Institute of Nature and Environmental Technology

Kanazawa University

Self-Review/Assessment Report

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Institute of Nature and Environmental Technology,

Kanazawa University

Contents

1. Preface

A multitude of problems related to environmental changes caused by human activities and nature itself pose some of the greatest challenges faced by science and technology in the 21st century. The Institute of Nature and Environmental Technology, established in 2002, was reorganised in 2007 to actively tackle such issues. The Institute places particular emphasis on environmental research on the north-eastern fringes of the continent of Asia with a special focus on areas that touch and encompass the Sea of Japan. The latter have been shown to be especially vulnerable to global environmental fluctuations. In 2014, the Institute set itself the objective of addressing the urgent environmental issues facing the general Sea of Japan area, and of formulating plans for a sustainable environment in the area in the future. Furthermore, in 2016, the Japanese Ministry of Education, Culture, Sports, Science and Technology designated the Institute an "International Joint Research Center for the Study of Environmental Changes due to Cross-border Pollution". The Institute has been aiming to make contributions at the domestic and regional level by examining the Sea of Japan area in particular. To this end, it has been investigating the origins and quantity of hazardous chemical substances, the nature of their cross-border transference, and the impact of these substances on human health and ecosystems.

The Noto Peninsula, which protrudes into the Sea of Japan, is an ideal observation base for analysing substances transported across national borders from the eastern fridges of the Asian continent. Furthermore, the oceanographic condition around the peninsula is also advantageous to the observation of the movement of harmful chemical substances. In technical terms, this refers to the recording of fluctuations in the inflow of substances from the East China Sea by utilising radionuclides as tracers.

In order to take advantage of the location of the Noto Peninsula, the Institute has set up two atmospheric observation supersites and a marine laboratory on the area. It has also established the Low-Level Radioactivity Laboratory, which is capable of measuring extremely low levels of radioactivity, as well as the Ogoya Underground Laboratory. The Botanical Garden there also evaluates organic substances that are harmful to human health. These unique facilities are a major feature of the institute. In addition, branch offices in China, South Korea and Russia have been set up, in order to establish overseas collaborative joint research bases.

The Institute, certified as a Joint Usage/Education Center by the Japanese Ministry of Education, Culture, Sports, Science and Technology in 2012, and re-certified in 2017 and 2022, has been offering courses that international non-Japanese speaking students can participate in as well as extensive practical training opportunities for

domestic students. Since receiving “Joint Usage / Research Center” certification in 2016 and then seeing this status renewed in 2022, the institute has been working on research that analyses and observes the quantities and transfer characteristics of harmful chemical substances over a wide area. It has also integrated research that has combined analyses of the atmospheric, marine, and terrestrial environment aimed at better understanding the dynamics of the targeted substances in the area. Fig.1 Intraorganization was modified in 2015.

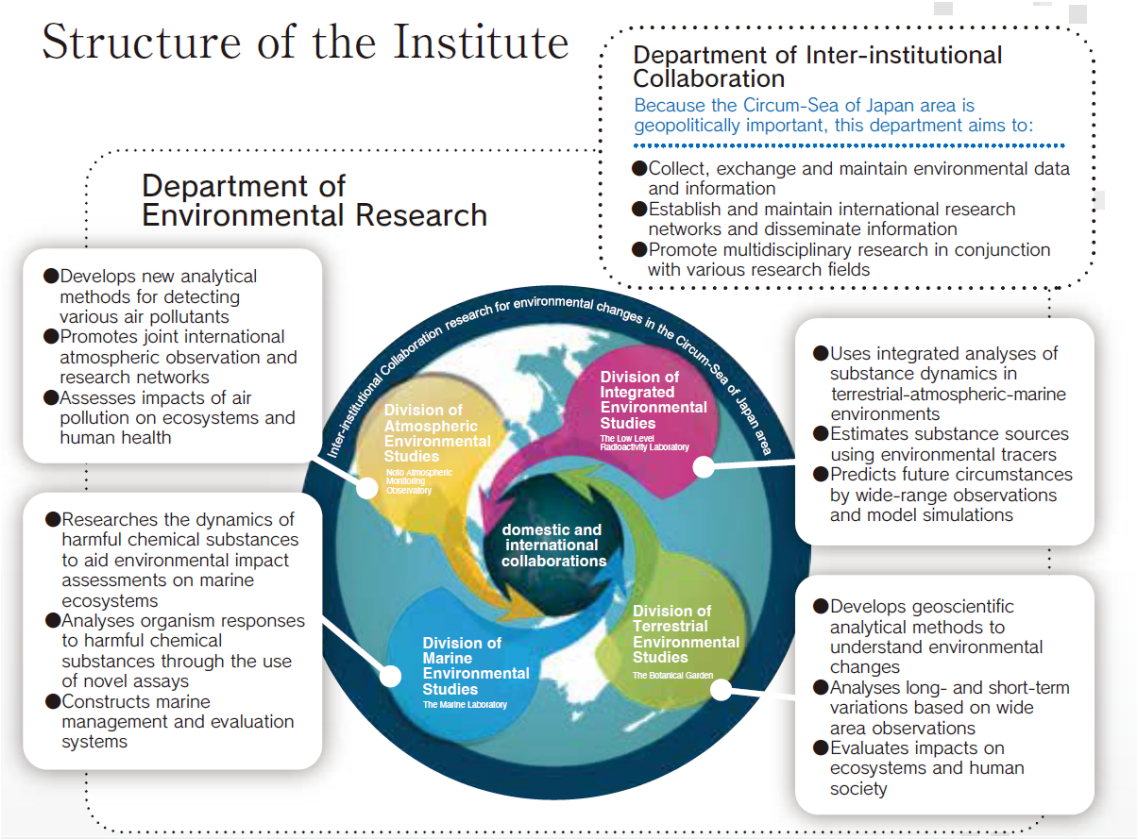

Fig.1 Structure of the Institute

Five years has passes since the last assessment of research and educational activities in the Institute held in 2017. This short report introduces our activities during the fiscal year 2017-2021 to help the assessment.

2. Division of Atmospheric Environmental Studies

[Ounline]

This division focuses on the atmospheric environment of the Circum-Sea of Japan area, an area of the world where high concentrations of atmospheric pollutants such as Asian dust (Kosa: yellow dust) and $PM_{2.5}$ have become one of the most prominent environmental issues. The aim of this division is to clarify their mechanisms of generation, transport, reaction, deposition, ecosystem and human consequences, and to better predict future pollution in the area. To this end, this division devises and develops new analytical methods and fosters joint international research networks. These activities and their results will make substantial contributions to the global efforts to mitigate atmospheric contamination.

[Monitoring]

Behaviors of Atmospheric Polycyclic Aromatic Hydrocarbons (PAHs) and Nitropolycyclic Aromatic Hydrocarbons (NPAHs) in East Asian

PAHs are a group of organic compounds consisting of two or more fused benzene rings, and NPAHs are their nitrated derivatives. PAHs and NPAHs are ubiquitous environmental pollutants. Atmospheric PAHs and NPAHs mainly originate from imperfect combustion and pyrolysis of organic matters, although some NPAHs are formed in the atmosphere via reactions of their parent PAHs such as 2-nitropyrene (2-NP) and 2-nitrofluoranthene (2-NFR). In urban areas, PAHs and NPAHs are mainly emitted from automobiles, power plants, domestic heating and industrial processes. Many PAHs and NPAHs have carcinogenic and/or mutagenic properties. Benzo[*a*]pyrene (BaP) and 1-NP are categorized in groups 1 (*carcinogenic to humans*) and 2A (*probably carcinogenic to humans*), respectively (*International Agency for Research on Cancer*). Several PAHs also exhibit estrogenic, antiestrogenic, antiandrogenic activities or reactive oxygen species producing activity.

Since 1990s, monitoring of atmospheric PAHs and NPAHs in the East Asian region has been carried out step by step. So far, we have investigated at 22 urban sites and 3 background sites in Japan, China, South Korea, Mongolia and Russia (Fig. 1). According to the recent results, the PAHs and NPAHs concentrations in Shenyang, Shanghai (China), and Vladivostok (Russia) highlighted the relatively poor air quality and relatively high threat to human health compared to those in Sapporo, Sagamihara, and Kirishima (Japan). These concentrations in summer were all lower than those in winter except for NPAHs concentration in Shanghai. In summer, due to the Pacific High prevailing, the air pollutants in 6 cities were all affected by clean air masses from the ocean to different extents. The PAHs and NPAHs in Japanese cities were mainly emitted

from domestic Japan and the ocean and could hardly be affected by sources originating from the East Asian mainland. In winter, heating systems were operating in northern China, leading to the PAHs and NPAHs concentrations in Shenyang increasing sharply. Moreover, due to the Siberian High prevailing, the air masses (containing many PAHs and NPAHs) mainly originated from northern China and Mongolia. These polluted air masses can affect the nearby (Vladivostok) and more distant areas (southern China and Japan), leading to the PAHs and NPAHs concentrations in these cities increasing to some extent. However, local traffic emissions still have relatively large effects. On the other hand, compared to those in other cities, although the total PAHs and 1-NP concentrations in Shenyang and Shanghai remained relatively high, they both decreased notably in recent years because the Chinese government paid more attention to environmental protection (Fig. 2.) (Yang et al., *Environ. Pollut.*, 2021).

International networking activities of ground-based observations

Since the establishment of Noto Ground-based Research Observatory (NOTOGRO) station in Suzu city 10 years ago, it has been home to many domestic and international collaborations and contributed in more than 20 peer-reviewed publications. To name a few, the NOTOGRO station joined an international campaign to inter-compare the regional cloud condensation nuclei (CCN) characteristics which involved 70 experts from 23 institutions and 14 countries (Schmale et al., *Atmos. Chem. Phys.,* 2018). The aerosol-cloud interaction continues to constitute the largest uncertainty in the anthropogenic radiative forcing on the climate system. The effort to minimizing such uncertainty requires long-term CCN measurements covering multiple locations and seasons. This unprecedented dataset was later utilized by a group of 39 researchers across the world to validate state-of-the-art general circulation and chemical transport model outputs (Fanourgakis et al., *Atmos. Chem. Phys.,* 2019).

Since 2020, the NOTOGRO station also joined a Korean lead project (the center for Fine Particle Research Initiative in East Asia considering National Differences: FRIEND) to characterize air pollution in Northeast Asia through a close international collaboration among four countries (Mongolia, China, Korea and Japan). The FRIEND project plans to simultaneously measure the air pollutants by deploying high temporal resolution instruments during intensive field campaigns and conduct modeling studies for the period of 4.5-years. The first intensive monitoring campaign of $PM_{2.5}$ and gaseous pollutants was conducted from December 15, 2020, to January 14, 2021 (Fig. 3). A large set of measurement data as well as daily filter samples were collected, and analyses are currently underway. An overview paper from the first intensive monitoring campaign has been submitted (Kim et al., *Atmos. Environ.*, 2022).

Decreased trend of $PM_{2.5}$ and BC observed at central and western Japanese islands

Interannual trend of $PM_{2.5}$ and BC in Noto (Suzu site of the NOTOGRO station) and Fukue were investigated by using the statistical trend analysis and chemical transport model, Regional Air Quality Model 2 (RAQM2ver3). Concentrations of $PM_{2.5}$ and BC in Noto and Fukue decreased with seasonal variation from 2013 to 2019. Total reduction of $PM_{2.5}$ mass concentrations in Noto and Fukue were 7.5 ± 5.0 and 3.8 ± 0.95 $\mu g/m^3$, which corresponds to 48% and 24% reduction against the initial concentrations, respectively. The reduction at Noto were larger than those at Fukue. Total reduction of BC in Noto from 2013 to 2019 were estimated to be 0.13 ± 0.1 $\mu g/m^3$, and the reduction account for 32%. Results with the source-receptor analysis by chemical transport model suggest that decrease of $PM_{2.5}$ and BC concentrations in Fukue and Noto is strongly influenced by decrease of transported amount from the Central China, Northern China, and Japan after 2015/2016. The ratios of $PM_{2.5}$/BC in Noto indicate that secondary production of $PM_{2.5}$ occurs during the transboundary transport and the contribution in summer is larger than those in winter (Fig. 4).

New particle formation in the lower boundary layer by using the kite observation

New particle formation (NPF) is the source of the atmospheric cloud condensation nuclei. Therefore, these are influence the cloud properties and Earth's energy balance, following then changing the atmospheric environment. Based on the ground based monitoring, the incomplete and weak NPF (starting from ~10 nm) at Fukue island (downstream) are suggesting the nucleation in upstream region. In order to investigate the NPF, the vertical distribution of atmospheric particle concentrations were observed at remote island, Fukue, by using the UAV. Vertical structure of atmospheric nanoparticle concentration (particle size > 6 nm and altitude < 1.2 km height) during NPF was observed using UAV at Fukue. Three different type of events were identified based on aerial observation (different vertical profile of nanoparticles), ground-based measurement (initial detected diameter of nanoparticle, starting time of NPF, and dominant chemical component in PM_1), and air mass backward trajectory analysis. The stronger NPF event (Event I, > 35,000 # cm^{-3}) with the particle size as small as 5 nm affected by the long range transport of air pollutants under a high pressure system was detected. In addition, sudden increase of particle number concentration and SO_2 concentration with weak NPF (onset diameter above than 10 nm) caused by the change of air mass origin (Event II, < 10,000 # cm^{-3}).

[Analytical method]

Development of novel particle measurement techniques

Atmospheric aerosols take variety of mixing states, and various chemical compounds are often found to co-exist within a single particle. Such internal mixing causes large particle to particle variability and significantly affect their fate, behavior and impacts within the atmospheric environment. There is a constant need for a novel analytical method capable of sensitively characterizing chemical and physical properties on a single particle basis.

Raman micro-spectroscopy is used to identify chemical compounds within an aerosol particle, however, its application has generally been limited to supermicron particles due to the optical diffraction-limit and inherently weak Raman signal. We have been working over the years to overcome these limitations by the help of SERS (Surface Enhanced Raman Spectroscopy) and developed a new condensation-growth coupled system capable of detecting sulfate and organics from a single nano-particle as small as 20 nm in diameter (Kunihisa et al., *Aerosol Sci. Technol.*, 2020). This is a significant step forward in the SERS related work since it pushed the detectable size limit down to well within the Aitken size range, therefore, could even be applied to studying atmospheric particle nucleation (Fig. 5).

A subset of aerosol particles would act as ice nuclei and thereby play an essential role in the cloud microphysics and water cycle. However, what makes a good ice nucleus remained unanswered for many years due to their scarcity. Through our new Individual Droplet Freezing Method (IDFM), we have demonstrated that atmospheric aging can alter the ice nucleating activity of Asian dust (Iwata and Matsuki, *Atmos. Chem. Phys.,* 2018), and that a certain fungal spore acts efficiently as ice nuclei and emitted selectively during rainy days (Iwata et al., *Atmosphere*, 2019).

Mixing of various organic and inorganic compounds can often form core-shell like structure and its physical properties depend largely on the particle coating. We have established a new method to directly evaluate the adhesion force of individual particles by using Atomic Force Microscopy (Ono et al., *Atmosphere*, 2020), and demonstrated that a certain Asian dust particle becomes more adhesive following long-range transport.

[Health effect]

Particulate PAH transport associated with adult chronic cough occurrence closely connected with meteorological conditions: A modelling study

PAHs are regarded as generating toxic adverse effects on human health. In this study, we analysed the relationship among atmospheric particulate PAHs (p-PAHs), cough occurrence via epidemiological research, and meteorological conditions using a

chemical transport model. According to previously reported epidemiological data observed from 4 January to 30 June 2011 in Kanazawa, a coastal site of the Sea of Japan, ambient p-PAH exposure is related to chronic cough occurrence in adult patients. Source receptor relationship (SRR) analysis revealed that a high cough occurrence was caused by exposure to high p-PAH levels in air masses transported from Central China (CCHN, 30-40°N) under westerly conditions. The p-PAHs transported from northern China (NCHN, >40°N) and the eastern part of Russia (ERUS) under northwesterly conditions also contributed to cough occurrence. The low equivalent potential temperature and geopotential height anomaly suggested that the p-PAHs emitted near the surface were suppressed and not transported upward but were instead transported horizontally near the surface in the boundary layer, resulting in high p-PAH concentrations arriving in Kanazawa. Our study findings suggest that the air mass transport pattern associated with meteorology strongly influences the high p-PAH concentrations causing adult chronic cough occurrence (Fig. 6).

Fig. 1 PAHs monitoring network in East Asia.

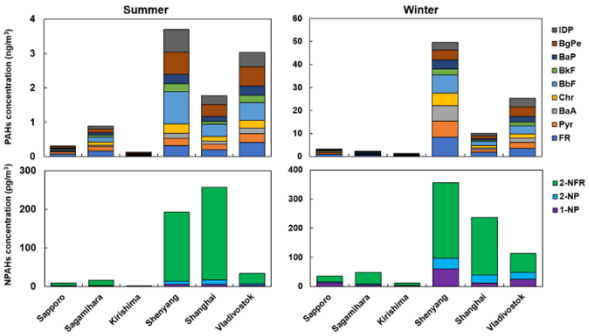

Fig. 2 PAHs and NPAHs concentrations in six cities.

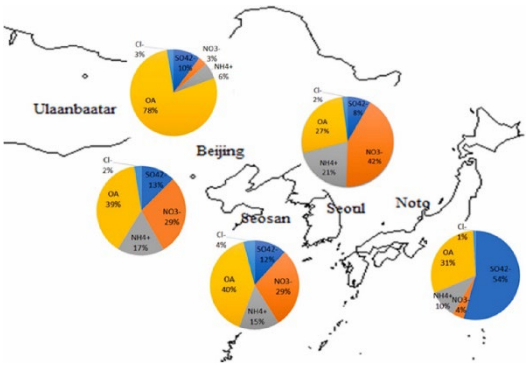

Fig. 3 Proportions of major chemical species in $PM_{2.5}$ (Ulaanbaatar and Seosan) and $PM_{1.0}$ (Beijing, Seoul and Noto) during the first intensive observation period.

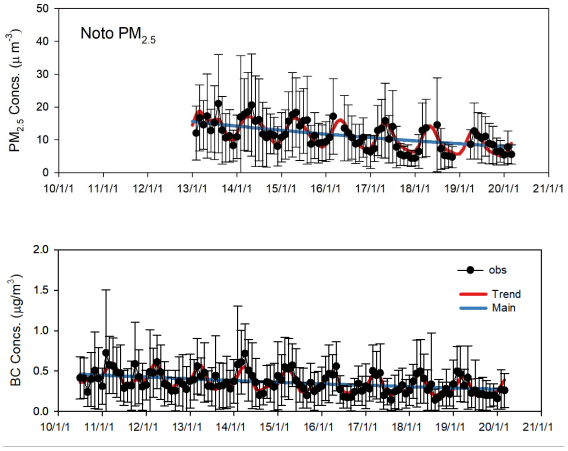

Fig. 4 Temporal variations of $PM_{2.5}$ and BC concentrations in Noto.

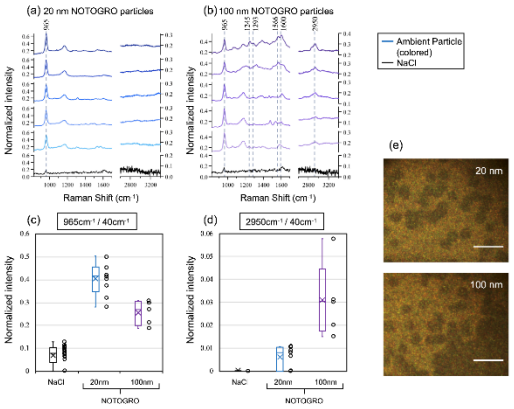

Fig. 5 (a, b) Normalized SERS (colored) spectra obtained from ambient particles in NOTOGRO, shown separately for mono- dispersed particles at 20 nm and 100 nm and pure water (black). (c, d) The normalized intensity of the peak at 965 cm^{-1} (sulfate) and 2950 cm^{-1} (Organics). (e) Optical images of particle traces found on SERS substrate following condensational growth. Scale bars: 10 μm.

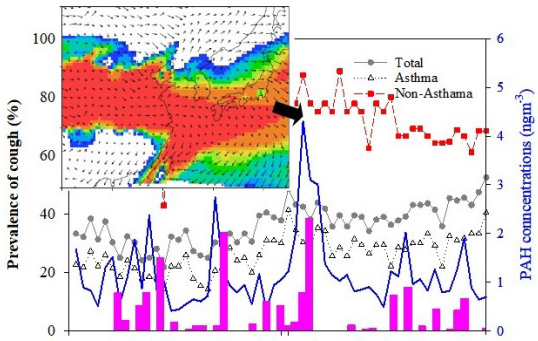

Fig. 6 Temporal variations in the daily prevalence of cough for the total patients and patients with and without asthma (%) versus the p-PAH concentration in Kanazawa city from 4 January to 30 June 2011. The bar indicates the per unit increase in the p-PAH concentration. These data were retrieved from Ayenda et al. (2016). An increase in the prevalence of cough occurred during the cross-border transport of PAHs from central China.

3. Division of Marine Environmental Studies

[Outline]

This division studies the influence of harmful chemical substances on marine organisms and the dynamics of these substances in the marine environment in the Circum-Sea of Japan area; particularly in coastal areas. This examination of marine biota is performed through the use of biochemical assays. The research performed aids in the development of marine environment evaluation systems and for this purpose involves international collaborations.

To achieve the purpose, we recently focus on the toxicity of polycyclic aromatic hydrocarbons (PAHs) on the basis of our basic studies for calcium metabolism. PAHs are widespread environmental contaminants derived from petroleum and generated through the incomplete combustion of fossil fuel, wood, and other organic materials. In the aquatic environment, PAH contamination occurs through storm water runoff. Atmospheric deposition of PAHs is now the largest source of aquatic PAH contamination. Furthermore, oil tankers are another anthropogenic source of PAHs and oil spills are correlated with the major shipping routes in marine environments. Moreover, accidental oil spills, such as those from the Deepwater Horizon, the Exxon Valdez, and the Nakhodka, have caused direct PAH pollution in the marine environment. PAH contamination is thus prevalent throughout the global marine system and directly affect marine organisms. In this division, thus, we recently studied the toxicity of PAHs and their metabolites in aquatic animals such as fish and sea urchin. We describe below.

[Fish scales as a suitable model for the analysis of pollutants including PAHs on bone metabolism]

A teleost scale is a bone-like tissue in which coexist osteoblasts (Fig. 1A), osteoclasts (Fig. 1B), and calcified bone matrix. The bone matrix, which includes type I collagen, osteocalcin, osteonectin, and hydroxyapatite, is present in scales as well as in mammalian bone (Ikegame et al., 2019; Kobayashi-Sun et al., 2020). Teleost scales have an important function in regulating blood calcium levels because teleost scales, which have both osteoblasts and osteoclasts, are known to function as potential internal calcium reservoirs similar to those in the endoskeletons of mammals. Using a functional calcium-regulating organ (teleost scales), we have developed a novel assay system (Honda and Suzuki 2020; Sekiguchi et al., 2021).

Using an assay system, we investigated the influence of PAHs on calcium metabolism. For example, injecting of BaA (10 ng/g body weight) to nibbler fish (marine teleost) induced both hypocalcemia and hypophosphatemia as a result of inhibition of both osteoclastic and osteoblastic marker mRNA expressions (Fig. 2) (Zanaty et al., 2020).

[Hydroxylated PAH metabolites cause osteoblast apoptosis and skeletal abnormalities in fish]

To study the toxicity of 3-hydroxybenzo[c]phenanthrene (3-OHBcP), a metabolite of benzo[c]phenanthrene (BcP), first we compared it with its parent compound, BcP, using an in ovo-nanoinjection method in Japanese medaka. Second, we examined the influence of 3-OHBcP on bone metabolism using goldfish. Third, the detailed mechanism of 3-OHBcP on bone metabolism was investigated using zebrafish and goldfish. The LC50s of BcP and 3-OHBcP in Japanese medaka were 5.7 nM and 0.003 nM, respectively, indicating that the metabolite was more than 1,900 times as toxic as the parent compound. In addition, nanoinjected 3-OHBcP (0.001 nM) induced skeletal abnormalities. Therefore, fish scales with both osteoblasts and osteoclasts on the calcified bone matrix were examined to investigate the mechanisms of 3-OHBcP toxicity on bone metabolism. We found that scale regeneration in the BcP-injected goldfish was significantly inhibited as compared with that in control goldfish. Furthermore, 3-OHBcP was detected in the bile of BcP-injected goldfish, indicating that 3-OHBcP metabolized from BcP inhibited scale regeneration. Subsequently, the toxicity of BcP and 3-OHBcP to osteoblasts was examined using an in vitro assay with regenerating scales. The osteoblastic activity in the 3-OHBcP (10-10 to 10-7 M)-treated scales was significantly suppressed, while BcP (10-11 to 10-7 M)-treated scales did not affect osteoblastic activity. Osteoclastic activity was unchanged by either BcP or 3-OHBcP treatment at each concentration (10-11 to 10-7 M). The detailed toxicity of 3-OHBcP (10-9 M) in osteoblasts was then examined using gene expression analysis on a global scale with fish scales. Eight genes, including APAF1, CHEK2, and FOS, which are associated with apoptosis, were identified from the upregulated genes. This indicated that 3-OHBcP treatment induced apoptosis in fish scales. In situ detection of cell death by TUNEL methods was supported by gene expression analysis. This study is the first to demonstrate that 3-OHBcP, a metabolite of BcP, has greater toxicity than the parent compound, BcP (Fig. 3) (Suzuki et al., 2022).

[Toxicity of PAHs on enzyme activities in marine fish]

To examine the influence of BaA on liver diseases, the blood marker enzyme activities for ALP, AST, LDH, and LAP were measured in the plasma of BaA-treated nibbler fish or untreated nibbler fish. Data are shown in Figure 4.

In the low-dose group, all markers tended to decrease. Significant differences in the ALP and LDH of the plasma of BaA-treated nibbler fish as compared to that of untreated nibbler fish were observed. At the higher dose, remarkable suppression of ALP was

obtained. These results indicate that BaA may affect liver diseases and emphasize the importance of prevention of aquatic PAH pollution (Zanaty et al., 2020).

[Toxicity of PAHs and OHPAHs to sea urchins]

Furthermore, we examined the effect of both PAHs and OHPAHs on the embryogenesis of sea urchins (Hemicentrotus pulcherrimus) (Sekiguchi et al., Fisheries Sci., 2018). PAHs including BaA are priority pollutants in the aquatic environment. Our previous study revealed that BaA and its metabolite, 4-monohydroxylated BaA (4-OHBaA) inhibit larval skeletogenesis in the sea urchin Hemicentrotus pulcherrimus.(Suzuki et al., Comp. Biochem. Physiol., 2015). Here we report studies to elucidate the target of skeletogenesis inhibition elicited by BaA and 4-OHBaA. First, we performed an in vitro experiment using isolated micromeres which give rise to the larval skeletogenic mesenchyme. However, skeletogenesis was not repressed by BaA and 4-OHBaA, implying that these chemicals indirectly influence on the formation of larval skeleton. Next, we analyzed their influence in vivo using embryos. Vascular endothelial growth factor (VEGF) that is expressed in the ectoderm and induces spicule formation, was inhibited by BaA and 4-OHBaA treatment. These chemicals also suppressed the expression of the heparan sulfate 6-O endosulfatase known as a VEGF signaling modulator. We therefore propose that BaA and 4-OHBaA effects on larval skeletogenesis occur via VEGF signaling (Fig. 5).

[Conclusions]

In aquatic animals, we discovered the toxicity of OHPAHs, metabolites of PAHs. Judging from the obtained data, the toxicity of OHPAHs is stronger than that of PAHs, at least in fish and sea urchins.

Furthermore, we indicated that PAHs or OHPAHs influences liver metabolism in marine fish. Thus, OHPAHs that occurred with accumulated PAHs may have toxic influences on aquatic animals, even if the PAH levels in the aquatic environments are low. Thus, we should emphasize the prevention of aquatic PAH pollution because of the toxicity of OHPAHs.

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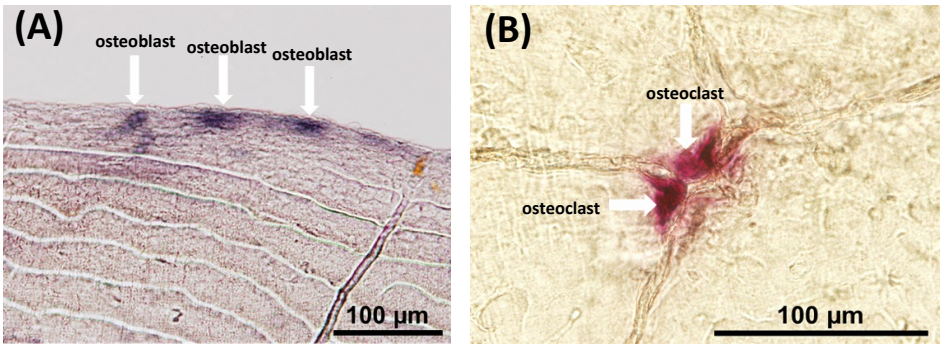


Fig.1 Teleost scales have an important function in regulating blood calcium levels because teleost scales, which have both osteoblasts (A) and osteoclasts (B), are known to function as potential internal calcium reservoirs similar to those in the endoskeletons of mammals (Ikegame et al., J. Pineal Res., 2019).

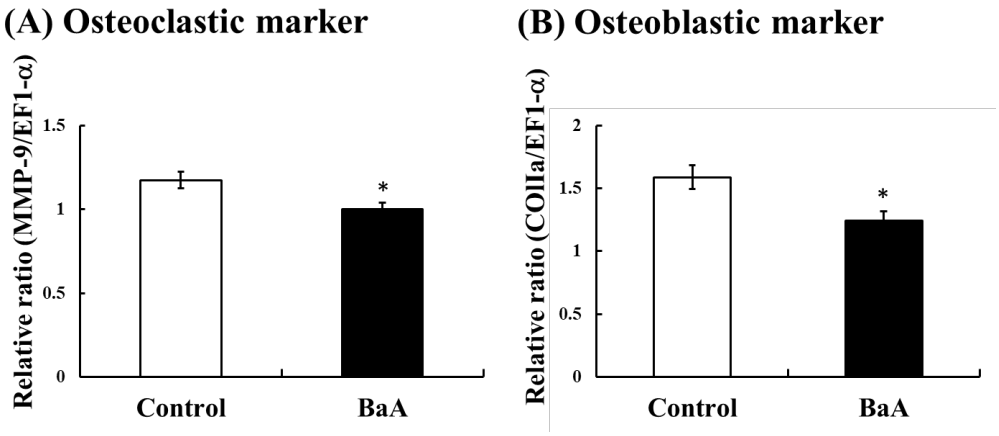


Fig.2 Efects of BaA on osteoclastic (A) and osteoblastic (B) marker mRNA expression in the scales of BaA-treated or untreated nibbler fish. BaA (10 ng/g body weight) was injected intraperitoneally (four times) into nibbler fish for 10 days during breeding. Thereafter, osteoclastic and osteoblastic marker mRNA expressions were examined in the control and experimental groups. * indicates a statistically significant dieffrence at $p < 0.05$ from the values in the control scales ($n = 8$). (Zanaty et al., Int. J. Environ. Res. Public Health, 2020)

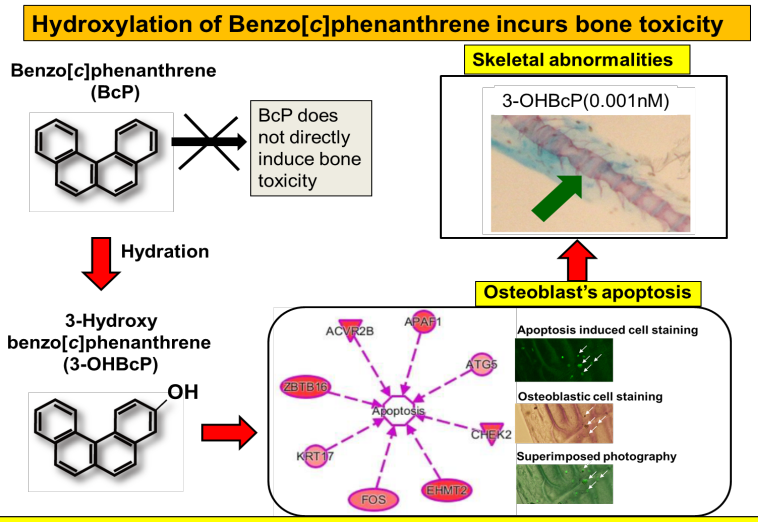


Fig.3 PAHs are toxic substances that induce various toxic stresses that result in apoptosis and endocrine disruption. The current findings suggest that it is not the parent compound (in this case, BcP) that is toxic but the hydroxylated congener that is truly biologically active. Thus, more attention to OHPAHs is warranted, in particular, those, such as members of the hydroxylated BcP group, which have many congeners but are understudied as compared to other OHPAHs (Suzuki et al., Ecotoxicol. Environ. Saf., 2022)

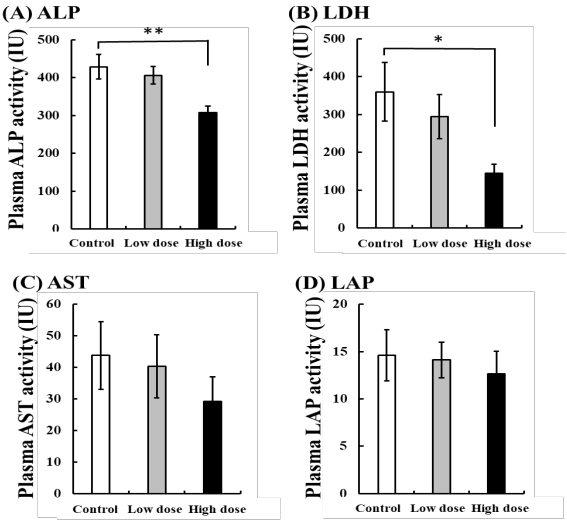


Fig.4 Toxicity in ALP (A), LDH (B), AST (C), and LAP (D) of the liver with BaA treatment. BaA (1 or 10 ng/g body weight) was injected intraperitoneally (4 times) into nibbler fish for 10 days during breeding. Thereafter, plasma liver markers were examined in the control and experimental groups. * and ** indicate statistically significant differences at $p < 0.05$ and $p < 0.01$, respectively, from the values in the control scales (n = 8). (Zanaty et al., Int. J. Environ. Res. Public Health, 2020)

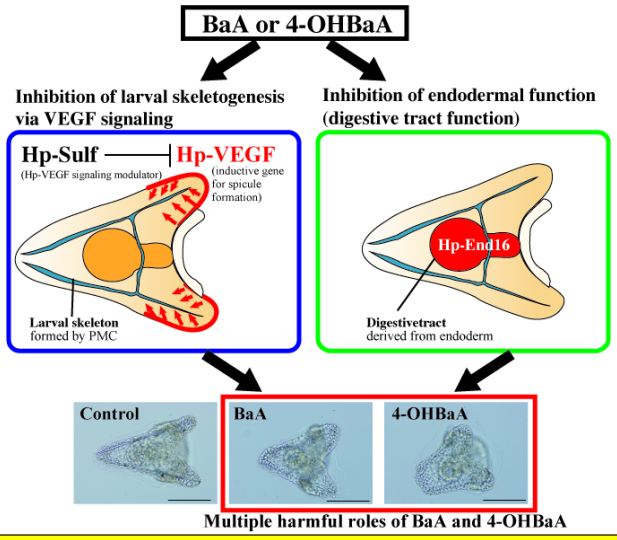


Fig.5 Hypothetical model of the effects of BaA and 4-OHBaA in pluteus larvae. BaA and 4-OHBaA inhibit the expression of Hp-VEGF localized in a part of ectodermal cells and also suppress the Hp-Sulf, which shows ubiquitous expression. Hp-VEGF induce the spicule formation. Hp-Sulf is expected to modulate Hp-VEGF signaling. In addition, Hp-Endo16 is expressed in the digestive tract and repressed by BaA and 4-OHBaA. Suppression of these genes expressions are expected to result in the phenotype of pluteus larva.

4. Division of Terrestrial Environmental Studies

[Outline]

The Sea of Japan area is characterized by the existence of diverse terrestrial environments. Aiming to understand their temporal and spatial changes and to clearly define the present, as well as predict future environmental circum- stances, this division develops geological and geochemical techniques to better understand short-term and long-term fluctuations. Further, this division uses biological approaches to evaluate the influence of human activities on terrestrial ecosystems and employs interdisciplinary approaches to develop their management strategies.

[Ecology and Conservation]

1) Climate change impacts on the potential distribution of threatened freshwater crustacean lineages

As climate change presents a major threat to biodiversity in the next decades, it is critical to assess its impact on species habitat suitability to inform biodiversity conservation. Species distribution models (SDMs) are a widely used tool to assess climate change impacts on species' geographical distributions. As the name of these models suggests, the species level is the most commonly used taxonomic unit in SDMs. However, recently it has been demonstrated that SDMs considering taxonomic resolution below (or above) the species level can make more reliable predictions of biodiversity change when different populations exhibit local adaptation. Here, we tested this idea using the Japanese crayfish (Cambaroides japonicus), a threatened species encompassing two geographically structured and phylogenetically distinct genetic lineages. The species-level SDM made future predictions that predicted much broader and severe impacts of climate change. However, the lineage-level SDMs led to reduced climate change impacts overall and also suggested that the eastern lineage may be more resilient to climate change than the western one.

2) Effects of farming practices on paddy field biodiversity

Agrochemical-free rice farming has attracted interest for restoring paddy field biodiversity and producing safe food. Odonata are commonly used as a biodiversity indicator in these lowinput farms. However, the effect of agrochemical-free rice farming on odonate diversity has rarely been assessed over the entire emergence period of these insects. We investigated whether different farming practices, such as conventional or natural (agrochemical-and fertilizer-free) cultivation, and associated water management strategies affect the emergence rates of Odonata in paddy field landscapes in central Japan (Fig. 1). Weekly exuviae sampling in 2017 and 2019 suggested that odonate assemblages differed between conventional and natural paddy fields, with a higher

number of taxa emerging from natural paddy fields. Contrary to expectations, conventional paddy fields had equivalent or higher emergence rates of all Odonata and two numerically dominant Sympetrum species. Our findings suggest that both conventional and natural paddy fields are important habitats for Odonata in Japan.

3) Developing resource-circulation farming using bamboo biomass resources

Bamboos have expanded their distribution in Japan in recent decades due to the abandonment of forest management practices following depopulation and aging in rural communities. Developing effective uses for bamboo biomass resources is highly desirable. In this study, we tested the effectiveness of ground bamboo as a new organic mulching material for agrochemical-free rice farming. Specifically, we tested whether ground culms and leaves with or without refrigeration/fermentation treatments influenced rice production differently. We found that straw weights were 1.5–2.1 times greater in the four ground bamboo treatments relative to controls. As a result, rice yields were 1.8–2.1 times higher in the four ground bamboo treatments relative to controls. Application of ground bamboo did not noticeably reduce rice grain quality. Thus, the use of ground bamboo mulch has considerable potential in increasing the yield of high-quality rice, at least on a small scale.

[Environmental Pollution and human health risk]

1) In order to investigate the actual situation of environmental pollution in the coastal area and intertidal zone, especially in the supratidal zone, environmental biomonitoring of the coastal area using wharf roach (Ligia spp.) has been conducted (Fig. 2). The target area is the coastal area of the Sea of Japan. As a result of analyzing the PAHs between the wharf roach and surrounding environmental samples in the coastal area and estimating the exposure route of PAHs, it was found that PAHs of three rings or less are mainly from food and soil. It was also suggested that PAHs with four or more rings were predominantly exposed to seawater and other environmental factors.

2) Biomonitoring of neonicotinoid pesticides and their metabolites in urine is being conducted to investigate the contamination status of neonicotinoid pesticides in humans and to evaluate their health effects, around the Sea of Japan. Urinary concentrations of neonicotinoid pesticides and metabolites were measured in three prefectures during the farming and off-farming seasons, and relatively high concentrations and frequencies of pesticides were detected in Niigata Prefecture during the farming season. This result suggests that pesticide contamination occurs universally through food, especially crops, regardless of season, but at the same time, it is thought that it also reflects the fact that the timing and amount of neonicotinoid pesticide use and the amount of exposure differ from region to region.

[Paleoenvironmental and Geological Studies]

1) We worked on environmental research in the arid area of Mongolia. Paleoenvironmental research in Mongolia has two advantages: (a) It locates in the middle of the continent with a high elevation of ~1500m, sensitive to solar incision fluctuation. Its climate is governed by three air mass, and depending on which air masses is dominant, the climate there is highly variable. (b) Human activity is less intense due to its low population, and the natural geographical environment is more or less preserved in the countryside. There are many lakes and rivers which have been lost recently because of the increase in the air temperature and decrease in precipitation, especially in the arid area. This may be attributable to the global warming caused by human activity. We studied changes in lake level and water chemistry by analyzing sediment cores from lakes, paleo-lake sediments at outcrops, and lake, river, and spring water. We compared their changes to the climate oscillation to understand how the climate impacts the environment related to water usage (Fig.3). We also found some of the toxic elements are captured in sediment, while some stay in the aqueous phase (Fig. 4).

2) Geological maps are the basis of all studies as well as various social services such as disaster prevention, environmental conservation and lifelong education. The stratigraphical survey has been carried out in the area of Kanazawa and its environs since 1994 in order to draw up high-resolution geological maps of the area.

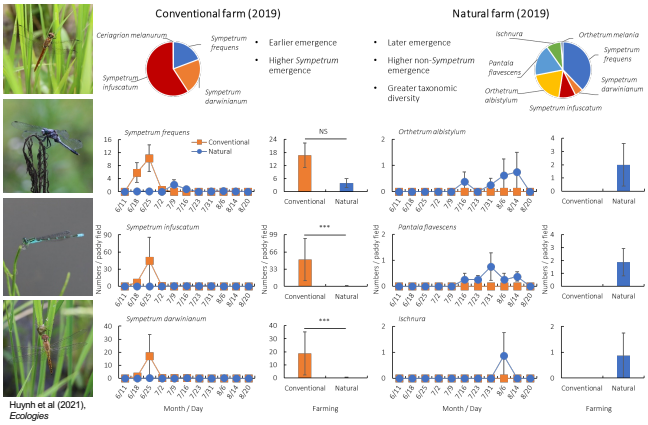
Miocene tuffaceous sandstone in the Komatsu area yields green jasper which was widely used for ornaments in the Yayoi Era of Japan (Fig. 5). In order to clarify geological and stratigraphical detail of the jasper, lithostratigraphical surveys and petrographical examinations of the tuffaceous sandstone have been carried out.

[Social and Cultural Contributions]

The Angkor World Heritage site of Cambodia is one of the most outstanding cultural heritage in the world. In order to conserve and protect the cultural heritage, natural environment and local society of the heritage (Fig. 6) and its environs, field examination and inspection have been carried out together with UNESCO and the Cambodian government.

Effects of farming practices on paddy field biodiversity —Do agrochemical-free paddy fields serve as refuge habitats for Odonata?—

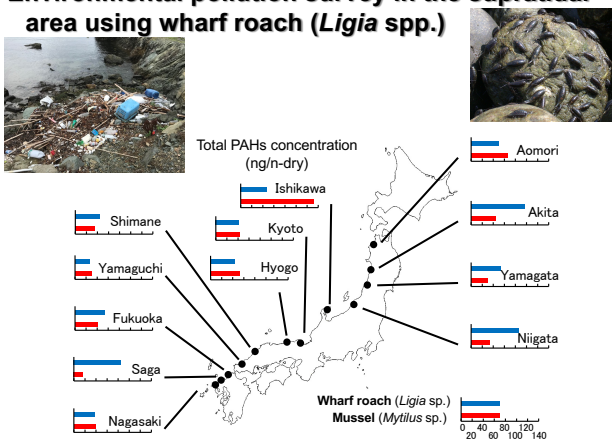
Fig.1



Huynh et al (2021), *Ecologies*

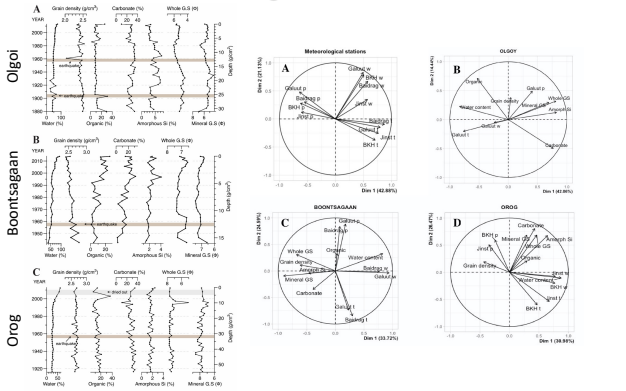
Environmental pollution survey in the supratidal area using wharf roach (*Ligia* spp.)

Fig.2



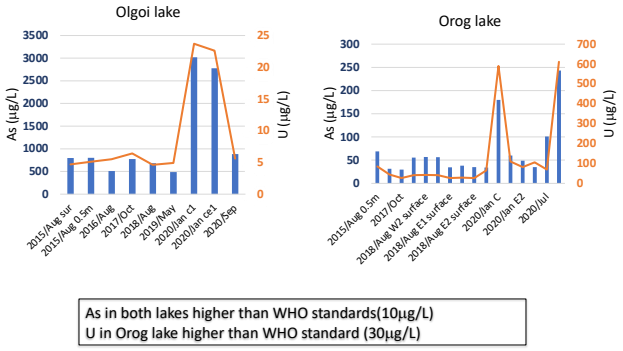
Relationship between sediment characteristics and Climate for Mongolian laes

Fig.3



As and U in Olgoi and Orog lakes, Mongolia

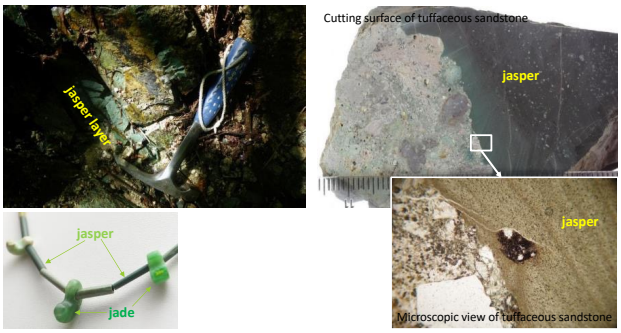
Fig.4



As in both lakes higher than WHO standards(10μg/L)
U in Orog lake higher than WHO standard (30μg/L)

Geological and stratigraphical detail of the jasper occurrence

Fig.5



Social and cultural contribution to the world heritage

Fig.6

5. Division of Integrated Environmental Studies

[Ontline]

The Low Level Radioactivity Laboratory is located in Nomi city, Ishikawa Prefecture. The division is composed of five research staff members (2017-2019) and four staff members (2020-2021). The hand-off training of Kanazawa University students and collaborative research activities with other universities and research institutes are carried out using radioactive measurement systems. This is aided by the extremely low background system constructed in the Ogoya Underground Laboratory.

In order to understand the migration behavior of chemical substances in the Earth's surface environment, it is necessary to study the transport processes within terrestrial, atmospheric, and marine environments, as well as their boundary zones. For this purpose, the division uses environmental tracers such as various stable and radioactive isotopes, and focuses on polycyclic aromatic hydrocarbons (PAHs), which is toxic chemical compounds and used as common pollutants of the K-INET. Though the use of environmental tracers and model simulations, the division carries out the integrated analysis of substances dynamics in the Circum-Sea of Japan area. The understanding fate of radiocesium released from the Fukushima Daiichi Nuclear Power Plant accident on 11 March 2011 is important to identify the transport with various processes (sorption, desorption, concentration, etc.). In final goal, we predict future circumstances by wide-range observation and model simulations, using integrated analysis of pollutants dynamics in terrestrial-atmospheric- marine environments.

[Ogoya Underground Laboratory]

Ogoya Underground Laboratory (OUL) is constructed in the Ogoya-Kuratani tunnel of the former Ogoya Copper Mine in Komatsu City. Measurement room is located in the middle of the tunnel where the overburden is 135m (270 m water equivalent) to reduce the background noise derived from secondary cosmic-ray components (muons and neutrons). Ge semiconductor detectors are shielded with the low background materials (^{210}Pb-free Pb and ^{60}Co-free Fe blocks) to achieve ultra-low background noise at about 1/100 of the aboveground value (Fig. 1). The 15 Ge detectors are installed in OUL and used for ultra-low level radioactivity measurements. The annual number of users was 771 (2017 FY), 615 (2018 FY), 685 (2019 FY), 576 (2020 FY), and 736 (2021 FY), respectively. During 2017-2019, in OUL, we particularly measured concentrations of radiocesium in seawater samples for assessments of Fukushima Dai-ichi Nuclear Power Plant accident, and other low-level radionuclides in environmental samples as geochemical tracers, including a joint usage/research collaborative researches (MEXT).

[Transport of ^{134}Cs in northern North Pacific Ocean]

Cesium-134, all of which are the 2011 Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident-derived considering its short half-life (2.06 y), is a useful tracer for seawater circulations after the supply from off northeastern Japan. The detection of ^{134}Cs in the Bering Sea in 2018 was ascribed to the arrival of FDNPP-derived radiocesium-rich Alaskan Stream water (Inoue et al., 2020). Subsequently, off the southeastern Hokkaido, Japan (Doto area), the distribution of ^{134}Cs concentrations in 2018–2019 indicated that ^{134}Cs was partially transported along with the East Kamchatka Current (EKC) from the northeastern North Pacific Ocean (Inoue et al., 2021). The ^{134}Cs concentrations at the surface in Doto area gradual increased from 2018 to 2020, and subsequently decreased to 2022 (Fig. 2a). It is considered that highly-^{134}Cs affected water masses had returned to Japan Islands side as the Oyashio Current (OYC) via EKC in the maximum effect in 2020 after the mixing with other currents and convection. Transport image of FDNPP-derived ^{134}Cs in the northern North Pacific Ocean is shown in Fig. 2b. Further study regarding ^{134}Cs circulation systems in the western North Pacific Ocean could be realized by the continual measurements of ^{134}Cs concentrations. This study is currently on-going by our research team.

[Dynamics of radionuclides released from the Fukushima Daiichi Nuclear Power Plant accident in freshwater environment]

A large amounts of radionuclides was released from the Fukushima Daiichi Nuclear Power Plant by the nuclear accident on March 11, 2011. Our laboratory has been investigating the dynamics of radiocesium in land (soil, rivers, and lakes) and coastal to pelagic marine environments. During 2017-2021, we performed the monitoring research at a fixed site in the upper Tone River, the lower Natsui River, the Abukuma River system, and rivers at Hamadori area in Fukushima Prefecture. The activity of ^{137}Cs exponentially decreased with increasing time after the accident, though the pulse input was observed at the rain events. Figure 3 shows temporal changes in the normalized dissolved ^{137}Cs concentration, $Cw(t)/D = Ae^{-(\lambda+k)/t}$, which Cw = ^{137}Cs concentration at time t, D: initial inventory of ^{137}Cs at t = 0, A: the weight of exponential component, λ+ k: effective decreasing coefficient, t: time (Ochiai et al., 2021). The regression lines was determined using data two years after the accident. The transport of dissolved ^{137}Cs from the watershed has decreased the 10 years since the accident. The slope of the regression lines is almost similar with the river systems except for the upper Tone River. Possible consideration is related to differences in watershed conditions at the lower reaches and the upper reach (Tone River). The summarized knowledge and dataset of geochemical and biological behavior of radiocesium, and tritium from the accident in freshwater systems were published as an academic book (Nagao, S. (ed.) Impacts of Fukushima

Nuclear Accident on Freshwater Environments. Springer, p.247. https://doi.org/10.1007/978-981-16-3671-4.) from Springer in 2021.

[Study on the transport processes of earth surface materials and atmospheric radionuclides in Nanao Bay]

To understand the transport processes of earth surface and atmospheric materials from terrestrial environment to downstream coastal area, studies on (1) transport processes of suspended sediment in Kumaki River in Noto Peninsula, (2) sedimentation processes in West Nanao Bay located in the downstream of Kumaki River were performed by using atmospheric radionuclides (^{7}Be, ^{210}Pb, ^{137}Cs). Additionally, (3) transport processes of these atmospheric radionuclides in the small reservoir and its catchment area.

Figure 4 shows ^{137}Cs and ^{210}Pb distribution, grain size, and magnetic susceptibility of bottom sediments in West Nanao Bay (Ochiai et al., 2022). Surface sediment concentrations and inventories of these radionuclides were relatively low in the middle and eastern areas of the bay and high in the western area, which has several major river mouths, suggesting that the contribution of riverine input affected only the western area. In the middle and eastern areas, the level of ^{137}Cs inventory was much lower than that of the soils, and the excess ^{210}Pb inventory was at the same level or lower. These results indicate that removal effects are stronger than accumulation effects (riverine input) in these areas. The patterns of radionuclides were consistent with the results, that grain size was relatively large in the middle and eastern areas, implying strong current conditions, and that fine sediment accumulated less in these areas.

[Spatial and temporal variations in marine polycyclic aromatic hydrocarbons at the coastal Sea of Japan]

Polycyclic aromatic hydrocarbons (PAHs), originating mainly from the incomplete combustion of fossil fuels and biomass and the leakage of petroleum products, are hazardous pollutants with adverse carcinogenic and mutagenic potential. Investigation of the environmental behavior of PAHs in the East Asian marginal seas is essential in assessing the ecological risk of anthropogenic PAHs, since these seas are one of the most fertile fishing ground. Concentrations of 13 phase-partitioned PAHs in seawater were monitored monthly off Okinawa Island, Oki Island, Noto Peninsula (Tsukumo Bay), Sado Island, Japan, during 2015–2021 to elucidate seasonal variations, main source, and transport pathways of PAHs in the southwestern Sea of Japan (Fig. 5A). Total concentrations of the 13 dissolved PAHs ($\Sigma_{13}PAH_{diss}$) component, contributing 95% of the $\Sigma_{13}PAH$ (= $\Sigma_{13}PAH_{diss}$ + $\Sigma_{13}PAH_{part}$) concentration, was in the range 0.43–9.32 ng L^{-1}

(mean 2.62, SD 2.08 ng L^{-1}), with higher values in summer–autumn during 2015–2019 (Fig. 5B). The main sources of dissolved PAHs were combustion products (Fig. 5C). Increasing dissolved PAH levels during July–October indicate that the area around southern Oki Island is impacted by PAH-rich summer continental-shelf water transported by the Tsushima Warm Current flowing from the East China Sea (Matsunaka et al., 2022).

[Surface and vertical distributions of polycyclic aromatic hydrocarbons in seawater at the East Asian marginal seas and Arctic Ocean]

This study clarified the surface distribution of PAHs in seawater at the East Asian marginal seas, North Pacific Ocean, and Arctic Ocean in 2019. $\Sigma_{13}PAH_{diss}$ at each ocean area in 2019 are illustrated in Fig. 6 A. $\Sigma_{13}PAH_{diss}$ in surface seawater were in the range 1.61–6.11 ng L^{-1}, with the relatively higher mean values were observed at the South China Sea, Sea of Japan, Okhotsk Sea, and Bering Sea and lower values at the East China Sea, western subarctic North Pacific, and Arctic Ocean. The $\Sigma_{13}PAH_{diss}$ in seawater collected from surface to 1000 m water depth at the South China Sea were in the range 1.96–4.12 ng L^{-1}, with the maximum values being recorded at water depth of 200 m, as found the PAH maximum layer (water depth of 100–150 m) at the East China Sea, Sea of Japan, and Okhotsk Sea in 2017 and 2019 (Fig. 6 B).

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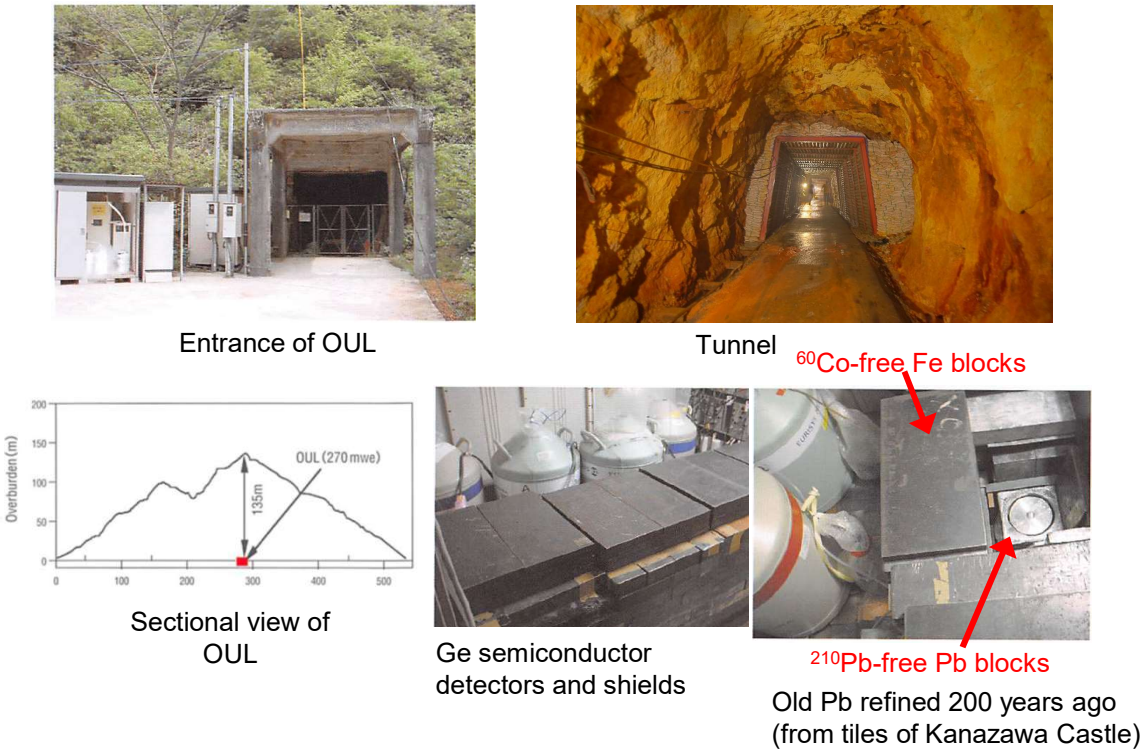

Fig.1 Outline of Ogoya Underground Laboratory (OUL).

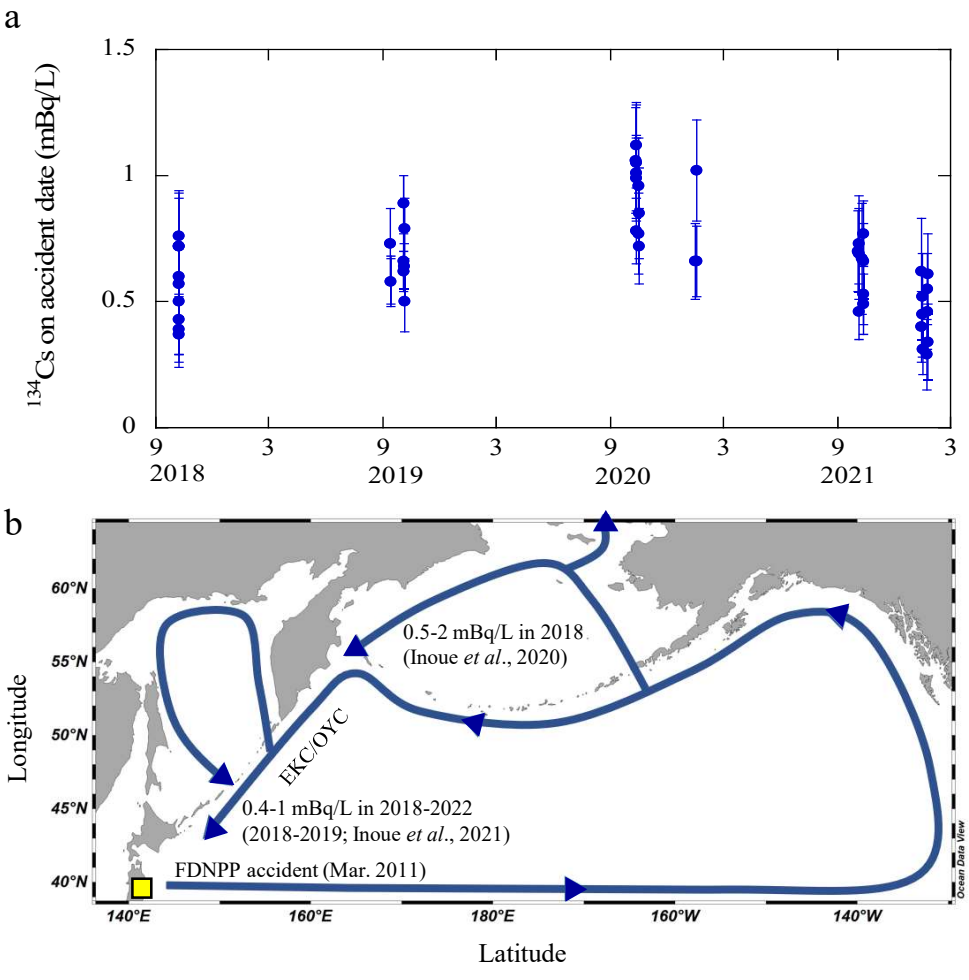

Fig. 2 a) Temporal variation of surface ^{134}Cs concentrations, decay-corrected to the FDNPP accident date, in Doto area, Hokkaido and b) the transport image in the northern North Pacific Ocean.

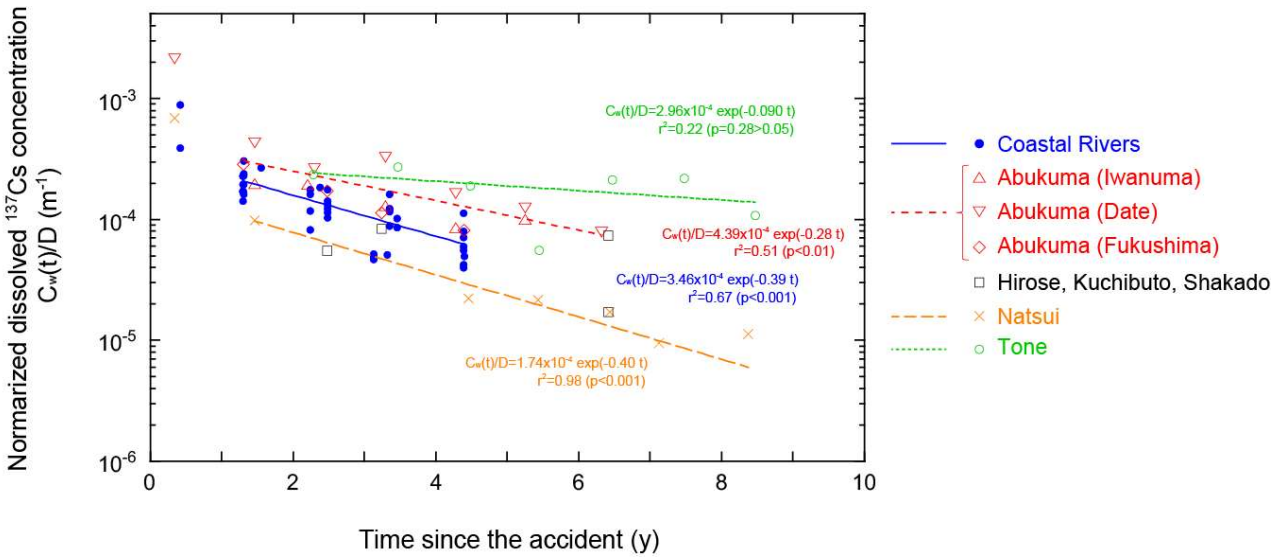

Fig.3 Temporal changes in the dissolved ^{137}Cs concentration in river water normalized by the ^{137}Cs inventory in the catchment area. Regression lines indicate the results for the coastal rivers in Fukushima area, Natui River, main stream of the Abukuma River, and Tone River, respectively.

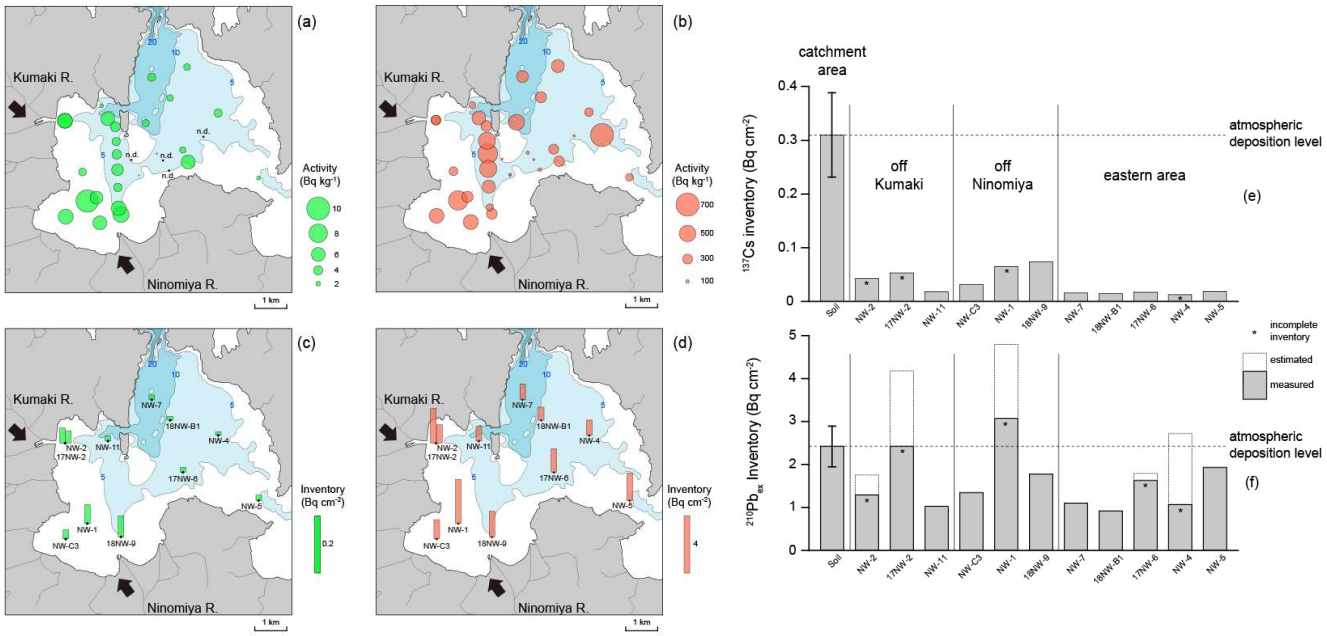

Fig.4 Spatial distributions of (a) ^{137}Cs and (b) ^{210}Pb$_{ex}$ concentrations in surface sediments and (c) (e) ^{137}Cs and (d) (f) ^{210}Pb$_{ex}$ inventories of sediment cores in the bay and the soil cores in the catchment area.

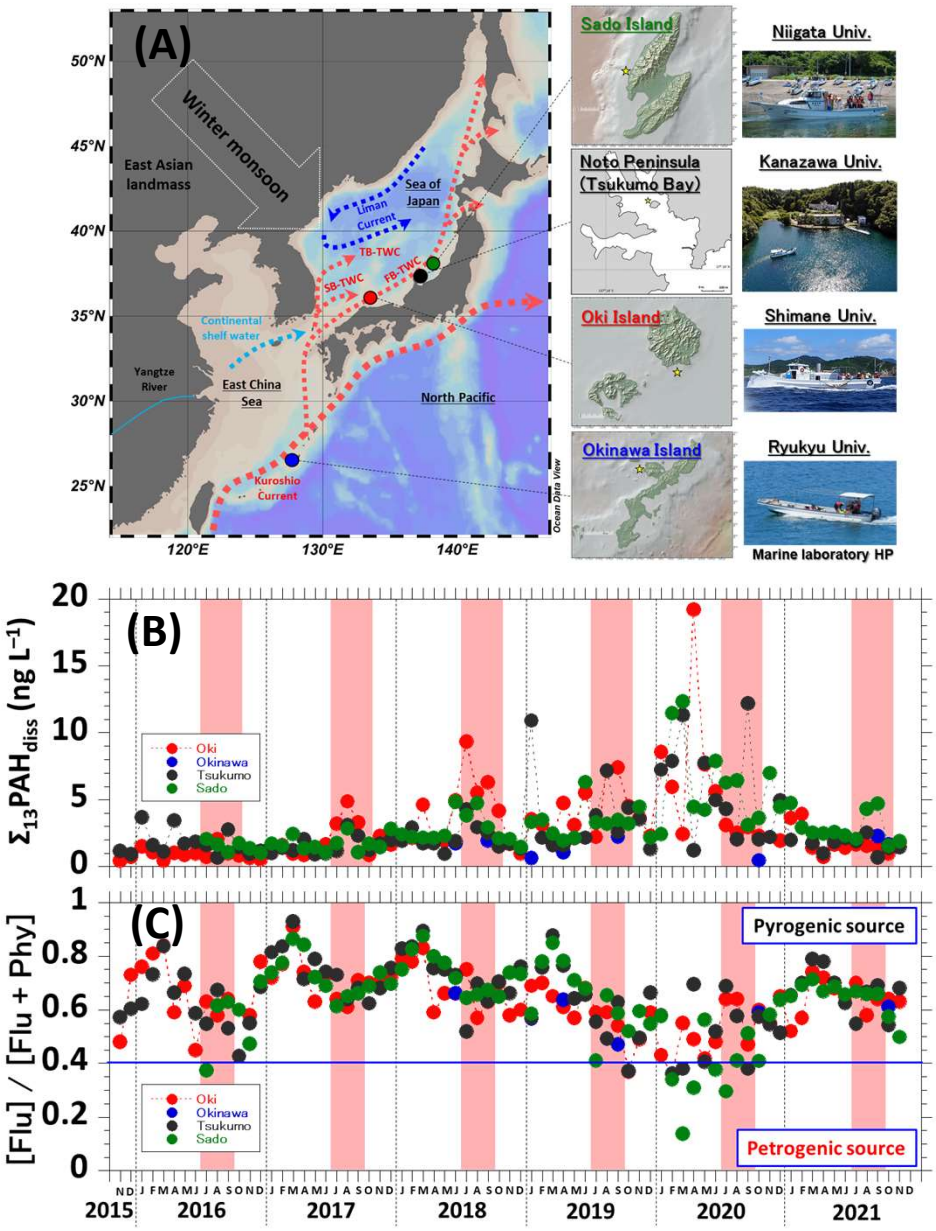

Fig.5 Schematic map of the triple branches of the Tsushima Warm Current (TWC), an offshoot of the Kuroshio Current, and the Liman Current in the Sea of Japan with the continental-shelf water and the Kuroshio Current in the East China Sea (A). Sampling sites for surface seawater near Okinawa Island, 2018–2021; Oki Island, 2015–2021; Noto Peninsula, 2015–2021; and Sado Island, 2016–2021. Monthly variations in the total concentrations of 3–6-ring PAHs for the dissolved phase (B) and PAHs diagnostic ratio of dissolved phase (C) in surface seawater at sites Oki, Okinawa, Noto, and Sado during 2015–2021. PAHs data (site Oki and Okinawa) during 2015–2019 are from Matsunaka et al. (2022).

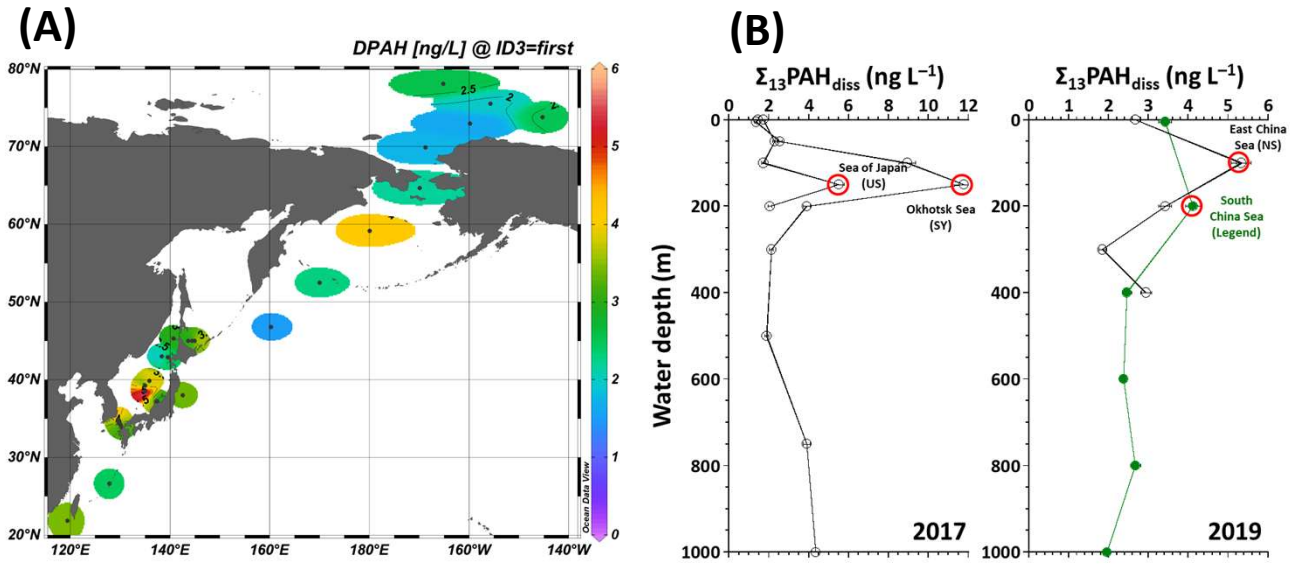

Fig.6 Surface distribution of total concentrations of 3–6-ring PAHs for the dissolved phase in seawater at the East Asian marginal seas and Arctic Ocean (A) and vertical profiles of total dissolved PAHs at the South China Sea, East China Sea, Sea of Japan, and Okhotsk Sea in 2017 and 2019.

6. Department of Inter-Institutional Collaboration

The Japan Sea Research Institute of Kanazawa University established in 1963 became the Department of Regional Studies at the Institute of Nature and Environmental Technology in 2007. Over again, the department was reorganised into the Department of Inter-Institutional Collaboration as a part of a reorganisation programme of the institute in 2016.

The Department of Inter-Institutional Collaboration manages the collection, exchange, and maintenance of environmental information in the area of entire East Asia while centred on the Circum-Sea of Japan, and it constructs and sustains international research networks in the area to disseminate information all over the world (Figs. 1 and 2). The department promotes interdisciplinary research in the area that is a geopolitically important region in the world. It also supports students' internationalisation education.

The Circum-Sea of Japan region in which industrialised countries are located can be recognised as a core of East Asia in various settings, environmental issues in particular. The department has been trying to expand its interdisciplinary activities to the whole East Asian region and to coordinate them. As a part of the activities, the department publishes a scientific journal named "Japan Sea Research ("Bulletin of the Japan Sea Research Institute, Kanazawa University" before 2017)" which contains original articles, short articles and so forth on in various research fields such as natural, human, and social sciences related only in the region (Fig. 3).

Further, the department has been organising a series of international thematic symposia "Rural and Urban Environment in the countries of the Modern Circum-Sea of Japan" since 2017, in order to understand the dynamics of the rural and urban societies in the circum-Sea of Japan countries (Fig. 4), because it is necessary to understand first the rural and urban environment which is the basement of Chinese society from the viewpoint of historical and environmental background in comparison with the society of Japan. The topics of the serious situation of China's air, water and soil pollution, which are negative aspects of industrial development have been also discussed.

The department supports students' internationalisation education at Kanazawa University. Since 2010, the department has been coordinating a students' international internship programme which sent ten undergraduate students belonging to various schools and colleges of the university to the APSARA National Authority of Cambodia which manages the conservation and maintenance of the Angkor World Heritage site as one of the most famous world heritage sites of UNESCO. The students have been engaged in the routines of the authority to learn environmental management such as

monitoring of groundwater levels, water quality surveys in local rivers and afforestation in the areas of the site (Fig. 5).

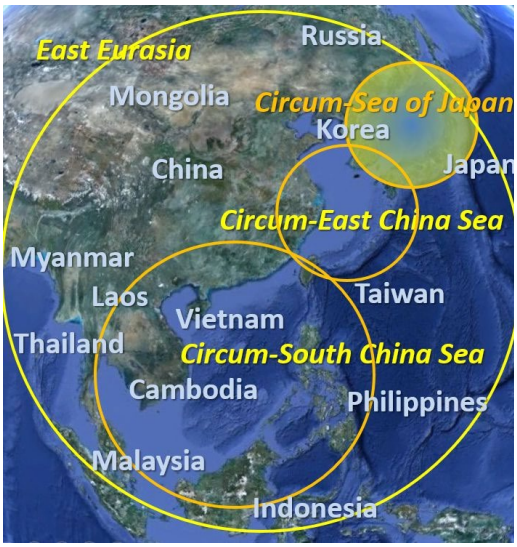

Fig.1 Geographical and geopolitical situation of the Circum-Sea of Japan region.

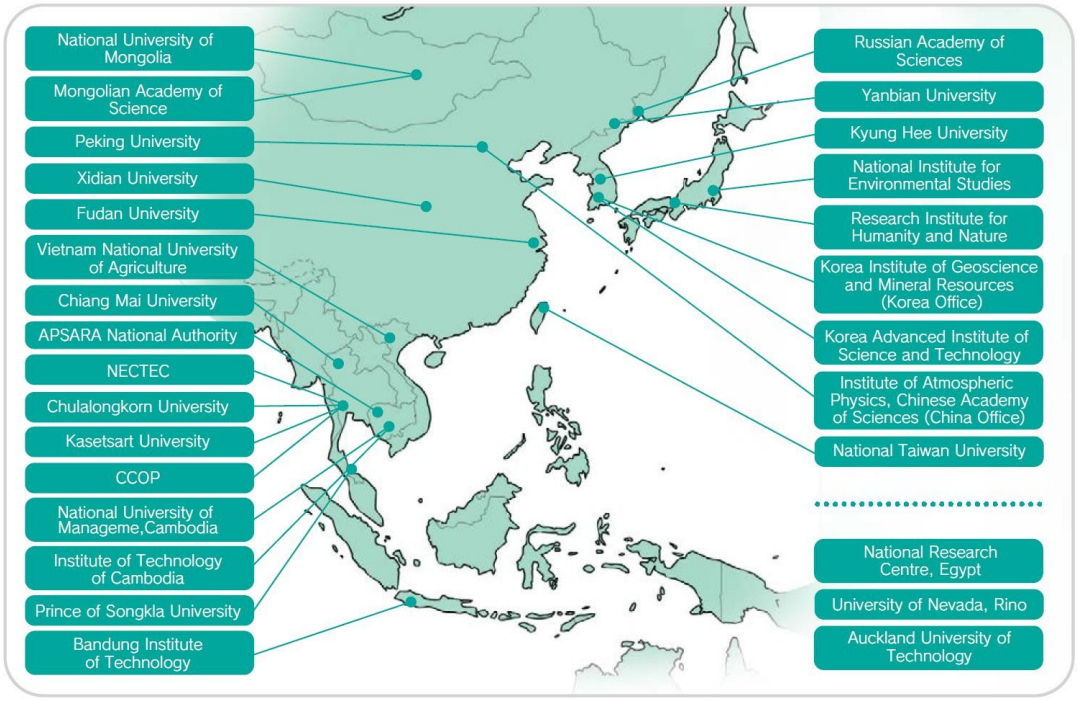

Fig.2 International and domestic network of the Institute of Nature and Environmental Technology

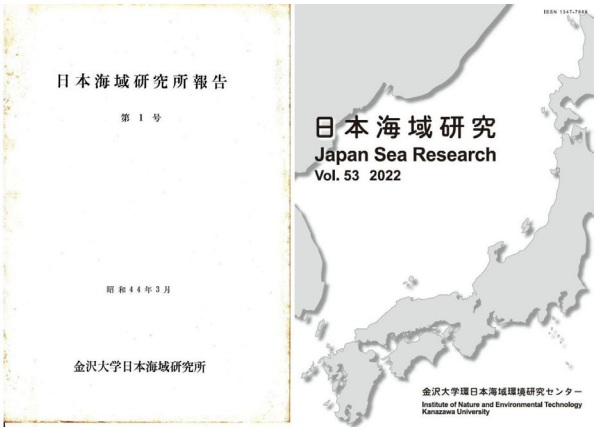

Fig.3 Covers of the journal "*Bulletin of the Japan Sea Research Institute, Kanazawa University*" volume 1 in 1969 and "*Japan Sea Research*" volume 53 in

Fig.4 The 3rd International Thematic Symposium held in East China Normal University, Shanghai in

Fig.5 International internship programme at the APSARA National Authority of Cambodia in 2019.

Publication list from fiscal year 2017 to 2021

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