Impacts of New Particle Formation on Short-term Meteorology and Air Quality as Determined by the NPF-explicit WRF-Chem in the Midwestern United States

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ABSTRACT

New particle formation (NPF) from nucleation and subsequent nuclei growth, which is frequently observed in the troposphere, is critical to aerosol-cloud interactions yet difficult to simulate. In this work, regional simulations with the fully coupled NPF-explicit WRF-Chem model link NPF to cloud properties and to changes in both meteorology and air quality in the U.S. Midwest during summer 2008. Simulations that include NPF have higher concentrations of condensation nuclei, as anticipated from the particle production associated with nucleation, leading to enhanced concentrations of cloud condensation nuclei (CCN) at high supersaturations. However, the online-coupled model develops a number of unexpected features that can be traced to a feedback loop involving aqueous (in-cloud) oxidation of sulfur combined with boundary layer NPF. Simulations with NPF (relative to simulations without) exhibit reduced PM_{2.5} sulfate mass, cloud dimming (reductions in the cloud frequency, CCN concentration at a low supersaturation, cloud optical depth, and cloud droplet number concentration), and enhanced surface-reaching shortwave radiation. This effect of NPF on the PM_{2.5} mass is mostly absent for other constituents of PM_{2.5}. The implications of this feedback loop, which is not considered in most climate and air quality modeling, are discussed.

Keywords: New particle formation; Meteorology; Air quality; Sulfate; WRF-Chem; Aerosol cloud interaction; Indirect climate effect.

INTRODUCTION

Atmospheric particles result from two source types: direct emissions or nucleation. Nucleation is a gas-to-particle conversion process which has been observed frequently throughout the world (Zhang *et al.*, 2011; Kerminen *et al.*, 2012; Yue *et al.*, 2016), and is a dominant source of particle number in the global atmosphere (Spracklen *et al.*, 2006; Merikanto *et al.*, 2009). Clusters of particles around 1 nm in size are introduced by nucleation (Kulmala *et al.*, 2000). Those clusters may grow via condensation to sufficiently large sizes (~100 nm) to serve as cloud condensation nuclei (CCN) (Merikanto *et al.*, 2009). Enhanced concentrations of CCN modify cloud amount, cloud optical properties, and precipitation efficiency of clouds and can thus impact climate (Twomey, 1977; Albrecht, 1989; Pincus and Baker, 1994; Paasonen *et al.*, 2013; Farmer *et al.*, 2015). Studies show

that up to 50% of global CCN are attributable to nucleation (Merikanto *et al.*, 2009; Yu and Luo, 2009; Wang and Penner, 2009). Detailed quantification of how new particle formation (NPF) affects climate remains uncertain, and the magnitude of aerosol indirect forcing is uncertain (Moore *et al.*, 2013; Fan *et al.*, 2016; Seinfeld *et al.*, 2016). While the influence of NPF in altering particle number and CCN concentrations is widely acknowledged (Yu and Luo, 2009; Gordon *et al.*, 2017), the potential feedbacks from NPF to other variables such as speciated aerosol mass, precipitation, and cloud liquid water content have not been investigated as widely and this work aims to close this research gap.

Two important characteristics of nucleation modeling studies are spatial scale (i.e., global or regional), and degree of coupling and feedbacks. Possible feedbacks that can be considered include those between NPF, clouds, radiation, aqueous aerosol chemistry, trace gases, and other meteorological variables.

Relative to global models, regional models are able to accommodate more computationally demanding process treatments and finer spatial resolutions; therefore, they are often used in conjunction with field campaigns and to assess the importance of various model processes and process

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feedbacks, often with a focus on the contribution of NPF to CCN. See for example Luo and Yu (2011) who used WRF-Chem + APM over the eastern U.S., Matsui *et al.* (2011, 2013) with the NPF-explicit WRF-Chem over East Asia, as well as other applications of NPF-explicit WRF-Chem variants (Cui *et al.*, 2014; Lupascu *et al.*, 2015). Similar to the aforementioned papers, our work *includes* assessment of the impact of NPF on CCN at regional scale using nucleation explicit WRF-Chem, but then *extends* beyond this metric to examine NPF's impact on a range of other climate- and health-relevant meteorological and air quality variables.

With respect to treatment of feedbacks, an important class of models are those with employing “offline” modeling systems; in other words, meteorology is prescribed and feedbacks between NPF aerosols, meteorology, clouds, and trace gases relevant to air quality are either limited or not considered at all.

More relevant to our aims of understanding the impact of feedbacks between NPF, clouds, and air quality are online coupled models. Several studies have investigated aerosol-meteorology interactions in the U.S., Europe and Asia by using the online WRF-Chem model (Zhang *et al.*, 2010b; Yang *et al.*, 2011; Shrivastava *et al.*, 2013; Berg *et al.*, 2015; Chen *et al.*, 2015; Forkel *et al.*, 2016; Liu *et al.*, 2016), often with a focus on direct and indirect effects of anthropogenic aerosols; they *include* nucleation, but do not *focus* on nucleation. One reason for this is the lack of aerosol representation below 39 nm in MOSAIC (the Model for Simulating Aerosol Interactions and Chemistry), the most detailed aerosol module in the WRF-Chem public release (Zaveri *et al.*, 2008). Furthermore, MOSAIC's binary nucleation parameterization underestimates PBL nucleation rates (Wexler *et al.*, 1994; Lucas and Akimoto, 2006; Zhang *et al.*, 2010a). Therefore, modifications to WRF-Chem are required for more detailed study of NPF impacts and to compare WRF-Chem to ultrafine and nuclei aerosol size distributions. The 20 size bin version of MOSAIC used in this paper (Matsui *et al.*, 2011) ranges from the lower bin boundary of 1 nm to upper bin boundary of 10,000 nm and was coupled to WRF-Chem as the NPF-explicit WRF-Chem.

Nearly all nucleation studies report an increase in CCN due to NPF. Combining this with the first indirect effect (Twomey, 1977) and the second indirect effect (Albrecht, 1989), it is commonplace to associate NPF with cloud brightening, extended cloud lifetime, and negative radiative forcing. However, more complex relationships are possible at regional scale (Rosenfeld *et al.*, 2008; Karydis *et al.*, 2011; Kerminen *et al.*, 2012; Sullivan *et al.*, 2018).

As an example where a fully coupled regional model can give counterintuitive results, Sullivan *et al.* (2018) used the NPF-explicit WRF-Chem model to study the impact of NPF on cloud radiative properties over the Midwestern U.S. for May 2008. Contrary to prior expectations, they found NPF decreased cloud albedo, resulting in positive radiative forcing relative to simulations excluding NPF.

As in Sullivan *et al.* (2018), the coupled modeling system used in this work allows exploration of feedbacks among aerosols, trace gases relevant to air quality, and

clouds over the continental U.S. in summer, providing an opportunity for replication of features reported by Sullivan *et al.* (2018), and adding tests for the roles of aqueous chemistry and biogenic secondary aerosol in the cloud dimming feedback process.

Furthermore, the NPF-explicit WRF-Chem model allows us to quantify the effects of nucleation on air quality, a topic that very few groups have reported on. The lack of documented results regarding interplay between NPF and air quality is largely due to extensive representation in the air quality literature by offline models at both regional and global scales (Merikanto *et al.*, 2009; Yu and Luo, 2009; Jung *et al.*, 2010; Yu and Hallar, 2014; Scott *et al.*, 2015; Baranizadeh *et al.*, 2016; Croft *et al.*, 2016; Gordon *et al.*, 2017). Some models couple aerosols to various model processes, but detailed assessment of processes in moderate to high SO₂ regions are typically not reported; furthermore, differences in spatio-temporal resolutions, emissions, chemical process treatments, and aerosol size resolution make replication in higher resolution regional models useful. See, for example, Wang and Penner (2009), Kazil *et al.* (2010), Wang *et al.* (2011), Wang *et al.* (2013), Matsui and Mahowald (2017) and model intercomparisons such as Quaas *et al.* (2009), Myhre *et al.* (2013, 2017), Mann *et al.* (2014), and Tsigaridis *et al.* (2014).

In this work, we use the NPF-explicit WRF-Chem model to quantify the effects of nucleation on regional meteorology and air quality, including the modulating influence of aqueous chemistry. The primary analysis area is the Midwestern U.S., a 12-state region mainly covered by agricultural lands interspersed with forests, and home to 21% of the U.S. population. The work is relevant to understanding aerosol-cloud interactions and modeling feedbacks between NPF, air quality, and meteorology in similar mid-latitude temperate agricultural regions with moderate PM_{2.5} (e.g., 5–15 µg m⁻³) and SO₂ (e.g., 0.5–3 ppb). A number of measurement studies are available to complement model-based studies of aerosols and clouds in the region (Pryor *et al.*, 2010; Sheridan *et al.*, 2012; Asmi *et al.*, 2013; Wagner *et al.*, 2015; Toon *et al.*, 2016; Bullard *et al.*, 2017).

METHOD

WRF-Chem Modeling System

NPF-explicit WRF-Chem, as described in Matsui *et al.* (2011), was applied to this study without modification (settings were modified as explained below). Key features of NPF-explicit WRF-Chem include extension of the MOSAIC aerosol module (Zaveri *et al.*, 2008) to allow 20 size bins from 1 nm to 10,000 nm, implemented in WRF-Chem 3.1.1 (Grell *et al.*, 2005; Fast *et al.*, 2006). Activation nucleation and binary nucleation parameterizations are used in the PBL and FT, respectively. The formation rate of activated clusters at 1 nm in the PBL (J) is calculated in the model by $J = A \times [\text{H}_2\text{SO}_4]$, where A is the empirical rate coefficient (s⁻¹) and $[\text{H}_2\text{SO}_4]$ is gas-phase sulfuric acid concentration. The prefactor variable A is set to 10⁻⁷ s⁻¹ for this study, consistent with previously published values ranging from

10^{-8} to $2 \times 10^{-5} \text{ s}^{-1}$ (Kuang *et al.*, 2008; Cui *et al.*, 2014; Sullivan *et al.*, 2018).

In the NPF-explicit WRF-Chem employed in this study, aerosol particles influence grid-resolved cloud properties explicitly because size-resolved aerosols are an input to the Lin microphysics scheme and act as cloud nuclei (Lin *et al.*, 1983). CCN are determined by the model at 6 supersaturations (0.02%, 0.05%, 0.1%, 0.2%, 0.5%, and 1.0%) via Köhler theory, using particle number, size and composition. Particle activation is calculated by comparing ambient supersaturation with calculated critical supersaturation (Abdul-Razzak and Ghan, 2000). When cloud droplets evaporate, the model returns activated aerosol to the interstitial phase. The simulated cloud droplet number distribution is used by the Goddard scheme to calculate incoming solar radiation (Chou and Suarez, 1994). Cloud droplet number and cloud water mixing ratio are used for computing effective cloud-droplet size and COD. Sub-grid cumulus clouds in this study are simulated by the Grell-3D parameterization (Grell *et al.*, 2005). Cumulus cloud properties are influenced through meteorological feedbacks by aerosols indirectly as aerosol activation is only simulated in the resolved clouds in this work. In more recent public releases of WRF-Chem (beginning with version 3.8), aerosols can be online coupled with the convective

parameterization (Berg *et al.*, 2015).

Model and emissions configurations are listed in Table 1. No biomass burning emissions are included in the simulations. A $36 \times 36 \text{ km}$ resolution domain over the continental U.S. is used with 34 vertical layers on top at about 16 km above ground level. An area corresponding to the Midwestern U.S. (79.5°W – 103.5°W , 36.5°N – 48.5°N) comprises a data analysis sub-region. The simulation period covers 20 June to 22 July 2008, with 3 days for model spin-up.

Numerical Experiments

Table 2 lists the primary numerical experiments performed in this study. In case N11A1, both PBL and FT nucleation are included, and aqueous chemistry is considered as well, and N11A1 is the base simulation. In case N01A1, only FT nucleation is included, while in the N00A1 case, both PBL and FT nucleation are turned off. For these three cases, the simulations are run continuously for the entire modeling period without reinitialization. N11A0 and N01A0 cases are one-week simulations (excluding 3-day spin-up) evaluating sensitivity to aqueous chemistry from 20 June to 30 June. Sensitivity tests are performed in two additional scenarios to explore the impact of model resolution and maximum supersaturation (S_{max}).

Table 1. Model configuration.

Atmospheric processes	Model option	Reference
Meteorology		
Longwave radiation	RRTM	Mlawer <i>et al.</i> (1997)
Shortwave radiation	Goddard	Chou <i>et al.</i> (1994)
Land surface model	Noah LSM	Chen and Dudia (2001)
PBL scheme	YSU	Hong <i>et al.</i> (2006)
Cumulus	Grell 3D	Grell and Devenyi (2002)
Cloud microphysics	Lin	Lin <i>et al.</i> (1983)
Initial and boundary conditions	NCEP FNL (Final) Operational Global Analysis data	https://rda.ucar.edu/datasets/ds083.2/
Chemistry		
Gas-phase chemistry	CBMZ	Zaveri and Peters (1999)
Photolysis	Fast- J	Wild <i>et al.</i> (2000)
Aerosol condensation	MOSAIC-4	Zaveri <i>et al.</i> (2008)
Aerosol coagulation	COAGSOLV	Jacobson <i>et al.</i> (1994)
Aqueous-phase chemistry	Bulk approach	Fahey and Pandis (2001)
Anthropogenic emissions	2005 NEI	
Biogenic emissions	MEGAN	Guenther <i>et al.</i> (2006)
Sea salt emissions	online	Monahan <i>et al.</i> (1986) and Gong (2002)
Dust emissions	online	Shaw <i>et al.</i> (2008)
Initial and boundary conditions	MOZART	Emmons <i>et al.</i> (2010)

Table 2. Model experiments.

Case name	PBL Nucleation	FT Nucleation	Aqueous Chemistry
N11A1	AN	BHN	On
N01A1	No nucleation	BHN	On
N00A1	No nucleation	No nucleation	On
N11A0	AN	BHN	Off
N01A0	No nucleation	BHN	Off

^a AN: activation nucleation. BHN: binary homogeneous nucleation.

Initial and boundary conditions are the same for all simulations, and nudging is not used.

Evaluation Data Sources

Surface meteorology, including 2 m temperature (T2), 2 m water vapor mixing ratio (Q2), 10 m wind speed (WS10) and wind direction (WD10) for a total of 97 stations in the Midwest are used (Table S1). Soundings of temperature (T) and relative humidity (RH) at Davenport, Iowa, are used for evaluating vertical profiles of meteorology. Surface measurements of bulk and speciated aerosol concentrations (PM_{2.5}, SO₄²⁻ and black carbon), SO₂, and MODIS AOD are compared with model output (data sources in Table S1). AERONET AOD from Bondville, IL, and size-resolved particle number concentrations from West Branch, Iowa (Pettibone, 2009), are also used in model evaluation.

RESULTS AND DISCUSSION

Model Evaluation

Measured and predicted values (meteorology and chemistry) from the N11A1 case are summarized in Table 3. Variables listed in Table 3 are also evaluated for N01A1 and N00A1. Results for those cases, and statistical metric definitions, can be found in supplemental information. Fractional bias (FB), fractional error (FE), mean bias (MB), mean error (ME), root mean square error (RMSE) and correlation coefficient (R) are reported, with percentage bias and error relative to measurement means. The performance statistics from different cases for both meteorology and species are generally similar. Noticeable sensitivity occurs for SO₄²⁻ and SO₂. For SO₄²⁻, N11A1 shows the best performance for all statistical metrics; for SO₂, the performance of N11A1 is either better or between that of N01A1 and N00A1.

Meteorology Evaluation

Predicted daily T2, WS10, WD10 and Q2 generally agree with observation. T2 is simulated with a correlation coefficient of 0.91 and a bias of 0.80 K. WS10 is persistently overestimated (MB: 0.59 m s⁻¹ [13.8%]; FE: 0.53; FB: 0.59). Similar positive biases are common in WRF-Chem studies, with possible attribution to horizontal resolution and unresolved topographical features (Zhang et al., 2010b; Tuccella et al., 2012). WD10 is simulated well

($r = 0.96$; MB: 4.36°). Q2 is also reproduced suitably (MB: 0.15 g kg⁻¹ [1.28%]; $r = 0.93$).

The model also reproduces vertical profiles of temperature and water vapor mixing ratio (Fig. S2). Water vapor mixing ratio near the surface was overestimated (underestimated) during nighttime (daytime). Variations are relatively small in the model which could be explained by the model's limited vertical resolution.

Air Pollutant Evaluation

As the purpose of the study is sensitivity to NPF, bias and error in simulating observed concentrations of air pollutants are most relevant to their influence on the sensitivity results.

Table 3 shows that the monthly average SO₄²⁻ concentration is overpredicted by 0.44 μg m⁻³ (20.1%), while the black carbon (BC) concentration is underpredicted by 0.07 μg m⁻³ (25.9%). A possible source of positive bias of SO₄²⁻ could be SO₂ overestimation. As shown in Table 3, SO₂ is predicted with a positive bias of 0.92 ppb (29.8%). Underestimation of BC may be the result of errors in anthropogenic emissions, the absence of wildfire emissions, and biases in the meteorological fields (for example, the overprediction of temperature indicates PBL height might be over-predicted as well). PM_{2.5} is underpredicted, which is expected, as this simulation did not include SOA formation except in sensitivity tests. Performance for PM_{2.5}, SO₄²⁻, and BC exceed the 75th percentile for r^2 , and are consistent with the middle 50% on bias and error metrics compared to prior U.S. photochemical grid modeling studies compiled by Simon et al. (2012). Using FB and FE results criteria from Morris et al. (2005), all chemical species fall into the “good” or “average” classifications. Comparing to more stringent and recent performance goals (Emery et al., 2017), black carbon meets suggested criteria for normalized mean bias (NMB), normalized mean error (NME), and r . PM_{2.5} meets two of three criteria (NMB exceeds 30%). SO₄²⁻ meets the correlation criteria (it exceeds r of 0.7) while slightly exceeding the NMB and NME recommended criteria.

Model-measurement comparison of AOD at 550 nm (Fig. S3) shows similar accuracy to previously reported results (Mashayekhi et al., 2014), with underprediction in the eastern U.S., as expected in summer modeled with SOA absent. Mean observed and modeled AOD measurement at the Midwest location of Bondville, IL, are 0.17 and 0.12, respectively (also shown in Fig. S3).

Table 3. Observed and modeled meteorological and chemical species.

	Mean obs	Mean model	FB	FE	MB	ME	RMSE	r
T2 (K)	295.50	296.30			0.80	0.93	1.26	0.91
WS10 (m s ⁻¹)	3.85	4.38			0.53	0.55	0.67	0.89
WD10 (°)	210.86	215.23			4.36	14.54	18.86	0.96
Q2 (g kg ⁻¹)	11.76	11.90			0.15	0.48	0.59	0.94
SO ₄ ²⁻ (μg m ⁻³)	2.21	2.65	0.20	0.43	0.45	0.99	1.72	0.81
BC (μg m ⁻³)	0.27	0.20	-0.22	0.50	-0.07	0.12	0.21	0.46
PM _{2.5} (μg m ⁻³)	9.15	6.02	-0.45	0.56	-3.13	4.08	6.17	0.56
SO ₂ (ppb)	3.09	4.01	0.27	0.73	0.92	2.57	3.83	0.30
CO (ppb)	272.16	219.60	-0.17	0.42	-52.49	106.04	137.49	0.33
O ₃ (ppb)	35.10	44.85	0.24	0.28	9.76	11.23	14.18	0.44

Particle Number Concentrations

CN_{10–63} (10 to 63 nm) are evaluated against 310 hours of measurements from West Branch, Iowa; they are reproduced within a factor of two and with a correlation coefficient of 0.42 (Fig. 1). Maps of surface CN can be found in supplemental information. Period averaged CN_{10–63} from the model and observation are 9228 cm^{−3} and 4873 cm^{−3}, respectively. The model performance is similar to that of Matsui *et al.* (2013). They found CN₁₀ at two sites in the outflow regions of East Asia were overpredicted by factors of 1.6 and 1.7, respectively. Performance is improved over prior reported evaluation of aerosol number concentration in the region, where Spak and Holloway (2009) compared CMAQ simulations (36 km resolution) with hourly observations at Bondville. Yu *et al.* (2015) using GEOS-Chem-APM (CN_{10–100}) and Sullivan *et al.* (2018) using NPF explicit WRF-Chem (CN_{6–25}) both report hourly correlation coefficients of ~0.2–0.5, depending on parameterization and season. Mann *et al.* (2014) examined the global variation in particle size distribution using 12 global aerosol microphysics models. They found that the best estimate of the annual mean particle concentrations (for CN₃, CN₁₀ or CN₁₄) were within a factor 2 of the observations at all 13 sites, including Bondville, IL (Midwestern U.S.).

The N01A1 scenario predicted a lower mean CN_{10–63} (4735 cm^{−3}) that was closer to the observed mean but with an inferior correlation coefficient (0.33 vs. 0.42). Overprediction in the N11A1 case is most pronounced in the smallest sizes, caused in part by insufficient growth of nuclei due to our assumption of only inorganic secondary growth. Although this causes bias at the smallest sizes and a shift between observed (high number mean diameter) and simulated (lower

number mean diameter) size distributions, our results focus mainly on the potential for feedbacks between nuclei, clouds, precipitation, and aerosol mass in the coupled nucleation explicit WRF-Chem system. This potential is assessed by model-model comparisons, and robustness of conclusions is checked by sensitivity runs including SOA. However, due to the overprediction of nuclei and ultrafine particles in this study, and the lack of SOA in the base case runs, we likely overestimate some of the microphysical-chemical feedbacks. The magnitude of the effects reported here should be considered as an upper limit that should be refined by future work.

Influence of NPF on Key Modeled Parameters CN and CCN

Table 4 lists the monthly mean CN and CCN concentrations from different simulations. As expected, simulations that include nucleation in the FT (N01A1) or the FT and PBL (N11A1) have higher number concentrations of CN₂₀ relative to N00A1 (CN_x is used throughout for the number concentration larger than *x* nm). CN₅₀ and CN₁₀₀ behave similarly. FT nucleation has a strong impact on surface CN concentrations, consistent with other studies (Merikanto *et al.*, 2009; Spracklen *et al.*, 2010); however, study of the FT nucleation impacts on boundary layer air quality, meteorology, and cloud properties is beyond the scope of the current work.

Results for CCN at high supersaturations are similar to those for CN. At 0.5% and 1% supersaturation, higher particle number concentrations in simulations with nucleation lead to higher CCN concentrations (Table 4). However, CCN at low supersaturation (0.1%) are highest without nucleation

Fig. 1. Observed (black) and modeled (red, N11A1 simulation) CN (10–63 nm) at the West Branch, Iowa (WBI), site.

Table 4. Monthly-mean CN and CCN concentrations. Averages are calculated for the surface layer within the Midwest subdomain shown in Fig. S1, ± represent the spatial standard deviation of the monthly mean.

Cases	CN ₂₀	CN ₅₀	CN ₁₀₀	CCN _{0.1} ^a	CCN _{0.5} ^b	CCN _{1.0} ^c
N11A1	7198 ± 3011	3668 ± 1584	1430 ± 750	130 ± 80	1740 ± 759	3190 ± 1248
N01A1	5816 ± 2389	3448 ± 1474	1415 ± 751	145 ± 85	1651 ± 708	2866 ± 1070
N00A1	1418 ± 724	1159 ± 592	696 ± 367	164 ± 82	552 ± 252	675 ± 290

^a Number concentration of CCN at 0.1% supersaturation, in # cm^{−3}.

^b Number concentration of CCN at 0.5% supersaturation, in # cm^{−3}.

^c Number concentration of CCN at 1.0% supersaturation, in # cm^{−3}.

(N00A1) and decrease in simulations with nucleation. Nucleation generates more small particles inhibiting the growth of preexisting particles, as well as decreasing hygroscopicity through inhibited accumulation mode sulfate condensation. Similar decreases in CCN concentrations at low supersaturations were reported by Matsui *et al.* (2011), but with smaller sensitivity to NPF. Cloud microphysics and aqueous chemistry configurations matched between the two studies, while grid nudging and aerosol-radiation feedback settings differed.

NPF Effect on Boundary Layer Clouds

Similar to the decreases in CCN at low supersaturations when NPF is active in the boundary layer (assessed by calculating the difference N11A1 minus N01A1), boundary layer NPF suppresses cloud droplet number concentration (CDNC), cloud liquid water path (LWP), and COD. CDNC concentrations are highest over the eastern part of the Midwest in the N01A1 simulation. LWP and COD are also high at the same location (Fig. S5). Noticeable CDNC reductions, resulting from turning PBL nucleation on, can be seen in these high CDNC regions. Changes in LWP and COD are similar in direction to the CDNC changes. PBL nucleation also leads to lower cloud occurrence frequency. LWP and COD changes linked to CDNC changes have been found in previous studies as well, but in those instances, CDNC changes were not linked to NPF (Yang *et al.*, 2011; Makar *et al.*, 2015).

Sullivan *et al.* (2018) used NPF explicit WRF-Chem modeling system (based on ver. 3.6.1), and a nearly matching time period, with higher horizontal resolution (4 km), and with resolved cumulus clouds. Our results are similar in the direction of the effect, and are based on version 3.1.1 rather than 3.6.1. The differences between the works are complementary. The difference in model version suggest the effect is not significantly mediated by recent enhancements or structural choices. Our simulations indicate that the effect is resolved at regional scales (36 km and 9 km) and not dependent on local convective processes resolved at the higher resolution of Sullivan *et al.* (2018) (4 km), and that sensitivity tests addressing resolution are robust. Biogenic contribution was neglected in Sullivan *et al.* (2018) and considered in this work, as were the individual process feedbacks creating the dimming.

NPF Effect on Air Pollutant Concentrations

Fig. 2 shows the absolute impacts of PBL nucleation (N11A1–N01A1) in monthly averaged surface $\text{PM}_{2.5}$, CO , O_3 and SO_2 . A decrease in $\text{PM}_{2.5}$ mass concentration over the Midwest is shown in Fig. 2(a). Midwest mean $\text{PM}_{2.5}$ concentrations from the N11A1 and N01A1 simulations are 6.04 and 6.56 $\mu\text{g m}^{-3}$, respectively. By including boundary layer nucleation, $\text{PM}_{2.5}$ is reduced by 8.0% on average. This decrease in $\text{PM}_{2.5}$ is due to reduction in secondary inorganic species, especially SO_4^{2-} . Compared to the N01A1 simulation, Midwest surface SO_4^{2-} in the N11A1 simulation decreases by 0.37 $\mu\text{g m}^{-3}$ on average, accounting for 71.2% of the $\text{PM}_{2.5}$ reduction. Concentrations of other species are much less sensitive to NPF. Ground level CO

and O_3 also decrease slightly in most areas in the N11A1 simulation, likely due to increases in the average PBL height in the N11A1 simulation, as shown in Fig. 4(d). Changes in SO_2 concentrations are significant in magnitude (up to 20%) but spatially inhomogeneous. We are aware of no other studies reporting a reduction of $\text{PM}_{2.5}$ sulfate due to the inclusion of NPF in a model, and we discuss the causes and implications below.

The spatial pattern to the $\text{PM}_{2.5}$ decrease (Fig. 2(a)) is similar to that of the decrease in CCN concentration at low supersaturations, as is expected since larger particles that can activate at low supersaturations are a substantial portion of the mass distribution (Fig. 8).

In contrast to surface $\text{PM}_{2.5}$, surface SO_4^{2-} , and CCN concentrations at low supersaturations, AOD is left mainly unchanged from the inclusion of NPF in the simulation (Fig. 3(c)), and the changes are spatially similar to changes in sulfate column loading (Figs. 3(d)–3(f)). Surface sulfate changes are slightly more pronounced than column sulfate changes because of changes in the sulfate vertical profile and higher PBL height in simulations with NPF. The AOD increase in the western part of the Midwest (Fig. 3(i)) is likely due to aerosol water.

NPF Effect on Meteorology

Sensitivity of meteorology to NPF is assessed by comparing N11A1 and N01A1, as shown in Fig. 4. Two types of patterns are observed. In the eastern portions of the Midwest, decreases in average cloud cover occurred with corresponding decreases in water vapor mixing ratio (Q2), and increases in shortwave radiation (SWDOWN), surface temperature (T2), and PBL height. In western portions of the Midwest, the opposite set of changes to meteorology occur. These patterns are in Fig. 4 for SWDOWN (Panel a), Q2 (Panel b), T2 (Panel c) and PBL height (Panel d), and can be compared to changes in cloud optical depth (Fig. S5). Averaging across the entire domain, and considering grid cells with clouds in both N11A1 and N01A1, turning NPF on increases CN_{25} by 497 cm^{-3} (9.4% increase relative to N01A1 for these grid cells), but decreases cloud droplet number concentration (CDNC) from 66.2 cm^{-3} (average for N01A1 in these paired grid cells) to 58.4 cm^{-3} , a 11.8% decrease in CDNC. Since the Twomey effect establishes that the sensitivities of CDNC to increases in particle number are positive when all other factors (water availability, updraft velocity, hygroscopicity, etc.) are constant, this result runs counter to a priori expectations. See, for example, Moore *et al.* (2013), where for polluted conditions ($\text{CN} \sim 5,000 \text{ cm}^{-3}$), $\partial \ln(\text{CDNC}) / \partial \ln(\text{CN})$ is 0.21 (the corresponding median slope of all BL parcels at $\text{CN} \sim 5,000 \text{ cm}^{-3}$ in our work is -0.35). Reasons for the negative sensitivity in the current work are discussed below.

The magnitude of changes reaches up to +66.4 W m^{-2} for monthly averaged shortwave radiation. T2 is influenced by a variety of factors, such as shortwave radiation, soil moisture and soil temperature. In the southwest of the area analyzed, T2 decreases by more than 1 K, and Q2 increases by more than 1.6 g kg^{-1} , consistent with precipitation enhancement (Fig. S7). PBL heights increase by up to 90.5 m

Fig. 2. Absolute changes (N11A1–N01A1) in surface concentrations of (a) $\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$), (b) CO (ppb), (c) O_3 (ppb), and (d) SO_2 (ppb).

as a result of surface warming in most areas of the Midwest. Reduced PBL heights are simulated in locations where the N11A1 simulation has lower shortwave radiation and lower surface temperature relative to N01A1.

Discussion of $\text{PM}_{2.5}$ Decrease

The $\text{PM}_{2.5}$ reduction simulated with PBL nucleation on (mainly due to $\text{PM}_{2.5}$ sulfate reduction) is potentially important for a number of applications, and the modeling system was exploited to establish its cause. In general, a sulfate mass concentration decrease must be from either weakening of a SO_4^{2-} source, or strengthening of a sink. SO_4^{2-} sources in WRF-Chem are primary emissions, and gas phase and in-cloud oxidation of SO_2 . SO_4^{2-} is removed from the model atmosphere by wet and dry deposition. A number of model-model comparisons and sensitivity runs were conducted to investigate how SO_4^{2-} sources and sinks were changing. Key results from these tests are discussed below; a full list of tests and associated inferences is also reported (Table S4).

Column integrated SO_4^{2-} and SO_2 are compared in

Fig. 5 to see if decreased conversion from SO_2 to SO_4^{2-} explains the changes in SO_4^{2-} concentrations. Column integration is across model layers 1–10 (~2 km). In the SO_4^{2-} reduction regions, column SO_2 increases. For the most polluted Midwest region as shown in the red square in Fig. 7(a), the spatial average of column SO_2 and SO_4^{2-} changes are +101.8 and –94.5 mmol m^{-2} , respectively. The increase of SO_2 and decrease of SO_4^{2-} are approximately equal, and we attribute the bulk of the SO_4^{2-} mass decrease (in N11A1) to less robust oxidation of SO_2 . In other words, the source of SO_4^{2-} decreases, rather than the sink increasing.

The influence of wet deposition on SO_4^{2-} is investigated by comparing the timing of precipitation with the SO_4^{2-} time series in N11A1 and N01A1. For wet deposition to be an important factor in the influence of NPF on SO_4^{2-} , we expect model-model differences to be maximized, or to increase, during periods of significant precipitation. Significant precipitation events occurred on 24 June and 29 June (Fig. S6). However, SO_4^{2-} differences between model runs do not match temporal trends in precipitation.

Fig. 3. Monthly average of column (a–c) AOD, (d–f) SO_4^{2-} ($\mu\text{g m}^{-2}$) and (g–i) aerosol water ($\mu\text{g m}^{-2}$). From left to right: N11A1, N01A1, N11A1–N01A1.

Dry deposition is ruled out as a main cause of the modeled SO_4^{2-} decrease by comparison of ΔSO_4^{2-} at the surface to ΔSO_4^{2-} at model layer 7 (~850 m above ground). Since dry deposition acts at the surface, its effect would be most pronounced at the lowest model layer. However, ΔSO_4^{2-} (Fig. 6(a)) is similar at both elevations.

To elucidate the relative impacts of gas phase and aqueous chemistry in the weakening of sulfur oxidation, model runs were performed with aqueous chemistry turned off, but with PBL nucleation active (N11A0) or inactive (N01A0). The results of the sensitivity tests (for Midwest averaged surface $\text{PM}_{2.5}$ and SO_4^{2-}) are shown in Table 5. These additional runs both show a decrease in SO_4^{2-} from NPF, so the direction of the NPF effect is the same as in the longer base and N01A1 runs. But the magnitude of the effect is much larger when both NPF and aqueous chemistry are active. Specifically, a 1.1% sulfate decrease occurs without aqueous chemistry, compared to a 13.0% sulfate decrease with aqueous chemistry. We therefore conclude that NPF and aqueous chemistry act together as the primary

cause of the nucleation effect on $\text{PM}_{2.5}$ mass.

Fig. 7 shows mean surface ΔSO_4^{2-} . The result is consistent with the spatially averaged values from Table 5 discussed above. Without aqueous chemistry, the influence of NPF on SO_4^{2-} concentration is small (Fig. 7(b)) and centered in the eastern portion of the Midwest and eastern U.S. In other words, the combined effects of aqueous chemistry and NPF are needed to explain the bulk of the reduced SO_4^{2-} concentrations and reduced $\text{SO}_2 \rightarrow \text{SO}_4^{2-}$ conversion.

We also attribute a portion of the SO_4^{2-} reduction to decrease in the gas phase OH radical. The OH reduction and its potential causes are explored in supplemental material (Fig. S8). In summary, biogenic emissions (~10% increase due to higher temperatures and SW radiation) are enhanced and water vapor decreases in the NPF on simulations. Increased VOCs shorten the OH lifetime (Archer-Nicholls *et al.*, 2014; Yahya *et al.*, 2014). Dilution of SO_4^{2-} through PBL height increase was investigated and found to be a minor contribution, based on the insensitivity of primary aerosols to PBL nucleation.

Interactions of Aqueous Chemistry, Cloud Droplets, and Aerosol Size Distributions

With PBL nucleation on, cloud water in the lower

troposphere (0–2 km) decreases by over 30% in most parts of the region with high SO_4^{2-} (Fig. S9), reducing the extent of aqueous sulfate production. Cloud water content increases

Fig. 4. Absolute changes (N11A1–N01A1) in (a) downwelling solar radiation (W m^{-2}), (b) surface water vapor mixing ratio Q_2 (g kg^{-1}), (c) 2 m temperature (K), and (d) PBL height (m).

Fig. 5. Changes in column-integrated (a) SO_4^{2-} and (b) SO_2 concentrations in mmol m^{-2} due to NPF (N11A1–N01A1).

Fig. 6. Percentage change at 850 m of (a) SO_4^{2-} and (b) H_2O_2 , calculated by $(\text{N11A1}-\text{N01A1})/\text{N01A1} \times 100\%$.

Table 5. Midwest mean surface $\text{PM}_{2.5}$ and SO_4^{2-} in the sensitivity test period.

Concentration ($\mu\text{g m}^{-3}$)	N11A0	N01A0	$(\text{N11A0}-\text{N01A0})/\text{N01A0}$
$\text{PM}_{2.5}$	5.23	5.29	-1.1%
SO_4^{2-}	1.82	1.84	-1.1%

Fig. 7. Absolute change in surface SO_4^{2-} ($\mu\text{g m}^{-2}$) due to NPF while (a) enabling aqueous chemistry (N11A1–N01A1) and (b) while disabling aqueous chemistry (N11A0–N01A0).

in some areas, but these are generally in locations with low SO_2 concentrations. In the Midwest, the largest decreases in absolute and percentage SO_4^{2-} concentration are simulated in the most polluted region as shown in the red square in Fig. 7(a). In the following discussion of the influence of cloud chemistry, we limit our analysis to this area.

The decrease in time-averaged mean cloud liquid water is from less frequent cloud occurrence, and from decrease in LWC of simulated clouds. For frequency calculation, we use hourly outputs for layer 1–layer 10. Cloudy grid cells decreased from 3.08% (N01A1) to 2.44% (N11A1) with PBL nucleation enabled.

Fig. 8 shows the influence of NPF on the mean aerosol number distribution of the boxed area (Fig. 8(a)), and mean aerosol volume distribution (Figs. 8(b)–8(d)). Panels a–c are from simulations without aqueous chemistry (N11A0

and N01A0) in order to isolate the influence of NPF from the additional effect of aqueous chemistry. The increase in particle number at sizes below 20 nm is clearly visible (Fig. 8(a)), as is the increase in particle volume in the same size ranges, shown in linear scale (Panel b) and log scale (Panel c). Volume ratios, by bin, are shown in Panel d. For the cases without aqueous chemistry (A0 suffix) there is an increase of particle volume at small sizes, and a decrease at larger sizes. Particle number concentration decreases the most in bin 11 and 12, covering a size range from 100 nm to 251 nm. The increase of sulfate in the 1–100 nm range is $0.043 \mu\text{g m}^{-3}$, while the decrease of sulfate in the 100–2500 nm range was $0.146 \mu\text{g m}^{-3}$. With cloud chemistry turned off, total $\text{PM}_{2.5}$ sulfate still decreases by about 5% in the boxed area.

With aqueous chemistry active (Fig. 8(d)), ratios with the

A1 suffix), the decrease in particle volume at large sizes is much more pronounced than the cases without aqueous chemistry, and shifts to larger sizes.

NPF has an impact on the distribution of $S_{\max}$, which can be seen in comparison of N11A1 and N01A1; $S_{\max}$ is the maximum supersaturation reached in rising parcels. $S_{\max}$ is calculated by the parameterization of Abdul-Razzak (2000) and is influenced by the updraft velocity, particle concentration and size, and hygroscopicity. The critical size for activation is about 120 nm at $S_{\max} = 0.25\%$ and 200 nm at $S_{\max} = 0.1\%$, corresponding to bin 11–12 (Kerminen *et al.*, 2012).

Median $S_{\max}$ for N11A1 and N01A1 runs are 0.153% and 0.157%, respectively. The distribution (graphed in supplemental material Fig. S10) of $S_{\max}$ values shifts to lower values in N11A1 relative to N01A1, with the frequency of low $S_{\max}$ ($< 0.1\%$) at 13.8% versus 1.8% respectively. The cause of the changes in $S_{\max}$ has not been quantified in this work; however, we hypothesize that the decrease of $S_{\max}$ contributes to the feedback between NPF and reduction in cloud water.

Further showing the importance of $S_{\max}$ is a sensitivity test with repetition of runs N11A1 and N01A1 with $S_{\max}$ fixed at 1%. In the two sensitivity tests (ten days including spin up) $S_{\max}$ was coerced to 1.0% whenever a cloud is formed in a grid cell. This allowed more particles to be activated with nucleation, and eliminated the large region

of SO_4^{2-} decrease with NPF on (Fig. S10), replacing it with a mix of small SO_4^{2-} increases and decreases.

As described in the section on meteorological effects, considering grid cells with resolved clouds in both N11A1 and N01A1, turning NPF on (with aqueous chemistry) increases CN_{25} but decreases CDNC, contributing to the cloud dimming effect. Stratifying this by particle number, we find median sensitivities $\Delta \ln(\text{CDNC})/\Delta \ln(\text{CN})$, where the Δ is N11A1–N01A1, have positive medians for CN below $\sim 3000 \text{ cm}^{-3}$, and then have negative medians when CN is above $\sim 3000 \text{ cm}^{-3}$. The highest frequency of CN in comparing paired cloudy grid cells between N11A1 and N01A1 contained 3000–4000 cm^{-3} CN, and had a median $\Delta \ln(\text{CDNC})/\Delta \ln(\text{CN})$ of -0.35 .

Decreased hygroscopicity of CCN active particles may also contribute to the decrease in cloud LWC and cloud droplet number in simulations with active NPF. This likely occurs as NPF shifts sulfate mass to smaller particles (through nucleation), and decreases condensational growth of larger particles, as shown in Fig. 8. Condensational growth occurring on primary particles increases their hygroscopicity, facilitating activation.

In addition to liquid-water content, the conversion rate of SO_2 to SO_4^{2-} depends on cloud pH and oxidant concentrations (primarily dissolved H_2O_2 and O_3). The SO_2 – H_2O_2 reaction is roughly pH independent while SO_2 – O_3 reaction rates decreases under acidic conditions. Under

Fig. 8. Midwest particle number (a) and volume (b) distribution without cloud chemistry. Top figures are for all 20 bins. (c) is for particle volume distributions ($63.1 < D_p < 631 \text{ nm}$), and (d) particle number ratios for bin 9–16.

both N11A1 and N01A1, over 96% of clouds had $\text{pH} < 5$, consistent with H_2O_2 oxidation of SO_2 (Hung and Hoffmann, 2015). Fig. 6(b) shows the gas phase H_2O_2 at a height of 850 m above ground. The N11A1 case has higher levels of H_2O_2 in most regions of the Midwest, suggesting cloud water is a more important factor than H_2O_2 availability in decreasing SO_4^{2-} .

Sensitivity to Resolution and Resolved Cumulus Clouds

The NPF-explicit WRF-Chem model used in this paper considers online aerosol-cloud interactions in resolved clouds only. However, at 36 km horizontal resolution, both resolved and parameterized clouds are required. The results are tested for sensitivity to horizontal model resolution using a 9 km horizontal resolution nest within the 36 km domain. Simulations are ten days with 3-day spin-up for the N11A1 and N01A1 cases without cumulus parameterization, resulting in only grid-resolved clouds. Although the scale is not as fine as convection-permitting models (< 3 km horizontal resolution; Prein *et al.*, 2015), this sensitivity study represents a spatial scale where cumulus clouds arise in simulation. The 9 and 36 km results (Figs. S12–S14) are compared for absolute values and changes (BL NPF on vs. off) in surface SO_4^{2-} , CDNC, cloud LWC, and total precipitation. Values and spatial patterns were similar with the exceptions of precipitation (higher at 9 km resolution) and absolute surface SO_4^{2-} concentration (lower in 9 km simulation). Percentage changes in sulfate are similar in the 9 km simulation even though absolute concentrations are lower. WRF-Chem v3.8 and v3.9 permit cloud-aerosol interactions in sub-grid-scale convective clouds as described by Berg *et al.* (2015) and replication of our finding in nucleation-explicit variants of these later releases (and in other models) is an important next step. Berg *et al.* (2015) conducted sensitivity studies with and without aqueous chemistry in WRF-Chem including aerosol interaction with convective clouds, and report on changes to both sources and sinks of SO_4^{2-} , but do not extend the work to also include sensitivity to NPF.

Sensitivity to Biogenic Secondary Aerosol

A biogenic SOA sensitivity simulation employed a simplified biogenic SOA treatment adapted from the WRF-Chem NPF study of Cui *et al.* (2014). Fixed fractions of isoprene (4%), α -pinene (15%) and limonene (15%) are converted to biogenic SOA and condensed onto preexisting particles in proportion to the Fuchs corrected surface area of each aerosol section. For computational efficiency, the partitioning is assumed to be irreversibly to the aerosol phase. OC increases across the domain, and especially in the southeast U.S. Midwest mean OC increases from 0.49 to $1.39 \mu\text{g m}^{-3}$ due to the biogenic SOA (Fig. S16). Decreases in SO_4^{2-} , CDNC and cloud water still occur in the eastern U.S., in patterns and quantities very similar to the base simulations with no biogenic SOA (Fig. S17–S18).

Conceptual Model

A conceptual model of the processes in NPF explicit WRF-Chem leading to the influence of NPF on inorganic

particle mass, and modified through a cloud processing feedback, is drawn in Fig. 9. PBL nucleation increases the number of ultrafine particles, shifting the size distribution to smaller sizes, increasing mass at sizes below 100 nm, and decreasing mass at sizes above about 100 nm. The modified size distribution of aerosol particles leads to changes in cloud properties, specifically lower CDNC, lower cloud liquid water path (LWP), depressed S_{max} , and lower COD. Model sensitivity tests indicate that the changes in overall sulfate mass are due to a feedback process where reduction of cloud liquid water causes less SO_4^{2-} formation via aqueous chemistry. Changes in biogenic emissions, shortwave radiation, and OH concentrations contribute. Not shown in Fig. 9 are the (relatively minor) changes in PBL height (increasing on average in areas of high inorganic PM), and their effect of decreasing all surface pollutants such as CO, primary organic carbon, black carbon, and inorganic PM.

CONCLUSION

This is the first study quantifying the interactions between PBL nucleation, meteorology, and air quality. We have demonstrated a link between NPF and reduced SO_2 - SO_4^{2-} conversion over the Midwestern and Eastern U.S. in summer that is moderated through cloud activation and aqueous chemical processes. Furthermore, for the location and time period simulated, resolving NPF led to an indirect effect counter to the usual direction—with NPF causing fewer and dimmer clouds, supporting the results of Sullivan *et al.* (2018). The result is novel because while there are studies that have one or more key features of our work (explicit treatment of the nuclei mode from 1 to 10 nm, clouds coupled to meteorology and resolved clouds coupled to primary and secondary aerosols, biogenic emissions that are coupled online with radiation and meteorology, and sensitivity tests where aqueous chemistry and PBL nucleation are independently activated or not activated), a literature search found no studies reporting all of these features.

In our simulations, which predicted most air pollutants at a level of accuracy similar to that of contemporary models, some features of previous studies were reproduced, such as the enhancement of CN concentrations and CCN concentrations at high supersaturations. More interestingly, the concentration of CCN at low supersaturations decreased, and the inclusion of PBL nucleation changed meteorological and air pollution variables, often in unexpected ways (reductions in $\text{PM}_{2.5}$, SO_4^{2-} ; reductions in the cloud frequency, cloud optical depth, and cloud droplet number concentration; etc.). Sensitivity tests demonstrated that aqueous chemistry combines with PBL nucleation to form a feedback loop, drawn in Fig. 9, resulting in less frequent clouds and lower average cloud water content. This in turn reduces the amount of volume available for aqueous chemical reactions and further reduces sulfate production. Changes in OH and biogenic VOCs may also be involved. Alternative hypotheses for the SO_4^{2-} decrease (cloud droplet pH changes, changes in H_2O_2 , changes in dry and wet deposition, and boundary layer height increases) are insufficient for explaining the

Fig. 9. Selected features leading to decrease in inorganic $\text{PM}_{2.5}$. Colors indicate phase as aerosols (bold black), clouds (blue) and gases (red). (1) active nuclei production increases ultrafine particles and (through decreased condensation of H_2SO_4) decreases accumulation mode particles (Fig. 8); (2) CDNC and cloud LWC decrease; (3) in areas of peak concentration, $\text{PM}_{2.5}$ and SO_4 decrease (Figs. 2 and 7); (4) short wave radiation increases (Fig. 4); (5) SO_2 concentrations increase (Fig. 5); (6) conversion of SO_2 to SO_4 by aqueous chemistry decreases (Fig. 7); (7) the distribution of S_{max} shifts to lower values with NPF; and (8) isoprene concentrations are increased somewhat in key locations with NPF; (9) Decrease of OH radical concentration (Fig. S8); (10) Decrease of water vapor concentration (Fig. S8).

modeled results. Limitations in the simulations with regard to resolution (36 km and sub-grid clouds not coupled with aerosols) and no SOA were addressed with sensitivity runs, which showed the persistence of key sulfate and cloud effects. When compared to observations within the domain, simulated CN concentrations were generally higher, which indicates our findings on air quality and meteorological feedback processes might be an upper limit.

These substantial effects of NPF on sulfate and clouds only appear in our simulations with the simultaneous coupling of NPF, aerosols, aqueous chemistry, and meteorology. Replication in other fully coupled models is needed across a variety of resolution-season combinations, including in models with state-of-science treatments of SOA and aerosol-cloud-radiation interactions in cumulus clouds. Furthermore, observational or experimental evidence of these feedback loops is also needed. With these caveats, we have found a potentially important feedback process relevant to air quality, weather forecasting, and aerosol-cloud-climate studies.

ACKNOWLEDGMENTS

This research has been supported by NSF grant ATM-0748602, funds from the Center for Global and Regional Environmental Research (CD, AP, CS), and NASA NNX11AI52G (SS). HM was supported by the Japan Society for the Promotion of Science (JSPS), the Ministry of Education, Culture, Sports, Science, and Technology and the Japan Society for the Promotion of Science (MEXT/JSPS) KAKENHI (JP26740014, JP17H04709, JP26241003, and JP16H01770), MEXT Green Network of Excellence (GRENE) and Arctic Challenge for Sustainability (ArCS) projects, and the global environment research funds of the Ministry of the Environment, Japan (2-1403, 2-1703) (HM). We thank Yuyan Cui and Alma Hodzic for correspondence regarding Cui *et al.* (2014).

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be

found in the online version at <http://www.aaqr.org>.

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Received for review, May 7, 2018

Revised, September 2, 2018

Accepted, September 8, 2018