

# Workshop on Atmospheric deposition of aerosols and their effects on biogeochemical cycles and climate at II-409

<http://www.isee.nagoya-u.ac.jp/meetings/>

Agenda and abstracts

Version 3.1: December 24, 2019

## **Sunday 22<sup>nd</sup> December**

18:00 Ice breaker in the city center

## **Monday 23<sup>rd</sup> December**

**9:30 – 10:00** Travel cost reimbursement at I-726

**10:00 – 11:30** Closed small group meeting at I-717

**11:30 – 13:30** Lunch at Hananoki restaurant (Japanese)

**13:30 – 13:50** Self introduction

## **Session I: Effects of marine biological emission to the atmosphere on climate**

**Chair: Akinori Ito (JAMSETC)**

**13:50** *Introduction: CLAW hypothesis*

**Akinori Ito (JAMSETC)**

**14:00 – 14:20** *Biogenic sulfur compounds in aerosols over the subarctic North Pacific*

**Yoko Iwamoto (Hiroshima University)**

**14:20 – 15:00** *Biogeochemical linkage between marine atmosphere and surface seawater via organic matter*

**Yuzo Miyazaki (Hokkaido University)**

**15:00 – 15:20** **Coffee break**

**Chair: Michihiro Mochida (Nagoya University)**

**15:20 – 15:40** *Measurement of number concentrations of Bioaerosols in the Central Pacific and classification of fluorescent particles*

**Kaori Kawana (JAMSTEC)**

**15:40 – 16:00** *Substantially low ozone mixing ratios over the Pacific Ocean and implications from iodine chemistry*

**Yugo Kanaya (JAMSTEC)**

**16:00 – 16:20** **Open Discussions**

**Akinori Ito (JAMSETC)**

**16:30 – 17:30** *Closed small group meeting at I-817*

**18:00** **Dinner at Craigs Café**

**Tuesday 24<sup>th</sup> December**

**9:00** *Recent activities in Gas Transfer at Water Surfaces meeting slides delivered by*  
**Hiroshi Tanimoto (NIES)**

*Closing Session I*

**Session II: Effects of atmospheric deposition of aerosols on marine biogeochemistry**

**Chair: Akinori Ito (JAMSETC)**

**9:05** *Introduction: Martin's Fe hypothesis*

**Akinori Ito (JAMSETC)**

**9:10 – 9:50** *Some recent results on Asian dust at desert and coastal areas of China*

**Daizhou Zhang (Prefectural University of Kumamoto)**

**9:50 – 10:50** [Special guest seminar] *Atmospheric nutrients: sources, atmospheric processing and impacts on ocean ecosystems*

**Zongbo Shi (University of Birmingham)**

**10:50 – 11:50** **Open Discussions**

**11:50 – 13:00** **Lunch at chez Jiroud (French)**

**13:00 – 14:00** [Special guest seminar] *Fate of iron after deposited to the ocean--from a modelling perspective*

**Ying Ye (Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research)**

**14:00 – 14:40** *The current status of iron studies in the western Pacific Ocean*

**Jun Nishioka (Hokkaido University)**

**14:40** *Closing Session II*

**Akinori Ito (JAMSETC)**

## **Session I : Effects of marine biological emission to the atmosphere on climate**

### **Introduction: CLAW hypothesis**

**Akinori Ito (JAMSETC)**

#### **Terms of Reference:**

- Review and synthesize the current scientific information on the effects of marine biological emission to the atmosphere on climate, initiated by the CLAW hypothesis.
- Consider the likely changes in marine biological emission into the future and the potential biogeochemical consequences of such changes.
- Identify the key future research needs to reduce uncertainties in predictive capability in this area.
- Publish the results of this activity in the open peer-reviewed scientific literature.
- Interact with, and provide information to, leading relevant international groups including the Future Earth core projects such as SOLAS, and UN programs such as GESAMP.

#### **References**

- Charlson, R. J., J. E. Lovelock, M. O. Andreae, and S. G. Warren (1987), Oceanic phytoplankton, atmospheric sulfur, cloud albedo and climate, *Nature*, 326, 655–661, doi:10.1038/326655a0.
- GESAMP Working Group 38, Report on two workshops on the changes in the acid/base balance of the atmosphere and ocean and their subsequent impacts on air/sea chemical exchange. [http://www.gesamp.org/site/assets/files/1714/report\\_on\\_the\\_wg\\_38\\_workshops\\_feb\\_march\\_2.pdf](http://www.gesamp.org/site/assets/files/1714/report_on_the_wg_38_workshops_feb_march_2.pdf)
- Ito, A., W. Landing, and D. Hamilton, (2019), Atmospheric deposition of iron, ocean biogeochemistry and marine emission of biological aerosols, SOLAS OSC Discussion session reports, April 2019, Sapporo, Japan.
- Shi, Z., E. Achterberg, and A. Ito, (2015), Atmospheric deposition, ocean biogeochemistry and climate change, SOLAS OSC Discussion session reports, September 2015, Kiel, Germany.

## Biogenic sulfur compounds in aerosols over the subarctic North Pacific

**Yoko Iwamoto**

Hiroshima University

Longitudinal distribution of atmospheric sulfur compounds were obtained from an atmospheric observations over the subarctic North Pacific. The concentrations of methanesulfonic acid (MSA) in the Alaska gyre were almost twice of those in the western subarctic gyre. The spatial variation of MSA concentration was similar to that of DMS in both seawater and atmosphere. The high concentrations of DMS in the atmosphere could be caused by high wind speed in the Alaska gyre. Furthermore, composition of oceanic suspended particles including phytoplankton debris showed remarkable increase in coccoliths in the Alaska gyre, indicating that dominant species of phytoplankton also influenced MSA formation. Although about 80% of MSA was existed in fine mode aerosols, the coarse mode fraction of MSA were higher in the Alaska gyre than the western subarctic gyre. This result suggest that seasalts, which were mainly existed in coarse mode, increased due to the high wind speed, and a part of MSA adsorbed on the seasalt particles. Since the residence time of seasalt aerosols are shorter than fine-mode aerosols, the contribution of biogenic sulfur compounds to CCN could be overestimated in the marine atmosphere with abundant seasalt aerosols.

## **Biogeochemical linkage between marine atmosphere and surface seawater via organic matter**

Yuzo Miyazaki

Institute of Low Temperature Science

Hokkaido University

Ocean-derived atmospheric aerosols can affect radiative forcing via acting as cloud condensation nuclei and ice nuclei as well as affecting biogeochemical cycle of bioelements. Marine atmospheric aerosols largely consist of organic matter (OM) associated with phytoplankton and dissolved organic matter in seawater. In recent years, much effort has been devoted to examining linkages between the chemistry of sea spray aerosol (SSA) and the biological and chemical conditions of surface seawater (SSW).

We have conducted field measurements of atmospheric organic aerosols using research vessels as well as a high-altitude observatory over oceanic regions characterized by unique sea-surface microbial activities. These measurements have revealed origins and chemical transformation of OM in marine aerosols by direct comparison of chemical and biogeochemical characteristics between SSA and SSW. In this presentation, an overview of our field research will be given, which includes topics on biogeochemical linkage between marine atmosphere and surface seawater via OM. Also several key questions will be discussed, raised by our field studies toward quantitative understanding of marine-atmosphere interactions via atmospheric aerosols.

# Measurement of number concentrations of Bioaerosols in the Central Pacific and classification of fluorescent particles

Kaori Kawana<sup>1</sup>, Fumikazu Taketani<sup>1</sup>, Kazuhiko Matsumoto<sup>1</sup>, Takuma Miyakawa<sup>1</sup>,  
Akinori Ito<sup>1</sup>, Yutaka Tobo<sup>2</sup>, Yugo Kanaya<sup>1</sup>

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<sup>2</sup>National Institute of Polar Research

Bioaerosols originating from biological activity are enriched in the forest, soil and marine environments. Marine bioaerosols are transported from sea surface to the atmosphere with wind and they could drive climate change via cloud processes by acting as cloud condensation nuclei (CCN) and ice-nucleating particles (INP). In particular, bioaerosols from oceans are significant for INP activation since they may potentially act as INP at higher temperature than other types (e.g., dust and mineral). It means that bioaerosols could act as important INPs for the formation of mixed-phase clouds and low level clouds in the mid latitudes, as well as ice clouds in the polar regions. Nevertheless, distribution/behavior of biological particles over the ocean surface, air-sea interaction, and contribution in the cloud system are still unknown due to lack of observations in the ocean atmosphere, in contrast to land-based studies.

Recently, detection techniques of bioaerosols based on ultraviolet light-induced autofluorescence have been developed. Excitation/emission wavelength patterns may be useful for categorization of these particles. Our group promote the development for innovative instrument and identification of bioaerosols based on such fluorescence property. In the KAKENHI project from 2018, we focus on the origin of primary marine bioaerosols and relevance to INP formation and assess the distribution of bioaerosols over the ocean, in order to find the key factor that connects marine ecosystem, atmosphere, and cloud.

In this presentation, we will introduce the project and discuss preliminary results of cruise observation on R/V Mirai in the Central Pacific in March 2019, including number concentrations of biological particles in the seawater and air samples and organic gel particles from marine biota (e.g., Transparent exopolymer particles (TEP) and Coomassie stainable particles (CSP)).

# 中部太平洋域における生物起源バイオエアロゾルの数濃度測定と 蛍光パターン分類

川名 華織<sup>1</sup>、竹谷 文一<sup>1</sup>、松本 和彦<sup>1</sup>、宮川 拓真<sup>1</sup>、伊藤 彰記<sup>1</sup>、當房 豊<sup>2</sup>、金谷 有剛

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<sup>1</sup>海洋研究開発機構

<sup>2</sup>国立極地研究所

生物活動に由来するバイオエアロゾルは森林・土壌・海洋環境中に多く存在する。海洋環境においては、バイオエアロゾルは海洋表層から巻き上がりによって大気中へと輸送され、雲凝結核(CCN)や氷晶形成粒子(INP)として作用し、雲過程を通じた気候影響に寄与することが示唆されている。特に INP 形成において、バイオエアロゾルは他の IN 活性粒子(鉱物粒子、ダスト)と比べて高温条件下で IN 活性をもつことから、高緯度域での氷雲中のみならず、中緯度での混合層雲や低層雲中の INP 形成の起源として重要だと考えられる。しかしながら陸域に比べて海域での観測は稀少であり、海洋中のバイオエアロゾルの挙動や海洋—大気間での相互作用、雲形成への寄与等の学術的知見は乏しい。

近年、バイオエアロゾルの計測法として、複数の異なる波長の紫外光を照射した時の発光・励起に由来する自家蛍光を検出する方法が開発された。海洋研究開発機構・地球表層システム研究センターではこれまでに蛍光性粒子の高度計測や蛍光特性によるバイオエアロゾルの識別法の開発を推進してきた。2018 年度に開始した科研費プロジェクトでは、特に海洋バイオエアロゾルの起源及び INP との関連性に焦点を当て、自家蛍光・DNA 染色法の 2 つの異なる手法で蛍光性粒子を評価し、海洋生態系～大気～雲とのつながりを持つ指標を見出すことを目指している。

発表では本プロジェクトの紹介とともに、2019 年 3 月に中部太平洋域で実施された蛍光性粒子の連続観測結果について、現場で採取された表層海水試料・大気試料中の微生物数や海水中の生物起源有機態粒子(多糖類(TEP)、タンパク質 (CSP))との関連性を交えて、議論する。



## Substantially low ozone mixing ratios over the Pacific Ocean and implications from iodine chemistry

Yugo Kanaya<sup>1</sup>, Hisahiro Takashima<sup>1,2</sup>, Saki Kato<sup>2</sup>, Kazuyuki Miyazaki<sup>3,1</sup>,  
Fumikazu Taketani<sup>1</sup>, Takuma Miyakawa<sup>1</sup>, Takashi Sekiya<sup>1</sup>, Kengo Sudo<sup>4,1</sup>

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### Abstract

Constraints from ozone (O<sub>3</sub>) observations over oceans are needed in addition to those from terrestrial regions to fully understand global tropospheric chemistry and its impact on the climate. On R/V Mirai, we obtained hourly atmospheric concentrations of ozone (N=11,666) during 24 research cruise legs in the Southern, Indian, Pacific, and Arctic Ocean, covering from 67°S to 75°N during 2012-2017, and critically evaluated the state-of-the-art Tropospheric Chemistry Reanalysis version 2 (TCR-2), produced by assimilating a full suite of satellite observations to a chemical transport model. The TCR-2 systematically overestimated over the Western Tropical Pacific (125–165°E, 10°S–25°N), where substantially low ozone levels (<10 ppbv) were frequently observed, implying loss processes unaccounted in the model. We found that an additional loss rate of 0.25 ppb h<sup>-1</sup> in a particular grid (165–180°E and 0–15°N) best explained the gap, by analyzing residence time of observed air masses. Our global measurements of iodine monoxide (IO) by MAX-DOAS simultaneously made on board showed coincident maxima (~0.5 pptv) in this region, implying a hotspot of halogen chemistry unaccounted in the model. We discuss origins of reactive iodine species including marine ecosystems.

## **Session II : Effects of atmospheric deposition of aerosols on marine biogeochemistry**

### **Introduction: Martin's Fe hypothesis**

Akinori Ito (JAMSETC)

#### **Terms of Reference:**

- Review and synthesize the current scientific information on the effects of atmospheric deposition of aerosols on marine biogeochemistry, initiated by the Martin's Fe hypothesis.
- Consider the likely changes in atmospheric deposition fluxes into the future and the potential marine biogeochemical consequences of such changes.
- Identify the key future research needs to reduce uncertainties in predictive capability in this area.
- Publish the results of this activity in the open peer-reviewed scientific literature.
- Interact with, and provide information to, leading relevant international groups including the GESAMP, FeMIP, and Fe workshop.

#### **References**

- Ito, A., S. Myriokefalitakis, M. Kanakidou, N. M. Mahowald, R. A. Scanza, D. S. Hamilton, A. R. Baker, T. Jickells, M. Sarin, S. Bikkina, Y. Gao, R. U. Shelley, C. S. Buck, W. M. Landing, A. R. Bowie, M. M. G. Perron, C. Guieu, N. Meskhidze, M. S. Johnson, Y. Feng, J. F. Kok, A. Nenes, R. A. Duce, Pyrogenic iron: The missing link to high iron solubility in aerosols, *Science Advances*, 5, eaau7671, doi: 10.1126/sciadv.aau7671 (2019).
- Martin, J. H. (1990), Glacial-interglacial CO<sub>2</sub> change: The iron hypothesis, *Paleoceanography*, 5(1), 1–13, doi:10.1029/PA005i001p00001.
- Meskhidze, N., Völker, C., Al-Abadleh, H. A., Barbeau, K., Bressac, M., Buck, C., Bundy, R. M., Croot, P., Feng, Y., Ito, A., Johansen, A. M., Landing, W. M., Mao, J., Myriokefalitakis, S., Ohnemus, D., Pasquier, B., and Ye, Y. (2019), Perspective on identifying and characterizing the processes controlling iron speciation and residence time at the atmosphere-ocean interface, *Marine Chemistry*, 217, 103704, <https://doi.org/10.1016/j.marchem.2019.103704>.
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# Some Recent Results on Asian Dust at Desert and Coastal Areas of China

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**Abstract:** This presentation is a brief summary of our recent results relevant to the potential influences of Asian dust particles on marine ecosystem. The major results include (1) dust-sea salt interaction, (2) soil-derived sulfate, (3) back-ground like nitrate in desert air, (4) limited nitrate and sulfate production on dust particles in the continental air, (5) phosphorous in dust-influenced aerosol samples at a Chinese coastal city, and (6) iron in fog aerosols at the Chinese coastal city. Details of these results, except (6), were published in papers listed in the references. At the end of the presentation, I will introduce our recent research activities at a Chinese desert area and the major purposes and attempts of those activities.

## References (in the order of results number)

- (1): Zhang, D., Iwasaka, Y. and Shi, G. 2005. Sea salt shifts the range sizes of Asian dust. *EOS Trans. AGU* 86, 523.  
Zhang, D. 2008. Effect of sea salt on dust settling to the ocean. *Tellus* 68B, 641-646, doi:10.1111/j.1600-0889.2008.00358.x.
- (2): Feng Wu, et al. 2012: Soil-derived sulfate in atmospheric dust particles at Taklimakan desert. *Geophys. Res. Lett.* 39, L24803, doi:10.1029/2012GL054406.
- (3): Feng Wu, et al. 2014: Background-like nitrate in desert air. *Atmos. Environ.* 84, 39-43, doi: 10.1016/j.atmosenv.2013.11.043.
- (4): Feng Wu, et al. 2017: Limited production of sulfate and nitrate on front-associated dust storm particles moving from desert to distant populated areas in northwestern China. *Atmos. Chem. Phys.* 17, 14473-14484, doi:10.5194/acp-17-14473-2017.
- (5): Jinhui Shi, et al. 2019: Phosphorus solubility in aerosol particles related to particle sources and atmospheric acidification in Asian continental outflow. *Atmos. Chem. Phys.*, 19, 847–860, doi: 10.5194/acp-19-847-2019.
- (6): Jinhui Shi, et al. 2019: High production of soluble iron by aerosol acidification in fog. in submission to *GRL*.

## **Atmospheric nutrients: sources, atmospheric processing and impacts on ocean ecosystems**

**Zongbo Shi<sup>1</sup>, Clarissa Baldo<sup>1</sup>, Chao Zhang<sup>2</sup>, Huiwang Gao<sup>2</sup>, Akinori Ito<sup>3</sup>**

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Millions of tons of aerosol particles are transported to remote oceans each year. Aerosol particles are emitted to the atmosphere from both natural (e.g., such as desert and forest fires) and anthropogenic sources (e.g., such as shipping emissions, coal combustion, and wood burning). Once deposited, these particles provide the ecosystems with essential nutrients, such as iron, phosphorus, and nitrogen. This, in turn, stimulates primary production (a plant's ability to produce complex organic compounds from water, carbon dioxide, and simple nutrients) and enhances carbon uptake, and thus indirectly affects the climate. To simulate the impact of the nutrient deposition on the ecosystems, it is necessary to accurately estimate the flux of soluble nutrients (e.g., iron).

Soluble nutrients can be directly emitted to the atmosphere. Aerosol particles contain more or less nutrients with different speciation and mineralogy, depending on the sources. This speciation controls the solubility of nutrients such as iron and phosphorus, which ranges from below 0.1% to over 70%. This talk will review the recent advances in the emission, speciation, mineralogy, and solubility of nutrients in aerosol particles with different origins, and highlight the major uncertainties that hinder our ability to accurately simulate the emission of soluble nutrients to the atmosphere.

Soluble nutrients may also be produced through atmospheric processing. There has been much debate on the relative importance of the chemical processing to the production of soluble iron and phosphorus, thus the enhancement in iron and phosphorus solubility. We will review the recent advances in the observations on the acid dissolution of insoluble iron and phosphorus in particles, as well as laboratory measurements of the dissolution kinetics. Some recent modelling studies will also be presented to show the relative importance of the atmospheric processes to the soluble iron and phosphorus deposition flux to the oceans.

The extent of the impact of the nutrient deposition on the ecosystems depends on the properties of the aerosols (e.g., the ratio of different nutrients and the presence of toxic metals such as copper), the quantity deposited, and the ocean water conditions (such as nutrient limitation status). We will examine some of the recent observations and modelling studies on the impact of the nutrient deposition on marine ecosystems, and discuss the remaining challenges in simulating such impact.

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# **Fate of iron after deposited to the ocean--from a modelling perspective**

Ying Ye

Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research

## Abstract

Impact of iron, brought by atmospheric deposition into ocean, on marine iron cycle and productivity is on the one hand controlled by physical and chemical properties of iron-containing particles (e.g. particle size, iron content and solubility), and on the other hand, strongly influenced by the fate of this iron after entering seawater. Physical and chemical properties of iron in seawater (e.g. solubility, organic complexation and particle reactivity) determine how much iron remains in the dissolved phase. And biological cycling of iron and physical transport with water masses regulate its vertical and horizontal distribution.

In this talk, I would like to give an overview of our understanding of marine iron cycle and how this is reflected in the development of models of marine iron cycle. Some examples from recent work done at Alfred Wegener Institute will be used to illustrate the implementation of knowledge gained from measurements into models.

## **The current status of iron studies in the western North Pacific Ocean**

Jun Nishioka (Hokkaido University)

### **Abstract**

Although the subarctic North Pacific is high nutrient low chlorophyll region, where phytoplankton growth is limited by iron (Fe) availability, the area has high biological productivity. The overall picture of the processes for supplying Fe and macro-nutrients, however, are still not fully understood. In this study, we compiled observed data of chemical water properties including dissolved Fe (dFe) and nutrients, with physical parameter of vertical mixing, in the North Pacific including the sub-polar marginal seas and areas around the Kuril and the Aleutian island chains. Estimated nutrient upward flux from the data showed that the Kuril and the Aleutian ICs are the hot spots to return nutrients from the intermediate water to the surface water. These results highlighted an importance of sub-polar marginal seas and intermediate water formation for circulating micro- and macro-nutrients and controlling biological production in the subarctic Pacific. Whereas, to grasp overall picture of Fe supply to the western North Pacific, the information from oceanic side should be compiled with the knowledge of atmospheric Fe input, quantitatively. This is the next challenge under cooperation between SOLAS and GEOTRACES community.