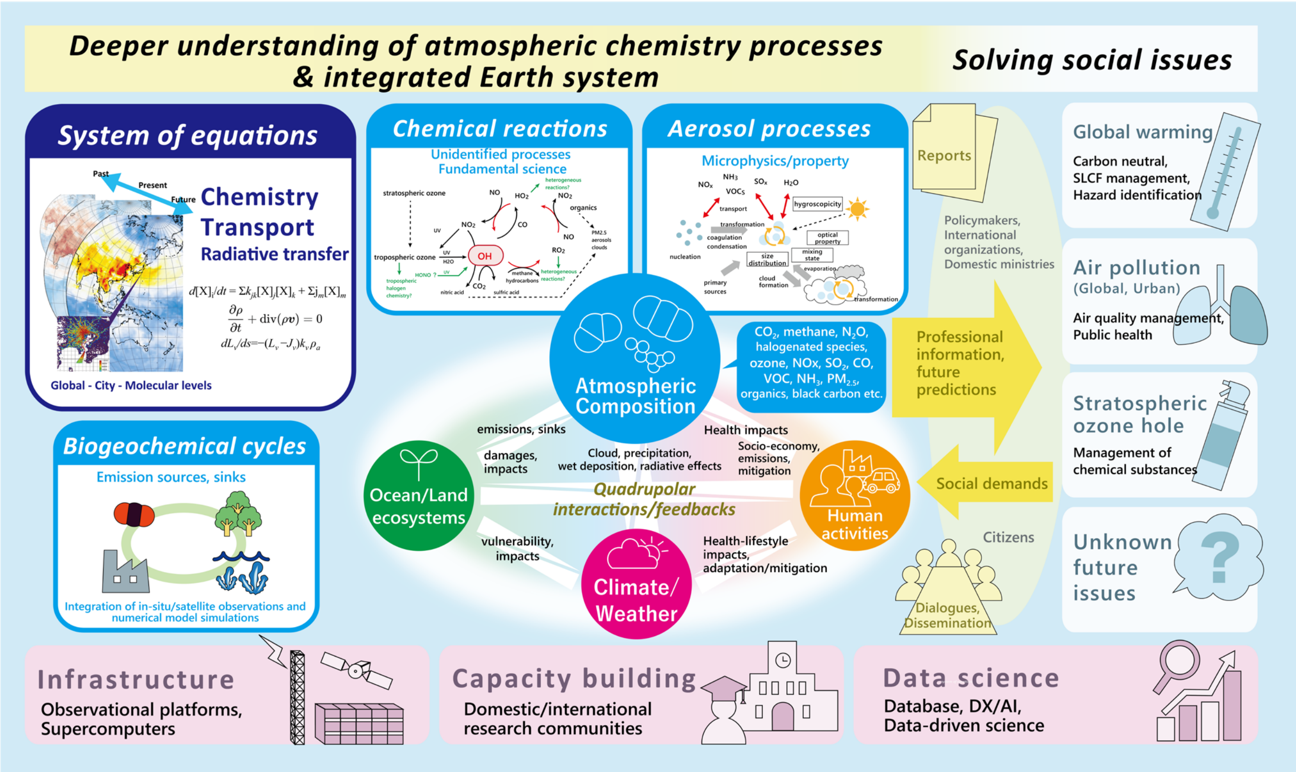
Future Vision for Atmospheric Chemistry 2022-32

- *Perspectives from Japan* -



IGAC subcommittee, Science Council of Japan

Japan National Committee of IGAC

Japan Society of Atmospheric Chemistry

Lead Authors

Yugo Kanaya (JAMSTEC)
Hiroshi Tanimoto (NIES)
Nobuyuki Takegawa (Tokyo Metropolitan University)
Michihiro Mochida (Nagoya University)

Contributing authors (from IGAC subcommittee)

Masayuki Takigawa (JAMSTEC)
Mitsuo Uematsu (The University of Tokyo)
Jing Zhang (Toyama University)
Satoshi Inomata (NIES)
Hitoshi Irie (Chiba University)
Nawo Eguchi (Kyushu University)
Yasuko Kasai (Tokyo Institute of Technology)
Jun'ichi Kurokawa (ACAP)
Naoko Saitoh (Chiba University)
Yasuhiro Sadanaga (Osaka Metropolitan University)
Kengo Sudo (Nagoya University)
Tsuyoshi Thomas Sekiyama (Meteorological Research Institute)
Sakae Toyoda (Tokyo Institute of Technology)
Tatsuya Nagashima (NIES)
Tomoki Nakayama (Nagasaki University)
Keiichiro Hara (Fukuoka University)
Jun Hirokawa (Hokkaido University)
Toshinobu Machida (NIES)
Atsuhi Matsuki (Kanazawa University)
Yuzo Miyazaki (Hokkaido University)
Shinji Morimoto (Tohoku University)
Seiichiro Yonemura (Prefectural University of Hiroshima)

Authors contributing to topical sections

Taku Umezawa (NIES)
Shinichi Enami (Tsukuba University)
Akinori Ito (JAMSTEC)
Hitoshi Matsui (Nagoya University)
Sho Ohata (Nagoya University)
Akira Mizuno (Nagoya University)
Yosuke Niwa (NIES)
Takuya Saito (NIES)
Prabir Patra (JAMSTEC)
Yosuke Sakamoto (Kyoto University/NIES)
Takashi Sekiya (JAMSTEC)
Tamaki Fujinawa (NIES)
Makoto Deushi (Meteorological Research Institute)
Tomohiro O. Sato (NICT)
Shinnosuke Ishizuka (Nagoya University)
Tetsuya Hama (The University of Tokyo)
Akihiko Ito (JAMSTEC)
Kazuhide Matsuda (Tokyo University of Agriculture and Technology)
Akira Tani (University of Shizuoka)
Fumikazu Taketani (JAMSTEC)
Yoko Iwamoto (Hiroshima University)
Jun Nishioka (Hokkaido University)
Yutaka Tobo (NIPR)
Syuichi Itahashi (CRIEPI/Kyushu University)
Naga Oshima (Meteorological Research Institute)
Kentaro Suzuki (The University of Tokyo)
Yousuke Sato (Hokkaido University)
Yosuke Yamashita (NIES)
Hideharu Akiyoshi (NIES)
Tomoo Nagahama (Nagoya University)
Tetsu Sakai (Meteorological Research Institute)
Yoshihiro Tomikawa (NIPR)
Hideaki Nakajima (NIES)
Takafumi Sugita (NIES)
Takatoshi Sakazaki (Kyoto University)

Contents

*Full version of "The Future of Atmospheric Chemistry 2022-32," including topical sections (in Japanese), is available at https://jpsac.org/sp1/

Executive Summary

1 Background

Global warming, which has been recognized as a future concern, has now morphed into a "climate crisis" that threatens our way of life. The root cause is unquestionably human activities and the increase in emissions of long-lived greenhouse gases (GHGs) such as CO_2 and short-lived climate forcers (SLCFs) including methane (IPCC 6th Assessment Report, 2021). In addition, health risks from atmospheric $PM_{2.5}$ and ozone are still an important problem that shortens the life expectancy of 4 million people per year worldwide (WHO Fact Sheets, 2022), and improvement is strongly desired. The tasks that the global atmospheric chemistry research community undertake have enlarged, from mere scientific clarification of the atmospheric processes of these trace constituents, to rationalizing "mitigation measures" such as emission reductions and their pathways, assessing uptake and buffering effects of the natural system, and to bringing about "essential solutions" to both climate and health problems, in pursuit of carbon neutral and wellbeing world. In order to fulfill the expected social role with the time-limited goals of carbon neutrality by 2050 and achievement of the SDGs by 2030, and to deepen the academic field at the same time, the basic stance that our research field should have and the research and development that we should promote in the future are clarified in this "Future Vision of Atmospheric Chemistry 2022-32" document. This follows up the previous two publications from the Japan national committee: "Atmospheric Chemistry Research in Japan 1989-1999: A Review of the Decade and Future Research Strategies" in 2000 and "Recommendations of the IGAC Subcommittee on Atmospheric Chemistry Research in Japan" in 2008. This time we choose seven themes and describes important R&D directions and issues. This Future Vision also summarizes the role to be played by our field in interdisciplinary, international, and domestic collaborations, as well as the necessary facilities, human resource development, and other issues. In particular, we will discuss how to best utilize the resources from Japan in the International Global Atmospheric Chemistry Program (IGAC) and its parent organizations, the International Commission on Atmospheric Chemistry and Global Pollution (iCACGP) and Future Earth (FE).

2 Progress in the past decade, current challenges, ultimate goals, and identified gaps

In formulating this Future Vision, we first drew up the "ultimate goal," identified global trends and Japan's strengths toward achieving that goal, identified important gaps in the current situation, and then proceeded to discuss what initiatives should be pursued over the next 10 years. As such, the readers can follow the discussion we made during its production and enhance his or her understanding. The review works were conducted in each of the seven sub-themes: "long-lived gases," "reactive gases," "physical chemistry," "atmosphere-land integration," "atmosphere-ocean integration," "aerosols, radiation and clouds," and "stratosphere and mesosphere" (Appendix A). Based on them, the ultimate goals were summarized into the following three points:

1) Deepening of atmospheric chemistry specific knowledge: For all atmospheric constituents, including greenhouse gases (GHGs), ozone depleting substances (ODSs), reactive gases that cause air pollution, and aerosols, changes in their concentrations and properties are to be expressed using equations and suitable parameters, enabling explanation of the mechanisms of phenomena and changes in the real atmosphere and future prediction. We are to fully understand the roles of atmospheric chemical processes (emissions, molecular reactions, transport and circulation, uptake and deposition, etc.) and the mechanisms of functions such as the nature's oxidizing capacity and purification system.

2) Interdisciplinary exploration of the Earth system: Interactions of atmospheric chemistry with land and ocean (including biosphere), coupling to climate systems (including cloud/radiation interactions) and health effects, are systematically understood.

3) Trans-disciplinary collaboration for solving social issues: Based on a deeper understanding of these chemical and the overall Earth systems, detection of changes in emissions, concentrations, and other key conditions is promptly made, to provide evidence for mitigation and adaptation measures for climate change, air pollution, etc., and to provide accurate future forecast information

to society.

In the past decade, "sophistication and integration of observations and numerical models" and "synthesis of methods for emissions estimation" have progressed worldwide, and new subjects (from mixing state/size-resolving modeling of aerosol particles to carbon neutrality) have emerged. Japanese research has been at the forefront of these new studies, best used its regional advantages to elucidate the Asian regional air pollution mechanisms, and has achieved unique research regarding the atmospheric chemistry theorem (such as OH radical reactivity evaluations). In addition, our strength has been demonstrated in elucidating sources/sinks of new important substances, such as iron oxide. At the same time, research structure has internationalized at IGAC and FE/WCRP (World Climate Research Program) and Japanese researchers have become more involved. Meanwhile, important substances (polyoxygenated organic compounds HOMs, bio-particles, ice nucleating particles etc.) have emerged whose cycles and functions need to be elucidated. In addition, the application and advancement of new methods (satellite observation, data assimilation, mass spectrometry, spectroscopic measurement, isotope analysis, etc.) have paved the ways to acquire key scientific knowledge and mechanism understanding in relation to climate/environmental issues. Furthermore, there is a need for strengthened collaboration that transcends the conventional units of academic research fields, in terms of the improvement of understanding and modeling of the Earth system, including the oceans and land, and dialogue with socioeconomic fields. In the future, in addition to continuing fundamental scientific exploration, we should pursue fundamental solutions to environmental problems and respond proactively to the demands of society.

3 Research Targets and Visions in the Next 10 Years

Based on this situation, the "tasks to be implemented in the next 10 years" were organized in the seven sub-groups and compiled in a cross-sectional manner, which were summarized into the following four points:

A) Gathering of atmospheric chemistry knowledge that contributes to solving social issues such as climate stabilization: Creation of socially relevant information on emissions and health effects through introduction of integrated approach on air pollutants and greenhouse gases and the promotion of satellite applications.

B) Explore key processes and systematize knowledge to solve remaining mysteries in atmospheric chemistry: Discover unknown processes, resolve inconsistencies between theory and reality, elucidate nonlinearity and multiphase chemistry, and gain comprehensive understanding from molecules to the entire Earth.

C) Interdisciplinary collaboration to improve understanding of the Earth system: Elucidation and integrated assessment of interactions (including health effects) and feedbacks among Earth's subsystems (ecosystems, atmospheric composition, climate and weather, human activities)

D) Enhancement of change detection capability by strengthening research infrastructure and improving long-term observation systems, promotion of human resource development, development of data science and distinctive international contributions

To accomplish these tasks, we will promote the advancement of understanding of atmospheric chemical processes within the field, we will actively strengthen collaboration with neighboring fields (meteorology and climate science, oceanography, cryosphere research, etc.) and the international research community to contribute to better understanding of the Earth system, and to solve social issues. We will also promote dialogues with every stakeholder relevant to carbon neutral society development, including those from academy, education, and private sectors as well as policy makers and citizens, based on state-of-the science knowledge and data. Such new visions are to be integrated during 2022-2032.

1 Ultimate Goals and Past Developments in the Field of Atmospheric Chemistry Research

(1) Aims of Atmospheric Chemistry Research

Atmospheric chemistry is a discipline that deals with the quantity and properties of substances (trace gases and particles) contained in the Earth's atmosphere, systematically understands the natural and anthropogenic processes involved in their changes, and leads to solutions to problems in the global atmospheric environment such as global warming and air pollution. The "ultimate goals" of this research field were reorganized through the discussion on the formulation of the "Future Vision of Atmospheric Chemistry 2022-32" as follows.

1) Deepening of atmospheric chemistry specific knowledge: For all atmospheric constituents, including greenhouse gases (GHGs), ozone depleting substances (ODSs), reactive gases that cause air pollution, and aerosols, changes in their concentrations and properties are to be expressed using equations and suitable parameters, enabling explanation of the mechanisms of phenomena and changes in the real atmosphere and future prediction. We are to fully understand the roles of atmospheric chemical processes (emissions, molecular reactions, transport and circulation, uptake and deposition, etc.) and the mechanisms of functions such as the nature's oxidizing capacity and purification system.

2) Interdisciplinary exploration of the Earth system: Interactions of atmospheric chemistry with land and ocean (including biosphere), coupling to climate systems (including cloud/radiation interactions) and health effects, are systematically understood.

3) Trans-disciplinary collaboration for solving social issues: Based on a deeper understanding of these chemical and the overall Earth systems, detection of changes in emissions, concentrations, and other key conditions is promptly made, to provide evidence for mitigation and adaptation measures for climate change, air pollution, etc., and to provide accurate future forecast information to society.

(*Please refer to Appendix A for the ultimate goals for each of the seven themes on which the basic work was conducted: "long-lived gases," "reactive gases," "physical chemistry," "atmosphere-land integration," "atmosphere-ocean integration," "aerosols, radiation and clouds," and "stratosphere and mesosphere". The above three goals are aggregate from these sections.)

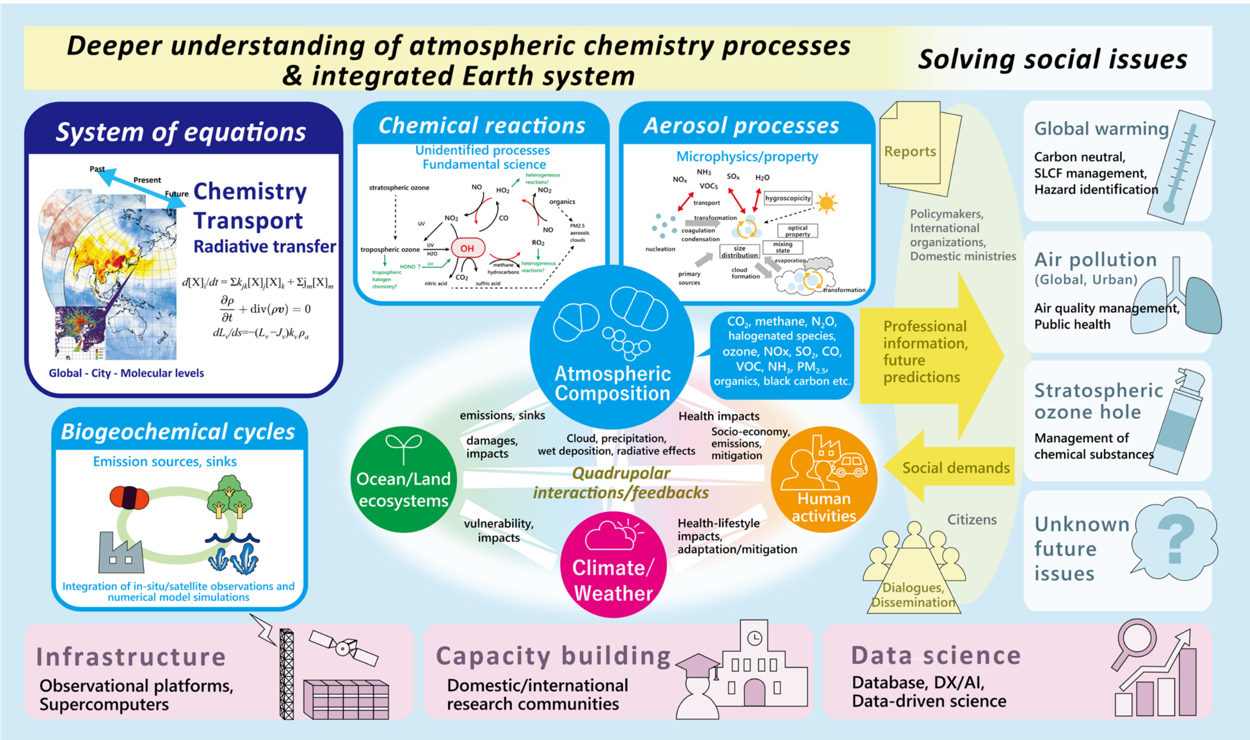


Figure 1. Conceptual diagram of the future atmospheric chemistry studies during 2022-32. (CC-BY4.0, https://jpsac.org/sp1/).

Table 1. Historical development of Atmospheric Chemistry in Japan.

	Research/Societal topics	**Approach**	**Progress, Society and Meetings**	**Publications**
1960-1979	Photochemical smog, Acid rain	Field observations, Chamber experiments	CACGP (1971) Aeronomy, Meteorology, Photochemistry, Chemical kinetics	
1980-1989	Stratospheric ozone destruction		IGAC (1988)	
1990-1999	Global warming Global atmospheric chemistry	Aircraft, Research Vessel, Stratospheric satellite observations, Campaign/long-term observations	Dawn of Atmospheric Chemistry Atmospheric Chemistry Symposium (1991) IGAC Fuji-Yoshida Conference (1994) Nobel Prize (S. Rowland, M. Molina, P. Crutzen, 1995) Atmospheric Chemistry Discussion meetings (1995-) Japan Expert Group of Atmospheric Chemistry (1999-2013)	"Atmospheric Chemistry Research in Japan 1989-1999: A Review of the Decade and Future Research Strategies"(2000)
2000-2009	Megacity, Multiphase chemistry, Climate model science	Numerical model, Tropospheric satellite observations	Nobel Prize (IPCC AR4, 2007)	"Recommendations of the IGAC Subcommittee on Atmospheric Chemistry Research in Japan" (2008)
2010-2019	Transboundary air pollution($PM_{2.5}$), radioactive species、Air quality	Observation-model integration, Systematization	Matured Atmospheric Chemistry Japan Society of Atmospheric Chemistry (2014-) iCACGP-IGAC Takamatsu Conference (2018)	
2020-2050	COVID19, SDGs, carbon neutral, climate change, adaptation, disaster prevention	Areal continuous /high-resolution satellite observations, Data assimilation	Nobel Prize (S. Manabe, 2021)	**This work (2023)**

A conceptual diagram representing the current activities to goals in the field of atmospheric chemistry research is shown in Fig. 1.

The spatial range is from "local" near our living space and emission sources, to "regional" areas such as Asia including Japan, or "global" including the polar regions, and the altitude range is from "troposphere" including "near the surface" to "stratosphere" and "upper layers". As shown in the uppermost part of the figure, the project will treat the entire process from deepening our understanding of "atmospheric chemical processes" and "Earth's integrated system" to "solving social issues" as one link in the chain. The chemical substances derived from both human activities and nature are comprehensively treated as "atmospheric composition" to elucidate elementary processes such as biogeochemical cycles of greenhouse gases, chemical reactions of reactive gases including secondary formation of pollutants, and changes in microphysics and properties of aerosol particles. These atmospheric constituents are considered as an important "subsystem" of the global environmental system, in parallel to human activities, climate and weather, and ocean, land, and ecosystems; the behavior of the entire Earth system is to be clarified, including interactions and feedbacks among these four subsystems. In order to solve social issues, the results of the natural-science-based analysis of the "current state", such as estimation of "emissions" as the driving force of modulation of the system, are utilized to make "future projections" based on simulations, and integrated knowledge such as "causal relationships" based on these results are provided to policymakers and administrative agencies to meet "social demands" (right part of Fig. 1). To promote such R&D and solutions to social issues, strengthening research infrastructure, human resource development, and deployment of data science are fundamental elements (lower part of Fig. 1). These fundamentals are common to the development of Earth and planetary science and environmental studies, and we aim to take a leading role in these areas in collaboration with many other fields.

In formulating the "Future Vision for Atmospheric Chemistry 2022-32," while being conscious of the long-term achievement of the aforementioned "ultimate goal,", we analyzed the "global trends" of the most recent decade and the "strengths of Japan's R&D efforts" evident during that period and in the historical context. Based on this analysis, the current "critical gaps" and further challenges were identified, and the discussion proceeded to what efforts should be promoted in the next 10 years. As in the case of the aforementioned ultimate goal, this work was also carried out for each of the seven themes (Appendix A), and then summarized here. Since the text from the seven themes exceeded 200 pages, it was published in separate volumes (Vol. 47 and 48 (linked from https://jpsac.org/sp1/) of the Archives of Atmospheric Chemistry Research (AACR), a journal of the Japan Society of Atmospheric Chemistry publishes). It should be noted that there we aimed to focus on "edgy" research topics and therefore some general topics, even if important, might have been left unmentioned.

(2) Roots and Historical Development of Atmospheric Chemistry Research

Table 1 summarizes the chronology and temporal evolution of atmospheric chemistry research. Since the 1990s, research topics have changed along with global-scale social issues such as stratospheric ozone depletion, transboundary air pollution, and global warming. In terms of research approaches, observations that were previously small-scale, have become more organized and integrated to larger scales with the development of satellite observations, etc., and are further being combined with numerical model simulations. Atmospheric chemistry, which has its origins in the fields of upper atmosphere, meteorology, photochemistry, and reaction kinetics, is now being studied by an international organization, the International Global Atmospheric Chemistry Program (IGAC, hereafter "IGAC"), as well as by the Commission on Atmospheric Chemistry and Global Pollution, CACGP (now iCACGP). Nobel prize winners have promoted the research field. In Japan, the "Japan Society of Atmospheric Chemistry (JpSAC)" and the "IGAC Japan National Committee", serving as a small subcommittee of the Joint FE/WCRP Subcommittee of the Committee on Environmental Studies and the Committee on Earth and Planetary Sciences of the Science Council of Japan" (hereinafter referred to as the "IGAC Subcommittee") have provided the pivotal initiatives, to organize IGAC international conferences in Japan, and twice-a-year conferences/symposiums, to foster discussion forum and develop the research community.

(3) Analysis of Research Trends in the Most Recent 10 Years and Further Challenges

In the course of these developments, during the analysis of "global trends" with particular attention to the most recent 10 years, we found that significant progress has been made in the sophistication and integration of observations and numerical models, as well as in the synthesis of multiple approaches in emissions estimations. In the former case, for example, an atmospheric chemistry transport model that expresses the emission distribution of each chemical species, meteorological fields, and chemical processes was used to interpret the processes of observed phenomena, and a new numerical model that resolves aerosol mixing states was constructed and verified through observations. The proportion of papers consisting only of observations or numerical models is decreasing. As for the latter, the "bottom-up method" which estimates emissions as the product of statistics of activity by socioeconomic sector (energy conversion, industry, transportation, residential, etc.) and emission factors, and the "top-down method" which estimates emissions inversely from observed atmospheric concentrations, have become more accurate individually and then synthesized. For example, the GCP (Global Carbon Project) evaluates both methods in an integrated manner, and high-level reports are published regularly. Furthermore, the last decade has been characterized by the emergence of new subjects (from social subjects such as climate crisis and carbon neutrality to the level of individual research subjects such as molecular-scale phase interfaces, aerosol state/size-specific modeling, etc.).

During the latest decade, "Strengths of Japanese research" include: new evaluation of iron oxide of industrial origin, which has attracted attention as a new substance affecting climate and ecosystems, by integrating observations and numerical models to clarify its contribution in reference to the conventionally known naturally occurring mineral dust-derived iron; evaluation of atmospheric oxidation theory through the quantification of the OH radical reactivity that governs the natural cleansing process; and the evaluation of source/sink flux changes through long-term, high-precision GHGs observations. Taking advantage of regional characteristics, we have also progressed with process studies and mechanism elucidation of $PM_{2.5}$ transboundary air pollution and have provided information on the origin contribution to the policymakers and stakeholders. Furthermore, during this period, the organization of research has also progressed at IGAC and FE/WCRP (World Climate Research Program), and Japanese researchers have become more involved.

On the other hand, as domestic and international research progresses, additional new important substances (e.g., highly oxygenated organic compounds known as HOMs, bio-particles, ice nucleating substances, etc.) have been discovered - in order to achieve the ultimate goal mentioned earlier, clarification of the circulation and functions of these substances is now required in addition to the conventional components. Besides, the application of new methods (satellite observation, data assimilation, mass spectrometry, spectroscopic measurement, isotope analysis, etc.) and the advancement of existing technologies have opened the way to obtain key scientific findings for elucidating natural scientific mechanisms and acquiring important information for solving social issues, as to be described in the next two sections. Furthermore, the need for improved understanding and modeling of the Earth system, including the oceans and land, the understanding of impacts from extraterrestrial space, and dialogue with socioeconomic sectors have emerged, requiring collaboration beyond the traditional units of individual disciplines.

2 Issues and Significance of Atmospheric Chemistry Research Today

(1) Efforts to be undertaken in the Next Decade

Looking ahead several decades to the achievement of the ultimate goal described in Chapter 1, and based on a consideration of historical developments and the progress made in the last decade, we summarized the "tasks to be implemented in the next decade" into the following four points (see more details about the tasks of the seven themes in Appendix A).

A) Gathering of atmospheric chemistry knowledge that contributes to solving social issues such as climate stabilization: Creation of socially relevant information on emissions and health effects through introduction of integrated approach on air pollutants and greenhouse gases and the promotion of satellite applications. {Chapters 6 on Long-lived gases, 3 on Reactive gases, 4 on Aerosol radiation clouds, 6 on Stratosphere and mesosphere} {the curly brackets refer to the chapter number of each topical sections (separate volume, https://jpsac.org/sp1/}.

B) Explore key processes and systematize knowledge to solve remaining mysteries in atmospheric chemistry: Discover unknown processes, resolve inconsistencies between theory and reality, elucidate nonlinearity and multiphase chemistry, and gain comprehensive understanding from molecules to the entire Earth. {Chapters 3 and 4 in Physical Chemistry, Chapter 2 in Reactive Gases}.

C) Interdisciplinary collaboration to improve understanding of the Earth system: Elucidation and integrated assessment of interactions (including health effects) and feedbacks among Earth's subsystems (ecosystems, atmospheric composition, climate and weather, human activities) {Atmosphere-land integration, atmosphere-ocean integration, aerosol radiation clouds Chapter 3, stratosphere and mesosphere Chapter 4}.

D) Enhancement of change detection capability by strengthening research infrastructure and improving long-term observation systems, promotion of human resource development, development of data science and distinctive international contributions {Chapter 2 on Aerosol Radiation Clouds, Chapter 3 on Stratosphere and Mesosphere}.

Here, A), B), and C) correspond to 3), 1), and 2) in Chapter 1, respectively, described as the ultimate goals, and D) is positioned as the strengthening of their common foundation.

In A), knowledge will be mobilized to solve social issues, with a focus on climate stabilization as a pressing issue. Sufficient scientific knowledge has to be obtained by the target year (e.g., carbon neutrality by 2050, SDGs by 2030, etc.) to disseminate the knowledge to society and promote change. Mitigation action cycles, evaluating changes of the state of the atmosphere and executing optimized actions have to be accelerated to satisfy the targets by their specific years. We have experienced scientific evaluation of societal actions such as reductions of ozone-depleting CFCs under the Montreal Protocol and sulfur limits from marine fuel oil (2020) tightened by IMO (International Maritime Organization). To respond to the societal needs of greenhouse gases evaluation, development of an advanced system to convert professional observations from "atmospheric environment observation satellites," into information that can be utilized by society without delay. In addition, it would be effective to conduct an integrated analysis of changes in GHGs and air pollutants, which have been addressed independently. The advantages of this approach include improving efficiency of evaluating energy combustion-related sources and emissions, which are common to both, and the fact that tropospheric ozone and aerosols, which are the major targets of air pollution mitigation, are short-lived climate forcing factors (SLCFs) and need to be managed in an integrated manner with GHGs to deter global warming. With not only climate stabilization but also air quality and health conservation in mind, we will strengthen our analysis and disseminate scientific information on "emission changes" and "climate and health impacts," which are emphasized as indicators of society's measures, along with other variables describing gas concentrations and aerosol properties.

B) explores the key processes that solve the remaining mysteries in atmospheric chemistry and establishes theories of the mechanisms that explain various phenomena. The discussion in the topical section included the importance of nitrous acid and heterogeneous reactions that is far from full understanding in the OH radical reaction system controlling the oxidative capacity of the atmosphere, and the formation pathway and yield of secondary-produced aerosols, and atmospheric chemical reactions in the polar regions. Studies will deepen the understanding of elementary processes, improve the understanding of "nonlinearity" such as ozone-producing chemical processes and "multiphase chemistry" including interfaces and heterogeneous reactions, and overcome the traditional barriers in linking local (micro) to global (macro) behavior in the real atmosphere. A combined process of physics and chemistry with challenges include quantification of the stratospheric temperature structure and atmospheric circulation changes at mid- and high latitudes due to the increase in greenhouse gases and HFCs.

C) is to deepen collaboration between atmospheric chemistry and neighboring fields to enhance an integrated understanding of the Earth system. As explained in Fig. 1, atmospheric composition is considered as one subsystem of the Earth system, and the mechanisms and feedbacks of the interaction of the four subsystems (ecosystem, weather/climate system, and human activities, and atmospheric composition), for example, changes of atmospheric composition caused by human activities affect climate; biogeochemistry influenced from climate change induces feedback on the ecosystems, are to be addressed. More specifically, this applies to the studies clarifying the effects of bioaerosols and secondary organic aerosols from terrestrial vegetation on climate/weather processes; changes in the supply of aerosols and precursors from the ocean to the atmosphere caused by global warming, ocean acidification, and changes in nutrient deposition from the atmosphere, affecting climate feedbacks via clouds; aerosol-cloud interactions diagnosed by simultaneous satellite observations of clouds, precipitation, and aerosols; and dynamic changes in Arctic wetlands, permafrost, forest fires, and ocean circulation and Earth system changes mediated by atmospheric chemistry.

With the target D), we aim to detect even smaller signals of change than before by continuing unique and long-term observations, strengthen the research infrastructure necessary to accomplish A) through C), and to foster the next generation of human resources while introducing new perspectives such as data science.

How to address the above four issues is also expressed in the conceptual diagram already shown (Fig. 1). To achieve these tasks, we will promote the advancement of understanding of atmospheric chemical processes within the field, we will actively strengthen collaboration with neighboring fields (meteorology and climate science, oceanography, cryosphere research, etc.) and the international research community to contribute to better understanding of the Earth system, and to solve social issues. We will also promote dialogues with every stakeholder relevant to carbon neutral society development, including those from academy, education, and private sectors as well as policy makers and citizens, based on state-of-the science knowledge and data. Such new visions are to be integrated during 2022-2032.

(2) Social Significance of the Atmospheric Chemistry Research

In August 2021, the Sixth Assessment Report (WG1) of the Intergovernmental Panel on Climate Change (IPCC) was released, describing that it is unequivocal that the root cause of global warming is human activity, which is the result of increased emissions of CO_2, methane, and short-lived climate forcers (SLCFs), and their concentrations (including ozone and aerosols) in the atmosphere, changing the global radiative budget (Fig. 2). It was also estimated that only about 0.4°C of temperature increase is left to achieve the Paris Agreement target of "1.5°C above pre-industrial levels," and that the amount of

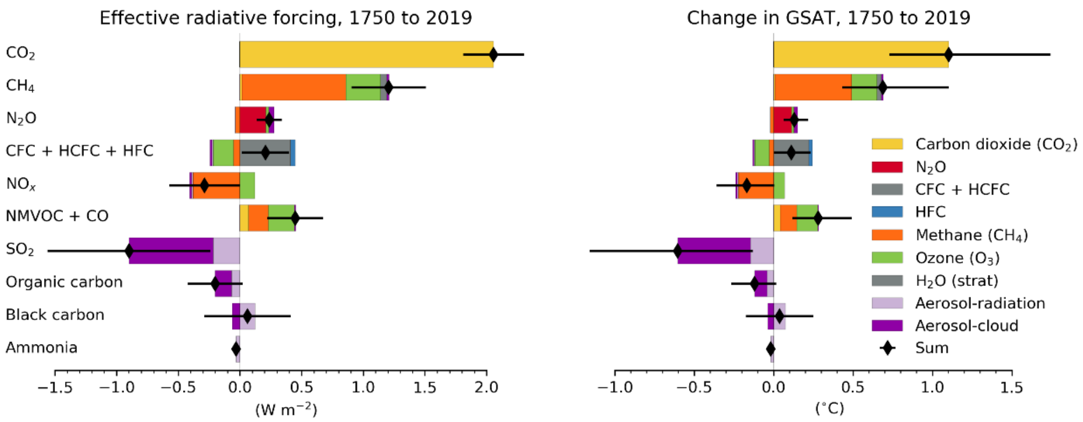


Figure 2. Contribution to effective radiative forcing (ERF) (a) and global mean surface air temperature (GSAT) change (b) from component emissions between 1750 to 2019 based on CMIP6 models (Thornhill et al., 2021; IPCC AR6 WG1 Chapter 6, Figure 6.12).

Table 2. WHO air quality guidelines and Japanese environmental standard values for $PM_{2.5}$ and surface ozone concentrations.

Air pollutants	Averaging periods	Air Quality Guidelines*		Japanese environmental standards
		WHO (2021)	WHO (2005)	
$PM_{2.5}$ (μg/m^3)	Annual	5	10	15
	24 hours	15	25	35
Ozone (μg/m^3)	Peak season	60 [ca. 30 ppb]	N/A	60 ppb, 1 hour
	8 hours	100 [ca. 50 ppb]	100 [ca. 50 ppb]	

* https://iris.who.int/handle/10665/345329

CO_2 emissions that can be allowed in the future (remaining carbon budget) is equivalent to only about 12 years of emissions in recent years. In this way, the issue of global warming, which has been recognized as a concern for the distant future since the end of the last century, has now changed into a "climate crisis" that requires urgent action. In 2020, Japan set a goal of carbon neutrality by 2050, and emission reductions are being sought in all socio-economic sectors. In estimating the remaining carbon budget, it is necessary to evaluate the contribution of temperature increase due to future changes in SLCFs. Therefore, in addition to CO_2, emissions of SLCFs, substances closely related to air pollution, have to be well managed together and their climate impact must be precisely understood.

Changes in atmospheric composition are directly linked not only to climate risks but also to air quality and health risks; health risks from $PM_{2.5}$ and ozone reduce life expectancy by 4 million per year worldwide (WHO Fact Sheets, 2022), and improvements are strongly desired. The recent relationship between atmospheric concentrations and ambient air quality standards in Japan shows that the attainment rate for $PM_{2.5}$ (daily average 35 μg m^{-3}, annual average 15 μg m^{-3}) is improving, but that for photochemical oxidants (ozone, 60 ppb at 1-hour value) is still almost zero. The World Health Organization (WHO) has rather tightened the guideline values for 2021 from the viewpoint of epidemiology, to 5 μg m^{-3} for $PM_{2.5}$ and about 30 ppb for ozone in the peak season (Table 2); therefore, significant improvements are needed. Furthermore, the biogeochemical cycles of reactive nitrogen (e.g., nitric acid) and phosphorus, which are important for marine and terrestrial ecosystems, are said to have crossed "planetary boundaries" and need to be better managed, including the atmosphere as a transport medium.

Under these circumstances, the research field of atmospheric chemistry, which deals with the science of CO_2 and air pollutants such as methane, $PM_{2.5}$, ozone, and nitrogen oxides, has an increasing societal need to respond to the demand for problem solving in collaboration with adjacent fields and society. Moreover, a new mission has emerged, such as providing evidence based on expertise.

In order to respond to social demands for problem solving, it is useful to promote natural science understanding and provide evidence to social science and policy makers for action, keeping in mind the concept of DPSIR, which has originated from Europe as a causal framework for environmental problems (Fig. 3). DPSIR stands for Driver (root cause), Pressure (load), State (condition), Impact (effect), and Response (countermeasure or response). When applied to air quality issues, P stands for "emissions", S for "change in air component concentrations", and I for "impact on climate and health. Scientific clarification of the causal relationship between them is essential, as to be shown in Chapter 2 (3). Then, the acquired knowledge should be smoothly and without delay deployed to policy makers, international organizations, and domestic ministries in an easy-to-understand manner to promote transformation of D (global social economy) through R (domestic and international measures and responses). We disseminate expertise in the field to society, for example, by pinpointing regions and sectors to target with emission reduction measures, evaluating the costs and benefits of measures in collaboration with economic and social engineering fields, providing knowledge on the current state of the atmosphere and emissions, future projections, and global integrated system change, and supporting decision making. Creating awareness of social transformation through dialogue with citizens is also important. The measurement of atmospheric composition by satellite is expected to make significant progress in terms of horizontal resolution, making it possible to track changes in GHG emissions by country, city, and even by individual plant location, which has been difficult in the past. The visualization of emission reduction efforts from time to time will be of great significance to the private sector, which is promoting information disclosure on its response to climate change issues,

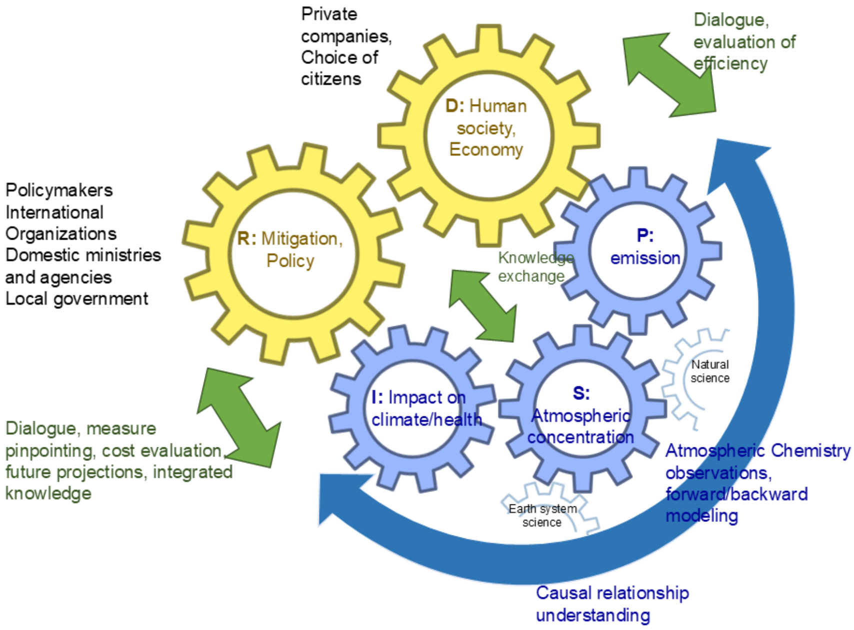


Figure 3. Schematic diagram of the linkage with society from the field of atmospheric chemistry (based on expertise in atmospheric chemistry, the causal relationship between socioeconomics - emission loads - atmospheric constituent concentration state - climate and health effects will be clarified (blue), so that policies based on scientific evidence (yellow) will appropriately lead to social transformation and mitigation of emission loads. In this way, a problem-solving cycle will be implemented based on the concept of DPSIR (see text). In order to solve problems in the required timescale, the assessment of emission and concentration changes will be expedited, and the dialogue with policy makers, international organizations, and national ministries and agencies will be extended (green) regarding the results of current analysis and future forecast information. Demonstrate the success or failure of emission reduction policies through further observations. To accelerate social transformation, we will provide information for private companies and citizens to make choices. The gears symbolize cause-and-effect relationships and the reliable transfer of information.

driving social change, and risk management through ESG (Environmental, Social, and Governance) activities and TCFD (Task Force on Climate-related Financial Disclosure) reporting. This dimension of activity can also generate new services such as consulting. In this context, it is also important to increase the number of human resources who can bridge the gap between expertise in atmospheric chemistry and socioeconomic activities.

(3) Scientific Values of Atmospheric Chemistry Research: Academic Research Direction

Regarding the scientific value and academic direction of research to be undertaken over the next 10 years, it is clearly important to pursue and understand individual atmospheric chemical processes (emissions, molecular reactions, atmospheric transport, global circulation, natural uptake and deposition, etc.) in depth, based on researchers' sense of curiosity. This section will focus on two overarching issues: "transition from the underdetermined system to the overdetermined system" and "establishing a hierarchical understanding of the system".

In tackling various problems in atmospheric chemistry, we often face the "underdetermined problem," in which the number of unknowns exceeds the number of equations because the amount of observation is insufficient compared to the number of variables to be accurately determined. For example, even if the atmospheric concentration of a substance can be accurately measured at a given geographical location, that alone does not quantitatively uniquely determine the multiple processes that cause changes in atmospheric concentrations, such as emissions and chemical production, loss, and inflow fluxes. In reality, the atmosphere is an open system in which boundary conditions such as emissions change with time, and the concentrations and properties of many different substance groups change over four dimensions (in space and time). The situation is further complicated by the fact that only limited aspects of the true state can be observed, from which the key processes (sometimes unknown processes) must be quantitatively evaluated. It is like an attempt to infer an underlying mechanism or theory from a jigsaw puzzle with missing pieces. However, atmospheric chemistry transport models, which represent meteorological fields, dynamics, and chemical processes, have made it possible to link the limited information from observations, and thus a joint analysis based on both observations and models is now being enabled in smarter ways. In the future, it will be important to move out from this under-determined state as aided by the increasing amount of observational information combined with intelligent methods such as data assimilation techniques.

In recent years, it has become possible to measure the geographical distribution of concentrations of multiple atmospheric constituents from satellites in a spatially continuous manner and with high horizontal resolution {Reactive Gases Chapter 2, see thematic volume for more details}. Geostationary satellites of the atmospheric environment have also emerged, making it possible to track temporal variations during the day. It is significant to analyze multiple substances in combination in order to unravel common processes, as there may be common emission sources, common transport, relationships associated with chemical reactions, represented as "covariance relationships" among multiple substances. Thus, one of the keys to convert the underdetermined problem to an overdetermined system will be to improve the quality and quantity of information that can be extracted by using dense temporal and spatial observations of multiple components and numerical models that are validated by such observations.

For example, the Japanese GOSAT-GW satellite with TANSO-3 sensor, scheduled for launch in fiscal year 2024, will simultaneously measure CO_2 and methane, which are important GHGs, as well as NO_2, a marker of energy combustion emission sources at high temperatures, such as cities and power plants. On the other hand, some small-scaled events are beyond the reach of satellite observations; therefore, it is important to expand the Earth observation network (ground, aircraft, and satellite) in a complementary manner. Data assimilation methods that combine information between components and in time and space, and the basic improvement of atmospheric chemistry transport models that form the basis for such methods, will be effective for analyzing such vast amounts of observational information. This integrated analysis will also require strategies and ideas for accurately quantifying uncertainties in several important factors and adequately linking them to the analysis of changes in atmospheric compositional state.

In addition to the use of such multi-component measurement information with high spatial and temporal resolution, advanced use of isotopic information and simultaneous measurement of concentrations and fluxes (reactivity) can also facilitate the transition to an overdetermined system. Such efforts should be promoted as well in the future. Another important goal of our research field is to clarify "properties of substances" and their changes, such as optical properties of aerosols, properties as cloud condensation nuclei CCN, etc., and indicators of health effects including reactive oxygen species. Therefore, increasing the amount of information on the properties is essential. Such an approach will be important to elucidate the full life cycle of primarily-emitted species, those produced in the atmosphere such as ozone and $PM_{2.5}$, and yet unidentified important substances, being affected by natural and anthropogenic emissions, atmospheric chemical processes, transport processes, and loss processes. It will also be a useful approach to solving remaining central problems in atmospheric chemistry, such as elucidating the behavior of OH radicals, which dominate atmospheric oxidative capacity and natural cleansing processes, but whose global-scale variations are not yet captured.

The second point, "hierarchical system understanding," aims to systematically understand the climate and related materials science. This begins with the systematization of the understanding within the field of atmospheric chemistry, for our case, then propagates to the higher levels of understanding of the natural system aspects of the global surface material cycle, including land and ocean in contact with the atmosphere, and adds the human dimension with bi-directional influences. To establish the theory of atmospheric chemistry, accumulation of knowledge of elementary reaction process units and systematization of atmospheric chemical equations are important. There are many areas that require further efforts, such as multiphase chemistry including liquid and gas phases and their interfaces. There is also a need to better link the chemical composition/microphysical properties of aerosols with their radiative properties. Simultaneously, it is important to gain a systematic understanding of the ocean, atmosphere, and land as Earth subsystems, and to gain a quantitative and consistent understanding of the mass exchange fluxes and interactions among these subsystems and human activities. For example, it is important to fully reconcile the assessment of CO_2 exchange between the land and the atmosphere from the perspective of atmospheric processes with that from the perspective of terrestrial vegetation in order to reach a common understanding. Lastly, we promote understanding of the mechanisms of interactions of atmospheric chemistry not only with natural systems but also with human activities, including the interaction of climate change with emissions from socioeconomic activities and anthropogenic land use change, as well as the mechanisms of action of health effects caused by changes in atmospheric composition. With this approach, we will be able to deepen atmospheric chemistry research as a discrete field and then progressively contribute to interdisciplinary Earth system research and even to transdisciplinary collaboration to solve societal problems. We should also strive to link the "knowledge" gained about the atmospheric chemistry system to universal knowledge that can be applied to other systems as well.

(4) Current Status and Future of Interdisciplinary Collaboration

In order to achieve the 10-year plan as described, in addition to deepening research and development within the field, there is a strong need to strengthen or start collaboration with neighboring fields, as illustrated below.

Earth System Modeling Science

In the CMIP coupled model intercomparison experiments directly linked to the IPCC report, many scenario experiments are conducted that include changes in atmospheric trace gases and aerosols in addition to the CO_2 changes. Two Japanese models, MRI-ESM2.0 from Meteorological Research Institute and MIROC-ES2L mainly from Japan Agency for Marine-Earth Science and Technology (JAMSTEC) have participated in the experiments, during CMIP6. Further participation in these Earth system model experiments and comparative evaluation of these models, with process-based atmospheric chemistry models and with observations, are desirable in the future. Among the feedbacks between land and ocean including ecosystems and atmospheric composition, for example, we promote studies on aerosol and ozone formation from the reactions of biogenic volatile organic compounds (BVOCs), evaluation of the impact on climate caused by ice nuclei formation from bioaerosol/dust particles, and global warming feedbacks of further GHGs emissions due to increased forest fires and thawing permafrost.

Economics and Social Engineering

The creation of anthropogenic emission inventories of CO_2 and SLCFs is a direct interface with the economic and civil engineering fields; highly accurate information is essential as a basis for emission reduction measures. Even if effective emission reduction measures for CO_2, SLCFs, and air pollutants are found as potential solutions on the scientific side, they will not be socially effective unless they are accompanied by economic rationality. Thus, the knowledge about the marginal abatement cost and the cost-benefit analysis becomes significant. It is also important to provide and disseminate consulting services and expertise to support ESG activities and TCFD reporting from private companies regarding GHG reduction.

The global socioeconomic changes, brought about by measures to prevent the spread of new coronavirus infections, can be clearly seen from our field's global satellite observations that measure the amount of NO_2 in the atmosphere; the changes were quantified by regions and time periods. This analysis has virtually evaluated the effects of future emission controls on air quality improvement and climate stabilization. In addition, preparing for and tracking changes in air quality that occur before and after unexpected and sudden changes in social systems, for example, nuclear accidents and volcanic eruptions, is another important interface between atmospheric chemistry and socioeconomics. The longer the basic data is maintained, the more valuable it will be to the scientific community in the field. The benefit is also used to evaluate the effectiveness of the control measures taken.

Satellite Earth Observation

Earth observation satellites have been closely linked to the field of atmospheric chemistry. In the past, the atmospheric chemistry community made only limited connection, e.g., contributions to the development of satellite sensors, but recently this activity has become even more active owing to the continuous efforts of those involved. In order to rationalize national-level investment on meaningful Earth observation satellite projects, the Task Force on Future Space Development, and its Remote Sensing Subcommittee, which is jointly managed by various academic societies, has begun discussions and coordination across mission applications. The Japan Society of Atmospheric Chemistry has joined this activity and, as an active party, is enhancing cooperation and mutual understanding by making proposals such as atmospheric environment measurements from geostationary satellites and by evaluating other missions. The Science Council of Japan's "Subcommittee on the Future Vision of Earth Observation Satellites", under the Committee on Earth and Planetary Sciences, also discusses and promotes the scientific and social significance of Earth observation satellites (see Chapters 3 and 4).

Computational Science

As represented by the supercomputers "K computer" and "Fugaku", remarkable progress has been seen in computational science, contributing to a better understanding of a wide range of phenomena from the molecular level to the global scale (see Chapter 4). As for atmospheric chemistry, there are already many studies making use of these facilities, such as theoretical calculations related to aerosols and other interfacial reactions, and the refinement of climate impact predictions for CO_2 and SLCFs. Furthermore, data analysis is now being conducted using AI and machine learning, which is expected to lead to the discovery of phenomena from weak signals, that might otherwise be hidden by internal variabilities in classical statistical analysis.

On the other hand, the number of experts specializing in computational science in atmospheric chemistry is limited. The activity has not yet reached the level of developing innovative algorithms that could lead to significant breakthroughs. As a result, many of the atmospheric chemistry process schemes used in models are basically derived from knowledge developed in other countries. In the future, it will be necessary to work closely with researchers in the mathematical sciences who specialize in computational science to develop originality from Japan's own algorithms. It is hoped that synergistic effects with the expansion of observational data will help bridge the gap between observations and models.

Health, medicine, and life sciences

There is a long history of research, mainly epidemiological studies, on factors that adversely affect health via the respiratory tract, such as ozone and $PM_{2.5}$. In recent years, studies that go further into physical and chemical processes, such as the dynamics of multiphase and heterogeneous systems including mass exchange in alveoli, oxidative stress induced by organic aerosols and peroxides generated in the atmosphere, or the transfer of insoluble solid particles into the circulatory system, have become active. This is an important interdisciplinary area between atmospheric chemistry and medical and life sciences. In addition to air-borne infectious diseases such as influenza and coronaviruses, it is also important to explore the relationship between atmospheric composition and infectious diseases such as Kawasaki disease, the cause of which is not yet fully identified. Although Europe and the United States have taken the lead in such research, it is important to bring about new developments in epidemiological research and toxicity assessment based on the sources of pollutants and climate unique to Asia and the racial characteristics of Asia. It is hoped that the potential of the atmospheric chemistry research field will be demonstrated in promoting such research.

3 International and domestic collaboration

(1) Collaboration with International Projects

In atmospheric chemistry research, it is equally important to share and collaborate with the international research community in addition to improving cutting-edge research results in order to achieve a comprehensive understanding of the global scale and to solve problems.

The International Global Atmospheric Chemistry Project (IGAC) and the International Commission on Atmospheric Chemistry and Global Pollution (iCACGP) play a major role in this international collaboration. When IGAC was launched, the executive committee, called the Scientific Steering Committee (SSC), consisted mainly of European and U.S. researchers, but now researchers from Japan, China, as well as South and Southeast Asia have joined the SSC and are active in the organization. It is important for Japan to continue to contribute internationally through its involvement in the SSC, as well as to inform the Japanese research community of global trends and developments.

IGAC and iCACGP hold regular international conferences (to be held jointly every two years after 2022) and play an important role as a place where the seeds of international collaboration are born at the grassroots and where future leaders are nurtured. In Japan, the Fujiyoshida Conference in 1994 (about 170 foreign and 90 Japanese participants from 27 countries), the Nagoya Conference in 1997 (about 90 foreign and 90 Japanese participants), and the Takamatsu Conference in 2018 (725 participants from 45 countries) were successfully held from the early period of the Japanese atmospheric chemistry community development. It was an opportunity to gain recognition and to dramatically increase international exchange and collaboration. In recent years, about 40% of the participants in international conferences have been Early Career Scientists (defined by IGAC as graduate students and researchers within 3 years of receiving their degrees), and these international conferences are becoming increasingly important for the development of the next generation of atmospheric chemists.

As of 2023, IGAC is promoting 11 Activities (Table 3) and 6 Working Groups, each of which plays a leadership and capacity-building role in international collaboration. These regular activities also play an important role as a forum for the continued promotion and development of international collaboration.

The topics covered by these Activities often have overlap with the topical theme covered by this Future Vision from Japan; in this context, in addition to the researchers currently participating from Japan, a wide range of age groups, including young researchers and students from Japan should be able to be involved in the IGAC Activities in the future to increase their direct contributions.

The Working Group, on the other hand, is a category launched by IGAC in 2010 for regions that are important for atmospheric chemistry but have weak international representation and recognition because of the underdeveloped research community in those regions. It aims to strengthen grassroots networks of researchers and promote international collaboration with researchers in other regions; currently such regions include Africa, North and Latin America, China, Japan, Monsoon Asia, and the Southern Hemisphere. The Japan National Committee (http://igacproject.org/JapanNationalCommittee), which covers Japan, simultaneously has a role of the

Table 3. IGAC's Activities.

Acronym	Activity	Japanese translation
PACES	Air Pollution in the Arctic: Climate, Environment and Societies	北極の大気汚染と気候・環境・社会
Allin-Wayra	Small sensors for atmospheric science	大気科学のための小型センサ
AMIGO	Analysis of Emissions using Observations	大気観測からの排出量解析
ACAM	Atmospheric Composition and the Asian Monsoon	大気組成とアジアモンスーン
BBURNED	Biomass Burning Uncertainty: ReactioNs, Emissions and Dynamics	バイオマス燃焼の不確実性：反応・排出・ダイナミクス
CCMI	Chemistry-Climate Model Initiative	化学気候モデリングイニシアティブ
GEIA	Global Emissions Initiative	地球規模の排出量研究イニシアティブ
MAP-AQ	Monitoring, Analysis and Prediction of Air Quality	大気質のモニタリング・解析・予測
CATCH	The Cryosphere and Atmospheric Chemistry	寒冷圏と大気化学
TOAR-II	Tropospheric Ozone Assessment Report -Phase II	対流圏オゾン評価報告書第２期
DEBITS	Deposition of Biogeochemically Important Trace Species	生物地球化学的に重要な微量成分の沈着

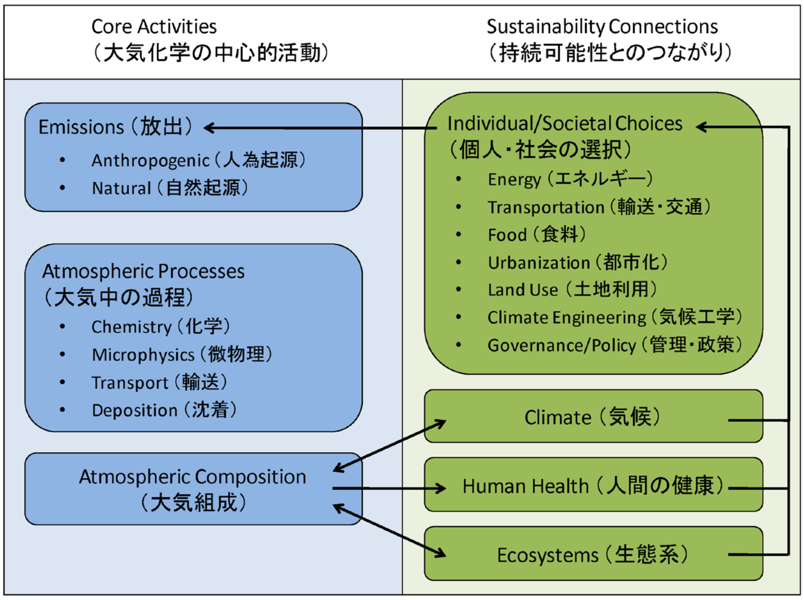


Figure 4. IGAC's vision diagram.

IGAC subcommittee of the Science Council of Japan. These double roles have resulted in increased international exposure and recognition, as well as in the domestic fields. Further mutual developmental relationships should be maintained in the future. Japan can also play a significant role in the Monsoon Asia and Oceania Networking Group (MANGO), a working group in monsoon Asia where atmospheric chemistry researchers had been tied only loosely and not well connected to the international community.

In addition to preparing for the aforementioned international conference, iCACGP's activities include planning an IAMAS session on atmospheric chemistry for the International Union of Geodesy and Geophysics (IUGG) international conference. The position of atmospheric chemistry in Earth system science should be reviewed from time to time, such that it can serve as an interface platform for broad collaboration with neighboring fields, including terrestrial and oceanic chemistry. On the other hand, Future Earth, an international research program launched by the International Geosphere-Biosphere Programme (IGBP) merged with neighboring programs, is another parent organization of IGAC (in parallel to iCACGP). Future Earth runs Global Research Networks (GRN), including IGAC, and there we can ideally play an important role in strengthening the atmospheric chemistry community's contribution to society and collaboration around sustainability. Future Earth has a Japan Hub with a Secretary General, and is also supported by institutions including National Institute for Environmental Studies and the University of Tokyo.

Of the 17 Goals of the SDGs, atmospheric chemists can play a significant role in climate change action, clean energy, etc. However, the perspective of "air quality," which affects health, is not easily seen at that Goals level, and the atmospheric chemistry community needs to work on this issue in the future. In Asia, in particular, the health hazards of air pollution and disasters coupled to climate change may become more serious, and further contributions from atmospheric chemistry research are needed. Furthermore, it is important to deepen collaboration with SOLAS (Surface Ocean-Lower Atmosphere Study), iLEAPS (Integrated Land Ecosystem-Atmosphere Processes Study) and with GCP (Global Carbon Project), which are adjacent fields under the Future Earth umbrella, to incorporate new perspectives and human resources into atmospheric chemistry as well. As described above, evolution as an Earth system science and contribution to a sustainable society are two important axes for future atmospheric chemistry research (Fig. 4).

In addition to IGAC and iCACGP, there are many other international research committees/organizations/bodies with which key collaboration should be established. The UN and its international organizations are the good examples. The World Meteorological Organization (WMO), through a mechanism called the Research Board, has established the Environmental Pollution and Atmospheric Chemistry Scientific Steering Committee (SSC-EPAC) (https://community.wmo.int/governance/commission-membership/research-board/ssc-epac), a committee closely related to atmospheric chemistry. In particular, the GAW (Global Atmosphere Watch) Programme plays an important role in atmospheric monitoring of greenhouse gases and air pollutants. GAW has seven subcommittees relevant to Ozone, UV radiation, Greenhouse Gases, Aerosols, Precipitation Chemistry, Reactive Gases, and GURME (Urban Weather and Environment). The JMA aims for a wide range of contributions, from involvement in data centers and data quality control, such as the World Data Centre for Greenhouse Gases (WDCGG), which the JMA is responsible for, to the maintenance of observing stations (even beyond the GAW stations) and the provision of observation data. There is also an Expert Team that discusses and reports on the technical aspects of research-oriented infrastructure, and initiatives that link science to service to society, such as GAFIS (Global Air Quality Forecasting and Information System), IG3IS (Integrated Global Greenhouse Gas Information System), covering air quality and health, and urban greenhouse gas emissions, respectively. The United Nations Environment Programme (UNEP) has launched initiatives such as the Climate and Clean Air Coalition (CCAC) and the International Methane Emissions Observatory (IMEO) to promote SLCFs-related research and development and building international momentum for SLCFs.

In East Asia, the East Asian Network for Acid Deposition Monitoring (EANET) started in 2001 has been playing a major role, and is expected to play a further role in addressing not only the acid rain but also ozone/aerosol pollution, and climate change issues through SLCFs; the potential anticipated from Japanese atmospheric chemistry researchers through the Japan-based Asian Center for Air Pollution Research (ACAP) is great. With regard to air pollution issues over a wider region, it is also important to contribute to the Task Force on Hemispheric Transport on Air Pollution (TF HTAP) through MICS-Asia (Model Inter-Comparison Study for Asia), for which ACAP serves as the secretariat.

The CEOS (Committee on Earth Observation Satellites) under the GEO (Group on Earth Observations) has been working with the GEOSS (Global Earth Observation System System) to develop a satellite-based Earth observation system and its specific groups include Atmospheric Composition-Virtual Constellation (AC-VC). Japan, while cooperating with South Korea, which launched a geostationary satellite for observing atmospheric pollutants in 2020, will deepen international collaboration in terms of Earth observation from space and strengthen its leadership in Asia through promotion of GOSAT-GW and newly proposed missions {see topical theme volumes relevant to Long-lived greenhouse gases and Reactive gases}.

International collaboration is also important in terms of climate change issues. In particular, at COP, the Conference of the Parties to the United Nations Framework Convention on Climate Change (UNFCCC), each country set policy targets toward 2050 carbon neutrality in order to achieve the 2°C/1.5°C target of the Paris Agreement. For this, understanding greenhouse gas emissions and their trends together with decarbonization pathways, and integration of reduction strategies of SLCFs are important, where atmospheric chemists can play an important role. In this context, involvement in the IPCC is also important. The Japanese atmospheric chemistry research community has produced a number of Author Team members (Lead Author, Review Editor, etc.) for the Fifth and Sixth Assessment Reports of the IPCC. Particularly, a separate chapter on SLCFs has been established in the Sixth Assessment Report. In the Seventh Assessment cycle, "2027 IPCC Methodology Report on Inventories for Short-lived Climate Forcers" is to be officially produced and contributions with expertise from atmospheric chemistry research community are anticipated. In addition to IPCC, the Arctic Council has also been working on SLCFs through the EGBCM (Expert Group on Black Carbon and Methane) and AMAP (Arctic Monitoring and Assessment Program). In particular, in order to reduce black carbon and methane emissions, observer countries of the Arctic Council (including Japan, China, South Korea, India) that have reported their emissions are invited to join the Expert Group on an equal footing with Arctic countries (Canada, Denmark, Finland, Iceland, Norway, Russia, Sweden, and the United States), even if they are non-Arctic countries. The provision of scientific knowledge is a key element of the Arctic Council's efforts to promote the conservation and protection of the Arctic. The related activities from Japan may enhance the country’s international position, and from this perspective, discussions with international legal scholars and policy makers are also important.

Involvement in international observational campaigns and model intercomparison experiments is also important. For example, NASA has conducted aircraft observations of atmospheric chemistry, known as the Global Tropospheric Experiment (GTE), around the world. In Asia, there were the Japan-based PEM-West A and B (1991 and 1994) and TRACE-P (2001). In 2016, NASA's KORUS-AQ aircraft observations in Asia were based in Korea, focusing on a specific area over Korea to directly observe emissions and chemical transformation from emission sources. Also in 2018, a German-led EMeRGe-Asia aircraft observation was based over

Taiwan. Going forward, NASA plans ASIA-AQ, the successor to KORUS-AQ, in 2024 to observe emission sources and chemical transformations over a larger area covering East and Southeast Asia in conjunction with satellites and numerical models. Aircraft observation is an indispensable element for verifying satellite observations and understanding integrated three-dimensional atmospheric composition changes. As Japan has yet to acquire an aircraft for research purposes, we will continue to propose and encourage the academic community and ministries to acquire an aircraft dedicated to earth observation in collaboration with other academic societies. For the time being, it is also important for Japan to synchronize its involvement in ground and shipboard observations with such opportunities, and to participate by bringing its own originally developed observation equipment and building up a track record.

In all of the above international collaboration, the challenge is how to effectively disseminate new findings and mature results from Japan, given the current situation in which the scientific community tends to be centered on Europe and the United States, and China is significantly increasing its presence in the world. Initiatives such as regional reports that international high-level reports rely on, and leadership in the Asia-Oceania region, such as the Asia Oceania Geosciences Society (AOGS), would be effective. In addition, Japanese researchers can play a role in contributing to the creation of appropriate rules by providing expert perspectives and fair and neutral evaluations to prevent the spread of ineffective initiatives and proposals that take advantage of carbon neutrality.

(2) Related Work and Committees of Domestic Ministries and Agencies

The Japan Society of Atmospheric Chemistry (JpSAC), which has its origin in the "Atmospheric Chemistry Discussion Group (Taiki Kagaku Kenkyukai)" and "Atmospheric Chemistry Discussion Meetings (Taiki Kagaku Touronkai)" started in 1999 and 1995, has contributed to the development and collaboration of atmospheric chemistry researchers in Japan. In 2006, we joined the Japan Geoscience Union (JpGU), and in 2007, the "Symposium on Atmospheric Chemistry," one of the two research meetings we had been holding, was transformed into the "Atmospheric Chemistry Session" at the JpGU regular annual (springtime) meetings. Such involvement in JpGU not only promotes atmospheric chemistry to researchers in other fields, but also serves to strengthen ties with Earth science fields related to atmospheric chemistry. In addition to JpGU, many members of JpSAC hold membership of the Geochemical Society of Japan, the Japan Society for Atmospheric Environment, the Meteorological Society of Japan, the Oceanographic Society of Japan, and others, and on some occasions they co-sponsor or support research meetings and sessions, thus promoting cooperation with surrounding fields. In the future, as a field that constitutes the Earth's surface system, it will be important to determine how we can contribute to comprehensive scientific understanding and how we can effectively disseminate scientific knowledge to society in terms of our involvement in global environmental issues. In this regard, the JpSAC's positioning, strengths, and weaknesses should be periodically analyzed, and future collaboration should be undertaken with an awareness of complementarity with neighboring societies.

We already mentioned that the JpSAC is closely related to IGAC. But the same also applies to WCRP, and SOLAS and iLEAPS of Future Earth. The scope is unique in that it is not limited to the tropospheric atmosphere, but also covers the exchange between the troposphere and stratosphere, the carbon cycle and reactive materials, and biogeochemistry and interaction with the terrestrial ecosystem and the ocean surface layer. It is resilient to the changing trends in the discipline and the pressure to restructure organizations and communities in response to societal needs for global environmental issues over the next 10 to 20 years.

Currently, the JpSAC is a cooperative academic organization of the Science Council of Japan, and it is important to be involved in diverse activities. The IGAC subcommittee links Japanese atmospheric chemistry research to the IGAC international collaboration. It should play the key role in addressing further issues proposed in the Master Plan for Large Research Programs and the Future Science Promotion Initiative, such as how to reconcile improvements in climate change mitigation with other values such as health, energy, and food. Also, our involvement in the Science Council of Japan's "Cross-cutting meetings on Carbon Neutral (Net Zero)" and the dissemination of the latest findings on the atmospheric composition changes related to climate change, not only to Division III (Science and Engineering) of the Science Council of Japan, but also to Division I (Humanities and Social Sciences) and Division II (Life Sciences) will enhance our recognition. The latest trends and issues in this field are reflected in the "Biennial Report on Research and Development in the Field of Environment and Energy" published by the Center for Research and Development Strategy, Japan Science and Technology Agency (JST), and should be used as a

Table 4. Japanese domestic and governmental committees to which Atmospheric Chemistry research community contributes.

省庁・委員会等 Ministry, committee	Discussion contents
環境省中央環境審議会・微小粒子状物質等専門委員会 (MOE, $PM_{2.5}$ and O_3 committee)	It discusses measures to reduce domestic concentrations of $PM_{2.5}$ and photochemical oxidants (ozone) and to achieve standard values from an expert perspective, taking into account the effects of transboundary air pollution and decarbonization.
経済産業省 VOC 排出削減効果の定量的評価に向けた検討等業務検討会 (MITI, VOC reduction committee)	From the viewpoint of economic and industrial promotion, the committee is considering measures to promote the reduction of VOCs as precursors of ozone, taking into account regional and industry-specific characteristics rather than a uniform domestic approach, and is discussing the scientific basis for such measures from a professional standpoint.
文部科学省・気象庁・環境省・経済産業省 IPCCAR6WG1 国内幹事会、IPCCAR6 国内連絡会 (4 ministries, IPCC committee)	Organized mainly by the author team members of IPCCAR6, it exchanges information among chapters and WGs and discusses how to expand Japan's contribution to the IPCC.
国土交通省気象庁品質評価科学活動懇談会 (JMA, GAW related observational quality assurance)	Provides expert opinions and scientific and technical advice on the quality improvement of measurements of GHGs, ozone, aerosols, radiation, etc. in accordance with the WMO/GAW Global Atmospheric Monitoring Program conducted by the Japan Meteorological Agency.
文部科学省地球観測推進部会 (MEXT, GEO related committee)	Implementation policies and plans are formulated to promote earth observation in cooperation with related ministries and agencies.

starting point for future cross-disciplinary efforts.

The followings are the examples of contributions to relevant tasks and committees of domestic ministries and other organizations that use atmospheric chemistry expertise to translate scientific findings into solutions to societal problems (Table 4). Though not included in the table, other committees that meet on a permanent or ad hoc basis include the Ministry of the Environment's Committee on Continuous Monitoring of Air Pollution and the Japan Aerospace Exploration Agency's Committee on Satellite Observation Missions.

These activities should be further expanded in the future in response to the growing social needs to address global climate change and air pollution issues. Another possibility would be to lead policy contributions and increase flexibility there. For example, the Ministry of the Environment submits the "Annual National Report on Black Carbon/Methane Emissions" to the EGBCM of the Arctic Council via the Ministry of Foreign Affairs, but the process is complicated because multiple departments are involved affected by the jurisdiction of the ministry. It is difficult to say that the ministry, which is the point of contact, is currently well connected to researchers who have accurate information and cutting-edge scientific knowledge. The IPCC has officially decided to develop a methodology report for the SLCF inventory in the 7th Assessment cycle, and the process has started as of 2023. In order for Japan to respond to these developments without delay and with scientific rationality, it is important for the research community side to start preparation and examination requesting smooth administrative support and also to lead the IPCC's new activity. In this way, there is much that can be done from the research side to strengthen cooperation among administration, practice, and research.

Atmospheric chemists can play a significant role not only at the national level, but also at the local government level, contributing to environmental impact assessment committees of prefectures and cities. In doing so, we could contribute more in the future to "decarbonization," the reduction of greenhouse gas emissions resulting from human activities. The Japanese government has set a reduction target of carbon neutrality in 2050 and a 46% reduction in greenhouse gas emissions in 2030 (compared to 2013 levels), which is also a global commitment. Despite the internationality, specific measures and verification are often localized and handled at the municipal level. Atmospheric chemistry, which deals with not only greenhouse gases but also air pollutants emitted into the atmosphere from anthropogenic or natural sources, covers not only the construction of emission inventories of these substances but also inverse analysis estimating emissions from atmospheric concentration changes. This will lead to improved emission knowledge on a real-time basis to facilitate reduction measures and contribute to simultaneous mitigation of climate change and air pollution. Such research and development are currently being conducted at the subcontinent and national level, but with future improvements in observation and modeling techniques, it will become possible to understand the current situation at the city/street scales, and the time will come when local governments can make a more direct policy contribution to decarbonization and air pollution control measures.

Despite such an important role, there are significant challenges regarding the long-term maintenance and coordination of Earth observation related to atmospheric chemistry: since the "Strategy for the Promotion of Earth Observation" document published in 2004, related to the GEO Strategic Plan, and now based on the "Implementation Policy for Earth Observation in Japan in the Next Decade" in 2015, Japanese ministries including MEXT, the Japan Meteorological Agency, and the Ministry of the Environment have established and maintained coordinated atmospheric observations. In reality, however, there are many situations in which each ministry/institution under them has been forced to terminate observations due to budget cuts, such as ozone sonde observations. It is desirable to strengthen the system by, for example, enhancing cooperation between the research community and the ministries to soften the impact of the termination of observations. It is hoped to establish the SLCFs field in the virtual Earth Observation Collaboration Center, for example, to overcome these difficulties.

4 Expansion of the platform for atmospheric chemistry research and development and challenges

(1) Platforms and Resources for Atmospheric Chemistry Research

Observations of the dynamics of atmospheric constituents at various temporal and spatial scales require multiple platforms for in-situ measurements or remote sensing, depending on the observation target. Typical platforms that have been used include satellites for long-term observations over wide areas and the entire globe, aircraft and ships for understanding the spatial distribution of a region, and ground stations suitable for long-term monitoring, observations of surface and vertical distribution conditions, and operation of advanced measurement equipment.

Satellite observation of trace gases and aerosols is a powerful method for determining their spatiotemporal distribution over a wide area, and is expected to become even more important in atmospheric chemistry. Applications include evaluation of atmospheric model simulations, source estimation by inverse calculations, and forecasting by data assimilation. Japan has successfully conducted satellite observations of CO_2 and methane with the GOSAT series (GOSAT, GOSAT-2). An observation mission with a third satellite (GOSAT-GW, to be launched in FY2024), which will add NO_2, is under preparation. The areal continuum observations with kilometer-class resolution are expected to provide direct assessment of nonlinear atmospheric chemical processes and evaluation of individual sources. In addition, observations of the ozone layer by the Improved Layer Around the Atmosphere Spectrometer (ILAS/ILAS-II) onboard the ADEOS/ADEOS-II satellite, and observations of halogen chemicals involved in ozone depletion by the Superconductive Submillimeter-wave Emission Sounder (JEM/SMILES) onboard the International Space Station have been made in the past. The JEM/SMILES is a superconducting submillimeter-wave radiation sounder aboard the International Space Station. Atmospheric observation by satellites is one of the distinctive efforts of Japanese atmospheric chemistry research, and it is expected to contribute to the global academic community by continuously maintaining and strengthening its infrastructure. Satellite observations of reactive gases and aerosols have been continuously progressing in the United States and Europe, providing an extremely important base of observation data for Japanese researchers for collaboration. Also, geostationary satellite observations by the GEMS mission from Korea, which started recently, will be a powerful tool for atmospheric chemistry research in the Asia-Pacific region. It is also important for Japan to promote and strengthen research in collaboration with these overseas satellite observations. In future satellite observation projects, including those we are proposing, it is expected that the dynamics of atmospheric constituents that have already been observed by satellite will be better tracked by improving the temporal and spatial resolution of the observations, with better characterization of localized sources such as cities. It is also desirable to expand the capabilities of detecting and quantifying new trace constituents such as volatile organic compounds (VOCs). Discussions in the "Earth Observation Satellite Future Vision Subcommittee" of the "Earth and Planetary Science Committee" of the SCJ, and those in the "Task Force Meeting and Remote Sensing Subcommittee," which consists of 25 academic societies, should be continued and proposals for publicly solicited missions should be pursued for realization. In addition, the Japan Society of Atmospheric Chemistry should promote industry-government-academia collaboration and the use of data to solve social issues with CONSEO (Consortium for Satellite Earth Observation), as being its official member.

Aircrafts are excellent platforms for understanding the spatial distribution of atmospheric constituents, including vertical profiles, and have strengths in the measurement of diverse gaseous constituents that cannot be captured by current satellites and in the observation of aerosols that have complex properties such as composition and particle size. Japan's efforts in atmospheric chemistry research include A-FORCE aircraft observations and observational research using overseas aircraft platforms such as NASA's DC-8 during the mission TRACE-P. However, there is virtually no support system for aircraft observation research in the Earth science field in Japan; researched need to rely on the use of short-term chartered aircraft funded by external sources. This makes it difficult for domestic researchers to participate in aircraft observations. Also, the infrastructure for accumulating and developing observation techniques remains extremely weak. Therefore, it is necessary to expand the scope of aerial observation by establishing a support system. In the "Master Plan for the 24th Scientific Research Program" (Master Plan 2020) of the Science Council of Japan, "Promotion of Climate and Earth System Science Research by Aircraft Observation" was selected as one of the priority large-scale research programs that should be promoted promptly among the large-scale scientific research programs. If the introduction of an aircraft dedicated to Earth observation as proposed in this plan is realized, it is expected to greatly advance the acquisition of data on the dynamics and sources of atmospheric trace gases and aerosols, their interaction with the land surface and oceans, and their relationship with climate and weather, which cannot be obtained from the ground or satellite platforms. In this plan, it is proposed that the Center for Orbital and Sub-orbital Observations at the Institute of Space and Earth Environment, Nagoya University, will be the core of the joint use and operation of the project. In addition, related to this plan, the Meteorological Society of Japan has compiled a report entitled "Research Plan for the Promotion of Climate and Earth System Science System by Aircraft Observation." It is hoped that related academic institutions and organizations collaborate and continue to study the introduction of Earth observation aircraft.

In the CONTRAIL (Comprehensive Observation Network for Trace gases by AIrLiner) project, aircraft observations by national research institutes in collaboration with private operators are also a powerful tool for atmospheric chemistry research. In addition to the use of aircraft, it is worth considering the use of private platforms for atmospheric chemistry research, for example, drones and other related platforms. In collaboration with the private sector, it is necessary to pay attention to the establishment of a stable and fundamental observation system, since there is an aspect of relying on the generosity of the private sector. Recent examples of aircraft observations in the East Asia region include the NASA research project KORUS-AQ, an aircraft observation study of the area around the southern Korean Peninsula conducted in cooperation between U.S. and South Korean researchers. Japan's commitment to international aircraft observations in the Asian region should also be strengthened. In addition, the use of balloons is effective for the observations to the stratosphere, and the establishment of a sustainable observation system is desirable.

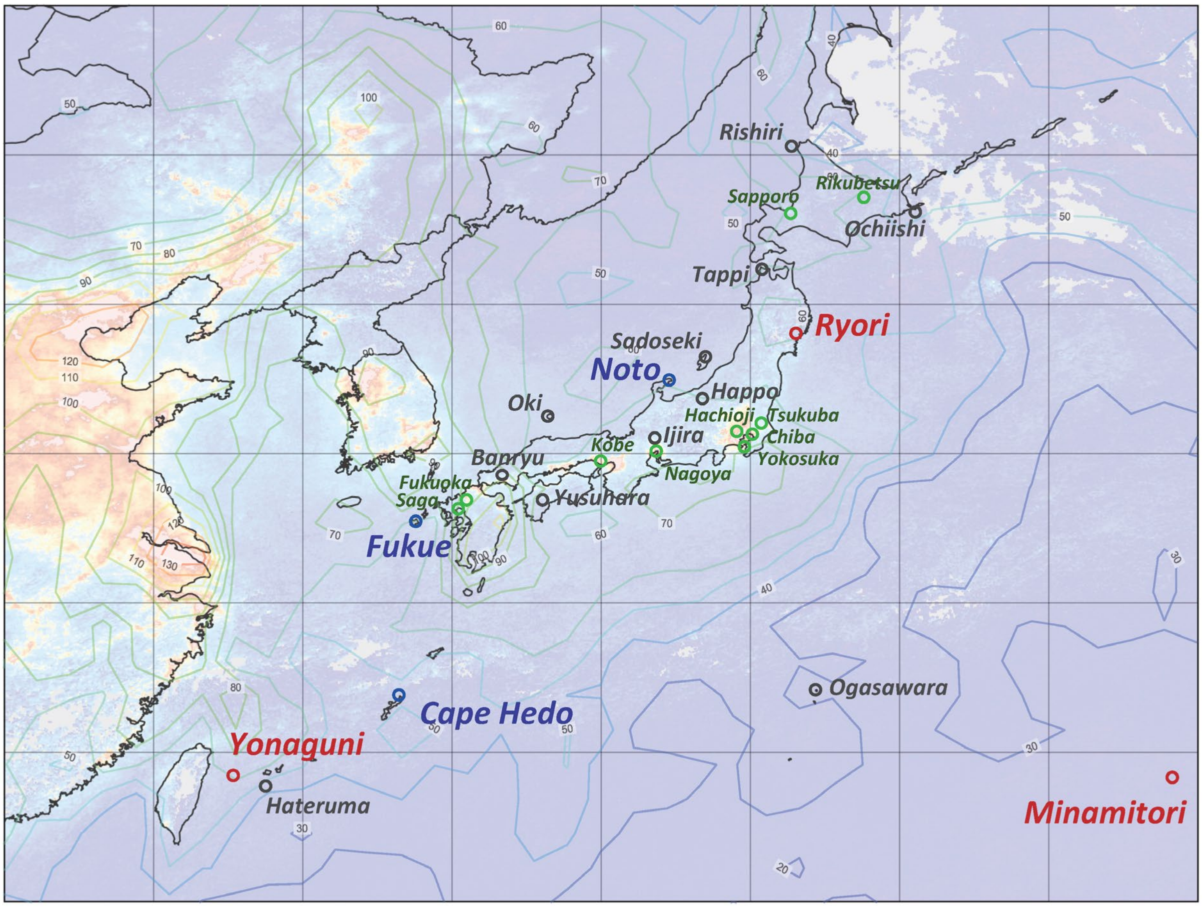


Figure 5. Domestic ground-based observation sites (red: JMA; blue and gray: supersites and stations maintained by the research community and laboratories; green: remote sensing measurement points. Background fill is tropospheric NO_2 column concentration from satellite TROPOMI. (emission distribution can be read), contour lines are typical spring near-surface ozone concentration distribution from data assimilation TCR-2)

Vessels are an important platform for understanding spatial distribution over the ocean, which cannot be covered by ground-based observations. Generally, the degree of freedom of the observation equipment that can be installed is much greater than that of aircraft, which have strict limitations on payload capacity. Shipboard observations are also important for research on chemical processes unique to ocean regions. It is also an important platform for understanding the atmospheric environment in the polar regions. The availability of opportunities for marine atmospheric observations is comparatively favorable, using the academic research vessels Hakuho Maru/Shinsei Maru and the oceanographic research vessels including R/V Mirai. However, the number of projects targeting atmospheric chemistry observation is not necessarily abundant, and there is still plenty of room for the atmospheric chemistry field to take the lead in shipboard observation. A new Arctic research vessel, named Mirai II, which is currently under construction for delivery in FY2026, will be operated by the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) and will continuously measure the basic components of atmospheric composition. JAMSTEC should serve as a focal point for national and international collaborative observations, accommodating additional equipment from other institutions to enhance the comprehensiveness of the research cruise missions. It is also important to continue discussions on a future successor to the Antarctic research vessel Shirase.

Ground-based observation networks (Fig. 5) are important platforms for both long-term monitoring and intensive observations of atmospheric processes using advanced instrumentation. Long-term monitoring platforms include the Cape Hedo Atmospheric and Aerosol Monitoring Station of the National Institute for Environmental Studies, the Fukue Atmospheric Environment Observatory maintained by Chiba University as a representative institution, and the Noto Supersite of Kanazawa University. The Ministry of the Environment's network (EANET) of long-term monitoring of air pollutants and acid deposition covering the north and south East Asia is also unique, and it is hoped to continue. In particular, the community has made significant achievements in the field of atmospheric chemistry in the long-term observation of pollutant outflows from the Asian continent on islands and coastal areas, and therefore its maintenance and development are strongly desired in the future. Domestic facilities are sometimes inferior to the facilities and equipment at Korean supersites, for example, and this is even a barrier to inviting Western researchers to conduct international joint observation and research. The ground-based observation system for aerosol concentration and particle size is still lacking, and should be expanded in the future with a view to contributing to the assessment of the climate impact of aerosols.

The development of research platforms is not only necessary in atmospheric observations, but also in laboratory experimental studies to understand the elementary processes of atmospheric chemistry. In particular, large reaction chambers are powerful tools in experiments to trace chemical transformations of gaseous components and aerosols. The smog chamber at the National Institute for Environmental Studies (NIES) has been a central platform for chamber experiments in Japan, and has produced important results. Since the maintenance and management of the chambers and associated instrumentation at the individual researcher level is an excessive burden, it is desirable for the atmospheric chemistry community to consider the maintenance and development of reaction experiment platforms for the continuation and development of process research in the future. In particular, it is desirable to consider linking the platform of experiments using cloud chambers to atmospheric chemistry research on themes related to ice crystal nuclei, etc., in addition to the use of chambers for tracking chemical reactions. The issue of new particle formation in the atmosphere is an important remaining issue that covers gases and aerosols with trace atmospheric components, but the CLOUD experiment at the European Organization for Nuclear Research (CERN) is currently the only facility enabling such research on the mechanism of new particle production. It is desirable for Japan to commit itself to such advanced experimental research overseas, and to consider its own experimental platform for taking the lead in indoor experimental research.

Computers are an essential research resource in numerical model simulations widely used in atmospheric chemistry. Atmospheric chemistry simulations can be roughly classified into online models coupled with weather and climate models and offline models that take the calculation results of weather and climate models as input data. In the former case, the number of forecast variables increases dramatically, resulting in a very large increase in CPU resources and memory usage. In the latter case, in addition to these, storage resources and I/O communication costs will increase markedly. Because of the burden of transferring data files, on-premise or private cloud servers are often used. On-premise servers, i.e., servers in the form of equipment installed and operated in a facility managed by the user, include RIKEN's Fugaku, the Earth Simulator of the Japan Agency for Marine-Earth

Science and Technology, and large computers of various organizations. But in the future, options may expand to the use of public cloud servers and virtual servers. Currently, we are in a transitional period from actual equipment to virtual equipment, and as computer performance and available resources develop, it is desirable to actively propose and promote atmospheric chemistry research that takes advantage of them. At present, atmospheric simulation relies heavily on large parallel computers equipped with vector or scalar conventional CPUs, while general-purpose computers equipped with GPUs for machine learning calculations have recently become available. In the future, it will be difficult to achieve the remarkable improvement in CPU performance that has been achieved so far, and it will be necessary to take into account speed-up through efficient parallel processing by utilizing GPUs. In addition to porting old physicochemical model calculations to GPUs, it will also be necessary to develop a method to utilize machine learning for atmospheric chemical simulations (so-called proxy modeling). It is necessary to sort out what kind of computer resources will be necessary or available in the future, depending on the objectives and calculation methods of atmospheric chemistry simulations.

(2) Maintenance and Use of Research Data

In recent years, it has become common for researchers to be required to disclose the data they use when publishing papers in academic journals, and the ground is being prepared to promote the use of data among researchers. As data repositories such as PANGAEA are being developed overseas, it is necessary to consider how Japan can contribute to data disclosure and use. In the field of atmospheric chemistry, there are many observational variables and it is expected that higher resolution data will become even more common in the future, and therefore, a large-scale storage under stable operating conditions with DOI assignment functionality would be desirable. It is also desirable to promote discussions on data collection, storage, and distribution, including coordination with existing efforts such as the World Data Center for Greenhouse Gases (WDCGG), one of the World Data Centers (WDC), and the ADS of the National Institute of Polar Research (NIPR). It is preferrable to consider efforts to develop or centralize a portal for all atmospheric chemistry observation data in Japan, as well as new initiatives that incorporate the needs of the atmospheric chemistry field. In promoting the use of these atmospheric observation data, it is also important to promote standardization of measurement methods. In this context, it is also necessary to consider how to collect and archive observation data that are not readily amenable to standardization, especially for advanced measurements.

Meteorological reanalysis data and emission inventories have long been shared, mainly through overseas efforts, and are important resources for atmospheric modeling research and other studies. The maintenance and development of these research resources, which link experiments and observations with modeling research, will continue to be important in the future. In addition to meteorological data provided by the Japan Meteorological Agency, the European Center for Medium-term Weather Forecasting (ECMWF), and the National Centers for Environmental Prediction (NCEP) in the United States, there is MERRA-2 provided by the National Aeronautics and Space Administration (NASA), which includes information on trace chemical constituents. In Japan, there are also efforts for aerosol reanalysis products (JRAero) and the second-generation chemical reanalysis data set (TCR-2) including reactive gases as well, and further expansion of data and information on trace constituents is desired. Regarding emission inventories, Japan has experience in developing emission inventories of air pollution- and climate change-related substances in the Asian region (e.g., REAS), and is strongly expected to continue contributing to the development of inventories for the Asian region and to the provision of guidelines for emission reduction measures.

Investment from academia in data science is expected to grow further in the future. Even though atmospheric chemistry is one of the disciplines that deals with large data sets, collaboration with data science could lead to the development of atmospheric chemistry from new angles. Although atmospheric chemistry research using artificial intelligence approaches is still in its infancy, it is a method with great potential for future development. As an example, an effort to apply deep learning to model global OH radical fluctuations and analyze the causes of fluctuations has been reported overseas. How to improve the current data archiving system and to develop the infrastructure related to big data should be considered when promoting the use of artificial intelligence methods such as machine learning.

(3) Human Resource Development

For the continued development of atmospheric chemistry in Japan, it is also important to sort out and improve issues related to the development of human resources to support it. While opportunities for learning about chemistry and physics, which form the basis of atmospheric chemistry, are available in secondary education, opportunities for learning geology, including topics on climate change and air quality, which atmospheric chemistry involves, are considered to be much less common than in other science subjects, mainly due to the subject selection for university entrance exams. It is desirable to improve the connection between high schools and universities in Earth science education by strengthening support from the academic community for geoscience education in high schools and by developing undergraduate university courses that take into account students with limited experience in high school geoscience studies. Secondary education institutions are focusing on developing the next generation of human resources with the ability to solve environmental and social problems from a global perspective, with the SDGs and other goals in mind, and therefore the atmospheric chemistry research field could make a strong positive contribution. In addition, education of atmospheric chemistry in the universities is mainly conducted in graduate schools, and there are few opportunities for it to be covered in undergraduate courses. There is room to consider ways to attract students' interest in atmospheric chemistry through support for undergraduate education, such as the expansion of teaching materials that can be easily handled in undergraduate classes. Atmospheric chemistry laboratories in the graduate school accept a wide range of students with backgrounds in chemistry and physics who are interested in global environmental issues. The fact that this is a field of study in which a wide range of expertise, not limited to geoscience, can be utilized would be regarded as a strength. In addition, it is desirable to widely promote the possibility of creating knowledge that directly leads to fundamental solutions to social issues, such as handling data on the concentration of substances that serve as improvement indicators for global warming and air pollution, and presenting effective reduction measures. Researchers from national research institutes provide support for higher education in the capacity of affiliated faculty members. By effectively utilizing the abundant research resources of these research institutes, human resource development at universities could be greatly enhanced.

As a career path for students who have studied atmospheric chemistry after obtaining their degrees, there are open positions for young researchers, mainly at the National Institute for Environmental Studies and other national institutions, and the employment situation surrounding young researchers who have just obtained their degrees is not very poor. However, as in other academic fields, it is not easy to obtain a permanent post, and the situation is such that researchers continue to work for a fixed-term post and aim to be selected for a permanent post or a tenure-track post (a post that can be transferred to permanent after examination). At universities, in addition to post-doctoral researchers employed with external funds, there are also tenure-track positions available for young researchers. But there is generally fierce competition among related fields for each a position, and therefore the production of excellent young researchers in atmospheric chemistry to be able to compete is a major key to the continuation and development of atmospheric chemistry education at universities. In addition, there is a concern that academic research positions are not an attractive option for young people, not only in the field of atmospheric chemistry, but also in other academic fields in Japan, because there are insufficient competition opportunities for positions. In addition, with Japan's nominal wages stagnating, the treatment of researchers, including post-doctoral fellows at universities or research institutes, may be losing their international competitiveness and hindering the attraction of talented young people from overseas. It is necessary to keep a close eye on whether the situation surrounding employment in this field will improve as a result of government measures to support young researchers.

Enhancing contacts with manufacturing and industrial sectors related to atmospheric chemistry would be worth. Collaboration that could lead to the development of cutting-edge measurement methods and consulting services would also be considered. It is worth considering not only from the perspective of strengthening atmospheric chemistry research, but also from the perspective of creating diverse career paths and exchanging human resources between industry and academia. Producing human resources with expertise in atmospheric chemistry to the private sector and society can be effective. Expectations are high for the use of data science for the global environment, and the use of satellite observation data on atmospheric composition on the Google Earth Engine, etc., to disseminate this information to industry and to exchange experts with adjacent fields will help to secure human resources in a broader sense. Promotion of gender equality is one of the important issues in employment in the field of atmospheric chemistry. The Japan Society of Atmospheric Chemistry (JpSAC) is actively increasing the ratio of women in leadership positions, and the Gender Equality and Human Resources Development Committee of JpSAC supports childcare during research meetings and conducts exchange programs for female members. In addition, the Committee actively supports and encourages young people who will lead the next generation both nationally and internationally.

Atmospheric chemistry is a field of study that deals with the atmosphere of the entire Earth and issues common to all humankind, such as global warming, making it easy to share research subjects with overseas researchers and to promote international exchange. Taking this advantage, there is room to promote international human resource development based on human resource exchange. Proactive development of international joint research is desirable, not only for the promotion of atmospheric chemistry research in Japan, but also as an opportunity to foster young researchers in Japan. It is also an important duty of Japan's academic community in the world to contribute to the development of young researchers overseas. The short course initiatives for young researchers at the iCACGP-IGAC 2018 international conference held in Takamatsu is a good example and such activities should also be spread to the domestic atmospheric chemistry community. It is also promoted that the domestic joint-use laboratories will take the initiative in inviting young researchers from overseas, leading to international exchanges in the future.

Appendix A: Summary from seven topical sections

Table A-1. Summary table for the topic **Greenhouse gases and ozone depleting substances**

(Authors: Taku Umezawa, Yosuke Niwa, Takuya Saito, Naoko Saitoh, Sakae Toyoda)

1. Ultimate Goals	✓ To quantitatively understand the causes of changes in atmospheric concentrations of GHGs and ODS from the viewpoints of emission sources and sinks, and to enable accurate future predictions based on an understanding of the circulation mechanism. ✓ To provide the international community with accurate information on the origin and distribution of emissions of target gases in order to contribute to the formulation of effective emission reduction measures under the Paris Agreement and the Montreal Protocol.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ The continuously developed atmospheric observations, emission inventories, atmospheric chemistry transport models, and terrestrial ecosystem models have enabled integrated comparative analysis of top-down and bottom-up methods, and an international project for this purpose has been organized, in which many Japanese researchers have participated. ✓ The data processing and utilization of column-averaged concentrations of CO_2 and CH_4 by the Japanese GHG observation satellite, GOSAT/TANSO-FTS, has made great progress. ✓ With the entry into force of the Paris Agreement, observation and modeling studies for understanding anthropogenic GHG emissions at regional and urban scales have been actively conducted. ✓ The importance of the Asian region as a source of GHG and ODS emissions is increasing, and Japanese observation data, which can detect source variability in the Asian region with high sensitivity, is highly valuable, and continuous provision of high-quality data is required. ✓ In Japan, unique observation platforms have been developed and maintained not only on the ground but also in balloons, aircraft, and ships, etc., and the results of observations of related tracers that are useful for GHG circulation analysis have been reported based on the development of unique technologies. ✓ Atmospheric chemistry transport models and terrestrial ecosystem models have been independently developed by several research groups in Japan and are highly recognized worldwide.
3. Current major gaps	✓ Observation data for continental inland and tropical regions are still insufficient at the global scale, and even higher-density observation data are required at the regional and urban scales, but there are significant technical and cost challenges in maintaining high-precision observations, developing analysis systems, and deploying new observations. ✓ Observation data from diverse platforms are increasing dramatically, but there are challenges to fully utilize diverse data for surface flux inverse analysis, such as ensuring uniform data quality and upgrading models to make the best use of the data.
4. Key questions and hypothesis to drive research and development during 2022-32	✓ How to evaluate whether the Paris Agreement and Montreal Protocol are working? What kind of spatio-temporal density, measurement accuracy, and observation platform observation data are needed? How should model analysis systems be developed to take advantage of diverse observational data? ✓ How will the sources and sinks of GHGs and ODS in the ocean and terrestrial ecosystems and atmospheric chemistry change with climate change, ocean acidification, ozone depletion and recovery, etc.? How will Arctic wetlands, permafrost, and forest fires be particularly affected by potentially dramatic changes?
5. Collaborations	GCP, RECCAP, NOAA, AGAGE, IGAC

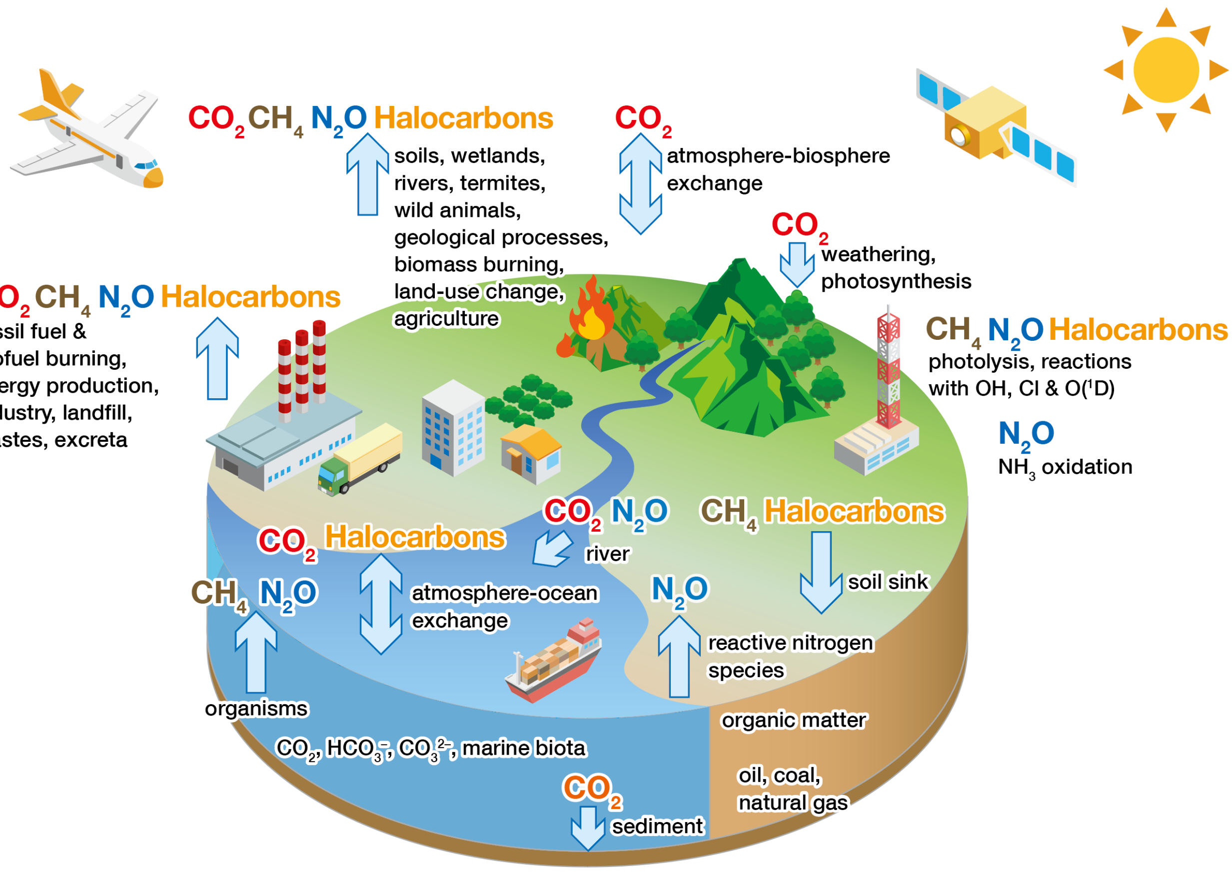


Figure A-1. Schematic diagram of global circulation of CO_2, CH_4, N_2O, and halocarbons.

Selected References (Greenhouse gases and ozone depleting substances)

Aoki, S., T. Nakazawa, S. Murayama & S. Kawaguchi (1992), Measurements of atmospheric methane at the Japanese Antarctic Station, Syowa, *Tellus*, 44B, 273–281.

Akiyama, H., Yan, X. & Yagi, K. (2006), Estimations of emission factors for fertilizer-induced direct N_2O emissions from agricultural soils in Japan: Summary of available data, *Soil Science and Plant Nutrition*, 52, 774–787, doi: 10.1111/j.1747-0765.2006.00097.x

Belikov, D. A. et al. (2013), Simulations of column-averaged CO_2 and CH_4 using the NIES TM with a hybrid sigma isentropic (s- θ) vertical coordinate, *Atmos. Chem. Phys.*, 13, 1713–1732, doi: 10.5194/acp-13-1713-2013.

Belikov, D. A., N. Saitoh, P. K. Patra, & N. Chandra, (2021), GOSAT CH_4 vertical profiles over the Indian Subcontinent: Effect of a priori and averaging kernels for climate applications, *Remote Sens.*, 13, 1677, doi:10.3390/rs1301677.69

Chandra, N., S. Hayashida, T. Saeki & P. K. Patra (2017), What controls the seasonal cycle of columnar methane observed by GOSAT over different regions in India?, *Atmos. Chem. Phys.*, 17, 12633－12643.

Chandra, N. et al. (2021), Emissions from the oil and gas sectors, coal mining and ruminant farming drive methane growth over the past three decades, *J. Meteor. Soc. Japan*, 99, 309– 337, doi: 10.2151/jmsj.2021-015.

Frey, M. et al. (2019), Building the COllaborative Carbon Column Observing Network (COCCON): long-term
stability and ensemble performance of the EM27/SUN Fourier transform spectrometer, *Atmos. Meas. Tech.*, 12, 1513－1530, doi: 10.5194/amt-12-1513-2019.

Fujita, R. et al. (2018). Temporal variations of the mole fraction, carbon, and hydrogen isotope ratios of atmospheric methane in the Hudson Bay Lowlands, Canada, *J. Geophys. Res. Atmos.*, 123, 4695–4711, doi: 10.1002/2017JD027972

Fujita, R. et al. (2020). Global and regional CH_4 emissions for 1995–2013 derived from atmospheric CH_4, δ13C - CH_4, and δD - CH_4 observations and a chemical transport model, *J. Geophys. Res. Atmos.*, 125, e2020JD032903, doi: 10.1029/2020JD032903.

Ghosh, A. et al. (2015), Variations in global methane sources and sinks during 1910–2010, *Atmos. Chem. Phys.*, 15, 2595–2612, doi: 10.5194/acp-15-2595-2015.

Goto, D., S. Morimoto, S. Aoki, P. K. Patra & T. Nakazawa (2017a), Terrestrial biospheric and oceanic CO_2 uptake estimated from long-term measurements of atmospheric CO_2 mole fraction, δ13C and $\delta(O_2/N_2)$ at Ny-Alesund, Svalbard, *J. Geophys. Res. Atmos.,* doi: 10.1002/2017JG003845.

Goto, D. et al. (2017b), Vertical profiles and temporal variations of greenhouse gases in the stratosphere over Syowa Station, Antarctica, *SOLA*, 13, 224–229, doi: 10.2151/sola.2017- 041.

Hayashida, S., A. Ono, S. Yoshizaki, C. Frankenberg, W. Takeuchi & X. Yan (2013), Methane concentrations over Monsoon Asia as observed by SCIAMACHY: Signals of methane emission from rice cultivation, *Remote Sens. Env.*, 139, 246－256.

Hirota, M., K. Nemoto, A. Murata & K. Fushimi (1991), Observation of Carbon Dioxide Concentrations in Air and Surface Sea Water in the Western North Pacific Ocean, *Oceanogr. Mag.*, 41, 19-28.

Iida, Y. et al. (2015), Trends in *p* CO_2 and sea–air CO_2 flux over the global open oceans for the last two decades. *J Oceanogr,* 71, 637–661, doi: 10.1007/s10872-015-0306-4.
Ishidoya, et al. (2012), Time and space variations of the O_2/N_2 ratio in the troposphere over Japan and estimation of the global CO_2 budget for the period 2000–2010, *Tellus B*, 64:1, doi: 10.3402/tellusb.v64i0.18964.
Ishidoya, S., et al. (2020), O_2 : CO_2 exchange ratio for net turbulent flux observed in an urban area of Tokyo, Japan, and its application to an evaluation of anthropogenic CO_2 emissions, *Atmos. Chem. Phys.*, 20, 5293–5308, doi: 10.5194/acp-20-5293-2020.
Ito, A. & M. Inatomi (2012), Use of a process-based model for assessing the methane budgets of global terrestrial ecosystems and evaluation of uncertainty, *Biogeosciences*, 9, 759–773, doi: 10.5194/bg-9-759-2012.
Kondo, M., T. Saeki, H. Takagi, K. Ichii & K. Ishijima (2016), The effect of GOSAT observations on estimates of net CO_2 flux in semi-arid regions of the Southern Hemisphere, *SOLA*, 12, 181－186.
Kondo, M. et al. (2020), State of the science in reconciling topdown and bottom-up approaches for terrestrial CO_2 budget, *Glob. Change Biol.*, 26: 1068–1084, doi: 10.1111/gcb.14917.
Kuze, A., H. Suto, M. Nakajima & T. Hamazaki (2009), Thermal and near infrared sensor for carbon observation Fourier transform spectrometer on the Greenhouse Gases Observing Satellite for greenhouse gases monitoring, *Appl. Optics*, 48, 6716－6733.
Kuze, A., N. Kikuchi, F. Kataoka, H. Suto, K. Shiomi & Y. Kondo (2020), Detection of methane emission from a local source using GOSAT target observations, *Remote Sens.*, 12, 267, doi: 10.3390/rs12020267.
Ishijima, K., T. Nakazawa, S. Sugawara, S. Aoki & T. Saeki (2001), Concentration variations of tropospheric nitrous oxide over Japan, *Geophys. Res. Lett.*, 28(1), 171–174, doi: 10.1029/2000gl011465.
Ishijima, K. et al. (2007), Temporal variations of the atmospheric nitrous oxide concentration and its δ15N and δ18O for the latter half of the 20th century reconstructed from firn air analyses, *J. Geophys. Res. Atmos.*, 112, D03305, doi: 10.1029/2006JD007208.
Ishijima, K. et al. (2022), Understanding Temporal Variations of Atmospheric Radon-222 around Japan using Model Simulations, *J. Meteorol. Soc. Japan*, 100(2), 343－359, doi: 10.2151/jmsj.2022-017.
Machida, T. et al. (2008), Worldwide measurements of atmospheric CO_2 and other trace gas species using commercial airlines, *J. Atmos. Ocean. Tech.*, 25, 1744– 1754, doi: 10.1175/2008JTECHA1082.1.
Maki, T. et al. (2010), New technique to analyse global distributions of CO_2 concentrations and fluxes from non-processed observational data, *Tellus B*, doi: 10.1111/j.1600- 0889. 2010.00488.x.
Maksyutov, S. et al. (2021), Technical note: A high-resolution inverse modelling technique for estimating surface CO_2 fluxes based on the NIES-TM–FLEXPART coupled transport model and its adjoint, *Atmos. Chem. Phys.*, 21, 1245–1266, doi: 10.5194/acp-21-1245-2021.
Matsueda, H., H. Y. Inoue (1996), Measurements of atmospheric CO_2 and CH_4 using a commercial airliner from 1993 to 1994, *Atmos. Environ.*, 30, 1647–1655.
Matsueda, H., H. Y. Inoue & M. Ishii (2002), Aircraft observation of carbon dioxide at 8–13 km altitude over the western Pacific from 1993 to 1999, *Tellus B*, 54:1, 1-21, doi: 10.3402/tellusb.v54i1.16645.
Morimoto, S., T. Nakazawa, K. Higuchi & S. Aoki (2000), Latitudinal distribution of atmospheric CO_2 sources and sinks inferred by δ13C measurements from 1985 to 1991, *J. Geophys. Res. Atmos.,* 105(D19), 24315–24326, doi: 10.1029/2000JD900386.
Morimoto, S., T. Nakazawa, S. Aoki, G. Hashida & T. Yamanouchi (2003), Concentration variations of atmospheric CO_2 observed at Syowa Station, Antarctica from 1984 to 2000, *Tellus B*, 55, 170-177.
Morimoto, S., R. Fujita, S. Aoki, D. Goto & T. Nakazawa (2017), Long-term variations of the mole fraction and carbon isotope ratio of atmospheric methane observed at NyAlesund, Svalbard from 1996 to 2013, *Tellus B*, 69:1, 1380497, doi: 10.1080/16000889.2017.1380497.
Nakazawa, T., K. Miyashita, S. Aoki & M. Tanaka (1991), Temporal and spatial variations of upper tropospheric and lower stratospheric carbon dioxide, *Tellus B*, 43:2, 106-117, doi: 10.3402/tellusb.v43i2.15254.
Nakazawa, T. et al. (2002), Variations of stratospheric trace gases measured using a balloon-borne cryogenic sampler, *Adv. Space Res.*, 30(5), 1349–1357.
Nakazawa, T. (2020), Current understanding of the global cycling of carbon dioxide, methane, and nitrous oxide, *Proc. Jpn. Acad., Ser. B*, 96, 394–419, doi: 10.2183/pjab.96.030.
Nara, H. et al. (2012), Effect of air composition (N_2, O_2, Ar, and H_2O) on CO_2 and CH_4 measurement by wavelength scanned cavity ring-down spectroscopy: calibration and measurement strategy, *Atmos. Meas. Tech.*, 5, 2689–2701, doi: 10.5194/amt-5-2689-2012.
Nara, H. et al. (2014), Emissions of methane from offshore oil and gas platforms in Southeast Asia, *Sci. Rep.*, 4:6503, doi: 10.1038/srep06503.
Nara, H. et al. (2017), Emission factors of CO_2, CO and CH_4 from Sumatran peatland fires in 2013 based on shipboard measurements, *Tellus B*, 69:1, 1399047, doi: 10.1080/16000889.2017.1399047.
Nishihashi, M. et al. (2019), Greenhouse gases and air pollutants monitoring project around Jakarta megacity, *IOP Conf. Ser.: Earth Environ. Sci.*, 303, 012038, doi: 10.1088/ 1755-1315/303/1/012038.
Niwa, Y., Tomita, H., Satoh, M. & Imasu, R. (2011), A three dimensional icosahedral grid advection scheme preserving monotonicity and consistency with continuity for atmospheric tracer transport, *J. Meteor. Soc. Japan*, 89, 255–268, doi:10.2151/jmsj.2011-306.
Niwa Y. et al. (2014), Seasonal Variations of CO_2, CH_4, N_2O and CO in the Mid-Troposphere over the Western North Pacific Observed Using a C-130H Cargo Aircraft, *J. Meteorol. Soc. Japan*, 92(1), 55－70, doi: 10.2151/jmsj.2014-104.
Niwa, Y. et al. (2017), A 4D-Var inversion system based on the icosahedral grid model (NICAM-TM 4D-Var v1.0) – Part 2: Optimization scheme and identical twin experiment of atmospheric CO_2 inversion, *Geosci. Model Dev.*, 10, 2201–2219, doi: 10.5194/gmd-10-2201-2017.
Niwa, Y. et al. (2021), Estimation of fire-induced carbon emissions from Equatorial Asia in 2015 using in situ aircraft and ship observations, *Atmos. Chem. Phys.*, 21, 9455–9473, doi: 10.5194/acp-21-9455-2021.
Nomura, S., H. Mukai, Y. Terao, T. Machida & Y. Nojiri (2017), Six years of atmospheric CO_2 observations at Mt. Fuji recorded with a battery-powered measurement system, *Atmos. Meas. Tech.*, 10, 667–680, doi: 10.5194/amt-10- 667-2017.
Nomura, S. et al. (2021), Measurement report: Regional characteristics of seasonal and long-term variations in
greenhouse gases at Nainital, India and Comilla, Bangladesh, *Atmos. Chem. Phys.*, 21, 16427–16452, doi: acp-21-16427-2021.
Oda, T. & S. Maksyutov (2011), A very high-resolution (1 km×1 km) global fossil fuel CO_2 emission inventory derived using a point source database and satellite observations of nighttime lights, *Atmos. Chem. Phys.*, 11, 543–556, doi: 10.5194/acp-11-543-2011.

Patra, P. K. et al. (2016), Regional Methane Emission Estimation Based on Observed Atmospheric Concentrations (2002− 2012), *J. Meteorol. Soc. Japan*, 94(1), 91−113, doi: 10.2151/jmsj.2016-006.
Patra, P. K., et al. (2018), Improved Chemical Tracer Simulation by MIROC4.0-based Atmospheric Chemistry- Transport Model (MIROC4-ACTM), *SOLA*, 14, 91−96, doi: 10.2151/sola.2018-016.
Patra, P. K. et al. (2021), Methyl chloroform continues to constrain the hydroxyl (OH) variability in the troposphere. J. *Geophys. Res.. Atmos.*, 126, e2020JD033862, doi:10.1029/2020JD033862.
Saeki, T. & P.K. Patra (2017), Implications of overestimated anthropogenic CO_2 emissions on East Asian and global land CO_2 flux inversion, *Geosci. Lett.* 4, 9, doi: 10.1186/s40562- 017-0074-7.
Saikawa, E. et al. (2014), Global and regional emissions estimates for N_2O, *Atmos. Chem. Phys.*, 14(9), 4617−4641, doi: 10.5194/acp-14-4617-2014.
Sasakawa, M. et al. (2013), Aircraft and tower measurements of CO_2 concentration in the planetary boundary layer and the lower free troposphere over southern taiga in West Siberia: Long-term records from 2002 to 2011, *J. Geophys. Res. Atmos.*, 118, 9489− 9498, doi: 10.1002/jgrd.50755.
Sasakawa, M. et al. (2017). Temporal characteristics of CH_4 vertical profiles observed in the West Siberian Lowland over Surgut from 1993 to 2015 and Novosibirsk from 1997 to 2015, *J. Geophys. Res. Atmos.*, 122, 11261−11273, doi: 10.1002/ 2017JD026836.
Sawa, Y. et al. (2015), Seasonal changes of CO_2, CH_4, N_2O, and SF_6 in the upper troposphere/lower stratosphere over the Eurasian continent observed by commercial airliner, *Geophys. Res. Lett.*, 42, doi: 10.1002/2014GL062734.
Sugawara, S. et al. (1997), Vertical profile of the carbon isotopic ratio of stratospheric methane over Japan, *Geophys. Res. Lett.*, 24, 23, 2989−2992.
Sugawara S., et al. (2018), Age and gravitational separation of the stratospheric air over Indonesia, *Atmos. Chem. Phys.*, doi: 10.5194/acp-18-1819-2018.
Sugawara, H., et al. (2021), Anthropogenic CO_2 emissions changes in an urban area of Tokyo, Japan, due to the COVID-19 pandemic: A case study during the state of emergency in April − May 2020, *Geophys. Res. Lett.*, 48, e2021GL092600, doi: 10.1029/2021GL092600.
Takahashi, T., et al. (2009), Climatological mean and decadal change in surface ocean *p* CO_2, and net sea−air CO_2 flux over the global oceans, *Deep-Sea Res Pt II,* 56:554−577.
Tanaka, M., T. Nakazawa & S. Aoki (1983), Concentration of atmospheric carbon dioxide over Japan, *J. Geophys. Res. Atmos.,* 88(C2), 1339−1344, doi: 10.1029/JC088iC02p01339.
Terao, Y. et al. (2011), Interannual variability and trends in atmospheric methane over the western Pacific from 1994 to 2010, *J. Geophys. Res. Atmos.,* 116, D14303, doi: 10.1029/2010JD015467.
Tohjima, Y. et al. (2000), Variations in atmospheric nitrous oxide observed at Hateruma monitoring station, *Chemosphere Global Change Sci.*, 2(3-4), 435−443.
Tohjima, Y. et al. (2002), Analysis and presentation of in situ atmospheric methane measurements from Cape Ochi-ishi and Hateruma Island, *J. Geophys. Res. Atmos.,* 107, D12, 4148, doi: 10.1029/2001JD001003.
Tohjima, Y., H. Mukai, T. Machida, Y. Nojiri & M. Gloor (2005), First measurements of the latitudinal atmospheric O_2 and CO_2 distributions across the western Pacific, *Geophys. Res.Lett.*, 32, L17805, doi: 10.1029/2005GL023311.
Tohjima, Y., H. Mukai, S. Hashimoto & P. K. Patra (2010), Increasing synoptic scale variability in atmospheric CO_2 at Hateruma Island associated with increasing East-Asian emissions, *Atmos. Chem. Phys.*, 10, 453−462, doi: 10.5194/acp-10-453-2010.
Tohjima, Y. et al. (2014), Temporal changes in the emissions of CH_4 and CO from China estimated from CH_4/CO_2 and CO/CO_2 correlations observed at Hateruma Island, *Atmos. Chem. Phys.*, 14, 1663−1677, doi: 10.5194/acp-14-1663- 2014.
Tohjima, Y., H. Mukai, T. Machida, Y. Hoshina & S.-I. Nakaoka (2019), Global carbon budgets estimated from atmospheric O_2/N_2 and CO_2 observations in the western Pacific region over a 15-year period, *Atmos. Chem. Phys.*, 19, 9269−9285, doi: 10.5194/acp-19-9269-2019.
Tohjima, Y. et al. (2020), Detection of fossil-fuel CO_2 plummet in China due to COVID-19 by observation at Hateruma, *Sci. Rep.*, 10, 18688, doi: 10.1038/s41598-020-75763-6.
Toyoda, S. et al. (2008), Isotopomeric characterization of N_2O produced, consumed, and emitted by automobiles, *Rapid Commun. Mass Spectrom.*, 22, 603−612. doi: 10.1002/rcm.3400.
Toyoda, S. et al. (2011), Characterization and production and consumption processes of N_2O emitted from temperate agricultural soils determined via isotopomer ratio analysis, *Glob. Biogeochem. Cycles*, 25, GB2008. doi:10.1029/2009GB003769.
Toyoda, S. et al. (2013), Decadal time series of tropospheric abundance of N_2O isotopomers and isotopologues in the Northern Hemisphere obtained by the long-term observation at Hateruma Island, Japan, *J. Geophys. Res. Atmos.,* 118, 3369−3381, doi: 10.1002/jgrd.50221.
Toyoda, S. et al. (2018), Vertical distributions of N_2O isotopocules in the equatorial stratosphere, *Atmos. Chem. Phys.*, 18, 833−844, doi: 10.5194/acp-18-833-2018.
Tsuboi, K. et al. (2013), Evaluation of a new JMA aircraft flask sampling system and laboratory trace gas analysis system, *Atmos. Meas. Tech*., 6, 1257−1270, doi: 10.5194/amt-6- 1257-2013.
Umezawa, T. et al. (2012a), Carbon and hydrogen isotopic ratios of atmospheric methane in the upper troposphere over the Western Pacific, *Atmos. Chem. Phys.*, 12, 8095−8113, doi: 10.5194/acp-12-8095-2012.
Umezawa, T., T. Machida, S. Aoki, & T. Nakazawa (2012b), Contributions of natural and anthropogenic sources to atmospheric methane variations over western Siberia estimated from its carbon and hydrogen isotopes, *Glob. Biogeochem. Cycles*, 26, GB4009, doi: 10.1029/2011GB004232.
Umezawa, T. et al. (2014), Variations of tropospheric methane over Japan during 1988 − 2010, *Tellus B*, 66, 23837, doi:10.3402/tellusb.v66.23837.
Umezawa, T. et al. (2018), Interlaboratory comparison of δ13C and δD measurements of atmospheric CH_4 for combined use of data sets from different laboratories, *Atmos. Meas. Tech.*, 11, 1207−1231, doi: 10.5194/amt-11-1207-2018.
Yokohata, T. et al. (2020), Future projection of greenhouse gas emissions due to permafrost degradation using a simple numerical scheme with a global land surface model, *Prog. Earth Planet. Sci.*, 7:56, doi: 10.1186/s40645-020-00366-8.
Wada, A., H. Matsueda, Y. Sawa, K. Tsuboi & S. Okubo (2011), Seasonal variation of enhancement ratios of trace gases observed over 10 years in the western North Pacific, *Atmos. Environ.*, 45, 2129−2137, doi: 10.1016/j.atmosenv.2011.01.043.

Watanabe, F. et al. (2000), Interannual variation of growth rate of atmospheric carbon dioxide concentration observed at the JMA's three monitoring stations: Large increase in concentration of atmospheric carbon dioxide in 1998, *J. Meteorol. Soc. Japan*, 78, 673-682.

Yoshida, Y. et al. (2011), Retrieval algorithm for CO_2 and CH_4 column abundances from short-wavelength infrared spectral observations by the Greenhouse gases observing satellite, *Atmos. Meas. Tech.*, 4, 717－734.

Yoshida, Y. et al. (2013), Improvement of the retrieval algorithm for GOSAT SWIR XCO_2 and XCH_4 and their validation using TCCON data, *Atmos. Meas. Tech.*, 6, 1533－1547.

Zeng, J., Y. Nojiri, P. Landschutzer, M. Telszewski & S. Nakaoka (2014), A Global Surface Ocean fCO_2 Climatology Based on a Feed-Forward Neural Network, *J. Atmos. Oceanic Technol.*, 31, 1838–1849, doi: 10.1175/JTECH-D-13-00137.1.

Table A-2. Summary table for the topic **Reactive gases**

(Authors: Yugo Kanaya, Kengo Sudo, Prabir K. Patra, Yosuke Sakamoto, Takashi Sekiya, Tamaki Fujinawa, Hiroshi Tanimoto, Nawo Eguchi, Naoko Saitoh, Yasuko Kasai, Makoto Deushi, Tomohiro O. Sato)

1. Ultimate Goals	✓ Be able to express various processes such as photochemical reactions, atmospheric transport, deposition, etc. in equations and, given emissions and boundary conditions, be able to predict future concentration-time evolution and budget of a group of atmospheric trace gas constituents. To be able to quantitatively understand the role of each process. ✓ The ability to link the causes of concentration changes to changes in emissions, quantify the contribution of each component to climate change as short-lived climate forcing factors (SLCFs), including feedbacks, and health effects, and to propose mitigation measures. ✓ The ability to provide accurate chemical weather forecasts to society based on the laws of physics and chemistry.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ The worldwide progress of satellite observations, high-precision in-situ observations, and standardization has greatly improved the spatio-temporal coverage of information on atmospheric constituent concentrations, and has facilitated the integration of observations and numerical models. ✓ Japan's strengths include: understanding the causes of regional ozone and $PM_{2.5}$ pollution in East Asia, evaluation of OH radical reactivity and heterogeneous uptake coefficients of radicals, process knowledge from isotopes, development of multiple Earth system models, progress in atmospheric chemistry transport modeling and data assimilation, and satellite observation analysis technology developed with GOSAT and others, emission inventories.
3. Current major gaps	✓ Lack of understanding of the OH radical reaction system, which is at the heart of atmospheric chemistry ✓ Lack of understanding of the nonlinearity of ozone chemistry and the spatial distribution of production rate and limiting factors ✓ Insufficient quantification of causal relationships between changes in atmospheric composition and emissions, and rapid assessment of emission inventories to optimize reduction measures ✓ Lack of information on vertical distribution (including stratospheric) and daily variations even with improved satellite observations ✓ Lack of early detection observation information for prediction of sudden events
4. Key questions and hypothesis to drive research and development during 2022-32	✓ Can the gap between methods in global OH estimation (hemispheric gradient, trend) be filled? ✓ Will satellite observation systems with kilometer-level horizontal resolution and continuous areal distribution and integrated greenhouse gas (GHG)-air quality (AQ) analysis significantly advance our understanding of nonlinearities and limiting factors in ozone chemistry and our assessment of important emission sources? ✓ Can AQ analysis also provide scientific evidence for decision making on climate stabilization? ✓ If ammonia, VOCs, etc. are integrated into numerical models in addition to NOx, etc., will the prediction of ozone, $PM_{2.5}$, etc. be improved (will this provide knowledge on food issues in addition to climate change and health issues)? ✓ Can we improve not only chemical weather forecasting and future prediction of the Earth system, but also the current numerical forecast model of meteorology by improving our understanding of the interaction between atmospheric chemistry, meteorology, and the Earth system, and by incorporating key observation information, thereby providing useful information to society?
5. Collaborations	✓ IGAC, iCACGP, COSPAR, CEOS, SPARC

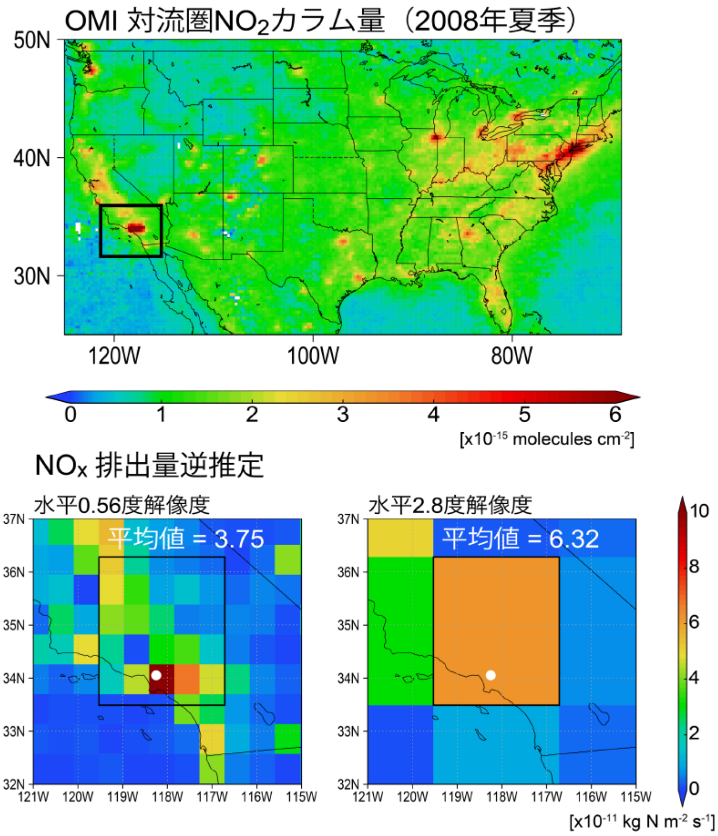


Figure A-2. Estimated NOx emissions from data assimilation system with two different horizontal resolutions based on OMI satellite observations of tropospheric NO_2. (modified from Sekiya et al. (2021))

Selected References (Reactive gases)

Akimoto, H. et al. (2019), Comparison of surface ozone simulation among selected regional models in MICS-Asia III – effects of chemistry and vertical transport for the causes of difference, *Atmos. Chem. Phys.*, 19, 603–615.

Akimoto, H. et al. (2020), Discrepancies between MICS-Asia III simulation and observation for surface ozone in the marine atmosphere over the northwestern Pacific Asian Rim region, *Atmos. Chem. Phys.*, 20, 15003–15014.

Fujinawa, T. et al. (2021). First concurrent observations of NO_2 and CO_2 from power plant plumes by airborne remote sensing, *Geophysical Research Letters*, 48, e2021GL092685, doi:10.1029/2021GL092685.

Ha, P. T. M., R. Matsuda, Y. Kanaya, F. Taketani & K. Sudo (2021), Effects of heterogeneous reactions on tropospheric chemistry: a global simulation with the chemistry–climate model CHASER V4.0, *Geosci. Model Dev.*, 14, 3813–3841.

Hattori, S. et al. (2021), Isotopic evidence for acidity-driven enhancement of sulfate formation after SO_2 emission control, *Sci. Adv.*, 7(19), eabd4610.

Hayashida, S., M. Kajino, M. Deushi, T. T. Sekiyama & X. Liu (2018), Seasonality of the lower tropospheric ozone over China observed by the Ozone Monitoring Instrument, *Atmos. Environ.*, 184, 244-253.

Inoue, K., K. Tonokura, K. & H. Yamada (2019), Modeling study on the spatial variation of the sensitivity of photochemical ozone concentrations and population exposure to VOC emission reductions in Japan, *Air Qual. Atmos. Health*, 12, 1035-1047.

Irie, H. et al. (2021), Continuous multi-component MAX-DOAS observations for the planetary boundary layer ozone variation analysis at Chiba and Tsukuba, Japan, from 2013 to 2019, *Progress in Earth and Planetary Science*, 8, 31.

Ishino, S. et al. (2017), Seasonal variations of triple oxygen isotopic compositions of atmospheric sulfate, nitrate, and ozone at Dumont d'Urville, coastal Antarctica. *Atmos. Chem. Phys.*, 17(5), 3713– 3727.

Ishino, S. et al. (2021), Regional Characteristics of Atmospheric Sulfate Formation in East Antarctica Imprinted on 17O Excess Signature, *J. Geophys. Res. Atmospheres*, 126, e2020JD033583.

Itahashi, S., R. Mathur, C. Hogrefe, S. L. Napelenok & Y. Zhang (2020), Modeling stratospheric intrusion and trans-Pacific transport on tropospheric ozone using hemispheric CMAQ during April 2010 – Part 2: Examination of emission impacts based on the higher-order decoupled direct method, *Atmos. Chem. Phys.*, 20, 3397–3413.

Kanaya, Y. et al. (2007), Chemistry of OH and HO_2 radicals observed at Rishiri Island, Japan, in September 2003: Missing daytime sink of HO_2 and positive nighttime correlations with monoterpenes, *J. Geophys. Res.*, 112, D11308, doi: 10.1029/2006JD007987.

Kanaya, Y. et al. (2012), Comparisons of observed and modeled OH and HO_2 concentrations during the ambient measurement period of the HOxComp field campaign, *Atmos. Chem. Phys.*, 12, 2567–2585.

Kanaya, Y. et al. (2019), Ozone and carbon monoxide observations over open oceans on R/V *Mirai* from 67° S to 75° N during 2012 to 2017: testing global chemical reanalysis in terms of Arctic processes, low ozone levels at low latitudes, and pollution transport, *Atmos. Chem. Phys*., 19, 7233–7254

Kanaya, Y. et al. (2020), Rapid reduction in black carbon emissions from China: evidence from 2009–2019 observations on Fukue Island, Japan, *Atmos. Chem. Phys.*, 20, 6339–6356.

Kohno, N., J. Li, Y. Sakamoto & Kajii, Y. (2020), Rate constants of CH_3O_2 + NO_2 ↔ $CH_3O_2NO_2$ and $C_2H_5O_2$ + NO_2 ↔ $C_2H_5O_2NO_2$ reactions under atmospheric conditions, *Int. J. Chem. Kinet.*, 53, 571-582.

Kurokawa, T. & T. Ohara (2020), Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3, *Atmos. Chem. Phys.*, 20, 12761–12793, doi: 10.5194/acp-20-12761-2020.

Li, J. et al. (2020), Total hydroxyl radical reactivity measurements in a suburban area during AQUAS–Tsukuba campaign in summer 2017. *Sci. Total Environ.* 740, 139897.

Miyazaki, K. et al. (2017), Decadal changes in global surface NOx emissions from multi-constituent satellite data assimilation, *Atmos. Chem. Phys*., 17, 807–837.

Miyazaki, K., et al. (2020a), Evaluation of a multi-model, multiconstituent assimilation framework for tropospheric chemical reanalysis, *Atmos. Chem. Phys*., 20, 931–967.

Miyazaki, K. et al. (2020b), Updated tropospheric chemistry reanalysis and emission estimates, TCR-2, for 2005–2018, *Earth Syst. Sci. Data*, 12, 2223–2259.

Miyazaki, K. et al. (2021), Global tropospheric ozone responses to reduced NOx emissions linked to the COVID-19 worldwide lockdowns, *Sci. Adv.*, 7 (24), eabf7460.

Nagashima, T., K. Sudo, H. Akimoto, J. Kurokawa, & T. Ohara (2017), Long-term change in the source contribution to surface ozone over Japan, *Atmos. Chem. Phys.*, 17, 8231- 8246.

Nakajima, T. et al. (2017), Model depiction of the atmospheric flows of radioactive cesium emitted from the Fukushima Daiichi Nuclear Power Station accident. *Progress in Earth and Planetary Science*, 4, 2, doi:10.1186/s40645-017- 0117-x.

Nakashima, Y., N. Kamei, S. Kobayashi & Y. Kajii, (2010), Total OH reactivity and VOC analyses for gasoline vehicular exhaust with a chassis dynamometer. *Atmos. Environ.* 44, 468-475.

Nakashima, Y. et al. (2014), Total OH reactivity measurements in ambient Air in a southern Rocky Mountain Ponderosa Pine Forest during BEACHON-SRM08 summer campaign. *Atmos. Environ.* 85, 1-8.

Oshima, N. et al. (2012), Wet removal of black carbon in Asian outflow: Aerosol Radiative Forcing in East Asia (AFORCE) aircraft campaign, *J. Geophys. Res.*, 117, D03204, doi:10.1029/2011JD016552, 2012.

Patra, P. et al. (2014), Observational evidence for interhemispheric hydroxyl-radical parity, *Nature*, 513, 219–223, doi: 10.1038/nature13721.

Patra, P. K. et al. (2017), The Orbiting Carbon Observatory (OCO-2) tracks 2-3 peta-gram increase in carbon release to the atmosphere during the 2014-2016 El Nino, *Sci. Rep.*, 7, 13567.

Patra, P. K. et al. (2021), Methyl chloroform continues to constrain the hydroxyl (OH) variability in the troposphere. *J. Geophys. Res. Atmospheres*, 126, e2020JD033862. doi: 10.1029/2020JD033862.

Sadanaga, Y. A. Yoshino, S. Kato & Y. Kajii (2005), Measurements of OH reactivity and photochemical ozone production in the urban atmosphere, *Environ. Sci. Technol.,* 39, 8847-8852.

Sakamoto, Y., A. Yabushita, M. Kawasaki & S. Enami, (2009), Direct emission of I2 molecule and IO radical from the heterogeneous reactions of gaseous ozone with aqueous potassium iodide solution, *J. Phys. Chem. A*, 113, 7707- 7713, doi: 10.1021/jp903486u.

Sakazaki, T. et al. (2013), Diurnal ozone variations in the stratosphere revealed in observations from the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) on board the International Space

Station (ISS), *J. Geophys. Res. Atmospheres*, 118, 7, 2991—3006, doi: 10.1002/jgrd.50220

Sato, K. et al. (2017), Total OH Reactivity Measurements for the OH-Initiated Oxidation of Aromatic Hydrocarbons in the Presence of NOx. *Atmos. Environ.* 171, 272-278.

Sato, T. O., T. Kuroda & Y. Kasai, Novel index to comprehensively evaluate air cleanliness: the Clean aIr Index (CII), *Geoscience Communication*, 3, 233-247, doi:10.5194/gc-3-233-2020, 2020.

Sekiya, T. et al. (2020), Global bromine- and iodine-mediated tropospheric ozone loss estimated using the CHASER chemical transport model, *SOLA*, doi: 10.2151/sola.2020-037.

Sekiya, T. et al. (2021), Impacts of horizontal resolution on global data assimilation of satellite measurements for tropospheric chemistry analysis, *J. Adv. Model. Earth Syst.*, 13, e2020MS002180.

Sekiyama, T. T. & M. Kajino (2020), Reproducibility of surface wind and tracer transport simulations over complex terrain using 5-, 3-, and 1-km-Grid Models, *J. Appl. Meteorol. Climatol.*, 59, 937-952.

Someya, Y. et al. (2020), Atmospheric ammonia retrieval from the TANSO-FTS/GOSAT thermal infrared sounder, *Atmos. Meas. Tech.*, 13, 309–321.

Sudo, K., M. Takahashi & H. Akimoto (2003), Future changes in stratosphere-troposphere exchange and their impacts on future tropospheric ozone simulations, *Geophys. Res. Lett.*, 30, 2256, doi:10.1029/2003GL018526, 24.

Takashima, H. et al. (2022), Full latitudinal marine atmospheric measurements of iodine monoxide, *Atmos. Chem. Phys.*, 22, 4005–4018, doi:10.5194/acp-22-4005-2022, 2022.

Takegawa, N. et al. (2004), Removal of NOx and Noy in Asian outflow plume: Aircraft measurements over the western Pacific in January 2002, *J. Geophys. Res.*, 109, doi:10.1029/2004JD004866.

Taketani, F. et al. (2012), Measurement of overall uptake coefficients for HO_2 radicals by aerosol particles sampled from ambient air at Mts. Tai and Mang (China). *Atmos. Chem. Phys.* 12, 11907-11916.

Taketani, F., Y. Kanaya & H. Akimoto (2013), Kinetic studies of heterogeneous reaction of HO_2 radical by dicarboxylic acid particles.

International Journal of Chemical Kinetics 45,560-565.

Takigawa, M. et al. (2020), Can Delhi's pollution be affected by crop fires in the Punjab region?, *SOLA*, 16, 77-80.

Tanimoto, H., K. Ikeda, R. van der A, S. Garivait & K. F. Boersma (2015), Interannual variability of nitrogen oxides emissions from boreal fires in Siberia and Alaska during 1996–2011 as observed from space, *Environ. Res. Lett*., 10, 065004.

Toride, K. et al. (2021), Potential of mid-tropospheric water vapor isotopes to improve large-scale circulation and weather predictability, *Geophys. Res. Lett.*, doi: 10.1029/2020GL091698.

Tsunogai, U. et al. (2010), Tracing the fate of atmospheric nitrate deposited onto a forest ecosystem in Eastern Asia using Δ17O, *Atmos. Chem. Phys.*, 10, 1809–1820, doi: 10.5194/acp-10-1809-2010, 2010.

Ueda, K., H. Nitta & H. Odajima (2010), The effects of weather, air pollutants and Asian dust on hospitalization for asthma in Fukuoka, *Environ. Health Prev. Med.*, 15, 350-357, doi: 10.1007/s12199-010-0150-5.

Xu, H. et al. (2021), Determination of the triple oxygen isotopic composition of tropospheric ozone in terminal positions using a multistep nitrite-coated filter-pack system. *Rapid Commun. Mass Spectrom*. 35, e9124. doi: 10.1002/rcm.9124.

Yamaji, K. et al. (2006), Analysis of the seasonal variation of ozone in the boundary layer in East Asia using the Community Multi-scale Air Quality model: What controls surface ozone levels over Japan?, *Atmos. Environ*., 40, 1856-1868.

Yoshino, A. et al. (2012), Air quality diagnosis from comprehensive observations of total OH reactivity and reactive trace species in urban central Tokyo, *Atmos. Environ.,* 49, 51-59.

Zhou, J., K. Murano, N. Kohno, Y. Sakamoto & Y. Kajii (2020), Real-time quantification of the total HO_2 reactivity of ambient air and HO_2 uptake kinetics onto ambient aerosols in Kyoto (Japan), *Atmos. Environ.*, 223, 117189, doi: 10.1016/j.atmosenv.2019.117189._

Table A-3. Summary table for the topic **Laboratory experiments and physical chemistry**

(Authors: Shinichi Enami, Shinnosuke Ishizuka, Tetsuya Hama, Satoshi Inomata)

1. Ultimate Goals	✓ Understanding of all physical and chemical processes that occur in the atmosphere. ✓ Derivation of reaction rate constants, uptake coefficients, and photolysis rate constants, taking into account the solvent effects of molecular heterogeneity and the peculiarities of interfaces. ✓ Based on the above data, all physico-chemical processes that occur in the atmosphere (chemical composition, volatility distribution, hygroscopicity, and optical properties for aerosols) can be computationally reproduced and predicted.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ The understanding of gas-phase reactions that lead to the formation of aerosols has been advanced worldwide. ✓ The importance of multiphase reactions involving interfaces has been recognized. ✓ Experimental equipment and theoretical calculations have been developed to elucidate the nature of interfaces. ✓ In Japan, original research results based on physical chemistry have been produced. ✓ The rise of emerging Asian countries and Japan's geographical advantages (joint research and personnel exchange).
3. Current major gaps	✓ The gap between laboratory experiments (simple systems and high concentrations) and real atmospheres (complex systems and low concentrations). ✓ The detection of intermediates to determine the formation mechanism of final products. ✓ What fraction of the aerosol components can be analyzed and quantified correctly? ✓ The understanding of the interactions between aerosols and clouds is not sufficient. ✓ The impact of reactions involving oceanic microlayers and ice and snow, where there is mass transfer to and from the atmosphere, has not been adequately evaluated. ✓ Interfacial phenomena can only be treated qualitatively and not quantified (e.g., nonlinear increase in reaction rate constants at the interface).
4. Key questions and hypothesis to drive research and development during 2022-32	✓ Can we improve the sensitivity of measurement instruments and the accuracy of multi-component simultaneous high time-resolution measurement devices? ✓ Can we correctly understand the formation mechanism of monomers and dimers (especially HOM) in the oxidation process and the chemical reaction process in the particle phase? ✓ How can we (simply) organize the production yield of secondary aerosols by key factors (oxidants, temperature, humidity, amount and properties of existing particles)? Are there any other factors that we are overlooking? ✓ Is it possible to capture the chemical transformation process of aerosols microscopically (e.g., with optical tweezers)? (Example: Capture aerosols in the air with optical tweezers, keep them for a few days, and measure them spectroscopically (Raman, IR). Can we obtain evidence of compositional changes due to OH oxidation during the day and NO_3 oxidation at night?) ✓ Understanding of phenomena peculiar to the interface. ✓ What are the breakthroughs for quantification of heterogeneity (complexity) and interfacial phenomena?
5. Collaborations	✓ IGAC, Molecular Science Society, Synthetic Organic Chemistry

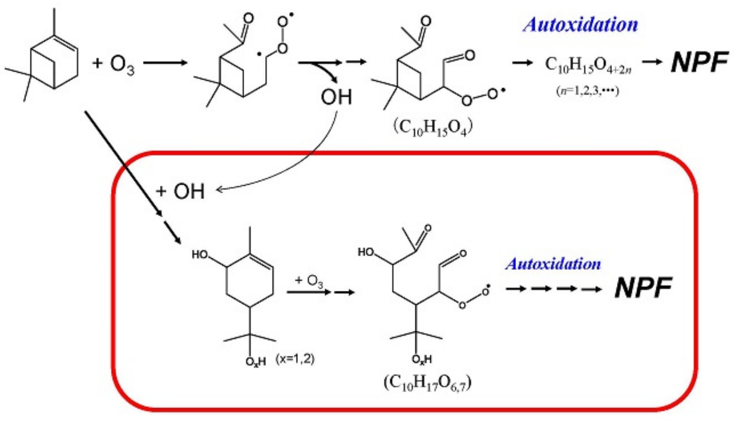


Figure A-3. New particle formation pathways from oxidation of α-pinene (from Inomata (2021)).

Selected References (Laboratory experiments and physical chemistry)

Akimoto, H. & J. Hirokawa (2020), Field Observations of Secondary Organic Aerosols, in *Atmospheric Multiphase Chemistry*, edited, doi:https://doi.org/10.1002/9781119422419.ch8.

Deng, Y. et al. (2021), Temperature and acidity dependence of secondary organic aerosol formation from α-pinene ozonolysis with a compact chamber system, *Atmos. Chem. Phys.*, *21*(8), 5983-6003, doi:10.5194/acp-21-5983-2021.

Enami, S. (2021), Fates of Organic Hydroperoxides in Atmospheric Condensed Phases, *J. Phys. Chem. A*, *125*(21), 4513-4523, doi:10.1021/acs.jpca.1c01513.

Enami, S., S. Ishizuka & A. J. Colussi (2019), Chemical signatures of surface microheterogeneity on liquid mixtures, *J. Chem. Phys.*, *150*(2), 024702.

Enami, S., H. Mishra, M. R. Hoffmann & A. J. Colussi (2012), Protonation and oligomerization of gaseous isoprene on mildly acidic surfaces: Implications for Atmospheric Chemistry, *J. Phys. Chem. A*, *116*(24), 6027-6032, doi:10.1021/jz100322w.

Enami, S., Y. Sakamoto & A. J. Colussi (2014), Fenton chemistry at aqueous interfaces, *Proc. Natl. Acad. Sci. U. S. A.*, *111*(2), 623-628, doi:10.1073/pnas.1314885111.

Endo, T. et al. (2018), Spherical Spontaneous Capillary- Wave Resonance on Optically Trapped Aerosol Droplet, *J. Phys. Chem. C*, *122*(36), 20684-20690, doi:10.1021/acs.jpcc.8b03784.

Hatakeyama, S. & H. Akimoto (1994), Reactions of Criegee intermediates in the gas-phase, *Res. Chem. Intermed.*, *20*(3-5), 503-524, doi:10.1163/156856794x00432.

Hiranuma, Y., K. Kaniwa, M. Shoji & F. Mafune (2011), Solvation Structures of Iodide on and below a Surface of Aqueous Solution Studied by Photodetachment Spectroscopy, *J. Phys. Chem. A*, *115*(30), 8493-8497, doi:10.1021/jp204195t.

Iinuma, Y., S. Ramasamy, K. Sato, A. Kołodziejczyk & R. Szmigielski (2021), Structural Characterisation of Dimeric Esters in α-Pinene Secondary Organic Aerosol Using N2 and CO_2 Ion Mobility Mass Spectrometry, *Atmosphere*, *12*(1), 17.

Inomata, S. (2021), New Particle Formation Promoted by OH Reactions during α-Pinene Ozonolysis, *ACS Earth Space Chem.*, *5*(8), 1929-1933, doi:10.1021/acsearthspacechem.1c00142.

Inoue, K.-i., C. Takada, L. Wang, A. Morita & S. Ye (2020), In Situ Monitoring of the Unsaturated Phospholipid Monolayer Oxidation in Ambient Air by HD-SFG Spectroscopy, *J. Phys. Chem. B*, *124*(25), 5246-5250, doi:10.1021/acs.jpcb.0c03408.

Ishiyama, T., A. Morita & T. Miyamae (2011), Surface structure of sulfuric acid solution relevant to sulfate aerosol: molecular dynamics simulation combined with sum frequency generation measurement, *Phys. Chem. Chem. Phys.*, *13*(47), 20965-20973, doi:10.1039/C1CP21920K.

Ishizuka, S., A. Matsugi, T. Hama & S. Enami (2020), Interfacial Water Mediates Oligomerization Pathways of Monoterpene Carbocations, *J. Phys. Chem. Lett.*, *11*(1), 67-74, doi:10.1021/acs.jpclett.9b03110.

Matsuoka, K., Y. Sakamoto, T. Hama, Y. Kajii & S. Enami (2017), Reactive Uptake of Gaseous Sesquiterpenes on Aqueous Surfaces, *J. Phys. Chem. A*, *121*, 810－818, doi:10.1021/acs.jpca.6b11821.

Nagashima, K. et al. (2016), Direct Visualization of Quasi-Liquid Layers on Ice Crystal Surfaces Induced by Hydrogen Chloride Gas, *Cryst. Growth Des.*, *16*(4), 2225-2230, doi:10.1021/acs.cgd.6b00044.

Nagashima, K., G. Sazaki, T. Hama, K.-i. Murata & Y. Furukawa (2018), Uptake Mechanism of Atmospheric Hydrogen Chloride Gas in Ice Crystals via Hydrochloric Acid Droplets, *Cryst. Growth Des.*, *18*(7), 4117-4122, doi:10.1021/acs.cgd.8b00531.

Nagata, Y. et al. (2019), The Surface of Ice under Equilibrium and Nonequilibrium Conditions, *Acc. Chem. Res.*, *52*(4), 1006-1015, doi:10.1021/acs.accounts.8b00615.

Negishi, K. & J.-Y. Kohno (2019), Low-Frequency Raman Scattering in Colliding Benzene Droplets, *J. Phys. Chem. A*, *123*(42), 9158-9165, doi:10.1021/acs.jpca.9b06354.

Nojima, Y., Y. Shioya, H. Torii & S. Yamaguchi (2020), Hydrogen order at the surface of ice Ih revealed by vibrational spectroscopy, *Chem. Commun.*, *56*(33), 4563-4566, doi:10.1039/D0CC00865F.

Nojima, Y., Y. Suzuki & S. Yamaguchi (2017), Weakly Hydrogen-Bonded Water Inside Charged Lipid Monolayer Observed with Heterodyne-Detected Vibrational Sum Frequency Generation Spectroscopy, *J. Phys. Chem. C*, *121*(4), 2173-2180, doi:10.1021/acs.jpcc.6b09229.

Qiu, J., S. Ishizuka, K. Tonokura, A. J. Colussi & S. Enami (2019a), Water Dramatically Accelerates the Decomposition of a-Hydroxyalkyl-Hydroperoxides in Aerosol Particles, *J. Phys. Chem. Lett.*, *10*, 5748-5755.

Qiu, J., S. Ishizuka, K. Tonokura & S. Enami (2019b), Interfacial vs Bulk Ozonolysis of Nerolidol, *Environ. Sci. Technol.*, *53*, 5750－5757, doi:10.1021/acs.est.9b00364.

Qiu, J., Z. Liang, K. Tonokura, A. J. Colussi & S. Enami (2020), Stability of Monoterpene-Derived α- Hydroxyalkyl-Hydroperoxides in Aqueous Organic Media: Relevance to the Fate of Hydroperoxides in Aerosol Particle Phases, *Environ. Sci. Technol.*, *54*(7), 3890-3899, doi:10.1021/acs.est.9b07497.

Sakamoto, Y. & K. Tonokura (2012), Measurements of the Absorption Line Strength of Hydroperoxyl Radical in the n3 Band using a Continuous Wave Quantum Cascade Laser, *J. Phys. Chem. A*, *116*(1), 215-222, doi:10.1021/jp207477n.

Sakamoto, Y., A. Yabushita, M. Kawasaki & S. Enami (2009), Direct Emission of I2 Molecule and IO Radical from the Heterogeneous Reactions of Gaseous Ozone with Aqueous Potassium Iodide Solution, *J. Phys. Chem. A*, *113*(27), 7707-7713, doi:10.1021/jp903486u.

Sazaki, G., S. Zepeda, S. Nakatsubo, E. Yokoyama & Y. Furukawa (2010), Elementary steps at the surface of ice crystals visualized by advanced optical microscopy, *Proc. Natl. Acad. Sci. U. S. A.*, *107*(46), 19702, doi:10.1073/pnas.1008866107.

Sekimoto, K., D. Fukuyama & S. Inomata (2020), Accurate identification of dimers from α-pinene oxidation using high-resolution collision-induced dissociation mass spectrometry, *J. Mass Spectrom.*, *55*(6), e4508, doi:https://doi.org/10.1002/jms.4508.

Shiraiwa, M. et al. (2017), Aerosol health effects from molecular to global scales, *Environ. Sci. Technol.*, *51*(23), 13545-13567.

Suzuki, T. & J.-Y. Kohno (2014), Simultaneous Detection of Images and Raman Spectra of Colliding Droplets: Composition Analysis of Protrusions Emerging during Collisions of Ethanol and Water Droplets, *J. Phys. Chem. B*, *118*(21), 5781-5786,

doi:10.1021/jp503285w.
Tajiri, T. et al. (2013), A Novel Adiabatic-Expansion-Type Cloud Simulation Chamber, *J. Meteor. Soc. Jpn. Ser. II*, *91*(5), 687-704, doi:10.2151/jmsj.2013-509.
Takenaka, N. & H. Bandow (2007), Chemical Kinetics of Reactions in the Unfrozen Solution of Ice, *J. Phys. Chem. A*, *111*(36), 8780-8786, doi:10.1021/jp0738356.
Yajima, R., Y. Sakamoto, S. Inomata & J. Hirokawa (2017), Relative Reactivity Measurements of Stabilized CH2OO, Produced by Ethene Ozonolysis, Toward Acetic Acid and Water Vapor Using Chemical Ionization Mass Spectrometry, *J. Phys. Chem. A*, *121*(34), 6440-6449, doi:10.1021/acs.jpca.7b05065.

Table A-4. Summary table for the topic **Terrestrial ecosystem and atmospheric chemistry**

(Authors: Michihiro Mochida, Akihiko Ito, Kazuhide Matsuda, Akira Tani)

1. Ultimate Goals	✓ To understand the dynamics and exchange of long-lived gases, especially greenhouse gases, on a multi-scale and with high accuracy over the short to long term. To elucidate the mechanisms of changes in the exchange rates and contribute to impact assessment and planning of adaptation and mitigation measures by quantitatively predicting the budget associated with global environmental changes. ✓ To deepen understanding of deposition and exchange processes of atmospheric substances, to model the phenomena, and to improve the prediction accuracy of chemical transport models. ✓ To elucidate the mechanisms of BVOC emissions and factors influencing them at the molecular, cellular, tissue, individual, and community levels, and to elucidate the BVOC-mediated effects on air quality and climate. ✓ To quantify the effects of terrestrial ecosystems on the atmospheric aerosol budget and enable future projections. To understand them in terms of the climatic effects of aerosols and the relationship between climate and vegetation.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ Dramatic increase in the amount of observation data by ground and satellite, and implementation of integrated analysis of greenhouse gases. Japan has contributed with highly accurate atmospheric observation data and budget estimation using atmospheric transport and terrestrial material circulation models. iLEAPS has been implemented, and interdisciplinary research on air-land material exchange has been revitalized in Japan. ✓ In the beginning of this century, flux observations for various chemical components and deposition surfaces were conducted using new technologies, and models were validated and compared. The new technologies have revealed components and surfaces with large uncertainties in deposition rates. ✓ Research on molecular to ecological levels, such as plant-plant communication and environmental response to release, and the development and widespread use of real-time measurement devices have progressed. BVOC research on land modification and urban green space progressed in Japan. ✓ Improved understanding of the formation mechanisms of biogenic secondary organic aerosols (BSOA), including the formation of highly oxidized organic molecules (HOMs) by autoxidation. Advances in bioaerosol research based on molecular tracer analysis and fluorescence measurements. In Japan, contributions were made to laboratory experiments and field observations related to BSOA.
3. Current major gaps	✓ Large discrepancies in model intercomparison results for greenhouse gases. Severe uncertainties in emission inventories as well. Monitoring system needs to be expanded to capture changes associated with events that are difficult to predict. ✓ Discrepancies between theoretical and observed deposition rates, inability to reproduce upward emission phenomena, and contribution to the refinement of transport and deposition simulations for transboundary air pollution control. ✓ It is necessary to measure the amount and concentration of BVOC emissions, to develop satellite observation methods to estimate them, and to develop inexpensive analytical methods. Measurement equipment is expensive, and this is not a field that young researchers can easily tackle. ✓ Challenges in understanding the amount and characteristics of BSOA production and its representation in atmospheric models. Bioaerosol measurement methods are not well established; resources available for BSOA observations are not being utilized. ✓ In understanding the material cycles of atmospheric and terrestrial ecosystems and the impact of their changes on human society, there is a lack of academic knowledge on BVOCs and aerosols, and efforts to link such knowledge to social implementation are lacking.
4. Key questions and hypothesis to drive research and development during 2022-32	✓ The existence of long-term attenuation of CO_2 fertilization effects, the impact of future deforestation and fires on the carbon budget, and the impact of air pollutants on greenhouse gas exchange. More sophisticated observation and modeling methods are required to provide a consistent explanation of the global budget. Efforts are needed to provide scientific data for the Global Stocktake of the Paris Agreement. ✓ Acquisition of highly accurate datasets through strategic observations based on flux observations targeting forests in particular, in collaboration with Asia-Flux and others, will lead to a better understanding of the process and improve the accuracy of model predictions. ✓ Development of new measurement and analysis methods for BVOCs and lower cost analytical equipment. Establishment of an environment for new researchers to enter the field by organizing cross-disciplinary groups and launching projects in atmospheric chemistry, ecology, plant physiology and ecology, and other fields. ✓ To deepen our understanding of the BSOA production process and explain the amount and characteristics of BSOA production; to quantify the impact of BSOA and bioaerosol production on climatic processes and to determine the presence and extent of feedback mechanisms on vegetation. ✓ A comprehensive understanding of interactions (e.g., GPP promotion and CO_2 fixation by nitrogen deposition) across fields (e.g., CO_2, GPP, BVOC, BSOA) will help elucidate undiscovered processes and improve quantification of the material balance.

	✓ To improve quantitative predictability of the dynamics of total organic matter involving terrestrial ecosystems, including the effects of human activities. ✓ We will promote research on atmospheric and terrestrial ecosystems for an integrated understanding of the Earth's surface systems, and based on this, promote social implementation of the same themes.
5. Collaborations	✓ IGAC, GCP, iLEAPS, FLUXNET, iCACGP etc.

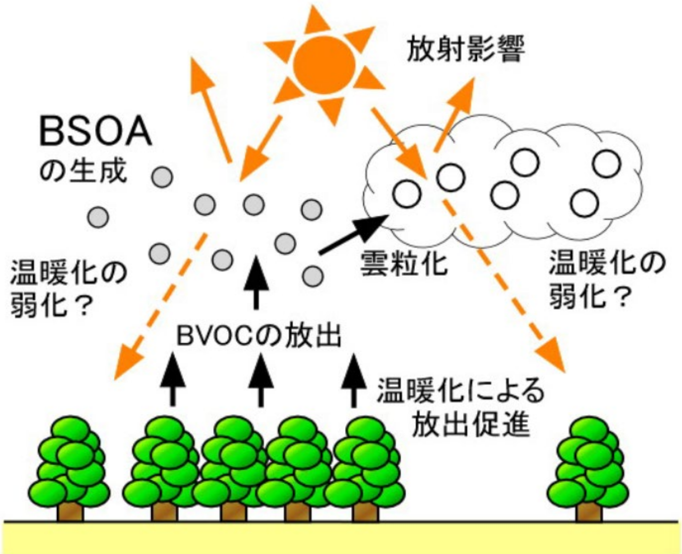


Figure A-4. Negative feedback via the BSOA formation.

Selected References (Terrestrial ecosystem and atmospheric chemistry)

Chandra, N., et al. (2021), Emissions form the oil and gas sectors, coal mining and ruminant farming drive methane growth over the past three decades, *J. Meteor. Soc. Jpn.*, 99, 309–337, doi:10.2151/jmsj.2021-015.

Deng. Y. et al. (2019), Diurnal variation and size dependence of the hygroscopicity of organic aerosol at a forest site in Wakayama, Japan: their relationship to CCN concentrations, *Atmos. Chem. Phys.*, 19, 5889–5903, doi:10.5194/acp-19- 5889-2019.

Fukui, T., K. Kokuryo, T. Baba and A. Kannari (2014), Updating EAGrid2000-Japan emissions inventory based on the recent emission trends, *J. Jpn. Soc. Atmos. Environ.*, 49, 117-125, doi.org/10.11298/taiki.49.117 (in Japanese).

Itahashi, S. et al. (2020), MICS-Asia III: overview of model intercomparison and evaluation of acid deposition over Asia, *Atmos. Chem. Phys.*, 20, 2667–2693, doi:10.5194/acp-20-2667-2020.

Ito, A. (2019), Disequilibrium of terrestrial ecosystem CO_2 budget caused by disturbance-induced emissions and non- CO_2 carbon export flows: a global model assessment, *Earth Syst. Dynam.*, 10, 685–709, doi:10.5194/esd-10-685-2019.

Izuta, T. (Ed.) (2017), Air Pollution Impacts on Plants in East Asia, Springer Tokyo, doi:10.1007/978-4-431-56438-6.

Jung, M., et al. (2020), Scaling carbon fluxes from eddy covariance sites to globe: synthesis and evaluation of the FLUXCOM approach, *Biogeosciences*, 17, 1343–1365, doi:10.5194/bg-17-1343-2020.

Katata, G., K. Matsuda, A. Sorimachi, M. Kajino & K. Takagi (2020), Effects of aerosol dynamics and gas-particle conversion on dry deposition of inorganic reactive nitrogen in a temperate forest. *Atmos. Chem. Phys.*, 20, 4933–4949. doi:10.5194/acp-20-4933-2020.

Kawamiya, M., T. Hajima, K. Tachiiri, S. Watanabe & T. Yokohata (2020), Two decades of Earth system modeling with an emphasis on Model for Interdisciplinary Research on Climate (MIROC), *Prog. Earth Planet. Sci.*, 7, 64, doi:10.1186/s40645-020-00369-5.

Kokubu Y, S. Hara & A. Tani (2020), Mapping seasonal tree canopy cover and leaf area using Worldview-2/3 satellite imagery: A megacity-scale case study in Tokyo urban area, *Remote Sens.*, 12, 1505, doi:10.3390/rs12091505.

Kondo, M., et al. (2020), State of the science in reconciling topdown and bottom-up approaches for terrestrial CO_2 budget, *Global Change Biol.*, 26, 1068–1084, doi:10.1111/gcb.14917.

Kurokawa, J. & T. Ohara (2020), Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3. *Atmos. Chem. Phys.*, 20, 12761–12793. Doi:10.5194/acp-20-12761-2020

Masui N, et al. (2020), Does ozone alter the attractiveness of Japanese white birch leaves to the leaf beetle Agelastica coerulea via changes in biogenic volatile organic compounds (BVOCs): An examination with the Y-tube test, *Forests*, 11, 58, doi:10.3390/f11010058.

Matsuda, K., I. Watanabe, V. Wingpud, P. Theramongkol & T. Ohizumi (2006), Deposition velocity of O_3 and SO_2 in the dry and wet season above a tropical forest in northern Thailand, *Atmos. Environ.*, 40, 7557–7564, doi:10.1016/j.atmosenv.2006.07.003.

Matsuda, K., I. Watanabe, K. Mizukami, S. Ban & A. Takahashi (2015), Dry deposition of $PM_{2.5}$ sulfate above a hilly forest using relaxed eddy accumulation, *Atmos. Environ.*, 107, 255–261, doi:10.1016/j.atmosenv.2015.02.050.

Nakahara, A., K. Takagi, A. Sorimachi, G. Katata & K. Matsuda (2019), Enhancement of dry deposition of $PM_{2.5}$ nitrate in a cool temperate forest. *Atmos. Environ.*, 212, 136−141, doi:10.1016/j.atmosenv.2019.05.053.

Niwa, Y. et al. (2021), Estimation of fire-induced carbon emissions from Equatorial Asia in 2015 using in situ aircraft and ship observations, *Atmos. Chem. Phys.*, 21, 9455−9473, doi:10.5194/acp-21-9455-2021.

Patra, P. K., et al. (2016), Regional methane emission estimation based on observed atmospheric concentrations (2002−2012), *J. Meteor. Soc. Jpn.*, 94, 91−113, doi: 10.2151/jmsj.2016-006.

Sato, K. et al. (2018), Studying volatility from composition, dilution, and heating measurements of secondary organic aerosols formed during α-pinene ozonolysis, *Atmos. Chem. Phys.*, 18, 5455−5466, doi:10.5194/acp-18-5455-2018.

Tani, A. & T. Mochizuki (2021), Review: Exchanges of volatile organic compounds between terrestrial ecosystems and the atmosphere. *J. Agric. Meteorol*., 77, 66−80, doi:10.2480/agrmet.D-20-00025.

Tani, A., S. Tobe & S. Shimizu (2010), Uptake of methacrolein and methyl vinyl ketone by tree saplings and implications for forest atmosphere, *Environ. Sci. Technol.*, 44, 7096− 7101, doi:10.1021/es1017569.

Tsunogai, U. et. al. (2010), Tracing the fate of atmospheric nitrate deposited onto a forest ecosystem in Eastern Asia using Δ17O, *Atmos. Chem. Phys.*, 10, 1809−1820, doi:10.5194/acp-10-1809-2010.

Tsunogai, U., S. Daita, D. D. Komatsu, F. Nakagawa & A. Tanaka (2011), Quantifying nitrate dynamics in an oligotrophic lake using Δ17O, *Biogeosciences*, 8, 687−702, doi:10.5194/bg-8-687-2011.

Yamaga, S. et al. (2021), Trends of sulfur and nitrogen deposition from 2003 to 2017 in Japanese remote areas, *Environ. Pollut.*, 289, 117842, 10.1016/j.envpol.2021.117842.

Yazaki, K., G. I. Arimura & T. Ohnishi (2017), 'Hidden' terpenoids in plants: their biosynthesis, localization and ecological roles, *Plant Cell Physiol.*, 58, 1615−1621, doi:10.1093/pcp/pcx123.

Xu, M., K. Kasahara, A. Sorimachi & K. Matsuda (2021), Nitric acid dry deposition associated with equilibrium shift of ammonium nitrate above a forest by long-term measurement using relaxed eddy accumulation, *Atmos.*
Environ., 256, 118454, doi:10.1016/j.atmosenv.2021.118454.

Zhu. C., K. Kawamura, Y. Fukuda, M. Mochida, & Y. Iwamoto (2016), Fungal spores overwhelm biogenic organic aerosols in a midlatitudinal forest, *Atmos. Chem. Phys.*, 16, 7497−
7506, doi:10.5194/acp-16-7497-2016.

Table A-5. Summary table for the topic **Marine ecosystem and atmospheric chemistry**

(Authors: Akinori Ito, Yuzo Miyazaki, Fumikazu Taketani, Yoko Iwamoto, Yugo Kanaya, Jun Nishioka)

1. Ultimate Goals	✓ Humanity has caused global atmospheric pollution and global warming, and we have entered the era known as the Anthropocene. It is important to understand the essential aspects of the effects of these anthropogenic global environmental changes on atmospheric chemistry and terrestrial-ocean ecosystem interactions, as well as the climate feedbacks caused by these effects, and to evaluate and predict these effects in an integrated manner. Therefore, the ultimate goal is to "clarify the climatic effects of the interaction between atmospheric trace elements and marine biological activities and their impacts on marine ecosystems and their climate feedbacks" during the Anthropocene period by integrating observations, experiments, and numerical modeling studies.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ Integrated studies on marine organisms - atmospheric aerosol chemistry and physical processes - and their effects on cloud condensation nuclei and ice nuclei evolution have been conducted domestically and internationally. ✓ Atmospheric chemistry observation data by ships were accumulated in Japan, mainly in the North Pacific Ocean. ✓ An integrated research system of cross-disciplinary observations and models on the atmosphere-ocean budget of organic matter, iron, and reactive nitrogen was established. ✓ New information on the composition of oceanic atmospheric aerosols was accumulated through offshore observations using advanced measurements and analyses, such as isotope analysis and fluorescence analysis. ✓ In the North Pacific subarctic region, knowledge on the distribution and circulation of iron in the interior of the ocean was accumulated. ✓ The ocean model suggests that combustion-derived iron is more efficient than mineral-derived iron for nutrient fertilization.
3. Current major gaps	✓ The modeling of the oceanic climate and weather systems is still insufficient in terms of a comprehensive understanding of the processes and amounts of organic matter supplied from surface seawater to the atmosphere, and their effects on cloud condensation nuclei and ice nuclei evolution, and cloud precipitation processes. ✓ The understanding of the supply and origin of atmospheric components that affect marine ecosystems in each ocean region is still insufficient. ✓ There is a lack of validation data from integrated shipboard observations synchronizing atmospheric and oceanic parameters and field experiments such as mesocosms in the Arctic Ocean, North Pacific Ocean, Indo-Pacific Ocean, and South Pacific Ocean. ✓ Lack of understanding of interactions between organic matter and nutrients in the ocean-atmosphere boundary layer.
4. Key questions and hypothesis to drive research and development during 2022-32	✓ With global warming, ocean acidification, and changes in nutrient deposition to the ocean, how does the quantity and quality of aerosols and precursors emitted from the ocean to the atmosphere change, and to what extent do positive or negative climate feedbacks occur via their functions such as cloud condensation and ice nucleation? ✓ Quantify the response of marine ecosystems (e.g., primary production) to the supply of various nutrients and inhibitors from the atmosphere to the ocean. ✓ Long-term integrated atmospheric and oceanic shipboard observations and modeling studies of atmospheric and oceanic circulation in the Arctic Ocean, North Pacific Ocean, Indian Ocean, and South Pacific Ocean will be conducted. ✓ We will integrate observations, experiments, and modeling studies of organic matter and nutrients that have been conducted separately, not only in atmospheric chemistry but also in oceanography, and develop them into cross-disciplinary research.
5. Collaborations	✓ The Oceanographic Society of Japan, Meteorological Society of Japan, Geochemical Society of Japan, Aerosol Society of Japan, SOLAS, CATCH, BEPSII, GESAMP, GEOTRACES, IMBeR

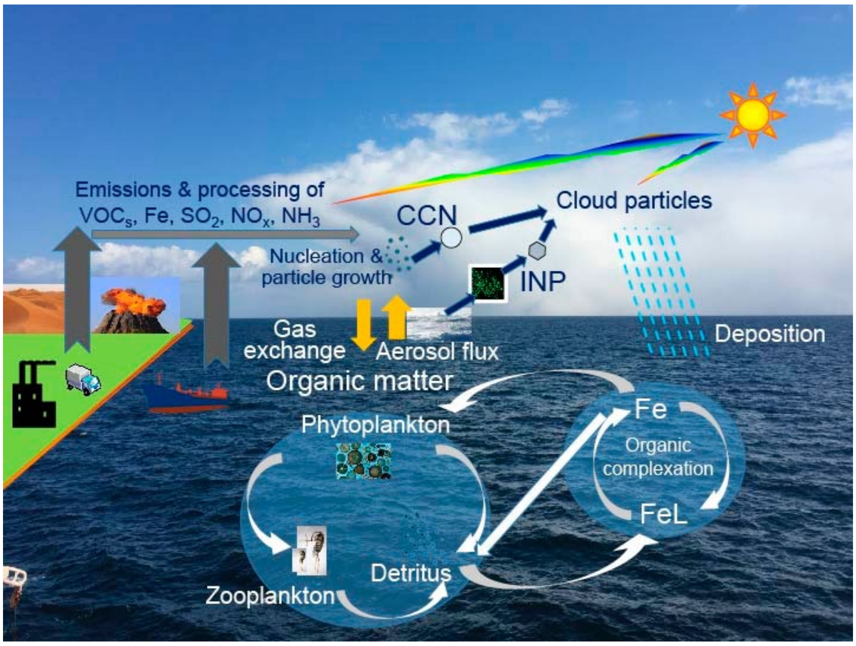


Figure A-5. Schematic diagram of interactions between atmospheric aerosols and marine ecosystems and their climate relevance in the Anthropocene.

Selected References (Marine ecosystem and atmospheric chemistry)

Dobashi, T. et al. (2022), Marine nitrogen fixation as a possible source of atmospheric water-soluble organic nitrogen aerosols in the subtropical North Pacific, submitted

Hamasaki, K. et al. (2019), Microbial community dynamics in sea surface microlayer and sea spray aerosols observed in coastal inlets of Japan, *SOLAS Open Science Conference*.

Inoue, J., Y. Tobo, F. Taketani & K. Sato (2021), Oceanic supply of ice-nucleating particles and its effect on ice cloud formation: A case study in the Arctic Ocean during a cold air outbreak in early winter, *Geophys. Res. Lett.*, 48, e2021GL094646, doi.org/10.1029/2021GL094646.

Itahashi, S., H. Hayami, I. Uno, X. Pan & M. Uematsu (2016), Importance of coarse-mode nitrate produced via sea salt as atmospheric input to East Asian oceans, *Geophys. Res. Lett*., 43, 5483- 5491, doi:10.1002/2016GL068722.

Ito, A. & M. Kawamiya (2010), Potential impact of ocean ecosystem changes due to global warming on marine organic carbon aerosols, *Global Biogeochem. Cycles*, 24, GB1012. doi.org/10.1029/2009GB003559.

Ito, A., G. Lin & J. E. Penner (2014), Reconciling modeled and observed atmospheric deposition of soluble organic nitrogen at coastal locations, *Global Biogeochem. Cycles*, 28, 617-630, doi.org/10.1002/2013GB004721.

Ito, A. et al. (2019), Pyrogenic iron: The missing link to high iron solubility in aerosols, *Sci. Adv.*, 5, eaau7671, doi.org/10.1126/sciadv.aau7671.

Ito, A., Y. Ye, A. Yamamoto, M. Watanabe & M. N. Aita (2020a), Responses of ocean biogeochemistry to atmospheric supply of lithogenic and pyrogenic iron-containing aerosols, *Geol. Mag.*, 157, 741-756, doi.org/10.1017/S0016756819001080.

Ito, A. et al. (2020b), Evaluation of aerosol iron solubility over Australian coastal regions based on inverse modeling: Implications of bushfires on bioaccessible iron concentrations in the Southern Hemisphere, *Prog. Earth Planet. Sci.*, 7, 42, doi.org/10.1186/s40645-020-00357-9.

Ito, A., Y. Ye, C. Baldo & Z. Shi (2021), Ocean fertilization by pyrogenic aerosol iron, *npj Clim. Atmos. Sci.*, 4, 30, doi.org/10.1038/s41612-021-00185-8.

Iwamoto, Y. et al. (2011), Biogeochemical implications of increased mineral particle concentrations in surface waters of the northwestern North Pacific during an Asian dust event, *Geophys. Res. Lett.*, 38, L01604, doi.org/10.1029/2010GL045906.

Iwata, A. & A. Matsuki (2018), Characterization of individual ice residual particles by the single droplet freezing method: a case study in the Asian dust outflow region, *Atmos. Chem. Phys.,* 18, 1785-1804, doi.org/10.5194/acp-18-1785-2018.

Kawai, H. et al. (2019), Significant improvement of cloud representation in the global climate model MRI-ESM2, *Geosci. Model Dev.*, 12, 2875–2897, doi.org/10.5194/gmd- 12-2875-2019.

Kawamiya, M., T. Hajima, K. Tachiiri, S. Watanabe & T. Yokohata (2020), Two decades of Earth system modeling with an emphasis on Model for Interdisciplinary Research on Climate (MIROC). *Prog. Earth Planet. Sci.*, 7, 64, doi.org/10.1186/s40645-020-00369-5.

Kawana, K., K. Matsumoto, F. Taketani, T. Miyakawa & Y. Kanaya (2021), Fluorescent biological aerosol particles over the central Pacific Ocean: covariation with ocean surface biological activity indicators, *Atmos. Chem. Phys*. 21, 15969-15983, doi.org/10.5194/acp-21-15969-2021.

Kurisu, M., K. Sakata, M. Uematsu, A. Ito & Y. Takahashi (2021), Contribution of combustion Fe in marine aerosols over the northwestern Pacific estimated by Fe stable isotope ratios, *Atmos. Chem. Phys.*, 21, 16027−16050, doi.org/10.5194/acp-21-16027-2021.

Kurokawa, J. & T. Ohara (2020), Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3, *Atmos. Chem. Phys.,* 20, 12761-12793, doi.org/10.5194/acp-20-12761-2020.

Maki, T. et al. (2018), Long-range-transported bioaerosols captured in snow cover on Mount Tateyama, Japan: impacts of Asian-dust events on airborne bacterial dynamics relating to ice-nucleation activities, *Atmos. Chem. Phys.*, 18, 8155-8171, doi.org/10.5194/acp-18-8155-2018.

Misumi, K. et al. (2021), Slowly sinking particles underlie dissolved iron transport across the Pacific Ocean, *Glob. Biogeochem. Cycle*, 21115, doi.org/10.1029/2020GB006823.

Miyakawa, T. et al. (2015), Ground-based measurement of fluorescent aerosol particles in Tokyo in the spring of 2013: Potential Impacts of non-biological materials on autofluorescence measurements of airborne particles, *J. Geophys. Res. Atmos.* 120, 1171-1185, doi:10.1002/2014JD022189.

Miyakawa, T. et al. (2022), Shipborne measurements of aerosol particles over the Southern Ocean in the late Austral summer of 2017: Insights into cloud formation, in preparation.

Miyazaki, Y. et al. (2018), Chemical transfer of dissolved organic matter from surface seawater to sea spray water soluble organic aerosol in the marine atmosphere, *Sci. Rep.*, 8, 14861, doi:10.1038/s41598-018-32864-7.

Miyazaki, Y. et al. (2020), New index of organic mass enrichment in sea spray aerosols linked with senescent
status in marine phytoplankton, *Sci. Rep.*, 10, 17042, doi:10.1038/s41598-020-73718-5.

Moteki, N. et al. (2017), Anthropogenic iron oxide aerosols enhance atmospheric heating, *Nat. Commun.*, 8, 15329, doi.org/10.1038/ncomms15329.

Moteki, N. (2020), Capabilities and limitations of the single particle extinction and scattering method for estimating the complex refractive index and size-distribution of spherical and non-spherical submicron particles, *J. Quant. Spectrosc. Radiat. Transfer*, 243, 106811, doi.org/10.1016/j.jqsrt.2019.106811.

Moteki, N. (2021), Measuring the complex forward-scattering amplitude of single particles by self-reference interferometry: CAS-v1 protocol, *Opt. Express*, 29(13), 20688-20714, doi.org/10.1364/OE.423175.

Myriokefalitakis, S. et al. (2018), Reviews and syntheses: the GESAMP atmospheric iron deposition model intercomparison study, *Biogeosciences*, 15, 6659-6684, doi.org/10.5194/bg-15-6659-2018.

Nakane, M., T. Ajioka & Y. Yamashita (2017), Distribution and Sources of Dissolved Black Carbon in Surface Waters of the Chukchi Sea, Bering Sea, and the North Pacific Ocean, *Front. Earth Sci.*, 5, 34, doi: 10.3389/feart.2017.00034.

Nishino, S. et al. (2011), Enhancement/reduction of biological pump depends on ocean circulation in the sea-ice reduction regions of the Arctic Ocean, *J. Oceanogr.*, 67, 305-314, doi.org/10.1007/s10872-011-0030-7.

Nishioka, J. et al. (2021), A review: iron and nutrient supply in the subarctic Pacific and its impact on phytoplankton production, *J. Oceanogr.*, 77, 561-587, doi.org/10.1007/s10872-021-00606-5.

Takemura, T., T. Nakajima, A. Higurashi, S. Ohta, & N. Sugimoto (2003), Aerosol distributions and radiative forcing over the Asian Pacific region simulated by Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS), *J. Geophys. Res.*, 108, 8659, doi.org/10.1029/2002JD003210.

Taketani, F. et al. (2013), Measurement of fluorescence spectra from atmospheric single submicron particle using laser-induced fluorescence technique, *Journal of Aerosol Science*, 58, 1-8, doi.org/10.1016/j.jaerosci.2012.12.002.

Taketani, F. et al. (2018), Seasonal response of north western Pacific marine ecosystems to deposition of atmospheric inorganic nitrogen compounds from East Asia, *Sci. Rep.*, 8(1), 9324, doi.org/10.1038/s41598-018-27523-w.

Tanimoto, H., S. Kameyama, T. Iwata, S. Inomata & Y. Omori (2014), Measurement of air-sea exchange of dimethyl sulfide and acetone by PTR-MS coupled with gradient flux technique, *Environ. Sci. Technol*. 2014, 48, 1, 526–533, doi.org/10.1021/es4032562.

Tobo, Y. (2016), An improved approach for measuring immersion freezing in large droplets over a wide
temperature range, *Sci. Rep.*, 6, 32930, doi:10.1038/srep32930.

Tobo, Y. et al. (2020), Seasonal trends of atmospheric ice nucleating particles over Tokyo, *J. Geophys. Res. Atmos.*, 125, e2020JD033658, doi.org/10.1029/2020JD033658.

Uetake, J. et al. (2020), Airborne bacteria confirm the pristine nature of the Southern Ocean boundary layer, *Proc. Natl. Acad. Sci. USA.*, 117 (24), 13275-13282, doi.org/ 10.1073/pnas.2000134117.

Yoshizue, M. et al. (2019), Individual particle analysis of marine aerosols collected during the north-south transect cruise in the Pacific Ocean and its marginal seas, *J. Oceanogr.*, 75, 513-524, doi.org/10.1007/s10872-019-00519-4.

Zhu, C. et al. (2020), Characterization of microplastics on filter substrates based on hyperspectral imaging: laboratory assessments, *Environ. Pollut.*, 263, 114296, doi.org/10.1016/j.envpol.2020.114296.

Table A-6. Summary table for the topic **Aerosol-radiation/cloud interactions**

(Authors: Hitoshi Matsui, Sho Ohata, Yutaka Tobo, Atsushi Matsuki, Syuichi Itahashi, Naga Oshima, Kentaro Suzuki, Yousuke Sato)

1. Ultimate Goals	✓ The number concentration of aerosols by particle size and total mass concentration by aerosol composition should be known and predicted. ✓ The aerosol particle and the total aerosol as a whole should be characterized and predicted in terms of optical properties, hygroscopicity, cloud condensation nucleus properties, ice nucleating properties, and so on. ✓ The aerosol-radiation interaction and aerosol-cloud interaction processes should be well represented using numerical models validated and constrained by in-situ and satellite observations, to achieve reliable climate and environmental impact predictions.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ The aerosol composition and physicochemical properties (e.g., hygroscopicity, cloud condensation nucleus properties, ice crystal nucleating properties, etc.) have been measured and laboratory experiments and atmospheric observation data of various aerosols have been accumulated worldwide. Correspondingly, numerical models have been improved to represent aerosol properties such as mixing state, hygroscopicity, and cloud condensation nucleation properties, as well as processes such as new particle formation, secondary organic aerosol formation, and wet deposition. In Japan, observations and modeling studies have accumulated knowledge on aerosol emission, transport, chemical transformation, deposition, and other processes, taking advantage of the country's geographical location downstream from the Asian continent, and have improved estimates of the effects of transboundary air pollution and radiative forcing of aerosols. ✓ Satellite observations, general circulation climate models, and global cloud-resolving models have developed in an interactive manner, and our understanding of aerosol-cloud interactions has advanced. In satellite observations, the combined use of active and passive sensors has advanced our observational understanding of cloud response to aerosol perturbations. By utilizing the world's most powerful large computers, such as the K computer and the Fugaku computer, Japan has led the research on aerosol-cloud interactions using global cloud-resolving models.
3. Current major gaps	✓ In the latest IPCC 6th Assessment, the range of uncertainties in the estimates of aerosol-radiation interactions and radiative forcing from aerosol-cloud interactions has not decreased since the 5th Assessment, and the variability in the estimates among climate models remains large. The variability in estimates among climate models is still significant. Estimates of the physico-chemical properties and spatio-temporal distribution of aerosols, which determine these radiative forcings, also continue to show large inter-model variability. ✓ The temporal and spatial distribution of aerosols, especially in the polar regions, the open ocean, and the upper troposphere, is not well understood and reproduced by numerical models. The aerosol dynamics in the past based on ice core analysis and the validation of numerical models are also insufficient. ✓ The understanding of the atmospheric abundance and dynamics of aerosols that act as ice nucleating particles (INPs) and the mechanisms that form ice crystals is inadequate. ✓ The long-term monitoring stations in Asia, South America, Africa, Oceania, and Siberia are relatively insufficient compared to those in Europe. ✓ Understanding of the response of cloud/precipitation systems to aerosol perturbations is inadequate. In particular, it is necessary to understand the response of clouds including ice at the elementary process level. ✓ Model representations of aerosol, cloud, and precipitation processes are not well constrained by observations.
4. Key questions and hypothesis to drive research and development during 2022-32	✓ Can the uncertainty ranges in the estimation of radiative forcing from aerosol-radiation interactions and aerosol-cloud interactions be reduced by accumulating field and satellite observation data, integrating observations and numerical models, and refining and increasing the resolution of numerical models? ✓ Can the knowledge of aerosol processes (mixing state, formation process and hygroscopic properties of organic aerosols, transformation process of black carbon, emission and formation process of brown carbon, new particle formation, etc.) obtained from laboratory experiments and observations be appropriately incorporated into the numerical model? ✓ What kind of surface structure of the INP induces the formation of ice crystals? Can long-term observations of INP at multiple locations clarify the dynamics of INP? Can we develop new measurement techniques for this purpose? ✓ Can we establish a sustainable long-term observation network of aerosol optical properties, particle size distribution, cloud condensation nuclei, INP, and chemical composition in East Asia, including Japan, in addition to ground-based remote sensing observations? ✓ Can we quantitatively predict future changes in air pollutants? Can the project contribute to the development of emission scenarios that enable us to address both air pollution and climate change? ✓ Can we introduce new elements into the numerical model system that are not fully considered in most existing aerosol models (e.g., ice nucleating properties, composition of mineral dust, bioaerosols, etc.) and clarify their spatio-temporal distribution and radiative forcing? ✓ Can we estimate and evaluate the interaction between aerosols and climate more quantitatively and

	comprehensively, including the response of not only anthropogenic aerosols but also naturally occurring aerosols to climate change? ✓ Can a new satellite observation mission with simultaneous cloud radar, precipitation radar, and lidar elucidate the interactions between microphysical and dynamical processes that complicate the cloud response to aerosols? How can this combined observational information be used to provide observational diagnostics of important elementary processes in aerosols, clouds, and precipitation? ✓ In general circulation climate models (GCMs), can we elucidate the effects of aerosols on mixed phase clouds, including ice, and the resulting radiative effects and interactions between convective clouds and aerosols? Can global cloud-resolving models (GCRMs) reproduce past climate conditions and link them to climate studies, and can aerosol-cloud interactions be studied in global LES models in a seamless manner that can be quickly linked to GCRMs? ✓ How can the representation of aerosol, cloud, and precipitation processes in these models be constrained and refined based on satellite observations? ✓ How can we achieve both high accuracy and high speed in modeling elemental processes by using machine learning?
5. Collaborations	✓ Meteorological Society of Japan, The Japanese Society of Snow and Ice, Japan Geoscience Union, Japan Association of Aerosol Science and Technology, IGAC, ESA-JAXA EarthCARE satellite mission, NASA AOS satellite mission

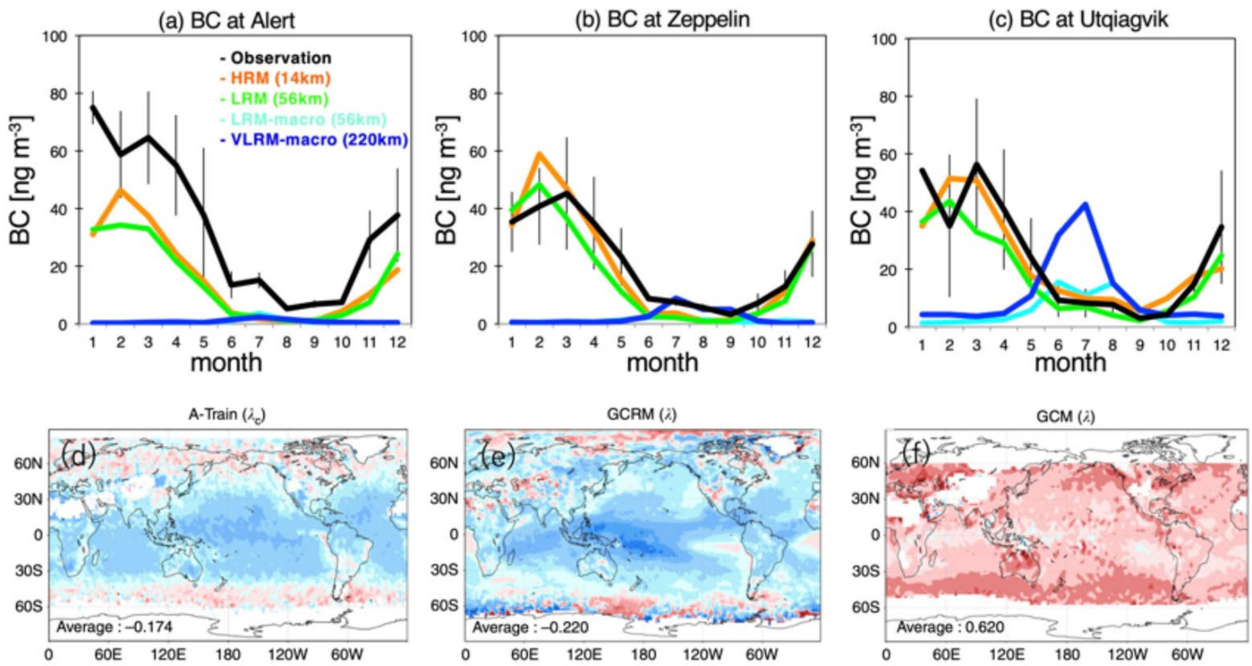


Figure A-6. (a-c) Seasonal change in the observed and modeled black carbon mass concentrations in the Arctic. (d-f) Susceptibility of cloud increase per aerosol increase (A-Train, NICAM, GCM(MIROC)). (adopted from Sato et al. (2016, 2018) and Goto et al. (2020).

Selected References (Aerosol-radiation/cloud interactions)

Aikawa, M. et al. (2010), Significant geographic gradients in particulate sulfate over Japan determined from multiple-site measurements and a chemical transport model: Impacts of transboundary pollution from the Asian continent, *Atmos. Environ.*, 44. 381-391, doi:10.1016/j.atmosenv.2019.10.025.

AMAP, (2021), Impacts of Short-lived Climate Forcers on Arctic Climate, Air Quality, and Human Health. Summary for Policy-makers. Arctic Monitoring and Assessment Programme (AMAP), Tromso, Norway. 20 pp

Aoki, T. et al. (2011), Physically based snow albedo model for calculating broadband albedos and the solar heating profile in snowpack for general circulation models. *J. Geophys. Res. Atmos.*, *116*, D11114, doi:10.1029/2010JD015507.

Aoki, T., S. Matoba, J. Uetake, N. Takeuchi & H. Motoyama (2014), Field activities of the "Snow Impurity and Glacial Microbe effects on abrupt warming in the Arctic" (SIGMA) Project in Greenland in 2011-2013. *Bull. Glaciol. Res.,* 32, 3-20, doi:

10.5331/bgr.32.3.
Chen, Q., F. Ikemori & M. Mochida (2016), Light absorption and excitation-emission fluorescence of urban organic aerosol components and their relationship to chemical structure, *Environ. Sci. Technol.*, *50*, 10859−10868, doi:10.1021/acs.est.6b02541.
Eguchi, K. et al. (2011), Modulation of cloud droplets and radiation over the north Pacific by sulfate aerosol erupted from mount Kilauea. *SOLA*, 7, 077-080, doi:10.2151/sola.2011-020.
Gen, M., et al. (2019), Electrospray surface-enhanced Raman spectroscopy (ES-SERS) for studying organic coatings of atmospheric aerosol particles. *Aerosol. Sci. Tech.*, 53(7), 760-770, doi.org/10.1080/02786826.2019.1597964.
Glis, J. et al. (2021), AeroCom phase III multi-model evaluation of the aerosol life cycle and optical properties using ground and space-based remote sensing as well as surface in situ observations. *Atmos. Chem. Phys.*, 21, 87−128, doi:10.5194/acp-21-87-2021.
Goto, D. et al. (2020), Global aerosol simulations using NICAM.16 on a 14 km grid spacing for a climate study:
improved and remaining issues relative to a lowerresolution model. *Geosci. Model Dev.*, 13(8), 3731−3768.
Hara, K. et al. (2021), Characterization of aerosol number size distributions and their effect on cloud properties at Syowa Station, Antarctica, *Atmos. Chem. Phys.*, 21, 12155−12172, doi.org/10.5194/acp-21-12155-2021.
Hattori, S. et al. (2021), Isotopic evidence for acidity-driven enhancement of sulfate formation after SO_2 emission control, *Sci. Adv.* 7, 19, eabd4610, doi:/10.1126/sciadv.abd4610.
Higurashi, A. & T. Nakajima (1999), Development of a two-channel aerosol retrieval algorithm on a global scale using NOAA AVHRR. *J. Atmos. Sci.*, 56, 924-941.
Iizuka, Y. et al., (2018), A 60 year record of atmospheric aerosol depositions preserved in a high-accumulation dome ice core, Southeast Greenland. *J. Geophys. Res. Atmos.*, 123, doi:/10.1002/2017JD026733.
Ikeda, K. & H. Tanimoto (2015), Exceedances of air quality standard level of $PM_{2.5}$ in Japan caused by Siberian wildfires, *Env. Res. Lett.,* 10, 105001, doi: 10.1088/1748- 9326/10/10/105001.
Inoue, J., Y. Tobo, F. Taketani & K. Sato (2021), Oceanic supply of ice-nucleating particles and its effect on ice cloud formation: A case study in the Arctic Ocean during a cold air outbreak in early winter, *Geophys. Res. Lett.*, 48, e2021GL094646.
Isono, K., M. Komabayashi & A. Ono (1959), The nature and the origin of ice nuclei in the Atmosphere, *J. Meteorol. Soc. Jpn.*, 37, 211-233.
Isono, K. & T. Tanaka (1966), Sudden increase of ice nucleus concentration associated with thunderstorm, *J. Meteorol. Soc. Jpn.*, 44, 255-259.
Itahashi et al. (2018), A 15-year record (2001-2015) of the ratio of nitrate to non-sea-salt sulfate in precipitation over East Asia, *Atmos. Chem. Phys.*, 18. 2835-2852, doi:10.5194/acp-18-2835-2018.
Ito, A., G. Lin & J. E. Penner (2018), Radiative forcing by light absorbing aerosols of pyrogenetic iron oxides. *Sci. Rep.* 8, 7347, doi:10.1038/s41598-018-25756-3.
Ito, A. et al. (2019), Pyrogenic iron: The missing link to high iron solubility in aerosols, *Sci. Adv.,* 5, eaau7671, doi:10.1126/sciadv.aau7671.
Ito, A. et al. (2021), Ocean fertilization by pyrogenic aerosol iron, *npj Clim. Atmos. Sci.,* 4, doi:10.1038/s41612-021-00185-8.
Iwamoto, Y. et al. (2021), Aerosol-Cloud Interaction at the Summit of Mt. Fuji, Japan: Factors Influencing Cloud
Droplet Number Concentrations, *Appl. Sci.*, 11, 8439, doi.org/10.3390/app11188439.
Jones, C. D., et al. (2021), The climate response to emissions reductions due to COVID-19: Initial results from CovidMIP. Geophys. Res. Lett., 48, e2020GL091883, doi:10.1029/2020GL091883.
Kanaya, Y. et al. (2020), Rapid reduction in black carbon emissions from China: evidence from 2009–2019 observations on Fukue Island, Japan, *Atmos. Chem. Phys.,* 20, 6339–6356, doi:/10.5194/acp-20-6339-2020.
Kaneyasu, N. et al. (2014), Impact of long-range transport of aerosols on the PM2. 5 composition at a major metropolitan area in the northern Kyushu area of Japan. Atmos. Environ., 97, 416-425.
Kawai, K., H. Matsui & Y. Tobo (2021), High potential of Asian dust to act as ice nucleating particles in mixed-phase clouds simulated with a global aerosol-climate model, *J. Geophys. Res. Atmos.*, 126, e2020JD034263.
Kawamoto, K., T. Nakajima & T. Y. Nakajima (2001), A global determination of cloud microphysics with AVHRR remote sensing. *J. Climate*, 14, 2054-2068.
Kinase, T. et al. (2020), Concentrations and size distributions of black carbon in the surface snow of eastern Antarctica in 2011, *J. Geophys. Res. Atmos.*, 125, e2019JD030737, doi:/10.1029/2019JD030737.
Kobayashi, H. et al. (2014), Development of a polarization optical particle counter capable of aerosol type classification, *Atmos. Environ.* 97, 486–492, doi:10.1016/j.atmosenv.2014.05.006.
Kondo, Y., et al. (2009), Stabilization of the mass absorption cross section of black carbon for filter-based absorption photometry by the use of a heated inlet, *Aerosol Sci. Technol.,* 43, 741−756, doi:10.1080/02786820902889879.
Kondo, Y. et al. (2011a), Consistency and traceability of black carbon measurements made by laser-induced
incandescence, thermal-optical transmittance, and filter-based photo-absorption techniques. *Aerosol Sci. Technol.*, 45, 295–312, doi:10.1080/02786826.2010.533215.
Kondo, Y. et al. (2012), Reduction of black carbon aerosols in Tokyo: Comparison of real-time observations with emission estimates, *Atmos. Environ.*, *54*, 242–249, doi:10.1016/j.atmosenv.2012.02.003.
Kondo, Y. et al. (2016), Effects of wet deposition on the abundance and size distribution of black carbon in East Asia, *J. Geophys. Res. Atmos.*, 121, 4691–4712, doi:10.1002/2015JD024479.
Kurisu, M., Y. Takahashi, T. Iizuka & M. Uematsu (2016), Very low isotope ratio of iron in fine aerosols related to its contribution to the surface ocean, *J. Geophys. Res. Atmos.,*121, 11119−11136, doi:10.1002/ 2016JD024957. Kurisu, M., K. Sakata, M. Uematsu, A. Ito & Y. Takahashi (2021), Contribution of combustion Fe in marine aerosols over the northwestern Pacific estimated by Fe stable isotope ratios, *Atmos. Chem. Phys.,* 21, 16027−16050, doi:10.5194/acp-21-16027-2021.

Kurokawa, J. et al. (2013), Emissions of air pollutants and greenhouse gases over Asian regions during 2000-2008: Regional Emission inventory in ASia (REAS) version 2, *Atmos. Chem. Phys.*, 13. 11019-11058, doi:10.5194/acp-13-11019-2013.

Kurokawa, J. & T. Ohara (2020), Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3, *Atmos. Chem. Phys.*, 20. 12761-12793, doi:10.5194/acp-20-12761-2020.

Liu, M. & H. Matsui (2021), Improved simulations of global black carbon distributions by modifying wet scavenging processes in convective and mixed-phase clouds, *J. Geophys. Res. Atmos.,* 126, e2020JD033890, doi:10.1029/2020JD033890.

Lund, M. T. et al. (2018). Concentrations and radiative forcing of anthropogenic aerosols from 1750 to 2014 simulated with the Oslo CTM3 and CEDS emission inventory, *Geosci. Model Dev.*, 11, 4909–4931, doi:10.5194/gmd-11-4909-2018.

Matsui, H. (2017), Development of a global aerosol model using a two-dimensional sectional method: 1. Model design, *J. Adv. Model. Earth Syst.,* 9, 1921–1947, doi:10.1002/2017MS000936.

Matsui, H., D. S. Hamilton & N. M. Mahowald (2018a), Black carbon radiative effects highly sensitive to emitted particle size when resolving mixing-state diversity, *Nat. Commun.*, 9, 3446, doi:10.1038/s41467-018-05635-1.

Matsui, H. & M. Liu (2021), Importance of supersaturation in Arctic black carbon simulations, *J. Climate,* 34, 7843－7856, doi:10.1175/JCLI-D-20-0994.1.

Matsui, H. & N. Mahowald (2017), Development of a global aerosol model using a two-dimensional sectional method: 2. Evaluation and sensitivity simulations, *J. Adv. Model. Earth Syst.*, 9, 1887–1920, doi:10.1002/2017MS000937.

Matsui, H. & N. Moteki (2020), High sensitivity of Arctic black carbon radiative effects to subgrid vertical velocity in aerosol activation, *Geophys. Res. Lett.,* 47, e2020GL088978, doi:10.1029/2020GL088978.

Matsui, H. et al. (2011a), Impact of new particle formation on the concentrations of aerosols and cloud condensation nuclei around Beijing, *J. Geophys. Res.,* 116, D19208, doi:10.1029/2011JD016025.

Matsui, H. et al. (2011b), Seasonal variation of the transport of black carbon aerosol from the Asian continent to the Arctic during the ARCTAS aircraft campaign. *J. Geophys. Res.,* 116, D05202, doi:10.1029/2010JD015067.

Matsui, H. et al. (2014), Volatility basis-set approach simulation of organic aerosol formation in East Asia: implications for anthropogenic–biogenic interaction and controllable amounts, *Atmos. Chem. Phys.*, 14, 9513－9535, doi:10.5194/acp-14-9513-2014.

Matsui, H. et al. (2018b), Anthropogenic combustion iron as a complex climate forcer, *Nat. Commun.,* 9, 1593, doi:10.1038/s41467-018-03997-0.

Michibata, T., K. Suzuki, Y. Sato & T. Takemura (2016), The source of discrepancies in aerosol-cloud-precipitation interactions between GCM and A-Train retrievals. *Atmos. Chem. Phys.*, 16, 15413-15424, doi:10.5194/acp-16-15413-2016.

Michibata, T., K. Suzuki, M. Sekiguchi & T. Takemura (2019), Prognostic precipitation in the MIROC6-SPRINTARS GCM: Description and evaluation against satellite observations. *J. Adv. Mod. Earth Sys.*, 11, doi:10.1029/2018MS001596.

Michibata, T. & K. Suzuki (2020), Reconciling compensating errors between precipitation constraints and the energy budget in a climate model. *Geophys. Res. Lett.*, 47, e2020GL088340, doi:10.1029/2020GL088340.

Michibata, T., K. Suzuki & T. Takemura (2020), Snow-induced buffering in aerosol-cloud interactions. *Atmos. Chem. Phys.*, 20, 13771-13780, doi:10.5194/acp-20-13771-2020.

Mitchell, J. F. B., T. C. Johns, J. M. Gregory & S. F. B. Tett (1995), Climate response to increasing levels of greenhouse gases and sulphate aerosols. *Nature*, 376, 501-504.

Miyakawa, T. et al. (2016). Emission regulations altered the concentrations, origin, and formation of carbonaceous aerosols in the Tokyo metropolitan area. *Aerosol Air Qual. Res.*, 16, 1603–1614. doi:10.4209/aaqr.2015.11.0624.

Mori, T. et al. (2020a), Changes in black carbon and $PM_{2.5}$ in Tokyo in 2003–2017. *Proc. Jpn. Acad. Ser. B*, 96, 122-129, doi: 10.2183/pjab.96.010.

Mori, T. et al. (2020b), Seasonal variation of wet deposition of black carbon in Arctic Alaska. *J. Geophys. Res. Atmos.*, 125, e2019JD032240, doi:10.1029/2019JD032240.

Mori, T. et al. (2021), Seasonal variation of wet deposition of black carbon at Ny-Alesund, Svalbard, *J. Geophys. Res. Atmos.,* 126, e2020JD034110. doi:/10.1029/2020JD034110.

Moteki, N. (2020), Capabilities and limitations of the single particle extinction and scattering method for estimating the complex refractive index and size-distribution of spherical and non-spherical submicron particles, *J. Quant. Spectrosc. Ra.*, 243, 106811, doi:10.1016/j.jqsrt.2019.106811.

Moteki, N. (2021), Measuring the complex forward-scattering amplitude of single particles by self-reference interferometry: CAS-v1 protocol, *Opt. Express,* 29, 20688- 20714, doi:10.1364/OE.423175.

Moteki, N. & Y. Kondo (2007), Effects of mixing state on black carbon measurements by laser-induced incandescence, *Aerosol Sci. Technol.,* 41, 398-417, doi:10.1080/02786820701199728.

Moteki, N. & Y. Kondo (2010), Dependence of laser-induced incandescence on physical properties of black carbon aerosols: Measurements and theoretical interpretation. *Aerosol Sci. Technol.*, 44, 663–675. doi:10.1080/02786826.2010.484450.

Moteki, N., Y. Kondo & S. Nakamura (2010), Method to measure refractive indices of small nonspherical particles: Application to black carbon particles. *J. Aerosol Sci.*, 41, 513–521, doi:10.1016/j.jaerosci.2010.02.013.

Moteki, N., T. Mori, H. Matsui, & S. Ohata (2019), Observational constraint of in-cloud supersaturation for simulations of aerosol rainout in atmospheric models, *npj Clim. Atmos. Sci.*, 2:6, doi:10.1038/s41612-019-0063-y.

Moteki, N. et al. (2007). Evolution of mixing state of black carbon particles: Aircraft measurements over the western Pacific in March 2004. *Geophys. Res. Lett.*, 34, L11803, doi:10.1029/2006GL028943.

Moteki, N. et al. (2012), Size dependence of wet removal of black carbon aerosols during transport from the boundary layer to the free troposphere. *Geophys. Res. Lett.*, 39, L13802, doi:10.1029/2012GL052034.

Moteki, N. et al. (2017), Anthropogenic iron oxide aerosols enhance atmospheric heating. *Nat. Commun.* 8,

15329,doi:10.1038/ncomms15329.
Nakajima, T., A. Higurashi, K. Kawamoto & J. E. Penner (2001), A possible correlation between satellite-derived cloud and aerosol microphysical parameters. *Geophys. Res. Lett.*, 28, 1171-1174.
Nakajima, T. Y., H. Masunaga & T. Nakajima (2009), Near global scale retrieval of the optical and microphysical properties of clouds from the Midori-II GLI and AMSR data. *J. Rem. Sens. Soc. Japan*, 29, 29-39.
Nakajima, T. Y. & T. Nakajima (1995), Wide-area determination of cloud microphysical properties from NOAA AVHRR measurements for FIRE and ASTEX regions. *J. Atmos. Sci.*, 52, 4043-4059.
Nakajima, T. Y., K. Suzuki & G. L. Stephens (2010), Droplet growth in warm water clouds observed by the A-Train. Part II: A multi sensor view. *J. Atmos. Sci.*, 67, 1897-1907.
Nakajima, T. et al. (2020a), A development of reduction scenarios of the short-lived climate pollutants (SLCPs) for mitigating global warming and environmental problems, *Progress in Earth and Planetary Science*, 7:33. 21p, doi:10.1186/s40645-020-00351-1.
Nakajima, T. et al. (2020b), An overview of and issues with sky radiometer technology and SKYNET, *Atmos. Meas. Tech.*, 13, 4195–4218, doi.org/10.5194/amt-13-4195-2020.
Nakayama, T., K. Sato, M. Tsuge, T. Imamura & Y. Matsumi (2015), Complex refractive index of secondary organic aerosol generated from isoprene/NOx photooxidation in the presence and absence of SO_2, *J. Geophys. Res. Atmos.*, 120, 7777–7787, doi:10.1002/2015JD023522.
Nakayama, T. et al. (2010), Laboratory studies on optical properties of secondary organic aerosols generated during the photooxidation of toluene and the ozonolysis of α-pinene, *J. Geophys. Res.*, 115, D24204, doi:10.1029/2010JD014387.
Nakayama, T. et al. (2013), Wavelength and NOx dependent complex refractive index of SOAs generated from the photooxidation of toluene, *Atmos. Chem. Phys.*, 13, 531–545, doi:10.5194/acp-13-531-2013.
Ohata, S., N. Moteki, T. Mori, M. Koike & Y. Kondo (2016), A key process controlling the wet removal of aerosols: new observational evidence. *Sci. Rep.*, *6*, 34113.doi:10.1038/srep34113.
Ohata, S. et al. (2021a). Arctic black carbon during PAMARCMiP 2018 and previous aircraft experiments in
spring. *Atmos. Chem. Phys.*, 21, 15861–15881, doi:10.5194/acp-21-15861-2021.
Ohata, S. et al. (2021b). Estimates of mass absorption cross sections of black carbon for filter-based absorption photometers in the Arctic. *Atmos. Meas. Tech.*, 14, 6723–6748. doi:10.5194/amt-14-6723-2021.
Orikasa, N. et al. (2020), Seasonal variations of atmospheric aerosol particles focused on cloud condensation nuclei and ice nucleating particles from ground-based observations in Tsukuba, Japan, *SOLA*, 16, 212-219.
Oshima, N. & M. Koike (2013) Development of a parameterization of black carbon aging for use in general circulation models, *Geosci. Model Dev.* 6, 263–282, doi:10.5194/gmd-6-263-2013.
Oshima, N., M. Koike, Y. Zhang & Y. Kondo (2009a), Aging of black carbon in outflow from anthropogenic sources using a mixing state resolved model: 2. Aerosol optical properties and cloud condensation nuclei activities, *J. Geophys. Res.,* 114, D18202, doi:10.1029/2008JD011681.
Oshima, N. et al. (2009b), Aging of black carbon in outflow from anthropogenic sources using a mixing state resoled model: Model development and evaluation, *J. Geophys. Res.,* 114, D06210, doi:10.1029/2008JD010680.
Oshima, N. et al. (2012), Wet removal of black carbon in Asian outflow: Aerosol Radiative Forcing in East Asia (AFORCE) aircraft campaign, *J. Geophys. Res.,* 117, D03204, doi:10.1029/2011JD016552.
Oshima, N. et al. (2020), Global and Arctic effective radiative forcing of anthropogenic gases and aerosols in MRIESM2.0. *Progress in Earth and Planetary Science*, 7, 38. doi:/10.1186/s40645-020-00348-w.
Pan, X. et al. (2016), Polarization properties of aerosol particles over western Japan: classification, seasonal variation, and implications for air quality, *Atmos. Chem. Phys.* 16, 9863– 9873, doi:10.5194/acp-16-9863-2016.
Sato, Y. & K. Suzuki (2019), How do aerosols affect cloudiness? *Science*, 363, 580-581, doi:10.1126/science.aaw3720.
Sato, Y. et al. (2016), Unrealistically pristine air in the Arctic produced by current global scale models. *Sci. Rep.*, 6, 26561. doi:10.1038/srep26561.
Sato, Y. et al. (2018), Aerosol effects on cloud water amounts were successfully simulated by a global cloud-system resolving model. *Nat. Commun.*, 9(1), 985. doi:10.1038/s41467-018-03379-6.
Satoh, M. et al. (2014), The Non-hydrostatic Icosahedral Atmospheric Model: description and development. *Prog. in Earth and Planet. Sci.*, *1*(1), 18. doi:10.1186/s40645-
014-0018-1. Satoh, M. et al. (2019), Global Cloud-Resolving Models. *Current Climate Change Reports*. doi:/10.1007/s40641- 019-00131-0.
Sekiguchi, M. et al. (2003), A study of the direct and indirect effects of aerosols using global satellite data sets of aerosol and cloud parameters. *J. Geophys. Res.*, 108(D22), 4699, doi:10.1029/2002JD003359.
Sinha, P. R., et al. (2017), Evaluation of ground-based black carbon measurements by filter-based photometers at two Arctic sites. Journal of Geophysical Research: Atmospheres, 122, 3544–3572. doi:10.1002/2016JD025843.
Sinha, P. R. et al. (2018), Seasonal progression of the deposition of black carbon by snowfall at Ny-Alesund, Spitsbergen, *J. Geophys. Res. Atmos.,* 123, 997-1016, doi:/10.1002/2017JD028027.
Smith, C. J. et al. (2020), Effective radiative forcing and adjustments in CMIP6 models. *Atmos. Chem. Phys.,* 20(16), 9591–9618. doi:/10.5194/acp-20-9591-2020
Smith, C. J. et al. (2021), Energy budget constraints on the time history of aerosol forcing and climate sensitivity. *J. Geophys. Res. Atmos.,* 126, e2020JD033622. doi:/10.1029/2020JD033622
Suzuki, K., J.-C., Golaz & G. L. Stephens (2013a), Evaluating cloud tuning in a climate model with satellite observations. *Geophys. Res. Lett.*, 40, 4464-4468, doi:10.1002/grl.50874.
Suzuki, K., T. Y. Nakajima & G. L. Stephens (2010), Particle growth and drop collection efficiency of warm clouds as inferred from

joint CloudSat and MODIS observations. *J. Atmos. Sci.*, 67, 3019-3032, doi:10.1175/2010JAS3463.1.
Suzuki, K., G. L. Stephens, S. C. van den Heever & T. Y. Nakajima (2011), Diagnosis of the warm rain process in cloud-resolving models using joint CloudSat and MODIS observations. *J. Atmos. Sci.*, 68, 2655-2670, doi:10.1175/JAS-D-10-05026.1.
Suzuki, K., G. L. Stephens & M. D. Lebsock (2013b), Aerosol effect on the warm rain formation process: Satellite observations and modeling. *J. Geophys. Res.*, 118, doi:10.1029/2012JD018722.
Suzuki, K. & T. Takemura (2019), Perturbations to global energy budget due to absorbing and scattering aerosols. *J. Geophys. Res. Atmos.,* 124, 2194–2209. doi:/10.1029/2018JD029808
Suzuki, K. et al. (2004), A study of the aerosol effect on a cloud field with simultaneous use of GCM modeling and satellite observation. *J. Atmos. Sci.*, 61, 179-194.
Suzuki, K. et al. (2008), Global cloud-system-resolving simulation of aerosol effect on warm clouds. *Geophys.*
Res. Lett., *35*(19), L19817. doi:/10.1029/2008GL035449.
Suzuki, K. et al. (2015), Evaluation of the warm rain formation process in global models with satellite observations. *J. Atmos. Sci.*, 72, 3996-4014, doi:10.1175/JAS-D-14-0265.1.
Szopa, S. V. et al. (2021), Short-Lived Climate Forcers. In Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change [Masson-Delmotte, V., P. et al. (eds.)]. Cambridge University Press. In Press.
Takahashi, H., K. Suzuki & G. Stephens (2017), Land-ocean differences in the warm-rain formation process in satellite and ground-based observations and model simulations. *Q. J. R. Meteorol. Soc.*, 143:1804-1815, doi:10.1002/qj.3042.
Takami, A. et al. (2007), Transport of anthropogenic aerosols from Asia and subsequent chemical transformation, *J. Geophys. Res.*, 112, D22S31, doi:10.1029/2006JD008120.
Takemura, T., T. Nozawa, S. Emori, T. Y. Nakajima & T. Nakajima (2005), Simulation of climate response to
aerosol direct and indirect effects with aerosol transport-radiation model. *J. Geophys. Res.*, 110(D2), D02202. doi:10.1029/2004JD005029.
Takemura, T. & K. Suzuki (2019), Weak global warming mitigation by reducing black carbon emissions, *Sci. Rep.*, 9:4419, doi:10.1038/s41598-019-41181-6.
Takemura, T. (2020), Return to different climate states by reducing sulphate aerosols under future CO_2 concentrations, *Sci. Rep.*, 10:21748, doi:10.1038/s41598-020-78805-1.
Taketani, F. et al. (2016), Shipborne observations of atmospheric black carbon aerosol particles over the Arctic Ocean, Bering Sea, and North Pacific Ocean during September 2014, *J. Geophys. Res. Atmos.*, 121, 1914-1921, doi:10.1002/2015JD023648.
Tobo, Y. et al. (2013), Biological aerosol particles as a key determinant of ice nuclei populations in a forest ecosystem, *J. Geophys. Res. Atmos.*, 118, 10100-10110.
Tobo, Y. et al. (2019), Glacially sourced dust as a potentially significant source of ice nucleating particles, *Nat. Geosci.*, 12, 253-258, doi:10.1038/s41561-019-0314-x.
Tobo, Y. et al. (2020), Seasonal trends of atmospheric ice nucleating particles over Tokyo, *J. Geophys. Res. Atmos.*, 125, e2020JD033658.
Tomita, H., H. Miura, S. Iga, T. Nasuno and M. Satoh (2005), A global cloud-resolving simulation: Preliminary results from an aqua planet experiment. *Geophys. Res. Lett.*, 32(8), 1–4. doi:10.1029/2005GL022459
Uetake, J. et al. (2016), Microbial community variation in cryoconite granules on Qaanaaq Glacier, NW Greenland, *FEMS Microbiol. Ecol.*, 92, fiw127, doi: 10.1093/femsec/fiw127.
Uno, I. et al. (2009), Asian dust transported one full circuit around the globe, *Nat. Geosci.*, 2, 557-560,
doi:10.1038/NGEO583.
Uno, I. et al. (2020), Paradigm shift in aerosol chemical composition over regions downwind of China, *Scientific Reports*, 10:21748, doi:10.1038/s41598-020-63592-6.
Yamagami, M., F. Ikemori, H. Nakashima, K. Hisatsune & K. Osada (2019), Decreasing trend of elemental carbon concentration with changes in major sources at Mega city Nagoya, Central Japan, *Atmos. Environ.*, 199, 155−163. doi:10.1016/j.atmosenv.2018.11.014.
Yasunari, T. et al. (2018), Extreme air pollution events in Hokkaido, Japan, tracked back to early snowmelt and largescale wildfires over East Eurasia: Case studies. *Sci. Rep.* 8, 6413, doi:10.1038/s41598-018-24335-w.
Yoshida, A. et al. (2016), Detection of light-absorbing iron oxide particles using a modified single-particle soot photometer. *Aerosol Sci. Tech.*, 50, 1−4, doi:10.1080/02786826.2016.1146402.
Yoshida, A. et al. (2020), Abundances and microphysical properties of light‐absorbing iron oxide and black carbon aerosols over East Asia and the Arctic. *J. Geophys. Res. Atmos.*, 125, e2019JD032301, doi:10.1029/2019JD032301.
Zanis, P. et al. (2020), Fast responses on pre-industrial climate from present-day aerosols in a CMIP6 multi-model study, *Atmos. Chem. Phys.,* 20, 8381−8404, doi:/10.5194/acp-20- 8381-2020

Table A-7. Summary table for the topic **Stratosphere and Mesosphere**

(Authors: Nawo Eguchi, Yosuke Yamashita, Hideharu Akiyoshi, Tetsu Sakai, Tomoo Nagahama, Yoshihiro Tomikawa, Hideaki Nakajima, Takafumi Sugita, Takatoshi Sakazaki, Takuya Saito, Akira Mizuno)

1. Ultimate Goals	✓ Understanding of the structure of the boundary region between the troposphere and stratosphere and the relationship between troposphere-stratosphere dynamical coupling and mass exchange. ✓ Understanding of the effects of changes in trace constituents (GHGs, ozone, CFC substitutes, water vapor, etc.) on atmospheric circulation in the stratosphere. ✓ Understanding of ozone layer changes associated with climate change and the impact of ozone layer changes on climate. ✓ Understanding of the chemical effects of stratospheric aerosols from volcanic eruptions and large forest fires, and quantitative understanding of the contribution of anthropogenic aerosols to the stratosphere and their influx processes. ✓ Prediction of the impact of geoengineering to modify the stratospheric aerosol layer as a countermeasure against global warming and appropriate indicators based on such predictions. ✓ Understanding of the effects of solar activity (radiation and energetic particles) and magnetospheric phenomena that produce NOx and HOx through ion chemistry and alter mesospheric and stratospheric ozone, and their impact on climate.
2. Progress in the past 10 years: global development and Japanese uniqueness	✓ Japan has made valuable contributions to balloon observations in the tropics, such as SOWER, large atmospheric radars (Indonesia, PANSY in Antarctica), and UTLS region observations with CONTRAIL. ✓ Advancement of numerical models. Improved parameterization of cumulus clouds and gravity waves. Contribution to the refinement of UTLS processes by chemical transport models. ✓ Improvement of resolution with the development of computer resources. ✓ Increased use of numerical models to study the effects of the ozone layer on the climate, where the ozone distribution is calculated online in a chemical climate model rather than given. ✓ Japan's SMILES, equipped with the world's first superconducting detector operated by a mechanical cryogenic refrigerator and observing in a solar asynchronous orbit unrestricted by local time, produced unique results, including the discovery of diurnal changes in ozone and variations associated with solar eclipses. ✓ The super-pressure balloon enables in-situ stratospheric observations at about the same altitude for about one month. ✓ Ground-based FTIR has increased the number of studies that investigate global secular changes by globally unifying and optimizing analysis tools and molecular parameters. Column quantities can now be derived for absorption lines such as CFC and HFC, which have weak intensities. ✓ Microwave observations, both ground-based and satellite-based, provide data in the mesosphere and beyond, which is difficult to obtain with other instruments. Japan has advanced technology in the development of superconducting receivers with ultra-high sensitivity for SMILES and other ground-based radiometers.
3. Current major gaps	✓ The temporal and vertical resolutions of current observational data and models are insufficient for understanding the material circulation and interactions in the troposphere-stratosphere (-mesosphere). High resolution observations and models are needed to understand phenomena such as tropical cumulus convection and tropopause folding, which are small in scale but also important for mass transport. ✓ In order to refine future projections, it is necessary to understand the interactions between meteorological fields and trace constituents such as ozone, and to incorporate them appropriately into high-resolution chemical climate models. ✓ The global satellite program has been stagnant (i.e., plans for continuing missions have been delayed), which may limit the continuity of global data and the types of trace constituents that can be observed. This may hinder our understanding of long-term trends in trace constituent concentrations, understanding of transport processes (consistent with meteorological fields and transport of various trace constituents), and estimation of abundances of equivalent effective stratospheric chlorine (EESC), reactive nitrogen oxides (NOy), etc.
4. Key questions and hypothesis to drive research and development during 2022-32	✓ How do trace elements, especially short-lived chemical species, enter the stratosphere from the troposphere? What are the timescales and troposphere-stratosphere exchange time constants? ✓ How does the stratospheric temperature structure change as the influx of chemical species such as water vapor from the troposphere to the stratosphere (and vice versa) changes with increasing GHGs? ✓ Basically, the stratospheric cooling effect of increased CO_2 will continue to dominate, but it is possible that, contrary to international regulations, increased concentrations of HFCs and other species may act to increase temperatures in the lower stratosphere, partially reducing the cooling effect. What will be the atmospheric circulation in the mid- and high latitudes due to these changes in the temperature structure? ✓ To what extent do solar activity, magnetospheric activity, cosmic rays, and other energy inputs to the Earth's atmosphere affect the global environment? Do such effects propagate to the troposphere?
5. Collaborations	✓ SPARC, NDACC, SCOSTEP

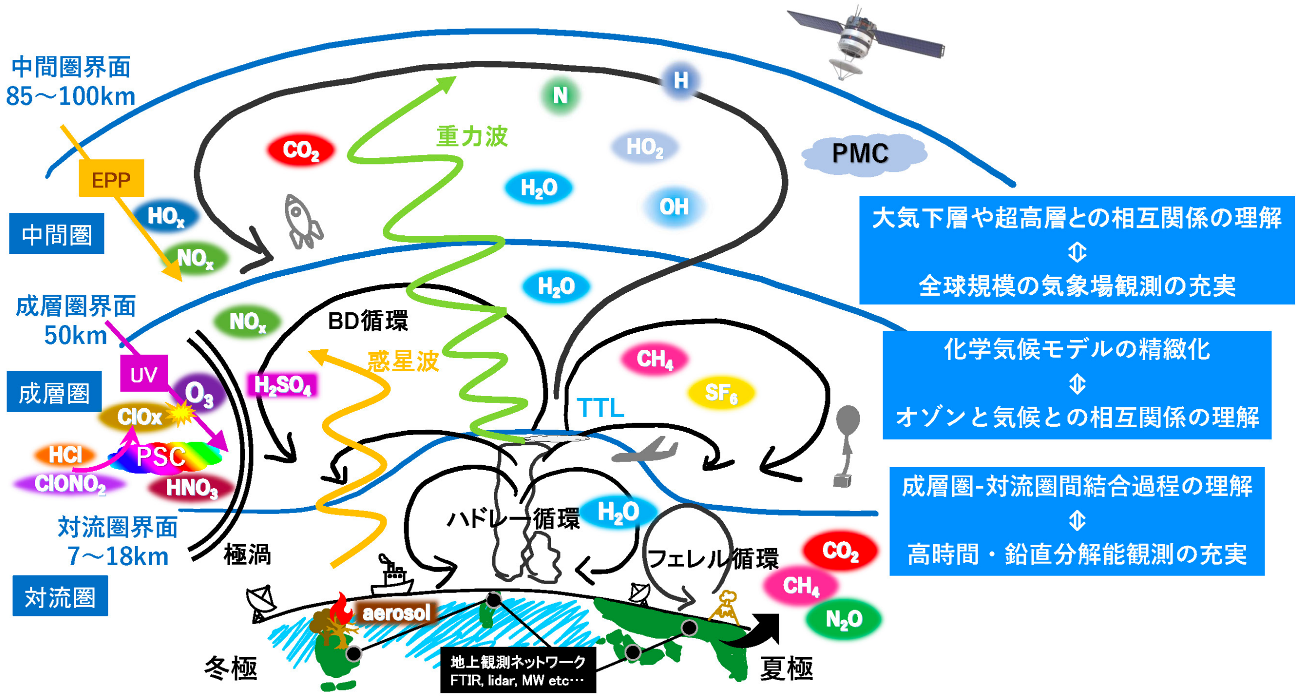


Figure A-7. Schematic diagram of phenomena and mechanisms relevant to atmospheric chemistry in the stratosphere and mesosphere.

Selected References (Stratosphere and Mesosphere)

Bisht, J. S. H. et al. (2021), Seasonal variations of SF_6, CO_2, CH_4, and N_2O in the UT/LS region due to emissions, transport, and chemistry, *Journal of Geophysical Research: Atmospheres,* 126, e2020JD033541, doi:10.1029/2020JD033541.

Chubachi, S. (1984), Preliminary results of ozone observation at Syowa station from February 1982 to January 1983, *Mem. Natl. Inst. Polar Res.*, *34*, 13-19.

Dupuy, E., H. Akiyoshi, & Y. Yamashita (2021), Impact of unmitigated HFC emissions on stratospheric ozone at the end of the 21st century as simulated by chemistry-climate models, *J. Geophys. Res.: Atmospheres*, *126*, e2021JD035307, doi: 10.1029/2021JD035307.

Fujiwara, M., P. Martineau, & J. S. Wright (2020), Surface temperature response to the major volcanic eruptions in multiple reanalysis data sets, *Atmos. Chem. Phys.*, 20, 345–374, doi: 10.5194/acp-20-345-2020.

Fujiwara, M. et al. (2021), Lower-stratospheric aerosol measurements in eastward-shedding vortices over Japan from the Asian summer monsoon anticyclone during the summer of 2018, *Atmos. Chem. Phys.*, 21, 3073–3090, doi:10.5194/acp-21-3073-2021.

Hattori, S., K. Kamezaki, & N. Yoshida (2020), Constraining the atmospheric OCS budget from sulfur isotopes, *Proc. Natl. Acad. Sci. U.S.A.*, 117(34), 20447–20452, doi:10.1073/pnas.2007260117.

Hayakawa, H. et al. (2021), The intensity and evolution of the extreme solar and geomagnetic storms in 1938 January, *Astrophys. J.*, 909 id. 197, 14pp., doi: 10.3847/1538-4357/abc427.

Imai, K. et al. (2015), SMILES observations of mesospheric ozone during the solar eclipse, *Geophys. Res. Lett.*, *42*, doi:10.1002/2015GL063323.

Kawatani, Y., & K. Hamilton (2013), Weakened stratospheric quasi-biennial oscillation driven by increased tropical mean upwelling, *Nature*, *497*, 478–481, doi: 10.1038/nature12140.

Kinoshita, T. et al. (2022), Toward Standard Radiosonde Observations of Waves and the Mean State in the 30–40 km Altitude Range Using 3–kg Balloons. *Journal of Atmospheric and Oceanic Technology*, doi: 10.1175/JTECH-D-21-0011.1.

Kodera, K., & Y. Kuroda (2002), Dynamical response to the solar cycle, *J. Geophys. Res.*, *107*, 4749, doi: 10.1029/2002JD002224.

Miyake, F., K. Nagaya, K. Masuda, & T. Nakamura (2012), A signature of cosmic-ray increase in AD 774–775 from tree rings in Japan, *Nature*, 486, 240–242, doi:10.1038/nature11123.

Miyazaki, K. et al. (2016), Inter-comparison of stratospheric mean-meridional circulation and eddy mixing among six reanalysis data sets, *Atmos. Chem. Phys.*, *16*, 6131–6152, doi: 10.5194/acp-16-6131-2016.

Miyoshi, Y. et al. (2021), Penetration of MeV electrons into the mesosphere accompanying pulsating aurorae, *Sci. Rep.,* 11, 13724, doi: 10.1038/s41598-021-92611-3.

Nakazawa, T. et al. (2002), Variations of stratospheric trace gases measured using a balloon-borne cryogenic sampler, *Adv. Space Res.*, 30(5), 1349-1357, doi: 10.1016/S0273- 1177(02)00551-3.

Ouchi, M. et al. (2019), Development of a balloon-borne instrument for CO_2 vertical profile observations in the troposphere, *Atmos. Meas. Tech.*, 12, 5639–5653, doi:10.5194/amt-12-5639-2019.
Sakazaki, T. et al. (2013), Diurnal ozone variations in the stratosphere revealed in observations from the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) on board the International Space Station (ISS), *J. Geophys. Res. Atmos.*, 118, 2991− 3006, doi:10.1002/jgrd.50220.
Satoh, M. et al. (2014), The non-hydrostatic icosahedral atmospheric model: Description and development. *Prog. In Earth and Planet. Sci.*, 1(1), 1-32, doi: 10.1186/s40645-014-0018-1.
Tada, M., K. Yoshimura, & K. Toride (2021), Improving weather forecasting by assimilation of water vapor isotopes, *Sci. Rep.*, 11, 18067, doi: 10.1038/s41598-021-97476-0.
Takeda, M. et al. (2021), First ground-based Fourier transform infrared (FTIR) spectrometer observations of HFC-23 at Rikubetsu, Japan, and Syowa Station, Antarctica, *Atmos. Meas. Tech.*, 14, 5955-5976, doi: 10.5194/amt-14-5955-2021.
Toride, K. et al. (2021), Potential of Mid-tropospheric Water Vapor Isotopes to Improve Large-Scale Circulation and Weather Predictability, *Geophys. Res. Lett.*, 48, e2020GL091698, doi: 10.1029/2020GL091698.
Yamashita, Y. et al. (2010), Ozone and temperature response of a chemistry climate model to the solar cycle and sea surface temperature, *J. Geophys. Res.*, *115*, D00M05, doi:10.1029/2009JD013436.
Yoneyama, K. et al. (2021), Evaluation of LMS6 and RS41 radiosonde humidity data obtained during YMC-Boreal Summer Monsoon study in 2018 and 2020, *J. Meteor. Soc. Japan*, 99, 1115-1125, doi: 10.2151/jmsj.2021-054.