We thank both reviewers for their valuable and constructive comments that helped us to improve the manuscript significantly. We list point by point responses to their comments below and hope they find the new version sufficient to be published in ACP.

Responses to comments from Reviewer#1

This work described the contributions of Nordic anthropogenic emissions on air pollution and premature mortality over the Nordic region and the Arctic. Although this study provides important results and is well written, there remain some concerns in the current manuscript. First, one issue is that the results are unsatisfactory discussed with only a few references in the case of premature mortality. Are the results coherent to other studies, years, countries? Which are the limitations of your method?

Major comments:

Comment: L113: Why 2015? Justify the selected year.

Response: We have now added the following (lines 115-118): "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."

Comment: L231: Why you use exposure-response functions for chronic mortality based on data from United States from the year 2002? Didn't you find any study from your Nordic region? What is about short-term exposure for PM2.5? Are the results from United States really applicable in the Nordic region? How could influence the use of ERF from different studies? Why you use short-term effects only for specific pollutants and for others only chronic mortality? I recommend including table S2 as regular table, as only with this table the paragraph is understandable. The paragraph is not very clear. I would also include some limitation here about the use of exposure-response functions or discuss the limitation. Explain and justify the reason for selecting the specific exposure-response functions from these studies.

Response: We have now extended the health impact assessment section to discuss these concerns from the two reviewers.

Comment: Fig. 5-9: I think you should use stacked percent plot, which could help to compare sectorial emissions. In order to see the total, you could label each bar. In the actual form, it is not possible to compare correctly some sectorial categories.

Response: The figures have been modified accordingly.

Comment: L448-: Why only PM2.5? NOx? SO2? O3? Please indicate the confidence interval for the mortality estimations, particularly in Table 4. The same for the cost estimations.

Response: NO_2, O_3 and SO_2 are mostly associated with acute health impacts, although there are some recent studies suggesting chronic impacts from O_3. However, the current version of the EVA model does not yet include these impacts. This is now also added to the text (lines 226-231).

Minor comments:

Comment: L77: please use capital letter for Primary PM2.5 (PPM2.5)

Response: Corrected accordingly.

Comment: L79: "secondary inorganic PM2.5 (SIA)" should it be secondary inorganic aerosol?

Response: Corrected accordingly.

Comment: L123: DHEM, although the abbreviation was defined in the abstract here you should repeat it for the reader. The same for EVA in point 2.2. It is not advisable to use abbreviations as titles.

Response: Corrected accordingly.

Comment: Fig1: Please use a better graphical representation. For example, treemap, circular packing or something else in which the graph is easier to read.

Response: Fig.1. has been modified accordingly.

Comment: L206: It should be "European Environmental Agency (EEA)".

Response: Corrected accordingly.

Comment: L232: remove last ")".

Response: Removed.

Comment: L305: "eacvh" -> each;

Response: Corrected.

Comment: "Figure 4" If you use figure reference in text at beginning, it would be better the style "Figure X". Please, revise the whole document.

Response: the text is modified accordingly.

Comment: L307: "The figure" -> the bars or graph?; subindex missing for PM2.5

Response: We have now modified the text.

Responses to comments from Reviewer#2

In this modelling study, PM and O3 exposure and associated premature mortalities in the Nordic countries have been attributed to anthropogenic emission sources (sectors) in each of the countries. The attribution is based on the tagging methodology which is more accurate than linearized source-receptor relations, commonly used under conditions of limited CPU resources.

General comments:
The material obtained from the modelling is potentially relevant, however my feeling is that the results are not optimally evaluated and presented, and too much attention is given to less relevant issues. In my opinion, the major conclusion of this study is that 80-85% percent of the air pollution impacts in the Nordic countries are coming from sources outside that region. This observation is reported in the abstract and in the conclusions, however without giving any further consideration on the (policy) relevance of this finding. It basically means that national air quality measures in the Nordic country apparently barely can contribute to improving air quality. Given this outcome, one can wonder what is the relevance of making a detailed discussion of individual other Nordic countries contributions by sector to PM2.5 in the receptor country (i.e. attribution by country and by sector of the tiny orange part in the bar graphs of Fig. 4).

Based on Figure 4, the key question is: which source regions and sectors are then contributing to the gray portion of the stacked bars? Hence, if the aim of the study is "to identify emission sectors that should be targeted for mitigation to decrease air pollution levels in the Nordic countries" (L108) the authors have clearly overlooked the major contributing factors. If the authors still want to focus specifically on the Nordic countries' contribution only, this should be better motivated and framed in the introduction. (Besides, I also wonder what is the motivation for a separate health impact assessment for specifically the low-populated Arctic region >67 N).

Response: We thank the reviewer for the detailed comments. We have tried to address all the concerns in the following sections. The main aim of the study is to identify how much each Nordic country is contributing to the air pollution levels and exposure in the Scandinavian region and the Arctic. We agree that it is also very relevant and interesting how much is coming from rest of the world on a sectoral basis, however this is not the main objective of this study, and it requires additional sectoral simulations. It is also true that population-wise, the Arctic is not much relevant regarding health impact assessment, as it is for e.g. Europe. However, it is also interesting to have some first estimates of how much the Arctic population is affected from air pollution that is mainly transported from rest of the world. We tried to make this more clear in the end of the introduction section.

Specific comments:

1. Throughout the text: use subscript in chemical names and PM2.5

Response: Corrected accordingly.

2. L34 – 37 and throughout the paper: please use consistent naming for the sectors in text and figure legends; "non-industrial" and "residential" are interchanged. It is better to use just one consistent name (preferably "residential"). Same for "Agriculture and Waste" and "Others" (preferably use "Agriculture + Waste")

Response: We have modified. The text, the plots and the tables accordingly.

3. L33: ': : :80% of the PM2.5 concentration was attributed to transport from outside: : :'

Response: Corrected.

4. L34-35: If 80% of PM2.5 comes from outside, how can residential combustion (inside the country) be responsible for 60% of total PM2.5 mass?

Response: We mean that out of the 20% originating inside the region, 60 % is coming from non-industrial combustion. We have modified the sentence accordingly (lines 35-36).

5. L38 OC is said to be the major contributor. In the main section however nothing has been mentioned on the chemical mass balance of the PM2.5 in each country, and further ammonium nitrate has never been mentioned. Was this compound considered as a PM2.5 constituent? Earlier studies have suggested that ammonium nitrate is the dominant PM2.5 component in NW Europe (e.g. Lelieveld et al., 2015).

Response: We have now added a modelled PM2.5 chemical composition section under Section 3.2.1 (lines 345-352).

6. L39: if the tagging method was used, the contribution of the residential sector should not be 'suggested' from the chemical composition but result directly from the tagged species & sector?

Response: The tagging method identifies the contribution from all non-industrial combustion sources in the residential and commercial heating, including residential wood burning.

7. L62: in Sweden originates

Response: Corrected.

8. L66: instead of 'geographic' maybe better use 'spatial'?

Response: Changed accordingly.

9. It is unclear to me how a country contributes to x% of European PM2.5. Do you mean that the European-wide population-weighted PM2.5 concentration (i.e. exposure) contains x% PM2.5 emitted in that country (L77-79)? Please formulate more precisely.

Response: The cited study showed that Sweden contributed to 1.4% of the European Primary PM2.5 (PPM2.5) mass concentrations.

10. L94: Can you be more quantitative on how 'low' air pollution levels are, e.g. relative to WHO target levels? Does 'low' refer to country-area-mean or population exposure? Does this also apply to urban locations?

Response: We have modified the sentence accordingly (lines 95-97) as: "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 40 $\mu g\ m^{-3}$ as well as the WHO limit value of 20 $\mu g\ m^{-3}$ (EEA, 2018)."

11. L96-104: not sure if the climate impact of BC is relevant in this context. Define 'SLCFs' (L103)

Response: We have removed that sentence from the text.

12. L109 'decrease'

Response: Corrected accordingly.

13. DEHM: What is the time resolution of the model output for the pollutants? Are O3 values produced at 1h time steps? Does the model include natural PM components (in particular seasalt)?

Response: We have modified the section as (lines 138-141):

"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).",

and as (lines 143-148):

"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 PM2.5 or PM10 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."

14. L151: Actually the large contribution of NH3 in 'Others' is due to Agriculture (only).

Response: Modified accordingly.

15. Figure 1 is difficult to read and would benefit from a different layout (e.g. stacked bar plots, and using a different Y-scale for each component). Alternatively, as the total values are already given in the table, the plot could show the different relative contributions by sector (stacked bar or pie plots).

Response: We have changed Figure 1.

16. L165: The tagging method keeps track: : :

Response: Corrected accordingly.

17. L167: What do you mean by "background concentrations"? Usually this refers to concentrations in absence of anthropogenic emissions. L168: not clear what is meant by "in parallel". In general this paragraph is rather difficult to understand for those not really confident with the tagging method. Would it be possible to include some mathematical equation(s) that express the basic principle(s)?

Response: We have now modified the section. The technical details for the tagging scheme was already provided in Brandt et al. (2013), which is referred to in the manuscript..

18. Model evaluation: L181: these data are not shown in the supplemental material (I presume they are presented in Tale 3 instead). Is the model resolution of 50kmx50km high enough to reproduce urban background concentrations? Does the model include a sub-grid treatment to simulate the urban increment (for PM) or titration decrement (for O3)? Table S1 should include as well station data for PM2.5, SOMO35, SO2 (i.e. all the metrics used for the health impact assessment). The references to data sources for the 4 countries can easily be moved to Table S1 so the section from L184 – L214 can be shortened.

Response: 50 km resolution is coarse to reproduce the urban gradients. We have added some discussion for this in the text. This is why, we only do the comparison with urban and regional background stations. Data sources has been moved to the supplementary material.

19. Section 2.2 (EVA) could contain some more information on the exposure-response functions and RRs used both for PM and O3. Table S2 should be explained better. ERFs are commonly expressing the relative risk. It is strange to see the exposure-response coefficient expressed as mortalities per concentration unit. There must be an underlying calculation, involving exposed population number and baseline mortalities.

Please expand this. Why is SO2 included as a risk but not NO2? WHO (2013, HRAPIE project) recommends PM, O3 and NO2 as major risk factors, but not SO2 and CO. It is not clear if CO was considered here: CO ERF is not mentioned in Table S2, but section 2.2 mentions it is included in EVA.

Response: This section has been extended following the suggestions from both reviewers.

20. L225: "EVA calculates and uses annual mean: : :" change to: "EVA uses annual mean: : :"

Response: Modified accordingly.

21. Are natural PM components included in the PM2.5 exposure? L235: "full range of anthropogenic concentrations"?

Response: Changed accordingly (lines 238-239) as: "…simulated total (anthropogenic and natural) $PM_{2.5}$ mass.".

22. L247-248: not clear if the perturbations were done for each individual SNAP sector, or for the combined sectors in case of Industry and Others

Response: The perturbation is done for the combined snaps in case of Industry and Others.

23. L253-255: what is the outcome of this comparison between 100% and scaled 30% perturbation?

Response: We apologize for the error. This experiment was removed from the study and not conducted due to time limitation within the project.

24. Table 3 should also include the mean values for O3 and PM2.5. What about the model evaluation for SO2 and NOx?

Response: We have now added a Table S2 in the supplementary for the model evaluation for NO_2 and SO_2 and text in the manuscript (lines 308-313).

25. Use consistent symbols for the correlation coefficient (either r or R)

Response: Changed to *r* throughout the text.

26. Do the Taylor diagrams add essential new information to what is given in Table 3? What can be concluded from the diagrams that is not emerging from the table? Is model performance better/worse for specific station type?

Response: The Taylor diagrams show a station by station evaluation while the table gives an overall comparison based on the mean of stations in each country. In addition, we have added the following text (lines 310-316): "In all countries, lower NMB values are calculated for O3 over the regional background stations compared to urban background stations, where the overestimations are higher. Regarding PM2.5, no such conclusions can be drawn die to very limited number of regional background stations in Denmark and Norway. In Finland, lower NMB values for PM2.5 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."

27. L270-274: the discussion does not correspond to what is given in Table 3: r is not >0.7 for Norway. If the % overestimation refers to the NME, then for O3 it is rather 20% for Denmark, Finland and Sweden, not 10%. For PM2.5, the R for Norway in the table is 0.35, not 0.7. A relatively good R does not necessarily imply a good model performance: mean biases of the order of -3g/m3 are significant as can be seen in *the NME (underestimation of 40 to 50% for all countries). Could this be due to a natural component in the measurements that was not considered in the model? What can be concluded from this model evaluation in the context of the further analysis? How robust are the model results?*

Response: We have now modified this section accordingly.

28. L301: "near de ground surface": change to "near the surface"

Response: Corrected

29. L301: 'Caused by the.." change to "Based on the prevailing : : :"

Response: Changed accordingly.

30. L305 eacvh: each

Response: Corrected.

31. Why does Fig. 4 not include NH4 and NO3? The sum BC+OC+SO4 is much lower than total PM2.5. What makes up the remaining PM2.5 mass? If NO2 and CO are not used in the impact evaluation why show the values?

Response: We have now added a paragraph for the modelled aerosol chemical composition (lines 353-360).

32. Why is the same analysis not made for O3 or SOMO35?

Response: We have done a similar analysis and the results are discussed in lines 376-379.

33. L307-308: this is a very important observation and raises immediately the question about the sources of this major rest contribution. This observation should not be left undiscussed.

Response: we have now added some discussion in the text (line 370-371).

34. Please increase text font in Figures 5 to 7.

Response: Modified accordingly.

35. L324: 7 g m-3

Response: Corrected.

36. L328: again: what is the contribution of ammonium nitrate?

Response: A section discussing chemical composition is now added to the text (lines 353-360).

37. In Section 3.2.1, the text is quite repetitive, basically repeating for each combination of source country x and receptor country z the contribution for each chemical component. How relevant is this separation in components in the context of the formulated scope of this study (i.e. to identify the emission sectors that should be targeted for mitigation)? To answer this question it is more relevant to show for each of the receptor regions how much is being contributed (1) from the country's own emissions (2) from other Nordic countries (3) from the rest of the world (by difference). I would suggest to move Figs. 5-8 to the supplemental information. Instead, for each relevant exposure metric (PM2.5, O3 and maybe SO2, NO2, CO in the supplemental material), a figure could then be presented for each receptor country (DK, NO, FI, SE), *with each bar representing a sector, and within each (stacked) bar a contribution from within the country, from other Nordic countries, and from the rest of the world (and maybe an additional bar for the sum of all sectors) as in attached figure 1 (made up with arbitrary numbers).*

Response: We agree with the reviewer and indeed it is an interesting approach. However, this is not he main scope of the paper. The main focus is to calculate how much each Nordic country is responsible, on a sectoral basis, of their pollution in the Nordic and Arctic regions. In addition, this approach requires to make additional perturbation simulations for the present sectors for each country.

38. Similar comment for the Arctic (Fig. 9) where a graph could show the contributions by sector from each Nordic country and the rest of the world. But what is the relevance of considering specifically the >67 N area for health impacts? The contribution from the Scandinavian countries are very low, also here it would be interesting to see what the major contributors to this receptor region are.

Response: Please see above response.

39. Are the concentrations and % shown in Figs 4 – 8 referring to exposure (i.e. population-weighted concentrations) or grid-area-weighted mean? To answer the formulated scientific question it should be exposure. For SE, NO and FI which have large portions of uninhabited area there could be a significant difference between area and population-weighted average.

Response: This is a very good point and we thank the reviewer for raising this. All the numbers and plots are now updated to present the population-weighted contributions and concentrations.

40. If the graphs are produced as suggested, including PM2.5 and O3, the grid maps Fig 10 and 11 add little new information and they could be omitted (or transferred to the supplemental information)

Response. We keep the figures based on the responses to comments 37 and 38.

41. If the grid maps are kept, please adapt the color scale of the O3 grid maps. Use the same range for the 4 maps, and make an upper limit that extends further above zero (now it seems that everything is colored red because the scale is cut off at a too low limit).

Response: Maps are now modified.

42. L405: what do you mean with "...are mainly calculated in the source country itself."

Response: We have modified the sentence (lines 482-484) as: "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."

43. L 406 "Zealand region" has no meaning to a readership not familiar with the regional naming details.

Response: We have removed this.

44. L405 – 407 ("Danish anthropogenic: : :towards south") I can't follow the reasoning here: titration leads to a -4 to -5% contribution, but also to a +1% increase south? Also, as the scale stops at 0, this cannot be observed in graph 10a.

Response: We have now modified this sentence (lines 484-485) and the figure accordingly.

45. What is the share of O3 and SO2 in the acute mortalities?

Response: We have now added the following sentence (line 537-540): "Results also show that SO2 is almost responsible for 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 O3 in the region by fresh NO emissions, leading to low mortality due to O3-exposure."

46. L469: Given the fact that PM2.5 is the major risk factor in mortalities, why is the contribution of AGR so dominant in DK (compared to the small share in Fig 5)? Is this because the population exposure was taken into account? How is the share of sectors in the mortalities evaluated? By using the same proportion as in the population-weighted PM composition? In table 5 it would be useful to put in brackets which share in total mortalities in each receptor country the numbers represent - e.g. 422 (13%).

Response: We have now added the following sentence (line 568-570): "As seen in the figure, agriculture and waste management sectors can have significant share in the premature mortality (e.g. Denmark) due to the dominant contribution of NH4 aerosols in the region."

47. L503 – 510: this is no new information because the costs are proportional to the mortalities for which it was already stated which sectors are dominating (L496 - 474). Further, when making recommendations on which sectors to address in order to "substantially reduce the costs of air pollution", the authors seem to have overlooked that 80 - 85% of the pollution health impact is imported from other regions.

Response: We have moved the figure to the supplement. We have also added the following to the end of the paragraph (lines 600-602): "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 downwind, which would require a coordinated effort in Europe."

48. L566 It is not 50% of total but 50% of premature deaths caused by the Nordic countries (the latter being 16% of total premature mortalities).

Response: we have modified as follows (lines 654-661): "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."

49. L579 -578: To my opinion this is the most relevant conclusion of this study. It leaves the reader with the feeling that the less relevant part of the data has been analyzed in too much detail, leaving this essential part untouched: : :

Response: Please refer to the response to the general comment and comment 37.

References

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D. and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525(7569), 367–371, doi:10.1038/nature15371, 2015.

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.

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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 originates from south-western Europe, while the largest contribution to short-term exposure originates from south-eastern Europe (Jönsson et al. 2013).

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 40 $\mu g\ m^{-3}$ as well as the

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WHO limit value of 20 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.

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

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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.

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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.

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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).

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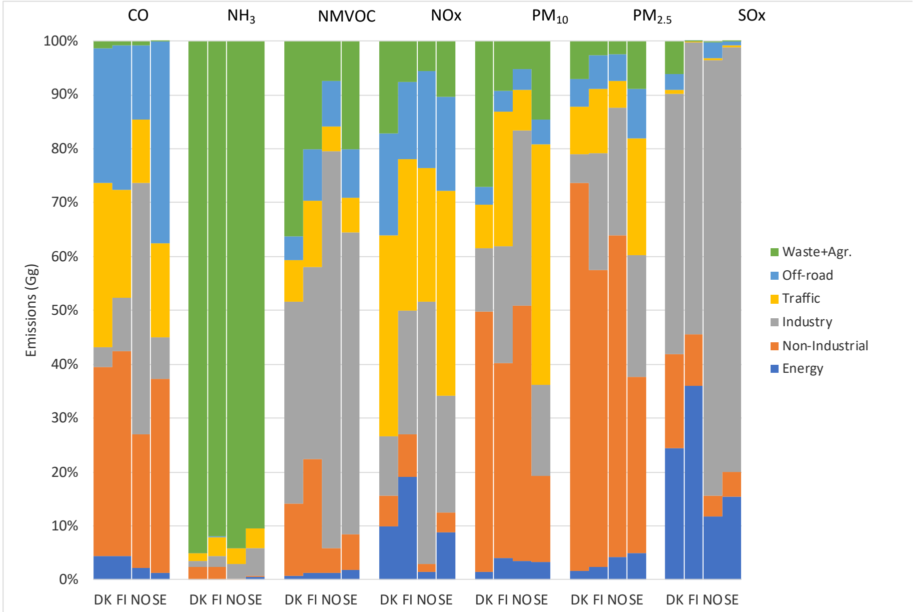
Table 1. Total pollutant emissions in the Nordic countries (in Gg) in 2015.

	CO	NH_3	NMVOC	NOx	SO_2	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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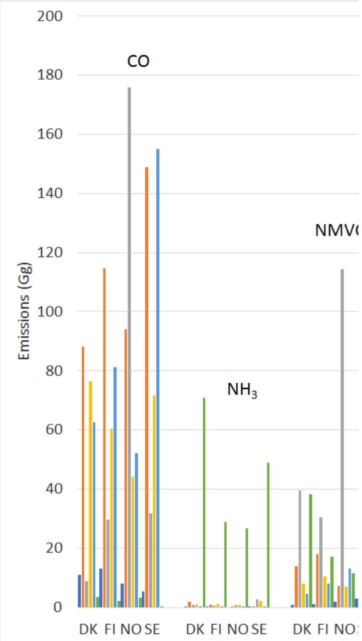

Figure 1. Relative distributions (%) of sectoral emissions of major air pollutants in the Nordic countries.

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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.

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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

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(*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 (in cases, days, or episodes), *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.

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, 2008). 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

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The measurement data for Finland represents regional and urban background levels. Data from the global and regional background stations are reported to European Monitoring and Evaluation Programme (EMEP) under the CLRTAP (Convention on Long-range Transboundary Air Pollution, http://www.unece.org/env/lrtap), and are available at http://ebas.nilu.no. The data for the urban background stations are reported at the 'Air Quality in Finland' web pages by the Finnish Meteorological Institute (https://en.ilmatieteenlaitos.fi/air-quality). ¶ ... [1]

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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, €$_{2013}$**
	(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

^{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%.

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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

2. Results and Discussion

2.1. 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 sptial 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 the overestimations are higher. Regarding PM$_{2.5}$, no such conclusions can be drawn die 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. Table S2 shows the same comparisons for NO$_2$ and SO$_2$. 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. 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).

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	*MB* (µg m^{-3})	*NMB* (%)	*RMSE* (µg m^{-3})	*r*	*MB* (µg m^{-3})	*NMB* (%)	*RMSE* (µg m^{-3})

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Denmark	0.81	5.67	0.09	11.60	0.75	-3.41	-0.32	6.22
Finland	0.74	4.77	0.10	12.44	-0.03	-0.80	-0.16	3.83
Norway	0.64	12.02	0.27	18.31	0.35	-2.56	-0.36	4.52
Sweden	0.74	7.00	0.13	13.25	0.59	0.33	0.08	3.23

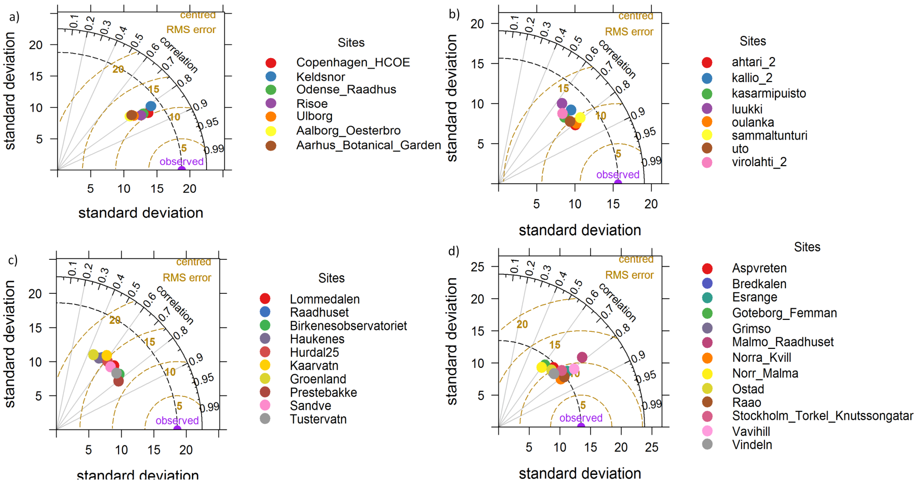

Figure 2. Taylor diagrams for daily mean O_3 for all stations in a) Denmark, b) Finland, c) Norway and d) Sweden.

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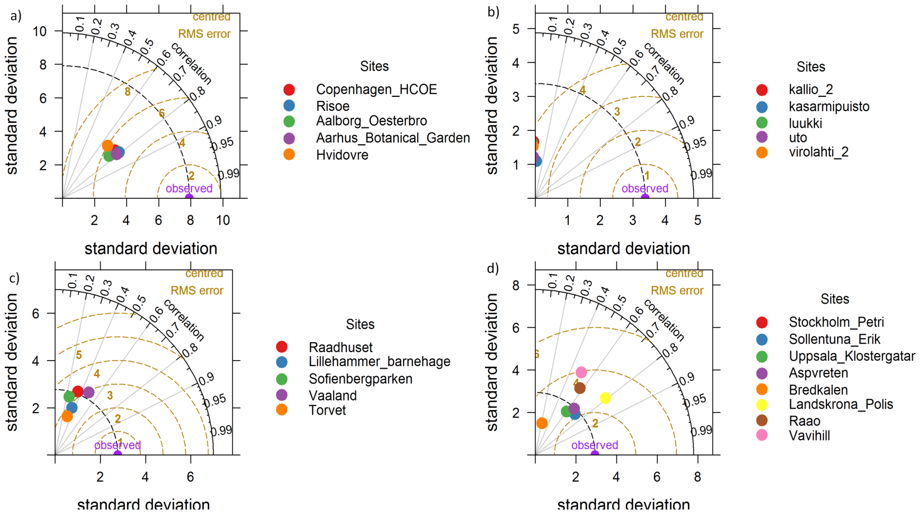

Figure 3. Taylor diagrams for daily mean PM$_{2.5}$ for all stations in a) Denmark, b) Finland, c) Norway and d) Sweden.

2.2. Sectoral contributions to surface concentrations

2.2.1. 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).

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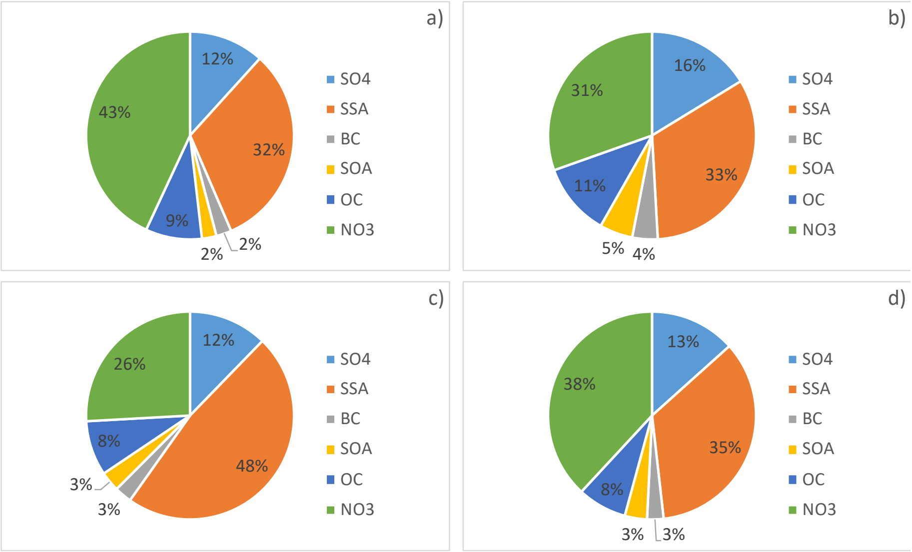

Figure 4. Simulated surface $PM_{2.5}$ chemical composition over a) Denmark, b) Finland, c) Norway, and d) Sweden.

Figure 5 compares the contribution of the total contribution of each Nordic country on the surface concentrations over the country itself, with contributions from rest of the Nordic countries and rest of the world. 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 downwind. 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.

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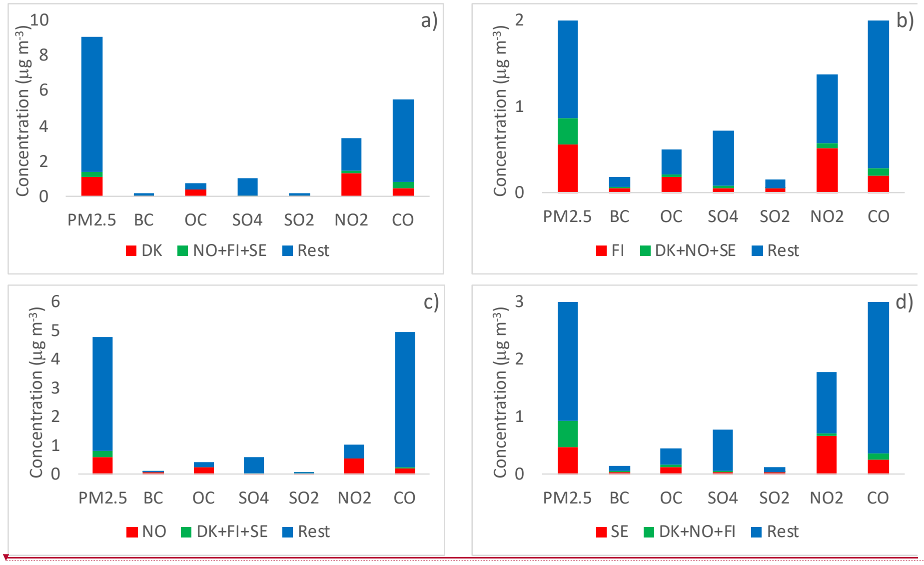
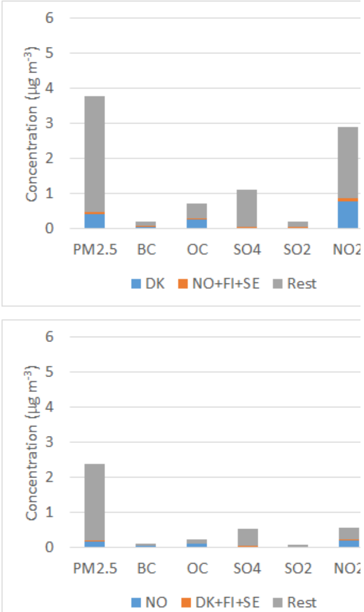


Figure 5. Absolute contributions of national, Scandinavian and other sources on the surface levels of major air pollutants over a) Denmark, b) Finland, c) Norway and d) Sweden. Note that CO concentrations are divided by 20 to scale with other pollutants.

Danish emissions contribute to only 1.14 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.



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Danish emissions contribute to only 10…144…µg m^{-3} (130…) of the surface $PM_{2.5}$ concentrations over Denmark (3.….18…µg m^{-3}), while contributions to other Nordic countries are about less than …1… (Fig.…igure 65…. Non-industrial combustion (SNAP2), which is dominated by residential…on-industrial combustion, is responsible for 0.3624…µg m^{-3} (605…) of the Danish contribution to surface $PM_{2.5}$ concentrations over Denmark. Residential…on-industrial combustion contributes to 0.2216…µg m^{-3} (5660…) of the Danish contribution to surface organic carbon (OC) concentrations over the country, suggesting the importance of residential…on-industrial wood burning for heating. Industry contributes to 0.017…µ□… m^{-3} (351…) 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 1.020.55…µg m^{-3} (~792 ... [6]

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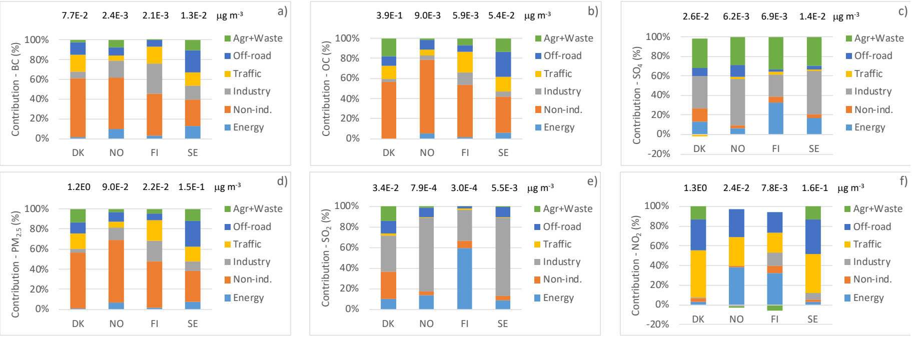

Figure 6. Population-weighted sectoral contributions of Danish emissions on surface a) BC, b) OC, c) SO_4, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Denmark.

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.

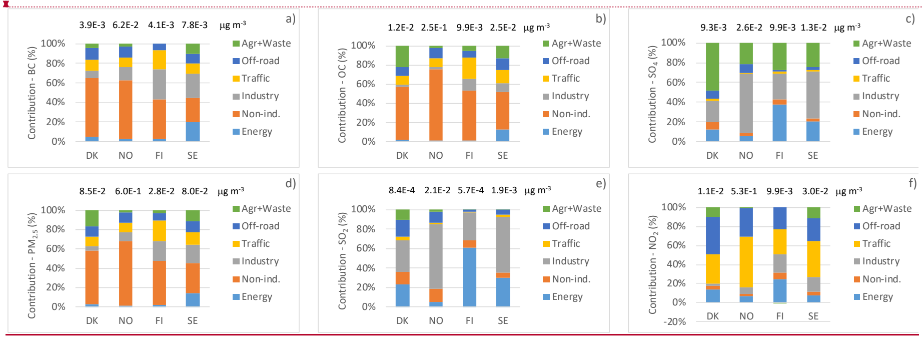

Figure 7. Population-weighted sectoral contributions of Norwegian emissions on surface a) BC, b) OC, c) SO_4, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Norway.

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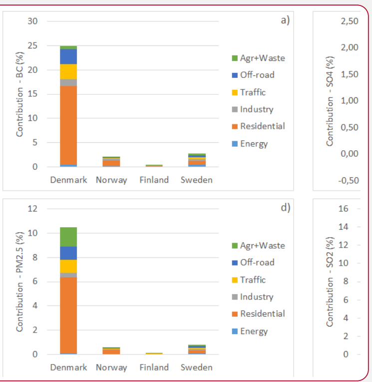

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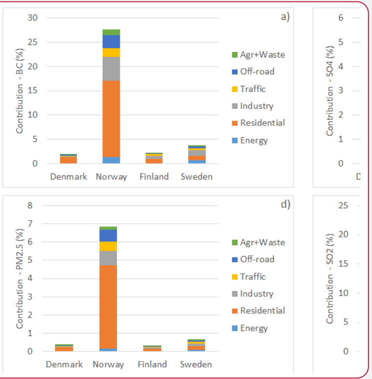

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Figure 8 shows the contributions of Finnish emissions on the pollutant levels over the Nordic countries. Similar to Denmark and Norway, non-industrial combustion is the major source of pollution over Finland, although contributions are lower compared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

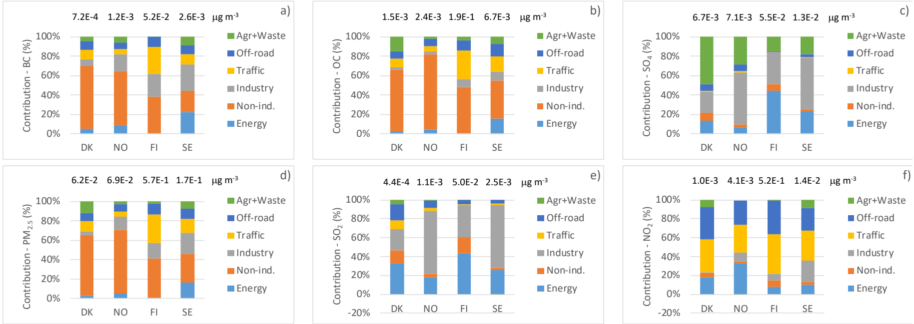

Figure 8. Population-weighted sectoral contributions of Finnish emissions on surface a) BC, b) OC, c) SO_4, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Finland.

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.

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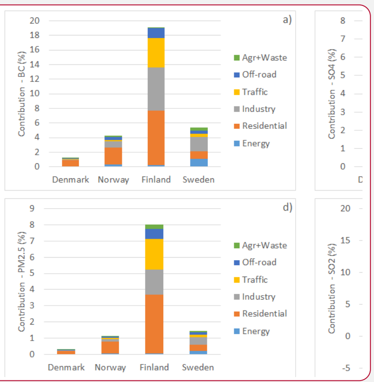

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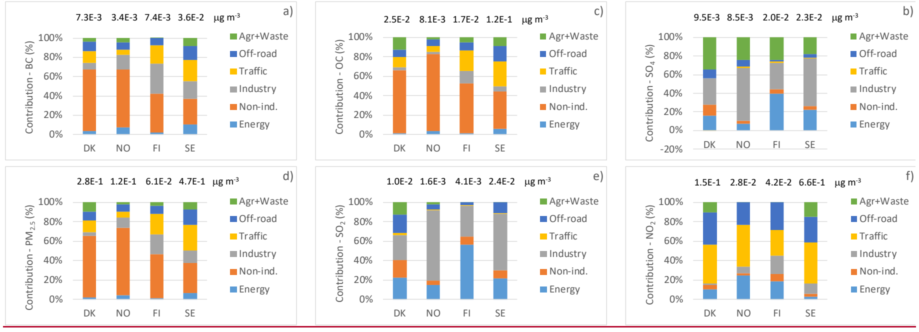

Figure 9. Population-weighted sectoral contributions of Swedish emissions on surface a) BC, b) SO_4, c) OC, d) $PM_{2.5}$, e) SO_2 and f) NO_2 over the Nordic countries. The labels above the bars show the absolute total contribution in µg m^{-3} from all the sectors in Sweden.

2.2.2. 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.

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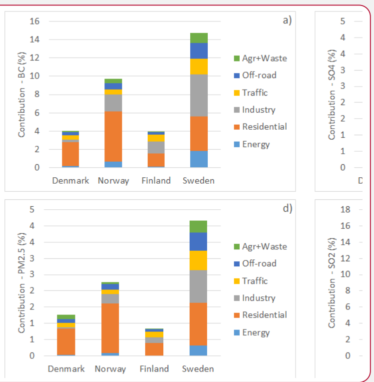

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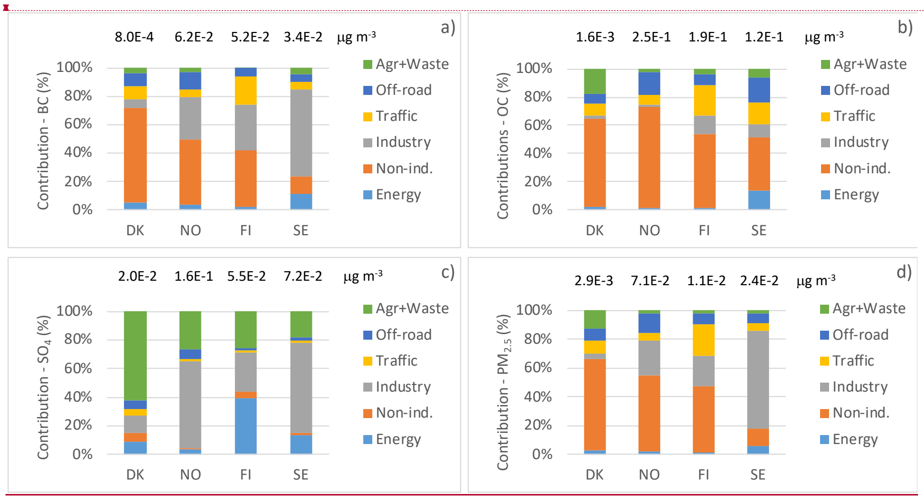
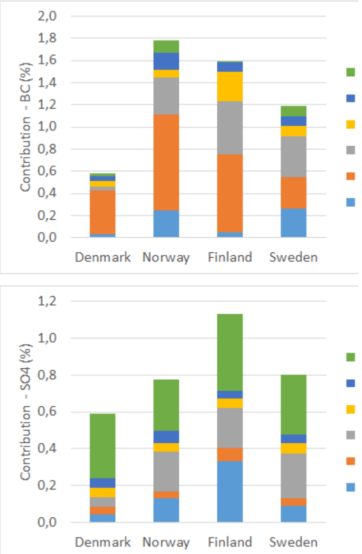


Figure 10. Population-weighted sectoral contributions from a) Denmark, b) Norway, c) Finland and d) Sweden to the surface aerosol levels over the Arctic (north of 67ºN). The labels above the bars show the absolute total contribution in μg m^{-3} from all the sectors in each source country.

2.2.3. *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) leads 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.



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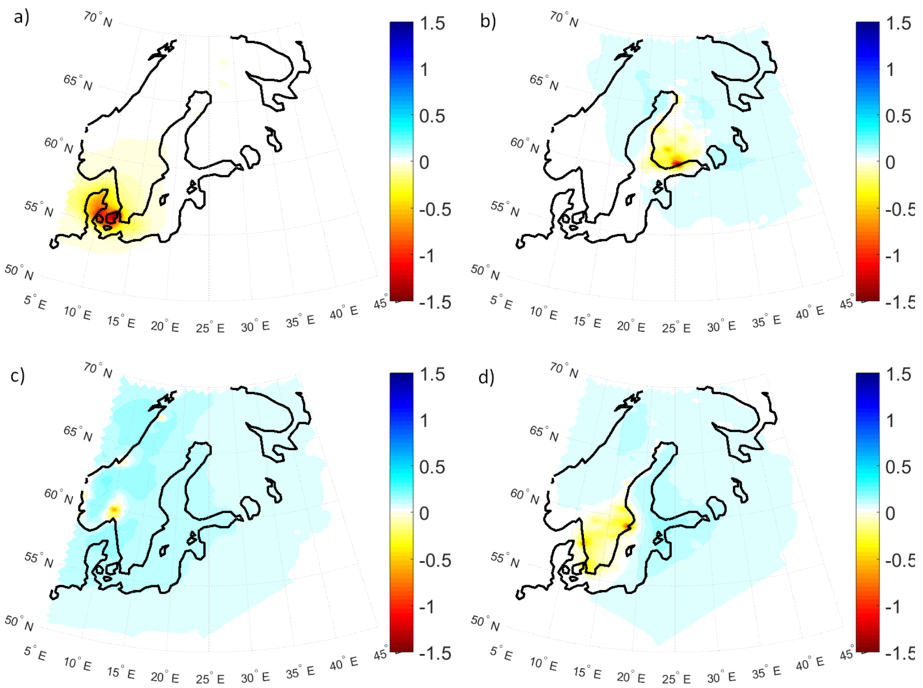

Figure 11. Spatial distributions of annual population-weighted mean absolute contributions (µg m^{-3}) of total emissions from a) Denmark, b) Finland, c) Norway, and d) Sweden to surface O_3 levels in the Nordic region.

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).

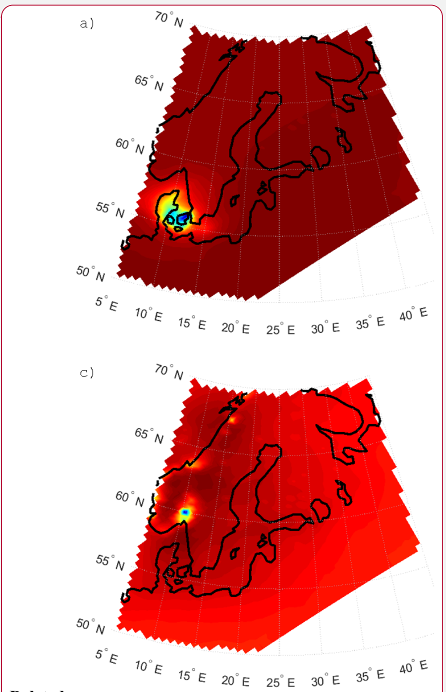

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Figure 12 also shows the impact of anthropogenic emissions from each Nordic country to the surface PM$_{2.5}$ over the Arctic. Overall, the impacts are very small, around a few per cent, as seen in the figure. The Danish emissions (Figure 12a) have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

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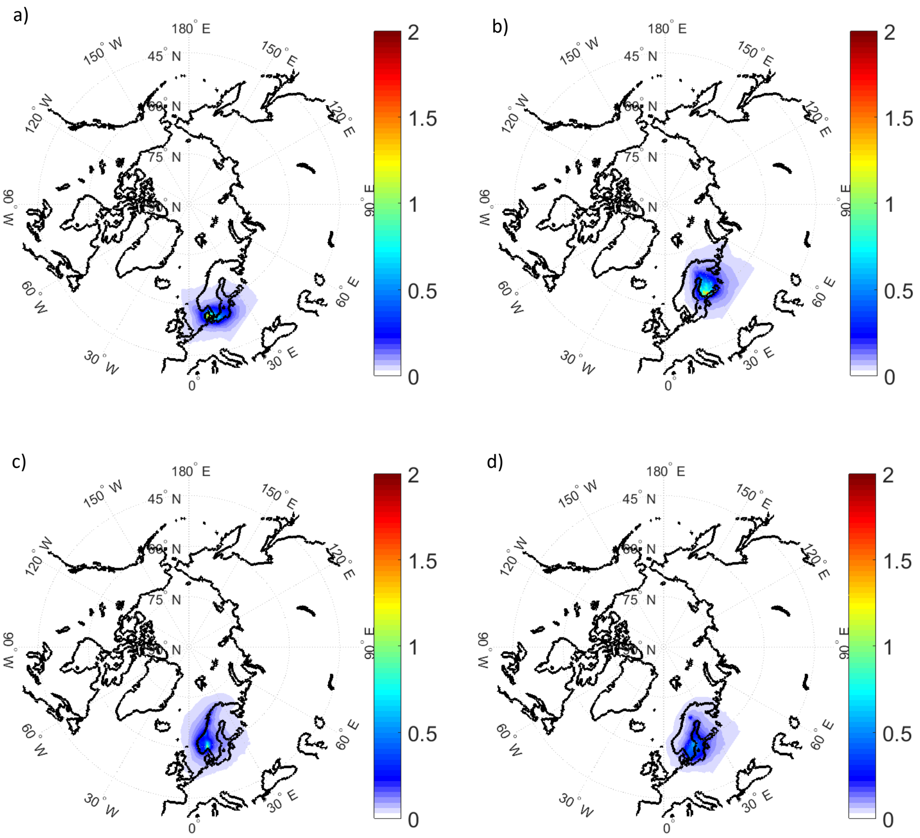

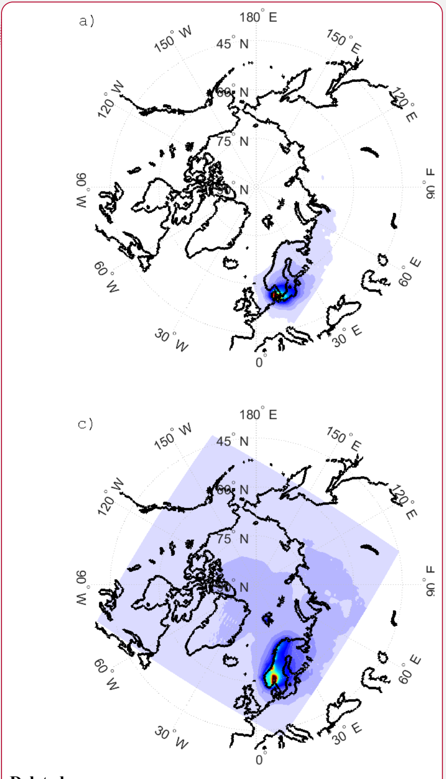

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Figure 12. Spatial distributions of annual population-weigthed mean absolute contributions (µg m^{-3}) of total emissions from a) Denmark, b) Finland, c) Norway, and d) Sweden to surface PM$_{2.5}$ levels over the Nordic and the Arctic regions (north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

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2.3. 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. 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 almost responsible for 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 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.

	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
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
Cost (million Euros)					
Acute	30 [29 30]	28 [27 28]	9 [9 10]	38 [37 38]	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
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

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.

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]**

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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+Waste sectors 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.

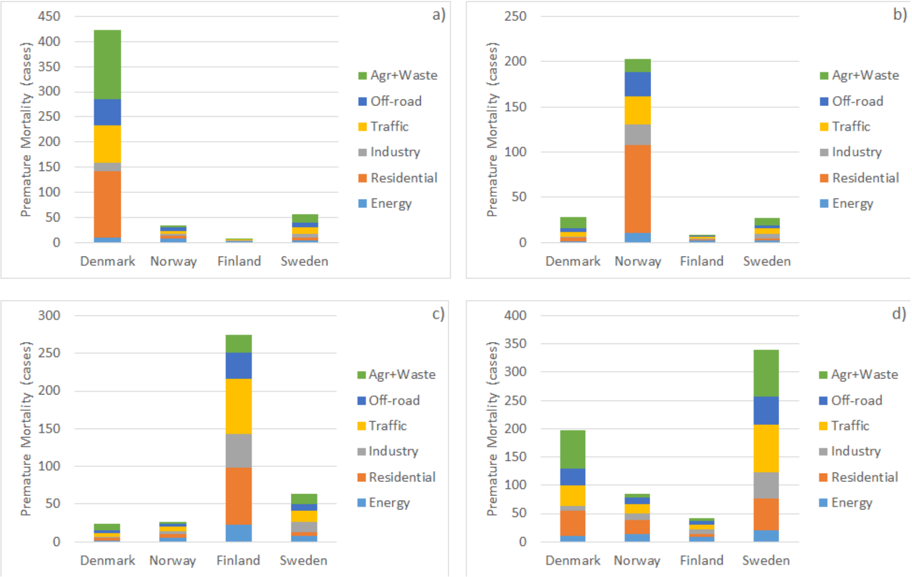

Figure 13. Source contributions from the anthropogenic emissions of a) Denmark, b) Norway, c) Finland, and d) Sweden to total premature mortality (acute+chronic) in the Nordic countries.

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.

Source	Receptors			
	Denmark	Finland	Norway	Sweden
Denmark	261 [256 266]	14 [14 15]	17 [17 18]	122 [119 124]

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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]

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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 downwind, which would require a coordinated effort in Europe.

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3. 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%.

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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 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.

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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.

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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).

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