**Wintertime Arctic Haze: Modelling wintertime sea-spray aerosols - a model-sensitivity study under Arctic Haze conditions**

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**Abstract.** Anthropogenic and natural emissions contribute to enhanced concentrations of aerosols, so-called Arctic Haze, in the Arctic winter and early spring. Models still have difficulties reproducing available observations. Whilst most attention has focused on the contribution of anthropogenic aerosols, there has been less focus on natural components such as sea-spray aerosols (SSA) – including sea salt sulphate and marine organics, which, under Arctic Haze conditions, can be significant contributors to fine and coarse mode aerosols, particularly in coastal areas. A model-sensitivity study of wintertime Arctic aerosol abundances shows evidence for enhanced local sources of SSA at Utqiagvik (northern Alaska, United States) during winter 2014. Models tend to underestimate sub-micron, and overestimate super-micron SSA in polar regions, including in the Arctic region. Quasi-hemispheric runs of the Weather Research Forecast model, coupled with chemistry model (WRF-Chem) are compared to aerosol composition data at remote Arctic sites to evaluate the model performance simulating wintertime Arctic Haze. Results show that the model overestimates sea salt (sodium and chloride) and nitrate and underestimates sulphate aerosols. Inclusion of more recent model used here, which includes a widely used SSA source function based on Gong et al. (1997), results in significantly improved model performance compared to observations at remote Arctic sites, notably for coarse mode sodium and chloride which are reduced. The improved model also simulates more realistic contributions of SSA to total mass for different aerosol modes, inorganic aerosols at different sites, ranging from 20-93% in the observations. The sensitivity of modelled SSA to processes influencing SSA production are examined in regional runs over northern Alaska (United States) where the Two thirds of the improved model performance
is from inclusion of the dependence on SSTs. Simulation of nitrate aerosols is also improved due to less heterogeneous uptake of nitric acid on SSA in the coarse mode and related increases in fine mode nitrate. This highlights the importance of interactions between natural SSA aerosols and inorganic anthropogenic aerosols contributing to Arctic Haze. Simulation of organic aerosols and the fraction of sea-salt sulphate are also improved compared to observations. However, the model underestimates episodes of high SSA, particularly in the with elevated observed concentrations of SSA components, and non-sea-salt sulphate at some Arctic sites, notably at Utqiaġvik (sub-micron, that were observed in winter 2014 during field campaigns at the Barrow Observatory, aerosols). Possible reasons are explored in higher resolution runs over northern Alaska for periods corresponding to Utqiaġvik – A field campaign in January and February 2014. The addition of a local source of marine organics is also included following previous studies showing evidence for an important contribution from marine emissions. Model results show relatively small sensitivity to aerosol dry removal with more sensitivity (improved biases) to using SSA marine organics, based on the campaign data, increases modelled organic aerosols over northern Alaska. However, underestimation compared to previous available data suggests that local natural sources from open leads as well as local anthropogenic sources are underestimated in the model. Missing local anthropogenic sources may also explain low modelled (sub-micron) non-sea-sulphate at Utqiaġvik (and certain other Arctic sites). The introduction of a higher wind speed dependence based on sub-micron data reported from an Arctic cruise. Sea-ice fraction, including sources from SSA emissions, also based on Arctic data, reduces biases in modelled sub-micron SSA while sea-ice fractions, including open leads, is are shown to be an important factor controlling modelled super-micron SSA, rather than sub-micron SSA. The findings of this study support analysis of the field campaign data pointing out SSA over the north coast of Alaska. The results presented here show that that modelled SSA are more sensitive to wind speed dependence, but that realistic modelling of sea-ice distributions is needed for simulation of local SSA, including marine organics. This study supports findings from the Utqiaġvik field campaign that open leads are the primary source of fresh and aged SSA, including marine organic aerosols, during wintertime at the Barrow Observatory, Utqiaġvik. Nevertheless, episodes of high observed SSA are still underestimated by the model at this site, possibly due to missing sources such as SSA production from breaking waves. An analysis of the observations and model results does and do not suggest an influence from blowing snow and frost flowers to SSA during the period of interest. Reasons for the high concentrations of sub-micron SSA observed at this site, higher than other Arctic sites, require further investigation. To improve model simulations of Arctic wintertime aerosols, new field data on processes influencing wintertime SSA production, in particular for fine mode aerosols, are needed as well as improved understanding about possible local anthropogenic sources.

1 Introduction

The Arctic region is warming faster than any other region on Earth (Allan, 2021). Greenhouse gases due to carbon dioxide, in particular carbon dioxide, and also due to short-lived climate forcers like methane, ozone and tropospheric ozone and aerosols have a significant impact on the environment, with a particularly strong warming effect in the Arctic region (AMAP, 2015; Allan, 2021). During winter and early spring, aerosols also affect clouds (aerosol-cloud indirect effects) and,
more specifically, cloud droplet number concentration and size by increasing the long-wave emissivity of clouds (long-wave warming effect) (AMAP, 2015; Allan, 2021)(Zhao and Garrett, 2015; Horowitz et al., 2020). This region is influenced by enhanced concentrations of aerosols (including sulphate (SO$_4^{2-}$). At this time of year, elevated aerosol concentrations of black carbon (BC), nitrate (NO$_3^-$), black carbon (BC)-non-sea salt (nss) sulphate (SO$_4^{2-}$) and organic aerosols (OA) during winter and spring are observed in the Arctic, a phenomenon called known as Arctic Haze (Rahn and McCaffrey, 1980; Barrie et al., 1994; Quinn et al., 2002). Transport of aerosols and their precursors from mid-latitudes anthropogenic emissions contribute to Arctic Haze (Heidam et al., 2004; Quinn et al., 2007; Law et al., 2014). Local within and near Arctic anthropogenic and natural sources also contribute to Arctic Haze during wintertime and the winter-spring transition (Law et al., 2017; Schmale et al., 2018; Kirpes et al., 2019). During wintertime 14% of organic mass at Alert originated from gas flaring in northern Russia (Leaitch et al., 2018). For example, gas flaring from Russia contributes to black carbon at Alert (northern Canada) and Utqiagvik (northern Alaska) (Stohl et al., 2013; Qi et al., 2017; Xu et al., 2017; Marelle et al., 2018). Metal industry and combustion sources, such as power generation, from Siberia (e.g. Kola peninsula) were identified as sources of pollution at Villum station, Greenland during winter and spring (Nguyen et al., 2013). Metal smelting from Siberia also contributes to SO$_4^{2-}$ at Zeppelin during wintertime (Hirdman et al., 2010). A more recent study by Winiger et al. (2019) showed that during wintertime Arctic sites, such as Utqiagvik, Alert, Zeppelin, are influenced by fossil fuel combustion emissions. Petroleum extraction on the North Slope of Alaska, including Prudhoe Bay, was found to influence aerosol distributions, composition, and particle growth at Utqiagvik, with enhanced growth of ultrafine particles (Kolesar et al., 2017; Kirpes et al., 2018).

, due to transport of anthropogenic aerosols and precursors from mid-latitude sources as well as within-Arctic emissions (Heidam et al., 2004; Quinn et al., 2007; Law et al., 2014, 2017; Schmale et al., 2018). Natural aerosol sources also contribute to Arctic Haze such as dust, volcanic emissions and sea-spray aerosols (SSA) (Rahn et al., 1977; Barrie and Barrie, 1990; Quinn et al., 2002). Dust is not only transported from mid-latitudes sources (Asia, Africa), but it is also produced within the Arctic, with local dust contributing up to 85% to total dust burden in the Arctic also contribute to wintertime Arctic aerosol burdens (Barrie and Barrie, 1990; Quinn et al., 2002; Zwaanink et al., 2016; Kirpes et al., 2018), with SSA also peaking in the wintertime (Zwaanink et al., 2016; Schmale et al., 2022). During wintertime, fresh SSA (including sodium ions (Na$^+$), chloride ions (Cl$^-$), sea-salt (ss) SO$_4^{2-}$ and marine organics) can be a significant fraction of particulate matter, 40% of super-micron (1 to 10 $\mu$m particle diameter) and 25% of sub-micron (up to 1 $\mu$m particle diameter) (Quinn et al., 2002). While studies have largely focused on anthropogenic sources of Arctic Haze influencing, in particular BC and SO$_4^{2-}$, there have been fewer studies on the contribution of SSA, the focus of this study. The primary mechanism leading to the formation of SSA is the heterogeneous uptake on SSA surfaces leading to inorganic aerosol formation (Su et al., 2022) which can influence the ability of models to simulate Arctic Haze. In this study, we focus on SSA under wintertime Arctic Haze conditions.

SSA are produced by bubble bursting (jet-drop and film-drop formation) on the sea surface due to wind stress during whitecap formation (Monahan et al., 1986). For this reason, wind speed is a significant parameter affecting SSA productivity (Russell et al., 2010; Saliba et al., 2019). Arctic SSA emissions also depend on sea surface temperatures (SSTs)
and salinity (Jaegle et al., 2011; Sofiev et al., 2011; Revell et al., 2019). Frost flowers and blowing snow have also been proposed as a source in polar regions during wintertime (Xu et al., 2013; Huang and Jaegle, 2017). SSA is composed primarily of sodium (Na\(^+\)), chloride (Cl\(^-\)), organics and sea-salt (ss) SO\(_4^{2-}\). SSA may influence cloud formation, including Arctic mixed-phase clouds (Adachi et al., 2022), since they can act as cloud condensation nuclei (CCN) (Quinn et al., 2017), or organics may contribute to ice nucleating particles (INPs) (Burrows et al., 2013). Arctic warming is leading to a decrease of in summer sea-ice during summertime and, as a result, less and and thinner sea-ice is forming during wintertime (Stroeve et al., 2012). Thus, new SSA sources, such as open ocean and leads, may contribute more in the future to the total aerosol burden. Increases in the area of the open ocean or more open leads in sea-ice may increase winter SSA over Arctic coastal regions, impacting CCN concentrations and radiative forcing potentially influencing radiative forcing (Ma et al., 2008; Ma et al., 2008; Eidhammer et al., 2010; Porta et al., 2020).

A detailed analysis of in-situ aerosol composition in Utqiagvik revealed that, due to long-range transport from the North Pacific (due to strong winds in source regions, such as in the Pacific Ocean), sub-micron SSA peaks in winter and early spring, while super-micron SSA peaks in summer, due to sea-ice retreat. The ability of models to capture wintertime Arctic aerosols has largely focused on the evaluation of anthropogenic Arctic Haze components, in particular BC and SO\(_4^{2-}\) (e.g. Eckhardt et al., 2015; Quinn et al., 2002). However, in winter, super-micron SSA mass concentrations increase in the presence of open leads, while sub-micron SSA appear to be more influenced by long-range transport (May et al., 2016; Kirpes et al., 2019). Whaley et al. (2022). Whaley et al. (2022) showed that, in general, models underestimate SO\(_4^{2-}\) and BC in winter. Kirpes et al. (2018) analysed atmospheric particle samples collected in winter. Very few model studies have assessed both anthropogenic and natural aerosols, like SSA, and these models do not make generally the distinction between nss and ss components of SO\(_4^{2-}\) or OA. A recent study by Moschos et al. (2022a) estimated that wintertime Arctic OA have largely anthropogenic origins but a possible contribution from local marine organics was not considered. An analysis of single-particle data from a field campaign near Utqiagvik, northern Alaska, in winter 2014 in Utqiagvik. They found that the samples were influenced by air masses from the Arctic Ocean to the north and Prudhoe Bay oilfields to the east. Aged SSA were always internally mixed with secondary SO\(_4^{2-}\), or with both SO\(_4^{2-}\) and NO\(_3^-\) and reduced chlorine, suggesting anthropogenic influence from background Arctic Haze or Alaskan oil-field emissions. Kirpes et al. (2019) concluded that, showed that, in addition to organic-sulphate haze aerosols, there were abundant fresh SSA, based on the presence of Na\(^+\) and Cl\(^-\), Cl\(^-\), magnesium and sulfur in ratios similar to seawater, including marine organic aerosols, that were produced locally from open leads, with indications of and included marine OA originating from secretions from sea ice algae and bacteria based on observed enrichments in carbon/sodium (C:Na\(^+\)) ratios. Previous studies of the Arctic and North Atlantic during wintertime and the winter-spring transition also showed that the majority (Kirpes et al., 2018, 2019). It can be noted that during winter, fresh SSA can be a significant fraction of particulate matter, contributing to 40% of super-micron (1 to 10 µm particle diameter) and 25% of sub-micron organic mass (OM) is highly correlated with Na\(^+\) concentrations (up to 1 µm particle diameter) aerosol mass (Russell et al., 2010; Shaw et al., 2010; Frossard et al., 2011; Leaitch et al., 2018; Quinn et al., 2002). Frost flowers with organic salt coatings have also been proposed as a possible source of wintertime SSA (Xu et al., 2013), although Kirpes et al. (2019) found
no evidence of frost flowers or blowing snow as a potential source, supporting the findings of older studies (Roscoe et al., 2011). Regional and global models have difficulties capturing wintertime Arctic Haze composition and often underestimate Kirpes et al. (2018) also measured aged SSA, which made up the majority of the sub-micron number fraction during their campaign at Utqiagvik (formerly known as Barrow). These aged SSA were internally mixed with secondary SO$_4^{2−}$ and BC (Bond et al., 2013; Eckhardt et al., 2015; Sato et al., 2016; Schwarz et al., 2017; Whaley et al., 2022), while the contribution of SSA to Arctic Haze remains poorly evaluated (Kirpes et al., 2019). Representation of or both SO$_4^{2−}$ and NO$_3^{−}$, and Cl$^{−}$ was depleted, indicating that multi-phase reactions had occurred during transport. The aged SSA were sampled in air masses influenced by background Arctic Haze and regional northern Alaskan oil field emissions. Their findings support an earlier analysis of Ny-Alesund (Svalbard) data showing that aged SSA were always internally mixed with NO$_3^{−}$, SO$_4^{2−}$, and organics (Chi et al., 2015). In fact, heterogeneous reactions occurring on the surface of SSA concentrations in models has been improved over recent years, but with less focus on the Arctic winter. For example, SSA source functions with updated dependencies on wind speeds, sea surface temperatures (SSTs) or salinity (Revell et al., 2019; Jaeglé et al., 2011; Sofiev et al., 2011) have led to improve simulation of super-micron SSA. However, sub-micron SSA is often still underestimated (Sofiev et al., 2011; Huang and Jaeglé, 2017), and sub-micron emissions of SSA from frost flowers and blowing snow have been included in models, involving uptake of sulphuric, nitric or organic acids, and associated Cl$^{−}$ displacement, are more evident in aged SSA (Xu et al., 2013, 2016; Huang and Jaeglé, 2017; Li et al., 2018; Chen et al., 2016). Modelled SSA including a source of frost flowers captures better monthly SSA concentrations at Alert during wintertime, while a source of blowing snow overestimates observations (Huang and Jaeglé, 2017; Marelle et al., 2021). At Utqiagvik during January and February a source of blowing snow improves modelled SSA; however it still cannot explain the high observed SSA, while the blowing snow explains high observed SSA in the Antarctic (Huang and Jaeglé, 2017). Chen et al. (2016) showed that uncertainties in modelled SSA can have a significant impact on sub-micron and super-micron NO$_3^{−}$ due to heterogeneous uptake of nitric acid (HNO$_3$) on SSA which produces NO$_3^{−}$. Uptake of sulphuric acid on SSA in the marine boundary layer can result in SO$_4^{2−}$ production (Alexander et al., 2005; Li et al., 2018) and Wu et al. (2019) emphasised the importance of heterogeneous reactions occurring on SSA for improved simulation of SO$_4^{2−}$ and NO$_3^{−}$ size distributions. Since nitric and sulphuric acid have largely anthropogenic origins this highlights important links between natural SSA and anthropogenic inorganic aerosols.

In this study, the performance of the This study is motivated by the findings of Kirpes et al. (2018, 2019) about wintertime Arctic aerosols, including SSA coated with marine organics, in northern coastal Alaska. Our main objectives are to assess the ability of a regional chemical-aerosol model (Weather Research Forecast model, coupled with chemistry (WRF-Chem)) is examined with regard to its ability to simulate Arctic Haze composition as well as SSA components, including ss-SO$_4^{2−}$ and marine organics. The model is first evaluated against available data) to simulate wintertime aerosols, in particular SSA, under Arctic Haze conditions, and to examine model sensitivity to processes which may be influencing SSA aerosols over northern Alaska, in particular. We also assess the observed and modelled contribution of SSA to total inorganic aerosols during Arctic wintertime.

Firstly, we focus on improving the model SSA emission scheme over the wider Arctic and the sensitivity to more recent treatments of SSA wind-speed and SST dependencies, is investigated. Inclusion of a marine organic source is also examined
The findings of Kirpes et al. (2019) are used as a basis for a more focused regional study to evaluate modelled Arctic wintertime aerosol composition in northern Alaska during winter. This includes updating the wind speed dependence, including a dependence on SSTs, and adding sources of marine OA and ss-SO$_4^{2-}$. Due to the links between SSA and other inorganic aerosols, contributing to Arctic Haze, model results are evaluated against observations of all aerosol components at remote Arctic sites. To our knowledge, these aspects have not been considered in previous studies using either WRF-Chem or other models (e.g., Whaley et al. (2022)). Secondly, we investigate the sensitivity of modelled SSA to processes influencing SSA, including organics and other inorganic aerosols, at Utqiagvik using the improved model run at higher resolution over northern Alaska for periods corresponding to the Kirpes et al. (2018, 2019) campaign. The sensitivity of the model results to processes influencing SSA production and concentrations are investigated including aerosol dry deposition, the addition of a local source of marine organics, wind speed dependence and sea-ice fraction. Missing local sources of marine organics are also examined. Fractions are investigated based on the findings of Kirpes et al. (2019). A companion paper, Ioannidis et al. (2022) (in prep.), examines the contribution of remote and regional anthropogenic emissions to ArcticBC in northern Alaska and northern Russia during wintertime. This field campaign and other data collected in the Arctic. A possible contribution from frost flowers or blowing snow is also considered.

The model setup, including the emissions are anthropogenic and natural emissions, is described in Section 2. The observed aerosol composition aerosol observations used to evaluate the model performance are introduced in Section 3. The model runs, including sensitivity simulations, together with results Details about the SSA emission scheme in the base model version, together with improvements to this scheme, are presented in Sections 4 and 5. First, in Section 4, simulated Arctic Haze, focusing on SSA, is evaluated at remote Arctic sites. Second, Section 4. Evaluation of simulated SSA, as well as other inorganic and organic aerosols, against Arctic observations, are presented in Section 5, the results together with an estimation of the contribution of SSA to total inorganic aerosols. Results from the regional study over northern Alaska during wintertime and sensitivity of results to processes influencing SSA production in the model are presented. are presented in Section 6. The implications of our findings for the simulation of Arctic Haze aerosols and conclusions SSA under wintertime Arctic Haze conditions are presented in Section 6. (Conclusions).

2 WRF-Chem

2.1 Model Setup

WRF-Chem chemical-transport model version 3.9.1.1 is used to simulate quasi-hemispheric and regional Arctic Haze aerosols and to examine local SSA sources over northern Alaska. WRF-Chem in this study. It is a fully coupled, online meteorological and chemical transport mesoscale model (Grell et al., 2005; Fast et al., 2006). Recent improvements into the WRF-Chem model over the Arctic are included in the version used in this study here (Marelle et al., 2017). The model setup, including meteorological and chemical schemes, is shown in Table 1. Briefly, Yonsei University (YSU - boundary layer), Model Version 5 similarity (MM5 - surface layer) and Noah-Multiparameterization Land Surface Model (NOAH MP, land surface model) are used. More details about the NOAH MP scheme are given in APPENDIX A.
Table 1. WRF-Chem model setup. The source functions for sea-spray emissions and their main updates are summarised below. CONTROL includes only Gong et al. (1997), while HEM_NEW includes updates to the SSA emission scheme. See text for details.

<table>
<thead>
<tr>
<th>Parameterization</th>
<th>Options</th>
</tr>
</thead>
<tbody>
<tr>
<td>Planetary boundary layer</td>
<td>Yonsei University (YSU) - (Hong et al., 2006)</td>
</tr>
<tr>
<td>Surface layer</td>
<td>Pennsylvania State / NCAR Mesoscale Model Version 5 (MM5) similarity (Grell et al., 1994; Jiménez et al., 2012)</td>
</tr>
<tr>
<td>Land surface</td>
<td>NOAH MP (Niu et al., 2011)</td>
</tr>
<tr>
<td>Microphysics</td>
<td>Morrison (Morrison, 2009)</td>
</tr>
<tr>
<td>SW &amp; LW radiation</td>
<td>Rapid Radiative Transfer Model (RRTMG - Iacono and D. (2008))</td>
</tr>
<tr>
<td>Cumulus parameterization</td>
<td>Kain-Fritsch with cumulus potential (KF-CuP) (Berg et al., 2013)</td>
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Chemistry (WRF-Chem)

- **Aerosol model**: MOSAIC 8-bins (Zaveri et al., 2008)
- **Gas-phase chemistry**: Statewide Air Pollution Research Center SAPRC-99
- **Photolysis**: Fast-J (Wild et al., 2000)

**CONTROL**

- **Sea-salt emissions**: Gong et al. (1997)
- **Sea-spray emissions (marine organics)**: Fuentes et al. (2010, 2011)
- **Satellite wind speed dependence, SST dependence**: Salisbury et al., 2014; Jaeglé et al., 2011
- **ss-SO$_4^{2-}$ source**: Kelly et al., 2010

All the various well-known processes for aerosols in the atmosphere, like nucleation, evaporation, coagulation, condensation, dry deposition, aerosol/cloud interactions and aqueous chemistry, are included in the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC, Zaveri et al. (2008)). MOSAIC treats all the major aerosol species, such as SO$_2^{2-}$, NO$_3^-$, Cl$^-$, ammonium (NH$_4^+$), Na$^{+}$, calcium (Ca$^{2+}$), BC, and OA. Reactive inorganic species such as potassium (K$^+$) and magnesium (Mg$^{2+}$) are not modelled in MOSAIC. The size distribution of each aerosol species is represented by eight bins, from $39\text{ nm to }0.0391$ micrometres ($\mu$m) to 10 $\mu$m: $[0.0391 \text{ to } 0.0781]$, $[0.0781 \text{ to } 0.1562]$, $[0.1562 \text{ to } 0.3125]$, $[0.3125 \text{ to } 0.625]$, $[0.625 \text{ to } 1.25]$, $[1.25 \text{ to } 2.5]$, $[2.5 \text{ to } 5.0]$ and $[5.0 \text{ to } 10.0]$ in $\mu$m. Each bin is assumed to be internally mixed, and both mass and number are simulated. As a result, aerosols are aged when emitted (coagulated with other species). The MOSAIC version used in this study also includes 18 irreversible heterogeneous reactions (see Table 1 in Zaveri et al. (2008)), such as the reaction of HNO$_3$ on sodium chloride (NaCl) to form sodium nitrate (NaNO$_3$), with depletion.
of Cl−. The reaction between NaCl and sulfuric acid (H2SO4) to produce sodium sulfate (Na2SO4), with associated Cl− depletion, is also included. Nighttime chemistry, notably heterogeneous hydrolysis of dinitrogen pentoxide leading to HNO3 formation, is also included (Archer-Nicholls et al., 2014). The applied MOSAIC version includes secondary organic aerosol (SOA) formation from the oxidation of anthropogenic and biogenic species (Shrivastava et al., 2011; Marelle et al., 2017) and is combined with SAPRC-99 gas-phase chemistry. In the base model, OA is the sum of SOA and anthropogenic emissions of organic matter (OM). Aqueous chemistry in grid-scale (Morrison, 2009) and subgrid-scale clouds (Berg et al., 2015) is also included. Aerosol sedimentation in MOSAIC is calculated throughout the atmospheric column based on the Stokes velocity scheme, as described in Marelle et al. (2017).

2.2 Emissions

This section provides details about the emissions that are used in the simulations. More details are provided about SSA emissions since this is the focus of this study. Wet removal of aerosols by grid-resolved stratiform clouds (precipitation) includes in-cloud and below-cloud removal by rain, snow, and graupel by Brownian diffusion, interception, and impaction mechanisms following Easter et al. (2004) and Chapman et al. (2009). Wet-removal due to subgrid-scale convective clouds (Berg et al., 2015) is also included in this MOSAIC version and described in previous studies (Marelle et al., 2017; Raut et al., 2017).

2.1.1 Anthropogenic and natural emissions

2.2 Anthropogenic and natural emissions

Anthropogenic emissions are from the Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants version 6 (ECLIPSE v6b) inventory, with a resolution of 0.5° x 0.5° (Whaley et al., 2022), including emissions of organic matter (OM). Emissions of dimethyl sulfide (DMS), mineral dust, and lightning NOx and nitrogen oxides (NOx) are calculated online in the model (see Marelle et al. (2017) and references therein). Dust emissions in MOSAIC are calculated following Shaw et al. (2008). Biogenic emissions for 2014 are calculated online using Model of Emissions of Gases and Aerosol from Nature (MEGAN) model (Guenther et al., 2012). Details about the treatment of SSA emissions and their improvement in the model are provided in Section 4 and summarised in Table 1.

2.2.1 Sea-spray emissions

In the control simulation, sea-salt emissions of Na+ and Cl− are included. They are calculated per particle radius, with 1000 sub-bins per MOSAIC bin, using the density function dF/dr (in particles m−2 s−1 μm−1) from (Gong et al., 1997) (G97 from now on) which represents the rate of seawater droplets forming per unit area (sea surface) and per increase of particle radius and its derived from the source function based on laboratory experiments described in Monahan et al. (1986) (MO86 from now-on).
Figure 1. WRF-Chem simulation domains: (a) d01 is the 100km domain and (b) d02 is the 20km domain. d02 shows sea-ice fractions interpolated at 20km obtained from NCEP FNL (at 1°×1° resolution (NCEP, 2000)). See text for details.

\[
\frac{dF}{dr} = 1.373 \times U_{10}^{3.41} \times r^2 \left(1 + 0.057 \times r^{1.05}\right) \times 10^{1.19e^8}
\]

where \( F \) is a function of \( U \) and \( r \), \( r \) is the particle radius at relative humidity (RH) equal to 80%, \( U \) the 10m-elevation wind speed and \( B = \frac{(0.380 - \log r)}{0.650} \). The source function is applied for particles with dry diameters of 0.45 \( \mu \)m or more. For particles with dry diameters less than 0.45 \( \mu \)m, a correction is applied to the formula based on O’Dowd et al. (1997). This approach is based on the whitecap method, where the emission flux scales linearly with the fraction of ocean area covered by whitecaps. Over open ocean, the whitecap fraction, \( W(U) \), is determined as a function of wind speed (Monahan and Muirheartaigh (1980) or MO80 from now on):

\[
W(U) = 3.84 \times 10^{-6} \times U_{10}^{3.41}
\]

This expression for \( W(U) \) is included implicitly in Equation (1) following details provided in MO80. In the base version of WRF-Chem SSA emissions are calculated for every grid cell, which is open ocean or salt water lakes. In this study, the grid cell which is covered by sea-ice is considered and then the fraction of that ice-free grid is used. In this way, SSA emissions from open leads are taken into account.

WRF-Chem simulation domains: d1 is the 100km domain and d2 is the 20km domain.
2.3 Simulations

Two simulation domains on a polar stereo- graphic projection are used in this study, as shown in Figure 1. The first (parent) domain (d01) covers a large part of the Northern Hemisphere with 100 × 100 km horizontal resolution. The boundary and initial conditions are derived from National Centres for Environmental Prediction Final meteorological reanalysis data (NCEP FNLI4°x1°), (NCEP (2000) meteorological reanalyses) and Model for OZone And Related chemical Tracers (MOZART, Emmons et al. (2010)). The nested domain (d02), run at a horizontal resolution of 20 × 20 km, covers continental Alaska, a small area of northwest Canada, and the Chukchi and Beaufort Seas (see Figure 1). 50 vertical levels and grid nudging are used for the 100 km resolution domain, while calculating spectral nudging parameters as in Hodnebrog et al. (2019), spectral nudging, following Hodnebrog et al. (2019), is implemented in the nested domain. WRF-Chem temperatures and winds are nudged at each dynamical step to the reanalysis, which are and updated every 6 hours, above the atmospheric boundary layer.

The simulations performed in this study are discussed in sections 4 and 5. Simulations at 100 km Two simulations at 100 km are performed, one using the base model version (CONTROL) and one using CONTROL plus improvements to the SSA emissions (HEM_NEW) (see Section 4). They are run for 4 months from November 2013 until the end of February 2014, with the first two months considered as spin-up. The model is run results are evaluated over the wider Arctic in Section 5. Results from HEM_NEW are then used as boundary conditions for regional runs over northern Alaska at 20 km for two different periods (23–28 January 2014 and 24–28 February 2014) corresponding to the campaign which took place in Utqiagvik and campaign described earlier (Kirpes et al. (2018, 2019) Kirpes et al., 2018, 2019). KRP18 and KRP19 from now on, respectively, see also sub-section 3.2). For these simulations, the initial, and boundary conditions are derived from the quasi hemispheric simulation A In this case, the model is run for 4 days prior to the beginning of each campaign and considered as spin-up. Results from a series of sensitivity runs are performed to examine processes affecting SSA emissions over northern Alaska. They are summarized in Table 3 and discussed in detail in Section 5. At 20 km for all the simulations, 4 days prior to the beginning of the campaign considered spin up, on a regional scale are discussed in Section 6. In all runs, the model results are output every 3h.

3 AerosolsAerosol Observations

The sites discussed in this section are shown in Figure 1.

3.1 Routine monitoring sites

Surface mass concentration data (for aerodynamic diameters (defined as dₐ) < 10 μm), from the EMEP (European Monitoring and Evaluation Programme) dataBASe (EBAS - http://ebas.nilu.no) for Zeppelin, Ny-Alesund, Norway (78.9N, 11.88W) and Alert, Canada (82.5N, -63.3W), are used to evaluate the quasi hemispheric 100 km model simulations together with total suspended particulate (TSP) (cutoff at 20 μm) data from Villum Research Station, Station Nord, Greenland (81.6N, -16.7W),
referred to as Villum from now on (reporting total suspended particulates). The data are collected on a daily (Zeppelin) and weekly (Villum, Alert) basis. At Alert, and Zeppelin (Aas et al., 2021) observations for Na$^+$, Cl$^-$, NH$_4^+$, Cl$^-$, NO$_3^-$ and total SO$_4^{2-}$ measured with ion chromatography are used (Sharma et al., 2019). This is also the case at Zeppelin (Aas et al., 2021). At Villum, the same observations are collected using a filter-pack over a week and analysed using ion-chromatography (Cl$^-$, total SO$_4^{2-}$), ion chromatograph (Na$^+$) and segmented flow analysis (NH$_4^+$) and cation ion chromatography (Na$^+$). For all the EBAS stations, the units of observed inorganic aerosols (NH$_4^+$, total SO$_4^{2-}$, NO$_3^-$) are converted to model units—mass concentrations (µgm$^{-3}$), to compare to model results, using the ratio of molar weights of NH$_4^+$-NO$_3^-$, SO$_4^{2-}$ to molar weights of nitrogen or sulphur, respectively. With regard to measurement uncertainties, EBAS documentation notes, in the case of Alert only, that there are uncertainties of around uncertainties ranging between 33% and 36% in Na$^+$, for Na$^+$, total SO$_4^{2-}$, NO$_3^-$ and Cl$^-$ respectively, and higher uncertainties (43%) for NH$_4^+$ at Alert. These high uncertainties may be related to uncertainties in the size cut-off of sub-micron filters. Uncertainties in coarse particle observations are based on the difference between high-volume (TSP) filters collected outside and sub-micron filters collected inside.

Surface mass concentration data, diameter less than 2.5 µm ($d_a <$ 2.5 µm) − mass concentration data from the Interagency Monitoring for Protected Visual Environments (IMPROVE) database is also used for model evaluation for Simeonof (55.3N, -160.5W), a sub-Arctic site on the Aleutians islands, south of Alaska and an inland site, Gates of the Arctic (66.9N, -151.5W). GoA from now on, which is located 391 km south-east of Utqiaġvik – town in northern Alaska (see Fig. 1). The samples are collected on-site (e.g. Simeonof site) over 24 hours every three days (http://views.cira.colostate.edu/fed/QueryWizard/Default.aspx, Malm et al. (1994))). At these two sites observations of Na$^+$, Cl$^-$, OC$^+$, Cl$^-$, organic carbon (OC), NO$_3^-$ and total SO$_4^{2-}$ are used. To compare with the OC observations at the two Alaskan sites, modelled OA is divided by 1.8, the reported ratio of OM/OC in the documentation for these two stations (Malm et al., 1994). In this study, mass concentration data with diameter $d$ ≤ 2.5 µm are defined as fine mode aerosols, while diameter $d_a <$ 10 µm are defined as coarse mode aerosols.

Sub-micron (d$< 1$ µm) and super-micron (1.0 < d$< 10$ µm) surface mass concentration data from the National Oceanic and Atmospheric Administration (NOAA) Barrow observatory Observatory (71.3N, -156.8W), near Utqiaġvik town (Utqiaġvik from now on), is also used in this study, with daily and weekly temporal coverage, respectively. The sampling site is located 8 km northeast of Utqiaġvik, 20 km town, 3km southwest of the Arctic Ocean, covered with snow during winter and 20m above mean sea level (msl), with a prevailing, east-northeast wind off the Beaufort Sea. Concentration data (Na$^+$, Cl$^-$, NH$_4^+$, Na$^+$, Cl$^-$, NO$_3^-$ and total SO$_4^{2-}$ - mass concentrations) are determined by ion chromatography (Quinn et al., 1998) and are sampled only for wind directions between 0 and 130 degrees (with 0 degrees indicating north). According to Quinn et al. (2002) measurement uncertainties of measurement uncertainties in sub-micron SSA components and SO$_{nss}$-SO$_4^{2-}$ are below 4%, while for 1.0 ± 6.1% µgm$^{-3}$ (concentration ± 95% uncertainty). The uncertainties in sub-micron NH$_4^+$ they are below 0.2 ± 7.8% µgm$^{-3}$. The uncertainties in super-micron aerosols can be up to 7 times higher than for sub-micron aerosols, since 7 daily super-micron samples are collected for every (weekly) super-micron sample. Measurement uncertainties are also due to sampling losses in the inlets. At Utqiaġvik, for particles with a diameter up to 10 µm, losses in the inlet system, from all loss
mechanisms, are estimated to be less than 10%, and losses of particles between the diameters of 0.01 and 1 μm are below 5% \cite{Sheridan2001}. Observed:

At each site observed ss-SO$_4^{2-}$ is calculated from observed Na$^+$ concentrations and the mass ratio of SO$_4^{2-}$ to Na$^+$ in seawater of 0.252 \cite{Bowen1979, Calhoun1991} and nss-SO$_4^{2-}$ is the difference between total SO$_4^{2-}$ and ss-SO$_4^{2-}$. Note that, in some cases, observed nss-SO$_4^{2-}$ has small negative concentrations, due to depletion of ss-SO$_4^{2-}$ through fractionation processes \cite{Quinn2002}. We note that, apart from the sub-micron observations at Utqiagvik and fine mode observations from the IMPROVE database, there are no other sub-micron or fine-mode observations collected routinely in the Arctic, as also reported recently by Schmale et al. \cite{2022}. Finally, data from a scanning mobility particle sizer (SMPS), located at Utqiagvik, are used and measures the particle size distribution at high temporal resolution, with a size range from 8.6 to 985 nm (0.0086 to 0.985 μm). Here, only observations at Utqiagvik are used since observations at the other Arctic sites used in this study are not available for winter 2014, as discussed by Freud et al. \cite{2017}.

Evaluation of modelled aerosol composition (runs CONTROL and HEM_NEW) against in situ observations of (a) coarse mode aerosols (d$_a < 10 \mu$m) at Alert, Canada (standard temperature pressure (STP) conditions), (b) TSP aerosols (d$_a < 10 \mu$m) at Villum, Greenland and (c) coarse mode aerosols (d$_a < 10 \mu$m) at Zeppelin, Norway in UTC. The black line shows model results from the CONTROL run; the red line shows the HEM_NEW run, while observations are shown as blue crosses. Villum and Alert observations are weekly averages, and the corresponding model weekly averages are shown as black diamonds for CONTROL and red pentagons for HEM_NEW. Zeppelin observations are daily 24h averages. Observations are shown only when they are available. See the text for details about the observations and model runs.

The model Stokes diameter (r$_d$) aerosol diameter is converted to aerodynamic diameter using the d$_a$ using the Seinfeld and Pandis \cite{1998} formula. Thus, the diameter of modelled sub-micron particles is up to 0.73 μm (including the first four MOSAIC bins and a fraction of the 5th-5th bin), and super-micron particle diameters are between 0.73 to 7.3 μm (fraction 5th-bin, 6th and 7th-5th bin, 6th and 7th bins and fraction 8th-8th bin). Seven MOSAIC bins and a fraction of the 8th-8th bin are used (modelled stokes-Stokes r$_d ≤ 7.3 \mu$m) to compare with Alert and Zeppelin observations (aerodynamic d$_a < 10 \mu$m; coarse mode). All model aerosol bins are used to compare with observations at Villum, where the observations are reported as total suspended particulates (TSP); i.e. there is no cutoff-TSP. For each site, modelled aerosols are estimated at the same conditions (temperature, pressure) as the reported observations. Overall, particles at Alert, observed total OC is assumed to include SOA, anthropogenic OA emissions and marine organics. Thus, from now on it will be referred to as tOC, to distinguish from OA defined earlier and OM. Aerosol measurements with different size ranges (up to 1.0 μm, 2.5 μm, and 10 μm) are used to validate the model performance in each domain results.

### 3.2 Campaign data

Details about the field campaign (January 23–27 and February 24–28, 2014) measurements near Utqiagvik, Alaska can be found in KRP18 and KRP19. Briefly, atmospheric particles were collected using a rotating micro-orifice uniform deposition impactor located 2–m–2m above the snow surface at a site located 5–km–5km across the tundra from the NOAA Barrow Observatory and inland from the Arctic Ocean. The sampled particles were analysed by computer-controlled scanning electron
microscopy with energy scattering X-ray spectroscopy (CCSEM-EDX) to determine the individual particle morphology and elemental composition. The analysed samples were collected either during daytime or nighttime, and only when wind directions were between 75 and 225 degrees, corresponding to minimise local pollution influence. Data analysis provided information about the different chemical components as a fraction of the total number of particles sampled observed during the campaign.

4 Processes influencing Model SSA over the wider Arctic emission treatments and their contribution to wintertime Arctic aerosols. This section focuses on evaluating the capability of the model to simulate Arctic Haze aerosols during wintertime and improving model treatments of SSA. Briefly, updates

This section introduces the treatment of SSA emissions in the base simulation (CONTROL), sea-salt emissions are calculated using the G97 parametrization scheme, including the MO80 whitecap method. All the updates described below are included in a new quasi-hemispheric simulation (HEM_NEW) with the aim to improve the model. This includes addition of marine organics (Fuentes et al., 2010), using a more recent whitecap method (Salisbury et al., 2014), including the dependence of SSA emissions on SST (Jaeglé et al., 2011), and the addition of a ss-SO$_4^{2-}$ component, based on Kelly et al. (2010). HEM_NEW simulation is then evaluated (sub-section 4.6) compared to CONTROL and the observations at the different sites followed by a discussion of the new results.

4.1 Anthropogenic and natural aerosols in the Arctic

First considering the observations, at remote sites such as Alert (Fig 2a), observed Na$^+$, Cl$^-$ and NO$_3$ coarse mode mass concentrations do not exceed 0.3, 0.5 model version of WRF-Chem using the MOSAIC aerosol scheme, followed by a description of the updates to the SSA emissions implemented in the model. The model is run with the original scheme (CONTROL run) and 0.09 $\mu$g m$^{-3}$, respectively, during the study period. Total SO$_4^{2-}$ (sum of ss-SO$_4^{2-}$ and nss-SO$_4^{2-}$) reach 0.44 $\mu$g m$^{-3}$, which is mostly nss-SO$_4^{2-}$, as ss-SO$_4^{2-}$ does not exceed 0.09 $\mu$gm$^{-3}$, likely to be due to long-range transport from sources in north central, western, northwest Russia and Europe (Leaitch et al., 2018). Similar magnitudes have been reported in previous studies during winter months (Leaitch et al., 2018). NH$_4^+$ peaks at 0.06 $\mu$gm$^{-3}$ and originates from Russia and Europe during winter (Leaitch et al., 2018). At Villum (Fig 2b), observed TSP Na$^+$, Cl$^-$ and NO$_3$ reach up to 0.12, 0.13 and 0.06 $\mu$g m$^{-3}$, respectively. These concentrations are lower than at Alert which could be explained by the fact that during winter the sea surrounding Villum station is frozen (Nguyen et al., 2013). Total SO$_4^{2-}$ does not exceed 0.2 $\mu$g m$^{-3}$ and is mostly nss-SO$_4^{2-}$ (up to 0.18 $\mu$g m$^{-3}$) with the updates (HEM_NEW run). Results from both runs are evaluated against observations in the Arctic, in Section 5.

4.1 SSA emissions - CONTROL run

SSA emissions in MOSAIC are calculated per particle radius, with 1000 sub-bins per MOSAIC bin, assuming that sea-salt is a simple mix of pure NaCl and using the density function dF/dr (in particles m$^{-3}$) based on (Massling et al., 2015) Gong et al. (1997) and is the dominant component of Arctic Haze at this site (Lange et al., 2018)
NH$_3^+$ concentrations at Villum are up to 0.1 μg m$^{-3}$. In the high Arctic, Na$^+$ could potentially also originate from anthropogenic sources which could account for up to 35% of total Na$^+$ (Barrie and Barrie, 1990). Note that this source is not included in the model, or in models generally. Higher Na$^+$, Cl$^-$ and NO$_3^-$ concentrations are observed at Zeppelin (coarse mode) reaching up to 3.0, 5.9 and 1.8 μg m$^{-3}$, respectively (Fig. 2c). Total SO$_4^{2-}$ does not exceed 0.8 μg m$^{-3}$ and might originate from metal smelting in Siberia (Hirdman et al., 2010). ss-SO$_4^{2-}$ contributes up to 0.8 μg m$^{-3}$ of the total SO$_4^{2-}$. Note that, in some cases, nss-SO$_4^{2-}$ has small negative concentrations, due to depletion of ss-SO$_4^{2-}$ through fractionation processes (Quinn et al., 2002).

Observed NH$_3^+$ does not exceed 0.5 μg m$^{-3}$ during the study period.

At Simeonof, an ice-free sub-Arctic island in south-western Alaska, high concentrations of fine mode Na$^+$ and Cl$^-$ are observed of up to 2.1 and 1.0 μg m$^{-3}$, respectively (Fig. 3a), especially at the beginning of January 2014, with low values of NO$_3^-$ (peaking at 0.25 μg m$^{-3}$). Total SO$_4^{2-}$ reaches 1.0 μg m$^{-3}$ and is mostly nss-SO$_4^{2-}$ (0.9 μg m$^{-3}$), while the contribution of ss-SO$_4^{2-}$ is smaller (up to 0.3 μg m$^{-3}$). Lower concentrations of fine mode Na$^+$ and Cl$^-$ (up to 0.35 μg m$^{-3}$) are observed at Gate of the Arctic (Fig. 3b), a non-coastal site located 404 km south east of Utqiaġvik in the Brooks Range Mountains, while NO$_3^-$ peaks at 0.45 μg m$^{-3}$. Total SO$_4^{2-}$ peaks at 0.64 μg m$^{-3}$ (G97 from now on). The G97 source function represents the rate that seawater droplets form per unit area (sea surface) and 0.56 μg m$^{-3}$ is nss-SO$_4^{2-}$ possibly due to local anthropogenic emissions originating from the North Slope of Alaska oilfields which may affect the measurements although this site is located inland (391 km) south of the oilfields. The contribution of ss-SO$_4^{2-}$ is insignificant (no more than 0.08 μg m$^{-3}$) at this site.

At Utqiaġvik, observed super-micron (1.0 < d$_{eq} < 10.0$ μm) per increase of particle radius. The fraction of Na$^+$ and Cl$^-$ concentrations reach 1.2 μg m$^{-3}$ (Fig. 4b), while NO$_3^-$ peaks at 0.2 μg m$^{-3}$. Super-micron SO$_4^{2-}$ and NH$_3^+$ do not exceed 0.16 and 0.009 μg m$^{-3}$, respectively. Super-micron NH$_3^+$ concentrations are insignificant (Quinn et al., 2002). However, there is more ss-SO$_4^{2-}$ (up to 0.18 μg m$^{-3}$) than nss-SO$_4^{2-}$. On the other hand, observed super-micron Na$^+$ is calculated using the molar weight of Na$^+$, Cl$^-$ and NO$_3^-$ at Utqiaġvik peak at 2.0, 2.2, and 0.9 μg m$^{-3}$ respectively (Fig. 4a). Note that based on the findings of KRP18, only 1%, by number, of the particles across the 0.15-1.0 μm and Cl$^-$ and then the fraction of Cl$^-$ is estimated, with the total being equal to 1. The G97 density function derived from the source function is based on laboratory experiments described in Monahan et al. (1986) (MO86 from now on):

$$\frac{dF}{dr} = 1.373 \times U_{10}^{3.41} \times r^{3}(1 + 0.057 \times r^{1.05}) \times 10^{1.19e^{-8}}$$

(1)

where $F$ is a function of $U$, the 10m-elevation wind speed, $r$, the particle radius at relative humidity (RH) equal to 80%, and $B = (0.380 - logr)_{0.45}$. The source function is applied for particles with dry diameters of 0.45 μm size range corresponded to fly-ash and dust, as compared to 50-90% from SSA across the same size range. This supports the assumption of Na$^+$ being primarily from SSA during this study. High sub-micron observed total SO$_4^{2-}$ (mostly nss-SO$_4^{2-}$) concentrations were measured at Utqiaġvik and peak at 2.4 μg m$^{-3}$, possibly due to local influence from Prudhoe Bay oil fields to the east (KRP18, KRP19), a magnitude much higher than super-micron SO$_4^{2-}$, also reported for Utqiaġvik by Quinn et al. (2002). Enhanced nss-SO$_4^{2-}$ during this period at Utqiaġvik could also be due to transport from mid-latitude sources, as well as due to transport and oxidation of SO$_2$ to SO$_4^{2-}$ near and within the Arctic region or more (equivalent to model particle diameters). For particles with dry diameters less than 0.45 μm, a correction is applied based on reported data in O’Dowd et al. (1997), since G97...
overestimates the production of small particles \citep{Gong2003,DeLeeuw2011}. G97 is based on the whitecap method, where the emission flux scales linearly with the fraction of the ocean area covered by whitecaps. Over open ocean, the whitecap fraction, \(W(U)\), is determined as a function of wind speed \citep{Barrie1984,Monahan1980} (MO80 from now on):

\[
W(U) = 3.84 \times 10^{-6} \times U_{10}^{3.11}
\]

(2)

This expression for \(W(U)\) is included implicitly in Equation (1) following details provided in MO80. In the base version, SSA emissions are calculated for every grid cell, which is open ocean or salt-water lakes. In this study, the grid cell which is covered by sea-ice is considered and then the fraction of that ice-free grid is used. In this way, SSA emissions from open leads are taken into account. It can be noted that this SSA scheme, based on \cite{Gong1997}, is still being used in global and regional models \cite[e.g.][]{Quinn2002}. Sub-micron \(\text{nss-SO}_{4}^{2-}\) peaks at 0.5 \(\mu\)g m\(^{-3}\). Observed \(\text{NH}_{4}^{+}\) is higher compared to the other remote Arctic sites (up to 0.34 \(\mu\)g m\(^{-3}\)) the Community Multiscale Air Quality Modeling System (CMAQ), \citep{Gantt2015}. Goddard Earth Observing System (GEOS)-Chem, \citep{Huang2017}, or in other models \cite[e.g.][]{Goddard2014}, also provide temporal variation during January and February follows that of \(\text{nss-SO}_{4}^{2-}\) due to \(\text{NH}_3\) reaction with acidic \(\text{SO}_{4}^{2-}\) aerosol near source regions outside of the Arctic \citep{Quinn2002} or due to regional sources of \(\text{NH}_3\), e.g. combustion of fossil fuels \citep{Whaley2018}.

Finally, only two sites provide total organic carbon (tOC) observations. Here, observed total organic carbon is assumed to include secondary organic aerosols, anthropogenic organic carbon emissions and marine organics. Thus, from now on it will be referred as tOC, to distinguish from OA and OM defined earlier. tOC ranges between 0.15 and 0.3 \(\mu\)g m\(^{-3}\) at Simeonof and 0.15 and 0.5 \(\mu\)g m\(^{-3}\) at Gates of the Arctic during January and February 2014.

Evaluation of the CONTROL simulation shows that the model overestimates observed fine/coarse mode (LOTUS-EUROS) \citep{Barthel2019} to simulate SSA, despite being relatively old. However, modelling studies have shown that G97 overestimates super-micron and TSP Na\(^+\) and Cl\(^-\) at most sites, and especially at Simeonof (by up to 15 \(\mu\)g m\(^{-3}\)), Zeppelin (Fig. 2c) (by up to 5.0 \(\mu\)g m\(^{-3}\)), Utqiaġvik (by up to 0.3 \(\mu\)g m\(^{-3}\)) and Gates of the Arctic (Fig. 3b) (by up to 4.0 \(\mu\)g m\(^{-3}\)) at each site. The CONTROL simulation also overestimates \(\text{NO}_{3}^{-}\) by up to 0.5 \(\mu\)g m\(^{-3}\) at each site. On the other hand, this simulation captures \(\text{NH}_{4}^{+}\) variability quite well at Alert, Villum and Utqiaġvik (super micron) \cite[see also biases and RMSEs (Root Mean Square Error) in APPENDIX C and Tables C1, C2 and C7 respectively.], whilst it overestimates \(\text{NH}_{4}^{+}\) at Zeppelin by up to 0.4 \(\mu\)g m\(^{-3}\). CONTROL includes only the \(\text{nss-SO}_{4}^{2-}\) component, however it captures observed variability of total \(\text{SO}_{4}^{2-}\) at Zeppelin (coarse mode), Villum (TSP) and Utqiaġvik (super-micron), but underestimates total \(\text{SO}_{4}^{2-}\) at Gates of the Arctic (fine mode) and Alert (coarse mode) by 0.5 and 0.2 \(\mu\)g m\(^{-3}\), respectively. In addition, the model underestimates SSA \cite[e.g.][]{Jaeglé2011}, JA11 from now on) or underestimates sub-micron Na\(^+\), Cl\(^-\), \(\text{SO}_{4}^{2-}\) and \(\text{NH}_{4}^{+}\) at Utqiaġvik. It also underestimates OA at the two sites compared to the measurements. In the following sections, model improvements SSA \cite[e.g.][]{Archer-Nicholls2014, Gantt2015}.

4.2 Updates to SSA emissions - HEM_NEW run
Here, updates to the model treatments of SSA emissions are described. Biases and RMSEs in $\mu$g m$^{-3}$, are given in APPENDIX C for all sites and available aerosol species at each location. They are included in the run HEM_NEW, which is also used as boundary conditions for the higher resolution runs over northern Alaska.

Evaluation of modelled aerosol composition (runs CONTROL and HEM_NEW) against in situ aerosol observations of fine mode ($d_a \leq 2.5 \mu m$) (both sites) at (a) Simeonof, Aleutians Islands, Alaska and (b) Gates of the Arctic, north of Alaska in local Alaskan time (AKST). The black line shows model results from the CONTROL run; the red line shows the HEM_NEW run, while observations are shown as blue crosses. Simeonof and Gates of the Arctic observations are 24h averages every three days and the corresponding model daily averages are shown as black diamonds for CONTROL and red pentagons for HEM_NEW. Observations are shown only when they are available. See the text for details about the observations and model runs.

### 4.3 Marine organics

Recent data analysis studies (Saliba et al., 2019; Kirpes et al., 2019) have suggested that marine organics associated with SSA contribute significantly to natural aerosol composition as ocean biomass can influence SSA number concentrations and diameter. In the CONTROL run, marine organics are not activated; however a source code is included in the model by Archer-Nicholls et al. (2014). For this reason, the parameterisation included in G97. However, Archer-Nicholls et al. (2014) implemented a scheme in the SSA emission module of MOSAIC based on Fuentes et al. (2010, 2011) (F10 and F11 from now on, respectively) is activated in the MOSAIC scheme to include a source flux for marine organics with dry diameters up from 0.003 to 0.45 $\mu m$, that is coupled to G97 for larger particles. This scheme is activated in HEM_NEW simulations. F10 is applied from the lower aerosol bin, namely 39 nm. The scheme is based on an analysis of data from a cruise in mid-latitudes mid-latitude cruise investigating the influence of dissolved organic matter OM on the production of sub-micron SSA. The F10 SSA source function also depends on MO80 whitecap coverage – in this study, organic and high wind speed dependence. Organic fractions equal to 0.2 for the first and second MOSAIC bins, 0.1 for the third bin and 0.01 for the remaining bins are used following the high biogenic activity scenario which assumes high C:Chlorophyll-a (Chl-a) ratios (see Lee et al. (2010)). F11 found that higher particle organic fractions are expected in algal bloom regions with high C:Chl-a ratios and Chl-a varying between 0.4-10 $\mu g L^{-1}$. The use of the F11 high biogenic activity option in our simulations is justified since MODIS-Aqua satellite data (https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MY1DMW_CHLORA&date=2014-12-01) for January-February 2014 show that Chl-a south of Alaska and along the west coast of the United States varied between 0.3 and 3.0 $\mu g L^{-1}$. Fujiki et al. (2009) also found that Chl-a varied between 0.4 and 1.0 $\mu g L^{-1}$ at six stations south of the Aleutian Islands, Alaska, during a sub-Arctic cruise in autumn 2005. Details about the F10 SSA source function are given in APPENDIX B. In the case of the model uses a source for marine organics, then OA is the sum of SOA, anthropogenic emissions of OM and Thus, in the HEM_NEW run, OA also includes marine organics.
A more detailed analysis of possible regional sources of marine organics focusing on northern Alaska, is presented in Section 6.

4.3 Whitecap method

In agreement with previous modelling studies, e.g. (Jaeglé et al., 2011), JA11 from now on and (Spada et al., 2013; Revell et al., 2019; Hartery et al., 2020), the CONTROL simulation produces too much coarse mode and TSP Na+ and Cl-

4.2.1 Wind speed dependence

As noted earlier, SSA emissions are highly dependent on wind speed and sea state (presence of whitecaps). The G97 parametrization, which depends on a scheme, and the related parameterisation by Gong (2003), depend on the whitecap method and thus have a high wind speed dependence (see Eq. 1). has been widely adopted to simulate SSA emissions in global and regional models, e.g. JA11 and Barthel et al. (2019). Several studies have tried to improve upon the whitecap method(W(U10)); especially for super-micron SSA. Callaghan et al. (2008) used an automated whitecap extraction technique to derive two whitecap expressions that differ from MO80, which are based on cubed relationships for U10. For sub-micron SSA, Ovadnevaite et al. (2012) showed that source functions, such as Gong (2003), based on the MO80 wind speed dependence, are responsible for an overestimation of the SSA emission flux. They found a lower wind speed dependence for small particles, based on an autumn field study off the west coast of Ireland. Other factors, such as the wave field (Salisbury et al., 2013) surfactant (amphiphilic organic material) activity (?) and or fetch-dependent threshold for breaking waves (Revell et al., 2019; Hartery et al., 2020), have also been shown to affect whitecap lifetime, with implications for SSA production. In a study by Goddijn-Murphy et al. (2011) Salisbury et al. (2014) analysed Marine Aerosol Production (MAP) whitecap data, in combination with analysis of in situ and satellite data, satellite data from Quick Scatterometer (QuikSCAT) for winds and waves. The satellite data (QuikSCAT) were used to derive an expression with a lower wind speed dependence compared to MO80(Salisbury et al., 2014), SALI14 from now on. Here, the SALI14 parametrization is implemented parameterisation is implemented, instead of the MO80 whitecap fraction expression since it is based on satellite data analysis providing information with global coverage including the Arctic (e.g. Chukchi Sea and Barents Sea during autumn) and south of Alaska:

\[
W(U) = 4.60 \times 10^{3.5} \times U_{10}^{2.26}
\]  

(3)

Based on Figure 2 in SALI14, the seasonal mean of W(U10) using Eq. 3 is lower at high latitudes latitudes above 40N and 40S compared to MO80 during autumn and winter. By using this more recent whitecap fraction expression in the quasi-hemispheric simulation, super-micron SSA concentrations decrease overall within the Arctic (not shown here). More specifically, super-micron Cl- and Na+ decrease more south of Alaska, by up to 20 µg m⁻³ (Aleutians Islands) and less north of Alaska, by up to 0.5 µg m⁻³. NO₃⁻ also decreases slightly over continental Alaska, by up to 0.5 µg m⁻³, due to increased heterogeneous formation on SSA.
Evaluation of modelled aerosol composition (runs CONTROL and HEM_NEW) against in-situ observations at Barrow Observatory, near Utqiagvik, Alaska for (a) super-micron and (b) sub-micron in UTC and in STP conditions. The black line shows model results from the CONTROL run; the red line shows the HEM_NEW run, while observations are shown as blue crosses. Sub-micron observations are daily averaged and super-micron observations are weekly averages. The corresponding model daily/weekly averages are shown as black diamonds for the CONTROL simulation and as red pentagons for the HEM_NEW. Observations are shown only when there are available. See the text for details about the observations and model runs.

4.3 SST-dependence

Recent data analysis studies pointed out that wind-

4.2.1 SST dependence

Wind speed alone cannot predict SSA variability, and it is important also to also include a dependence on SSTs for SSA prediction as pointed out by, for example, data-based studies in the Arctic (Saliba et al., 2019; Liu et al., 2021b). Recent modelling studies and mid-latitudes, such as Ovadnevaite et al. (2014). Modelling studies also showed that the application of a SST dependence improves simulated SSA concentrations compared to observations (Jaeglé et al., 2011; Sofiev et al., 2011; Spada et al., 2013; Barthel et al., 2019), but not implemented in WRF-Chem. More specifically, previous studies tested different SSA source functions, with and without SST dependence, and reported that including such a SST dependence improves model results, regardless of the SSA source function wind speed dependence employed (Spada et al., 2013; Grythe et al., 2014; Barthel et al., 2019). However, uncertainties still remain about the influence of SSTs on SSA production (Revell et al., 2019), including the role of other factors, such as seawater composition (Callaghan et al., 2014) or wave characteristics (e.g. wave speed and breaking wave type, Callaghan et al. (2012)), which might be more important than SSTs alone. In this study, the JA11 SST correction factor is applied when SSTs are between -2°C and 30°C to evaluate the effect of SST-SSTs on sub- and super-micron SSA emissions in the Arctic. In our simulations, SSTs are provided by the reanalyses data, in this case, FNL, and in the presence of sea-ice, SST is set equal to -1.75°C. In that case, the SST correction factor is set to the minimum value based on Barthel et al. (2019).

4.3 Sea-salt sulphate

Standard versions of the WRF-Chem model do not include-

4.2.1 Sea-salt sulphate

A source of ss-SO$_4^{2-}$ is included in the MOSAIC SSA emission scheme (HEM_NEW), since it was not included in the base model version (CONTROL). The mass fraction of ss-SO$_4^{2-}$ can be estimated to be $0.25 - 0.252$ of the Na$^+$ mass...
Figure 2. Average absolute differences in super-micron aerosol mass concentrations (in \( \mu g m^{-3} \)) between HEM_NEW and CONTROL during January and February 2014 at the surface. The black x in northern Alaska shows where Utqiaġvik is located. The black circle shows Alert, Canada, the black diamond shows Villum in Greenland, while the black pentagon shows Zeppelin, Svalbard. Total SO\(_4^{2-}\) is shown. All the results are shown north of 50N. Note the different scales.

Fraction \( + \) mass fraction based on (Kelly et al., 2010; Neumann et al., 2016) Kelly et al. (2010) and applied in WRF-Chem to calculate Neumann et al. (2016). The fraction of ss-SO\(_4^{2-}\) is subtracted from the fraction of Na\(^+\), Cl\(^-\), and marine OA. Note that the total fraction of Na\(^+\), Cl\(^-\), marine organics \( + \), Cl\(^-\), marine OA, and ss-SO\(_4^{2-}\) is equal to 1.0, and additional mass is emissions are not added. We find that, on average, the mass fraction of ss-SO\(_4^{2-}\) emissions in our simulations is around 9.9% of the total SSA emissions. This can be compared with the CMAQ model where the ss-SO\(_4^{2-}\) emissions are estimated to be 9.97% of the total SSA emissions in our simulations (Kelly et al., 2010).

4.3 Discussion

Average differences in
5 Evaluation of simulated wintertime SSA and other aerosols over the Arctic

First, absolute differences in simulated aerosol concentrations between the HEM_NEW and CONTROL simulations are shown in Figures 5 and 6 for results, averaged over January and February 2014, are presented. Model results from the two runs are then evaluated against available observations of, not only Na\(^+\) and Cl\(^-\), but also OA and SO\(_4^{2-}\) which now include a sea-salt component, and NO\(_3^-\) which is affected by heterogeneous reactions on SSA. We also show NH\(_4^+\) for completeness. Lastly, we compare observation-based and modelled contributions of SSA to total wintertime inorganic aerosol concentrations during winter 2014.

5.1 SSA emission updates: HEM_NEW versus CONTROL

Average absolute differences in super-micron and sub-micron aerosols inorganic aerosols between the HEM_NEW and CONTROL are shown in Figures 2 and 3, respectively. The updated model HEM_NEW simulates less super-micron Na\(^+\) by up to 20 \(\mu\)g \(\mu\)gm\(^{-3}\), and Cl\(^-\) by up to 30 \(\mu\)g \(\mu\)gm\(^{-3}\), especially south of Alaska and north of the Atlantic Ocean. This is due to the combined effect of using a lower wind speed dependence and including the SST dependence (Fig. 5). These decreases lead to an overall decrease (up to 2.5 \(\mu\)g \(\mu\)m\(^{-3}\)) in 2). Inclusion of a SST dependence leads to a larger

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**Figure 3.** The same as Figure 2, but for sub-micron aerosol mass concentrations.
decrease in locally produced super-micron $\text{NO}_3^{-}$ and $\text{Cl}^{-}$ over the Arctic and sub-Arctic ice-free regions, due to lower temperatures north of 50N, compared to using the lower wind speed dependence, based on SAL14, which has a smaller effect. Overall, one-third of the super-micron reductions can be attributed to the lower wind speed dependence and two-thirds to the SST dependence. Super-micron $\text{NO}_3^{-}$ over continental and coastal regions and the North Atlantic. This is in agreement with Chen et al. (2016) who examined the influence of SSA on $\text{NO}_3^{-}$ and reported that overestimation of SSA can lead to an overestimation of super-micron $\text{NO}_3^{-}$ due to is also lower (by up to 1.0 $\mu$gm$^{-3}$) due to less formation of $\text{NO}_3^{-}$ via heterogeneous uptake of nitric acid ($\text{HNO}_3$) $\text{HNO}_3$ on SSA. These reactions involving heterogeneous uptake of acid gases also produce HCl, thus depleting $\text{Cl}^{-}$ relative to $\text{Na}^{+}$ (Su et al., 2022). The presence of sea-ice also plays a role. Smaller decreases in $\text{Na}^{+}$ and $\text{Cl}^{-}$ are found north of Alaska (Beaufort Sea) compared to ice-free regions such as the northern Atlantic Ocean. The local influence of sea-ice fraction and open leads on SSA production over northern Alaska are examined further in Section 6. Furthermore, due to the addition of marine organics and $\text{ss-SO}_4^{2-}$ component in the model in HEM_NEW, there is more super-micron SO$_4^{2-}$, of by up to 2 $\mu$g m$^{-3}$ $\mu$gm$^{-3}$, and super-micron OA, by up to 0.6 $\mu$gm$^{-3}$, over marine regions. Super-micron NH$_4^{+}$ also increases (by up to 0.2 $\mu$g m$^{-3}$) in regions, such as Siberia and North of Europe, coinciding with decreases and increases in slightly increases up to 0.15 $\mu$gm$^{-3}$ over regions where $\text{NO}_3^{-}$ and $\text{SO}_4^{2-}$, respectively. Super-micron OA increases by up to 0.6 $\mu$g m$^{-3}$ due to the inclusion of marine organics. During winter, the Beaufort Sea, located north of Alaska is covered by sea-ice. Here, the implemented changes lead to increases.

There are smaller decreases in super-micron $\text{Na}^{+}$ and $\text{Cl}^{-}$ compared to ice-free regions such as the Aleutians islands, e.g., Simeonof site (Fig. 2a) further south. The local effect of sea-ice fraction and open leads on SSA production is examined further in 5.4.

Average differences in super-micron aerosol mass concentrations ($\mu$ g m$^{-3}$) at the surface between HEM_NEW and CONTROL during January and February 2014. The black star in northern Alaska shows where Utqiaġvik is located. The black circle shows Alert, Canada, the black diamond shows Villum in Greenland, while the black pentagon shows Zeppelin, Svalbard. Average differences in sub-micron aerosol mass concentrations ($\mu$ g m$^{-3}$) and at the surface between HEM_NEW and CONTROL during January and February 2014. The black star in northern Alaska shows where Utqiaġvik is located. The black circle shows Alert, Canada, the black diamond shows Villum in Greenland, while the black pentagon shows Zeppelin, Svalbard.

On the other hand, the effect of model updates on sub-micron $\text{Na}^{+}$ is smaller, with decreases of HEM_NEW sub-micron $\text{Na}^{+}$ compared to CONTROL, by up to 0.25 $\mu$g m$^{-3}$ $\mu$gm$^{-3}$, south of Alaska and in the North Atlantic (Fig. 6) due to use of 3. Again, this is due primarily to the introduction of the SST dependence. When using SALI14 lower wind speed dependence (SALI14 instead of MOS80). The lifetime of SSA, estimated to be between 1 to 4 days over open ocean, in the Arctic and during wintertime (Rhodes et al., 2017; Xu et al., 2016; Huang and Jaeglé, 2017; Hoppel et al., 2002), could explain the small decrease of alone, there is a small decrease in sub-micron CI$^{-}$ over continental coastal areas (e.g. south of Alaska) in HEM_NEW. This could also affect long-range transport of $\text{Na}^{+}$ and a small increase in sub-micron SSA from oceanic regions leading to $\text{Na}^{+}$ over the Arctic. Sub-micron CI$^{-}$ also decreases over continental regions, such as northeast United States of America (USA) and Siberia. Sub-micron OA increases by up to 1.5 $\mu$g m$^{-3}$ due to inclusion of the F10 parametrization. Note that including $\text{ss-SO}_4^{2-}$ leads to a decrease in $\text{Na}^{+}$ and CI$^{-}$ fractions per bin since no additional mass is added. In contrast to
super-micron NO$_3^-$ areas, where NO$_3^-$ and HNO$_3$ are higher due to anthropogenic sources (Fig. 3). Heterogeneous uptake on SSA reduces Cl$^-$ and increases sub-micron NO$_3^-$ increases by 2.5 by up to 6.0 $\mu$gm$^{-3}$ over sources regions and total SO$_4^{2-}$ increases due to ss-SO$_4^{2-}$ component. Also, sub-micron NH$_4^+$ slightly increases, showing similar patterns to sub-micron in HEM_NEW over continental regions while the increases over the Arctic Ocean are smaller. This is in contrast to super-micron NO$_3^-$ and SO$_4^{2-}$, probably due to a potential shift in the balance between (NH$_4^+$) decreases. These results are consistent with the study of Chen et al. (2016), also using WRF-Chem with MOSAIC, who noted that since SSA are primarily present in the coarse (super-micron) mode, this favours the formation of NaNO$_3$ and SO$_4$ and which is thermodynamically stable, and limits the formation of NH$_4$NO$_3$.

To investigate the variations in modelled NO$_3^-$, SO$_4^{2-}$ and NH$_4^+$, the mean neutralized factor ($f$) is calculated (not shown here) as the ratio of NH$_4^+$ to the sum of (2SO$_4^{2-}$+NO$_3^-$), in molar concentrations, following, for example Fisher et al. (2011). For sites in the Arctic with available observations of these aerosols. When $f$ is equal to 1 aerosols are more neutralized, while when $f$ < 1 then aerosols are acidic, and more acidic when $f$ is closer to 0 (Fisher et al., 2011). At all sites, except Zeppelin, higher molar concentrations were observed for SO$_4^{2-}$ compared to NO$_3^-$ and NH$_4^+$. At Utqiaġvik, the average observed value of $f$ is equal to 0.15 for which is semi-volatile (Chen et al., 2020). Therefore, lower super-micron aerosols, whilst in the model $f$ decreases from 0.7 to 0.66. This implies that observed SSA in HEM_NEW, results in less super-micron aerosols are more acidic while in the model they are more neutralized (Fisher et al., 2011), probably because modelled NH$_4^+$ decreased more than SO$_4^{2-}$ and NO$_3^-$ (Fig. 4a) between the two simulations. There is less super-micron NH$_4^+$ in the model than the sum 2SO$_4^{2-}$+NO$_3^-$, as in observations, however observed 2SO$_4^{2-}$+ and more super-micron NO$_3^-$ is much higher than modelled. Observed super-micron aerosols at Utqiaġvik are more acidic compared to.. We also note that, for these reasons, sub-micron aerosols for which $f$ equals 0.34. For sub-micron aerosols, HEM_NEW has an average $f$ value of 0.08 compared to 0.01 in the CONTROL run. The increase in modelled sub-micron $f$ could be due to the bigger increase in modelled NH$_4$NO$_3$ between the two simulations (Fig. 4b). However, in the observations, the higher also increases, by up to 1.5 $\mu$gm$^{-3}$, especially over continental areas, and displays similar regional patterns to sub-micron $f$ is because the sum 2SO$_4^{2-}$+NO$_3^-$ is much higher than NH$_4^+$. At Alert (coarse mode), model $f$ increases from 0.14 (CONTROL) to 0.19 (HEM_NEW), with observed $f$ equal to 0.21, implying that model and observations are acidic, in contrast to Utqiaġvik modelled super-micron aerosols. Similar values of $f$ are found for Zeppelin (coarse mode) and Villum (TSP)(0.12 for CONTROL, 0.13 and 0.18 for HEM_NEW, respectively) with observed aerosols (0.34 and 0.36 respectively) being less acidic at these sites. Overall the model inorganic aerosols are mostly too acidic compared to the observations. This could be due to underestimation of anthropogenic sources of NH$_4$ on the above sites, originating from mid-latitudes. It can be noted that in the model is assumed that all of the aerosol species are internally mixed. However, in reality some of the NO$_3^-$ and... Inclusion of marine organics linked to SSA, leads to increases in sub-micron OA, by up to 1.5 $\mu$gm$^{-3}$, and total SO$_4^{2-}$ are observed to be mixed with SSA (KRP18). Based on that, the calculated $f$ for observations would be biased (too acidic), as some of the NO$_3^-$ and SO$_4$ increases due to the addition of ss-SO$_4^{2-}$ are present as Na$_2$SO$_4$ and NaNO$_3$.

Figures 2.
Table 2. Biases, in $\mu$gm$^{-3}$, averaged over January and February 2014 for the CONTROL and HEM_NEW simulations compared to the observations. NA stands for not available.

<table>
<thead>
<tr>
<th></th>
<th>CONTROL</th>
<th>HEM_NEW</th>
<th>CONTROL</th>
<th>HEM_NEW</th>
<th>CONTROL</th>
<th>HEM_NEW</th>
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<tr>
<td></td>
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<td>Cl$^-$</td>
<td>NO$_3^-$</td>
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<td>nss-SO$<em>{4}^{2-}$/ss-SO$</em>{4}^{2-}$</td>
<td>NH$_4^+$</td>
<td>OA</td>
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<td></td>
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<td>0.25</td>
<td>0.06</td>
<td>-0.02/0.04</td>
<td>0.011</td>
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<td>0.27</td>
<td>0.25</td>
<td>0.14</td>
<td>0.05</td>
<td>0.04/0.07</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
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<td>4.9</td>
<td>0.1</td>
<td>0.13</td>
<td>0.2</td>
<td>0.05</td>
<td>0.24/0.12</td>
<td>0.01</td>
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</tr>
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<td>-0.26</td>
<td>0.26</td>
<td>0.13</td>
<td>0.005</td>
<td>0.006/-0.02</td>
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<td>-0.065</td>
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<td>-0.2</td>
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</tr>
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</table>

5.2 Evaluation against observations

Model results are evaluated against available observations of aerosols at different sites as shown in Figures 4, 5, and 3, showing the effect of all the modifications (HEM_NEW) compared to CONTROL and the observations at four Arctic and one 6. These figures are grouped according to the size ranges of the measurements at the different sites as discussed in Section 3.1. Mean biases and root mean square errors (RMSEs) between the observations and the model results are given in Table 2 and Table C.1 (APPENDIX C), respectively. In the following the main findings are discussed by aerosol component.

SSA (Na$^+$ and Cl$^-$): Updates to the treatment of SSA emissions in HEM_NEW greatly improves modelled SSA over the Arctic with notable reductions in biases and RMSEs in Na$^+$ and Cl$^-$ compared to observations at Alert, Zeppelin ($d_\text{a} < 10$ µm), Villum (TSP), Gates of the Arctic (GoA) (fine mode), and the sub-Arctic sites. At the two remote high-Arctic sites surrounded by sea ice (Alert and Villum, Figure 2a,b) site, Simeonof (fine mode). Overall, HEM_NEW captures better Na$^+$ and Cl$^-$ variability, with a small overestimation at Villum (maximum 0.2 $\mu$ g m$^{-3}$). Biases, in $\mu$ g m$^{-3}$, at Alert for Na$^+$ and Cl$^-$ decrease from 0.81 to 0.12 and from 1.05 to -0.03, respectively. Model results also improve at Villum for Na$^+$ and Cl$^-$ with biases reduced from 1.3 to 0.25 and from 1.9 to 0.22 $\mu$ g m$^{-3}$, respectively. The spatial variability between the observed Na$^+$ and Cl$^-$ at the different sites, in particular, the lower observed concentrations at Villum, which is surrounded by sea-ice at this time of year, and higher concentrations at Simeonof and Zeppelin. The extent to which sea-ice is present near different sites is an important factor. For example, the high variability in modelled SSA at Villum at the end of January and the middle of February 2014 is likely to be due to fluctuations in sea-ice fraction around the site, as seen (0.9-1.0) in the FNL sea-ice reanalysis product (varies between 0.93 and 1.0 fully covered, in January and February). Also, analyses). At Utqiagvik, the model captures super-micron Na$^+$, whereas Cl$^-$ is now underestimated due to Cl$^-$ depletion.
Sub-micron Na\(^+\) and Cl\(^-\) are still underestimated in HEM_NEW captures better NO\(_3\)^- and NH\(_4\)^+ at Alert while slightly overestimates total SO\(_2\)^- (see APPENDIX C, with average biases of about -0.5 \(\mu\)g\(^m\(^{-3}\) for Na\(^+\) and -0.12 \(\mu\)g\(^m\(^{-3}\) for Cl\(^-\) with higher biases during episodes with elevated observed SSA. Sub-micron SSA at this site may have been transported to the Arctic from the Pacific Ocean (Quinn et al., 2002; May et al., 2016), and thus model underestimations may point to deficiencies in the SSA source function further south or issues related to long-range transport or to wet and dry deposition treatments in the model. However, the fact that the model agrees better with observations over the wider Arctic, as well as at sub-Arctic Simeonof, provides confidence in the modelled long-range transport as a source of Arctic (sub-micron) SSA. Simulated SSA also compares well with reported weekly averaged sub-micron Na\(^+\) mass concentrations collected during January and February 2014 at Alert (0.1 \(\mu\)g\(^m\(^{-3}\) observed, up to 0.08 \(\mu\)g\(^m\(^{-3}\) modelled) (Leaitch et al., 2018). We also note that, at Utqiagvik, while May et al. (2016) attributed sub-micron SSA to long-range transport, KRP18 estimated that 42% of their analysed samples in the sub-micron range were fresh SSA with chemical signatures similar to sea-water, with 18% classed as partially aged with enhanced anthropogenic components (S, N) and depleted Cl\(^-\), and the remainder included organics and sulphate particles. Thus, model discrepancies may also be due to local processes influencing SSA over northern Alaska. This is investigated further in Section 6.

**Nitrate:** Improved SSA treatments in HEM_NEW also lead to improved simulation of NO\(_3\)^- at some sites, notably Simeonof, GoA, Alert, Villum and biases are reduced (see Table C1). At Villum, HEM_NEW captures better SO\(_2\)^- compared to CONTROL run, slightly underestimates NH\(_4\)^+ end of February, but still overestimates NO\(_3\)^- (see APPENDIX C, Table C2). Similar results are found for Zeppelin where HEM_NEW simulates better Na\(^+\), Cl\(^-\), 2). While modelled super-micron NO\(_3\)^- and NH\(_4\)^+ but overestimates SO\(_2\)^-. More specifically, HEM_NEW slightly underestimates observed Na\(^+\), Cl\(^-\) and NH\(_4\)^+, but the model results improve in this site. SSA updates also improve modelled at Utqiagvik is improved, the model still underestimates certain periods when elevated sub-micron NO\(_3\)^- (see APPENDIX C, Table C3).

At Simeonof, HEM_NEW captures better Na\(^+\), Cl\(^-\) and is observed, also the case at GoA and Simeonof. The improved behaviour of modelled NO\(_3\)^- variability during winter 2014 and, due to the inclusion of marine organics, the model simulates more tOC, although it still underestimates observed variability. Calculated biases decrease from 1.4 to 0.3, 2.0 to 0.1, 0.12 to 0.08, 0.08 to 0.05 \(\mu\)g \(m\(^{-3}\) for Na\(^+\), Cl\(^-\), is, in general, due to reductions in Na\(^+\) and Cl\(^-\), leading to less NO\(_3\)^- production in the coarse mode, especially close to or just downwind of major anthropogenic emission regions at mid-latitudes, and a shift to more NO\(_3\)^- and tOC, respectively. Also, the addition of in the fine mode, as discussed previously. These effects are most evident at Utqiagvik, where the model can be compared to sub- and super-micron data. Comparison with data from other sites is with either total, coarse plus fine mode, or fine mode aerosol observations, and therefore includes both increases and decreases in simulated NO\(_3\)^-. Overall, these results illustrate the importance of correctly simulating SSA and its effects on anthropogenic aerosols. While observed NO\(_3\)^- concentrations are generally lower than other aerosol components, such as Na\(^+\), Cl\(^-\) or nss-SO\(_2\)^-, during Arctic winter, a recent trend analysis study showed that NO\(_3\)^- is clearly increasing at Alert, especially during the winter months (Schmale et al., 2022). Such increases in NO\(_3\) may be due to increased NO\(_3\)^- formation due to lower acidity following SO\(_2\) reductions, that outweigh reductions in NO\(_x\) emissions at mid-latitudes (Sharma et al., 2019).
However, increases in SSA over the Arctic Ocean, due to reductions in ice-covered waters, may also explain these changes (e.g. Browse et al. (2014)) although no significant trends in Na⁺ have yet been detected (Schmale et al., 2022).

**Sulphate**: Figures 4, 5 and 6 show observed ss-SO$_4^{2-}$ in and total-SO$_4^{2-}$ together with results from CONTROL and HEM_NEW leads to improvements (biases, RMSEs) in. With regard to total simulated SO$_4^{2-}$ even if the model occasionally underestimates by up to 0.6 µg m$^{-3}$. Similar patterns are found for the Gates of the Arctic in northern Alaska. Na⁺ and Cl⁻ are lower in the addition of ss-SO$_4^{2-}$ improves the model results, for example, at Simeonof where observed fine mode ss-SO$_4^{2-}$ makes a significant contribution (30-80%, up to 0.3 µgm$^{-3}$) to total SO$_4^{2-}$. ss-SO$_4^{2-}$ also contributes between 10-40% of total SO$_4^{2-}$ at Alert and Villum and modelled ss-SO$_4^{2-}$ agrees better with the observations. The remainder is nss-SO$_4^{2-}$, a dominant component of Arctic Haze resulting from long-range transport from sources in Russia and Europe at these sites (Leaitch et al., 2018; Lange et al., 2018). Model results are at the lower end (up to 0.3 µgm$^{-3}$) of reported sub-micron nss-SO$_4^{2-}$.
Figure 5. Evaluation of modelled aerosol composition against observations at Utqiagvik for (a) super-micron and (b) sub-micron in UTC and in STP conditions. Observations are shown only when available. The lines and the symbols are the same as in Figure 4. See the text for more details. Note the different scales.

mass concentrations (0.3-1.1 \(\mu\)gm\(^{-3}\)) at Alert during winter 2014 (Leaitch et al., 2018). On the other hand, HEM_NEW while modelled NO\(_3^-\) and tOC also improve, with biases decreasing for all the four species (0.56 to 0.16, 0.70 further overestimates total observed SO\(_2^-\) at Zeppelin due to the inclusion of ss-SO\(_2^-\) especially during certain episodes with elevated concentrations. We note that Zeppelin is a mountain site at 471m, and thus discrepancies with the observations may also be due to issues simulating the vertical distribution and transport of nss-SO\(_2^-\) from Eurasian source regions (Hirdman et al., 2010). At Utqiagvik, on the northern coast of Alaska, most of total observed super-micron \(\text{SO}_4^{2-}\) is ss-SO\(_2^-\) (up to 0.09, 0.26 to 0.18 \(\mu\)gm\(^{-3}\), around 80%), and 0.24 to 0.21 \(\mu\)g m\(^{-3}\) for Na\(^+\), Cl\(^-\), NO\(_3^-\) and tOC, respectively, see also APPENDIX C for RMSEs). The inclusion of ss-SO\(_2^-\) in HEM_NEW simulates more total improves agreement with the observations. With regard to total sub-micron \(\text{SO}_4^{2-}\) at this site but still underestimates the observations, in particular, high mass concentrations are observed at Utqiagvik compared to other Arctic sites, consisting mostly of nss-SO\(_2^-\). Here, the contribution of \(\text{SO}_4^{2-}\), peaking at 2.4 \(\mu\)gm\(^{-3}\), much higher than total super-micron \(\text{SO}_4^{2-}\) (peaking at 0.5 \(\mu\)gm\(^{-3}\)), as also reported by Quinn et al. (2002). However, the model underestimates nss-SO\(_4^{2-}\) at this site. As noted by KRP18 and KRP19, this is likely to be due to the local influence from the North Slope of Alaska (NSA) oil fields to the east. In a companion paper, Ioannidis et al. (2022, in prep.) the influence of
These regional emissions on BC at Barrow is investigated during winter 2014 finding that up to 30-50% of BC may originate from this source. Indeed, at GoA, 391km inland from the coast, and south of the NSA oil fields, the contribution of nss-SO$_4^{2-}$ is more important and ss-SO$_4^{2-}$ is minimal, as shown in Fig. 3b. Thus, the model underestimation could negligible. The model captures total fine mode SO$_4^{2-}$ (peaking up to 0.64 μgm$^{-3}$) at this site and the addition of ss-SO$_4^{2-}$ does not affect the results. As well as local sources, model difficulties simulating sub-micron nss-SO$_4^{2-}$ at Utqiagvik may be due to issues related to long-range transport of nss SO$_4^{2-}$, such as wet deposition, or to missing local anthropogenic sources (e.g., Prudhoe Bay oilfields). Additional wintertime production of underestimation in transport of mid-latitude sources, SO$_4^{2-}$ via mechanisms not requiring sunlight may also contribute formation mechanisms under dark, cold winter conditions may also be lacking in the model. For example, high concentrations of hydroxymethane sulphonate (HMS) have been measured recently during winter in Fairbanks, Alaskan Interior. McCabe et al. (2006) (Campbell et al., 2022) suggested that there is contributing to secondary SO$_4^{2-}$ at Alert during wintertime from metal catalyzed production during Arctic winter, although only a small contribution from HMS to SO$_4^{2-}$ was found in observations at Oliktok Point, situated within NSA oil fields (Liu et al., 2021a). Oxidation of SO$_2$ oxidation of S(IV) (10–18%). Results from HEM_NEW also underestimate tOC at Gates of the Arctic, possibly due to underestimation of marine organics (see discussion in next section) or missing regional or remote sources by ozone in alkaline SSA could also contribute up to 9% to SO$_4^{2-}$ formation (Alexander et al., 2005). However, the version of WRF-Chem used here does not include such reactions, in common with many chemistry-aerosol models run over the Arctic (Whaley et al., 2022).

Figure 4 compares results from CONTROL and HEM_NEW with observations for Ammonium: NH$_4^{+}$ observations are available at all sites except for Simeonof and GoA. Observed NH$_4^{+}$ concentrations are very low (below 0.2 μgm$^{-3}$) at Alert and Villum, with higher concentrations observed at Zeppelin. Overall there is a good agreement between the model and measurements, with very low biases and RMSEs in both runs, apart from an underestimation of elevated NH$_4^{+}$ at Zeppelin. At Utqiagvik, there is good agreement with super-micron (weekly averages) and sub-micron (daily averages) aerosols at Utqiagvik. While CONTROL overestimates SSA and NO$_3^{-}$ and underestimates SO$_4^{2-}$ (only NH$_4^{+}$ except for periods with higher observed NH$_4^{+}$ (up to 0.1 μgm$^{-3}$). However, the model underestimates periods with elevated sub-micron NH$_4^{+}$, of up to 0.4 μgm$^{-3}$, which is higher compared to the other sites. Temporal variations in NH$_4^{+}$ during January and February 2014, generally, follow nss-SO$_4^{2-}$, in general, HEM_NEW captures better observed super-micron Na$^+$, Cl$^-$, NO$_3^{-}$ and NH$_4^{+}$ aerosols during the simulation period (Fig. 4a)(see also Appendix C). The Na$^+$ bias decreases from 0.3 to 0.07 μg·m$^{-3}$ but Cl$^-$ is now underestimated (bias decreases from 0.27 to 0.26 μg·m$^{-3}$), due to the introduction of the SST dependence (not shown). Also, there is more super-micron as NH$_4^{+}$ preferentially forms ammonium bisulfate and, to a lesser extent, ammonium sulfate in the particle phase (Schmale et al., 2022), and they have common anthropogenic origins. Previous studies also noted that NH$_4^{+}$ is two times higher at Utqiagvik than at Alert, Zeppelin and Villum, while SO$_4^{2-}$ in concentrations are similar at all three stations (Schmale et al., 2022), possibly suggesting differences in aerosol acidity at different sites. This is also found in this study based on the observations and modelled results (HEM_NEW and the model slightly underestimates observed SO$_4^{2-}$ by about 0.1 μg·m$^{-3}$. Super-micron OA is smaller in magnitude compared to the other aerosol components). It is therefore interesting to investigate the effect of the improved SSA emissions on modelled aerosol acidity. For this, we estimate
Figure 6. Evaluation of modelled aerosol composition against in-situ aerosol observations with \( d_a < 10 \mu m \) (a) at Alert, Canada (standard temperature pressure (STP) conditions), (b) at Zeppelin, Svalbard and (c) TSP aerosols at Villum, Greenland in UTC. Observations are shown only when available. The lines and the symbols are the same as in Figure 4. See the text for more details. Note the different scales.
the neutralisation factor \( f \), following *Fisher et al. (2011)*. The results are discussed in APPENDIX D. CONTROL tends to predict more acidic aerosols than observed. Based on the observations, most acidic aerosols are found at Alert, Zeppelin and Utqiagvik (super-micron), with somewhat less acidic aerosols at Villum and Utqiagvik (sub-micron). This is improved to some degree in HEM_NEW with aerosols becoming less acidic at some sites, notably at Alert and Villum, due to decreases in simulated \( \text{NO}_3^- \). However, super micron OA mass concentration measurements are not available in winter 2014 to evaluate the modelled sub-micron aerosols at Utqiagvik are less acidic than the observations, due to the underestimation of nss-\( \text{SO}_4^{2-} \). Overall, modelled super-micron SSA concentrations decrease the updates to SSA emissions lead to somewhat less acidic anthropogenic aerosols over the Arctic, again highlighting the importance of interactions between SSA and other inorganic aerosols.

**Organic aerosols:** Only two sites provide TOC fine-mode observations ranging from between 0.15-0.3 \( \mu \text{g m}^{-3} \) at Simeonof and 0.15 and 0.5 \( \mu \text{g m}^{-3} \) at GoA during January and February 2014. The inclusion of marine organics in HEM_NEW at other remote sites (Fig. 2 and 3) in better agreement with the observations compared to the CONTROL run improves modelled OA, especially at the coastal Simeonof site. Since observations at other sites are not available for winter 2014, results are compared with other reported measurements. *Shaw et al. (2010)* reported sub-micron OA at Utqiagvik equal to 0.3 \( \mu \text{g m}^{-3} \) during winter 2008 (November to February). However, a more recent study by *Moschos et al. (2022a)* reported lower wintertime OA concentrations (\( d_a < 10 \mu \text{m} \)) at this site (around 0.1 \( \mu \text{g m}^{-3} \)), attributed mostly to primary-anthropogenic or haze OA originating from Eurasia. However, modelled OA for the same size range is only up to 0.05 \( \mu \text{g m}^{-3} \). At Villum, *Nielsen et al. (2019)* also reported higher sub-micron OA observations, peaking at 2.2 \( \mu \text{g m}^{-3} \) in February 2015, attributed mostly to Arctic Haze influence (up to 1.1 \( \mu \text{g m}^{-3} \)) with secondary influences from hydrocarbon-like organics (up to 1.0 \( \mu \text{g m}^{-3} \)) and marine sources (up to 0.2 \( \mu \text{g m}^{-3} \)). Modelled OA in HEM_NEW at this site does not exceed 0.1 \( \mu \text{g m}^{-3} \). Overall, the model underestimates Arctic OA in common with many other models (*Whaley et al., 2022*). These discrepancies may be due missing or underestimated anthropogenic or natural sources. For example, it is known that there are large uncertainties in anthropogenic OA emissions (*Marelle et al., 2017*). The possibility of a wintertime marine OA source over northern Alaska is explored further in Section 6.

On the other hand, while HEM_NEW (Fig. 4b) represents better periods with low concentrations of sub-micron \( \text{Na}^+ \) and \( \text{Cl}^- \) at Utqiagvik in January and February 2014 (up to 0.3 \( \mu \text{g m}^{-3} \)), it still underestimates episodes with very high observed \( \text{Na}^+ \) and \( \text{Cl}^- \), especially at the end of February 2014. The model simulates better \( \text{NO}_3^- \) but underestimates \( \text{NH}_4^+ \) and \( \text{SO}_4^{2-} \), especially at the beginning of January and end of February 2014. Sub-micron OA at 100 km ranges between 0.01 and 0.15 \( \mu \text{g m}^{-3} \). However, observations of OA at Utqiagvik during this period are not available with which to validate the model. *Barrett et al. (2015)* and *Barrett and Sheesley (2017)* showed that OC at Utqiagvik is influenced by primary and secondary biogenic carbon and fossil fuel carbon, with air masses originating from the Arctic Ocean, Russian and Canadian Arctic. More specifically, *Barrett and Sheesley (2017)* made measurements of OC (diameter less than 10 \( \mu \text{m} \)) collected during winter 2012-2013 northeast of Utqiagvik and reported average OC of 0.22 \( \mu \text{g m}^{-3} \). To compare directly with the model results we divide the modeled value by 1.4 (Fig. 5). In that case, modelled super-micron tOC at Utqiagvik is three times less than the observed OC, showing that the model lacks sources of OC. *Shaw et al. (2010)* reported sub-micron OM equal to 0.43 \( \mu \text{g m}^{-3} \).
**Table 3.** Calculated fractions of observed and modelled (HEM_NEW) SSA to total inorganic aerosol mass concentrations (summed from available observations at each site). For each site, SSA are defined as the sum of Na\(^{+}\), Cl\(^{-}\) and ss-SO\(_4^{2-}\). Total is defined as the sum of SSA and inorganic aerosols. Inorganic is the sum of nss-SO\(_4^{2-}\), NH\(_4^{+}\) and NO\(_3^{-}\) for each station except for Simeonof and Gates of the Arctic GoA where inorganic is the sum of nss-SO\(_4^{2-}\) and NO\(_3^{-}\). Note that NH\(_4^{+}\) is rarely internally mixed within SSA aerosol, because most NO\(_3^{-}\) and SO\(_4^{2-}\) forms via Cl\(^{-}\) (e.g. NaCl + HNO\(_3\) → NaNO\(_3\) + HCl). Total\(_{all}\) below is defined as the sum of SSA, nss-SO\(_4^{2-}\), NH\(_4^{+}\), NO\(_3^{-}\), BC, OA and dust (model only). The aerosol size for SSA, Total and Total\(_{all}\) varies per station and corresponds to observed aerosol sizes as described in Section 3.

<table>
<thead>
<tr>
<th>Sites</th>
<th>SSA/Total [obs]</th>
<th>SSA/Total [HEM_NEW]</th>
<th>SSA/Total(_{all}) [HEM_NEW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simeonof (fine mode)</td>
<td>0.73</td>
<td>0.84</td>
<td>0.74</td>
</tr>
<tr>
<td>Gates of the Arctic GoA (fine mode)</td>
<td>0.20</td>
<td>0.44</td>
<td>0.33</td>
</tr>
<tr>
<td>Utqiaγvik-sub-micron</td>
<td>0.60</td>
<td>0.22</td>
<td>0.13</td>
</tr>
<tr>
<td>Utqiaγvik-super-micron</td>
<td>0.93</td>
<td>0.57</td>
<td>0.54</td>
</tr>
<tr>
<td>Alert (coarse mode)</td>
<td>0.59</td>
<td>0.54</td>
<td>0.45</td>
</tr>
<tr>
<td>Villum (TSP)</td>
<td>0.32</td>
<td>0.63</td>
<td>0.52</td>
</tr>
<tr>
<td>Zeppelin (coarse mode)</td>
<td>0.56</td>
<td>0.75</td>
<td>0.62</td>
</tr>
</tbody>
</table>

during winter 2008 (November to February) at Utqiaγvik, almost double the simulated OA at Utqiaγvik. Their analysis showed that OM was correlated with organic and inorganic seawater components with the air masses originating along the coastal regions of the Northwest Territories of Canada. Also, the model results can be compared with weekly average sub-micron OM data collected at Alert (Leaitch et al., 2018) and Fig. 2. At Alert, OM reaches up to 0.25 µ g m\(^{-3}\) in February 2014, which is almost double compared to the model results for Utqiaγvik, North Slope of Alaska and Alert (Fig. 6). At Villum, a recent study by Nielsen et al. (2019) showed that OA peaks at 2.2 µ g m\(^{-3}\) at the beginning (21 to 28 February 2015) of their study period. Their study shows that the majority of OA is mostly due to Arctic Haze influence (up to 1.1 µ g m\(^{-3}\)) with secondary influence, due to hydrocarbon-like organics (up to 1.0 µ g m\(^{-3}\)) and a marine influence (up to 0.2 µ g m\(^{-3}\)). Reasons for these differences on modelled and observed OA are investigated in the next section focusing on regional processes affecting SSA near northern Alaska.

Previous studies pointed out that SSA are an important contributor in the-

### 5.3 Contribution of SSA to total inorganic aerosols

Lastly, we assess the contribution of SSA to total inorganic aerosols in the Arctic during wintertime since previous studies noted that they can make an important contribution to total sub-micron and super-micron mass fraction in the Arctic during wintertime. A recent study by fractions at this time of year (Quinn et al., 2002; May et al., 2016; Kirpes et al., 2018, 2019). Moschos et al. (2022b) showed that during wintertime SSA dominates also showed SSA dominates wintertime PM\(_{10}\) (particulate matter with diameter \(d_{10} \leq 10 \mu m\)) mass concentrations at remote Arctic sites, including Alert (56%), Baranova (41%) (Russia), Gruvebadet (44(Norway)), Utqiaγvik (66%), Villum (32%), and Zeppelin (65%). In contrast, at sites such as Tiksi
(northern Russia) and Pallas (Finland), SO$_4^{2-}$ and OA dominate (70% and 55%, respectively). To investigate the contribution of SSA to total mass concentrations during the period of this study, the observed and modelled fraction fractions of SSA to "total" (SSA plus inorganic) aerosols are estimated (see Table 2). However, it 3. It should be noted that this fraction varies between sites since not all components were measured.

Overall, taking into account the observations available at each site, the fraction of SSA to total SSA +plus inorganics is higher at all the coastal sites (Utqiagvik, Alert, Simeonof, Villum) and Zeppelin ranging from 54 to 93%. Only at the Gates of the Arctic and Villum stations GoA and Villum is the fraction of SSA is smaller (20% and 32%). The modelled, respectively, SSA fractions, calculated using the HEM_NEW SSA fraction shows similar patterns (fraction ranges results, show similar patterns compared to the observations with fractions ranging between 44% and 84%) compared to the observations. An exception is sub-micron modelled SSA at, is Utqiagvik where the modelled fraction is lower than in the observations due to low modelled simulated sub-micron SSA concentrations. When taking into account all aerosol components in the model, including OA, BC and dust, SSA is dominant at Simeonof, Utqiagvik (super-micron), Zeppelin and Villum (more than 54%), whereas at Alert, SSA contributes about 45%. This analysis shows that SSA is an important fraction of total fine-mode, super-micron, coarse mode and TSP aerosols in the inorganic aerosols at most Arctic coastal sites during wintertime.

6 Regional processes influencing SSA over northern Alaska

In this section, processes which could affect SSA emissions on a regional scale over northern Alaska are examined. In general, the improved model simulates better observed super-micron, TSP, fine and coarse mode SSA, NO$_3^-$ and SO$_4^{2-}$. Overall, the results presented here show that the simulation of Arctic SSA, and other inorganic and organic aerosols, is improved as a result of the updated SSA emission treatments. In particular, simulated aerosols, including the coarse mode or super-micron fraction, are improved compared to the observations. The results also show that it is important to include natural SSA emissions of ss-SO$_4^{2-}$ aerosols at different sites in the Arctic but the model has difficulties capturing sub-micron SSA during wintertime at Utqiagvik. Possible reasons for these discrepancies are investigated in model runs at 20 km resolution during the campaign periods in January and February 2014 with boundary and initial conditions from HEM_NEW. The sensitivity of modelled SSA to various processes is examined including aerosol dry deposition over snow/ice, inclusion of local marine organic aerosols, higher wind speed dependence for sub-micron SSA and representation of sea ice fraction. The possible role of blowing snow and frost flowers is also discussed. Details about the simulations are provided in Table 2. and marine organics, although the latter are highly uncertain. Missing anthropogenic sources could also be contributing underestimation of OA and nss-SO$_4^{2-}$. Many models in the recent AMAP model evaluation of Arctic composition also showed similar discrepancies, attributed to issues with anthropogenic emissions, or model transport, deposition and aerosol formation (Whaley et al., 2022). The results presented here also confirm the importance of interactions between SSA and other inorganic aerosols via heterogeneous uptake, affecting mass concentrations and size distributions, notably NO$_3^-$, and thus model ability to capture wintertime Arctic Haze.

5.1 Aerosol dry deposition
Table 4. Description of the regional-scale WRF-Chem model simulations including at 20km resolution over northern Alaska. See text for details about SSA treatments in the regional runs.

<table>
<thead>
<tr>
<th>Simulation Name</th>
<th>Regional simulations</th>
</tr>
</thead>
<tbody>
<tr>
<td>ALASKA_CONTROL_FEB_JAN</td>
<td></td>
</tr>
<tr>
<td>DRYDEP_FEB-NEW_ALASKA_JAN</td>
<td></td>
</tr>
<tr>
<td>ALASKA_CONTROL_FEB</td>
<td></td>
</tr>
<tr>
<td>LOC_ORG_FEB</td>
<td></td>
</tr>
<tr>
<td>SSA_WS_DEP_FEB</td>
<td></td>
</tr>
<tr>
<td>NEW_ALASKA_FEB</td>
<td></td>
</tr>
</tbody>
</table>

ALASKA_CONTROL_JAN Control run for January 2014 (same setup as ALASKA_CONTROL_FEB) NEW_ALASKA_JAN including all updates in

Previous studies have shown the importance of including wet and dry removal treatments in models (Witek et al., 2007; Eckhardt et al., 2015; Sofiev et al., 2011) estimated that dry deposition, including sedimentation, could contribute more than 50% to SSA removal, especially for super micron SSA. JA11, using model treatments for dry deposition from Zhang et al. (2001) over land and Slinn (1982) over ocean, reported that the loss of super micron SSA is dominated by dry deposition. In the quasi-hemispheric simulations, dry deposition velocities are calculated in MOSAIC based on the Binkowski and Shankar (1995) parametrization. Here, the Zhang et al. (2001) scheme is applied over Alaska, in which the dry deposition velocities are calculated taking into account the different land categories, in contrast to MOSAIC scheme, which uses universal values for processes such as Brownian diffusion and Schmidt number (Slinn and Slinn, 1980; Slinn, 1982). Zhang et al. (2001) has been used in previous studies, for example, by Fisher et al. (2011) and Huang and Jaeglé (2017). These studies applied aerosol dry deposition velocities of $3.0 \times 10^{-4}$ m s$^{-1}$ over snow and ice surfaces for all aerosol diameters and the dry deposition velocity is calculated as a function of aerosol diameter. Zhang et al. (2001) includes detailed treatments of deposition processes, such as Brownian diffusion, impaction, interception, gravitational settling and particle rebound, which highly vary depending on land surface type. Certain parameters link to interception, such as collection efficiency by interception, or impaction processes (e.g. Stokes number) over specific land use categories (such as ice/snow and open ocean), are calculated without considering the radius of surface collector (Giorgi, 1988), but using kinematic viscosity of air, gravitational settling velocity of particle, friction velocity (Slinn, 1982; Seinfeld, 1986). Thus, dry deposition velocities over ice/snow and open ocean are set equal to $3.0 \times 10^{-4}$.

6 Regional processes influencing SSA over northern Alaska

Possible processes affecting SSA emissions on a regional scale over northern Alaska are now examined in more detail, and $1.9 \times 10^{-3}$ m s$^{-1}$, respectively, for both sub- and super-micron aerosols, following Nilsson and Rannik (2001), who reported dry deposition velocity measurements from an Arctic Ocean expedition in 1999. In that way, the influence of more realistic dry deposition velocities on SSA aerosols is examined during wintertime.
First row shows the average values of aerosol mass concentrations for sub-micron during February campaign. Average differences at the surface between DRY DEP FEB and ALASKA CONTROL FEB (second row), and between LOC ORG FEB and DRY DEP FEB (third row) during February campaign for sub-micron Na\(^+\), OA, NO\(_2\)^ – (µ g m\(^{-3}\)). See text and Table 2 for detailed description of the model runs.

Figures 7 and 8 show the effect of this modification for sub- and super-micron SSA and NO\(_2\)^ –, respectively (differences between DRY DEP FEB and ALASKA CONTROL FEB runs). Sub-micron Na\(^+\), OA and NO\(_2\)^ – decrease very slightly, whereas super-micron Na\(^+\), OA and NO\(_2\)^ – increase by up to 0.6, 0.02 and 0.3 µ g m\(^{-3}\), respectively, with the largest increase over in particular, those which may explain low modelled sub-micron SSA at Utqiaġvik. Model simulations are run at 20 km over northern Alaska for shorter periods in January and February 2014 corresponding to the KRP18 measurement campaign. The boundary and initial conditions are taken from HEM NEW. The sensitivity of modelled SSA to a local source of marine organic aerosols, wind speed dependence and the representation of sea-ice areas or regions with snow cover. These changes in modelled sub- and super-micron aerosols are due to differences between the dry deposition velocities in the two schemes. Over model grids covered with snow or ice and open ocean MOSAIC dry deposition velocities are smaller (larger) for sub-micron (super-micron) in magnitude compared to reported velocities by Nilsson and Rannik (2001). During wintertime over northern (inland) Alaska, all the grid cells during the simulations are snow covered. Based on these results, and the fact that super-micron Na\(^+\) and Cl\(^-\) are slightly underestimated at 100 km and Utqiaġvik (see section 4.6), the following simulations use the observed dry deposition velocities reported by Nilsson and Rannik (2001).

First row shows the average values of aerosol mass concentrations of super-micron during February campaign. Average differences at the surface between DRY DEP FEB and ALASKA CONTROL FEB (second row), and between LOC ORG FEB and DRY DEP FEB (third row) in super-micron Na\(^+\), OA, NO\(_2\)^ – (µ g m\(^{-3}\)) during February campaign. See text and Table 2 for detailed description of the model runs.

These results show that sub- and super-micron (mostly) SSA are sensitive to different dry deposition parametrization in WRF-Chem. To address potential uncertainties in dry removal treatments and their influence on SSA regionally, a series of fraction is investigated (see Table 4 for details about the simulations). Differences between runs with and without specific sensitivity tests are also performed. Firstly, correct modelling of aerosol dry deposition depends on the ability of the model to capture the structure of the Arctic boundary layer including vertical temperatures and winds. Model results at 20 km and 100 km horizontal resolution are compared against hourly in-situ 2 m, 10 m temperatures and 10 m wind speeds or temperature and wind speed profiles up to 4 km for January and February 2014 (see figures in APPENDIX D with calculated bias and RMSE for the two periods). Observed wind speeds during January ranged between 4.7 and 14.1 m s\(^{-1}\) and wind directions were mostly easterly (77 to 135 degrees). During February, wind speeds ranged between 0.4 and 13.3 m s\(^{-1}\) and wind directions were mostly easterly, except from 22 UTC 25 February to 11 UTC 26 February when the winds were westerly. In general, the model performs well at 20 km, and better than at 100 km, in terms of temperature and winds, although it slightly underestimates observations at examined sequentially for the surface. On the other hand, there are small discrepancies, of up to 10 degrees, between modelled (at 20 km) and observed wind direction at the Barrow site, near Utqiaġvik town, except at 26 February when these discrepancies are up to 70 degrees.
To examine further causes of variability in modelled dry removal of SSA, a sensitivity test is carried out where aerosol dry deposition and gravitational settling are switched off during a windy day, 28 February 2014. On this day, 10 m wind speeds at period, before evaluating a run including the main changes against observations at Utqiaġvik varied between 7 and 13.5 m s\(^{-1}\) and were easterly (104 to 130 degrees). This corresponds to a period when observed sub-micron Na\(^+\) and Cl\(^-\) concentrations were high, around 1.4 and 2.0 \(\mu\)g m\(^{-3}\), respectively (see Figure 10b). During this day the model captures quite well observed wind speeds and directions, with small differences of up to 2 m s\(^{-1}\) and up to 10 degrees differences in wind direction (not shown here). In this sensitivity run, the model simulates more super-micron SSA (an increase up to 0.8 \(\mu\)g m\(^{-3}\)), which is expected due to the influence of gravitational settling on super-micron particles. The increase on sub-micron SSA is smaller. However, observations of dry deposition of different aerosols are needed to better constrain the model in the Arctic, during the January and February campaign. The possible role of blowing snow and frost flowers is also addressed.

![Figure 7](image)

**Figure 7.** Average absolute differences between LOC ORG FEB and ALASKA CONTROL FEB during the February campaign for sub-micron Na\(^+\), OA, NO\(_3^-\) (\(\mu\)g m\(^{-3}\)). All the results are shown at the surface. See text and Table 4 for more details. Utqiaġvik is shown by the black dot. Note the different scales.

### 6.1 Local source of marine organics

For the simulations at 100 km, the F10 parametrization is used in the 100km HEM NEW run is based on C:Chl-a from a cruise at mid-latitudes. Whilst phytoplankton blooms may not be expected in the high Arctic winter, previous studies have shown evidence of sea ice biological activity under low light conditions coupled with decreased sea ice in the Arctic (Krembs et al., 2002; Lovejoy C., 2007; Hancke et al., 2018). In addition, Analysis of data collected over the Arctic and North Atlantic during winter, and the winter-spring transition, showed that the majority of sub-micron OM is highly correlated with Na\(^+\) concentrations (Russell et al., 2010; Shaw et al., 2010; Frossard et al., 2011; Leaitch et al., 2018). More specifically, Russell et al. (2010) (RUS10 from now on RUS14) analysed samples from the International Chemistry Experiment in the Arctic Lower Troposphere (ICEALOT) cruise and found that most organic mass in clean regions of OM in the
North Atlantic and the Arctic is composed of carbohydrate-like compounds containing organic hydroxyl groups from primary ocean emissions. Frossard et al. (2014) (FRSS14 from now on) investigated the sources and composition of atmospheric marine aerosol particles based on the analysis of various samples, including those from ICEALOT, reporting that ocean-derived organic particles include primary marine organic aerosol (OA). In particular, they calculated the ratio of OC:Na$^{+}$ as a metric for comparing the composition of model-generated primary marine aerosol and seawater previously used by Hoffman and Duce (1976), and reported OC:Na$^{+}$ ratios of 0.45 for atmospheric marine aerosol particles. KRP19 also reported that during their campaign, during their campaign in 2014 almost all the individual SSAs, almost all individual SSA had thick organic coatings (made up of marine saccharides with average C:Na mole ratios of 0.5 and 0.3 for sub-micron and super-micron SSA, respectively) made up of marine saccharides. They also identified open sea-ice leads, sea-ice leads, enriched with exoplymeric substances, as contributing to organic OA in winter SSA.

Here, elemental fractions for sub- and super-micron aerosols, sampled during the KRP19 campaigns, are used to better constrain modelled marine emissions (mOC). More specifically, the marine OC emissions, The ratio of sub- and super-micron OC:Na$^{+}$ is calculated, following FRSS14, and using the elemental fractions from KRP19, as an indicator of the presence of a local source of marine organics. The organic fraction of the total SSA for the high organic activity scenario SSA emissions in WRF-Chem is increased from 0.2 to 0.4 for sub-micron (1st to 5th MOSAIC 8 bins) and 2nd MOSAIC bins), from 0.1 to 0.4 for the 3rd MOSAIC bin and from 0.01 to 0.11 for super-micron (6th to 8th MOSAIC 8 bins) the remaining MOSAIC bins. Note again that no additional SSA mass is added. Figures 7 and 8 show the sensitivity of the model results to including a larger marine organic fraction. Sub- and super-micron OA concentrations increase on average by a small amount, by up to 0.09 and 0.12 $\mu$g $\cdot$ m$^{-3}$, respectively, especially south-west of Alaska and along coastal areas, including Utqiaġvik. Sub-micron Na$^{+}$ and NO$_3^-$ slightly decrease (0.005 $\mu$g $\cdot$ m$^{-3}$) around Utqiaġvik region, and super-micron Na$^{+}$ and NO$_3^-$ decrease north-west of Utqiaġvik.

KRP19 reported measured sub-micron organic carbon volume fractions based on analysis of 150 SSA particles between 0.3 $\mu$m and 0.6 $\mu$m comparable to organic carbon volume fractions observed in SSA produced in mid latitude algal bloom regions. This suggests the presence of significant organic carbon associated with locally produced SSA on the coast of northern Alaska. There are only two available daily observations at Gates of the Arctic GoA during the February campaign simulation period to evaluate the model results. The model captures better observed tOC at the end of February in the run (LOC_ORG_FEB) with higher organic fraction (not shown here). However, it underestimates tOC on 25 February when the observed tOC reached 0.33 $\mu$g $\cdot$ m$^{-3}$ (see APPENDIX F). As mentioned previously, this discrepancy could also be due to missing local anthropogenic sources related to North Slope oil field emissions (Gunsch et al., 2020).

In the following runs, marine organics based on the calculated ratio of OC:Na$^{+}$ are included instead of OA sources. Higher OA fractions in the super-micron leads to lower Na$^{+}$ and, as result lower NO$_3^-$, As indicated above in Section 5, a decrease in super-micron NO$_3^-$ results in an increase in sub-micron NO$_3^-$, Sub-micron Na$^{+}$ increases probably due to the formation of NaNO$_3$ in the model. In the following runs, higher organic fractions are used instead of those from F10, considering the importance of local SSA marine sources at Utqiaġvik (KRP19). By including a local source of marine organics in the model.
Figure 8. Average differences in mass concentrations of (a) sub-micron $\text{Na}^+$, OA, NO$_3^-$, in $\mu$g m$^{-3}$ between SSA_WS_DEP_FEB and LOC_ORG_FEB. (b) The map on the left shows the average value of SSA emission fluxes in $\mu$g m$^{-2}$ s$^{-1}$ during the February campaign and the map on the right shows the average differences between SSA_WS_DEP_FEB and LOC_ORG_FEB emission fluxes in $\mu$g m$^{-2}$ s$^{-1}$. All the results are shown at the surface. Utqiagvik is shown by the black dot. Note the different scales.

...this leads to a better agreement with the findings of the previous studies discussed in section 4.6. It can be noted that there are only sporadic measurements of OA/OC at remote Arctic sites and detailed long-term observations are not available which might help to better constrain model simulations.

6.2 Wind-speed sensitivity to sub-micron SSA emissions dependence

In the regional runs presented so far, the 100km HEM_NEW run, a lower wind speed dependence based on satellite data was used since it improves modelled SSA compared to observations in the 100 km runs (see in section 4.3). At many sites over the Arctic as discussed in Section 5. However, RUS10 found evidence for higher wind speed dependence during the ICEALOT cruise in the Arctic based on data collected during the Arctic leg of the ICEALOT cruise. They found that wind speed is a good predictor of a marine factor, calculated using positive matrix factorization, for sub-micron organic...
mass-matter (OM$_{sea}$). Their analysis showed a high correlation between OM$_{sea}$, sub-micron sodium (Na$_{\pm}^{+}$1) concentration and wind speed at 18-m-18m (correlation r equal to 0.90 for the North Atlantic and Arctic region, see Table S3 Supplementary Material in RUS10). Average OM$_{sea}$ concentrations (0.2 $\mu$g m$^{-3}$) reported by RUS10 for the eastern Arctic Ocean are about half those reported at Utqiaġvik, for example, by Shaw et al. (2010) during wintertime.

Average differences in mass concentrations of (a) sub-micron Na$_{\pm}^{+}$, OA, NO$_{3}^{-}$, in $\mu$g m$^{-3}$, at the surface between SSA_WS_Dep_FEB and LOC_ORG_FEB. Grey star indicates the location of Utqiaġvik. (b) The map on the left shows the average value of SSA emission fluxes in $\mu$g m$^{-2}$s$^{-1}$ during February campaign and the map on the right shows average differences between SSA_WS_Dep_FEB and LOC_ORG_FEB in $\mu$g m$^{-2}$s$^{-1}$.

In a sensitivity simulation, the run, results from RUS10 are used to include a higher wind speed dependence for sub-micron SSA. This linear dependence differs from the power dependencies included in G97, SAL14, and other studies, but is based on empirical relationships determined from analysis of data collected in the Arctic. Equations (5) and (6) from the RUS10 analysis for the Arctic legs of their cruise are applied to the model as a correction factor:

$$\text{Na}^+1\text{Na}^+1 = 0.022 \times U_{18} - 0.012$$  \hspace{1cm} (4)

$$\text{OM}_{sea} = 0.025 \times U_{18} - 0.049$$  \hspace{1cm} (5)

where $U_{18}$ is wind speed at 18-m-18m in ms$^{-1}$, ranging for wind speeds between 2 and 14 m s$^{-1}$ (Figure 2, RUS10). RUS10 used Na$_{\pm}^{+}$1 as a proxy for sub-micron NaCl, and subsequently SSA, because Na$_{\pm}^{+}$1 equalled sub-micron Cl$_{\pm}^{-}$1 on a molar basis for the North Atlantic and Arctic sampling regions. Thus, Equation (5) is also used to estimate a correction factor for Cl$_{\pm}^{-}$. Here, wind speeds in the first model layer are used, i.e. around 26 m-26m. Differences in $U_{18}$ and $U_{26}$ reach a maximum of 1 m s$^{-1}$ (see Fig.D1 in APPENDIX D). Following RUS10, E1 in APPENDIX E), Comparisons with radiosonde data at Utqiaġvik shows that the model performs well in terms of winds and temperatures (see APPENDIX E) and the role of meteorology on aerosols is not discussed further here. The correction factors are only applied in the model to the to simulated number and mass of the SSA emissions when modelled wind speeds are between 2 and 14 m s$^{-1}$, and when RUS10-calculated sub-micron SSA concentrations emissions are greater than model calculated SSA. In this way, SSA emissions are enhanced during periods of higher wind speeds.

To illustrate the sensitivity of the results to applying this correction, Fig. 9 shows differences in sub-micron aerosol concentrations and mass concentrations compared to the run including local marine organics, as well as model SSA emission fluxes, the latter being the sum of dry mass emissions calculated in the model. Overall, this leads to The SSA emission flux is affected over ice-free model grids leading to increased SSA production east and west of Utqiaġvik (by up to 0.015 $\mu$g m$^{-2}$s$^{-1}$) while the highest increases are southwest of Alaska (by up to 0.035 $\mu$g m$^{-2}$). This results in an increase of 0.25, 0.19 and 0.11 $\mu$g m$^{-3}$ in sub-micron Na$_{\pm}^{+}$, NO$_{3}^{-}$ and OA, respectively, over the Utqiaġvik region and southwest Alaska during the February campaign. The SSA emission flux is influenced directly by the area in the model grid which is ice-free. This leads to SSA production east and west of Utqiaġvik while the highest values are southwest of Alaska. By adding the RUS10 correction, SSA emission fluxes increase slightly by up to 0.035 $\mu$g m$^{-2}$s$^{-1}$ along the southwest Alaskan coast, and by up to 0.015 $\mu$g m$^{-2}$s$^{-1}$ around Utqiaġvik. RUS10 showed that sub-micron SSA and wind speed are well correlated over open ocean.
Figure 9. Average differences between ALASKA_NEW_FEB and SSA_WS_DEP_FEB showing the effect of switching from FNL to ERA5 sea-ice fractions during the February campaign for (a) SSA emission fluxes \( \mu g m^{-3} \mu g m^{-2} s^{-1} \), (b) sub-micron mass concentration of Na\(^{+}\) and (c) super-micron mass concentration of Na\(^{+}\). The grey star shows all the location of results are shown at the surface. Utqiaġvik is shown by the black dot. Note the different scales.

Thus, a correction factor to sub-micron SSA, based on in-situ data, improves sub-micron model SSA and could be included in future simulations for studies focusing on these results further illustrate the sensitivity of SSA emissions to wind speeds, in this case affecting fine mode aerosols. These results are in contrast to previous studies finding stronger wind speed dependencies for larger SSA particles, such as Liu et al. (2021b) who analysed aircraft data, including over the Arctic. However, size dependent source functions need to be developed for the Arctic region.

6.3 Sea-ice fractions

The sensitivity of modelled SSA to prescribed sea-ice fractions during wintertime and the role of leads, is also investigated since KRP19 already pointed out the importance of using realistic sea-ice distributions to simulate marine aerosols. High spatial resolution images of sea-ice cover are available, including during the Polar Night, from a marine radar operating on top of a building in downtown Utqiaġvik town (71°17’13” N, 156°47’17” W), 22.5m above sea level, with a range of up to 11km to the northwest (http://seaice.alaska.edu/gi/data/barrow_radar) (Druckenmiller et al., 2009; Eicken et al., 2011). May et al. (2016) previously showed increased super-micron Na\(^{+}\) mass concentrations during periods of elevated wind speeds and lead presence, in a multiyear study using the sea ice radar data at Utqiaġvik. Between 23-28 January 2014, when the winds at Barrow observatory Utqiaġvik were easterly, the radar showed that the coastal area east of Utqiaġvik featured open leads (KRP19). From 24-28 February 2014, the west coastal area also featured leads as also shown by Moderate Resolution Imaging Spectroradiometer (MODIS) satellite images (KRP19). To examine the sensitivity of modelled SSA emissions to sea-ice cover, ERA5 sea-ice fractions with a resolution of 0.25\(^\circ\) x 0.25\(^\circ\) are used instead of FNL fraction at 1.0\(^\circ\) x 1.0\(^\circ\) resolution. Note that only sea-ice fraction field is different, while the rest of the meteorological fields are from FNL.
Figure 10. Evaluation of modelled aerosol composition against in-situ observations at Utqiagvik during the (a) January and (b) February 2014 campaign in UTC (00z,12z) and STP conditions. The black lines show model results from the ALASKA_CONTROL_JAN/FEB; the red lines show the ALASKA_NEW_JAN/FEB, while the daily observations are shown as blue crosses. The corresponding model daily averages are shown as black diamonds for the ALASKA_CONTROL_JAN/FEB runs and as red pentagons for ALASKA_NEW_JAN/FEB runs. Green circles show observed ss-SO$_4^{2-}$. The grey lines and pentagons show modelled ss-SO$_4^{2-}$ for ALASKA_NEW_JAN/FEB, while dark turquoise pentagons show modelled ss-SO$_4^{2-}$ for ALASKA_CONTROL_JAN/FEB. See the text for more details.

Results for February are shown in Fig. 10. The SSA emission flux (Fig. 10a) increases over a small region around west of Utqiagvik and across the North Slope of Alaska due to decreased sea-ice fraction but decreases but decreases just to the east of Utqiagvik and southwest of Alaska (e.g. Selawik Lake and Norton Bay) due to increased sea-ice fraction. Sub-micron Na$^{+}$ slightly increases along the north coast of Alaska and around Utqiagvik, by up to 0.1 $\mu g.m^{-2}$ and around Utqiagvik $\mu g.m^{-3}$ (see Fig. 10b) and 9b). Larger super-micron Na$^{+}$ increases are simulated by up to 0.4 $\mu g.m^{-3}$ around Utqiagvik, and decreases by up to 0.4 $\mu g.m^{-3}$ southwest of Alaska (Fig. 10c). The model results for January indicate that there is less sea-ice in the region around Utqiagvik and south west of Alaska. Therefore, higher 9c). Higher SSA emission fluxes were simulated for February (0.035 $\mu g.m^{-2}.m^{-3}.s^{-1}$) compared to January (0.015 $\mu g.m^{-2}.m^{-3}.s^{-1}$) maps, since there is more sea-ice in the region around Utqiagvik and south west of Alaska in the January simulation (not shown here).
Time-series during a) January and b) February 2014 of sub-micron mass concentrations of Na\(^+\), Cl\(^-\), NO\(^3\), NH\(^+\), SO\(^2\)\(^-\), in \(\mu\)g m\(^{-3}\), simulation period. Model simulations are validated against in-situ sub-micron aerosols at Utqiaġvik, Alaska, in UTC (every 12h; 00z, 12z). The black line shows model results from the CONTROL run; the red line shows the ALASKA_NEW run, while the daily observations are shown in blue crosses. The corresponding model daily averages are shown as black diamonds for the control simulation and as red pentagons for the ALASKA_NEW runs. See the text for details about the observations and model runs.

Two further simulations are performed to examine model sensitivity to sea-ice fraction. First, ERA5 sea-ice fractions are set equal to 0 (ice-free conditions) zero to the north, west, and east of Utqiaġvik to examine the effect of having ice-free conditions and the presence of open leads locally (as seen by the radar). Second, ERA5 sea-ice fractions are set equal to 0.75 north, west, east of Utqiaġvik and northwest of Alaska. In both cases, the model is run on a windy day (28 February 2014). The first sensitivity test leads to an increase in SSA emission fluxes by up to 0.2 \(\mu\)g m\(^{-2}\) s\(^{-1}\), where \(\mu\)g m\(^{-2}\) s\(^{-1}\) when sea-ice fraction equals zero (not shown) and 1.2 \(\mu\)g m\(^{-2}\) s\(^{-1}\) and 0.05 \(\mu\)g m\(^{-2}\) s\(^{-1}\) in super-micron and sub-micron Na\(^+\), respectively. The second sensitivity test yields similar results. This is because ERA5 sea-ice fractions are higher (more sea-ice) than the test case (0.75) leading to an overall increase small increases in the SSA emission flux of up to 0.02 \(\mu\)g m\(^{-2}\) s\(^{-1}\), especially east of Utqiaġvik, affecting primarily. Again, super-micron SSA (increases of up to 1.5 \(\mu\)g m\(^{-3}\)) rather \(\mu\)g m\(^{-3}\) are affected more than sub-micron SSA, probably due to the short simulation period.

These results illustrate the regional sensitivity of super-micron SSA, in particular, to the prescribed sea-ice fraction and point out the need of improving this in models. Regarding rather than sub-micron SSA, which is less sensitive to local-to-prescribed sea-ice in these model simulations, there is the possibility that missing-fractions. Missing mechanisms influencing sub-micron SSA emissions may need to be included in the model such as SSA production of, in particular, ultrafine particles, from breaking waves in the surf zone for particles with diameters between 1.6 and 20 \(\mu\)m \((\text{De Leeuw et al., 2000})\) or diameters ranging between 0.01–0.132 (ultrafine), 0.132–1.2 and 1.2–8.0 \(\mu\)m \((\text{Clarke et al., 2006})\), which would be important and in the ice-free ocean. However, information about wave-breaking activity in the surf zone during winter along the northern Alaskan coast is needed to address this.

### 6.4 Evaluation against observations in northern Alaska

The model is also run for January 2014 including all the updates described above (see Table 2 and section 2.3). Fig. 11 shows the comparison between runs. Results from runs at 20km with and without all these updates—the main changes included in the sensitivity tests (local source of marine organics, higher wind speed dependence and ERA-5 sea-ice fractions) are compared to sub-micron aerosol observations at Utqiaġvik during for both the January and February campaigns. Note that the focus of this section is on sub-micron SSA, as there are not detailed 2014 campaign (see Fig. 10). We note that there are no super-micron observations during the periods of the simulations due to their weekly temporal variation and due to the fact that the model still underestimates observed-simulation periods due to the weekly sampling frequency. It is interesting to compare to these periods since the observations show different behaviours. While observed sub-micron SSA at Utqiaġvik...
There are differences in the observations between the two periods. While sub-micron observed Na\(^+\) and Cl\(^-\) did not exceed 1 µg m\(^{-3}\) during January, observed sub-micron Na\(^+\) and Cl\(^-\) concentrations reached up to 2.5 µg m\(^{-3}\) in February. They did not exceed 1 µgm\(^{-3}\) during January. As noted earlier such high concentrations of Na\(^+\) and Cl\(^-\) were not observed at Alert and Villum during January and February 2014. This could be explained by the fact that these two sites are entirely surrounded by more sea ice in winter. Overall, the model simulates better observed The January run, including all the updates (ALASKA_NEW_JAN), captures better sub-micron Na\(^+\) and Cl\(^-\) in January but still underestimates concentrations \(^+\) and Cl\(^-\) (reduced biases and RMSEs - see APPENDIX F, Table F1) although it underestimates observations by up to 0.3 and 0.6 µg m\(^{-3}\) and 0.8 µgm\(^{-3}\) (Fig. 10a), respectively, while sub-micron NO\(_3^-\) is slightly overestimated. Biases in January decrease from −0.31 to −0.16, −0.50 to −0.33 and −0.04 to 0.039 µg m\(^{-3}\), respectively. On the other hand, and RMSEs (see APPENDIX F, Table F2) are also slightly improved for February although sub-micron Na\(^+\), Cl\(^-\), Cl\(^-\) are still underestimated in the run including all the updates (ALASKA_NEW_FEB) by up to 2.0 µg m\(^{-3}\) in February indicating that there are missing processes in the model linked to 1.5 µgm\(^{-3}\). Slightly more NO\(_3^-\) is simulated in January, even if the model still underestimates the elevated observations. Up to six times more OA is simulated during both periods in better agreement with reported observations (Moschos et al., 2022b) as discussed in section 5.2. The sensitivity tests discussed in this section do not directly address the model underestimation of elevated episodes of sub-micron SSA emissions, as discussed earlier. However, overall the results at Utqiaġvik in February, including all the updates (ALASKA_NEW_FEB), are better compared to the control simulation (ALASKA_CONTROL_FEB). Biases for Na\(^+\), Cl\(^-\), SO\(_4^{2-}\), since it is mostly nss-SO\(_4^{2-}\), and thus the changes are small (during both simulation periods). We note that these runs at 20km are for periods including elevated aerosols and are thus more challenging for the model to reproduce. Runs at higher resolution may be needed to better resolve, for example, sea-ice distributions.

Observations of particle number concentration are also used to validate the regional model results at Utqiaġvik (see Fig. 11). High number concentrations are observed during both periods, up to 10\(^3\) particles per cm\(^3\), especially for particle sizes less than 20 nm. Freud et al. (2017) reported similar wintertime magnitudes in the accumulation mode (diameter range 100-150 nm) at Utqiaġvik, averaging between 1 x 10\(^2\) and NO\(_3^-\) decrease from 1.29-2 x 10\(^2\) particles per cm\(^3\), whereas the magnitude is smaller for particles with diameters less than 50nm. They also noted that particle number concentrations are higher at Utqiaġvik and Tiksi (in northern Russia) compared to other Arctic sites (Alert, Villum and Zeppelin). The model tends to underestimate observed number concentrations, especially in the 4\(^{th}\) (312.5 to 1.18, −1.90 to −1.78 and −0.20 to −0.11 µg m\(^{-3}\)), respectively. During both months, the model lacks SO\(_4^{2-}\) due to missing local anthropogenic sources, as discussed in section 4.6 and due to small contribution from ss-SO\(_4^{2-}\) as is shown also in section 4.1 for different Arctic sites. Missing aqueous phase reactions, such as the oxidation of SO\(_2\) by ozone in alkaline SSA aerosols (Alexander et al., 2005), are missing from the model and might explain these high discrepancies compared to 625.0 nm) and 5\(^{th}\) (625.0 to 1250 nm) MOSAIC bins, even if the model compares better in January when measured number concentrations are lower. This is consistent with the evaluation of sub-micron SSA and other aerosol components, particularly for episodes when observed aerosols were enhanced (Fig. 10). Note that the model results cannot be compared to measurements smaller than 39 nm because MOSAIC does not represent these aerosols explicitly.
and nucleation is parameterised. Inclusion of a source function to account for ultrafine SSA emissions, for example, from breaking waves at the surf zone, may lead to improved model results (Clarke et al., 2006).

Overall the results the results presented so far in this Section show that modelled sub-micron observations at Utqiaġvik. Also, the variations in modelled NH$_4^+$ between the ALASKA_CONTROL and ALASKA_NEW for January and February simulations are small. The model underestimates observed NH$_4^+$, which peaks at 0.2 µg m$^{-3}$ during February. Calculated SSA (Na$^+$, Cl$^-$), and as a consequence NO$_3^-$, are more sensitive to using a higher wind speed dependence than sea-ice fractions, over northern Alaska, based on estimated biases and RMSEs for all aerosol species and for January and February campaigns are given in APPENDIX F.

Comparison with data from Gates of the Arctic (see APPENDIX E) shows that there are not significant differences between the control run and including all the updates in February 2014. The model still underestimates observed IOI due to missing local anthropogenic sources and overestimates SO$_4^{2-}$, NO$_3^-$, Na$^+$ and Cl$^-$ each test simulation (not shown here). Sea-ice fractions have a greater effect on super-micron SSA mass concentrations. Modelled sub-micron OA are more sensitive to a higher wind speed dependence and, to a lesser extent, the introduction of an additional source of local marine organics. However, due to short period of the simulation only two observations are available, thus more detailed observations are needed to examine further the reason why the model differs from the observations at this site the latter is highly uncertain.
Table 5. Average sub-micron modelled and observed depletion factors, following Frey et al. (2020), during the campaign in January and February 2014 at Utqiaġvik. Model results for ALASKA_NEW_JAN and ALASKA_NEW_FEB simulations are shown here, respectively. Observations refer to sub-micron data from NOAA. See text for details.

<table>
<thead>
<tr>
<th>Depletion Factors</th>
<th>Model</th>
<th>Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>January campaign</td>
<td></td>
<td></td>
</tr>
<tr>
<td>DF(_{\text{SO}_4^{2-}})</td>
<td>-0.77(-0.94)</td>
<td>-7.56</td>
</tr>
<tr>
<td>DF(_{\text{Na}^{+}})</td>
<td>-1.05(-0.95)</td>
<td>-0.09</td>
</tr>
<tr>
<td>February campaign</td>
<td></td>
<td></td>
</tr>
<tr>
<td>DF(_{\text{SO}_4^{2-}})</td>
<td>-4.8(-2.2)</td>
<td>-2.15</td>
</tr>
<tr>
<td>DF(_{\text{Na}^{+}})</td>
<td>-1.1(-1.2)</td>
<td>-0.19</td>
</tr>
<tr>
<td>DF(_{\text{Br}^{-}})</td>
<td>-</td>
<td>0.063</td>
</tr>
</tbody>
</table>

6.5 Are blowing snow and/or frost flowers a source of sub-micron SSA during wintertime at Utqiaġvik?

Lastly, we consider whether enhanced SSA, in particular in the sub-micron size range, at Utqiaġvik could be due to blowing snow or frost flower sources. We noted earlier that As noted earlier, KRP19 found no evidence of blowing snow or frost flowers at this site but that SSA originated from open leads during wintertime. The findings of KRP19 Their findings are supported by the earlier laboratory study of Roscoe et al. (2011) who reported that frost flowers are not an efficient source of SSA. However, an older study by Shaw et al. (2010) found that during winter at Utqiaġvik surface frost flowers formed on the forming on sea and lake ice are a source of ocean derived OM. Modelling studies that have included a source of blowing snow and frost flowers suggest that they are contributing to SSA at this time of year at Utqiaġvik, Alert and Zeppelin (Xu et al., 2013, 2016; Huang and Jaeglé, 2017; Rhodes et al., 2017).

To investigate whether blowing snow or frost flowers could also be a source of SSA during the campaigns at Utqiaġvik, depletion factors are estimated following Frey et al. (2020) Frey et al. (2020) (FR20 from now on). FR20 reported that blowing snow was the main source of SSA rather than frost flowers and open-leads in Antarctic wintertime, based on SO\(_4^{2-}\) and Br\(^{-}\) bromide (Br\(^{-}\)) depletion in SSA being indicative of a blowing snow origin, and not seawater. Other studies also suggested that blowing snow and frost flowers near Utqiaġvik are characterised by SO\(_4^{2-}\) depletion compared to seawater (Douglas et al., 2012; Jacobi et al., 2012). Here, depletion factors are calculated using modelled and observed sub-micron aerosol concentrations mass concentrations during the campaign periods. More specifically, total SO\(_4^{2-}\) depletion relative to Na\(^{+}\) (DF\(_{\text{SO}_4^{2-}}\)\(\text{Na}^{+}\)), Na\(^{+}\) depletion relative to Cl\(^{-}\) (DF\(_{\text{Na}^{+}}\)\(\text{Cl}^{-}\)), and Br\(^{-}\) depletion relative to Na\(^{+}\) (DF\(_{\text{Br}^{-}}\)\(\text{Na}^{+}\)) are calculated using the following equation:

$$\text{DF}_x = 1 - \frac{R_{\text{smpl}}}{R_{\text{RSW}}}$$

(6)
Table 6. Average modelled and observed molar ratios for sub-micron SSA, following Kirpes et al. (2019), during the campaign in January and February 2014 at Utqiaġvik. Model results from ALASKA_NEW_JAN and ALASKA_NEW_FEB simulations are used. Observations refer to sub-micron data from NOAA.

<table>
<thead>
<tr>
<th>Molar ratios</th>
<th>Model</th>
<th>Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>January campaign</td>
<td></td>
<td></td>
</tr>
<tr>
<td>total-SO$_4^{2-}$:Na$^+$</td>
<td>0.12</td>
<td>0.55</td>
</tr>
<tr>
<td>Cl$^-$:Na$^+$</td>
<td>0.71</td>
<td>1.1</td>
</tr>
<tr>
<td>February campaign</td>
<td></td>
<td></td>
</tr>
<tr>
<td>total-SO$_4^{2-}$:Na$^+$</td>
<td>1.5</td>
<td>0.2</td>
</tr>
<tr>
<td>Cl$^-$:Na$^+$</td>
<td>0.8</td>
<td>1.08</td>
</tr>
</tbody>
</table>

where $\frac{R}{x}$ is the mass ratio (x:y) of species $x,y$ in the model or in the sample (smpl) and in reference seawater (RSW) (Millero et al., 2008). A depletion factor (DF$_x$) between 0—zero (small) and 1 (strong) indicates 0–100% depletion, whereas DF$_x$ smaller than 0—less than zero indicates enrichment. Frey et al. (2020) FR20 suggested, based on depletion of SO$_4^{2-}$ relative to Na$_{ss}^{+}$, that most SSA originates from blowing snow on sea-ice with minor contributions from frost flowers, and not from open leads.

Average The average values of modelled and observed DFs are shown in Table 4. In January, observed Total SO$_4^{2-}$ is enriched relative to Na$^+$ in both the observations and the model results during both campaign periods, in contrast to FR20 who reported substantial depletion. In February, observed and model results both indicate SO$_4^{2-}$ enrichment relative to seawater, whereas in January, model results are less enrichment compared to the observations, possibly due to underestimation of nss-SO$_4^{2-}$. In January, observed total SO$_4^{2-}$ concentrations are 8.56 times more 7.56 times higher than in reference seawater, possibly due to internal mixing with anthropogenic SO$_{nss}$-SO$_4^{2-}$ from NSA oilfield emissions (+ as noted by KRP18), whilst in the model, Modelled total SO$_4^{2-}$ concentrations are 1.77 is less enriched than the observations (0.94 times higher than in reference seawater, showing enrichment in both cases (Table 4). Modelled and observed depletion factors also show enrichment in February. This is in contrast with results from Frey et al. (2020) who reported substantial depletion. They also reported ), likely due the model underestimation of nss-SO$_4^{2-}$. FR20 did report a case of enrichment due to possible contamination from the ship, an anthropogenic source. Our modelling results and the observations at Utqiaġvik indicate enrichment of SO$_4^{2-}$ relative to Na$^+$, suggesting that blowing snow and frost flowers are not a source of SSA, at least during these campaigns. Previous studies (Douglas et al., 2012; Jacobi et al., 2012) suggested that blowing snow and frost flowers near Utqiaġvik are characterised by SO$_4^{2-}$ depletion compared to seawater. Na$^+$ depletion relative to Cl$^-$ during both campaigns The Na$^+$ depletion factor also shows enrichment during both campaigns, albeit more negligible in the observations than in the model. Observed Na$^+$ depletion relative to Cl$^-$ is 1.09 or 1.19 times more than in reference seawater, during January and February, respectively. Our analysis suggests that blowing snow and frost flowers are not a significant source of SSA, at least during the campaign.
SSA can also play an important role in polar tropospheric ozone and halogen chemistry through the release of active bromine during spring (Fan and Jacob, 1992; Simpson et al., 2007; Peterson et al., 2017). Reactions involving bromine are an important sink of ozone (O$_3$) (Barrie, 1986; Barrie et al., 1988; Wang et al., 2019a; Marelle et al., 2021) and also cause mercury oxidation (Schroeder et al., 1998). Br$^-$ depletion relative to Na$_{a+}$ is calculated only during February, since observed Br$^-$ was zero during the period of January campaign, and indicates a small depletion in reference seawater. The calculated observed mass ratio of Br$^-$ to Na$_{a+}$, based on the available observations of Br$^-$ during February, indicates a January campaign period. The results for February show a small depletion indicating a seawater origin. The observed mass ratio of Br$^-$ to Na$_{a+}$ to Na$_{a+}$ ranges between 0.0057 and 0.0059, while the mass ratio of Br$^-$ to Na$_{a+}$ to Na$_{a+}$ in reference seawater is equal to 0.006. On the other hand, Frey et al. (2020) FR20 reported no or little Br$^-$ depletion relative to Na$_{a+}$ due to Br$^-$ losses at the surface and small depletion further aloft (in Antarctica). For a more comprehensive analysis, observations are required at different locations and altitudes across coastal northern Alaska.

We note that the version of WRF-Chem used in this study does not include halogen chemistry. It has since been implemented in a later version by Marelle et al. (2021) to examine springtime ozone depletion events during March-April 2012 at Utqiaġvik. Heterogeneous reactions on sea-salt aerosols emitted SSA from the sublimation of lofted blowing snow were included. Their results suggested that blowing snow could be a source of SSA during spring although it should be noted that this version of the model overestimated SSA at remote model version, including blowing snow as a source of SSA, overestimated SSA ($d_4 < 10 \mu$m) at Arctic sites, such as Alert and Villum, when blowing snow was included as a source of SSA. Also, they during spring and did not examine wintertime conditions.

Finally, following KRP19, modelled and observed molar ratios of sub-micron Cl$^-$:Na$_{a+}$:Na$_{a+}$ and SO$_4^{2-}$:Na$_{a+}$ are estimated to further examine the origins of SSA and compare our findings with KRP19 (see Table 5). Observed 6). The averaged molar ratios of Cl$^-$:Na$_{a+}$ sub-micron Cl$^-$:Na$_{a+}$ and SO$_4^{2-}$:Na$_{a+}$ for January and February campaign periods derived here for the campaign periods (Table 6) agree with KRP19 (Cl$^-$:Na$_{a+}$:Na$_{a+}$ equal to 1.08, see KRP19 supplement - Table S3 and text.). This indicates, text and references within. They indicate a seawater origin (following Pilson (2012)), and confirms and confirm the findings of KRP19 that there was no evidence for blowing snow and frost flowers as a source of SSA during the campaign Utqiaġvik campaign, also in agreement with previous studies (May et al., 2016). Model averaged molar ratios are smaller in magnitude than the observations. Observed and modelled ratios differences in magnitude could be altered by these discrepancies could be due to the fact that the model underestimates sub-micron SSA and SO$_4^{2-}$, due to missing mechanisms for sub-micron SSA emissions and local/regional anthropogenic sources of SO$_4^{2-}$. Differences between observed and modelled Cl$^-$:Na$_{a+}$ ratios for the reasons noted earlier. Differences could also be due related to issues with modelled SSA lifetime and chemical processing during long range transport. Previous studies found that sub-micron SSA have larger chloride depletion than super-micron SSA (Barrie et al., 1994; Hara et al., 2002; Leck et al., 2002) - long-range transport.
2016 used Cl\textsuperscript{−}:Na\textsuperscript{+} molar ratio enrichment factors of Cl\textsuperscript{−}:Na\textsuperscript{+} as an indicator of long-range transport influence on SSA at Utqiaġvik. They reported that Cl\textsuperscript{−} depletion was larger for sub-\textsuperscript{aged} sub-micron than aged super-micron SSA due to a longer lifetime. On average during the simulation periods in January and February 2014, the results, in line with other studies (Leck et al., 2002; Hara et al., 2002). The regional results (ALASKA\_NEW\_FEB) indicate that modelled Cl\textsuperscript{−} sub-micron Cl\textsuperscript{−} has undergone significant atmospheric processing. This is consistent with the regional model results, influenced by the simulation at 100 km, has too much aged sub-micron SSA, while the contribution from locally produced sub-micron SSA may be too low in the model (modelled enrichment factors equal to 0.5 and 0.4 during January and February simulation periods, respectively, lower than the threshold (0.75) defined by May et al. (2016)). On the other hand, modelled enrichment factors for super-micron are equal to 0.6 and 0.85 to during January and February, respectively, indicating that there is a possibly background influence on super-micron SSA during January, while they are locally produced during February. KRP18 observing reported the presence of both nascent-fresh (locally-produced) SSA and aged (partially chloride-depleted) SSA Cl\textsuperscript{−}-depleted) SSA for sub-micron SSA, while super-micron were mostly fresh (KRP18, Figure 2).

Based on this analysis of depletion factors and molar ratios, little evidence suggest a blowing snow influence on the analysis above (including observations), there is little evidence suggesting that blowing snow or frost flowers are a significant source of SSA during the campaigns campaign at Utqiaġvik is found. Rather, the presence of predominantly easterly winds (s.s. 5,3) and the presence of leads east of Utqiaġvik (especially during February), suggests that the primary source of SSA was marine from open leads and open leads are an important primary source, in agreement with the findings of KRP19.

6.6 Conclusions

In this study, the ability of the WRF-Chem is used to investigate Arctic Haze composition at remote Arctic sites during wintertime model to simulate wintertime Arctic aerosols is assessed with a particular focus on SSA processes influencing SSA emissions and the contribution of SSA to Arctic Haze. Model performance is evaluated first in terms of reproducing aerosol composition in the Arctic before focusing on processes influencing SSA at regional scales over northern Alaska during winter 2014.

The control version of WRF-Chem overestimates super-micron, coarse mode and TSP SSA due under Arctic Haze conditions. The inclusion of updated treatments of SSA emissions leads to improved simulation of SSA over the wider Arctic compared to the still widely used Gong et al. (1997)-based source function included in the base model. Na\textsuperscript{+} and Cl\textsuperscript{−} biases are reduced by a factor of 7 to missing and out of date SSA emission treatments in the model. In particular, the addition of observations at Alert, Villum and Zeppelin, and by a factor of 4 compared to super-micron Na\textsuperscript{+} and Cl\textsuperscript{−} data at Utqiaġvik. The addition of the SST dependence has a larger effect on modelled SSA compared to updating the wind speed dependence, and is responsible for two-thirds of the reductions in super-micron/coarse mode SSA, due to low SSTs in the Arctic. The use of a more realistic lower wind speed dependence for SSA, based on satellite data, and inclusion of a dependence of SSA emissions on SSTs lead to improved results for also results in lower super-micron, coarse mode and TSP SSA and NO\textsubscript{3}\textsuperscript{−} over the Arctic. The latter has already been included in certain modelling studies. Also, recent data analysis studies in the Arctic have pointed out that wind speed alone cannot predict SSA production and that other mechanisms, such as SST dependence, are needed.
However, there are still uncertainties regarding the role of SSTs in SSA production. Other factors such as seawater composition, wave characteristics, fetch and salinity need to be considered in future versions of WRF-Chem. In this study, marine organic aerosol emissions are also activated in the model since they are an important component of SSA in the Arctic and globally. Inclusion of all these updates leads to improved representation of SSA over the wider Arctic. Modelled super micron SSA and salinity may also be playing a role and should be considered in future versions of WRF-Chem.

In addition, missing sources of ultrafine SSA particles, for example, due to breaking waves at the coast, could be included by defining the surf zone in the model (Clarke et al., 2006). In all cases, more field data is needed to understand and develop SSA source functions specific to the Arctic during winter.

Results from this study also highlight the importance of interactions between SSA and other inorganic aerosols, notably NO$_3^-$, which have largely anthropogenic origins, and contribute to wintertime Arctic Haze. Improved simulation of Na$^+$ and Cl$^-$ leads to less coarse mode and TSP SSA are reduced at all Arctic sites in more fine mode NO$_3^-$ in the model, in better agreement with the observations. Results for NO$_3^-$ are also improved overall. This is due to less formation of NO$_3^-$ via heterogeneous uptake of HNO$_3$. Inclusion of the SST dependence only has a small effect on sub-micron SSA in the Arctic. In the future, other SST dependencies could be considered such as that proposed by Sofiev et al. (2011) which could increase sub-micron SSA at low temperatures (Salter et al., 2015; Barthel et al., 2019). However, further field data studies are needed to confirm such dependencies in the Arctic, primarily in the coarse mode, and more NO$_3^-$ in the fine mode, in line with previous mid-latitude studies. As a result, simulated aerosols in the updated model are slightly less acidic in the Arctic, improving agreement with some Arctic sites, even if the model tends to have aerosols, which are too acidic (at some sites).

Marine organic aerosol and sub-micron sea-spray emissions are also activated in the model since they are an important component of SSA in the Arctic, and globally, and a source of ss-SO$_{4}^{2-}$ is also added to the model leading to improved modeled SO$_{4}^{2-}$ in the high Arctic (e.g. Alert) and Alaskan (e.g. Gates of the Arctic, Simeonof) sites.

Simulated OA is improved at the Simeonof sub-Arctic site with reduced biases, up to a factor of 4, although, in general, OA is underestimated at sites over the wider Arctic. The addition of ss-SO$_{4}^{2-}$ agrees well with ss-SO$_{4}^{2-}$ derived from the observations at most Arctic sites and leads to improved modelled total SO$_{4}^{2-}$. However, at sites such as Utqiagvik, which may be influenced by the Prudhoe Bay oilfields, the model still underestimates sub-micron SOZeppelin and Villum, which are dominated by nss-SO$_{4}^{2-}$ possibly due to missing anthropogenic emissions. Missing aqueous chemical formation of SO$_{4}^{2-}$ in dark conditions may also explain these discrepancies (e.g., this additional source results in further overestimation. While super-micron SO$_{4}^{2-}$ production from metal catalyzed O$_2$ oxidation of S(IV), McCabe et al. (2006)). Results from the improved quasi hemispheric run indicate a shift in the balance between (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$, with aerosols being less acidic than the base version of the model.

Overall, super-micron, coarse mode and TSP SSA, OA, SO$_{4}^{2-}$, NH$_4^+$ and NO$_3^-$ are improved in the HEM_NEW quasi hemispheric simulation compared to observations at Arctic sites, based on biases and RMSEs. However, the model underestimates sub-micron SSA at primarily of sea-salt origin, is captured better at Utqiagvik where there are episodes with significantly higher SSA compared to other Arctic sites on the northern Alaskan coast, sub-micron SO$_{4}^{2-}$, which is primarily nss-SO$_{4}^{2-}$, is underestimated.
at this site during episodes with elevated concentrations, and also at Gates of the Arctic further inland. Model discrepancies in OA and nss-SO$_4^{2-}$ may be due to missing local anthropogenic emissions, coupled with missing heterogeneous or dark reactions leading to secondary aerosol formation. In the case of OA, primary marine emissions may also be underestimated. It can be noted that such underestimations are a common feature in other models (Whaley et al., 2022). Uncertainties in model transport and wet and dry deposition processes may also be responsible for deficiencies in modelled wintertime Arctic aerosols (Whaley et al., 2022).

Model sensitivity to different processes affecting wintertime SSA over northern Alaska during winter is explored. KRP19 pointed out that there is sea ice biology influence is explored further with the aim to understand, in particular, model underestimation of sub-micron SSA at Utqiagvik during wintertime and winter 2014 when field data analysis showed that marine emissions are from open leads were an important source of organic aerosols at this location. In order to include local sources of marine organics, the ratio SSA, including marine organics (KRP18, KRP19). Based on observed ratios of OC:Na$^+$ is used leading to + from the KRP19 campaign, a local source of marine organics is included in model runs at 20km over northern Alaska. This results in higher modelled OA, in better agreement with previous measurements at this site (and at Alert) and its advised to be included in future WRF-Chem simulations in the Arctic region. To further explore the uncertainties on sub-micron SSA, and other sites such as Alert, although the model still tends to underestimate reported data.

The sensitivity of modelled SSA over northern Alaska to using a higher wind speed dependence, based on Arctic data, is also investigated. This leads to an increase in modelled sub-micron SSA, with the model performing better during January than in February. Model sensitivity to prescribed sea-ice fractions is also explored. In a run with ERA5 instead of FNL, sea-ice fraction is tested in the model. The results, in combination with different sensitivities changing sea-ice fraction, show that fractions, modelled super-micron SSA are more sensitive to sea-ice treatments than sub-micron SSA in the model. In general, modelled sub-micron SSA are more sensitive to the use of a higher wind speed dependence rather than the distribution of sea-ice. To improve model simulations in this region, field campaigns are needed to study processes influencing wintertime SSA production and to determine more realistic sea-ice fractions which vary on at least a daily basis. The use of satellite sea-ice data, combined with higher resolution simulations over Utqiagvik and coastal Arctic sites, will help to gain more detailed further insights into the influence of open leads on SSA production during wintertime. The results of this study also highlight that SSA dry removal is less important than the role of open leads in the Arctic during wintertime. The role of wet deposition on SSA is also examined. In that case, the precipitation flux is doubled and as result super-micron SSA decreased, but the sensitivity did not affect sub-micron SSA. Wet deposition is not addressed further in this study, because according to NOAA climate data recorded precipitation and snowfall was the lowest during February 2014. Wet deposition is addressed in details in the companion paper for BC. Our results suggest that further investigation is needed to determine more realistic dry deposition velocities over snow, ice and ocean in the Arctic and to derive more realistic sea-ice fractions, including the presence of open leads, which can vary over periods of days. The sensitivity of model results to using a higher wind speed dependence, based on data from Russell et al. (2010), is investigated for sub-micron SSA. This leads to small improvements in the model sub-micron SSA, with the model performing better during January than February period of the campaign open leads on SSA production, including marine organics, during wintertime.
Further analysis is required to understand the origins of, in particular, missing local anthropogenic sources could also explain some of the discrepancies in modelled sub-micron SSA in northern Alaska, and to improve their representation in the WRF-Chem model. For example, missing sources of sub-micron SSA, such as a source function for ultrafine SSA particles due to breaking waves (Clarke et al., 2006) could be included. Also, anthropogenic sources of Cl\(^-\) may need to be considered, such as road salt in urban areas (McNamara et al., 2020; Denby et al., 2016) or coal combustion, waste incineration, and other industrial activities (Wang et al., 2019b) which are not included in current global inventories. The model also lacks emission inventories, WRF-Chem, and models in general, also lack anthropogenic emissions of Na\(^+\). Anthropogenic sources of Na\(^+\) could be wastewater and sewage treatment systems, contamination from landfills and salt storage areas (e.g., Panno et al. (2006)). However, detailed +, which could possibly account for up to 30% of Na\(^+\), as noted by Barrie and Barrie (1990). However, the analysis of depletion factors and molar ratios at presented here for Utqiaġvik, Alaska showed that during the simulation period the shows that the main source of SSA are from marine emissions fresh SSA is from marine sources including open ocean or leads and there is no evidence of. We also find no evidence for frost flowers or blowing snow as a source of SSA + at least during the periods considered in this study at Utqiaġvik, in agreement with the findings of KRP19 and previous studies (May et al., 2016). Further insights into wintertime marine SSA sources, including organics are needed, as well as improved quantification of local anthropogenic emissions. Further observations from field measurements are needed to better understand SSA emissions and their dependencies.

This model study supports recent findings based on observations that SSA make an important contribution. Overall, we find that wintertime SSA at remote Arctic sites contribute between 54% and 84% to total inorganic SSA (observations and improved model results), in agreement with previous findings, that SSA are important contributors to super-micron (coarse mode, TSP) mass concentrations during wintertime at remote Arctic sites. Future work has to consider carefully possible sources of sub-micron SSA and their inclusion in models, in order to explain observed SSA during wintertime. Processes linked to the Ice fractures and the area of open ocean are likely to become more important with decreasing sea-ice cover in the Arctic due to as a result of climate warming. Observations This may lead to more SSA which can act as CCN or INPs with implications for Arctic aerosol-cloud indirect effects, notably long-wave radiative forcing which dominates in winter (Eidhammer et al., 2010; Partanen et al., 2014). As well as ground-based measurements, vertical profiles of SSA components including organic aerosols (often missing) are needed as are also needed to better understand SSA sources and their impacts on clouds. Such studies will ultimately help to improve understanding about processes and their treatments in models, and in order to reduce uncertainties in estimation of aerosol radiative effects, estimates of aerosol-cloud indirect radiative effects and the magnitude of the associated radiative cooling (Horowitz et al., 2020) or warming (Zhao and Garrett, 2015).

**Code availability.** The code used to calculate SSA emissions in this study is available on Zenodo as: Ioannidis et al. (2023)

https://doi.org/10.5281/zenodo.7502210
Data availability. All data used in the present paper for Zeppelin and Alert are open access and are available at the EBAS database infrastructure at NILU - Norwegian Institute for Air research: http://ebas.nilu.no/. Observations for Villum are obtained after personal communication with Henrik Skov. Observations from IMPROVE database can be obtained from: http://views.cira.colostate.edu/fed/QueryWizard/. Sub- and super-micron aerosol mass concentrations at Utqiagvik, Alaska can be obtained from the follow link: https://saga.pmel.noaa.gov/data/stations/.
Table A1. Land Surface model’s (NOAH MP) parametrization. "Opt_" indicates the namelist option for NOAH MP.

<table>
<thead>
<tr>
<th>NOAH MP Parametrization</th>
<th>On</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dynamic Vegetation (DVEG)</td>
<td>Ball-Berry [Ball et al. (1987)]</td>
</tr>
<tr>
<td></td>
<td>[Collatz et al. (1991)] [Collatz et al. (1992)]</td>
</tr>
<tr>
<td>Stomatal Resistance</td>
<td>[Bonan (1996)] [Bonan (1996)] [Sellers et al. (1996)]</td>
</tr>
<tr>
<td></td>
<td>[Collatz et al. (1991)] [Collatz et al. (1992)]</td>
</tr>
<tr>
<td></td>
<td>[Sellers et al. (1996)]</td>
</tr>
<tr>
<td>Surface layer drag</td>
<td>Original Noah [Chen et al. (1997)]</td>
</tr>
<tr>
<td>coefficient (opt_sfc)</td>
<td>[Chen et al. (1997)]</td>
</tr>
<tr>
<td></td>
<td>[Chen et al. (1997)]</td>
</tr>
<tr>
<td>Soil moisture for</td>
<td>Noah (soil moisture)</td>
</tr>
<tr>
<td>stomatal resistance (opt_btr)</td>
<td>[Chen et al. (1997)]</td>
</tr>
<tr>
<td>Runoff (opt_run)</td>
<td>TOPMODEL with groundwater [Niu et al. (2007)] [Niu et al. (2007)]</td>
</tr>
<tr>
<td></td>
<td>[Niu and Yang (2004)]</td>
</tr>
<tr>
<td>Soil permeability (opt_inf)</td>
<td>linear effects, more permeable [Yue Niue and liang Yang (2006)]</td>
</tr>
<tr>
<td></td>
<td>[Yue Niue and liang Yang (2006)]</td>
</tr>
<tr>
<td>Radiative transfer (opt_rad)</td>
<td>modified two-stream (gap = F(solar angle, 3D structure ...) &lt; 1-FVEG)</td>
</tr>
<tr>
<td>Precipitation (snow/rain) partitioning (opt_snf)</td>
<td>[Jordan (1991)] [Jordan (1991)]</td>
</tr>
<tr>
<td></td>
<td>[Jordan (1991)] [Jordan (1991)]</td>
</tr>
<tr>
<td>Soil temperature lower boundary (opt_tbot)</td>
<td>TBOT at ZBOT (8m) read from a file (original Noah)</td>
</tr>
<tr>
<td></td>
<td>[Yue Niue and liang Yang (2006)]</td>
</tr>
<tr>
<td>Soil/snow temperature time scheme (opt_sfc)</td>
<td>semi-implicit; flux top boundary condition</td>
</tr>
<tr>
<td></td>
<td>[Sakaguchi and Zeng (2009)] [Sakaguchi and Zeng (2009)]</td>
</tr>
<tr>
<td>Surface resistance to evaporation/sublimation (opt_rsf)</td>
<td>include phase change of ice</td>
</tr>
<tr>
<td>Glacier treatment (opt_gla)</td>
<td></td>
</tr>
</tbody>
</table>

1300 Appendix A

Following Monaghan et al. (2018), NOAH-MP parameter file MPTABLE.TBL has been modified, and it can be used for simulations over Alaska. These modifications improved the model’s capability to capture cold surface temperature and meteorological profiles (e.g. wind speed, relative humidity, temperature) over Alaska.

Appendix B

1305 Fuentes size-resolved sea-spray source flux
\[
\frac{dF_o}{d\log D_{p0}} = \frac{dF_p}{d\log D_{p0}} \times W = \frac{Q}{A_b} \times \frac{dN_T}{d\log D_{p0}} \times W
\]  

(Eq. B1)

where \(W(U)\) is Monahan and O’Muircheartaigh whitecap coverage, \(dF_p/d\log D_{p0}\) is the size-resolved particle flux per unit time and water surface covered by bubbles, \(D_{p0}\) is the dry diameters, \(Q\) is the sweep air flow, \(A_b\) is the total surface area covered by bubbles, \(dN_T/d\log D_{p0}\) is the particle size distribution (the sum of four log-normal modes) and is equal to:

\[
\frac{dN_T}{d\log D_{p0}} = \sum_{i=1}^{4} \frac{dN_{T,i}}{d\log D_{p0}} = \sum_{i=1}^{4} \frac{N_{T,i}}{\sqrt{2\pi} \times \log \sigma_i} \times \exp\left[-\frac{1}{2} \times \left( \frac{\log D_{p0}}{\log \sigma_i} \right)^2 \right]
\]  

(B2)

where \(i\) is the sub-index for the mode number and \(N_i, D_{p0g,i}\) and \(\sigma_i\) are the total particle number, geometric mean and geometric standard deviation for each log-normal mode. \(N_{T,i}\) and \(D_{p0g,i}\) are depending on parameters \(a_i\) and \(\beta_i\) derived from polynomial and exponential regressions defining the total particle number and geometric mean diameter of the log-normal modes, and can be found in Table 5 \textit{in Fuentes et al. (2010)}.
Table C1. Biases and RMSEs, in $\mu g m^{-3}$, are calculated for aerosols at the Alert, Canada, during January and February 2014 and for CONTROL and HEM_NEW simulations at 100 km compared to the observations. NA stands for Not available.

<table>
<thead>
<tr>
<th>Site</th>
<th>Na$^+$</th>
<th>Cl$^-$</th>
<th>NO$_3^-$</th>
<th>nss-SO$_2^{4-}$</th>
<th>nss-SO$_2^{4-}$/ss-SO$_2^{4-}$</th>
<th>NH$_4^+$</th>
<th>OA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alert</td>
<td>0.91</td>
<td>0.18</td>
<td>1.2</td>
<td>0.19</td>
<td>0.1</td>
<td>0.09/0.03</td>
<td>0.011</td>
</tr>
<tr>
<td>Villum</td>
<td>1.4</td>
<td>0.3</td>
<td>2.1</td>
<td>0.26</td>
<td>0.15</td>
<td>0.1</td>
<td>0.06/0.07</td>
</tr>
<tr>
<td>Zeppelin</td>
<td>4.4</td>
<td>0.3</td>
<td>6.5</td>
<td>0.4</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3/0.2</td>
</tr>
<tr>
<td>Utqiaqvik super-micron</td>
<td>0.37</td>
<td>0.25</td>
<td>0.48</td>
<td>0.51</td>
<td>0.3</td>
<td>0.17</td>
<td>0.05</td>
</tr>
<tr>
<td>Utqiaqvik sub-micron</td>
<td>0.66</td>
<td>0.67</td>
<td>0.361</td>
<td>0.364</td>
<td>0.162</td>
<td>0.158</td>
<td>0.88</td>
</tr>
<tr>
<td>GoA</td>
<td>0.9</td>
<td>0.3</td>
<td>1.2</td>
<td>0.3</td>
<td>0.3</td>
<td>0.2</td>
<td>0.16/0.08</td>
</tr>
<tr>
<td>Simeonof</td>
<td>2.5</td>
<td>0.6</td>
<td>3.7</td>
<td>0.7</td>
<td>0.23</td>
<td>0.2</td>
<td>0.25/0.19</td>
</tr>
</tbody>
</table>

Appendix C

In this APPENDIX, the biases and RMSEs are calculated and shown for each site, as shown in Fig. 1, and are shown in the tables below. Each table corresponds to a site and for the available observed aerosol concentrations, such as Na$^+$, Cl$^-$, SO$_2^{4-}$ (total and non-sea component), NO$_3^-$, NH$_4^+$ and OC. Bias is calculated as the difference between model simulation and observation, and aerosol component at 100 km.
Biases

| Table D1. Biases Estimated mean neutralized factor $f$ using the observations and RMSE the results from the two quasi-hemispheric simulations (100km), in $\mu$g m$^{-3}$, are calculated for aerosols at Villum Research station, Greenland, during January and February 2014 and for CONTROL and HEM_NEW simulations at 100km the different sites. $f$ is not estimated for Simeonof and GoA sites as there are not observations of NH$_4^+$.

| $\text{Na}^+$ | Alert |
| $\text{Cl}^-$ | Villum |
| $\text{NO}_3^-$ | Zeppelin |

| $\text{SO}_4^{2-}$ | 0.104 Utqiaġvik super-micron |
| $\text{Cl}$ | 6.480.72NO$_3^-$ 0.260.29NH$_4^+$ 0.0770.076SO$_4^{2-}$ 0.250.45 sub-micron |

| $\text{NO}_3^-$ | 0.230.20SO$_4^{2-}$ 0.250.26OA 0.1 |

1320 Appendix D

$=1em$ Biases and RMSEs, in $\mu$g m$^{-3}$, are calculated for aerosols at Gates of the Arctic, south of Alaska, during January and February 2014 and for CONTROL and HEM_NEW simulations at 100km. **RMSE RMSE** $\text{Na}^+$ 0.90.3Cl $\text{NO}_3^-$ 0.32 $\text{SO}_4^{2-}$ 0.20.2OA 0.280.26To investigate aerosol acidity, the mean neutralized factor ($f$) is calculated as the ratio of NH$_4^+$ to the sum of ($2\text{nss-SO}_4^{2-} + \text{NO}_3^-$), in molar concentrations, following Fisher et al. (2011), for sites in the Arctic with available observations of these aerosols. When $f$ is equal to 1 then aerosols are assumed to be neutralized, while when $f < 1$ then aerosols are acidic, and more acidic when $f$ is closer to zero (Fisher et al., 2011). In general, higher molar concentrations were observed for nss-SO$_4^{2-}$ compared to NO$_3^-$ and NH$_4^+$. Table D1 shows $f$ for observations and the two 100km simulations at the different sites. Overall, modelled $f$ increases due to the improved treatment of SSA and the associated influence on NO$_3^-$ via heterogeneous reactions. Since aerosols are assumed to be internally mixed in the model, NH$_4^+$ and nss-SO$_4^{2-}$ mass concentrations also vary between the two simulations. Thus, aerosols in HEM_NEW tend to be less acidic (e.g. at Alert and Villum), due to NO$_3^-$ decreases in the coarse-mode/TSP size range. This leads to better agreement with the observed $f$ at Alert, in particular. At Villum, observed aerosols are less acidic than in the model. This could be due to the fact that the model has more NH$_4^+$ compared to the observations. Only small changes are found at Utqiaġvik between the two runs, and the model tends to have aerosols which are slightly more acidic (super-micron) and less acidic (sub-micron) compared to the observations. The small increase in model sub-micron $f$ at Utqiaġvik could be due to the increase in sub-micron NO$_3^-$ and insignificant changes in NH$_4^+$ and nss-SO$_4^{2-}$. Differences with the observed values could be explained by underestimation of nss-SO$_4^{2-}$ at this site. The calculated $f$ for observations could also be biased low (too acidic), since some of the NO$_3^-$ and SO$_4^{2-}$ are present as Na$_2$SO$_4$ and NaN$_3$O$_3$ in the atmosphere, which are not measured.
Table E1. Biases and RMSEs, in \( \mu g m^{-3} \), are calculated between ALASKA_NEW_JAN, ALASKA_NEW_FEB and in-situ meteorological parameters derived from NOAA Baseline Observatories during the campaign’s periods in January and February 2014. Bias was calculated as the difference between model simulation and observations.

<table>
<thead>
<tr>
<th></th>
<th>January campaign</th>
<th>February campaign</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Bias</td>
<td>RMSE</td>
</tr>
<tr>
<td>2m Temperature</td>
<td>0.1</td>
<td>1.9</td>
</tr>
<tr>
<td>10m Temperature</td>
<td>-0.03</td>
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<tr>
<td>10m Wind speed</td>
<td>0.08</td>
<td>1.4</td>
</tr>
<tr>
<td>10m Wind direction</td>
<td>-11.2</td>
<td>13.2</td>
</tr>
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</table>

Biases and RMSEs, in \( \mu g m^{-3} \), are calculated for super-micron aerosols at Utqiaġvik, north of Alaska, during January and February 2014 and CONTROL and HEM_NEW simulations at 100km. RMSE RMSE Na\(^{+}\) 0.370.25Cl\(^{-}\) 0.480.51NO\(_3\)\(^{-}\) 0.30.17NH\(_4\)\(^{+}\) 0.003680.0037SO\(_2\)\(^{2-}\) 0.0050.06

Biases and RMSEs, in \( \mu g m^{-3} \), are calculated for sub-micron aerosols at Utqiaġvik, north of Alaska, during January and February 2014 and CONTROL and HEM_NEW simulations at 100km. RMSE RMSE Na\(^{+}\) 0.660.67Cl\(^{-}\) 0.3610.364NO\(_3\)\(^{-}\) 0.1620.158NH\(_4\)\(^{+}\) 0.1060.100SO\(_2\)\(^{2-}\) 0.8750.853

Appendix E

Surface observations are used to validate the meteorological conditions that occur over Utqiaġvik and Alaska in wintertime. The model is validated against the surface (hourly) observations obtained from National Oceanic and Atmospheric Administration / Earth System Research Laboratory / Global Monitoring Division (NOAA/ESRL/GMD) Baseline Observatories. Also, radiosondes data are used to evaluate the model’s performance at different altitudes. Radiosonde data (every 12h) are derived from Integrated Global Radiosonde Archive version 2 (IGRA 2) (Durre et al. (2018)). Site is located at latitude: \( 71.2889 \) and longitude: \( -156.7833 \). In the future, more detailed studies it is advisable to investigate the meteorological and synoptic conditions that occur at each site within Arctic. A detailed focus on the meteorology and removal treatments at lower latitudes to minimize potential uncertainties linked to transport errors in the model is also desirable.
Figure E1. Average temperatures, in degrees C, and wind speeds, in ms$^{-1}$, as a function of altitude (m), up to 4km, during (a,b) January and (c,d) February campaign in 2014, at Utqiagvik, Alaska. The observations are shown in black (circle). The blue pentagon shows the model results for the CONTROL simulation (at 100km) and the red diamond shows the model results for the NEW_ALASKA_JAN and NEW_ALASKA_FEB simulation. Observations are derived from IGRA2 and are available every 12h (0Z and 12Z, UTC). For the comparison, model output at 0 and 12Z UTC are used. The corresponding horizontal lines show the standard deviation.
Figure E2. Time series of observed and modelled 2m and 10m temperature, and 10m wind speed, at Utqiaġvik (Barrow, Alaska, in UTC. The observations are shown in red and derived from the NOAA observatory. The blue line shows the results for the HEM_NEW simulation at 100km, while the black line shows the results for ALASKA_NEW_JAN and ALASKA_NEW_FEB simulations at 20km. The observations are hourly, while the model output is every 3h.

Appendix F

Here the bias and RMSE are shown between ALASKA_NEW_JAN and ALASKA_NEW_FEB and the observations for Utqiaġvik at 20km.

Appendix G

This APPENDIX shows the comparison for the Gates of the Arctic site at 20 km, for ALASKA_NEW_FEB and ALASKA_CONTROL_FEB. The observations are only available for the February campaign, daily averaged in local Alaskan time every three days.

Model inter-variability during February campaign. Model simulations are validated against aerosols at the gates of the Arctic site, north of Alaska. The black line shows ALASKA_CONTROL_FEB simulation and the black symbol the daily averaged values. The red line shows ALASKA_NEW_FEB simulation and the red pentagon the daily averaged values. The blue star indicates averaged daily observations. Observations and model are in local Alaskan time. Observed and modelled $SO_2^-$ is total $SO_2^-$.
**Table F1.** Biases and RMSEs, in $\mu g m^{-3}$, are calculated for aerosols at Utqiaġvik, north of Alaska, during January 2014 and for ALASKA_CONTROL_JAN and ALASKA_NEW_JAN simulations at 20km.

<table>
<thead>
<tr>
<th></th>
<th>ALASKA_CONTROL_JAN</th>
<th>ALASKA_NEW_JAN</th>
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<tbody>
<tr>
<td></td>
<td>Bias</td>
<td>RMSE</td>
</tr>
<tr>
<td>Na$^{+}$</td>
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<tr>
<td>Cl$^{-}$</td>
<td>-0.50</td>
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<tr>
<td>NO$_3^{-}$</td>
<td>-0.040</td>
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<tr>
<td>SO$_2^{-}$</td>
<td>-0.4</td>
<td>0.440</td>
</tr>
<tr>
<td>NH$_4^{+}$</td>
<td>0.0380</td>
<td>0.040</td>
</tr>
</tbody>
</table>

**Table F2.** Biases and RMSEs, in $\mu g m^{-3}$, calculated for aerosols at Utqiaġvik, north of Alaska, during February 2014 and for ALASKA_CONTROL_FEB and ALASKA_NEW_FEB simulations at 20km.

<table>
<thead>
<tr>
<th></th>
<th>ALASKA_CONTROL_FEB</th>
<th>ALASKA_NEW_FEB</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Bias</td>
<td>RMSE</td>
</tr>
<tr>
<td>Na$^{+}$</td>
<td>-1.30</td>
<td>1.40</td>
</tr>
<tr>
<td>Cl$^{-}$</td>
<td>-1.91</td>
<td>1.92</td>
</tr>
<tr>
<td>NO$_3^{-}$</td>
<td>-0.20</td>
<td>0.40</td>
</tr>
<tr>
<td>SO$_2^{-}$</td>
<td>-1.03</td>
<td>1.321</td>
</tr>
<tr>
<td>NH$_4^{+}$</td>
<td>0.0970</td>
<td>0.10</td>
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</tbody>
</table>

Also, the table below shows biases and RMSEs, in $\mu g m^{-3}$, for all available aerosol species at the Gates of the Arctic.

<p>| | | | | |</p>
<table>
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</thead>
<tbody>
<tr>
<td></td>
<td>Bias</td>
<td>RMSE</td>
<td>Bias</td>
<td>RMSE</td>
</tr>
<tr>
<td>Na$^{+}$</td>
<td>0.346</td>
<td>0.341</td>
<td>0.157</td>
<td>0.157</td>
</tr>
<tr>
<td>Cl$^{-}$</td>
<td>0.48</td>
<td>0.47</td>
<td>0.240</td>
<td>0.230</td>
</tr>
<tr>
<td>NO$_3^{-}$</td>
<td>0.244</td>
<td>0.244</td>
<td>0.157</td>
<td>0.157</td>
</tr>
<tr>
<td>SO$_2^{-}$</td>
<td>0.137</td>
<td>0.137</td>
<td>0.157</td>
<td>0.157</td>
</tr>
</tbody>
</table>
Author contributions. The first author (EI) implemented the updates, performed the simulations and the analysis, and drafted the paper. KSL designed the study and contributed to the interpretation of results and the analysis, and to the writing of the paper. JCR, LM and TO contributed to discussions about the model setup and simulations. JCR, LM, KP, RMK, PKQ and LU contributed to the analysis and interpretation of the results. AM and HS contributed to the interpretation of the results. All co-authors contributed to the paper and the discussions about the results, as well as for the revision of the manuscript and have approved the final version.

Competing interests. The authors declare that they have no conflict of interest.

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