

# **US SOLAS Science Plan**

## **TITLE PAGE**

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## Table of Contents

<b>1. Executive Summary .....</b>	<b>4</b>
<b>2. Introduction .....</b>	<b>7</b>
2.1 Background and history .....	7
2.2 Ocean-Atmosphere Interaction Committee .....	7
2.3 Workshop rationale and summary.....	8
<b>3. US Contributions to International SOLAS .....</b>	<b>9</b>
3.1 Theme 1. Greenhouse gases and the oceans .....	9
3.2 Theme 2. Air-sea interface and fluxes of mass and energy.....	11
3.3 Theme 3. Atmospheric deposition and ocean biogeochemistry.....	13
3.4 Theme 4. Interconnections between aerosols, clouds, and marine ecosystems .....	15
3.5 Theme 5. Ocean biogeochemical controls on atmospheric chemistry .....	17
<b>4. Crosscutting Topics and Research Questions.....</b>	<b>19</b>
4.1 Sea-surface microlayer.....	19
4.1.1 Role of bubbles in the formation and decay of the sea-surface microlayer .....	20
4.1.2 Particulate matter .....	20
4.1.3 Thermal and photochemical reactions.....	20
4.1.4 Microlayer sampling.....	21
4.2 Surfactants .....	21
4.2.1 Surfactant chemistry, including sources and sinks in the surface ocean .....	21
4.2.2 Surfactant physical properties and effects on SOLAS-relevant processes .....	22
4.3 Air-sea gas exchange.....	22
4.3.1 Turbulence is the fundamental driver of air-sea gas exchange.....	23
4.3.2 High wind speed regions .....	24
4.3.3 Coastal areas .....	24
4.3.4 Sea ice.....	24
4.4 Halogens, reactive nitrogen, and redox chemistry.....	25
4.4.1 Ocean-atmosphere interactions involving halogens .....	26
4.4.2 Ocean-atmosphere interactions involving reactive nitrogen .....	26
4.4.3 Oxidation state of the marine boundary layer and surface ocean .....	26
4.5 Air-Sea Exchange and Evolution of Atmospheric Aerosols .....	27
4.5.1 Physical drivers of open-ocean wind waves.....	28
4.5.2 Bubble plume dynamics and bubble rafts.....	28
4.5.3 Primary marine aerosol production and evolution .....	28
4.5.4 Aerosol nutrients and bioavailability.....	29
<b>5. Implementation.....</b>	<b>30</b>
5.1 Leveraging Existing Platforms and Programs.....	30
5.1.1 Atmospheric Towers.....	30
5.1.2 Autonomous Vehicles.....	30
5.1.3 Specialized Platforms .....	30

5.1.4 Lab Facilities .....	31
5.2 Field Campaigns: Past and Future .....	31
5.3 Remote Sensing Technologies .....	32
5.4 Numerical and Data Analysis Tools .....	32
5.5 Model development .....	33
5.5.1 Modeling of marine aerosols.....	33
5.5.2 Nutrient cycling models.....	33
5.6 Data Management .....	34
<b>6. Leadership, Coordination, and Communication .....</b>	<b>35</b>
<b>7. References.....</b>	<b>36</b>
<b>Appendix 1. 2019 US SOLAS Workshop Documents .....</b>	<b>45</b>

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# US SOLAS SCIENCE PLAN

## 1. Executive Summary

The Surface Ocean – Lower Atmosphere Study (SOLAS) (<http://www.solas-int.org/>) is an international research initiative focused on understanding the key biogeochemical-physical interactions and feedbacks between the ocean and atmosphere that are critical elements of climate and global biogeochemical cycles. Following the release of the SOLAS Decadal Science Plan (2015-2025) (Brévière et al., 2016), the Ocean-Atmosphere Interaction Committee (OAIC) was formed as a subcommittee of the Ocean Carbon and Biogeochemistry (OCB) Scientific Steering Committee to coordinate US SOLAS efforts and activities, facilitate interactions among atmospheric and ocean scientists, and strengthen US contributions to international SOLAS. In October 2019, with support from OCB, the OAIC convened an open community workshop, *Ocean-Atmosphere Interactions: Scoping directions for new research* with the goal of fostering new collaborations and identifying knowledge gaps and high-priority science questions to formulate a US SOLAS Science Plan. Based on presentations and discussions at the workshop, the OAIC and workshop participants have developed this US SOLAS Science Plan. The first part of the workshop and this Science Plan were purposefully designed around the five themes of the SOLAS Decadal Science Plan (2015-2025) (Brévière et al., 2016) to provide a common set of research priorities and ensuring a more cohesive US contribution to international SOLAS. The following high-level priorities emerged under each of the international SOLAS Science themes (Fig. 1, section 3):

### **Theme 1: Greenhouse gases and the ocean**

- Constrain key drivers of natural variability in air-sea fluxes (e.g., climate indices, volcanic eruptions, circulation, etc.)
- Expand capacity to observe and quantify carbon fluxes along the aquatic continuum and constrain impact of extreme events in land-ocean and coastal carbon fluxes
- Constrain uncertainties and temporal variability of processes underlying regional air-sea fluxes, particularly in high  $\Delta p\text{CO}_2$  regions (e.g., coastal upwelling regions, tropical and high latitude oceans, western boundary currents, etc.)
- Expand observational capabilities and coverage to better quantify fluxes of other potent GHGs (e.g., methane and nitrous oxide) and improve understanding of associated processes

### **Theme 2: Air-Sea Interface and Fluxes of Mass and Energy**

- Improve capacity to measure and quantify gas exchange in poorly measured environments with high uncertainty (high winds/waves, shallow coastal waters, regions with partial sea ice cover)
- Improve understanding of fundamental processes that determine gas transfer (waves, wave breaking, sea spray, bubbles, surfactants)
- Develop more sophisticated gas transfer parameterizations through improvement of existing techniques (e.g., eddy covariance), as well as application of remote sensing, numerical methods, and new findings from lab and field studies

### **Theme 3: Atmospheric Deposition and Ocean Biogeochemistry**

- Expand observational capabilities (e.g., autonomous, remote sensing, ship-based, time-series, etc.) and coverage (regions outside the North Atlantic) to better constrain atmospheric deposition fluxes on different spatial and temporal scales in a variety of environments
- Improve representation of atmospheric deposition processes in models
- Develop a mechanistic understanding of the biological and chemical factors that determine the bioavailability of deposited nutrients

- Develop reference materials, standardized sampling and laboratory protocols, and a recommended core set of parameters for field collection and pursue international intercalibration efforts to improve intercomparability of atmospheric deposition measurements

#### **Theme 4: Interconnection Between Aerosols, Clouds, and Marine Ecosystems**

- Conduct laboratory and field studies to better understand the processes that drive the production and air-sea exchange of volatile organic compounds (VOCs) and primary organic aerosol
- Explore the physical processes that help shape cloud condensation nuclei (CCN) properties of ocean-derived aerosols
- Examine remote sensing observables that can help link biological activity in the ocean to aerosols and clouds

#### **Theme 5: Ocean Biogeochemical Controls on Atmospheric Chemistry**

- Determine physical, photochemical and ecological controls on VOC production and removal
- Examine the impact of climate change and episodic events (e.g., hurricanes, algal blooms) on marine VOC production and emissions
- Study the biochemical basis for VOC formation

Workshop participants also identified crosscutting topics (Fig. 2, section 4) that span multiple SOLAS themes and are ripe for new discovery. Research activities on these topics such as collecting observations, expanding observational capabilities across platforms, conducting process studies in the laboratory and field, improving modeling capabilities and representation of key processes, and developing and applying new numerical approaches have the capacity to advance the field of air-sea research:

- **Sea-surface microlayer:** Priorities include the role of bubbles in its formation and decay, particulate matter composition and cycling, thermal and photochemical reactions, and development of standardized approaches for sampling.
- **Surfactants:** Priorities include identifying major sources and sinks (e.g., dissolved organic carbon (DOC) in dissolved, colloidal, and particulate forms) of marine surfactants and understanding their physical properties and associated effects on processes such as air-sea gas flux and aerosol growth and activation.
- **Air-sea gas exchange:** Priorities include studies of turbulence, a fundamental driver of air-sea gas exchange, as well as observations and process studies to improve parameterizations in high-wind speed, coastal, and sea ice-impacted regions.
- **Halogens, reactive nitrogen, and redox chemistry:** Priorities include more measurements of reactive bromine, chlorine, and iodine in the marine troposphere and surface ocean, shipboard investigations of alkyl nitrate formation and associated photochemical and biological processes, and ship- and laboratory-based studies aimed at identifying key processes that determine the oxidative state of the marine boundary layer and surface ocean.
- **Air-sea exchange and evolution of atmospheric aerosols:** Priorities include improved characterization of open-ocean wind wave and bubble plume dynamics and associated impacts on primary marine aerosol (PMA) production and air-sea exchange, improved understanding of how the ocean's physicochemical and biological state affects PMA properties and how those properties evolve during atmospheric transport, and better constraints on the bioavailability of aerosol-borne nutrients deposited in the surface ocean.

Implementation of US SOLAS research priorities outlined here will require a combination of community-led efforts and sustained communication and coordination to strengthen and expand the US air-sea research community and ensure robust contributions to SOLAS. Overarching recommendations include improved and expanded observations and platforms; broadening applications of airborne and

satellite remote sensing datasets; field and laboratory-based process studies; large community-coordinated interdisciplinary field campaigns; more advanced models, numerical and data analysis tools; establishment of community-vetted standard operating procedures and best practices; and universal adoption of FAIR (Findable, Accessible, Interoperable, Reusable, Wilkinson et al., 2016) data practices. Continued investments in leadership (e.g., OAIC), coordination of community workshops and activities, and support and capacity building of the next generation of air-sea researchers will ensure a productive and vibrant US SOLAS community in the future.

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## 2. Introduction

### 2.1 Background and history

The Surface Ocean – Lower Atmosphere Study (SOLAS) project (<http://www.solas-int.org/>) is an international research initiative focused on understanding the key biogeochemical-physical interactions and feedbacks between the ocean and atmosphere that are critical elements of climate and global biogeochemical cycles. SOLAS also seeks to link this knowledge with current societal issues. The following brief historical narrative of SOLAS, and US involvement in SOLAS progress is provided as context for the new US Science Plan that follows.

SOLAS is jointly sponsored by the Scientific Committee on Oceanic Research (SCOR), the international Commission on Atmospheric Chemistry and Global Pollution (iCACGP), and the World Climate Research Programme (WCRP). SOLAS was launched as a core project of IGBP (International Geosphere-Biosphere Programme) in 2004, became part of Future Earth in 2015, and currently has 30 scientific national and regional networks worldwide.

Initial discussions with SCOR and IGBP about SOLAS began in 1997 with a task team meeting in London that produced IGBP Bulletin No. 31, 1997. The first SOLAS International Open Science/Planning Meeting was held in Damp, Germany (February 2000) with strong representation from the US research community. An international scientific steering committee (SSC) was assembled in May 2000 to develop a Science Plan. US scientists served on the SOLAS SSC and helped shape the first international SOLAS Science plan (SOLAS, 2004). There has been continual US representation on the SOLAS SSC and a US National Representative throughout the history of SOLAS.

In 2001, the National Science Foundation (NSF)'s Chemical Oceanography Program made an award to support a US-SOLAS Workshop aimed at developing a US-SOLAS science plan with direct links to the growing international SOLAS initiative. The workshop considered several interrelated fundamental science issues in ocean-atmosphere biogeochemical coupling, including the sulfur cycle, radiation and photochemistry, CO<sub>2</sub> and other climate-relevant gases, atmosphere-ocean nitrogen cycling, and boundary layer physics. Additional funding from the National Oceanic and Atmospheric Administration (NOAA), the Office of Naval Research (ONR), and the National Aeronautics and Space Administration (NASA) culminated in the first US-SOLAS workshop in Potomac, MD (May 2001). About 70 US scientists contributed to discussions that led to the first US-SOLAS Science Plan completed later that same year. In 2006, with contributions from 28 SSC and Advisory members, US-SOLAS followed with an updated Science Implementation Plan (McGillis, 2006) that contained specific recommendations for future US-SOLAS research that linked to the international SOLAS project. While US atmospheric and oceanographic scientists have since continued to collaborate and make globally recognized contributions to the understanding of ocean-atmosphere interactions, the 2006 science implementation plan has not been revisited. US air-sea research has since pressed ahead largely in alignment with specific funding opportunities rather than under a common umbrella of national research priorities.

The international SOLAS project reformulated Midterm Strategies (Law et al., 2013) to focus attention on critical areas of air-sea research and its relevance to societal needs and later developed the SOLAS Decadal Science Plan (2015-2025) (Brévière et al., 2016). An increasingly active US community contributed to these plans, participated in domestic and international SOLAS research collaborations, and SOLAS workshops and Open Science Meetings, but did so without a modern US SOLAS plan.

### 2.2 Ocean-Atmosphere Interaction Committee

The Ocean-Atmosphere Interaction Committee (OAIC) was formed in 2018 as a subcommittee of the Ocean Carbon and Biogeochemistry (OCB) SSC to strengthen communication and collaboration between ocean and atmospheric scientists in the US. The committee provides support for scientists working at the air-sea interface to overcome the disciplinary silos that result from separate atmospheric or oceanic science community platforms. The OAIC works to reinvigorate a network of US air-sea researchers in collaboration with the international SOLAS program. Specific roles of OAIC include:

- Representing the interests of the air-sea research community to the broader scientific community, as well as educators, decision-makers, and the general public
- Organizing topical air-sea workshops or organizing sessions at large national and international conferences
- Mentoring early-career scientists who wish to conduct air-sea research
- Facilitating early career participation in meetings and training activities focused on air-sea interactions
- Communicating opportunities and scientific findings to grow and strengthen a network of air-sea researchers
- Exploring potential sources and mechanisms for funding air-sea research

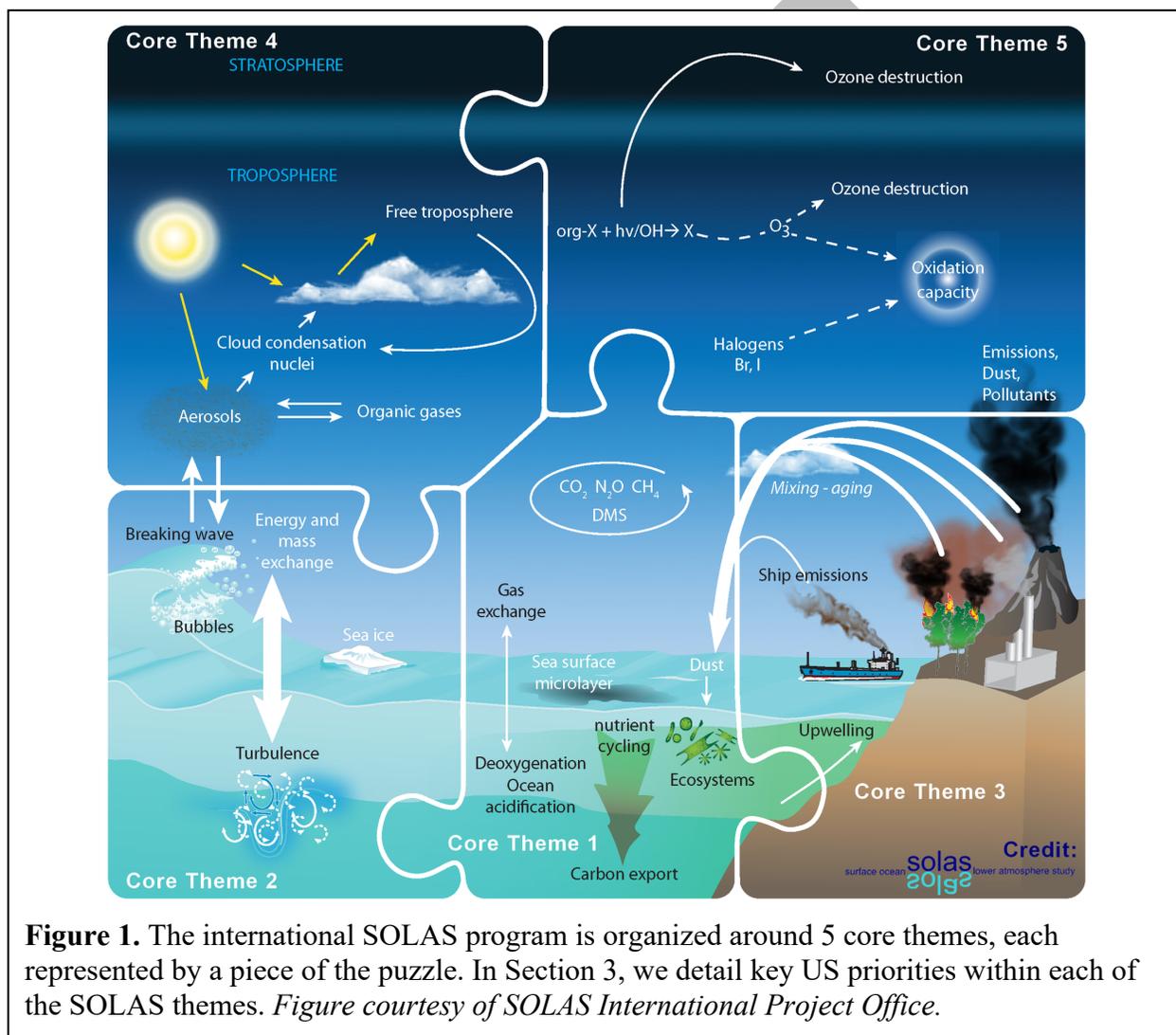
### 2.3 Workshop rationale and summary

The interface between the ocean and the atmosphere covers over 70% of the planet's surface area, and an improved understanding of the processes operating at this interface is key to our ability to predict future climate scenarios. As Earth System Models (ESMs) evolve, new understanding of air-sea interactions should be incorporated, including novel knowledge on high and extreme wind events, gas and aerosol exchange, heat exchange, biogeochemical and ecological feedbacks, and the multi-faceted impacts of large-scale climate indices such as the El Niño Southern Oscillation (ENSO). These phenomena impact the lower atmosphere and surface ocean across regional and global scales, but we have only begun to understand their intricacies. To date, there has been limited systematic coordination within the US air-sea research community, but there is a need to network, collaborate, and coordinate air-sea research activities.

With leadership from the [OAIC](#), OCB convened a [community workshop, \*Ocean-Atmosphere Interactions: Scoping directions for new research\*](#), on October 1-3, 2019 in Sterling, VA. The goal of this workshop was to bring together members of the US air-sea research community to facilitate new collaborations, identify knowledge gaps and high-priority science questions to motivate innovative research and contribute to international SOLAS efforts, and to formulate a US SOLAS science plan. The workshop included 59 members of the air-sea research community and representatives from NSF, NASA, and the US Carbon Cycle Science Program. Based on presentations and discussions at the workshop ([workshop website](#)), the OAIC and workshop participants developed a common set of science goals and research priorities outlined in this report that will serve to strengthen the US air-sea research community and foster a more cohesive US contribution to international SOLAS.

### 3. US Contributions to International SOLAS

The key air-sea interaction research priorities, collaborations, and critical issues identified by the US-SOLAS community at the workshop were quite detailed, diverse, and wide-ranging. We organized this report to articulate the strengths of potential US SOLAS questions and initiatives and potential synergy with the international SOLAS program. To this end, we first framed key US-SOLAS questions and activities within the themes identified in the SOLAS Decadal Science Plan (2015-2025) (Brévière et al., 2016, Fig. 1) (Section 3), and then we identified critical cross-cutting research areas (Section 4).



#### 3.1 Theme 1. Greenhouse gases and the oceans

Greenhouse gases (GHGs) such as carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O) are key regulators of Earth's temperature and climate. Atmospheric GHG concentrations are mainly regulated by physical and biogeochemical processes in the upper ocean. Steady progress has been made to improve quantitative constraints and process-level understanding of global N<sub>2</sub>O (Tian et al., 2020), CH<sub>4</sub> (Sauniois et al., 2020), and carbon (Friedlingstein et al., 2019) budgets, oceanic uptake of anthropogenic carbon

(Gruber et al., 2019), land-ocean carbon fluxes (Resplandy et al., 2018), and temporal variability in the ocean carbon sink (McKinley et al., 2020; Lovenduski et al., 2019; McKinley et al., 2017). Data products and databases such as the Surface Ocean CO<sub>2</sub> Atlas (SOCAT) and CH<sub>4</sub>/N<sub>2</sub>O database, [MEMENTO](#), are valuable resources for carbon cycle studies and biogeochemical model validation. Advances in sensor development and autonomous aircraft- and satellite-based observing technology will expand coverage and yield new insights on oceanic drivers of GHG cycling and their feedbacks on climate. The high-level US priorities that emerged related to oceanic GHG cycling and associated atmosphere-ocean dynamics are:

### **What are the sources of natural variability in air-sea gas exchange?**

Large-scale climate indices (e.g., ENSO, PDO, SAM) and other earth system processes interact to cause natural variations in air-sea gas exchange. Key components of these interaction are poorly constrained and thus limit predictive capacity, including:

- Implications of ocean circulation shifts
- Volcanic eruptions - deposition effects (fertilization) on ocean productivity and carbon uptake
- Local to regional biophysical interactions (e.g., submesoscale to mesoscale eddies, fronts)
- Carbon-climate feedbacks associated with physical and biological responses to anthropogenic warming, ocean acidification and deoxygenation, and increased nutrient loading from atmospheric and terrestrial inputs

### **What are the uncertainties and temporal variability in regional air-sea fluxes?**

Regional fluxes vary based on dynamics and response times, and observations to discern these differences are lacking. In particular, it is important to improve process understanding and observational coverage of air-sea fluxes in high  $\Delta p\text{CO}_2$  regions (e.g., coastal upwelling regions, tropical and high latitude oceans, western boundary currents, etc.). Thus we need to:

- Evaluate potential contributions of air-sea flux measurement uncertainties (observational gaps, seasonal biases, biases in air-sea gas exchange parameterizations, etc.) to model-data discrepancies in regional interannual and decadal variability in ocean carbon uptake and outgassing
- Explore impacts of ice (glaciers, sea ice) on high-latitude air-sea and land-ocean carbon fluxes
- Compare observation-based surface  $p\text{CO}_2$  products and associated methods (e.g., interpolation) and identify potential sources of discrepancy (e.g., skin temperature corrections, treatment/definition of surface  $p\text{CO}_2$ ) - e.g., [OCB Working Group Filling the gaps in observation-based estimates of air-sea carbon fluxes](#)
- Cross-calibrate regional measurements across platforms (e.g., ships, floats, gliders, moorings)

### **How can we improve estimates of riverine and coastal carbon fluxes for increased understanding of GHG exchange?**

While the coastal ocean makes up only a small percentage of the global ocean area, these dynamic systems cycle a disproportionate amount of carbon. With their high spatiotemporal variability, coastal and estuarine carbon fluxes are especially uncertain in the face of climate and land use changes. Closing the ocean carbon budget requires improvements in our capacity to measure and model coastal riverine, lateral, and air-sea fluxes. Specific recommendations include:

- Expand the capacity to measure extreme events by deploying autonomous observing assets to quantify episodic land-ocean fluxes and carbon delivery to coastal waters
- Apply novel observing tools and methods (e.g., remote sensing technology, isotope and tracer methods) to quantify and scale up lateral fluxes from tidal wetlands to coastal waters
- Use [NOAA's Carbon Tracker](#) as a potential roadmap for integrating diverse measurements with models

- Deploy novel platforms (e.g., animals in the Southern Ocean, gliders on shelves, Saildrones in Arctic, etc.) equipped with sensors to measure ocean carbon system parameters

### **What are the sea-to-air fluxes of other greenhouse gases of interest?**

N<sub>2</sub>O and CH<sub>4</sub> are potent GHGs with large natural sources, sinks, and reservoirs, and are thus deemed a high priority for future observation and prediction, particularly given the dearth of oceanic CH<sub>4</sub> (~20,000 measurements in MEMENTO) and N<sub>2</sub>O (~120,000 measurements in MEMENTO) measurements relative to CO<sub>2</sub> (>28 million pCO<sub>2</sub> measurements in SOCAT v2020). The [SCOR Working Group 143 “Dissolved N<sub>2</sub>O and CH<sub>4</sub> measurements: Working towards a global network of ocean time series measurements of N<sub>2</sub>O and CH<sub>4</sub>”](#) worked to establish a common framework for measurement, standardization, and data reporting to improve quality and inter-comparability of these oceanic GHG measurements (Wilson et al., 2018). Although considerable progress has been made to standardize measurements and reporting, the natural cycles of CH<sub>4</sub> and N<sub>2</sub>O and the driving physical and biogeochemical processes underling these cycles remain poorly characterized. Oceanic emissions of N<sub>2</sub>O and CH<sub>4</sub> occur naturally via a range of microbial processes, and production and consumption of these GHGs in the ocean are highly sensitive to climate and environmental changes. A recent [OCB Scoping Workshop Oceanic Methane and Nitrous Oxide: The present situation and future scenarios](#) convened researchers to plan and prioritize future CH<sub>4</sub> and N<sub>2</sub>O measurements in the global ocean and assess current understanding of the production and consumption mechanisms, emissions, and spatiotemporal distributions (Wilson et al., 2020), specifically targeting the following priorities:

- Determine gas hydrate destabilization and fate of released CH<sub>4</sub>
- Study microbial dynamics of denitrification- and nitrification-driven N<sub>2</sub>O production and links to oxygen minimum zone evolution
- Evaluate the impact of physical processes such as upwelling and diapycnal mixing on GHG advection to the surface ocean and flux to the atmosphere
- Determine the impacts of ocean acidification, deoxygenation, and warming on GHG production rates and associated climate and environmental feedbacks on interannual (e.g., ENSO) and multidecadal timescales
- Examine N<sub>2</sub>O-CO<sub>2</sub> feedbacks associated with deposition-driven bloom initiation and subsequent biological drawdown of CO<sub>2</sub> and N<sub>2</sub>O

## **3.2 Theme 2. Air-sea interface and fluxes of mass and energy**

Fluxes across the air-sea interface are an essential connection between the ocean and atmosphere. These fluxes are difficult to measure directly, and thus are most often parameterized in terms of more readily available measurements such as wind speed. Important progress has been made in quantifying air-sea fluxes over the last decade but there is still considerable uncertainty on how to best calculate air-sea exchange in “non-standard” environments, such as coastal regimes, ice covered regions, high wind-speed environments, and estuaries. Additionally, questions remain over which fundamental processes and variables should be included in air-sea gas transfer velocity parameterizations. Furthermore, while a wide variety of methods have been successfully applied to investigate air-sea exchange, it is essential to continue to evaluate these methods as they often disagree, especially in marginal environments. Air-sea fluxes of gases, energy and momentum are discussed in this section. Air-sea fluxes of primary aerosols are discussed in Section 3.4. The following questions emerged as high US priorities to advance progress on SOLAS Theme 2.

### **In what environments does air-sea exchange have the greatest uncertainty?**

Much progress has been made in constraining key drivers and developing robust parameterizations of air-sea gas exchange in the open ocean. However, large uncertainties remain for undersampled systems (e.g.,

high-wind environments, coastal regions, ice-covered regions), in part because of a lack of understanding of the key processes that drive air-sea exchange. Air-sea momentum, mass and heat exchange in extreme wind conditions are very complex and not well understood. There is increasing evidence that the air-sea interface changes fundamentally at or approaching hurricane-force winds, in which whitecaps, spray, and spume dominate the wave boundary layer, acting to limit (Donelan et al., 2004; Takagaki et al., 2012; Potter et al., 2015; Curcic and Haus, 2020) and perhaps even reduce the aerodynamic drag (Powell et al., 2003; Jarosz et al., 2007; Sanford et al., 2011). At high wind speeds ( $>25 \text{ m s}^{-1}$ ), bubble processes associated with breaking waves are important drivers of air-sea gas exchange, but are not well constrained or parameterized due to logistical challenges (D'Asaro and McNeil, 2007). Studies to date have yielded conflicting results on the impact of high wind speeds on the form of the wind speed-gas transfer relationship, particularly whether or not gas transfer eventually plateaus at high wind speeds. There are also large uncertainties in air-sea exchange in partially ice-covered environments, since ice dampens air-sea gas exchange, whereas ice leads may experience increased turbulence relative to the open ocean. Traditionally, gas exchange in ice-covered environments has been treated similarly to open ocean settings and simplistically scaled by the fraction of the ice cover, which fails to capture key underlying physical, chemical, and biological processes (Crabeck et al., 2014; Else et al., 2013; Fransson et al., 2017; Gourdal et al., 2020). Coastal and estuarine systems are responsible for a large drawdown of  $\text{CO}_2$ , and yet gas transfer in these systems is still highly uncertain due to limited fetch, coastal currents, shallow waters, and increased surfactants. Research priorities include:

- Identify and characterize physical and biogeochemical drivers to better quantify and predict air-sea exchange in ice-covered systems
- Explicitly measure gas exchange in partially ice-covered systems using a variety of methods that integrate over different spatial and temporal scales
- Improve existing estuarine-specific parameterizations and determine whether there are better correlates to use for estuarine gas exchange than wind speed, water depth and current velocity
- Conduct more detailed investigations of air-sea exchange of gases, energy, and heat at very high-wind speeds, using autonomous in situ sensors (e.g., during hurricanes), in order to accurately model air-sea exchange fluxes at high wind speeds

### **What are the fundamental processes and variables needed in air-sea exchange models?**

Air-sea exchange fundamentally depends on turbulence, but since turbulence is typically not measured directly in field studies, air-sea exchange parameterizations are often based solely on wind speed/stress. For example, many existing gas transfer parameterizations (Wanninkhof, 2014) do not explicitly include bubbles, yet bubbles produced from breaking waves at the sea surface represent an important transfer mechanism from the atmosphere to the surface ocean for relatively insoluble gases such as oxygen. It is unclear how well the few parameterizations that include bubbles (Liang et al., 2013; Deike and Melville, 2018) work in different environments. Whitecap fractional coverage is typically parameterized in terms of wind speed, has orders of magnitude variability (Anguelova and Webber, 2006) and the parameterizations are uncertain. Furthermore, organic surfactants on the sea surface (see Sections 4.1 and 4.2) can affect gas diffusion across the air-sea interface and can strongly dampen short-wavelength (cm-scale) waves on the ocean surface. In laboratory and field experiments, surfactants have been shown to suppress air-sea gas transfer by as much as 30% (Frew et al., 1990, Salter et al., 2011). The following activities are required to improve estimates of air-sea gas exchange:

- Quantify relationships between turbulence and key variables such as water temperature, air temperature, humidity, and wave statistics to inform the development of more robust air-sea exchange parameterizations for use in multiple environments
- Expand gas exchange parameterizations to explicitly include gas transfer through bubbles, sea spray and other phenomena relevant at high wind speeds (e.g., wind history, rain, etc.)

- Conduct field-based bubble studies to investigate the role of bubbles at high wind speeds and determine bubble penetration depth and size distributions in a range of oceanographic settings, especially during high-wind events such as hurricanes
- Improve understanding of whitecaps, the surface expression of wave breaking, and a myriad of boundary layer processes by conducting holistic field-based studies in a range of environments
- Conduct field studies to quantify how the composition, concentration, and origin of surfactants determine their influence on air-sea exchange

### **What new methods can be used to improve our understanding?**

Traditional gas exchange experiments have been conducted in the field using several techniques, including eddy covariance of CO<sub>2</sub> or DMS, tracer release experiments using <sup>3</sup>He and SF<sub>6</sub>, and mass balance approaches using noble gases. However, results from these techniques do not always agree, and the origins of these discrepancies are not fully understood. Models are also useful tools for quantifying air-sea gas exchange and can be used to derive wave statistics that can be used to calculate wave property-based estimates of gas exchange. Inclusion of wave properties in gas transfer parameterizations may allow better estimation of bubble effects on gas exchange. What new approaches can be used to measure gas exchange and its key drivers?

- Expand the eddy covariance technique to other gases, and further evaluate the uncertainties and limitations of this technique
- Conduct laboratory studies to target specific mechanisms of gas exchange, particularly those relevant to under-studied environments, and conduct a comparative analysis to assess the transferability of these findings to the field
- Increase use of remote sensing methods to provide much needed global coverage and information on properties relevant to air-sea exchange such as wave age, surface roughness, and whitecap fraction (Anguelova and Bettenhausen, 2019)
- Use data assimilation of surface properties to improve air-sea exchange parameterizations. Continue to use and adapt ocean inversion methods to derive air-sea exchange

### **3.3 Theme 3. Atmospheric deposition and ocean biogeochemistry**

Atmospheric deposition delivers nutrients (e.g., iron) to the remote ocean (Duce et al., 1991; Jickells et al., 2005) that are known to limit marine primary production and affect the cycling of other nutrients such as nitrogen (N) and phosphorus (P) (Falkowski et al., 1998; Kim et al., 2011; Moore et al., 2013). Thus, atmospheric deposition has large-scale impacts on the ecology and biogeochemical cycles in the upper ocean. Steady progress has been made to advance the process-level understanding of atmospheric nutrient deposition to the oceans and improve their representation in global biogeochemical models. However, the temporal and spatial coverage of atmospheric deposition sampling is limited, and there remains substantial uncertainty in the parameterization of deposition processes in models. To overcome these challenges and gain a better understanding of atmosphere-ocean biogeochemical interactions, the following fundamental questions emerged as high US priorities to advance progress on SOLAS Theme 3.

#### **How can we improve the estimates of atmospheric nutrient deposition in models?**

Most atmospheric models still treat deposition as a seasonal cycle; mechanistic understanding and complexity of atmospheric deposition should be better represented in models with inclusion of important factors such as speciation, solubility, residence time, and boundary conditions. Priorities needed to improve models include:

- Advocate for collaboration between modelers and observational scientists to discuss critical questions, data issues, and important parameters to measure
- Identify fundamental processes and variables needed to improve atmospheric deposition models
- Explore higher-frequency and mesoscale variability through process studies, with the goal of improving mechanistic understanding and model parameterizations on a regional scale

- Move from data to mechanistic models to parameterizations that can be used in global models.
- Merge in situ observations (e.g., ships, buoys, gliders), satellite data, and numerical models to develop global estimates of deposition fluxes and their impacts on upper ocean biogeochemical processes to better understand climate feedbacks and to project long-term trends

### **In what environments does atmospheric deposition have the greatest uncertainty? What are long-term depositional fluxes in regions other than the North Atlantic?**

Long-term atmospheric observational platforms are largely restricted to the North Atlantic. By contrast there is a dearth of depositional data in the Southern Ocean, a region that is under-sampled in terms of atmospheric deposition due to its remoteness and low aerosol concentrations (Winton et al., 2016; Gao et al., 2020) and the Arctic Ocean that is impacted by altered regional dust sources and atmospheric composition due to the warming (Crusius et al., 2011; Gao et al., 2019). Key research priorities to address these uncertainties in polar waters include:

- Establish long-term station-based time series measurements
- Leverage monitoring networks with infrastructure that is already in place to support atmospheric deposition measurements (e.g., physical oceanography efforts, LTER sites)
- Use ship-based seasonal expeditions, including sampling on non-dedicated scientific cruises
- Develop new instruments to deploy on autonomous platforms (e.g., saildrones, buoys) to extend spatial and temporal resolution
- Integrate satellite observations with *in situ* measurements

### **How can estimates of atmospheric deposition fluxes be better constrained?**

Constraining depositional fluxes of aerosols is one of the major uncertainties in our understanding of biogeochemical cycling of nutrients in the oceans. In particular, atmospheric wet deposition of nutrients to the ocean is poorly understood (Gao et al., 2003). Most studies have focused on concentration measurements in both aerosols and precipitation rather than fluxes, which has introduced substantial uncertainties to deposition estimates. Standardized methodologies have not been developed to quantify atmospheric deposition and to measure properties of key bioactive elements. These issues have contributed to uncertainties in reproducing observed spatial and temporal patterns of nutrient concentrations in the ocean. The following priorities will address these uncertainties:

- Standardize methodologies
- Use tracers such as beryllium-7 (Kadko et al., 2015), thorium-230/232 (Hayes et al., 2017), and lead-210 (Boyle et al., 2014) to improve wet and dry deposition flux estimates
- Identify uncertainty areas and biases associated with physical parameter measurements
- Conduct measurements to better characterize atmospheric deposition associated with episodic events such as hurricanes, volcanoes and biomass burning
- Compare atmospheric deposition in different forms (e.g., dust particles, rain, snow) and across different depositional environments (e.g., ice, ocean surface)

### **How does ocean biogeochemistry influence the conversion of atmospheric bioaccessible nutrients to oceanic bioavailable forms?**

Atmospherically delivered nutrients cannot necessarily be characterized as bioavailable because they are in aerosols and precipitation and may not be freely available to cross an organism's cell membrane. Nutrient-laden aerosols instead should be viewed as a bioaccessible, which is potentially accessible from the environment (Meskhidze et al., 2019). The bioaccessibility of atmospherically-delivered nutrients in seawater will vary among different bacterial and phytoplankton taxa and for species within taxa, due to differences in the cellular surface area and presence of multiple uptake mechanisms for the transport of

chemicals across the cell membrane. To better understand bioaccessibility and its impact on the ecology of the upper oceans several priorities are highlighted:

- Develop a mechanistic understanding of bioaccessibility of atmospherically-derived nutrients in marine plankton
- Improve ocean ligand modeling and ligand-nutrient (e.g., Fe) complex formation and its associated impact on bioavailability
- Examine atmospheric and oceanic ligands and their roles in nutrient complexation in the ocean
- Study the kinetics of aerosol-derived bioaccessible nutrient transformations into dissolved/colloidal/particulate phases in seawater to elucidate the role of in-situ speciation on the aerosol nutrient biogeochemistry
- Investigate whether global warming and other anthropogenic stressors synergistically alter the uptake of atmospheric nutrients and metals by marine biota

### **What is needed to establish Standard Operating Procedures (SOPs) and increase data comparability?**

Currently, different procedures have been applied to atmospheric deposition measurements, and many parameters have been operationally defined for different purposes among investigators. The standardization of aerosol nutrient measurement methodologies for atmospheric and oceanic disciplines is critical for improved representation of nutrient biogeochemistry. Critical needs include:

- Establish a formalized process for developing and using standard reference materials
- Standardize SOPs for field collection and laboratory analyses and archive them in the international Ocean Best Practices repository
- Establish a core set of parameters for field collection, including source, process, and flux tracers
- Coordinate international intercalibration studies with emphasis on using accessible consensus reference materials and standardized sampling protocols

## **3.4 Theme 4. Interconnections between aerosols, clouds, and marine ecosystems**

Clouds in the marine boundary layer (MBL) play an important but poorly constrained role in shaping Earth's climate (Carslaw, 2013). Their formation is strongly tied to the aerosol composition in the MBL, which is comprised of a mixture of marine-derived primary and secondary aerosols, non-marine derived secondary aerosols formed in the remote MBL, and continentally-derived aerosols. Ocean-derived and continental sources contribute both cloud condensation nuclei (CCN) and ice nuclei (IN), with CCN and IN number concentration, size, composition, and morphology all influencing the formation, radiative properties, and lifetime of clouds in the MBL. Primary marine aerosols (PMA) are often enriched in marine-derived organic matter including refractory dissolved organic carbon (RDOC), and living and recently produced marine organic material (see Section 4.1). Secondary ocean-derived aerosol includes sulfates generated by the oxidation of dimethyl sulfide (DMS) and secondary organic aerosol (SOA) derived from the oxidation of volatile organic compounds (VOCs) emitted from the ocean (Ayers and Gillett, 2000; Bates et al., 1992; Claeys et al., 2010). Ocean- and continentally-derived trace gases can either change the chemical composition and hygroscopic properties of existing particles or lead to new particle formation (NPF) (Charlson et al., 1987; Zheng et al., 2021). The emission of RDOC and biogenic compounds (both primary and secondary) into the atmosphere depends strongly on physical conditions including wind speed and seawater temperature (Salter et al., 2014; Zabori et al., 2012) and the biological community structure will affect the quantity and composition of primary marine organic aerosol and gaseous precursors required for secondary aerosol formation (Ault et al., 2013; Quinn et al., 2015; Wang et al., 2015). Current uncertainties in Earth system model climate change simulations are largely due to

our poor understanding of processes connecting aerosols, clouds, and marine ecosystems; achieving high confidence in model simulations critically depends on implementation of more realistic ocean ecosystem-aerosol-cloud interactions. The following fundamental questions remain as high priorities on SOLAS Theme 4.

### **What physical processes (e.g., sea-surface microlayer, bubbles) determine ocean-derived aerosols of different sizes and mixing-state that impact clouds?**

Sea salt dominates the SSA mass concentration over remote oceans (Seinfeld and Pandis, 2006).

However, studies have shown that sub-micrometer SSA can hold a significant organic mass fraction (O'Dowd et al., 2004; Keene et al., 2007; Gantt et al., 2011; de Leeuw et al., 2011) that could affect its light scattering, CCN, and IN properties. Most marine aerosol parameterizations implemented in current climate models were developed based on laboratory measurements of SSA production (Mårtensson et al., 2003; Gong, 2003; Sellegri et al., 2006; Long et al., 2011). These parameterizations predict wide ranges in SSA fluxes (Lewis & Schwartz, 2004; Grythe et al., 2014). Although recent parameterizations for SSA production are using shipborne measurements (Saliba et al., 2019), more studies need be conducted using particle flux measuring instrumentation that can distinguish ocean-derived primary particles from the long-range transportation of terrestrial aerosols and secondary aerosols formed through the successive oxidation of gas-phase precursors. To expand field measurements of marine aerosols and clouds, long-term monitoring sites at fixed field stations such as the Ragged Point facility in Barbados, Tudor Hill, Bermuda, Amsterdam Island, and Cape Grim can also be leveraged to increase the spatial and temporal coverage (see Section 5.1). Long-term monitoring sites such as the Bermuda Atlantic Time-series Study (BATS) and the Hawai'i Ocean Time-series (HOT) would be particularly beneficial, as they would provide rich data resources for ocean-atmosphere process studies. The following activities are needed:

- Analyze existing field measurements with marine aerosol, meteorological, and ocean biological data (e.g., PODEX, SEACRS, SABOR, NAAMES, AWARE, SOCRATES, AToM, LASIC, ENA) to explore detailed mechanisms that control the production and chemical composition of ocean-derived aerosols
- Conduct field campaigns in oceanic regions that are least represented in current parameterizations of ocean-derived aerosols and clouds (e.g., polar regions) to determine how processes specific to those regions are impacting aerosol characteristics
- Set up long-term measurement sites for marine aerosols and their gaseous precursors to explore how atmospheric properties (e.g., surface wind speed ( $U_{10}$ ), atmospheric stability), ocean physicochemical properties (e.g., SST, pH, salinity, whitecap fraction, DOC and POC, surface film coverage), biological indicators (e.g., organism type and abundance, Chlorophyll a concentration), and sea ice extent modify CCN and IN numbers over the ocean

### **What remote sensing observables can be used to link marine biological activity to aerosols and clouds?**

Satellites are the dominant means to characterize marine aerosols and aerosol-cloud-climate interactions in a changing climate because they provide global, long-term information on the spatiotemporal variability of many properties affecting marine aerosol production and removal. There are several past, existing, and planned remote sensing instruments supported through the U.S. and international programs that will improve characterization of marine aerosols and differentiate ocean-derived aerosols from long-range transport of continental emissions. Remotely sensed instruments combined with ground-based systems including ship-borne data acquisition initiatives are needed to establish links between aerosols, clouds, and marine ecosystems. In particular, it is important to:

- Achieve coincident (in time and space) retrievals of cloud physical properties, vertically-resolved aerosol physical and chemical properties, and ocean surface and sub-surface biogeochemical data
- Provide data at a signal-to-noise ratio that is high enough for retrieval of ocean ecosystem processes and aerosol speciation and loadings over the oceans

- Provide better coverage in the Arctic and Southern Ocean regions characterized by high cloudiness and low solar zenith angles
- Apply new and emerging observing technologies (i.e., UV-VIS-NIR-SWIR ocean color sensor, High Special Resolution Lidar, polarimeter, and "CubeSats", a shoebox sized satellite) for quantifying key processes at the atmosphere-ocean interface

### 3.5 Theme 5. Ocean biogeochemical controls on atmospheric chemistry

The oceans produce a plethora of volatile organic and inorganic compounds (VOCs, VICs) and organic surfactants. These compounds enter the atmosphere as gases or as part of primary marine aerosols, wherein they affect atmospheric chemistry in multiple ways, as detailed in the crosscutting section of this report (Section 4). VOCs, VICs and surfactants are produced through a myriad of biological, photochemical, and thermal processes in the sunlit oceans, but many details of the controlling processes are poorly constrained. Similar uncertainties persist regarding the impacts of ocean-derived organic matter on gas phase and aerosol chemistry in the atmosphere. Process studies, measurements, and modeling efforts focused on present-day and future scenarios are needed to assess short- and long-term impacts of these gases and surfactants on Earth's radiative balance, climate, and ocean-atmosphere feedbacks.

#### **What process studies and measurements will provide critical advancements to move the field forward?**

Directed field campaigns and laboratory studies are needed to address important data gaps, using emerging technologies in biochemistry, analytical chemistry, ocean engineering (e.g., gliders, buoys), remote sensing and imaging science (e.g., Imaging FlowCytobot). High priorities include:

- Examine rapidly changing polar oceans and atmosphere, ice-impacted waters, and the seasonal/diurnal variability in VOC and VIC cycling in all oceans
- Assess impacts of physical (e.g., UV-induced photoinhibition, shear stress) and ecological interactions (e.g., algal and microbial viral infection) on volatile gas and surfactant production and removal
- Investigate sea-surface microlayer photochemical/thermal reactions (e.g., reaction with atmospheric-derived ozone) and their impacts on air-sea fluxes and microlayer ecology
- Examine the impacts of episodic events such as hurricanes, harmful algal blooms (e.g., *Phaeocystis antarctica* in the Ross Sea) and coastal deoxygenation on atmospheric inputs of volatile gases or organic matter associated with primary marine aerosols
- Determine the underlying physiological functions and biochemical processes associated with volatile gas and surfactant cycling in the oceans

#### **How will climate change impact relevant biogeochemical processes in the long term?**

Climate change has resulted in increased atmospheric turbulence and cloudiness, ocean acidification, changes in seasonal ice patterns and extent of springtime sea-ice melt in polar regions. Climate fluctuations also impact concentrations of ozone and other reactive oxidants (e.g., OH radical) in the atmosphere. Climate-driven priority study areas concerning volatile gas and surfactant production, removal and fluxes include:

- Determine the effects of warming on the physics, photochemistry and ecology of the upper ocean in coastal and open-ocean settings
- Examine the impact of seasonal melt-induced "freshening" of the near-surface ocean on upper-ocean chemistry and ecology
- Investigate effects of temperature changes in polar regions on the nature of the seasonal ice, ice optical properties, ice porosity, ice photochemistry and ecology

- Evaluate changes in upper ocean turbulence and degree of cloudiness and their effect on sea-surface microlayer dynamics
- Ascertain impacts of warming-induced destabilization of gas hydrates and the release of volatile gases
- Establish whether ozone and other atmospheric oxidants react with sea-surface microlayer constituents and affect microlayer ecology, PMA formation and organic matter enrichment, and the formation of VOCs and VICs

### **What are important ocean-atmosphere feedbacks?**

Chemical processing of ocean-derived organic and inorganic gases and organic surfactants in the atmosphere and subsequent wet and dry deposition into the ocean may reinforce several feedbacks that warrant detailed investigation. It thus is imperative to:

- Determine if marine-derived organic matter reactions in the atmosphere produce compounds (e.g., ammonia) that re-deposit to the ocean and affect the ecology and associated production of gases and surfactants
- Ascertain the impact of photochemical processing of marine organic matter in PMA on volatile inorganic and organic gas production in the troposphere

### **Is there interannual or decadal variability in atmospheric chemistry due to changes in ocean biogeochemistry?**

This question is a key uncertainty in assessing feedbacks and ocean-atmospheric interactions and future climate assessments. To address it, it is necessary to:

- Use sustained data sets, such as those from Long-Term Ecological Research (LTER) sites, to ascertain the impact of photochemical processing of marine organic matter in PMA
- Conduct long term time series of a simpler set of relevant measurements at key sites to extrapolate from short field intensives that involve large ensembles of sophisticated measurements

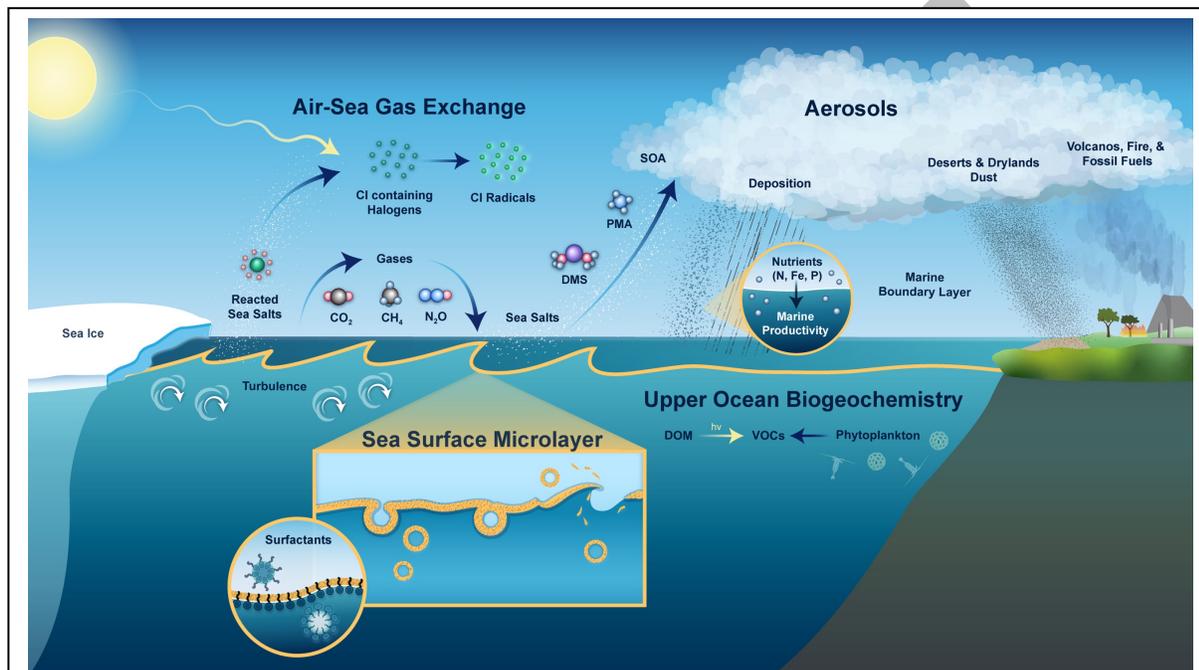
### **Are there potential experimental and methodological issues and limitations to consider?**

Any laboratory or field study to evaluate biological or chemical processes has associated uncertainties and limitations that are important to consider. Priorities include:

- Determine best methods to study microlayer reactions involving VOCs and VICs as products or reactants
- Compare VOC and VIC production in axenic algal cultures grown for many generations with “wild” populations
- Assess influence of bubbling on production rates of volatile gases or surfactants in algal cultures
- Determine if the reduced mixing dynamics of mesocosms affect biogeochemical processes and rates
- Develop data-handling platforms to process data streams from high-frequency data acquisition systems (e.g., Imaging FlowCytobot) used to study volatile gas cycling

## 4. Crosscutting Topics and Research Questions

High-priority topics that overlap with several of the five SOLAS themes were identified as cross cutting topics that warrant further discussion to highlight uncertainties and avenues for future research. These include the sea-surface microlayer, surfactants, aerosols, air-sea exchange, and atmospheric chemistry (Fig. 2). An overarching goal of investigating these crosscutting topics will be to determine (i) the extent to which increased understanding of these topics will improve oceanic and atmospheric chemistry models and (ii) the predicted changes in these systems in response to a changing climate.



**Figure 2.** Cross cutting themes, identified by US researchers as high priority topics for further research, include the sea surface microlayer (section 4.1), surfactants (section 4.2), air-sea gas exchange (section 4.3), halogens, reactive nitrogen and redox chemistry (section 4.4), and air-sea exchange and evolution of aerosols (section 4.5). This schematic shows how those topics are all interrelated processes occurring at the air-sea interface, upper ocean and lower atmosphere.

### 4.1 Sea-surface microlayer

The sea-surface microlayer is an important interface controlling the transfer of energy, material and gases between the oceans and the atmosphere, and as such has been a primary focus of the SOLAS program since its inception. The five SOLAS themes all include processes that affect and are affected by the sea-surface microlayer, including marine ecology (Themes 4 and 5), photochemistry, and free radical/thermal chemistry (Theme 2), greenhouse gases and gas exchange (Themes 1 and 2), primary aerosol production (Themes 2, 4 and 5), and atmospheric deposition (Theme 3). Our understanding of the sea-surface microlayer has evolved, uncovering numerous physical, biological, and chemical properties that are distinctly different from the underlying “bulk” seawater and overlying atmosphere. Several reviews highlight current knowledge and research gaps (Cunliffe et al., 2013; Soloviev and Lukas, 2014; Engel et al., 2017; Wurl et al., 2017; Galgani and Loiselle, 2019).

Organic matter in the sea-surface microlayer contains old carbon referred to as refractory organic carbon, classic surfactants including fatty acids and lipids, and compound classes with surfactant properties such as carbohydrates and combined amino acids (peptides and proteins) along with a myriad

of minor dissolved components and particulate matter that includes viruses and bacteria (Quinn et al., 2015; Facchini et al., 2008; O’Dowd et al., 2004; Prather et al., 2013; Russell et al., 2010; Kieber et al., 2016; Beaupre et al., 2019). The microlayer is formed and maintained by rising bubble plumes generated by breaking waves that deposit adsorbed material to the sea surface. Breaking waves also disrupt and destroy the microlayer mixing surfactants into the underlying seawater. Through dynamic interactions of breaking waves and rising bubble plumes, the sea-surface microlayer persists even under windy conditions and is therefore not just a “calm weather” phenomenon. However, the various properties and impacts of the microlayer that have been observed under calm conditions are largely unknown under windy conditions, primarily due to the lack of sampling and observations.

Although there has been significant progress in our understanding of the sea-surface microlayer, especially with respect to its composition, substantial uncertainties persist, as discussed below and in the following crosscutting sections on surfactants, air-sea gas exchange, and aerosols.

#### 4.1.1 Role of bubbles in the formation and decay of the sea-surface microlayer

As ocean wave-generated plumes of bubbles move downward and then upward to the sea surface, they become coated with surfactants (Section 4.2). Subsequent disintegration of bubble plumes at the sea surface injects material into the atmosphere as components of PMA (Section 4.5); most of the material adsorbed onto the bursting bubbles is deposited to the sea surface replenishing the sea-surface microlayer with living and nonliving dissolved and particulate inorganic and organic matter. As the main source of material to the microlayer, bubbles play a central role in microlayer dynamics. Despite their importance, we know very little about bubble plume dynamics or the effect of surfactants on these dynamics, which is critical to understanding air-sea exchange processes and PMA formation as discussed in Themes 1, 2, 4 and 5. Thus, it is high priority to:

- Evaluate interdependencies between wind speed and duration, sea surface roughness, bubble plume depth and scavenging properties, and sea-surface microlayer composition
- Study the physics of bursting bubbles in multilayer, interacting bubble rafts

#### 4.1.2 Particulate matter

Examination of microlayer constituents has primarily focused on dissolved components. However, living and nonliving particulate matter (POM) are also present, including anthropogenic material (e.g., plastics, Galgani and Loiselle, 2019) and in some cases concentrated, in the microlayer. Additionally, POM is likely produced from DOM during the bubble bursting process (e.g., gels), but virtually nothing is known regarding the role of particulate matter cycling and impacts in the sea-surface microlayer. Thus it is imperative to:

- Conduct studies to determine the photochemical formation of VOCs from microlayer POM photolysis
- Evaluate DOM to POM production fluxes from bursting bubble rafts
- Study biological processes and impacts on the chemical and biological composition of the sea-surface microlayer to determine, for example, their role in surfactant production and VOC formation
- Determine the surface-active properties of living and nonliving particles

#### 4.1.3 Thermal and photochemical reactions

Dissolved and particulate organic and inorganic matter concentrated in the sea-surface microlayer may react with other constituents in the microlayer or with constituents deposited to it from the MBL, which in turn may affect gas exchange (Thompson and Zafiriou 1983), PMA composition, and the processing of evolved marine and continentally-derived aerosols deposited to the sea surface. These reactions may be driven by sunlight (Chiu et al. 2017) or dependent on thermal reactions (e.g., reactions initiated by atmospherically-derived gases including ozone or NO<sub>x</sub>). Work has been done to understand hydrolysis

reactions involving reactive gases such as sulfur dioxide, carbon dioxide, and carbonyl compounds. However, overall, other than seminal work by Thompson and Zafiriou (1983) and more recent work by Chiu et al. (2017), there is very little known about the impacts of microlayer reactions on SOLAS-centered processes. The key research priority is to:

- Develop methodologies to study microlayer reactions including optical and molecular techniques
- Study microlayer reactions in a variety of environments

#### 4.1.4 Microlayer sampling

The sea-surface microlayer is conceptually defined as the interface between the oceans and atmosphere with unique physicochemical and biological properties from the underlying bulk seawater, with a thickness as thin as 1 micron. However, in practice the sea-surface microlayer is operationally defined by how the sea-surface microlayer is sampled and how samples are preserved for analysis. From this perspective, the microlayer can be as thin as 50 microns or as thick as 200 microns or greater. Therefore, the method of sampling and the microlayer thickness collected can result in very different conclusions regarding microlayer microbiomes, organic enrichments and physicochemical and photochemical processes. Hence it is necessary to:

- Standardize microlayer sampling and intercalibrate methods and procedures.
- Develop optical techniques and remote sensing methods to study the sea-surface microlayer so that sampling can be made of truly the microlayer and not the water below it

## 4.2 Surfactants

Surfactants are amphiphilic molecules that partition across and are enriched at air-water interfaces (e.g., the sea surface, surfaces of bubbles in the water column, aerosol surfaces), reducing the interfacial tension of those surfaces. Understanding the distribution and role of surfactants in the surface ocean and aerosols is central to all five SOLAS themes. Surfactants in the surface microlayer hinder gas exchange across the ocean-atmosphere interface of volatile compounds, including greenhouse gases (e.g., CO<sub>2</sub>, CH<sub>4</sub>, volatile organic compounds, N<sub>2</sub>O) and naturally produced ozone-depleting substances (e.g., halocarbons), (SOLAS theme 1). Surfactants impact the chemical and physical properties of the surface microlayer, which affect diffusive air-sea fluxes and alter the emission of primary marine aerosols (SOLAS theme 2). Surfactants influence surface-microlayer physical properties and play a critical role in how material from atmospheric deposition crosses the surface microlayer. Additionally, surfactants can alter the properties of atmospheric aerosol particles, such as particle size or aqueous solubility, and thus the likelihood they will be removed from the atmosphere through deposition (SOLAS theme 3). Surfactants affect aerosol properties by creating coatings and altering surface tension. A reduction in aerosol surface tension favors its growth into cloud droplets by reducing the required critical supersaturation. This may also alter the number and size of cloud droplets and thus the radiative properties of the clouds (SOLAS theme 4). Surfactants both at the sea surface and in particles may alter gas-particle interactions, including heterogeneous reactions, which affect chemical reactions in the atmosphere (SOLAS theme 5).

Understanding surfactants' impacts on SOLAS science is contingent on understanding the cycling of surfactants in the oceans. Several key uncertainties and research priorities are highlighted to improve our understanding of surfactants in the oceans and atmosphere.

#### 4.2.1 Surfactant chemistry, including sources and sinks in the surface ocean

Very little is known regarding sources and sinks of marine surfactants. In particular, it is not known if surfactants are photochemically produced, altered, or destroyed, or whether ecosystem processes affect surfactant concentrations and composition. Since phytoplankton are the primary source of dissolved organic matter (DOM) in the ocean, studies are needed to determine if there are predictable relationships between algal assemblages, their physiological status, and the composition of surfactants in the upper

ocean and atmosphere. Much of the DOM in the ocean is relatively old and recalcitrant (RDOM), and therefore decoupled from short-term local biological processes. Does this RDOC have surfactant properties, as suggested by Kieber et al. (2016)? Additionally, the extent to which remote sensing could be used to study the presence and concentrations of surfactants in the surface ocean should be investigated. To this end, it will be important to determine the optical properties of surfactants at the sea surface and in aerosols. Research is needed to:

- Determine how processes on a range of spatial and temporal scales contribute to the composition of different reservoirs of organic matter in the ocean, including surfactants.
- Determine the fraction of dissolved, colloidal, and particulate organic carbon that exhibits surfactant properties and whether these fractions and associated production and destruction mechanisms vary spatially and temporally.

#### 4.2.2 Surfactant physical properties and effects on SOLAS-relevant processes

Surfactants reduce the surface tension at interfaces and affect processes such as aerosol growth and activation, air-sea gas fluxes and bubble rafting. Current models of the growth of aerosol particles and their activation into cloud droplets often ignore the effect of surfactants on the surface tension and coatings of particles. While research has shown enrichment of organic compounds in the surface microlayer and in primary marine aerosol, the enrichment of surfactants, including potential composition-dependent enrichment, is still a topic of current study. At the sea surface and in marine aerosols, surfactant films may hinder the exchange of gases across the interface. Surfactants will influence bubble properties, including the lifetimes and formation of bubble rafts at the sea surface. This in turn may influence the production of primary marine aerosols and their resulting number and mass size distributions and chemical compositions. Very little is known about the influence of surfactants on these processes; critical areas of surfactant research span many fronts:

- Evaluate the role of surfactants in reducing mass transfer coefficients and air-sea gas fluxes
- Determine the influence of surfactants on the growth of aerosols in sub-saturated environments, and the surface tension of aerosols and their activation into cloud droplets
- Quantify the concentration, composition, and properties of surfactants in primary marine and atmospheric aerosols
- Directly measure surface tension depression in atmospheric aerosols and relate that surface tension to the fractions of organic mass and surfactants
- Identify the role of the structure and molecular composition of seawater surfactants on physical and environmental properties and processes, such as reduction of surface tension, mass exchange, aerosol enrichments, and bubble formation and bursting

#### 4.3 Air-sea gas exchange

Even though air-sea gas exchange most directly relates to SOLAS theme 2, it was identified as a cross cutting theme due to its connections to multiple other SOLAS themes. For example, air-sea exchange of CO<sub>2</sub> and other greenhouse gases, in deep ventilation regions as well as throughout the global oceans, is an important part of the cycle of these gases and is imperative for accurately representing GHG in models (theme 1). Secondary organic aerosols are produced by the oxidation of DMS and other VOC (e.g., halocarbons, carbonyl compounds, alkenes, alcohols, COS) produced in the ocean and then transferred, via air-sea gas exchange, into the atmosphere (theme 2). Indeed, many gases produced by biological and photochemical processes in the ocean are then transferred to the atmosphere affecting atmospheric chemistry and the oxidizing capacity (theme 5). Understanding the transfer of gases across the air-sea interface, therefore, is critical in predicting and understanding the chemistry in the upper ocean and atmosphere.

Uncertainties in air-sea gas exchange limit estimates of the global ocean uptake of atmospheric CO<sub>2</sub> (uncertainties are presently about  $\pm 0.5$  Gt C y<sup>-1</sup>). These uncertainties stem from both estimates of gas

transfer velocity ( $k$ ), and the air sea  $\text{CO}_2$  gradient ( $\Delta p\text{CO}_2$ ). The relative proportion of the total uncertainty in the global air-sea  $\text{CO}_2$  flux from  $k$  or  $p\text{CO}_2$  is not known (though there have been attempts to characterize the uncertainty from data-based approaches (Wrobel and Piskozub, 2016). A major effort is underway to estimate  $p\text{CO}_2$  from pH measurements on Biogeochemical-Argo (BGC-Argo) floats, but uncertainties persist due to assumptions required to calculate  $p\text{CO}_2$  from the pH measured aboard the floats (i.e., total alkalinity inferred from salinity, as well as error in the pH measurement itself). The relationship between wind speed and  $\text{CO}_2$  gas exchange at moderate wind speeds over the open ocean is fairly well quantified (Nightingale et al., 2000; Ho et al., 2006; Wanninkhof, 2014). However, there are very few observations of  $k$  and  $p\text{CO}_2$  in high wind and wave areas, in coastal and inland seas, and in ice-covered waters, which contribute to uncertainties in the ocean carbon sink that are reducible only through focused research efforts.

Not surprisingly, gas exchange models developed to parameterize gas fluxes in open-ocean settings do not work in coastal and ice-impacted environments, and they do not account for the effect of Langmuir Circulation and other processes on gas exchange rates. Coastal and inland areas are important because they undergo rapid biogeochemical cycling, and high rates of carbon sequestration. Because of their importance, coastal areas are identified as one of three key environments in all five core themes of SOLAS, but very little is known regarding gas exchange in these environments. Likewise, sea ice zones span  $\sim 10\%$  of the world oceans and are undergoing rapid changes due to global warming, including changes in sea ice extent. Sea ice was long assumed to inhibit air-sea gas and material exchange, but are now understood to be biologically and chemically rich and complex systems that exchange gases with both the atmosphere and underlying water (BEPSII, 2019). Not enough is known about  $k$  or the marine carbon cycle in these environments, and the SOLAS community is uniquely positioned to improve current understanding. Finally, Langmuir Cells generate turbulence and vertical mixing, and they are a common feature in the oceans. They have been suggested to affect gas exchange through the development of downwelling and upwelling currents in the Langmuir cells (Chiba and Baschek, 2010), but very little is known about their effect on gas transfer rates on a regional or global scale.

#### 4.3.1 Turbulence is the fundamental driver of air-sea gas exchange

In the open ocean, turbulence in the upper ocean is primarily generated by wind shear. In coastal areas, turbulence could be generated by wind, local tidal flows, depth-limited wave shoaling and breaking, and attenuated by suspended sediment. Attempts have been made to parameterize  $k$  in shallow water regions with easily measured proxies of bottom-generated turbulence (via current velocity and water depth) and wind-generated turbulence (via wind speed) (Ho et al., 2016), but it is not clear if these parameterizations are universally applicable. Additionally, there are not enough dissolved gas measurements in these environments to account for spatially heterogeneity and temporal dynamics; and often times ship scientists are required to turn off their measurement systems in the exclusive economic zone of other countries.

Gas exchange process studies in a variety of environments (see below for particularly understudied environments) should include deployments of  $^3\text{He}/\text{SF}_6$  dual tracers, eddy covariance (EC) methodologies, made concurrently with measurements of wind speed, waves, turbulence, bubbles, and surfactants. This suite of techniques and measurements, and novel others, should be used to:

- Undertake creative and comprehensive field efforts to improve measurements and parameterizations of  $k$  by directly including turbulence and other properties on which that gas exchange depends
- Make measurements of dissolved gases in areas where measurements are currently lacking, from research vessels, ships of opportunity, and autonomous platforms.
- Ascertain the importance of Langmuir Circulation on gas exchange

#### 4.3.2 High wind speed regions

High wind speed regions suffer from a lack of gas exchange measurements and are also under-sampled for  $p\text{CO}_2$  and other dissolved gases. Wrobel and Piskozub (2016) showed that the spread in  $k$  produces uncertainty in the global  $\text{CO}_2$  fluxes of less than 10% when applying only the various quadratic parameterizations (Nightingale et al., 2000; Ho et al., 2006; Wanninkhof, 2014). However, this uncertainty rises to 65% when including non-quadratic parameterizations (McGillis et al., 2001). A similar uncertainty is expected for the gas transfer of other gases (DMS, VOC, etc.) in high wind speed regions. Whereas these non-quadratic parameterizations are not considered representative of low wind speed environments, the uncertainty represented by the spread of all parameterizations at high wind speeds may be important. Parameterizations of the gas transfer velocity become highly uncertain at wind speeds  $>15 \text{ m s}^{-1}$ . In addition to the common methods used to measure  $k$ , studies in high-wind speed regions should include simultaneous measurements of  $k$  and wave and bubble characteristics (e.g., wave height and steepness, frequency of wave breaking, bubble size distributions). Additionally, it is imperative to design technologies to quantify air-sea gas exchange on moorings and/or autonomous platforms that can function in high-wind environments as well as to conduct gas exchange studies using ships and platforms analogous to the Floating Instrument Platform (FLIP). For example, in high-wind speed regions,  $p\text{CO}_2$  could be measured by Autonomous Surface Vehicles (ASVs) carrying high-precision  $\text{CO}_2$  sensors (Sutton et al., 2014) that can effectively sample regions with high spatial and temporal variability, and provide crossover points to validate BGC-Argo estimates of  $p\text{CO}_2$  from pH. In short, it is necessary to:

- Design new technologies particularly suited to high wind environments
- Use new and existing methods to quantify the exchange of a variety of gases in high wind conditions

#### 4.3.3 Coastal areas

These areas include inland seas, shelf areas, shallow tidal estuaries (mean depth  $< 5 \text{ m}$ ) with straight and meandering sections (short and long fetch), coral reef ecosystems, seagrass beds, mangrove-lined ecosystems, rocky shorelines, and steep relief coastlines (i.e. bluffs). As previously discussed, it is not advisable to use open-ocean gas exchange parameterizations in coastal areas. In addition to the common measurements used to study gas exchange in seawater, bathymetry, bottom roughness, momentum exchange, and current velocities should also be measured in coastal studies. Important priorities include:

- Conduct coastal gas exchange experiments to test existing gas exchange parameterizations and develop new parameterizations in various shallow water environments
- Use multiple techniques and measure more variables than typically measured in the open ocean in a variety of coastal environments to improve coastal air/sea gas fluxes
- Leverage remote sensing to determine the effect of sediment load (via turbidity of water), CDOM and surfactant concentrations, and whitecap coverage (for bubbles) on gas transfer rates

#### 4.3.4 Sea ice

Sea ice affects gas exchange by limiting direct transfer through the sea ice and perhaps by stimulating transfer within leads within the ice. To increase our understanding of gas exchange in sea ice affected waters, we need to assess existing parameterizations of sea ice physics (e.g., permeability, porosity, mushy-ice – Loose et al., 2014; Roberts et al., 2017; Jin et al., 2018) and biology (e.g., ice algae – Jin et al., 2018) along with the development of new parameterizations for ocean-ice-snow, ice-snow-air, and ocean-ice-snow-air gas and particle fluxes. This will require creating a consistent and inclusive data set for model validation for both the Arctic and Antarctic as well as process studies aimed at improving understanding of gas exchange in ice-covered regions

Ice characteristics such as ice extent, thickness, and concentration are accessible from remote sensing from autonomous platforms. In particular, remote sensing currently provides several products for sea ice extent and concentration, but they do not always agree at the sea-ice margin. There are new algorithms to estimate sea ice thickness and hence volume but these are hard to test and are not applicable to every sea ice region. Ice mass balance buoys deployed in Arctic sea ice are providing invaluable data; their design has been modified so they can survive summer ice melt conditions and record multi-year data. There are sensors already designed and well tested in Arctic conditions year-round for some atmospheric chemical species (BrO and other halogenated volatiles, O<sub>3</sub>, CO<sub>2</sub>; e.g., O-Buoy atmospheric chemistry network) that could be part of new persistent deployments. Sea ice thickness and optical properties can control the light field that will ultimately control sea-ice photochemistry and determine the phenology of ice algae vs. phytoplankton processes and associated food webs; these processes will result in varying fluxes of DOM and gases into the sea ice and underlying/surrounding seawater that will be available for gas and particle exchange to the overlying atmosphere on a seasonal basis. Overall, to better understand air-sea gas exchange in ice-covered regions it is necessary to:

- Conduct gas exchange process studies in partially ice-covered regions as well as in simulated environments (i.e. lab settings) to improve understanding of *k* in ice-covered regions
- Perform experiments to determine the impact of sea ice on fluxes of gases (e.g., CO<sub>2</sub>, O<sub>2</sub>, DMS, BrO) and aerosols (e.g., sea salt) between the ocean and the atmosphere, and to determine the impact of sea ice on gas concentrations (e.g., CO<sub>2</sub>, O<sub>2</sub>, DMS) in surface seawater
- Determine the impact of sea-ice biogeochemistry on dissolved gas cycling in sea ice
- Improve sea-ice remote sensing algorithms
- Develop sensors to be integrated with existing and/or new such autonomous, automated ice-tethered systems (e.g., NSF Arctic Observing Network)

#### 4.4 Halogens, reactive nitrogen, and redox chemistry

The global impact of tropospheric reactive halogens on global atmospheric chemistry is relatively unknown, but is thought to decrease the oxidation capacity of the atmosphere through reductions in ozone, hydrogen oxides (HOx), and nitrogen oxides (NOx). Therefore, understanding the sources, sinks, and redox reactions of halogenated compounds in marine air and the ocean are relevant to multiple SOLAS themes. Recent models suggest that the largest source of tropospheric reactive halogens derives from reactions on sea salt aerosol (Br, Cl) (Chen et al., 2018) and the ocean surface (I) (Sherwen et al., 2016), demonstrating a mechanistic link between the surface ocean and lower atmosphere (Themes 4, 5). Additionally, air-sea gas exchange can result in emission of gas-phase CH<sub>3</sub>Br and CH<sub>3</sub>I (Theme 2).

Dating back to the 1980s, observations of episodic ozone depletion events in the Arctic spring revealed a causal role for Br (Barrie et al., 1988), originating from chemical reactions involving bromine in the gas- and aerosol-phase (Evans et al., 2003). These heterogeneous reactions have recently been included in global chemical transport models, which suggest a significant impact of reactive halogens on tropospheric oxidants such as ozone and the OH radical (Parrella et al., 2012; Schmidt et al., 2016; Sherwen et al., 2016). This, along with reported oceanic emissions of CH<sub>3</sub>Br and CH<sub>3</sub>I (for review see Liss et al. 2014), indicates a significant role for halogens in atmospheric chemistry and control of the oxidative capacity of the atmosphere (Themes 4 and 5). Alkyl nitrates also represent a significant contribution of reactive nitrogen to the remote marine atmosphere (Themes 2, 4, and 5).

Photochemical and biological processes create reactive oxygen species (ROS; O<sub>2</sub><sup>-</sup>, <sup>1</sup>O<sub>2</sub>, HOOH, OH radical) in seawater and in ice (Powers and Miller, 2015; Diaz et al., 2013; Ossola et al., 2021; Kieber et al., 2003). Organic radicals are produced as well, and together, these radicals will alter redox reaction pathways on time scales controlled by changing solar irradiance, photon absorption, and ecological/biological function. Direct measurements of ROS in surface seawater are scarce but may alter microlayer, surfactant, and aerosol compositions and should be accurately defined. Photochemical

halogenation of DOM has been reported (Diego et al., 2014; Hao et al., 2017), but the consequences of these new compounds to halogen exchange and cycling are not known.

While recent model simulations demonstrate the importance of oceanic sources of reactive halogens and nitrates to atmospheric chemistry, the variability of these sources over space and time is not known. There are too few observations of tropospheric reactive halogens for a rigorous evaluation of model performance. The mechanisms for surface ocean sources and sinks for these reactive compounds are not well-defined. Short-lived reactive oxygen and organic radical species that populate the surface ocean are kinetically controlled and may create important temporal and spatial deviations from the thermodynamic oxidation state expected in oxygenated surface seawater. These may have consequences that link to biogeochemical cycles (Theme 5), the fate of atmospheric deposition of trace elements (Theme 3), halogen chemistry, and air-sea interactions through complex biogenic and chemical redox reactions.

#### 4.4.1 Ocean-atmosphere interactions involving halogens

The ocean is both a source and a sink for  $\text{CH}_3\text{Br}$ ,  $\text{CH}_3\text{I}$ , and a source of reactive bromine to both the troposphere and the stratosphere, with implications for both stratospheric ozone and altered oxidative capacity of the troposphere. Prior to the 1990s, the oceans were a net source of  $\text{CH}_3\text{Br}$  but with changes in anthropogenic emissions of  $\text{CH}_3\text{Br}$  there is now an open question as to whether or not the oceans have since become a net sink. Furthermore, the abundance of reactive halogens in the MBL (e.g.,  $\text{CH}_3\text{I}$  and  $\text{Cl}$  radicals) and the processes resulting in sinks and chemical production of these species from the ocean surface, either through direct emission or production from heterogeneous reactions on sea salt aerosol, are not quantified. Models estimating the oxidative state of the troposphere are poorly constrained, with important implications for the lifetime of GHGs such as methane. To address these gaps, it is imperative to:

- Make comprehensive gas-phase measurements in the marine troposphere of reactive bromine ( $\text{HOBr}$ ,  $\text{BrO}$ ,  $\text{BrNO}_3$ ,  $\text{HBr}$ ), chlorine ( $\text{ClO}$ ,  $\text{HOCl}$ ,  $\text{ClNO}_2$ ,  $\text{ClNO}_3$ ,  $\text{Cl}_2$ ,  $\text{HCl}$ ) and iodine ( $\text{HOI}$ ,  $\text{HNO}_3$ ) combined with aerosol-phase measurements of  $\text{Cl}^-$ ,  $\text{Br}^-$ , and  $\text{I}^-$
- Determine the saturation state of  $\text{CH}_3\text{Br}$ ,  $\text{CH}_3\text{I}$  and the distributions of organohalogens in the surface ocean
- Conduct mechanistic studies to elucidate sources and reaction pathways of  $\text{CH}_3\text{Br}$ ,  $\text{CH}_3\text{I}$  and related species

#### 4.4.2 Ocean-atmosphere interactions involving reactive nitrogen

While alkyl nitrates can be a significant component of reactive nitrogen in the marine troposphere, the controls for their production and emission from the ocean, spatial distribution and relationship with nitrite in seawater, are not fully understood. There is an open question as to whether the production of alkyl nitrates in the surface ocean is entirely photochemical or has an additional biological source. The observation that production of alkyl nitrates in the Atlantic is lower than in other open ocean systems cannot currently be explained. Consequently, there is a need to:

- Conduct shipboard observations of alkyl nitrates, together with appropriate incubations, culture work, in situ and physiological studies with related photochemical studies to establish relative significance for sources

#### 4.4.3 Oxidation state of the marine boundary layer and surface ocean

Specific controls on ozone flux to the surface ocean are not well understood and will need to be determined before quantitative models of the variations in the MBL oxidative state can be accurately modeled. There is evidence that ozone reacts with iodide in seawater but not with iodate (Martino et al., 2012). Consequently, careful analysis of iodide/iodate ratios in surface seawater may provide critical confirmation of ozone reactions and inform changes in MBL oxidation state. Photochemistry creates a myriad of redox active compounds and radical transients that could alter biogeochemical redox pathways

on a daily cycle, and would have a strong seasonality at critically important high latitudes. The presence of reactive transients could alter solubility of atmospherically deposited trace elements and nutrients, thus providing feedback for biogenic gas production and ecosystem function. Important priorities include:

- Perform shipboard and laboratory studies to validate measured ozone deposition flux/velocity at sea in the context of specific kinetics and mechanisms for ozone reactivity with the ocean surface (with a specific focus on microlayer and iodide/iodate measurements).
- Develop new in situ methodology for direct measurements of the oxygen and organic reactive transients as they relate to shifts in oxidative state on microscales relevant to dissolution and microlayer reactions,
- Investigate larger scale spatio/temporal influences on the oxidative state of the surface ocean

#### 4.5 Air-Sea Exchange and Evolution of Atmospheric Aerosols

The deposition of aerosols to the ocean surface and ejection of primary marine aerosol (PMA) from breaking waves are important components of all five SOLAS themes. For example, deposition of nutrient-laden aerosols to the ocean surface can stimulate primary productivity leading to the uptake of atmospheric CO<sub>2</sub> into the ocean while the atomic Cl produced via multiphase reactions involving PMA oxidizes finite amounts of atmospheric methane (Theme 1) (Meskhidze et al., 2003; Mahowald, 2011; Lawler et al., 2009; Wang et al., 2019). Energy exchange and associated wave action at the air-sea interface modulate both the production and deposition of aerosols as well as the exchange of gases that lead to the production of secondary organic aerosol (SOA) (Theme 2) (Meskhidze & Nenes, 2006; Claeys et al., 2010; Lewis and Schwartz, 2004). Halogen-radical chemistry involving marine aerosol accelerates the oxidation of NO<sub>x</sub> to HNO<sub>3</sub> and the oxidation of marine VOCs leading to the formation of marine-derived SOA (Keene et al., 2009). In addition, the pH-dependent gas-aerosol phase partitioning of HNO<sub>3</sub> and NH<sub>3</sub> modulate the size distributions and corresponding size-dependent dry-deposition rates of particulate NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> as well as the total (gas + aerosol) dry deposition fluxes of both HNO<sub>3</sub> and NH<sub>3</sub> to the ocean surface (Russell et al., 2003; Fischer et al., 2006; Smith et al., 2007; Baker et al., 2018). Both sets of processes modulate the atmospheric lifetimes and total deposition fields of nutrient N to the ocean thereby influencing biogeochemistry in near-surface seawater while the wet and dry deposition of dust, biomass burning, bioaerosols, combustion and other aerosol sources provide Fe and P to the ocean (Theme 3) (Barkley et al., 2019; Mahowald et al., 2005; Ito et al., 2019). Numerous studies have investigated the influences of marine aerosols including PMA and dust and, more recently, marine SOA as functions of size and composition on marine clouds (Theme 4) (Gantt et al., 2012; Mayer et al., 2020). Finally, biogeochemical processes in the surface ocean exert significant but poorly characterized controls on the size-resolved production fluxes of marine aerosol to the atmosphere and the associated impacts on atmospheric chemistry (Theme 5) (Gantt et al., 2010; Galí et al., 2018).

Significant advances have been made in modeling, in situ and laboratory measurements, and remote sensing of aerosols. Detailed atmospheric models have been developed for the production of sea spray aerosol and the delivery of nutrient-laden aerosols from different sources (e.g., mineral dust, combustion, biomass burning) to the open ocean. These models now routinely diagnose aerosol mobilization at the source region, in addition to transport and transformation in the atmosphere as well as removal by wet and dry deposition. Several past and ongoing international research efforts (such as GEOTRACES and NAAMES) have provided crucial insight needed for improved characterization of aerosol effects on ocean biogeochemistry and clouds. More frequent coverage over polar regions has improved the observations of transport of pollution, smoke, and glacial dust into the polar marine environment. New remote sensing instruments such as advanced optical spectrometers, polarimeters, and Lidars are highly promising in that they can now provide essential retrievals of phytoplankton biomass, pigment concentrations, physiology, and taxonomic diversity, improve atmospheric corrections for ocean color retrievals, and detailed vertical distribution of atmospheric aerosol type and refractive properties.

Despite such progress, substantial gaps remain in our knowledge of marine aerosols. For example, although the dynamics of bubble plumes have been investigated for decades (Thorpe, 1982), air injection rates as a function of sea state and the associated properties of detraining plumes that modulate the size-resolved production fluxes of PMA number, mass, and composition remain poorly constrained. Likewise, very little is known about the impacts of the physicochemical evolution and aging of marine aerosol on the oxidation of marine-derived-organic matter that affects SOA and PMA, the production and cycling of halogen radicals (Zhu et al., 2019), and the solubility of nutrients deposited to the surface ocean (Nenes et al., 2011; Shi et al., 2012). These significant knowledge gaps limit our current predictive capability for the multi-phase physical and chemical evolution of the marine troposphere and corresponding influences on Earth systems, including radiative transfer and climate.

Several interrelated processes that modulate marine aerosol production and evolution of marine atmospheric composition warrant investigation. Specific areas of uncertainty and key research to address these uncertainties are discussed below. Some of the research priorities discussed below are likely best answered through the establishment of additional long-term field sites to investigate the ocean's biological and biogeochemical responses to nutrient and toxic types of aerosol inputs (dust, combustion, and volcanic aerosols) (see Section 5.2). Additionally, research will be most useful if it is used to develop new or improved parameterizations of aerosol production, transport, solubility, and bioavailability that are included in models of various scales and complexities (see Section 5.5).

#### 4.5.1 Physical drivers of open-ocean wind waves

Virtually all parameterizations of marine aerosol production are driven directly or indirectly by 10-meter wind velocity ( $U_{10}$ ) as a proxy for wave action. However, oceanic wind waves are typically not in equilibrium with local conditions, and  $U_{10}$ -based approaches do not account for important non-linear processes (including influences of wind fetch and duration, bathymetry, and interactions between wind and wave fields) that modulate ambient sea state. Consequently,  $U_{10}$ -based approaches do not capture the wave dynamics that control the volume and injection depth of air entrained into the surface ocean and the associated marine aerosol production via bubble bursting during plume detrainment. Studies are thus needed to:

- Assess PMA fluxes using near sea-surface wind field dynamics
- Determine PMA physicochemical characteristics in different wind regimes

#### 4.5.2 Bubble plume dynamics and bubble rafts

Characteristics of bubble plumes (e.g., bubble size distribution, void fraction, bubble age) vary as functions of seawater type (e.g., biologically productive vs. oligotrophic). All of these factors (as well as plume depth) influence production fluxes of marine aerosol produced via bubble bursting during plume detrainment (e.g., Keene et al., 2017). Available evidence also suggests that surface bubble rafts modulate the production flux of PMA (Frossard et al., 2019), but the influences of raft dynamics on PMA production are largely unconstrained. Studies should:

- Investigate the physical (e.g., sea-state, temperature, raft thickness) and chemical (e.g., surfactant concentrations and composition) factors that modulate bubble plume characteristics and dynamics to develop a mechanistic understanding of and reliable predictive capability for PMA production
- Determine how and to what extent organic surfactants (Section 4.2) affect the physical and chemical properties of PMA produced via bubble bursting

#### 4.5.3 Primary marine aerosol production and evolution

Dependence of size- and composition-resolved production on major drivers such as plume dynamics, surfactants, and temperature remains poorly understood. Available evidence suggests that results from studies based on artificial seawater, coastal seawater, amended seawater, and/or incubation studies may not be representative of ambient aerosol production over the open ocean, either because of

methodological differences or natural variability. Furthermore, the physical and chemical processing of PMA during its atmospheric lifetime is poorly constrained (Zhou et al., 2008) and warrants further investigation. Thus it is necessary to:

- Conduct an intercomparison study to directly compare the performance of marine aerosol generators employed by different research groups
- Determine the factors that affect PMA production in open-ocean oligotrophic seawater based on the detrainment of representative bubble plumes
- Conduct studies to determine how the ocean's physicochemical and biological state affect the production flux, number size distribution, hygroscopic, and ice nucleating properties of PMA
- Perform field and laboratory studies to investigate the physicochemical evolution of organic and inorganic matter associated with PMA during multi-day exposure to atmospheric conditions and to assess the size and hygroscopicity-related changes in the context of the PMA importance to CCN and IN formation, tropospheric chemistry (e.g., halogen chemistry), and the recycling of marine-derived organic matter across the air-sea interface
- Investigate how the physicochemical characteristics of marine aerosols are transformed during atmospheric transport and interactions with clouds

#### 4.5.4 Aerosol nutrients and bioavailability

Comprehensive characterization of the effects of atmospheric deposition of aerosols on surface ocean productivity requires knowledge of aerosol-nutrient bioavailability after deposition to the surface ocean. However, the concept of nutrient bioavailability in seawater is extremely complex and poorly defined, often with different nomenclatures across disciplines leading to variable results. Overall, very little is known about the impact of wet and dry deposition of atmospheric constituents on the biogeochemistry and ecology of the upper ocean. To propel this emerging research area forward it is imperative to:

- Standardize methodologies to study nutrient bioavailability
- Investigate the ocean's biological and biogeochemical responses to nutrient and toxicant aerosol inputs
- Design experiments to address the fundamental question of how does solubility affect atmospheric deposition fluxes which in turn relates to changes in bioavailable nutrients in the ocean
- Target laboratory incubation experiments to elucidate how differences in bacterial and phytoplankton taxa or species within taxa affect bioavailability and nutrient uptake mechanisms
- Explore the potential co-influence of different nutrients (and trace metals), presence of different organic compounds (both in the atmosphere and in the ocean), aerosol speciation and solution pH, light, and microbial interactions affect nutrient bioavailability in aerosol pre- and post-depositional processes
- Use newly improved remote sensing tools – and continue to design new sensors – to increase understanding of aerosol-ocean biogeochemical interactions, such as the feedbacks between changes in aerosols sources, ocean physicochemical properties, and biological state

## 5. Implementation

### 5.1 Leveraging Existing Platforms and Programs

Existing atmospheric and oceanographic platforms and programs are critical for measurements that advance SOLAS science. Atmospheric towers, buoys, satellites, oceanographic time series, repeat hydrography, and autonomous surface and underwater vehicles (AVs) are all needed for progress in air-sea research.

#### 5.1.1 Atmospheric Towers

Atmospheric observations from land-based towers provide extensive, long term, vertically-distributed data that are difficult to replicate over the ocean. Similar data, however, when obtained close to the sea provide transformative insight and are of tremendous value. Six such atmospheric towers are located at (1) [Tudor Hill](#), Bermuda, linked to the Biological Institute for Oceans Science (BIOS), used for study of aerosol deposition relative to ocean productivity; (2) [Ragged Point](#), Barbados, linked to the University of Miami, used to examine long-range transport of African dust to the Caribbean for over 50 years; (3) [Mace Head](#), Ireland, linked to the National University of Ireland, collecting gas and aerosol samples from the N. Atlantic; (4) [Penlee Point Atmospheric Observatory](#), United Kingdom, measuring atmospheric composition and making eddy covariance observations of coastal air/sea exchange linked to seawater composition measurements in the Western English Channel; (5) [Cape Verde Atmospheric Observatory](#) measuring atmospheric composition in a region dominated by outflow of Saharan air masses; and (6) Cape Grimm, Tasmania, providing high latitude observations of Southern Ocean marine aerosols. These and other sites provide time-series data to document critical parameters such as dust, marine organic aerosols, and the flux of mass and momentum across the air-sea interface. These measurements are essential to constrain global climate models that link seasonal variability in atmospheric material deposition to responses in marine biogeochemistry. Future studies should incorporate existing infrastructure and time-series data, as well as identify crucial locations to establish new monitoring stations that would expand opportunities to link with process-level studies addressing wet and dry deposition, upper ocean nutrient cycling, and kinetics for aerosol-derived nutrient transformations into dissolved, colloidal, and particulate forms in seawater.

#### 5.1.2 Autonomous Vehicles

Ocean-going platforms, particularly research vessels, remain critically important for the study of many of the themes and cross-cutting questions described in Sections 3 and 4. Historically, research vessels capable of transporting innovative analytical systems to a wide range of oceanic environments have been crucial to building a global understanding of oceanographic and atmospheric processes. The advent of smaller, agile autonomous vehicles (AVs: Autonaut, Wave Glider, Saildrone, and underwater Gliders) have already proved their utility for obtaining robust, air- and water-side near-surface measurements to augment research vessel campaigns. Maturing AV technology will likely further increase their incorporation into ocean-going field studies important for SOLAS-oriented research. Bridging the substantial observational gap between traditional research vessels and AVs remains a challenge. While smaller AVs excel in agility and operational versatility, they have relatively limited payloads and endurance. Large vessels overcome these but are limited during extreme ocean-atmosphere conditions and they can strongly disrupt the air-sea interface. Novel approaches that link data between the platforms are needed.

#### 5.1.3 Specialized Platforms

Specialized platforms, such as the Floating Instrument Platform (FLIP), provided a tower-like platform deployed in the deep ocean specifically for air-sea interaction studies. However, FLIP has been divested and there is no modern equivalent. Modern platforms similar to FLIP would be useful to address many of the research questions previously discussed in this report. Moored or drifting buoys optimized for air-sea

interaction measurements, such as the Air-Sea Interaction Spar (ASIS) and Extreme Air-Sea Interaction (EASI) buoys, are especially useful for studying air-sea interactions during high-energy conditions. The surface-following capability of ASIS allows it to make directional wave and air-sea interaction flux measurements at high resolution with a minimal drag profile (Graber et al. 2000), making it especially valuable for near-surface measurements on both the atmospheric and oceanic side. EASI (Drennan et al., 2014) is a Navy Oceanographic Meteorological Automated Device (NOMAD) style heave-pitch-roll buoy specifically designed for high wind environments and long-term deployment. Both ASIS and EASI are capable of sampling highly energetic conditions where platforms or ships cannot safely operate.

#### 5.1.4 Lab Facilities

Current difficulties in making high-wind measurements and isolating key processes in field measurements make the combination of in situ observations and laboratory measurements highly desirable. As discussed in Section 3.2, current estimates of the rates of momentum, heat or gas exchange in extreme conditions are not reliable, largely due to the inability of ships to sample in hurricane or typhoon conditions. Controlled studies of air-sea exchange in high winds are therefore critical to improve numerical models used in operational hurricane prediction. For example, the [SURge-SStructure-Atmosphere INteraction](#) (SUSTAIN) laboratory at the University of Miami, one of a number of similar facilities around the world, can generate computer-controlled directional waves combined with direct wind forcing equivalent to  $U_{10} > 90 \text{ m s}^{-1}$ . The added ability to use fresh or filtered seawater at 5-35°C enables rigorous gas transfer studies. Discrete laboratory experiments are also useful for aerosol solubility studies (Section 4.5) and process studies related to multiple SOLAS themes. While much can be learned from these laboratory studies, in all cases careful consideration of how laboratory results are translated into real-world systems is critical.

## 5.2 Field Campaigns: Past and Future

The GasEx campaigns, the North Atlantic Aerosols and Marine Ecosystems Study (NAAMES), and EXPORTS Processes in the Ocean from RemoTe Sensing (EXPORTS) have generated valuable field data for the US SOLAS community while making clear the planning, logistical challenges, and agency engagement required for success. Large field campaigns muster the SOLAS community and provide unique collaborative and training opportunities for early career scientists. Funding agency coordination is effective to leverage resources for complementary research as was done for NAAMES and EXPORTS (NASA-led with NSF support). Engaging non-US partners promotes coordination and funding partnerships. The following considerations are helpful for SOLAS multidisciplinary field campaigns:

- Engage with international SOLAS and the non-US SOLAS community in the early stages of US-led field projects to maximize the added value that comes from international collaboration
- Identify existing field assets and/or programs for coordination and leverage
- Prioritize and clearly articulate measurements and compelling science questions
- Plan for logistical challenges and potential delays, especially in remote locations
- Develop and clearly communicate administrative and data management plans

Crosscutting US SOLAS themes (Section 4) will require specific activities for successful process studies and field campaigns to advance understanding, including:

- Shipboard air-sea gas exchange experiments longer than 2-3 weeks that coordinate with stable platforms in coastal systems and remote and autonomous platforms in high-wind speed regions (e.g., Pacific sector of Southern Ocean)
- Coordinated aircraft and shipboard Lagrangian studies in high-dust regions such as the North Atlantic (African dust), North Pacific (Asian and Alaskan dust), Southern Ocean (Patagonian dust), and Arctic (proglacial dust and long-range transport from lower latitudes) to elucidate atmospheric deposition effects on ocean biogeochemistry

- Investigations of VOC, trace gas, and aerosols produced directly and indirectly by microorganisms across a range of seasons and oceanographic provinces

Remote and extreme environments are chronically under-sampled with regard to SOLAS priorities and logistics for access and deployment of instruments are costly and challenging. Regardless, opportunities to expand data should be pursued. For example, recent ice-free passages have made the Arctic more accessible for observation of air-sea dynamics. New data on deposition of Alaskan glacial dust can be compared to impacts of Asian and domestic Fe deposition in HNLC regions, as well as similar Southern Ocean studies. Altered deposition and biogenic marine aerosol sources from changing productivity in newly ice-free areas can raise compelling questions that contrast the atmospheric and biological changes along the NW Passage and waters off the coast of Canada and West of Greenland (Baffin Bay and Nares Strait).

### 5.3 Remote Sensing Technologies

The US SOLAS community will work together with the remote sensing community to add value to existing data and provide science priorities for future airborne missions important to air-sea interactions. Thus, we will contribute directly to research areas identified in the Decadal Survey as priority funding areas for NASA, NOAA, and USGS. Existing assets with ongoing and archived data that can enhance US SOLAS goals include PACE, HypIRI, OMI, CALIPSO, CloudSat, MODIS, VIIRS, MISR, IceSat-2, and MOSAiC, which provide comprehensive observation of remote marine regions over long time-scales. The SOLAS and remote sensing communities should fund joint science projects, internships, fellowships, and postdoctoral opportunities, and use existing social media to advertise and raise interest.

The US SOLAS community should:

- Consider parameters that can be linked to space observables and unobservable extrapolations;
- Locate field studies in coordination with relevant satellite tracks to enhance scaling of in situ observations for cal-val efforts for new and established applications and models that link spatial scales useful for both scientists and policymakers;
- Use in-kind NASA services to gather satellite-related datasets for marine and satellite aerosol product validation. For example, Aeronet offers shipboard radiometers to measure atmospheric aerosol optical depths for use as a proxy of aerosol mass or number concentration.
- Engage the remote sensing community to identify critical key parameters (e.g., wave, bubble, or wind variables; greenhouse gas fluxes at the ocean surface; aerosol deposition rates) most useful for interpreting current and future satellite data.
- Contribute white papers for the next Decadal Survey so that SOLAS interests and expertise are represented in the development of future research priorities and missions.
- Explore novel applications of satellite remote sensing and autonomous (e.g., BGC-Argo – see section 5.1.2) data sets to address US SOLAS research priorities (e.g., SOLAS-sponsored workshops to highlight emerging observing technologies and discuss scientific applications).

### 5.4 Numerical and Data Analysis Tools

As an integrating activity, US SOLAS will seek to address the key challenges and opportunities that lie in connecting surface ocean and atmospheric data from a myriad of diverse platforms. We need to embrace rapidly-evolving data analysis tools, such as machine-learning approaches, to explore novel interpretations of existing data sets. Neural networks, the random-forest method, and other machine-learning techniques, together with more traditional statistical techniques, are increasingly used to address SOLAS- and OCB-centered activities such as air-sea gas exchange. Engaging with data scientists at all stages of research will optimize the value of an ever-growing oceanographic observation network and train students in emerging data tools.

Observing System Simulation Experiments (OSSEs) provide numerical model simulations of the ocean and/or atmosphere to test system design during the planning stage of field operations. Model output

is sampled by a hypothetical observing system with subsampled output then analyzed using techniques to be used on real observations. Reconstructed variables are compared to the known model “truth” where created systematic and/or random errors evaluate the ability of a proposed observing system to quantify a variable of interest. Various designs for an observing system can thus be “examined” to maximize performance relative to cost. When underlying models are statistically similar to the real ocean (e.g., resolves the mesoscale), this is an indispensable tool for field planning. Moreover, real field data resulting from OSSE optimized design can be assimilated into existing models or used to minimize errors in a state estimate. An iterative approach to improve models for subsequent OSSE-based field planning maximizes the value of data collection and analysis.

Targeted funding for data mining, development and application of numerical methods to analyze existing and future data, as well as the creation of tools to plan observational programs should be a high priority, thus ensuring maximal insight from valuable cross-disciplinary data sets.

## 5.5 Model development

Models are an important tool for scaling limited data sets for exploration of earth system processes and feedbacks. US SOLAS priorities for improved model performance include many of the themes and crosscutting topics covered in this report including but not limited to GHG, VOCs, gas exchange, upper ocean biogeochemistry and marine optics. Additionally, a sustained effort to enhance existing global databases and quantify critical processes will be essential to support further model development and evaluation. For example, many models rely on observation-based climatologies to estimate emissions (DMS: Lana et al, 2011; halocarbons: Ziska et al., 2013). The databases and techniques used to generate the climatologies themselves may need to be updated in light of recent work (Bell et al., 2021) and new measurements (e.g., satellite proxies for DMSO) and process-based observations. As two examples of ways in which models can advance US SOLAS priorities, primary marine aerosols (PMA) and nutrient cycling are discussed in more detail below.

### 5.5.1 Modeling of marine aerosols

Marine aerosols impact the simulation of low-level clouds and climate sensitivity. Depending on the application, models can simulate the production and removal of primary and secondary marine aerosols as well as their chemical composition, number size distribution, and contribution to the CCN budget. The US SOLAS community should help to further evaluate and refine parameterizations for improved representation of the following drivers in the aerosol models:

- Wind and wave (high-frequency) fields, especially at high wind speed
- Sea surface temperature and solar radiation
- Surfactants, seawater composition, biological activity
- Ocean-ice-snow interfaces
- Bubble plume dynamics

### 5.5.2 Nutrient cycling models

A growing number of models account for nutrient cycling (e.g., Fe, N, and to a lesser extent P) between the land, river, ocean, and atmosphere. Models are able to quantify natural and anthropogenic sources, size distributions and mineralogy, initial nutrient solubilities at source regions, mobilization of nutrients during atmospheric transport (via proton-promoted, organic ligand-promoted, and photochemical mechanisms), or wet and dry deposition. US SOLAS should help to improve parameterizations associated with nutrient cycling across air-water interfaces with the following activities:

- Improve accessibility to existing datasets (e.g., baseline monitoring data) and archival strategies for future campaigns to enhance model evaluation and development

- Better characterize the mix of nutrients associated with different sources (e.g., dust, biomass burning, volcanoes) and their physical form (e.g., dissolved, colloidal, particulate, inorganic, organic ligand-complexed), as well as nutrients in detritus and phytoplankton
- Provide increased data for model constraints of dissolved organic nitrogen and phosphorus
- Assess the impact of atmospheric-derived organics in modulating nutrient bioavailability
- Prioritize quantification of the Arctic paleorecords with respect to changes in nutrient source regions (dust, wildfires)

## 5.6 Data Management

Proper data management is a key tenet of modern science, as many instruments and observing systems can generate giga-scale amounts of data on very short temporal and small spatial scales. US SOLAS research is supported by multiple funding agencies, each with its own data reporting and management requirements. To maximize return on investment and ensure accessibility and advancement of knowledge, investigators must strive for timely data submission and work with data managers and repositories towards FAIR (Findable, Accessible, Interoperable, Reusable, Wilkinson et al., 2016) data practices. The collection and management of US SOLAS data should have the following minimal key features:

- Develop and document QA/QC protocols to ensure accurate results, using inter-laboratory intercalibrations and reference materials where applicable
- Work to enhance interoperability and searchability across multiple US agencies' data repositories and also interface with international repositories that host SOLAS data sets
- Streamline access and functionality for submitting and using US SOLAS data, as well as meeting requirements for funding agency reporting and data management specifications

## 6. Leadership, Coordination, and Communication

In recognition of the critical importance of atmospheric processes and air-sea exchange in the global carbon cycle, OCB recently established a [subcommittee on ocean-atmosphere interactions in partnership with SOLAS](#) to foster increased US coordination and scientific momentum on air-sea exchange of greenhouse gases and feedbacks with marine ecosystems and biogeochemical cycles (Section 2.2). The committee will provide a support system for scientists working at the air-sea interface, especially to overcome the disciplinary segregation that can result from separate atmospheric or oceanic science community platforms and funding. This subcommittee provides vital leadership by organizing interdisciplinary community workshops and activities that serve as incubators for new ideas and collaborations; exploring funding mechanisms; supporting and mentoring early-career SOLAS scientists. The committee is promoting communication to break down disciplinary barriers, building awareness among broad audiences of the importance of air-sea interactions and helping to grow the US SOLAS research area. OAIC members are drawn from an open community nomination process and serve a 3 year term, with the option to serve one additional term for a maximum of 6 years. The OAIC is 9 to 12 scientists with expertise at the boundary of atmospheric and marine science, as well as those with research more heavily weighted to either marine or atmospheric science, all of which make scientific contributions to the broader OCB and SOLAS communities in keeping with their subcommittee work. The OAIC includes at least one international member to build and strengthen ties to ongoing OAIC-relevant scientific research and activities outside the US, and one early career member to lead the subcommittee in planning activities and initiatives to support early career scientists working at this disciplinary interface. As part of, or in addition to the OAIC, topical contacts (e.g., remote sensing, large field campaign coordination, big data analysis) will be identified as a resource of knowledge, information, and community for US SOLAS. As an example, such a contact might help organize an inclusive and open meeting with the objective of aligning existing remote sensing resources with US SOLAS research questions, identifying synergies, developing hypotheses, and planning approaches for testing them.

The OCB Project Office and the SOLAS IPO provide vital coordination, communication, and outreach that benefit their scientific networks and broader audiences. The 2019 Air-Sea Interaction Workshop and community efforts going forward will provide an effective interdisciplinary model and forum for scientific inquiry and discussion. Benefits to the air-sea research community from such activities include inspiration for single-PI and larger-scale collaborative projects and proposals; field campaigns; manuscripts and special journal volumes; input to US and international science and observing initiatives and programs; training activities; and public education and outreach that make air-sea research topics more broadly accessible. The OAIC and project offices will identify and communicate potential research funding opportunities to the US SOLAS community and include federal agencies and other potential sponsors in US SOLAS planning discussions about research and observing priorities. Additionally, it will work with the SOLAS IPO to engage the international SOLAS community on large scale US-led SOLAS projects.

Training and education of the next generation of SOLAS scientists is a high priority. The OAIC includes an early career slot (within Ph.D. + 4 yrs.) and facilitates early career travel support that serves to grow, strengthen, and provide training and professional networking opportunities (e.g., SOLAS summer school, SOLAS open science meeting, International Symposium on Gas Transfer at Water Surfaces, etc.). Training programs and workshops on novel observing platforms, ocean optics, data analysis, and remote sensing will provide scientists with access to valuable tools and data to conduct SOLAS-relevant research.

Centralized communication is critical to support a growing network of US SOLAS researchers. The OCB and SOLAS IPO already serve as communication hubs for large networks and will be leveraged to engage and inform the US SOLAS community via the web, electronic newsletters, and social media about newly published research; permanent and postdoc positions; graduate student opportunities; and relevant meetings and funding opportunities. These project offices also provide access to infrastructure to support tele- and web conferences to help community coordination/engagement.

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## Appendix 1. 2019 US SOLAS Workshop Documents

This will include agenda and participant list

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