

Final Report for “Simulating the Arctic Winter Longwave Indirect Effects: A New Parameterization for Frost Flower Aerosol Salt Emissions” (DE-SC0006679) for 9/15/2011 through 9/14/2015

1. DOE Award Number: DE- SC0006679.

Name of Recipient: Scripps Institution of Oceanography, University of California, San Diego.

Project Title: Simulating the Arctic Winter Longwave Indirect Effects: A New Parameterization for Frost Flower Aerosol Salt Emissions

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Co-PI: Richard C.J. Somerville (Scripps-UCSD).

Collaborators: Susannah Burrows (PNNL), Phil Rasch (PNNL).

2. There are no authorized distribution limitation notices.

3. Executive Summary

Description of the Project: This project has improved the aerosol formulation in a global climate model by using innovative new field and laboratory observations to develop and implement a novel wind-driven sea ice aerosol flux parameterization. This work fills a critical gap in the understanding of clouds, aerosol, and radiation in polar regions by addressing one of the largest missing particle sources in aerosol-climate modeling. Recent measurements of Arctic organic and inorganic aerosol indicate that the largest source of natural aerosol during the Arctic winter is emitted from crystal structures, known as frost flowers, formed on a newly frozen sea ice surface [Shaw et al., 2010]. We have implemented the new parameterization in an updated climate model making it the first capable of investigating how polar natural aerosol-cloud indirect effects relate to this important and previously unrecognized sea ice source. The parameterization is constrained by Arctic ARM *in situ* cloud and radiation data. The modified climate model has been used to quantify the potential pan-Arctic radiative forcing and aerosol indirect effects due to this missing source. This research supported the work of one postdoc (Li Xu) for two years and contributed to the training and research of an undergraduate student. This research allowed us to establish a collaboration between SIO and PNNL in order to contribute the frost flower parameterization to the new ACME model. One peer-reviewed publication has already resulted from this work, and a manuscript for a second publication has been completed. Additional publications from the PNNL collaboration are expected to follow.

How this Research Adds to the Understanding of Climate Change:

Understanding climate change on regional scales requires a broad scientific consideration of anthropogenic influences that goes beyond greenhouse gas emissions to also include the effects of aerosol. . Incorporation of this missing sea ice aerosol source has markedly improved our understanding and ability to simulate high-latitude cloud-aerosol-radiation interactions on multiple time scales. The Arctic is Earth's most sensitive region to changes in the atmospheric composition of greenhouse gases, exhibiting nearly twice the global mean surface warming in response to industrial-era anthropogenic emissions [IPCC - Solomon et al., 2007]. This project has made significant scientific progress by investigating how frost flower particles in the Arctic region can be used to improve prediction of how aerosols, clouds, and climate will mutually interact to produce climate change at regional scales in response to anthropogenic forcing.

4. Comparison of the Actual Accomplishments with the Objectives

The *project objectives* were to improve scientific models about the potential response of the Earth's climate to increased greenhouse gas levels by:

1. *Developing and implementing a frost flower aerosol source parameterization, consisting of three tasks:*
 - a. *Innovative observational data analyses of Arctic winter aerosol*
 - b. *Mechanistic aerosol source parameterization for Global Climate Models*
 - c. *Parameterization evaluation and validation*

The actual accomplishment of the project include the development of the regional modeling development and implementation, as described by Xu et al. [2013], and the global modeling development and implementation, as described by Xu et al. [2016].

2. *Identify and interpret the Arctic aerosol-cloud interactions and climate implications, consisting of three types of studies:*
 - a. *Constrain the spatial extent of this new aerosol source*
 - b. *Investigate life cycles of clouds and aerosols and their interactions*
 - c. *Elucidate a possible cyrosphere-atmosphere feedback through aerosol emissions*

The actual accomplishment of the project include the identification and interpretation of cloud-climate interactions and climate implications in the regional model simulations, as described by Xu et al. [2013], and in the global model simulations, as described by Xu et al. [2016].

Overall, the project accomplished more than originally proposed because of synergies and collaborations with other projects. The regional modeling provided an impressive test bed to show local effects of frost flowers at the Barrow site and provided an expanded code base for use in future studies. The global modeling provided a long-term evaluation of the pan-Arctic effects of frost flowers and contributed to the development of important new modeling capabilities for Earth system modeling studies.

This project made extensive use of long-term measurements at the DOE ARM NSA site at Barrow, Alaska. The radiative measurements at this site provided ground-based validation of the calculated surface fluxes. This critical observational record allowed for validation of model parameterizations.

The incorporation of frost flowers in CESM in coordination with HiLAT is an advance was well beyond what was proposed, but will clearly be a lasting impact from our project as well as a dramatic extension of the HiLAT project. The no-cost extension allowed for critical overlap in time with the newly-started HiLAT project, and the resulting additional manuscript documents the advances made possible by this joint effort.

5. Summary of Project Activities

This research supported the work of one postdoc (Li Xu) for two years and contributed to the training and research of a graduate student (Jun Liu) and an undergraduate student (Allison Khoe). This research allowed us to establish a collaboration between SIO and PNNL in order to contribute the frost flower parameterization to the new ACME model. One peer-reviewed publication has already resulted from this work, and a manuscript for a second publication has been completed. Additional publications from the PNNL collaboration are expected to follow.

Completed Studies

Xu et al. [JGR, 2013] used a regional model to show the aerosol-cloud effects of frost flowers, clusters of highly saline ice crystals growing on newly formed sea ice or frozen lakes. Observations of particles derived from frost flowers in the Arctic were used to formulate an observation-based parameterization of salt aerosol source function from frost flowers. The particle flux from frost flowers in winter has the order of $10^6 \text{ m}^{-2} \text{ s}^{-1}$ at the wind speed of 10 m s^{-1} , but the source flux is highly localized to new sea ice regions and strongly dependent on wind speed. Xu et al. [2013] implemented this parameterization into the regional Weather Research and Forecasting model with Chemistry initialized for two wintertime scenarios. The addition of sea salt aerosol emissions from frost flowers increases averaged sea salt aerosol mass and number concentration and subsequent cloud droplet number. This change of cloud droplet number concentration increases downward longwave cloud radiative forcing through enhanced cloud optical depth and emissivity. The magnitude of this forcing of sea salt aerosols from frost flowers on clouds and radiation, however, contributes negligibly to surface warming in Barrow, Alaska, in the wintertime scenarios studied here.

Xu et al. [JGR, 2016] implements an observationally-based parameterization for estimating sea salt production from frost flowers in the Community Earth System Model (CESM). This work evaluates the potential influence of this sea salt source on the pan-Arctic (60°N-90°N) climate. Results show that frost flower salt emissions substantially increase the modeled surface sea salt aerosol concentration in the winter months when new sea ice and frost flowers are present. The parameterization reproduces both the magnitude and seasonal variation of the observed submicron sea salt aerosol concentration at surface in Barrow during winter much better than the standard CESM simulation without a frost-flower salt particle source. Adding these frost flower salt particle emissions increases aerosol optical depth by 10% and results in a small cooling at surface. The increase in salt particle mass concentrations of a factor of 8 provides nearly 3 times the cloud condensation nuclei concentration, as well as 10% increases in cloud droplet number and 40% increases in liquid water content near coastal regions adjacent to continents. These cloud changes reduce longwave cloud forcing by 4% and cause a small surface warming, increasing the downward

longwave flux at the surface by 2 W m^{-2} in the pan-Arctic under the present-day climate.

Continuing Studies

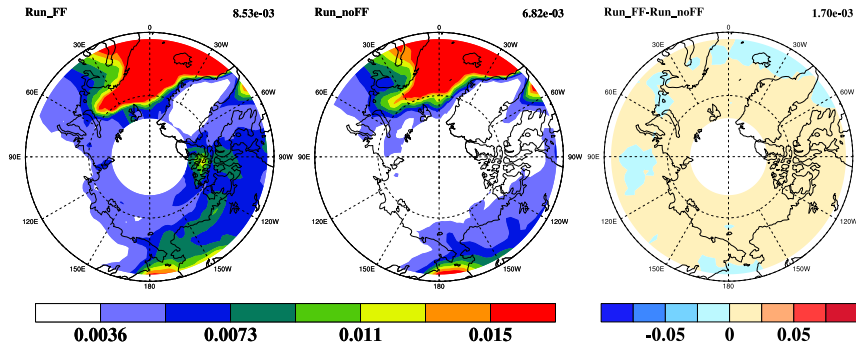
Collaborations with Susannah Burrows of PNNL will continue as part of her work on the recently funded HiLAT project. In addition, as part of the new DOE-ARM project AWARE, we will assess the contributions of frost flowers to aerosol particles at McMurdo station.

Other Funding and Activities that Contributed to this Project

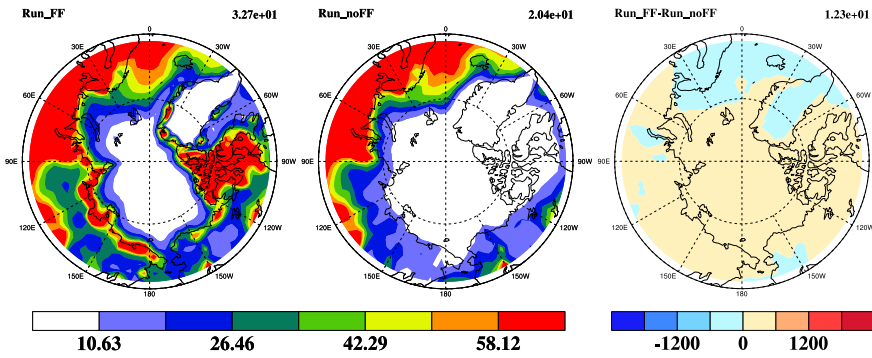
We have been able to achieve an important degree of synergy between this DOE project and ACME-related research at PNNL, including the HiLAT project.

Figures

a) Aerosol Optical Depth for Sea Salt



b) Accumulation mode aerosol number concentration



c) CCN at 0.1% Supersaturation

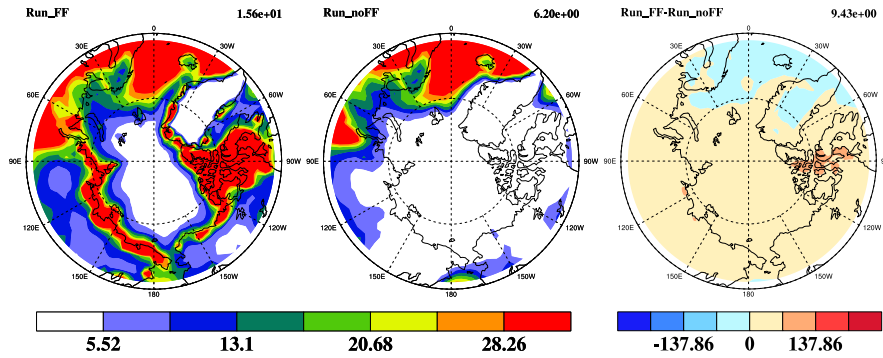


Figure 1: Wintertime (i.e., NDJF) averaged variables from the FF and noFF simulations and their absolute difference for: a) sea salt aerosol optical depth; b) accumulation mode aerosol number concentration ($\#/cm^3$); c) cloud condensation nuclei ($\#/cm^3$) at 0.1% supersaturation at surface at 936 hPa. The mean values averaged in the pan-Arctic region are shown in the right corner of each panel. Figure modified from Xu et al. [2016].

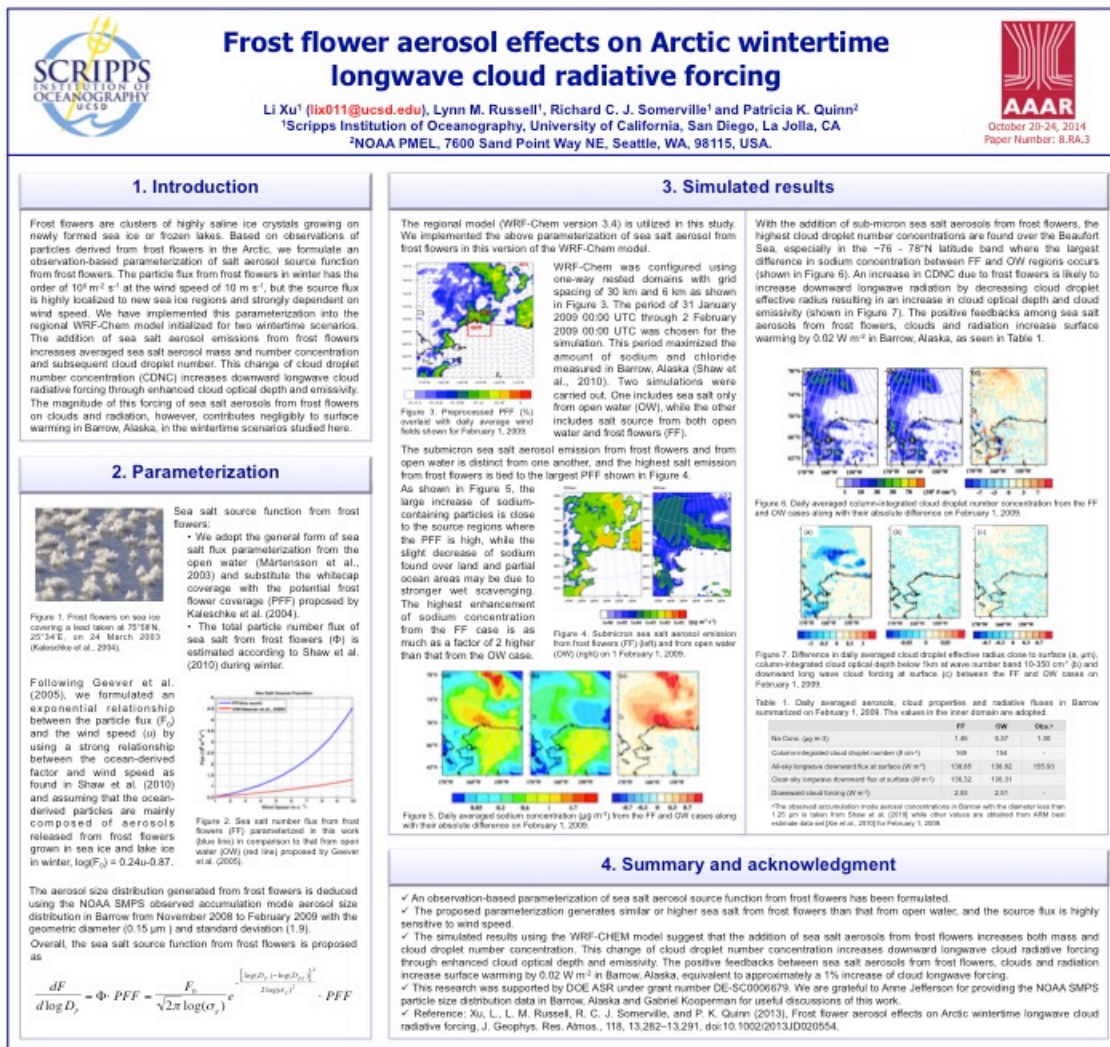


Figure 2: Poster presentation by Xu et al. at American Association for Aerosol Research Fall Meeting, 23 October 2014.

6. Products developed under the award: Publications, conference papers, or other public releases of results.

Publications:

- Xu, L., L. M. Russell, R. C. Somerville, and P. K. Quinn (2013), Frost flower aerosol effects on Arctic wintertime longwave cloud radiative forcing, *J. Geophys. Res. Atmos.*, 118, 13,282–13,291, doi:10.1002/2013JD020554.
- Xu, L., L. M. Russell, S. M. Burrows (2016), Potential aerosol sources from frost flowers in the pan-Arctic region, *J. Geophys. Res.*, in internal review.

Conference papers

- Xu, L., L. M. Russell, R. C. Somerville, and P. K. Quinn, Frost flower aerosol effects on Arctic wintertime longwave cloud radiative forcing, Atmospheric Sciences Research Meeting, Bolger Center, Potomac, Maryland, 11 March 2014.
- Xu, L., L. M. Russell, R. C. Somerville, and P. K. Quinn, Frost flower aerosol effects on Arctic wintertime longwave cloud radiative forcing, American Association for Aerosol Research Fall Meeting, 23 October 2014.

7. For projects involving computer modeling:

a. Model description, key assumptions, version, source and intended use

Our frost flower project aims to study the importance of sea salt aerosol productions from frost flowers on atmospheric aerosol abundance as well as their interaction with clouds and regional or global climate during the Arctic winter. To achieve this goal, we used one regional model, the Weather Research and Forecasting Model with Chemistry version 3.4 (WRF-Chem); one global model, the Community Earth System Model version 1.2.1 (CESM).

Both WRF-Chem and CESM models are uncontroversial, widely validated research tools with proven track records in the literature. WRF is a regional numerical weather prediction model developed through a multi-institutional endeavor headed at NCAR and is used for both forecasting and climate research applications. WRF-Chem is a modified version of the WRF model, which includes online chemistry, capable of simulating aerosol-cloud-radiation direct and indirect interactions at cloud-resolving scales. The chemistry component of the WRF model is described by *Grell et al.* [2005] and *Fast et al.* [2006]. The CESM model is a global climate model, composed of atmosphere, ocean, land surface, and sea ice components. A full scientific description of CESM is given by Hurrell et al. [2013].

In this work, we developed the regional model, WRF-Chem, with the implementation of the proposed observationally-based frost flower sea salt source productions to show the aerosol-cloud effects of frost flowers for two wintertime scenarios as described by Xu et al. [2013]. Additionally, we extend two local cloud events evaluated by Xu et al. [2013] to look at seasonal trends by adapting the same observationally-based parameterization for estimating sea salt production from frost flowers to the CESM. The CESM framework provides the opportunity to evaluate the potential influence of this sea salt source on aerosol-cloud interactions in the pan-Arctic (60°N-90°N) region.

b. Performance criteria for the model related to the intended use

Both WRF-Chem and CESM models used in this project have been validated throughout their development (WRF-Chem: *Grell et al.* [2005]¹, *Fast et al.* [2006]; CESM: Hurrell et al. [2013]).

c. Test results to demonstrate the model performance criteria were met

The performance of sea salt aerosol representations in WRF and CESM models were validated with respect to sea salt budgets including sources, sinks, burden and their residence time in the atmosphere in the pan-Arctic region as well as the entire globe in Xu et al. [2013, 2016]. The global sea salt budget is in accordance with the previous reference study of Liu et al. [2012].

¹ A list of WRF-Chem and CESM studies comparing model results to observations are at http://ruc.noaa.gov/wrf/WG11/References/WRF-Chem.references_July2012.htm and <http://www.cesm.ucar.edu/publications/>.

The primary references for test results are:

Xu, L., L. M. Russell, S. M. Burrows (2016), Potential aerosol sources from frost flowers in the pan-Arctic region, *J. Geophys. Res.*, in internal review.

Xu, L., L. M. Russell, R. C. Somerville, and P. K. Quinn (2013), Frost flower aerosol effects on Arctic wintertime longwave cloud radiative forcing, *J. Geophys. Res. Atmos.*, 118, 13,282–13,291, doi:10.1002/2013JD020554.

d. Theory behind the model, expressed in non-mathematical terms

Recent measurements of Arctic organic and inorganic aerosol indicate that the largest source of natural aerosol during the Arctic winter is emitted from crystal structures, known as frost flowers, formed on a newly frozen sea ice surface [Shaw et al., 2010]. There is not a single parameterization proposed to estimate sea salt aerosol productions from frost flowers. No one has used numerical models (including regional or global models) to study the interaction among aerosol, clouds, and radiation in polar regions due to this “missing” sea salt-containing wintertime particle source. Most global climate models (GCMs) include sea salt aerosol production from open water regions only. Two GCMs [Piot and Glasow, 2008; Mahowald et al., 2006] have added frost flower salt production by applying a sea salt source function from open water to regions of sea ice as a rough first approximation. Xu et al. [2013; 2016] fill a critical gap in the understanding of clouds, aerosol, and radiation in polar regions by addressing one of the largest missing particle sources in aerosol-climate modeling.

e. Mathematics to be used, including formulas and calculation methods

For mathematical details regarding the regional and global model used in this project, see *Grell et al.* [2005] for WRF-Chem and Hurrell et al. [2013] for CESM.

The WRF-Chem model was configured with aerosol treatments of the Model for Simulating Aerosol Interaction and Chemistry (MOSAIC) [Zaveri et al., 2008] with eight size bins to represent the aerosol size distribution [Fast et al., 2006]. Each bin is assumed to be internally mixed so that all particles within a bin are assumed to have the same chemical composition. Both mass and number are simulated for each size bin. Sea salt emissions from open oceans are computed online using fluxes derived from the predicted surface winds and boundary layer quantities as described by Gong et al. [2002]. The potential frost flower coverage (PFF) on which frost flowers grow was preprocessed and calculated using satellite-derived sea ice fraction from 0.5x0.5 degree gridded National Snow and Ice Data Center (NSDIC) SSM/I passive microwave data [Maslanik and Stroeve, 1999] along with the 1x1 degree gridded National Centers for Environmental Prediction (NCEP) reanalysis surface air temperature [Kalnay et al., 1996].

The CESM model uses a modal aerosol module with three size classes (MAM3), including the Aitken mode (dry diameter size range of 0.015-0.053 μm), accumulation mode (0.058-0.27 μm), and coarse mode (0.80-3.65 μm). For each

mode, the model predicts mass mixing ratios of internally mixed aerosol species and the total number mixing ratio. Aerosol particles outside cloud droplets (interstitial aerosols) and those within cloud droplets are tracked separately. Sea salt aerosols are calculated online as functions of wind speed and temperature. Sea salt particles do not act as good ice nuclei in this CESM version. Surface emission of open water sea salt small particles (diameter $<2.8 \mu\text{m}$) follows the method of Mårtensson et al. [2003], while larger particles (diameter $>2.8 \mu\text{m}$) follow the method of Monahan et al [1986]. For the sea salt particle production from frost flowers, we employ the observationally-based parameterization developed by Xu et al. [2013] in the accumulation mode with dry diameter size range of $0.058\text{-}0.27 \mu\text{m}$ in the MAM3 in the CESM model.

Following the formulation of sea salt aerosol production from breaking waves in open water [Martensson et al., 2003], we proposed a general form of the sea salt particle flux (F) from frost flowers, as follows.

$$\frac{dF}{d\log(D_p)} = \frac{\Phi \cdot e^{-\frac{[\log(D) - \log(D_g)]^2}{2\log(\sigma_g)^2}}}{\sqrt{2\pi} \log(\sigma_g)} \cdot PFF (m^2 s^{-1}), \quad (1)$$

where Φ is the total particle number flux produced per surface area of frost flowers per second and PFF stands for the potential frost flower coverage defined and parameterized in Kaleschke et al. [2004]. Following Geever et al. [2005], we formulated an exponential relationship between the particle flux and wind speed (u) using the relationship between the ocean-derived factor and wind speed identified in Shaw et al. [2010] and assuming that the ocean-derived particles are released from frost flowers grown in sea ice in winter,

$$\Phi = e^{0.24u - 0.84}, \quad (2)$$

We adopted the magnitude of the source flux of $10^6 \text{ m}^2 \text{ s}^{-1}$ [Geever et al. 2005], since Rankin et al. [2000] suggested the salt production from frost flowers has a similar magnitude to that from open water. Kaleschke et al. [2004] introduced PFF as a proxy for predicting the maximum surface area from which frost flowers may form on sea ice using a one-dimensional thermodynamic model of sea ice and the frost flower growth. The equations relevant to the calculation of PFF are listed below.

$$T_0 = \frac{-15.96 + \theta^{0.53} T_a}{8.4 + 1.33\theta^{0.53}} \quad (3)$$

$$g = 0.000785 e^{0.478(T_0 - T_a)} \quad (4)$$

$$F_t = F_{t-\delta t} + g(1 - F_{t-\delta t})\delta t \quad (5)$$

$$PFF = F_{max}(T_a)(1 - f_{seaice}) \quad (6)$$

where T_0 and T_a are sea ice surface temperature and air temperature,

respectively. θ is cumulative freezing days, $\theta = \int (T_f - T_a) dt$, where T_f is the freezing point of sea water, $T_f = -1.9^\circ\text{C}$. g is the frost flower growth rate in unit of hour^{-1} adopted from Martin et al. [1996] while F_t is the area coverage at a time step δt . As the frost flower growth rate decreases with the increase of sea ice thickness, the recursive equations in Eqn.(3)-(6) yield a maximum area coverage $F_{max}(T_a)$ for a given surface air temperature T_a and integration time (e.g., 5 days used in this work). Thus, PFF is obtained by weighting $F_{max}(T_a)$ with the potential new ice fraction, which is assumed to equal the open water fraction (that is one minus sea ice fraction) on which thin new ice form rapidly and subsequently frost flowers can grow.

In CESM, the potential frost flower (PFF) coverage is calculated in the CESM atmosphere module (CAM5) collecting prognostic sea ice fraction from the CESM ocean component (POP2). Assuming a frost flower lifetime of up to five days, the PFF coverage for each day was computed by integrating the values from the current day and four previous days. Kaleschke et al. [2004] showed that the largest difference in PFF coverage is less than 10% when the model integration time (i.e., the lifetime of frost flowers) varies from one day to five days at a surface air temperature of -40°C .

f. Whether or not the theory and mathematical algorithms were peer reviewed, and, if so, include a summary of theoretical strengths and weaknesses

The frost flower algorithms are peer-reviewed for publication in Xu et al. [2013; 2016]. In addition, both WRF-Chem and CESM have been extensively peer reviewed. Ongoing lists of select publications for WRF and CESM can be found at:

http://ruc.noaa.gov/wrf/WG11/References/WRF-Chem.references_July2012.htm
<http://www.cesm.ucar.edu/publications/>

g. Hardware requirements

The WRF-Chem model was integrated using distributed resources available through the National Science Foundation's XSEDE network. We ran the code on a Cray XT5 at the University of Tennessee's. WRF-Chem was configured with two nested domains with resolutions of 30 km and 6 km for domains 1 and 2, respectively. The coarsest grid (Domain 1) extended approximately from latitudes 60°N to 80°N and from 175°W to 140°W , covering northern Alaska, the Bering Strait, and the Beaufort Sea. Domain 2 was centered near Barrow. Both domains extended 27 layers in the vertical, from the surface to 100 hPa, with finer resolution near the surface. Under this configuration, the computational expense per one-day simulation was approximately 1 CPU-hours for WRF-Chem at 96 processor scale.

The CESM model was integrated using distributed resources available through the National Energy Research Scientific Computer Center (NERSC). We ran the code on a Cray XE6 petaflop system in the NERSC. In all experiments, CAM5, the atmospheric component of CESM, was configured at $1.9^\circ \times 2.5^\circ$ horizontal resolution with 30 hybrid vertical levels while the ocean/sea ice component of the

CESM, was configured in a grid of approximately 1-degree resolution (i.e., the grid of gx1v6). Under this configuration, and after optimization on our system architecture, the computational expense per year simulation was roughly 17 CPU-hours for the CESM run at 3456 total processor scale with 676 processors assigned to 6 components including atmosphere, ocean, land, sea ice, land-ice and the coupler.

Both WRF-Chem and CESM models have excellent check pointing and restart-capability such that long or expensive integrations could be reliably separated into shorter segments.

h. Documentation (e.g., user guide, model code)

The frost flower code modules for WRF (MOSAIC) and CESM are available at the project web sites.

The WRF user guide and model code can be found at <http://www.wrf-model.org>. The CESM user guide and model code are available on the NCAR website at <http://www.cesm.ucar.edu>.

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