



1 **Improved Prediction of Dimethyl Sulfide (DMS) Distributions in the** 2 **NE Subarctic Pacific using Machine Learning Algorithms**

3 Brandon J. McNabb¹ & Philippe D. Tortell^{1,2}

4 ¹Department of Earth, Ocean and Atmospheric Sciences, University of British Columbia, Vancouver, BC V6T 1Z4,
5 Canada

6 ²Department of Botany, University of British Columbia, Vancouver, BC V6T 1Z4, Canada

7 *Correspondence to:* Brandon J. McNabb (bmcnabb@eoas.ubc.ca)



8 **Abstract.** Dimethyl sulfide (DMS) is a volatile biogenic gas with the potential to influence regional climate as a
9 source of atmospheric aerosols and cloud condensation nuclei (CCN). The complexity of the oceanic DMS cycle
10 presents a challenge in accurately predicting sea-surface concentrations and sea-air fluxes of this gas. In this study,
11 we applied machine learning methods to model the distribution of DMS in the NE Subarctic Pacific (NESAP), a
12 global DMS hot-spot. Using nearly two decades of ship-based DMS observations, combined with satellite-derived
13 oceanographic data, we constructed ensembles of 1000 machine-learning models using two techniques, random
14 forest regression (RFR) and artificial neural networks (ANN). Our models dramatically improve upon existing
15 statistical DMS models, capturing up to 62% of observed DMS variability in the NESAP and demonstrate notable
16 regional patterns that are associated with mesoscale oceanographic variability. In particular, our results indicate a
17 strong coherence between DMS concentrations, sea surface nitrate (SSN) concentrations, photosynthetically active
18 radiation (PAR) and sea surface height anomalies (SSHA), suggesting that NESAP DMS cycling is primarily
19 influenced by heterogenous nutrient availability, light-dependent processes and physical mixing. Based on our
20 model output, we derive summertime, sea-air flux estimates ranging between 0.5-2.0 Tg S yr⁻¹ in the NESAP. Our
21 work demonstrates a new approach to capturing spatial and temporal patterns in DMS variability, which is likely
22 applicable to other oceanic regions.

23 1 Introduction

24 Dimethyl sulfide (DMS), a volatile biogenic gas, is an important component of the marine sulfur cycle.
25 This molecule contributes the largest fraction of bulk non-sea salt (NSS) sulfate emissions to the atmosphere (Bates
26 et al., 1992), where it is rapidly oxidized to form aerosols that act as cloud condensation nuclei (CCN; Charlson et
27 al., 1987; Hegg et al., 1991; Korhonen et al., 2008), potentially influencing regional albedo and climate (Charlson
28 et al., 1987; Ayers and Caine, 2007). Given its potential role in climate regulation, and recognized importance to
29 marine microbial metabolism (Vila-Costa et al., 2006) and food web interactions (Nevitt, 2008), substantial
30 research has focused on characterizing DMS dynamics in seawater. This work has revealed considerable
31 complexity in the oceanic DMS cycle, which has limited the development of simple predictive algorithms
32 describing spatial and temporal DMS distributions.

33 Oceanic DMS production and loss are tightly linked with the biological cycling of the related metabolites
34 dimethyl sulfoniopropionate (DMSP) and dimethyl sulfoxide (DMSO). DMS is believed to be primarily derived
35 from the cleavage of DMSP (Kiene and Linn, 2000), but it can also be cycled through biological DMSO reduction
36 (Spiese et al., 2009) and oxidation (Lidbury et al., 2016), and abiotically by light-dependent reactions (del Valle et



37 al., 2007; Royer et al., 2016). DMS cycling is influenced by suite of environmental and ecological factors, including
38 release from phytoplankton cells into the dissolved pool via grazing (Dacey and Wakeham, 1986), viral lysis (Malin
39 et al., 1998), or exudation. Oxidative stress generated by other variables such as temperature (Kirst et al., 1991),
40 salinity (Dickson and Kirst, 1987), UV radiation (Kinsey et al., 2016), and nutrient limitation (Bucciarelli et al.,
41 2013; Spiese & Tatarkov, 2014) may also enhance the cycling of DMSP and DMSO, which may regulate DMS
42 concentrations through cascading oxidative pathways (Sunda et al., 2002). Finally, variability in surface wind fields
43 can modulate the rates of DMS sea-air exchange, providing a significant source of heterogeneity in surface water
44 DMS concentrations (Royer et al., 2016). These examples illustrate the complex non-linearity of the oceanic DMS
45 cycle.

46 Over the past two decades, a number of approaches have been developed to model DMS distributions at
47 both global (Bock et al., 2021; Galí et al., 2018; Simó and Dachs, 2002; Vallina and Simó, 2007) and regional (Watanabe
48 et al., 2007) scales. These models have been largely based on linear regression techniques to estimate DMS using
49 one or two predictors. To date, these studies have focused on a number of variables, including ratio of chlorophyll
50 a (Chl-a) to mixed layer depth (MLD) (Simó and Dachs, 2002), sea surface temperature (SST) and nitrate (SSN)
51 (Watanabe et al., 2007), solar radiation dose (SRD) (Vallina and Simó, 2007), photosynthetically active radiation
52 (PAR) and modelled DMSP concentrations (Galí et al., 2018). Some of these models have demonstrated reasonably
53 good performance at global scales, but their predictive power is generally diminished at regional scales (Herr et al.,
54 2019), failing to accurately resolve important smaller-scale features (Belviso et al., 2003; Nemcek et al., 2008;
55 Royer et al., 2015; Tortell, 2005b).

56 In recent years, machine-learning algorithms have been increasingly used to derive predictions for non-
57 linear oceanic systems. For example, these methods have been successfully applied to describe the spatial and
58 temporal patterns of global methane flux (Weber et al., 2019) and carbon export (Roshan and DeVries, 2017). To
59 our knowledge, only two studies have thus far applied machine-learning to describe DMS distributions, with one
60 study focused on the Arctic (Humphries et al., 2012) and the other exploring a global domain (Wang et al., 2020).
61 Despite producing algorithms with reasonable predictive skill, these two studies found limited success in resolving
62 the underlying relationships driving DMS variability. This was partially due to a reliance on indirect sensitivity
63 tests assessing the importance of predictor variables, and also, potentially, from the large-scale averaging applied
64 to the underlying data fields ($1 \times 1^\circ$; 111 km^2). Analyses at higher spatial resolution may reveal mesoscale (roughly
65 20-200 km) and sub-mesoscale (roughly 1-20 km) patterns that would otherwise be obscured, thereby increasing
66 predictive strength.



67 Machine learning algorithms require large datasets for the training and testing process. Traditionally, DMS
68 measurements were based on time-consuming ship-board analysis of discrete samples, resulting in sparse data
69 coverage over much of the oceans. More recently, the development of several automated DMS measurement
70 systems (Royer et al., 2014; Saltzman et al., 2009; Tortell, 2005a) has provided marine DMS observations at a
71 significantly higher resolution, yielding greater spatial and temporal data coverage. These new datasets potentially
72 enable new insights into small-scale and regional patterns in oceanic DMS distributions, as well as the
73 characterization of oceanic DMS ‘hot-spots’. One such global DMS hotspot is the northeast subarctic Pacific
74 (NESAP) (Asher et al., 2017; Herr et al., 2019; Lana et al., 2011), a region encompassing both highly productive
75 coastal upwelling regimes, and off-shore, iron-limited waters (Martin and Fitzwater, 1988). Several factors have
76 been proposed to account for the elevated DMS production in the NESAP, including increased productivity from
77 entrainment and upwelling along coastal fronts (Asher et al., 2017), and the stimulation of DMS production in
78 response to oxidative stress in low iron waters (Sunda et al., 2002; Herr et al., 2020). Although multiple studies
79 have examined empirical relationships between DMS and various oceanographic factors in the NESAP (Watanabe
80 et al., 2007; Herr et al., 2019; Asher et al., 2017, 2011), these have all reported low predictive skill based on simple
81 linear correlation approaches. To date, machine-learning approaches have not been applied to describe DMS
82 distributions specifically in this region.

83 Here, we present an approach to modelling summertime NESAP DMS concentrations and sea-air fluxes
84 using ensemble random forest regression (RFR) and artificial neural network (ANN) machine-learning algorithms.
85 Our statistical models leverage field observations of DMS collected across the NESAP between 1997 to 2017 to
86 generate a summertime DMS climatology mapped at a higher spatial resolution than previous efforts (Simó and
87 Dachs, 2002; Vallina and Simó, 2007; Galí et al., 2018; Watanabe et al., 2007; Humphries et al., 2012; Wang et
88 al., 2020). This new modelling approach represents a significant improvement over previous methods and predicts
89 regional DMS distributions that are coherent with underlying patterns of oceanographic variability. Most notably,
90 the modelled DMS concentrations and sea-air fluxes can be explained, to a large extent, by regional and mesoscale
91 patterns in nutrient supply and physical mixing dynamics. Based on the output of our models, we present
92 summertime sea-air flux estimates in close agreement with previous studies (Herr et al., 2019; Lana et al., 2011),
93 further highlighting the importance of the NESAP as a globally-significant sulfur source to the atmosphere.



94 2 Methods

95 2.1 Data

96 A combination of data sources was used in training our machine-learning models to build a summertime
97 DMS climatology. For this study, we restricted DMS measurements to the months of June, July and August between
98 1997 to 2017 in the NESAP (43-60°N, 147-122°W). A total of 26,201 data points were obtained from the NOAA
99 PMEL repository (<https://saga.pmel.noaa.gov/dms/>; last accessed: February 3, 2021), including measurements
100 derived from purge and trap gas chromatography and membrane inlet mass spectrometry. The DMS data were
101 binned to a monthly resolution, regardless of year, and averaged into 0.25 x 0.25° grid cells.

102 Predictor data used to build our machine-learning models included the following variables derived from
103 the NASA Aqua MODIS satellite at level L3 monthly 0.036° resolution: sea surface temperature (SST), the ratio
104 of normalized fluorescence line height to chlorophyll a (nFLH:Chl-a), instantaneous and daily observed
105 photosynthetically active radiation (iPAR and PAR, respectively), particulate inorganic carbon (PIC), the
106 absorption of gelbstof and detritus at 433 nm ($a_{\text{cdm}}(443)$), and diffuse attenuation coefficients at 490nm (K_d).
107 Satellite-based PIC is considered as a proxy for the abundance of coccolithophores and other calcified
108 phytoplankton (Franklin et al., 2010), whereas the $a_{\text{cdm}}(443)$ product is considered a proxy for the distributions of
109 chromophoric dissolved organic matter (CDOM) (Nelson & Siegel, 2013), which is thought to be an important
110 photosensitizer of DMS (see Sect. 4.1). For observations prior to 2004, data were from either SeaWiFS or Terra
111 MODIS when SeaWiFS data was unavailable (e.g. nFLH and iPAR). As described below, K_d and PIC were later
112 excluded from the final models (see Sect. 2.6).

113 The following predictor variables were also used: 6-day averaged sea surface height anomalies (SSHA)
114 derived from the TOPEX/Poseidon satellites at 0.17° resolution; Level L4 ESA Sentinel-3 Copernicus monthly-
115 averaged 0.25° wind speeds; net primary productivity (NPP) from the Vertically-Generalized Production Model
116 (VGPM; Behrenfeld & Falkowski, 1997) at monthly 0.25° resolution; sea surface nitrate from the 2018 World
117 Ocean Atlas at monthly 1° resolution (Garcia et al., 2019); and mixed-layer depth (MLD) and sea surface salinity
118 (SSS) from the MIMOC climatology at 0.5° resolution (Schmidtke et al., 2013). Except for MIMOC data, all
119 predictors were restricted in time to the corresponding years of DMS sampling (1997 to 2017). Net community
120 productivity (NCP) was estimated from the algorithm of Li & Cassar, (2016; using NPP and SST). As with DMS
121 observations, predictor data were interpolated to a 0.25 x 0.25° average monthly resolution using linear radial basis
122 interpolation functions. Interpolation was constrained to the oceanic region by masking out land pixels using
123 ETOPO2 bathymetric (0.033° resolution) binned at 0.25 x 0.25° resolution. Data sources can be found in Table 1.



124

125 **Table 1. Data sources and spatial and temporal resolution of predictor variables used to develop the RFR and ANN algorithms. Data**
 126 **processing levels are indicated where relevant. All variables were used as predictors (excluding bathymetry) and post-processed to**
 127 **monthly-averaged, 0.25° resolution (see sections 2.1-2.2).**

Variable	Spatial Resolution (°)	Temporal Resolution	Source	Level
Sea Surface Temperature (SST)	0.036	6-Day Average	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS(2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Chlorophyll-Normalized Fluorescence (nFLH:Chl-a)	0.036	Monthly	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS (2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Instantaneous Photosynthetically Active Radiation (iPAR)	0.036	Monthly	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS (2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Daily Photosynthetically Active Radiation (PAR)	0.036	Monthly	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS (2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Particulate Inorganic Carbon (Calcite; PIC)	0.036	Monthly	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS (2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Absorbtion of Gelbstof and Detritus at 433 nm ($a_{cdm}(443)$)	0.036	Monthly	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS (2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Diffuse Attenuation Coeffiencents at 490 nm (K_d)	0.036	Monthly	SeaWiFS/AquaTERRA (1997-2003) or AquaMODIS (2004-2017): https://oceancolor.gsfc.nasa.gov/13/	3
Sea Surface Height Anomalies (SSHA)	0.17	Monthly	TOPEX/Poseidon: https://podaac.jpl.nasa.gov/dataset/SEA_SURFACE_HEIGHT_ALT_GRID_S_L4_2SATS_5DAY_6THDEG_V_J_PL1812	4
Monthly Wind Speeds	0.25	Monthly	ESA Sentinal-3 Copernicus: https://resources.marine.copernicus.eu/?option=com_csw&view=details&product_id=WIND_GLO_PHY_CLIMAT_E_L4_REP_012_003	N/A



Net Primary Productivity (NPP)	0.25	Monthly	Vertically-Generalized Production Model (VGPM): http://www.science.oregonstate.edu/ocean.productivity/	N/A
Sea Surface Nitrate (SSN)	1	Monthly	World Ocean Atlas 2018 (WO18): https://www.ncei.noaa.gov/access/world-ocean-atlas-2018/	N/A
Mixed Layer Depth (MLD)	0.5	Monthly	MIMOC Climatology: https://www.pmel.noaa.gov/mimoc/	N/A
Sea Surface Salinity (SSS)	0.5	Monthly	MIMOC Climatology: https://www.pmel.noaa.gov/mimoc/	N/A
Bathymetry	0.033	N/A	ETOPO2: https://rda.ucar.edu/datasets/ds759.3/	N/A



129

130 2.2 Machine-learning models

131 We compared the performance of random forest regression (RFR) and artificial neural network (ANN)
132 models at the regional scale. In both cases, the models were built as an ensemble of either 1000 individual decision
133 trees or individual networks to minimize bias in predictions. The input data were randomly divided for use in model
134 training (80%) and external testing (20%). Although RFR is not sensitive to large differences in predictor variance,
135 predictor data were standardized in both models by normalization to their respective mean and standard deviation.
136 Additionally, we applied an inverse hyperbolic sine (IHS) transformation to the DMS data prior to training. Testing
137 results indicated that IHS yielded slightly better performance than the more traditional logarithmic transformations
138 for both of our models.

139 Both our ANN and RFR models followed a similar design to Weber et al. (2019). Our ANNs were built
140 using a feed-forward framework consisting of a single input node, two hidden layers each consisting of 30 neurons
141 (using a sigmoidal activation function), and a single output layer (using a linear activation function). A Bayesian
142 L2 (Ridge) regularization parameter was tuned to minimize overfitting. Each individual decision tree within the
143 RFR was trained using the standard CART algorithm and constrained to a max depth of 25 decision splits, the
144 simplest configuration determined to perform well and minimize overfitting.

145 2.3 Sea-to-air fluxes

146 Sea-air DMS fluxes (F_{DMS} , $\mu\text{mol m}^{-2} \text{d}^{-1}$) were calculated from the monthly-averaged observed and
147 modelled DMS values for June, July and August. F_{DMS} was calculated using the gas transfer velocity (k , $\text{cm}^3 \text{hr}^{-1}$)
148 following the modified approach of Webb et al. (2019):

$$149 F_{DMS} = k(DMS)(0.24) \quad (1)$$

150 where the factor of 0.24 converts to the values to daily fluxes. Since our fluxes were calculated from our monthly
151 averaged models, the gas transfer velocity was calculated using the approach from Simó & Dachs (2002), as
152 modified by (Nightingale et al., 2000). This approach is necessary to correct for differences due to the non-linear
153 relationship between DMS and wind speed (Livingstone and Imboden, 1993) when using monthly-averaged,
154 satellite-derived wind speeds. Assuming a Rayleigh distribution ($\xi=2$), k can be defined as:

$$155 k = [5.88\eta\Gamma(1 + \frac{2}{\xi}) + 1.49\eta\Gamma(s)]Sc_{DMS}^{-0.5} \quad (2)$$



156 where η is the quotient of the wind speed (m s^{-1}) by the gamma function $\Gamma(s)$ (using $s = 1 + \frac{1}{\xi}$), and Sc_{DMS} is the
157 DMS-specific Schmidt number ($\text{cm}^3 \text{hr}^{-1}$) as defined by Saltzman et al. (1993):

$$158 \quad Sc_{DMS} = 2674 - 147.12(SST) + 3.72(SST^2) - 0.038(SST^3) \quad (3)$$

159 Regional summertime fluxes (\bar{F}_{DMS} , Tg) were calculated as the average quantity of DMS-sulfur emitted
160 over 92 days through the area of the mapped study region ($1.28 \times 10^7 \text{ km}^2$ or 85.0% of the total bounded area).

161 2.4 Comparison against existing algorithms

162 Simple linear regression (LR) and multiple linear regression (MLR) models were built for comparison
163 against the machine-learning algorithms. We also tested the performance of our RFR and ANN models against the
164 published algorithms of Simó & Dachs (2002), Watanabe et al. (2007), Vallina & Simó, (2007), and Galí et al.
165 (2018) (hereafter referred to as SD02, W07, VS07, and G18, respectively). SRD is calculated here using MLD as
166 described by Vallina & Simó (2007):

$$167 \quad SRD = \frac{PAR}{K_d \times MLD} \times (1 - e^{-K_d \times MLD}) \quad (4)$$

168 Each of the four algorithms was assessed using both their original coefficients and coefficients tuned to
169 our NESAP dataset using nonlinear least-squares optimization.

170 2.5 Controls on DMS variability

171 Principal component analysis (PCA) was applied to assess the relationships between DMS and the nine
172 predictors used to build the RFR and ANN ensembles. Additionally, non-parametric spearman rank correlations
173 were calculated between each variable and both the modelled and observed DMS concentrations. Correlation
174 analysis was also extended to assess the role of taxonomy on predicted DMS concentrations, using the outputs of
175 a chlorophyll-a based taxonomic algorithm by Hirata et al. (2011) with NESAP-tuned coefficients (Zeng et al.,
176 2018).

177 2.6 Sensitivity Tests and Predictor Selection

178 To inform our selection of grid size, we assessed the performance of both the RFR and ANN models using
179 grid cells ranging from 0.25 to 5° (Fig. 1). From this analysis, we found that model accuracy was highest at 0.25°
180 resolution (see Sect. 3.1). Smaller grid sizes would presumably further improve model accuracy, but at a
181 significantly higher computational cost.

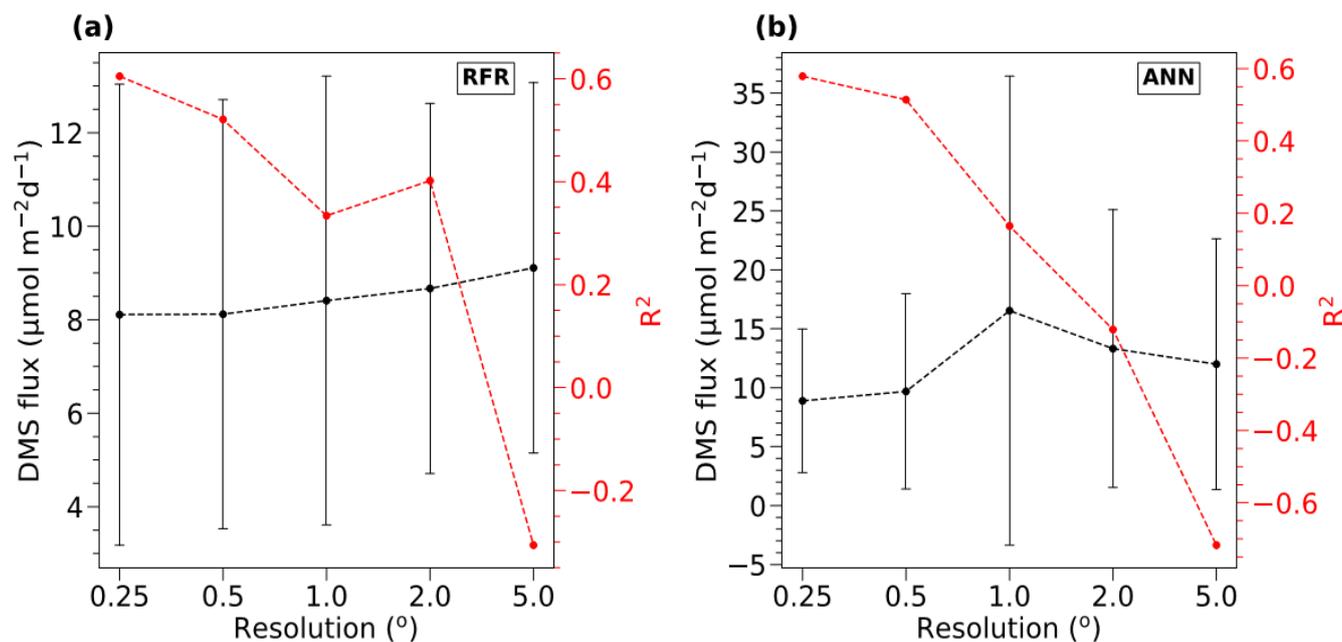


182 We also tested the influence of other biological predictor variables on the performance of the RFR and
 183 ANN models, using either NCP, NPP, Chl-a, or PIC. These sensitivity tests indicated no significant difference
 184 between the various biological predictor variables, although accuracy was slightly reduced when PIC was used.
 185 We therefore selected NCP as the biological predictor variable within our model framework. We also removed K_d
 186 as a predictor variable after further sensitivity testing indicated that its exclusion slightly improved results.

187 The inclusion of nFLH:Chl-a represents a proxy for iron limitation (see Sect. 4.1). However, fluorescence
 188 yields corrected for non-photochemical quenching (NPQ) have been suggested to yield a better iron limitation
 189 proxy than nFLH:Chl-a (Behrenfeld et al., 2009). We therefore calculated NPQ-corrected fluorescence yields (φ_f)
 190 by:

$$191 \quad \varphi_f = \frac{nFLH}{Chl-a \times \alpha \times S} \times \frac{iPAR}{iPAR} \quad (5)$$

192 where $\alpha = 0.0147 \times Chl - a^{-0.316}$ and $S = 100 \text{ mW cm}^{-2} \mu\text{m}^{-1} \text{sr}^{-1} \text{m}$ as described by Behrenfeld et al. (2009). Our
 193 tests indicated nFLH:Chl-a yielded slightly improved performance overall whereas φ_f decreased both models'
 194 performance. We therefore retained nFLH:Chl-a and excluded φ_f in our final model design.



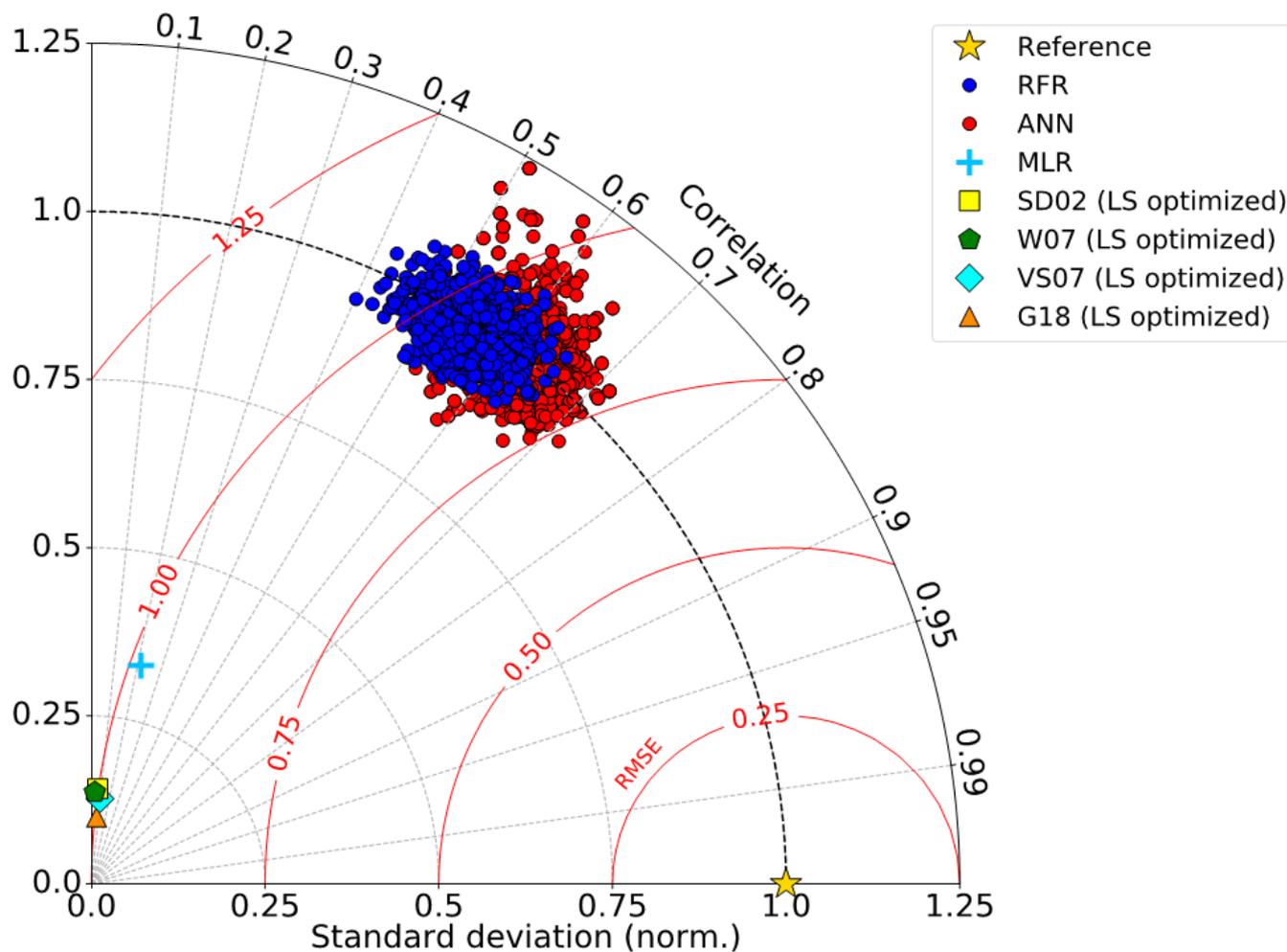
195
 196 Fig. 1. Sensitivity of RFR and ANN models to grid size resolution. DMS fluxes (green) and R^2 values (red) derived from sensitivity
 197 tests of (a) RFR and (b) ANN models to pixels resolutions of 0.25-5 $^\circ$. The negative R^2 values observed at the lowest resolution (largest
 198 grid cells) indicate that the predicted values explain less variance than the overall mean of the dataset.



199 **3 Results**

200 **3.1 Model evaluation**

201 To benchmark the performance of our RFR and ANN models, we first evaluated the predictive skill of four
202 existing empirical DMS algorithms (SD02, W07, VS07, & G18), in addition to simple and multiple linear
203 regression models. Previous studies have demonstrated that these empirical algorithms show strong predictive skill
204 ($R^2=0.53-0.84$) over large scales and in some oceanic regions (Simó and Dachs, 2002; Galí et al., 2018; Watanabe
205 et al., 2007), but significantly poorer performance in the NESAP (Herr et al., 2019). Consistent with these results,
206 we found that the SD02, W07, VS07, and G18 did not accurately predict NESAP DMS distributions, even with
207 regionally tuned coefficients (Fig. 2, $R^2=0-0.01$). We also found that simple and multiple linear regressions
208 performed poorly ($R^2=0-0.05$; Fig. 2, 3), yielding virtually no explanatory power for surface water DMS
209 distributions in the NESAP ($R^2\leq 0.05$).



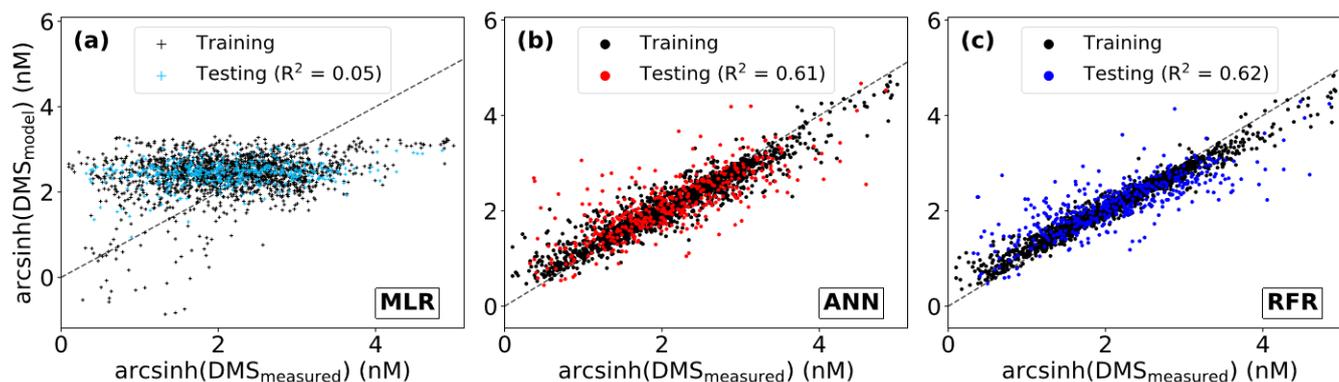
210

211 **Fig. 2.** Taylor Diagram showing comparative performance metrics of each individual Random Forest Regression (RFR)
212 and Artificial Neural Network (ANN) model (1000-model ensembles) against multiple linear regression (MLR) and
213 other statistical DMS models (See sections 2.1 and 2.4). The Pearson correlation coefficients (“Correlation”; outer
214 radius), root mean squared error (“RMSE”; red radial contours), and standard deviations (SDs; grey radial contours
215 from origin) are all computed with respect to the observed DMS samples after inverse hyperbolic sine (IHS)
216 transformation. The reference of a perfect model fit is shown with a gold star. SDs of the model outputs are normalized
217 to the SDs of the DMS observations. RMSE represents a normalized trigonometric derivation from both the correlation
218 coefficients and normalized SDs. Performance of the SDO2, W07, VS07, and G18 algorithms reported here are
219 calculated using regionally tuned coefficients to the NESAP derived from non-linear least-squares optimization (see
220 section 2.4).

221 Relative to other published modelling approaches, both the RFR and ANN models dramatically improved
222 the representation of NESAP DMS variability, achieving significantly higher predictive accuracy (Fig. 2, 3). The
223 collective ensembles of both the RFR and ANN models yielded strong performance, explaining up to 62% of the
224 observed DMS variability ($R^2=0.61-0.62$; Fig. 3). For individual models within the ensembles, the ANN method



225 provided slightly better results ($R^2=0.16-0.50$), compared to the individual RFR models ($R^2=0.16-0.43$). As
226 observed for predicted DMS concentrations, the models showed lower predictive power for sea-air DMS fluxes at
227 coarser resolution (Fig. 1).



229 **Fig. 3. Performance of three modelling approaches in predicting observed DMS distributions; (A) multiple linear**
230 **regression (MLR) (B) ensemble of Artificial Neural Networks (ANN) and (C) ensemble of Random Forest Regression**
231 **(RFR). For consistency, all predictions are partitioned by the Training and Testing datasets used to build the ensembles**
232 **(see section 2.2). Model performance (R^2) is computed only for the Testing dataset predictions. The dashed line**
233 **demonstrates a 1:1 relationship.**

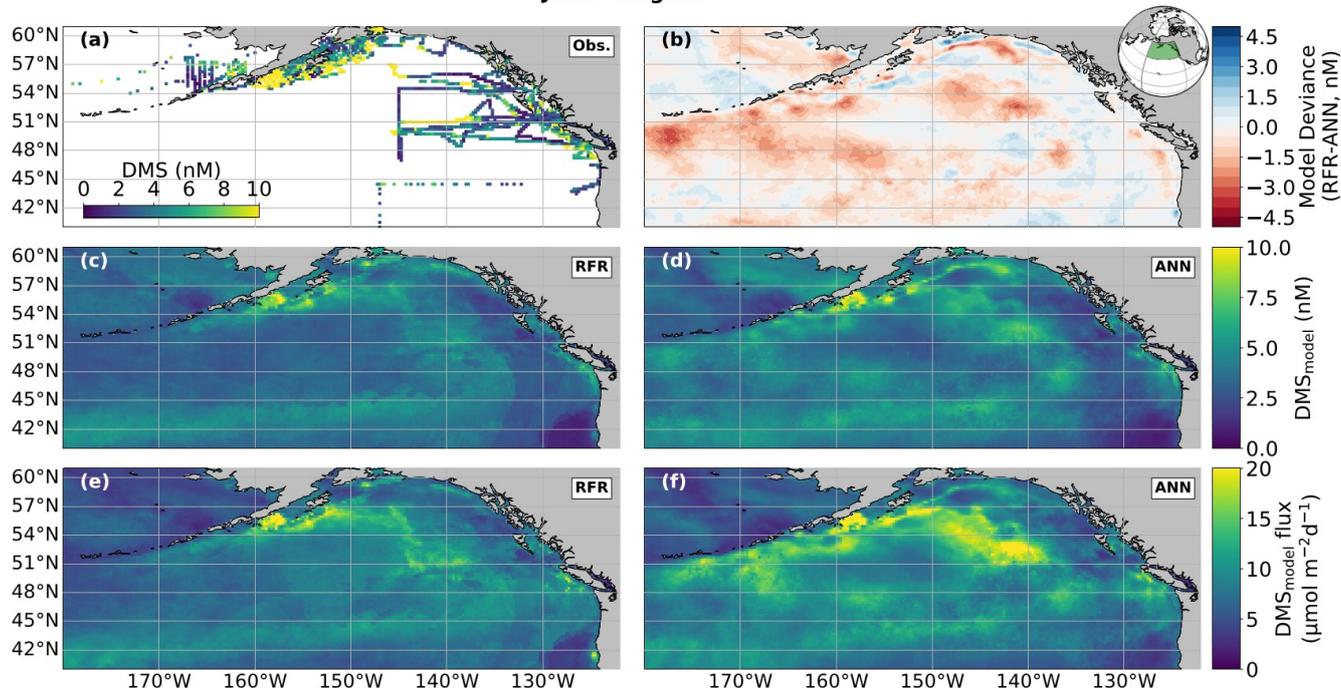
234 3.2 DMS distributions and sea-air fluxes

235 The predicted spatial distribution of DMS was generally consistent between observations and the RFR and
236 ANN methods (Fig. 4a,c,d). The average model derived DMS concentrations was 4.0 ± 2.1 nM and 4.7 ± 3.0 nM
237 (mean \pm SD) for the RFR and ANN ensemble models, respectively, with a similar range from 0.3 to 84.3 nM. In
238 both models, the highest DMS concentrations were largely constrained to coastlines and within the Alaska Gyre
239 adjacent to the Aleutian Islands (Fig. 4b-c, 8C). The greatest discrepancy between DMS concentrations from the
240 two models was observed in these regional ‘hotspots’, where the ANN models emphasize high DMS within the
241 Alaska gyre, while the RFR models emphasize elevated coastal DMS concentrations (Fig. 4b). The models deviated
242 on average by 0.49 nM, with the greatest offsets observed in an area of particularly sparse DMS observations in
243 the Alaska Gyre (Fig. 4a,b). Future observational data in this region should help improve model performance.

244



June – August



245

246 **Fig. 4. Predicted maps of sea surface DMS concentrations and sea-air fluxes. (a) Ship-based observations of mean summertime**
247 **(June-August) DMS concentrations used to construct the predictive models. (b) Differences between the (c) Random Forest**
248 **Regression (RFR) and (d) Artificial Neural Network (ANN) ensemble predicted DMS concentrations. (e,f) DMS sea-air fluxes**
249 **derived from the predicted DMS concentrations. Colormap ranges are restricted to illustrate trends, with <1% of DMS data**
250 **exceeding the colorbar limits. The inset map in (b) shows the NESAP study region as a shaded green patch in a global orthographic**
251 **projection.**

252

253 Sea-air DMS fluxes (Fig. 4e,f) derived from ANN predictions were 18% higher, on average, than RFR
254 predictions, largely due to higher predicted values in the Alaska Gyre (Fig. 4d-e, Table 2). The distribution of ANN
255 sea-air fluxes was also closer to ship-based observations (Fig. 5). Predicted regional fluxes ranged from 0.7 to 107
256 $\mu\text{mol m}^{-2}\text{d}^{-1}$ between the two models (Fig. 4e,f, 5), with the highest predicted DMS emissions in August, when
257 derived sea-air fluxes were approximately 1.5-fold greater than in June and July (Table 2). Our models yielded a
258 summertime integrated sea-air flux of 0.31 ± 0.19 Tg DMS-derived sulfur (equivalent to 0.5 to 2.0 Tg S yr^{-1} ; Table
259 2), in good agreement with recent estimates based on compiled ship-based observations (0.3 Tg; Herr et al., 2019)
260 and existing climatological estimates (Table 2; Lana et al., 2011). This summertime mean value is equivalent to
261 $\sim 4\text{-}8\%$ of total global DMS sea-air emissions annually, assuming an uncertainty ranging between 15 to 28 Tg S yr^{-1}
262 1 in global estimates (Bock et al., 2021). This result further emphasizes the NESAP as a globally significant DMS
263 source to the atmosphere.

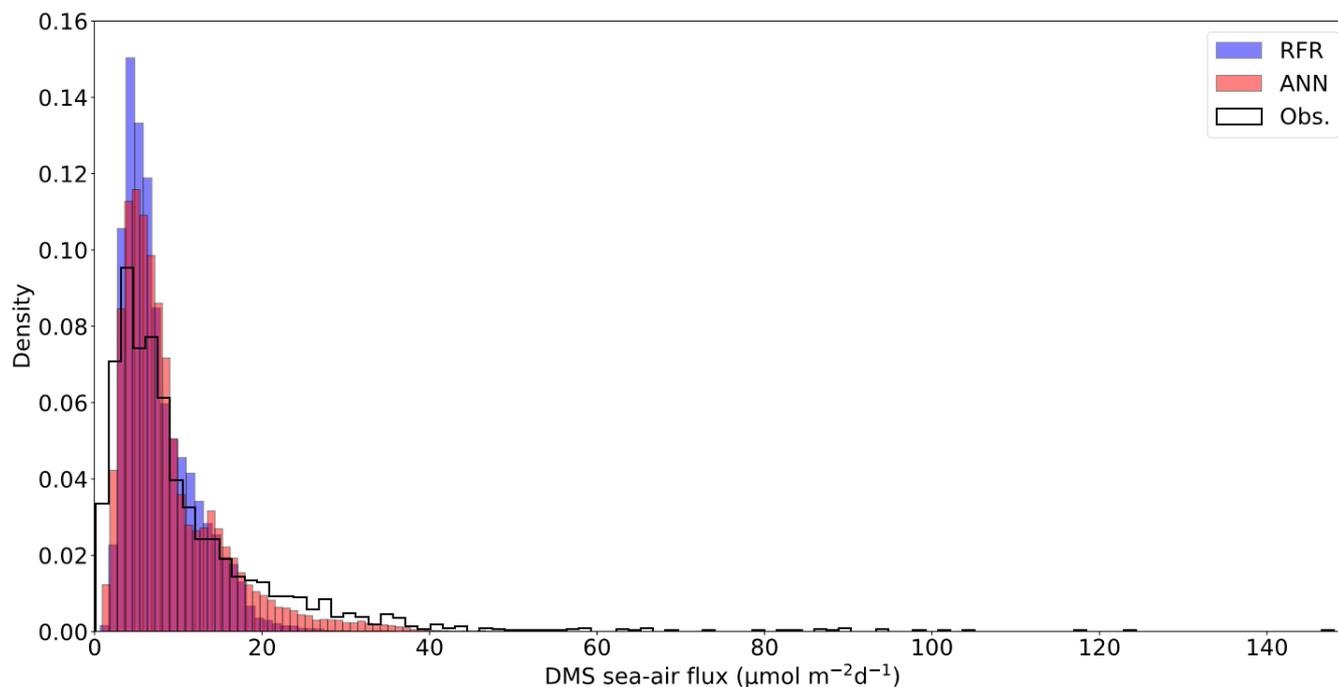


264 **Table 2. Monthly and mean summertime NESAP sea-air DMS fluxes. Fluxes (mean ± SD) are calculated from the**
 265 **Random Forest Regression (RFR) and Artificial Neural Network (ANN) model predictions (based on an ensemble of**
 266 **2000 models). NESAP sea-air flux derived from the Lana et al. (2011) climatology is shown for comparative purposes.**

267

	Annual Sulfur Emissions			
	RFR	ANN	This Study	Lana et al. (2011)
	$\mu\text{mol m}^{-2} \text{d}^{-1}$	$\mu\text{mol m}^{-2} \text{d}^{-1}$	Tg S	Tg S
June	5.9 ± 3.7	6.0 ± 3.9	0.22 ± 0.13	0.44 ± 0.20
July	6.5 ± 3.0	7.7 ± 3.8	0.26 ± 0.12	0.33 ± 0.17
August	10.8 ± 3.0	14.0 ± 3.8	0.45 ± 0.21	0.54 ± 0.21
June-August	7.7 ± 2.4	9.2 ± 3.4	0.31 ± 0.19	0.44 ± 0.21

268



269

270 **Fig. 5. Histograms of DMS sea-air flux distributions derived from the 1000-model ensemble random forest regression (RFR) and**
 271 **artificial neural network (ANN) predictions as well as cruise observations (Obs.). The sample sizes of both models are equivalent**
 272 **(n= 49,632) and are significantly higher than the observational dataset (n=2063). Note that the ANN better predicts the upper tail of**
 273 **DMS observations greater than 20 nM.**

274

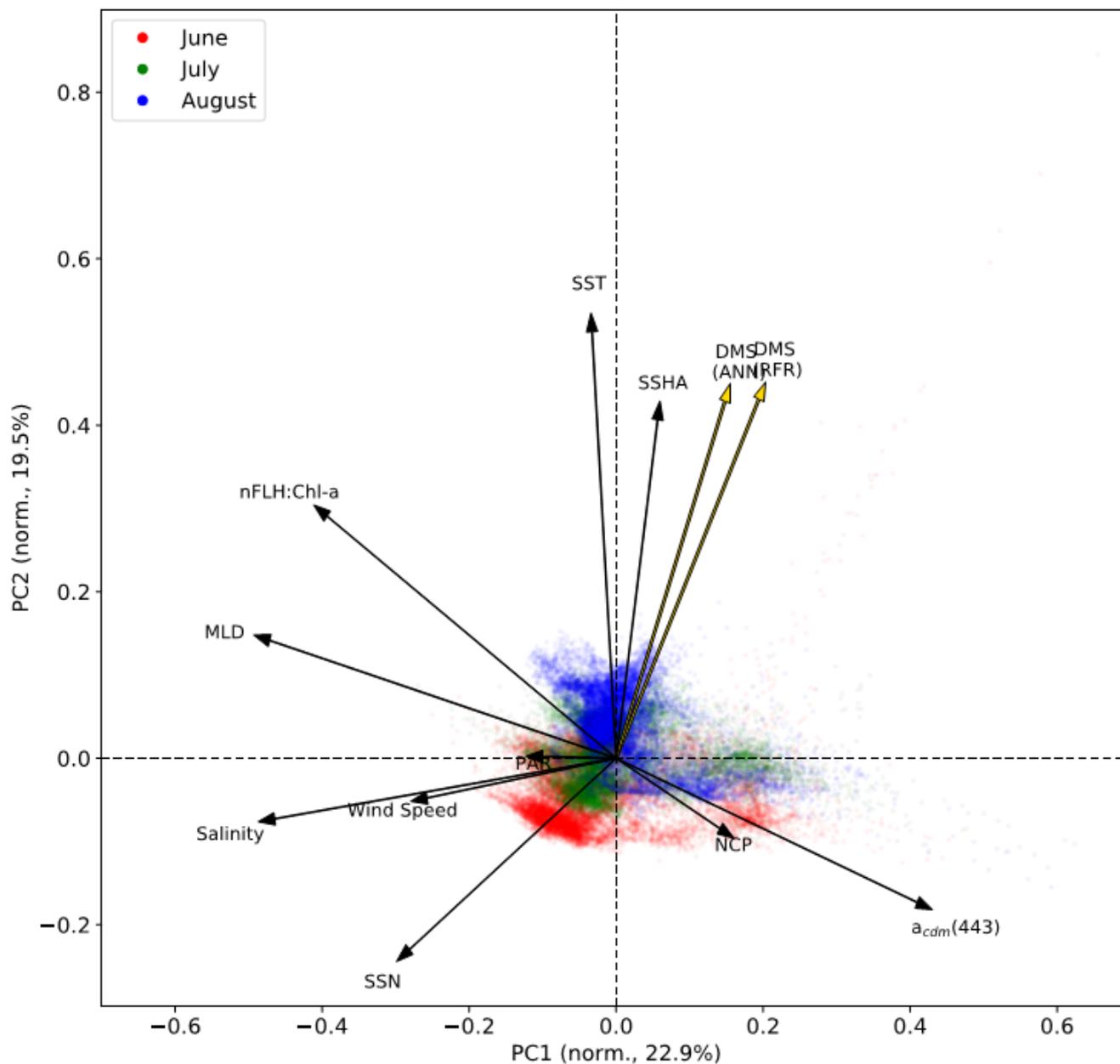


275 3.4 Drivers of DMS variability

276 In addition to modelling the spatial and temporal distribution of surface water DMS in the NESAP, we
277 examined the influence of different oceanographic variables as model predictors. As expected based on previous
278 work (Herr et al., 2019), no single predictor was found to exert a dominant control on modelled DMS distributions
279 from either the RFR or ANN models (Fig. 6, 7). Rather, the relationship between DMS and other oceanographic
280 variables exhibited significant region-specific patterns. One of the most compelling regional signatures was the
281 apparent relationship between DMS and SSHA. In both models, we found significant positive correlations between
282 DMS and SSHA ($\rho=0.35, 0.41$ for RFR and ANN, respectively) across the full spatial domain, with a particularly
283 notable relationship along the northern Alaskan coastline (Fig. 8, 9). Here, strong winds (Fig. 9j-l), coupled with
284 the northeastern Alaska current flow, produce two characteristic oceanographic features in the NESAP: strong,
285 semi-permanent mesoscale eddies collectively referred to as the Haida, Sitka and Yakutat eddies (Fig. 8a), and the
286 formation of the high nutrient, low chlorophyll (HNLC) Alaska Gyre (Fig. 8c; Okkonen et al., 2001; Whitney et
287 al., 2005). Both the monthly (Fig. 9a-i) and summertime-averaged (Fig. 8a,b) RFR and ANN-derived DMS
288 concentrations are low where these downwelling eddies form. In contrast, elevated DMS concentrations were
289 associated with the negative SSHA coastal upwelling areas (Fig. 8a,b), where phytoplankton productivity is
290 stimulated by nutrient inputs into the mixed layer.

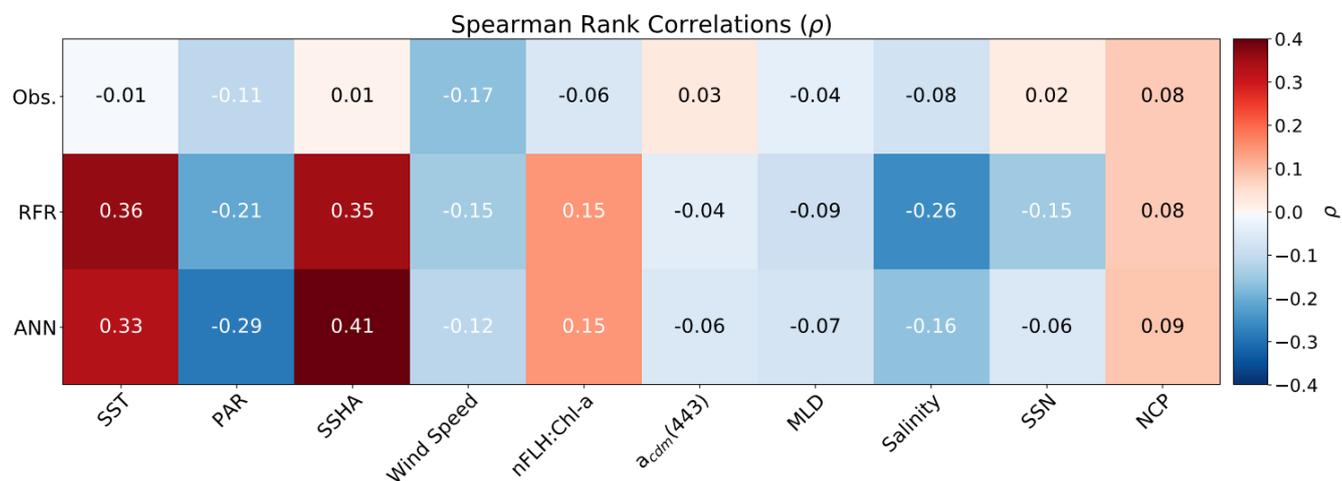


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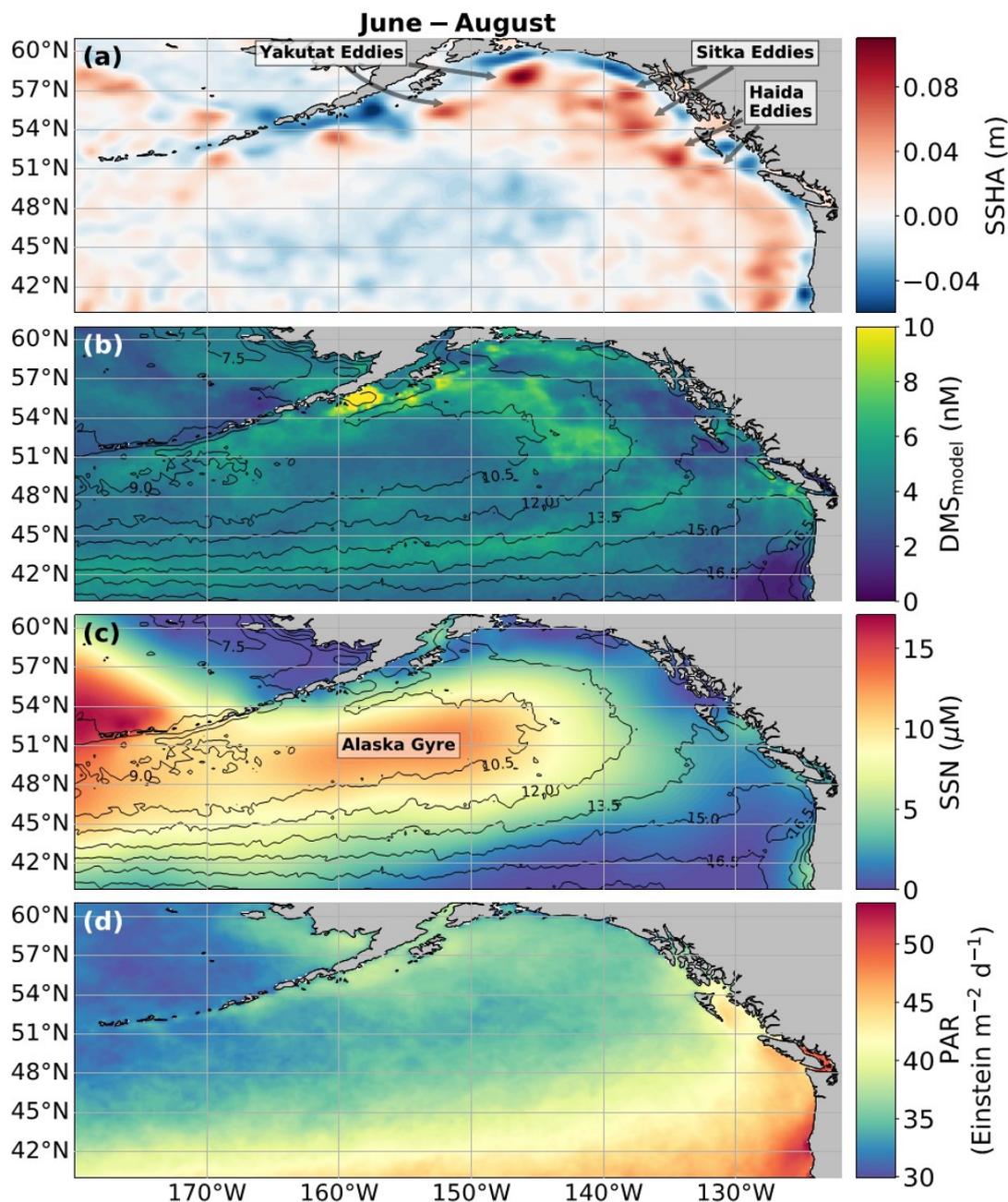
293 **Fig. 6. Principal Component Analysis (PCA) showing the relationships between variables used to construct the**
294 **predictive algorithms. Eigenvectors (arrows) are superimposed over the principal components (PCs; data points) for**
295 **the first two significant modes obtained from PCA. PCs are normalized and clustered by month (June-August, see**
296 **legend for colors), while the eigenvectors are grouped by ensemble model predictions (gold) and nine predictor**
297 **variables (black). The percentage of variance explained by each mode is indicated along the axes.**



298

299 **Fig. 7. Heatmap of Spearman rank correlations (ρ). Top row: observed DMS concentrations; middle row: RFR model**
 300 **predictions; bottom row: ANN model predictions. All model correlations are computed on the 1000-model ensembles.**

301 Modelled DMS concentrations also significantly correlated with hydrographic frontal patterns. We found
 302 significant correlations between DMS and SST ($\rho=0.36, 0.33$ for RFR and ANN, respectively) which suggested
 303 the central Alaska Gyre is an area of elevated DMS variability. Both models predict high DMS levels in the northern
 304 frontal zone of the gyre (140°W-145°W) between the 10.5 and 12°C isotherms and the southern frontal zone
 305 between (42°N-45°N) between the 13.5 and 15°C isotherms (Fig. 8b,c). By comparison, our models suggest that
 306 DMS concentrations are predominantly low in relation to high sea surface nitrate (SSN) concentrations within the
 307 HNLC gyre (Fig. 8, 9). As discussed below, the relationship between DMS and macronutrient concentrations in
 308 the HNLC waters of the central Gulf of Alaska could indicate an important role for iron limitation as a controlling
 309 factor in the DMS cycle. The presence of elevated summer nutrients in offshore waters is taken as a proxy for iron
 310 limitation, which increases over the course of the summer growing season.



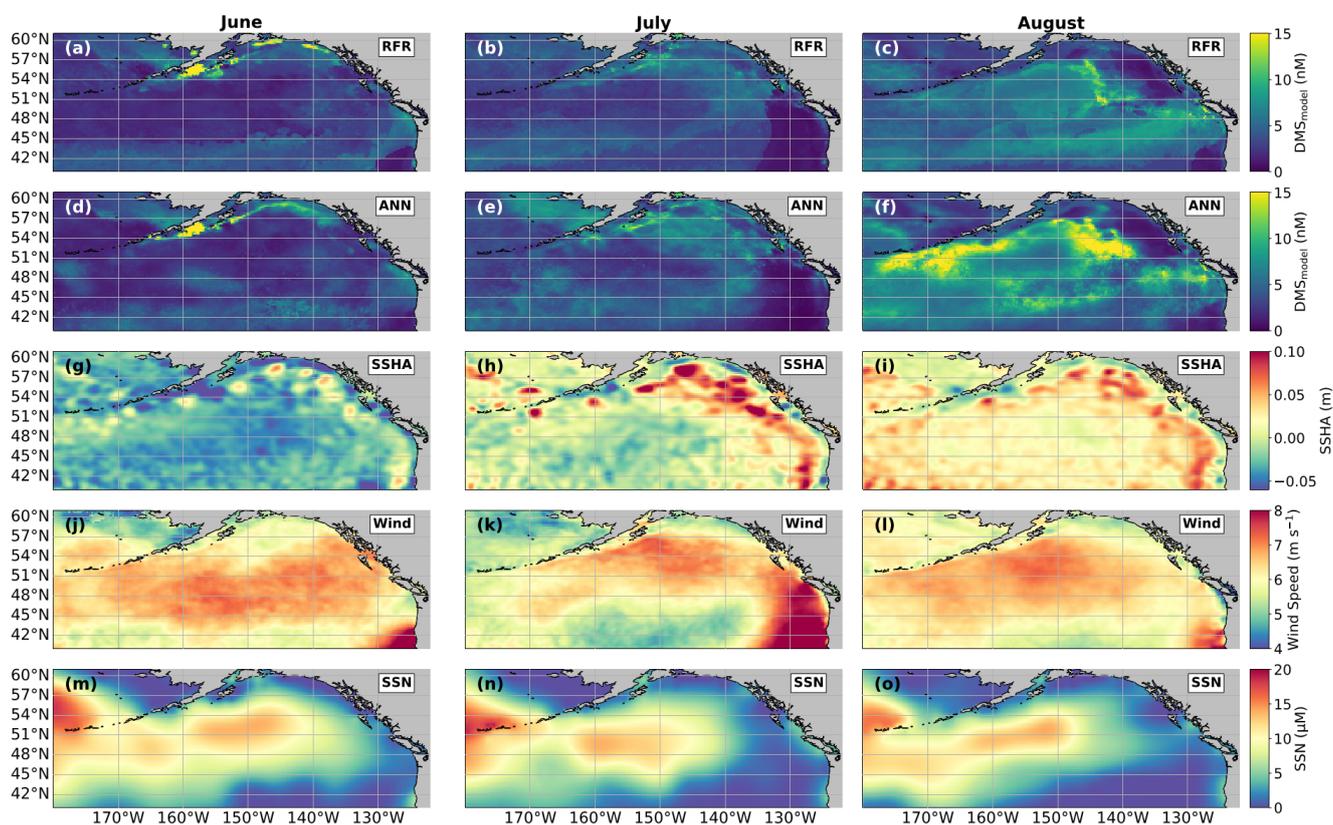
311

312 Fig. 8. Physical drivers of summertime (June-August) NESAP DMS distributions. (a) Sea surface height anomalies
313 (SSHA), (b) predicted DMS concentrations derived from the mean of all 2000 RFR and ANN machine learning models,
314 (c) sea surface nitrate (SSN) and (d) photosynthetically active radiation (PAR). Contours in (b,c) show sea surface
315 temperature (SST) isotherms. Coherent features of elevated sea-surface height indicate the presence of mesoscale
316 eddies, whereas nearshore low SSHA features reveal areas of upwelling. Colormaps ranges are restricted to illustrate
317 trends with <1% of data exceeding the colorbar limits.



318 Other variables appear to exhibit a more localized or minimal influence on DMS cycling. For instance,
319 both NCP and DMS are elevated in productive nearshore waters, but NCP generally correlates weakly with both
320 RFR- and ANN-derived DMS concentrations ($\rho=0.08$, 0.09 for RFR and ANN, respectively). Similarly, modelled
321 phytoplankton taxonomic composition (Hirata et al., 2011; Zeng et al., 2018) was not significantly correlated with
322 predicted DMS concentrations ($\rho<0.1$). Although strong, persistent winds appear to sustain low DMS
323 concentrations off the coast of Oregon and Vancouver Island (Fig. 9), wind speeds only weakly correlate with DMS
324 overall for the region ($\rho=-0.15$ and -0.12 for RFR and ANN, respectively). Additionally, high PAR in these areas
325 correspond with low DMS concentrations (Fig. 6d) and there is an overall negative correlation between PAR and
326 DMS for the region (Fig. 6, 7; $\rho=-0.21$ and -0.29 for RFR and ANN, respectively). Finally, despite hypothesized
327 links between DMS cycling and iron limitation in the NESAP (Levasseur et al., 2006; Merzouk et al., 2006),
328 nFLH:Chl-a ratios (taken as a proxy for phytoplankton iron stress; Behrenfeld et al., 2009; Westberry et al., 2013)
329 did not exhibit any coherent spatial patterns and only weakly correlated to our modelled DMS concentrations
330 ($\rho=0.15$ for both RFR and ANN, respectively).

331



332

333 **Fig. 9.** Predicted spatial and temporal (June-August) DMS distribution in relation to underlying oceanographic
334 variables. DMS concentrations predicted from (a-c) the Random Forest Regression (RFR) and (d-f) the Artificial
335 Neural Network (ANN) ensemble models are mapped alongside the monthly-averaged (g-i) sea surface height anomalies
336 (SSHA), (j-l) wind speeds (Wind), and (m-o) sea surface nitrate (SSN) for each month. Colormap ranges are restricted
337 to illustrate trends, with at most 1.5% of the data beyond the colorbar limits.

338 4 Discussion

339 The relative sparsity of DMS data in many oceanic regions and the complexity of DMS cycling have limited
340 previous attempts to model oceanic distributions of this compound (Simó and Dachs, 2002; Vallina and Simó,
341 2007; Galí et al., 2018; Watanabe et al., 2007; Herr et al., 2019). Taking advantage of expanding data resources,
342 we employed a new approach to statistically describe DMS distributions in the NESAP. Our results show that both
343 our RFR and ANN models substantially improved predictive strength over traditional empirical approaches (Fig.
344 2, 3), while identifying several key DMS relationships and regional patterns across the NESAP (Fig. 8, 9). Although
345 our statistical approach does not directly elucidate the underlying mechanisms driving these relationships, we can



346 nonetheless make some reasonable inductive inferences. These inferences are discussed below, along with the
347 implications of the improved predictive performance observed here.

348 **4.1 Relationships with other oceanographic variables**

349 Among the more prominent spatial relationships we observed was the coherence between predicted DMS
350 concentrations and SST, and the negative correlation between predicted DMS concentrations and sea surface nitrate
351 (SSN) within and surrounding the Alaska Gyre (Fig. 6-9). The DMS-nitrate relationship may be partially explained
352 by the so-called sulfur overflow hypothesis (Stefels, 2000), which suggests that nutrient-limited phytoplankton
353 increase DMSP production and its subsequent cleavage to DMS, in order to regulate intracellular sulfur quotas
354 when protein synthesis is limited (Hatton & Wilson, 2007; Kinsey et al., 2016; Simó & Vila-Costa, 2006; Spiese
355 & Tatarkov, 2014; Stefels, 2000). This pathway may help explain the higher predicted DMS concentrations
356 predicted at the northern extent of the Alaska Gyre, where SSN concentrations begin to decrease (Fig. 6). The
357 apparent relationship between DMS and nitrate could also result indirectly from the underlying effects of iron
358 limitation. Excess summertime nitrate concentrations are taken as evidence for iron limitation in the NESAP (Boyd
359 and Harrison, 1999; Boyd et al., 2004; Martin and Fitzwater, 1988; Whitney et al., 2005). Under iron-limiting
360 conditions, DMS is thought to function, together with DMSP and DMSO, as part of an antioxidant response to
361 oxidative stress (Sunda et al., 2002). This hypothesis suggests that iron limitation should stimulate net production
362 of DMS and DMSP (Bucciarelli et al., 2013; Sunda et al., 2002), which is inconsistent with the negative dependence
363 predicted between DMS and SSN (Fig. 8b,c).

364 Satellite-based, chlorophyll-normalized fluorescence has been suggested as an additional proxy for iron
365 limitation. Low iron conditions can lead to both a reduction in photosystem I relative to photosystem II (Strzepek
366 and Harrison, 2004), and an apparent increase in energetically-decoupled light harvesting complexes (Allen et al.,
367 2008; Behrenfeld & Milligan, 2013), resulting in elevated fluorescence-to-chlorophyll a ratios (nFLH:Chl-a)
368 (Westberry et al., 2013). To our knowledge, this proxy has not been widely investigated with respect to DMS
369 cycling. In our analysis, we found that nFLH:Chl-a ratios, and the NPQ-corrected fluorescence yields (ϕ_f), exhibited
370 only weak positive correlations with the RFR and ANN predicted DMS concentrations (Fig. 6, 7). Moreover,
371 neither of these metrics exhibited coherent spatial patterns with predicted DMS concentrations, suggesting a limited
372 role for iron in driving spatial patterns of DMS cycling within the NESAP. However, it is important to note the
373 potential temporal mismatch between our monthly DMS predictions and these more instantaneous metrics of iron
374 limitation, which reflect short-term physiological changes (days to weeks; (Behrenfeld et al., 2009; Westberry et
375 al., 2019) that depend on sporadic iron loading (e.g. aerosol deposition; Mahowald et al., 2009). Indeed, both natural



376 and artificial iron-fertilization events have thus far been detected from satellite-derived nFLH:Chl-a at daily
377 resolution (Westberry et al., 2013), in contrast to the monthly-averaged data used here. Therefore, modelling
378 frameworks utilizing shorter temporal scales may find a clearer connection between DMS cycling and iron
379 limitation using the chlorophyll-a fluorescence proxy.

380 Beyond nutrient limitation effects, ambient light fields are believed to exert significant direct and indirect
381 effects on DMS cycling (del Valle et al., 2007). Ultraviolet radiation has been noted to induce high DMS production
382 and turnover through a proposed cascading oxidation pathway, which acts to remove harmful reactive oxygen
383 species (Sunda et al., 2002; Archer et al., 2010). In contrast, more recent evidence has indicated the potential for
384 elevated DMS production in the NESAP from the reduction of DMSO due to light-induced oxidative stress over
385 diurnal cycles (Herr et al., 2020). However, our modelled DMS concentrations exhibited a negative correlation
386 with PAR (Fig. 6, 7), suggesting that incident light may predominantly drive DMS loss in the NESAP through
387 photolysis (del Valle et al., 2007) on regional and longer-term scales.

388 Since DMS does not have strong light absorption properties, the presence of photosensitisers is necessary for
389 the abiotic photooxidation of DMS (Brimblecombe and Shooter, 1986). To account for this process, our models
390 incorporated nitrate (SSN) and $a_{cdm}(443)$ (as a proxy for CDOM; Nelson & Siegel, 2013), both of which are thought
391 to be dominant photosensitisers of DMS in marine systems (Taalba et al., 2013; Bouillon and Miller, 2004, 2005;
392 Galí et al., 2016). In the NESAP, nitrate appears to exert a stronger influence than CDOM on the apparent quantum
393 yields (AQY) of DMS (Bouillon and Miller, 2004). In support of this, our results suggest a stronger negative
394 dependence of predicted DMS concentrations on nitrate compared to CDOM within the NESAP (Fig. 6, 7). We
395 note, however, that the DMS-nitrate relationship likely also reflects physiological impacts of nutrient limitation, as
396 discussed above. Nonetheless, our results are consistent with elevated rates of DMS photo-oxidation in the nitrate-
397 replete, low iron waters of the Alaska Gyre, where photolysis, coupled with potentially high DMS oxidation rates
398 due to iron-induced oxidative stress (Sunda et al., 2002), may explain the low predicted DMS concentrations (Fig.
399 8, 9). Further *in situ* work will be required to resolve the relative contributions of these biotic and abiotic processes
400 to DMS cycling within these areas.

401 Among all the statistical relationships we observed, perhaps the most striking was the association of DMS
402 variability with SSHA, particularly along the Alaskan coast and in relation to mesoscale eddies (Okkonen et al.,
403 2001; Whitney et al., 2005; Fig. 8, 9). To our knowledge, only one other study has linked SSHA to DMS within
404 the NESAP. Herr et al., (2019) demonstrated contrasting positive and negative correlations between DMS and
405 SSHA in offshore and coastal waters, respectively, in general agreement with our results. Presently, the underlying
406 mechanisms explaining the relationship between SSHA and DMS cycling remain unclear, yet it is likely that



407 physical mixing processes are important. For example, enhanced biological production is known to be stimulated
408 by eddy re-supply of iron and macronutrients via vertical advection and diffusion (Whitney et al., 2005; Bailey et
409 al., 2008). These nutrient supply processes would also be expected to influence DMS cycling, as outlined above.
410 Elevated abundances of high DMS-producers within eddies have been noted in the Sargasso Sea (Bailey et al.,
411 2008), while eddy-induced vertical transport likely supplements nearshore, current-driven upwelling that can also
412 resupply iron into the coastal waters of the NESAP (Cullen et al., 2009; Freeland et al., 1984). In addition, eddy
413 propagation can allow cross-shelf transport, distributing micronutrients to offshore waters (Fiechter and Moore,
414 2012), potentially contributing to the apparent elevated DMS concentrations in the outer Alaska gyre between the
415 10.5 and 12°C isotherms (Fig. 8). These mixing and transport mechanisms could partially explain the influence of
416 elevated productivity in driving increased nearshore and northern NESAP DMS concentrations (Fig. 4, 7-9),
417 representing a novel source of DMS variability in this region.

418 The taxonomic composition of plankton assemblages is also a likely source of variability influencing DMS
419 cycling. Significant changes to DMS production and consumption rates within the NESAP are expected in response
420 to variable microbial and phytoplankton taxonomy (Vila-Costa et al., 2006; Lidbury et al., 2016; Sheehan and
421 Petrou, 2020). Such taxonomic variability may, in turn, reflect transient community composition shifts in response
422 to mixing (Bailey et al., 2008), nitrate (Bouillon and Miller, 2004), and iron availability (Levasseur et al., 2006;
423 Merzouk et al., 2006). The monthly averaging used in our data processing removes autocorrelation associated with
424 individual sampling expeditions (Wang et al., 2020), but it may preclude capturing these transient taxonomic
425 responses. For instance, coccolithophores have long been believed to influence DMS cycling in the NESAP (Herr
426 et al., 2019; Asher et al., 2011), yet averaged calcite distributions did not yield increased predictive strength for
427 DMS concentrations in our analysis (see Sect. 2.6). Similarly, applying a chlorophyll-a based taxonomic algorithm
428 (Hirata et al., 2011; Zeng et al., 2018) yielded no further explanation of the DMS variability predicted. The
429 influence of taxonomic composition thus remains cryptic within our modelling framework.

430 **4.2 Implications of Improved Predictive Power**

431 As noted above, both the RFR and ANN approaches demonstrate significantly improved accuracy,
432 explaining up to 62% of observed DMS variability (Fig. 2, 3). This model performance is somewhat lower than
433 that achieved in the prediction of methane fluxes (Weber et al., 2019) and dissolved inorganic carbon dynamics
434 (Roshan and DeVries, 2017), where R^2 values ranging from 0.7 to 0.95 were obtained. Nonetheless, the dramatic
435 accuracy improvement of our algorithms over traditional methods (Fig. 2, 3) encourages the further use of these
436 techniques in modelling DMS distributions.



437 Improved predictive accuracy provides opportunities to gain insight into the mechanisms driving DMS cycling.
438 Our approach has yielded accurate DMS predictions at a 4 to 40-fold higher resolution than previous algorithms
439 (Simó and Dachs, 2002; Vallina and Simó, 2007; Galí et al., 2018; Watanabe et al., 2007), enabling the description
440 of mesoscale patterns and processes (Fig. 8). Extending these methods to sub-mesoscale resolution will enable
441 investigations into the dependence of DMS on finer-scale hydrographic processes, particularly stratification and
442 frontal dynamics, which have been increasingly linked to DMS cycling but remain unresolved mechanistically
443 (Asher et al., 2011; Royer et al., 2015). Moreover, coupling machine learning algorithms with biophysical and
444 tracer export models holds promise to resolve the contributions of eddy dynamics and upwelling intensity on DMS
445 variability, likely through nutrient availability and physiological mechanisms (Asher et al., 2011; Bailey et al.,
446 2008; Cullen et al., 2009). Recent work has also developed a new database of DMS apparent quantum yields (Galí
447 et al., 2016). As the availability of these measurements increases, simultaneous mapping of both DMS quantum
448 yields and concentrations will become feasible, enabling future studies to better parse out the contribution of
449 photolysis, physical mixing, and biological drivers of DMS cycling.

450 Although used in a diagnostic capacity here, our statistical models also hold potential for prognostic
451 applications. Frameworks utilizing shorter time scales will likely be able to detect underlying mechanisms
452 driving observed diel cycling (Galí et al., 2013; Royer et al., 2016), even if the underlying mechanisms are still
453 unresolved. We note, however, that caution will need be exercised as machine learning models have a tendency
454 to overfit noise (Weber et al., 2019; Roshan and DeVries, 2017; Wang et al., 2020), thus requiring appropriately
455 large training datasets and the use of known “future” observations to validate predictive accuracy in this context.
456 The significant variability in DMS cycling across oceanic regimes will likely also render predictions more
457 successful at regional, rather than global, scales (Galí et al., 2018; Royer et al., 2015). Nonetheless, prognostic
458 applications of these algorithms should be investigated to aid in the future development of improved mechanistic
459 models.

460 **5 Conclusions**

461 We have presented a statistical approach to modelling DMS distributions, which provides significantly
462 higher accuracy than traditional methods (Simó and Dachs, 2002; Vallina and Simó, 2007; Galí et al., 2018;
463 Watanabe et al., 2007; Lana et al., 2011), and yields estimates of the summertime NESAP DMS sea-air fluxes to
464 0.5-2.0 Tg S yr⁻¹ in agreement with previous findings (Herr et al., 2019; Lana et al., 2011). Our results further
465 underscore the importance of the NESAP to global DMS production and motivate further observations in



466 traditionally under-sampled areas such as the Alaska Gyre and Aleutian Islands. Although we are unable to directly
467 examine the mechanistic drivers of DMS variability, our findings suggest nutrient limitation, light-driven
468 processes, and eddy-induced mixing are potentially key drivers of DMS cycling in the NESAP. Future studies will
469 benefit from using such statistical algorithms, in conjunction with field-based process studies and mechanistic
470 models, to better understand the underlying dynamics and driving factors in the oceanic DMS cycle.

471 *Code availability.* The analysis in this study makes extensive use of the Numpy, Matplotlib, & Scikit-Learn libraries
472 in Python. The custom codes used can be downloaded at
473 https://github.com/bjmcnabb/DMS_Climatology/tree/main/NESAP or are available upon request from the
474 corresponding author.

475 *Data Availability.* DMS observations and predictor datasets are described in the Methods with relevant links to
476 repositories. Data from the Lana et al. (2011) climatology used for comparison in Table 2 are available via the
477 SOLAs project (retrieved from www.bodc.ac.uk/solas_integration/implementation_products/group1/dms/), where
478 the DMS sea-air fluxes were calculated as described in Sect. 2.3. The gridded climatologies produced from each
479 algorithm in this study can be obtained at
480 https://github.com/bjmcnabb/DMS_Climatology/tree/main/NESAP/Climatologies.

481 *Author Contribution.* BM and PT designed the study. Model code was written and implemented by BM. BM
482 prepared the manuscript with significant contributions from PT.

483 *Competing Interests.* The authors declare that they have no conflict of interest.

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487



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