acute (short-term) and chronic (long-term) mortality, related to acute exposure to O_3, and SO_2, and chronic exposure to $PM_{2.5}$, and the associated external costs. The EVA system requires gridded concentrations along with gridded population data, exposure-response functions (ERFs) for health impacts, which are recommended by the WHO (2013), and economic valuation functions of the impacts from air pollution. In addition, EVA uses population densities over fixed age intervals, corresponding to babies (under one year), children (under 15), adults (above 15 and above 30), and elderlies (above 65). The impacts of short-term exposure to O_3, and SO_2, and the long-term exposure to $PM_{2.5}$ are well established. EVA uses the annual mean concentrations of SO_2, and $PM_{2.5}$, while for O_3, it uses the SOMO35 metric that is defined as the annual sum of the daily maximum of 8-hour running average over 35 ppb, following WHO (2013) and EEA (2017).

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

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

where R is the response of the mortality rate or the years of life lost (in cases or days), δ_c denotes the pollutant concentration, P denotes the affected share of the population, and α an empirically determined constant for the particular health outcome. EVA uses ERFs that are modelled as a linear function, which is a reasonable approximation for the region of interest in the present study, as showed in several studies (e.g. Pope et al., 2000; the joint World Health Organization/UNECE Task Force on Health (EU, 2004; Watkiss et al., 2005)). However, some studies showed non-linear relationships, being steeper at lower than at higher concentrations (e.g. Samoli et al., 2005). Therefore, linear relationships may lead to overestimated health impacts over highly polluted areas. Exposure response functions (ERF) for all-cause chronic mortality due to $PM_{2.5}$ are based on Pope et al., 2002; Krewski et al., 2009), which are also recommended by the WHO (2013). These are the most extensive and up-to-date data, although there are ongoing studies in Europe, and in particular in the Nordic region to develop regional-specific ERFs (e.g. the Nordic WelfAir project: https://projects.au.dk/nordicwelfair/). The current version of the EVA system used in the present study does not include impacts due to exposure to NO_2. However, a new version is currently under development under the NordicWelfAir project.

Deleted: ,

Deleted: , or episodes

Deleted: *c*

Field Code Changed

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

Regarding short-term exposure to O_3, EVA uses the ERF recommended by the CAFE Programme (Hurley et al., 2005) and WHO (2013) that uses the daily maximum of 8-hour mean O_3 concentrations. There are also studies showing that SO_2 is associated with acute mortality, and EVA adopts the ERF identified in the APHENA study – Air Pollution and Health: A European Approach (Katsouyanni et al., 1997). Some recent studies also report the chronic effects from O_3 (e.g. Turner, 2016), however the current version of the EVA model does not include these effects. The ERFs used in EVA to calculate mortality are presented in Table 2.

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

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}	
Chronic mortality[1,4,], YOLL (PM)	1.138E-3 YOLL/μgm^{-3} (>30 years)	57,510 per YOLL
Infant mortality[5], IM (PM)	6.68E-6 cases/μgm^{-3} (<9 months)	2,298,148 per case

Deleted: >

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

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

2.3. Scenarios (response and contribution)

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

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

Table 3. Source sectors used in the perturbation scenarios.

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

3. Results and Discussion

3.1. Evaluation

Surface ozone and $PM_{2.5}$ concentrations calculated by the DEHM model have been evaluated using surface observations from the urban background and regional background monitoring stations in the Nordic countries. The comparison of the mean of all observed concentrations in each country and the corresponding modelled concentrations are presented in Table 4 while Figs. 2 and 3 present Taylor diagrams for each station in each Nordic country, giving insight to the spatial distribution of model performance. As seen in Table 3, temporal variation of O_3 levels are well reproduced by the DEHM model over all countries ($r > 0.6$), however with an overestimation of ~10% over Denmark, Finland and Sweden, and ~30% over Norway. The daily variations of $PM_{2.5}$ levels, averaged over all stations in each Nordic country are well reproduced for Denmark (r>~0.7), moderately over Norway and Sweden (r>0.4), and poorly (r~0) over Finland (Table 3). $PM_{2.5}$ concentrations are underestimated by up to 35% over Denmark, Finland and Norway, and overestimated by 8% over Sweden.

In all countries, lower *NMB* values are calculated for O_3 over the regional background stations compared to urban background stations, where values are overestimated. Regarding $PM_{2.5}$, no such conclusions can be drawn due to very limited number of regional background stations in Denmark and Norway. In Finland, lower *NMB* values for $PM_{2.5}$ are calculated for the regional background stations, while in Sweden, much lower *NMB* values are calculated for the urban stations. These differences reflect the underestimations in emissions as well as the coarse model resolution, as well as missing sources, in particular for PM, such as wind-blown and resuspended dust in the DEHM model. It should also be mentioned that the modelled PM does not contain residual water. Table S2 shows the same comparisons for NO_2 and SO_2. The underestimations in the modelled $PM_{2.5}$ levels imply an underestimated exposure to $PM_{2.5}$ levels, given the dominance of $PM_{2.5}$ in premature mortality. Similarly, the overestimations in O_3 levels can be attributed to the underestimated NO-titration (Table S2).

Deleted: the

Deleted: ions are higher

Deleted: i

Deleted: Differences in observed and modelled concentrations can be attributed to coarse model resolution as well as missing sources, in particular for PM, such as wind-blown and resuspended dust in the DEHM model.

Table 4. Model evaluation for the daily mean concentrations of O_3 and $PM_{2.5}$ for all the selected stations in the Nordic countries.

	O_3					$PM_{2.5}$				
	r	*Obs.* (µg m^{-3})	*MB* (µg m^{-3})	*NMB* (%)	*RMSE* (µg m^{-3})	*r*	*Obs.* (µg m^{-3})	*MB* (µg m^{-3})	*NMB* (%)	*RMS* (µg r
Denmark	0.81	59.59	5.67	0.09	11.60	0.75	10.77	-3.41	-0.32	
Finland	0.74	55.20	4.77	0.10	12.44	-0.03	5.05	-0.80	-0.16	
Norway	0.64	54.65	12.02	0.27	18.31	0.35	6.85	-2.56	-0.36	
Sweden	0.74	57.88	7.00	0.13	13.25	0.59	5.00	0.33	0.08	

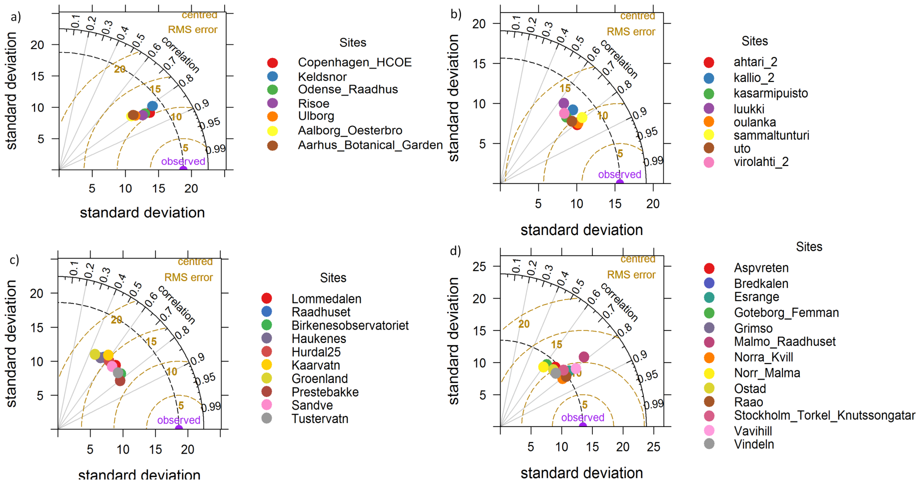


Figure 2. Taylor diagrams for daily mean O_3 for all stations in a) Denmark, b) Finland, c) Norway and d) Sweden.

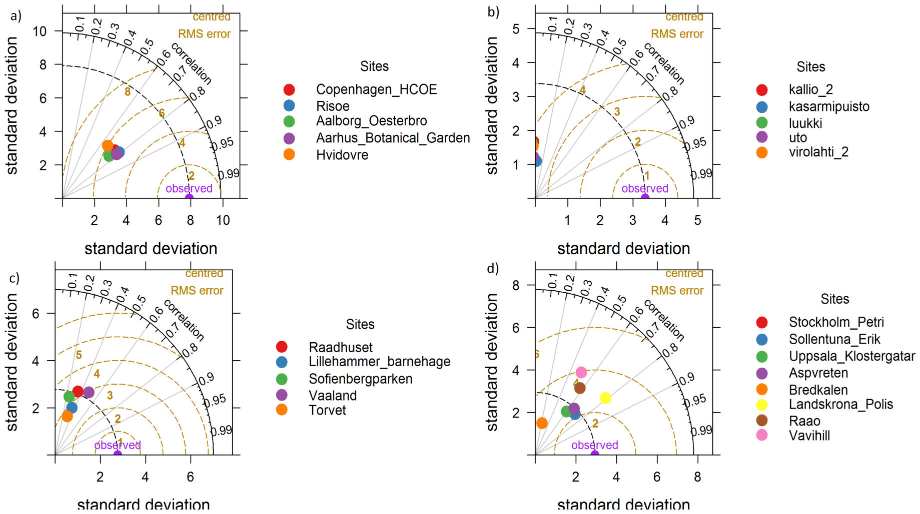


Figure 3. Taylor diagrams for daily mean $PM_{2.5}$ for all stations in a) Denmark, b) Finland, c) Norway and d) Sweden.

3.2. Sectoral contributions to surface concentrations

3.2.1. Nordic countries

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

Our simulations show that $PM_{2.5}$ mass concentrations over the Nordic countries are dominated by nitrate aerosols (30% - 45 %) and sea-salt (30% - 50%). SO_4 aerosols contribute 10 to 15% of $PM_{2.5}$ concentrations while OC contributes by 8-11%, and BC by 2-4% of the $PM_{2.5}$ mass. As SO_4 and NO_3 aerosols include NH_4 in DEHM, results suggest that NH_4 aerosols contribute by more than half of the $PM_{2.5}$ mass over the Nordic countries. The annual mean surface PM2.5 concentrations for Denmark, Finland, Norway and Sweden are calculated to be 9.1 μgm^{-3}, 4.4 μgm^{-3}, 4.8 μgm^{-3} and 5.8 μgm^{-3}, respectively. These values are in agreement with those reported by the EEA (2017), however underestimating by 12% (Denmark) up to 30% (Norway).

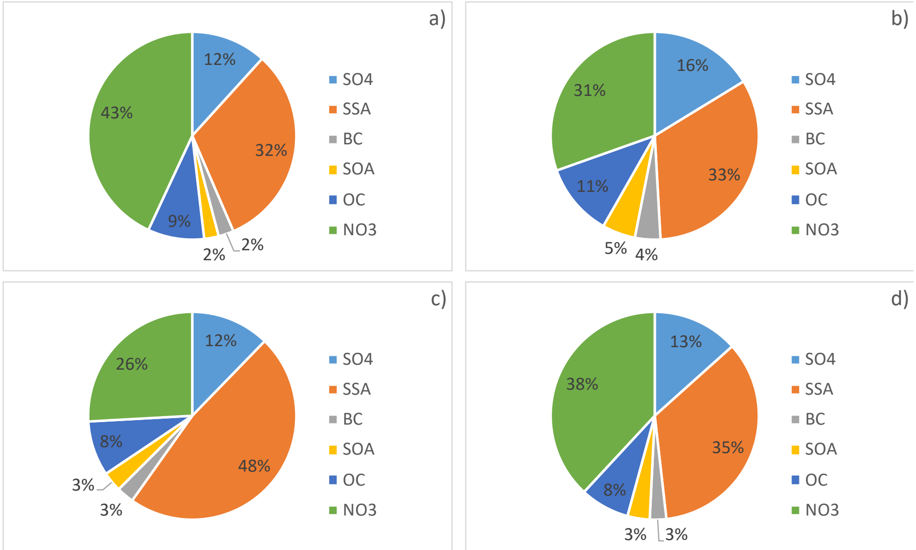
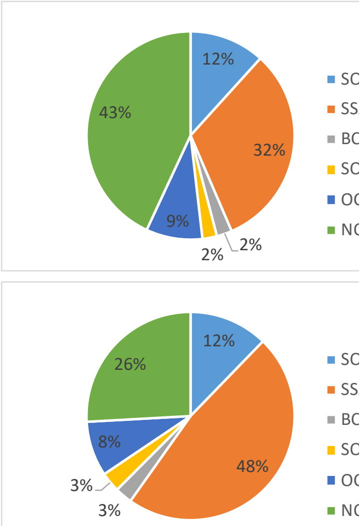


Figure 4. Simulated surface PM$_{2.5}$ chemical composition over a) Denmark, b) Finland, c) Norway, and d) Sweden.

Deleted:



Formatted: English (US)

Formatted: Danish

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 PM2.5 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 µg m^{-3} (highest in Denmark and lowest in Finland). Similar to PM$_{2.5}$, annual mean surface O$_3$ levels are also low (~30 µg m^{-3}). Similar analyses done for O$_3$ (not shown) show that O$_3$ levels are controlled largely regional, where the local sources in the Nordic countries lead to small sink of O$_3$ 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 O$_3$ is a regional background pollutant in Europe.

Formatted: Subscript

Deleted: downwind

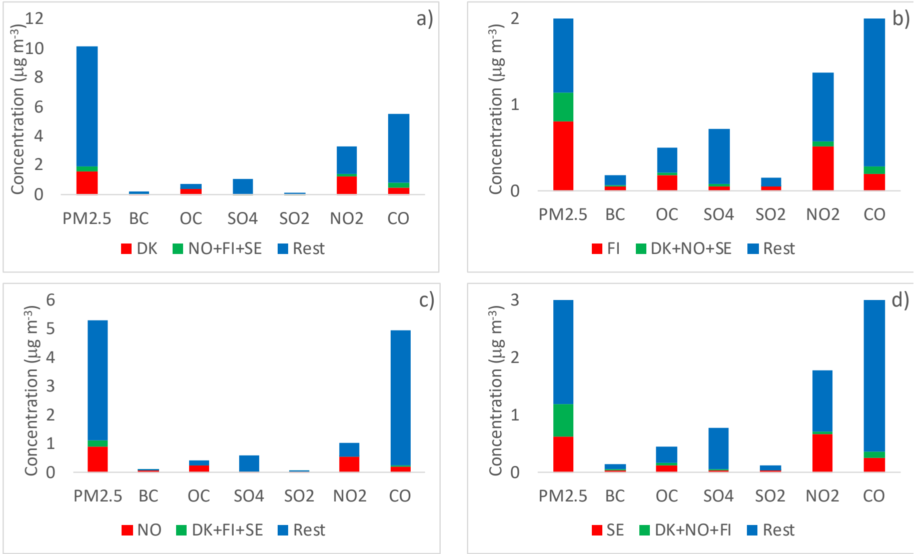
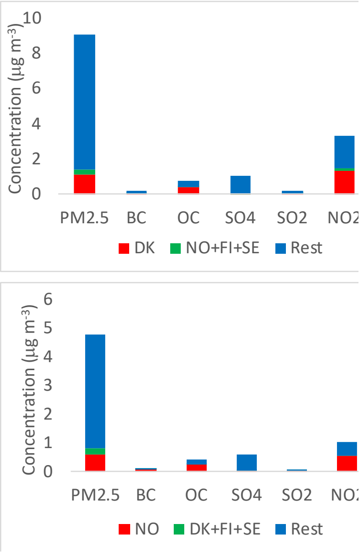


Figure 5. Absolute contributions of national, Scandinavian and other sources on the surface levels of major air pollutants over a) Denmark, b) Finland, c) Norway and d) Sweden. Note that CO concentrations are divided by 20 to scale with other pollutants.

Danish emissions contribute to only 1.14 µg m^{-3} (13%) of the surface $PM_{2.5}$ concentrations over Denmark (9.1 µg 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 µg m^{-3} (60%) of the Danish contribution to surface $PM_{2.5}$ concentrations over Denmark. Non-industrial combustion contributes to 0.22 µg 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 µg m^{-3} (35%) of the Danish contribution to the surface SO_2 concentrations over Denmark, while on-road and off-road transport contributes equally to the Danish share of the in surface NO_2 concentrations by 1.02 µg m^{-3} (~79% together). Agriculture and waste handling are important sources for surface SO_4 levels over Denmark as well as over the other Nordic countries, via the formation of ammonium sulfate ($(NH_4)_2SO_4$) due to the large ammonia (NH_3) emissions from these sectors. 0.26 µg m^{-3} of $PM_{2.5}$ over Denmark comes the other Nordic countries, with 0.03 µg m^{-3} coming from non-industrial combustion only.



Deleted:

Formatted: English (US)

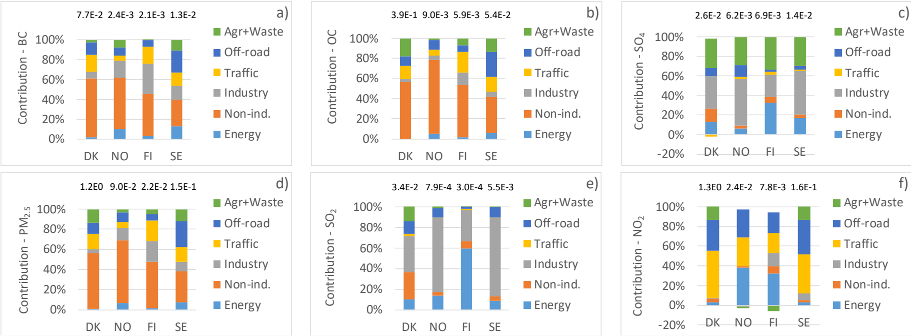


Figure 6. Population-weighted sectoral contributions of Danish emissions on surface a) BC, b) OC, c) SO_4, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Denmark.

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 µg m^{-3} (13%) of the surface $PM_{2.5}$ concentrations over Norway, while contributions to other Nordic countries are below 1%, except for NO_2, where on-road transport emissions from Norway contributes to almost 0.02 µg m^{-3} (42%) of the surface NO_2 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 µg m^{-3} (74%) of local contribution to the surface OC levels over Norway. Industry is a major source of surface SO_2 levels over Norway, contributing to 0.02 µg m^{-3} (66%) of the local contribution. 0.2 µg m^{-3} of $PM_{2.5}$ levels over Norway comes from the other Nordic countries, 0.02 µg m^{-3} being from non-residential combustion.

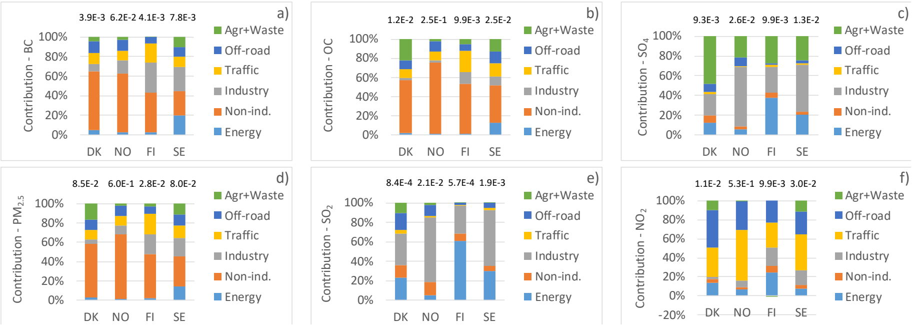


Figure 7. Population-weighted sectoral contributions of Norwegian emissions on surface a) BC, b) OC, c) SO_4, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Norway.

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 pollution over Finland, although contributions are lower compared to Denmark and Norway (0.19 µg m^{-3} (41%) of PM$_{2.5}$ and 0.11 µg m^{-3} (48%) of OC). Another noticeable difference is that energy production is also an important contributor to surface SO$_2$ (0.01. µg m^{-3}: %44) and SO$_4$ (0.03 µg m^{-3}: 44%) levels over Finland. 0.3 µg m^{-3} of PM$_{2.5}$ levels over Finland come from the other Nordic countries, 0.2 µg m^{-3} being from non-residential combustion. Finnish emissions, in particular industrial combustion, contribute largest to the air pollution over Sweden.

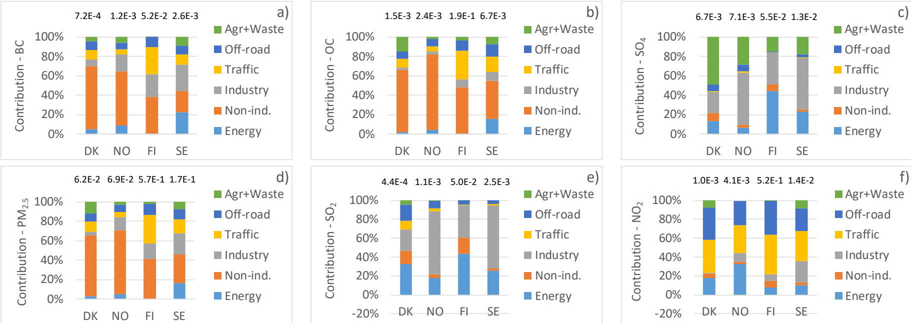


Figure 8. Population-weighted sectoral contributions of Finnish emissions on surface a) BC, b) OC, c) SO$_4$, d) PM$_{2.5}$, e) SO$_2$ and f) NO$_2$ over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Finland.

Contributions from the Swedish emission sources to surface pollutant levels over the Nordic countries are presented in Figure 9. Unlike other Nordic countries, Swedish emissions have larger contributions to pollution levels over the other Nordic countries, in particular over Norway. The figure also shows that Sweden does not experience as dominant contribution from non-industrial combustion (32%) like the other Nordic countries show. Swedish emissions from SNAP2 are much lower than for the rest of the Nordic countries (official emissions reported to the CLRTAP), most probably due to lower emission factors. Non-industrial combustion and industry contribute similarly to the surface PM$_{2.5}$ levels. Industry also has an important contribution to surface SO$_4$ levels (0.01 µg m^{-3}: 51%), as well to SO$_2$ (0.01 µg m^{-3}: 58%) and BC (0.006 µg m^{-3}: 18%). 0.5 µg m^{-3} of surface PM$_{2.5}$ levels over Sweden comes from the other Nordic countries, of which, 0.1 µg m^{-3} comes from non-residential combustion.

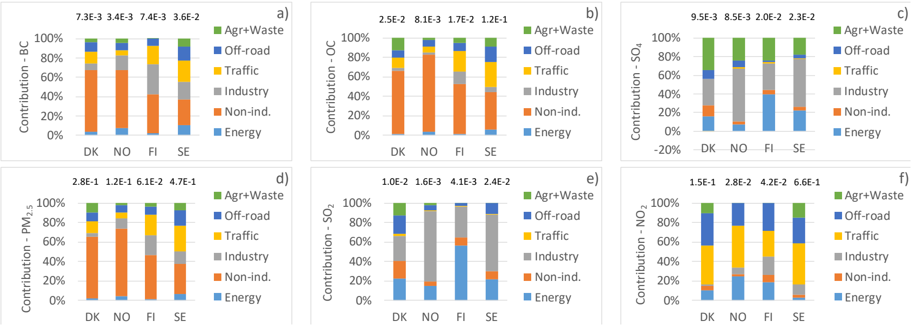


Figure 9. Population-weighted sectoral contributions of Swedish emissions on surface a) BC, b) SO_4, c) OC, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Sweden.

3.2.2. Arctic

The contributions of the emission sources in the different Nordic countries on the surface aerosol concentrations over the Arctic region (defined as the area north of 67 °N latitude) are presented in Figure 10. Results show that overall, Norway has the largest contribution to surface aerosol levels over the Arctic, while Denmark has the lowest contribution, although contributions are only a few percent. Norwegian emissions, in particular non-industrial combustion, contributes to about 2% of the surface BC levels over the Arctic. Non-industrial combustion in the Nordic countries is also the largest contributor to Arctic BC levels, except for Sweden, where industry plays a more important role. Non-industrial combustion is also the dominant contributor to OC levels over the Arctic. Sulfate levels are largely influenced by the contributed from the agriculture and waste treatment facilities over the Nordic countries. Contributions to Arctic $PM_{2.5}$ levels are similar to the contributions to the BC levels.

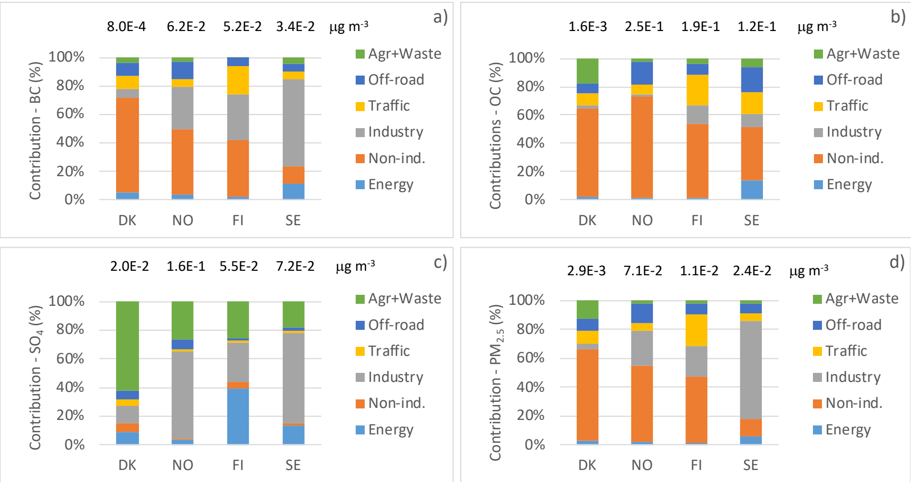


Figure 10. Population-weighted sectoral contributions from a) Denmark, b) Norway, c) Finland and d) Sweden to the surface aerosol levels over the Arctic (north of 67ºN). The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in each source country.

3.2.3. Spatial distributions of contributions

The geographical distributions of total anthropogenic emissions from each Nordic country to surface $PM_{2.5}$ and O_3 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 O_3 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 O_3 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 O_3 destruction over the area (Figure 11b). Finnish emissions also lead to an increase of surface O_3 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 O_3 levels (Figure 11c) are largest (up to 1µg m^{-3} : 2%) over the Oslo area and the impact extents over the northern part of Oslo with a slightly larger spatial contribution to O_3 levels compared to Denmark and Finland. The Swedish emissions have a larger geographical impact on the surface O_3 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.

Deleted: s

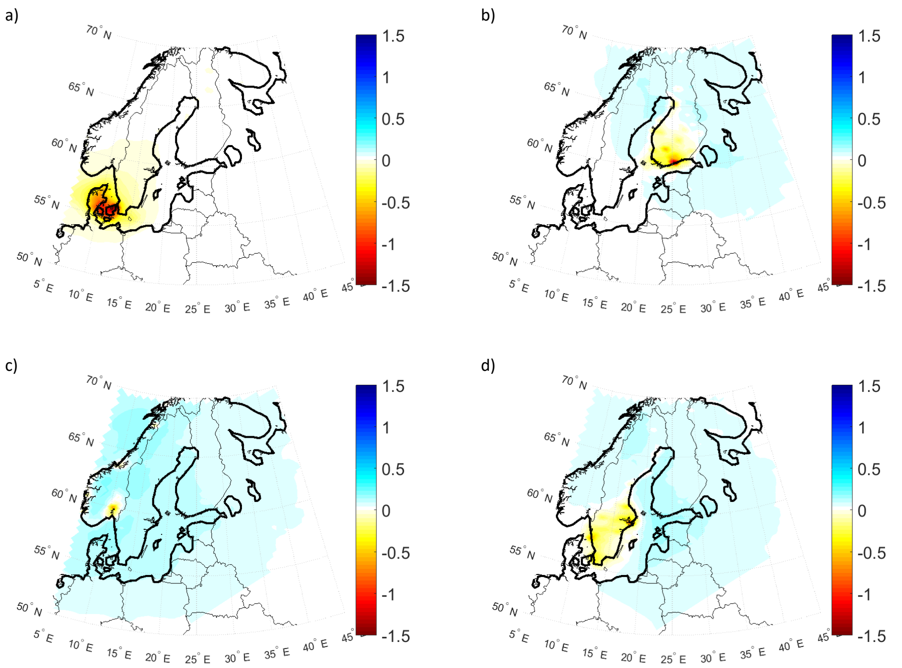
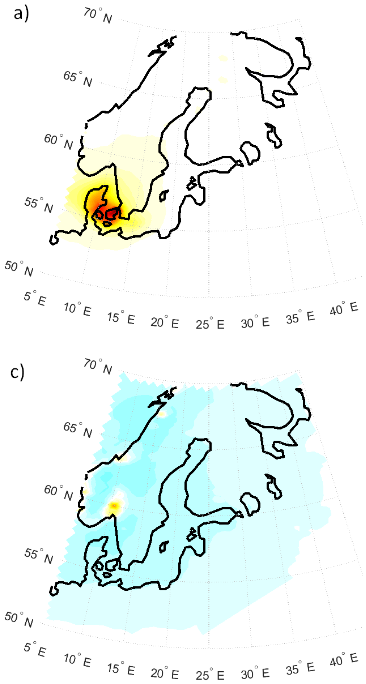


Figure 11. Spatial distributions of annual population-weighted mean absolute contributions (µg m^{-3}) of total emissions from a) Denmark, b) Finland, c) Norway, and d) Sweden to surface O_3 levels in the Nordic region.

Figure 12 shows the annual-mean absolute contributions of each Nordic country on the surface $PM_{2.5}$ levels in the entire model domain. Danish anthropogenic emissions are responsible for up to 20% of surface $PM_{2.5}$ levels over Denmark, with largest contributions over the capital region (Greater Copenhagen area) (Figure 12a). Danish land emissions also impact the surface $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ level around the capital region by up to 30%, while there is also a significant impact on surface $PM_{2.5}$ levels over Sweden by around 7% (Figure 12c). Finally, Swedish anthropogenic emissions have large contribution to surface $PM_{2.5}$ levels over the Stockholm area by around 15% and also contributes to $PM_{2.5}$ 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 $PM_{2.5}$ over the Arctic. Overall, the impacts are very small, around a few per cent, as seen in



Deleted:

the figure. The Danish emissions (Figure 12a) have a more local contribution compared to other Nordic countries and the impact does not reach above roughly 70 °N. The outflow from Finland, Norway and Sweden can reach to the central Arctic ocean over to the northern parts of Greenland, however contributions are around 1-2% (Figs. 12b-d).

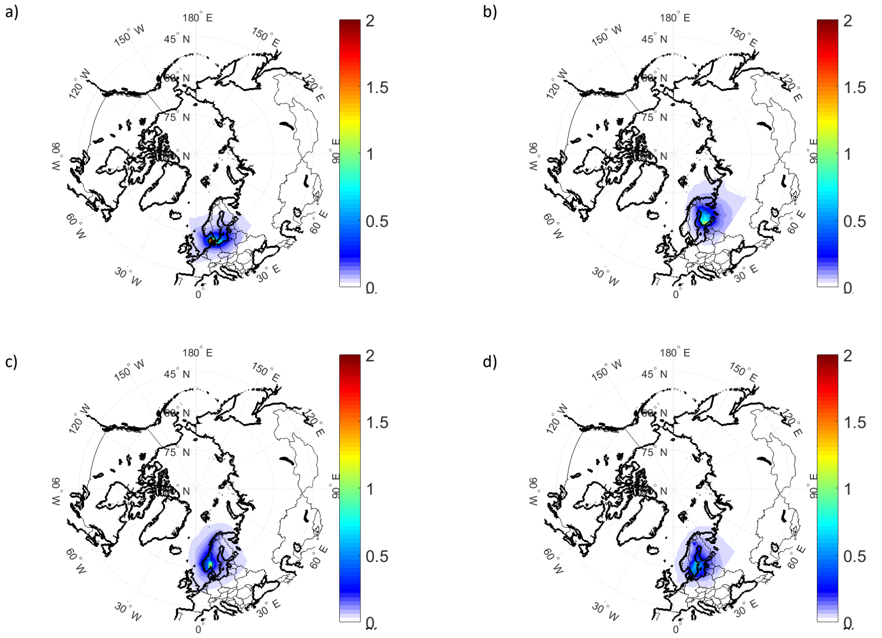
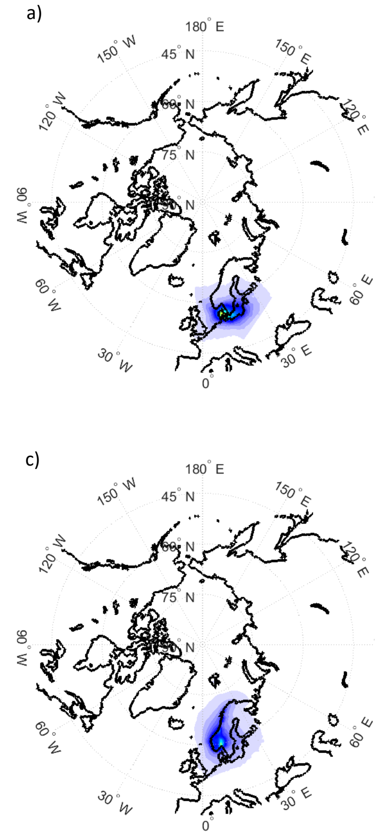


Figure 12. Spatial distributions of annual population-weighted mean absolute contributions (µg m^{-3}) of total emissions from a) Denmark, b) Finland, c) Norway, and d) Sweden to surface PM$_{2.5}$ levels over the Nordic and the Arctic regions (north of 67ºN).

3.3. Contribution to premature mortality and costs

The number of acute and chronic premature mortality in the four selected Nordic countries and the Arctic region (north of 67ºN), along with the associated costs are presented in Table 5. The 95% confidence intervals provided in the brackets are calculated by scaling the calculated health outcomes by the confidence intervals of relative risk (RR) presented in section 2.2 (RR= 1.062 [1.040-1.083]). As seen in the Table, chronic mortality due to PM$_{2.5}$ is the major source for premature mortality, as EVA calculates chronic mortality only due to exposure to PM$_{2.5}$ (see Table 2). The highest number of cases is calculated for Sweden (~4 200 cases), followed by Denmark (~3 500 cases), Finland (~1 800) and Norway (~1 700). Results also show that SO$_2$ is responsible for almost all acute mortalities in the region, which is consistent with earlier studies (e.g. Brandt et al., 2013). This is due to the decrease of O$_3$ in the region by fresh NO emissions, leading to low



Deleted:

Deleted: weigthed

Deleted: almost

mortality due to O_3-exposure. These numbers lead to an associated cost of more than 2 billion Euros in Sweden and Denmark and ~ 1 billion Euros in Finland and Norway. The number of premature death cases are comparable with existing literature (e.g. Brandt et al., 2013a for Denmark; Solazzo et al., 2018 for all four Nordic countries; EEA, 2017 for all four Nordic countries). In the Arctic region, the total number of premature mortality cases is calculated to be 94, 93 of which are due to exposure to $PM_{2.5}$ (chronic), leading to a cost of 58 million Euros.

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

	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]

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

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

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]**

Figure 13 shows the contributions of sectoral emissions from each Nordic country to the total premature death cases in 2015 in the different Nordic countries. Overall, Nordic countries contribute to low premature death cases in their Nordic neighbours (≤50). As seen in the figure, agriculture and waste management sectors together can have significant share in the premature mortality (e.g. Denmark) due to the dominant contribution of NH_4 aerosols in the region (Figure 4).

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

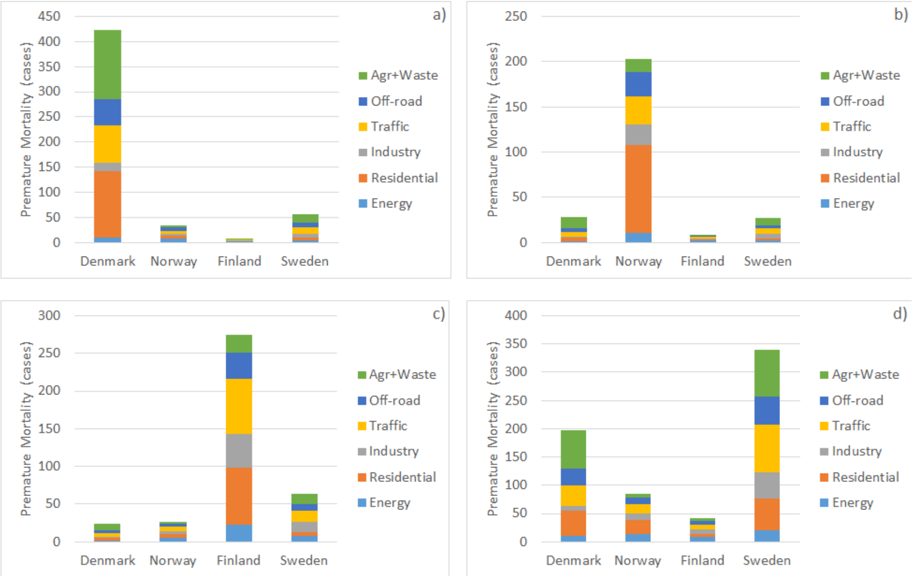


Figure 13. Source contributions from the anthropogenic emissions of a) Denmark, b) Norway, c) Finland, and d) Sweden to total premature mortality (acute+chronic) in the Nordic countries.

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

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

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]

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

Deleted: downwind

4. Conclusions

The sectoral contributions of land-based anthropogenic emission sources in the four Nordic countries; Denmark. Finland, Norway and Sweden, on air pollution levels and premature mortality in these countries and over the Arctic have been estimated using the DEHM/EVA impact assessment system for the year 2015. The chemistry and transport model, DEHM, was run with tagging mode in order to calculate inline the sectoral contributions based on 30% reductions of each sector separately. Using the modelled surface concentrations of O_3, SO_2 and $PM_{2.5}$, the EVA model calculated the acute (O_3 and SO_2) and chronic ($PM_{2.5}$) premature mortality due to exposure to these pollutants.

Results show that the Nordic countries are responsible for 5-10% of the regional background surface $PM_{2.5}$ concentrations in the countries itself. The non-industrial combustion (SNAP2), which is dominated by the non-industrial wood combustion, is responsible for 50% to 80% of the contribution to surface $PM_{2.5}$ in the Nordic countries. In Denmark, Finland and Norway, non-industrial combustion contributes largely to surface OC (by 60% - 80%). In Sweden, SNAP2 is responsible for 43% of the contribution to surface OC, while 43% comes from industrial activities. Similar to OC, BC is also dominated by non-industrial combustion (by 50%-65%), except for Sweden, where 25% originates from non-industrial combustion and 31% from industrial activities. The dominant source for surface SO_4 and SO_2 in all four Nordic countries is calculated to be industrial activities. In Norway and Sweden, around 70% of SO_2 are coming from industrial activities, while in Denmark and Finland, industrial activities are responsible for around 30% of SO_2. Off-road traffic is responsible for 21% of SO_2, while energy production is responsible for 50% of SO_2 in Finland. Industrial activities are also responsible for 60% of SO_4 in Norway and Sweden and 30% in Denmark and Finland. The dominant source for NO_2 is calculated to mobile sources, and the share between on-road and off-road traffic varies depending on the country. Almost 35% of NO_2 comes from on-road traffic in all four Nordic countries while off-road traffic contributes by 25% to 35%.

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

Anthropogenic emissions lead to a titration of around 4-5%, particularly over the source countries and lead to a very small surface O_3 increase (>1%) in the downwind regions. The largest impacts are calculated to be around the capital regions. Danish emissions also impact the surface $PM_{2.5}$ levels over the southern part of Sweden and Norway, by around 3%. Finnish emissions also have a small impact, lower than 3%, on the central part of Sweden and northern parts of Norway. Norwegian anthropogenic emissions impacts $PM_{2.5}$ levels over Sweden by around 7% while Swedish anthropogenic emissions contribute to $PM_{2.5}$ levels over the southwestern parts of Finland, by up to 5%. It should be noted that these results are calculated for a specific year, 2015, therefore transport from one country to others can significantly vary in different years due to meteorology, in particular wind speed and direction.

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

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

Author Contribution

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

Acknowledgements

This study has been conducted under the FREYA project, funded by the Nordic Council of Ministers, Climate and Air Pollution Group (grant agreement no. MST-227-00036). AU gratefully acknowledges the NordicWelfAir project funded by the NordForsk's Nordic Programme on Health and Welfare (grant agreement no. 75007). The work has also been funded by the Academy of Finland within the project GLOROIA and by the Research Council of Norway under the project BlackArc (contract no 240921).

REFERENCES

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

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

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

Deleted: ¶

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

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

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

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

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

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

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

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

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

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

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

Forsberg, B., Hansson, H.C., Johansson, C., Aureskoug, H., Persson, K., and Järvholm, B.: Comparative health impact assessment of local and regional particulate air pollutants in Scandinavia. Ambio, 34, 11-19, 2005.

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

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

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

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

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

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

Field Code Changed

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

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

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

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

Karvosenoja, N., Kangas, L., Kupiainen, K., Kukkonen, J., Karppinen, A., Sofiev, M., Tainio, M., Paunu, V.-V., Ahtoniemi, P., Tuomisto, J.T. and Porvari, P.: Integrated modeling assessments of the population exposure in Finland to primary PM2.5 from traffic and domestic wood combustion on the resolutions of 1 and 10 km, Air Qual. Atmos. Health, 4, 3–4, 179–188, 2011.

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

Kukkonen, J., López-Aparicio, S., Segersson, D., Geels, C., Kangas, L., Kauhaniemi, M., Maragkidou, A., Jensen, A., Assmuth, T., Karppinen, A., Sofiev, M., Hellen, H., Riikonen, K., Nikmo, J., Kousa, A., Niemi, J. V., Karvosenoja, N., Santos, G. S., Sundvor, I., Im, U., Christensen, J. H., Nielsen, O.-K., Plejdrup, M. S., Nøjgaard, J. K., Omstedt, G., Andersson, C., Forsberg, B., and Brandt, J.: The influence of residential wood combustion on the concentrations of PM2.5 in four Nordic cities, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-564, in review, 2019.

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

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

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

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

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

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

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

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

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

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

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

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

WHO: Health risks of air pollution in Europe – HRAPIE: Recommendations of concentration-response functions for cost-benefit analysis of particulate matter, ozone and nitrogen dioxide, World Health Organization, available at: 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.

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

Page 9: [1] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [2] Formatted Table **Ulas Im** **19/09/2019 11:03:00**

Formatted Table

Page 9: [3] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [4] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [5] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [6] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [7] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [8] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [9] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [10] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [11] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [12] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [13] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [14] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [15] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [16] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [17] Formatted Table **Ulas Im** **19/09/2019 11:19:00**

Formatted Table

Page 9: [18] Formatted **Ulas Im** **19/09/2019 11:17:00**

Font: (Default) Times New Roman, 11 pt

Page 9: [19] Formatted **Ulas Im** **19/09/2019 11:18:00**

Centred

Page 9: [20] Formatted **Ulas Im** **19/09/2019 11:17:00**

Font: 11 pt

Page 9: [21] Formatted **Ulas Im** **19/09/2019 11:18:00**

Font: (Default) Times New Roman, 11 pt

Page 9: [22] Formatted **Ulas Im** **19/09/2019 11:19:00**

Centred

Page 9: [23] Formatted **Ulas Im** **19/09/2019 11:18:00**

Font: 11 pt

Page 9: [24] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [25] Formatted **Ulas Im** **19/09/2019 11:17:00**

Font: (Default) Times New Roman, 11 pt

Page 9: [26] Formatted **Ulas Im** **19/09/2019 11:18:00**

Centred

Page 9: [27] Formatted **Ulas Im** **19/09/2019 11:17:00**

Font: 11 pt

Page 9: [28] Formatted **Ulas Im** **19/09/2019 11:18:00**

Font: (Default) Times New Roman, 11 pt

Page 9: [29] Formatted **Ulas Im** **19/09/2019 11:19:00**

Centred

Page 9: [30] Formatted **Ulas Im** **19/09/2019 11:18:00**

Font: 11 pt

Page 9: [31] Formatted **Ulas Im** **19/09/2019 11:03:00**

Font: 11 pt

Page 9: [32] Formatted **Ulas Im** **19/09/2019 11:17:00**

Font: (Default) Times New Roman, 11 pt

Page 9: [33] Formatted **Ulas Im** **19/09/2019 11:18:00**

Centred

Page 9: [34] Formatted **Ulas Im** **19/09/2019 11:17:00**

Font: 11 pt

Page 9: [35] Formatted **Ulas Im** **19/09/2019 11:18:00**

Font: (Default) Times New Roman, 11 pt

Page 9: [36] Formatted Ulas Im 19/09/2019 11:19:00

Centred

Page 9: [37] Formatted Ulas Im 19/09/2019 11:18:00

Font: 11 pt

Page 9: [38] Formatted Ulas Im 19/09/2019 11:03:00

Font: 11 pt

Page 9: [39] Formatted Ulas Im 19/09/2019 11:17:00

Font: (Default) Times New Roman, 11 pt

Page 9: [40] Formatted Ulas Im 19/09/2019 11:18:00

Centred

Page 9: [41] Formatted Ulas Im 19/09/2019 11:17:00

Font: 11 pt

Page 9: [42] Formatted Ulas Im 19/09/2019 11:18:00

Font: (Default) Times New Roman, 11 pt

Page 9: [43] Formatted Ulas Im 19/09/2019 11:19:00

Centred

Page 9: [44] Formatted Ulas Im 19/09/2019 11:18:00

Font: 11 pt