iLEAPS / IGAC-Japan 合同研究集会 2019

iLEAPS / IGAC-Japan Joint Workshop 2019

持続可能な地球社会の実現をめざす国際協働研究のプラットフォームであるフューチャー・ア ースには、大気/陸域境界のエネルギー・物質の交換やそれに関わる諸過程の解明を目指す iLEAPS、また、大気化学の視点から物質循環・大気過程・化学組成の解明を目指す IGAC などが 組織されています。iLEAPS・IGAC の学術分野の課題には、温室効果気体や生物起源揮発性有機 化合物(BVOC)の大気陸域間の収支や、気候と生態系の相互作用における大気微量成分の役割 など双方にまたがるものがあり、今後、これらの課題に両分野の研究者が連携して取り組むこと が重要になると考えられます。そこで本研究集会では、日本学術会議の iLEAPS・IGAC 小委員会 の委員を含む双方の学術コミュニティの研究者が集い、大気陸域間の相互作用に関わる研究につ いて話題提供と討論を行います。そして、両分野にまたがる重要課題を明確にし、その解明に向 けた今後の研究の展望を得ることを目指します。また、両分野が連携した研究の推進に向けて、 iLEAPS・IGAC 小委員会や国内の両コミュニティで今後重要となる取り組みを見出すことも本集 会の目標とします。

- 日時: 2019年9月6日(金) 13:00~9月7日(土) 16:00
- 場所: 名古屋大学宇宙地球環境研究所 研究所共同館 II-409 室(愛知県名古屋市千種区不老町) http://www.isee.nagoya-u.ac.jp/directions.html
- 主催: 名古屋大学宇宙地球環境研究所・日本学術会議 iLEAPS 小委員会・ 日本学術会議 IGAC 小委員会
- 幹事: 持田 陸宏(名古屋大学・日本学術会議 IGAC 小委員会副委員長) 檜山 哲哉(名古屋大学・日本学術会議 iLEAPS 小委員会委員長)

プログラム(各講演の時間配分:発表 15 分・質疑 10 分)

9月6日(金)・一日目

- 13:00~13:05 趣旨説明(持田 陸宏・名古屋大学)
- 13:05~13:10 iLEAPS 小委員会委員長 挨拶(檜山 哲哉・名古屋大学)
- 13:10~13:15 IGAC 小委員会委員長 挨拶(谷本 浩志・国立環境研究所)
- 座長:檜山 哲哉(名古屋大学)

13:15~13:40高橋 けんし (京都大学): Methane emission from woody stem and its
potential impact on atmospheric methane budget413:40~14:05高野 倫未 (大阪府立大学): Year-round measurements of methane fluxes in

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- urban area
- 14:05~14:30坂本 春樹 (東京大学): Modeling methane emissions from natural wetlands
using a terrestrial ecosystem dynamics model (S-TEDy)

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座長:谷本 浩志	(国立環境研究所)	
15:10~15:35	米村 正一郎(農研機構): Chemical kinetics of soil N ₂ O emission	8
15:35~16:00	松田 和秀(東京農工大学): Exchange of gaseous and particulate reactive nitrogen between the atmosphere and forests	9
16:00~16:25	金谷 有剛(海洋研究開発機構): Boreal climate change and fires: drivers of atmospheric composition change and impact on the Earth surface system	10
16:25~16:50	森野 祥平(名古屋大学): Interannual variation of summer precipitation and atmospheric water circulation over and around Alaska	11
16:50~17:15	佐藤 永(海洋研究開発機構):Reconstructing and predicting global potential natural vegetation with a deep neural network model	12
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- 9月7日(土)・二日目
- 09:00~10:00第 24 期・第 3 回 iLEAPS 小委員会(IGAC 小委員会メンバーのオブザーバー参
加可能・非公開)
- 10:00~10:15 休憩
- 座長:持田 陸宏(名古屋大学)

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11:30~11:55	鄧 彦閣(名古屋大学): Contribution of Biogenic Secondary Organic Aerosols to Cloud Condensation Nuclei Concentrations at a Forest Site in the Kii Peninsula, Japan	16
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(講演者の氏名・所属を記載)

Methane emission from woody stem and its potential impact on atmospheric methane budget

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It has been suggested that natural wetlands constitute a crucial contribution for atmospheric methane at present (IPCC, 2013). Anoxic wetland ecosystems are considered as the largest contributor to natural methane emissions, accounting for more than 20% of the global methane source. However, methane emissions from wetlands are highly variable, both spatially and temporally and at scales ranging from microtopographic to regional differences. To comprehend this variation fully and also to predict responses to climate change, an understanding of the intimate linkage between carbon cycling and methane emission in these systems is urgently required.

Recently, wetland-adapted trees have attracted a considerable attention because of its potential significance as a new emission source of methane (e.g., Gauci et al., 2010; Pangala et al., 2013), in which methane molecules produced by methanogens in soil are believed to be transported upward inside the stem and diffused to the atmosphere through woody stem surfaces. Additionally a recent study has suggested that methanogenic archaea can inhabit in heartwood, which might provide a source of methane emitted from woody stem (Yip et al., 2019). However, the magnitude and controls of tree-mediated emission processes remain unknown.

In our study, significance of plant-mediated methane transport was examined in a riparian wetland of a forest water catchment, in the central of Japan. A near-infrared laser spectroscopy instrument and closed chamber systems enables us in-situ continuous measurements of methane emission rates from the stem surfaces of Japanese alder trees (*Alnus japonica* (Thunb.) Steud.), revealing that meteorological conditions and soil environment are associated with the temporal variations in the methane emission rates. Individual differences in the magnitude of methane emission are also analyzed in detail.

Year-round measurements of methane fluxes in urban area

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Measurements of urban greenhouse gas emissions are important. In particular, methane (CH₄) fluxes were rarely measured in urban areas; thus its budgets and variabilities are highly uncertain. Limited studies reported that emission inventories underestimated urban CH₄ emissions. Continuous flux measurements and mobile measurements of the gas concentrations are effective tools for investigating the spatial-temporal variabilities of CH₄ emissions from urban areas.

In this study, CH₄ fluxes were measured at two locations in Sakai, Osaka, Japan, using the open-path eddy covariance method since 2018. The area of the city is about 150 km², and its population is approximately 840,000 residents. The site surrounding the city center (SAC) was characterized as a densely built-up, consisting of industrial, commercial, and residential areas. The site located at an edge of the university (OPU) was characterized as suburb, consisting of university buildings, vegetation, and residential areas. The mobile measurements of CH₄ concentrations were conducted around the two sites using a vehicle at the summer of 2019.

The SAC site emitted CH_4 throughout the year. The CH_4 fluxes showed a clear diurnal variation, but an unclear seasonal variation. Based on a mean diurnal cycle from October 2018 to July 2019, the CH_4 fluxes had a daytime maximum, about 50 nmol m⁻² s⁻¹, around 3PM, which was approximately four times greater than a nighttime minimum. The high CH_4 emissions during the daytime could be caused by high emissions from the vehicle traffic, natural gas consumption, and/or sewerage systems in the daytime. Because emissions from imperfect combustions of natural gas could seasonally change owing to seasonal change in use of gas-powered air conditioners, the unclear seasonal variations in CH_4 fluxes suggest that contributions of imperfect combustions could be small.

The OPU site emitted CH₄ throughout the year. The CH₄ fluxes showed a clear diurnal and seasonal variation. Based on a mean diurnal cycle from June 2018 to July 2019, the CH₄ fluxes had a daytime maximum, about 40 nmol m⁻² s⁻¹, around 1PM, which was approximately three times greater than the nighttime minimum. Although the diurnal variations in the CH₄ flux were observed in both the summer and winter, the daytime maximum was greater in the summer than other seasons. From July to September, based on a mean diurnal cycle, the daytime maximum of CH₄ fluxes was over 60 nmol m⁻² s⁻¹. Consequently, for the seasonal scale, higher CH₄ fluxes were observed in the summer than other seasons. Annual emissions of CH₄ in OPU site was about 18 g CH₄ m⁻² year⁻¹, which was similar to that observed in the city of Łód'z, Poland (Pawlak et al., 2016).

Hotspot increases of CH_4 concentration (up to 2.7 ppm) were detected within the flux footprint for SAC by the mobile measurements. Among the hotspots, some high CH_4 concentrations were reproducible, which could be caused by natural gas leakages. Previous studies in United States reported that high CH_4 concentrations over 10 ppm were observed due to natural gas leaks from distribution pipelines (von Fischer et al., 2017), but such high concentrations were not observed in this area. This indicates that the gas leakages in the study area were lower than those measured at United States.

Reference

Pawlak et al. (2016) Atm. Chem. Phys., 16, 8281-8294. von Fischer et al. (2017) Environ. Sci. Technol., 51, 4091-4099.

Modeling methane emissions from natural wetlands using a terrestrial ecosystem dynamics model (S-TEDy)

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The emission of greenhouse gases into the atmosphere is causing significant changes in climate. Methane is one of the important greenhouse gases, and natural wetlands account for 50% of the natural methane emissions. Tundra ecosystems in particular are estimated to include 14% of the global soil carbon and account for between 20 and 25% of the natural methane emissions. Climate change has the potential to increase methane emissions, and increase methane emissions would induce further climate change, resulting in a positive climate feedback. Therefore, the estimation of methane emission from wetlands including tundra ecosystems is an urgent issue.

In this research, methane and carbon dioxide emission from natural wetlands are simulated by using the SEIB-originated Terrestrial Ecosystem Dynamics (S-TEDy) model. In this model, the underground carbon and nitrogen dynamics are simulated based on a carbon-nitrogen cycle model developed by Porporato et al., 2003. Methane emission can be simulated by adopting anaerobic respiration into this carbon-nitrogen cycle. In this way, methane emission can be simulated considering the environmental changes (such as changes of plant growth or vegetation types) induced by climate change.

Reference

Porporato, A., D'Odorico, P., Laio F., Rodriguez-Irtube, I., 2003. Hydrologic controls on soil carbon and nitrogen cycles. I. Modeling scheme. Adv. Water Res., 26, 45–58

Estimating CH₄ fluxes across high latitude ecosystems using VISIT model

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High-latitude warming has increased greenhouse gas emissions from northern ecosystems, resulting in a positive feedback on the climate system. Process-based ecosystem models have been used to estimate highlatitude methane (CH₄) fluxes. These estimates have large uncertainties due to a lack of constraints on observational data. In this study, we simulated CH4 fluxes across high-latitude ecosystems using a process-based terrestrial ecosystem model, VISIT (Ito and Inatomi, 2012). The model was calibrated using 17 multi-year eddy covariance data set from cool temperate, boreal, and arctic research sites (71 site-years). In high-latitude ecosystems, the soil temperature profile and water table depth are major factors determining CH₄ flux; thus, we introduced a soil heat conduction component into VISIT for better representing soil thawing-freezing processes. In addition, water table depth was simulated by a water balance model optimized through observational data. Six parameters of VISIT were optimized using CO₂ and CH₄ fluxes at the high-latitude ecosystems, including fen, bog, lowland forest, and tundra sites. We selected Q_{10} (temperature coefficient for methane production), R_0 (constant rate factor for methane production) and Pox (fraction at which methane is oxidized during plantmediated transport) for optimizing the sub-model for CH4 flux. We classified the observation sites into four categories (peatland, forested peatland, dry tundra and wet tundra), and optimized model parameters for each category. The differential evolution method, a global search algorithm for parameters, was used to optimize the model parameters. As a cost function, the Nash-Sutcliffe model efficiency coefficient was maximized. We used the best model parameters for estimating the CO₂ and CH₄ budgets across the terrestrial wetlands north of 40°N. In this presentation, we will compare the simulated CO₂ and CH₄ fluxes based on the optimized and default parameters from 1901 to 2018. By comparing these fluxes with the top-down estimates, we will discuss uncertainties in the simulated fluxes across spatial and temporal scales.

Reference

Ito, A., & Inatomi, M. (2012). Biogeosciences, 9(2), 759-773.

Chemical kinetics of soil N₂O emission

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Introduction

It is needless to say that N_2O is one of the critical greenhouse gases in the atmosphere. N_2O is emitted through nitrification/denitrification processes induced by application of nitrogen fertilizers. So, to understand N2O dynamics in soil and its mitigation can be considered to be one very important theme in Future Earth Program. However, it is well known that predictability of N_2O emission from soil is poor, in contrast to CO_2 and CH₄ emissions from soils. The poor predictability of N_2O emission from soil is that many processes including non-biological production contribute to N_2O production in soil; N_2O is a by-product in the nitrification/denitrification processes, in contrast that CO_2 is a final product of oxidation under aerobic environments and that CH₄ is a final product under anaerobic environments. So, the present studies are mostly oriented towards direct relation between N_2O emissions and related agricultural managements from the mitigation point of view. Moreover, it is generally estimated that denitrification processes contribute more terrestrial soil N_2O emission than nitrification processes.

To understand N_2O emission soil, sophisticated system to monitor N_2O emission and other related gases under various controlled conditions in laboratory. We developed a system to monitor them in laboratory to be available for various gas-exchange processes of soil and other objects in agro-ecosystems. Using the system, we began to investigate chemical kinetics of soil N_2O emission. We show tentative results in this presentation.

Materials and Methods

The measurement system (Yonemura et al., 2019a; 2019b) was improved in that to in addition to the original bone system. The system can continuously measure simultaneous exchanges of CO₂, CH₄, H₂, CO, NO and N₂O by soil. The main point is: control of oxygen concentration to realize aerobic and anaerobic conditions, control of C_2H_2 concentration to control nitrification and denitrification processes.

As a test to control reaction of denitrification processes, we controlled the ambient O_2 concentration as low as possible, added N_2O itself and observed soil emissions of N_2O , NO, and N_2 . We hypothesized that N_2 emission increases linearly with increasing N_2O concentration, reflecting $N_2O \rightarrow N_2$ reaction in the denitrification processes.

Results

The results did not meet the hypothesis-- N_2 emission did not increase as increase of N_2O concentrations (Fig. 1). The more N_2O emission under higher moisture (50%WHC) shows that trap of N_2O by higher moisture is very important to the reaction to N_2 .

Discussion and Future Perspectives

Further, from the information of Fig.1. We now can calculate N_2O concentration in soil and reaction coefficients in soil (kinetics). Please being looking forward to our presentation.



Yonemura S, Uchida M, Iwahana G, Kim Y, Yoshikawa K, 2019a: Technical advances in measuring greenhouse gas emissions from thawing permafrost soils in the laboratory. Polar Science 19, 137-145.

Yonemura S, Kaneda S, Kodama N, Sakurai G, Yokozawa M, 2019b: Dynamic measurements of earthworm respiration. Journal of Agricultural Meteorology 75(2), 103-110.

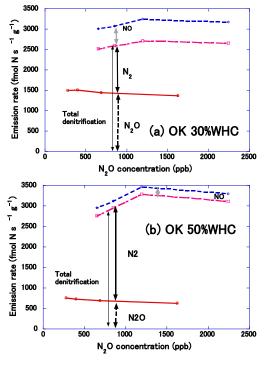


Fig. 1 N₂O, N₂ and NO emissions from (low land) soil. WHC means Water Holding Capacity.

Exchange of gaseous and particulate reactive nitrogen between the atmosphere and forests

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1. Introduction

The Asian region has been identified as a high-risk area for nitrogen deposition effects on ecosystems. According to a comparison of inferential models used to estimate dry deposition rates for reactive nitrogen across the NitroEurope network (Flechard et al., 2011), there was a large discrepancy in the deposition velocity (V_d) of nitrogen compounds such as HNO₃, NH₃, NO₃⁻ and NH₄⁺ among the models, especially at the forest surface. In recent PM_{2.5} simulations conducted using chemical transport models for the East Asian region, the modeled results tended to overestimate the PM_{2.5} nitrate concentration, and the simulated nitrate concentration was highly dependent on V_d of nitrogen compounds (Shimadera et al., 2014). On the other hand, the dry deposition process has rarely been researched for the East Asian region. Therefore a better understanding of the dry deposition process will improve not only the nitrogen deposition process at forests in East Asia, especially focusing on the researches made in the project "Mechanism of gas and particulate reactive nitrogen deposition for evaluation of nitrogen load to ecosystem (JSPS KAKENHI 16H02933)".

Measurements were done in three different types of forests located in Tokyo (FM Tama site), Hokkaido (Teshio site) and Thailand (Sakaerat site) by using the experimental towers established in each forest furing the period from 2016 to 2018. Vertical profiles of inorganic compounds in $PM_{2.5}$ at all sites, and those of HNO₃ and NH₃ at FM Tama and Teshio were measured. The fluxes and deposition velocities were measured at FM Tama site by relaxed eddy accumulation (REA) method (Matsuda et al., 2015). The denuder method and the filter pack method were used for the sampling of the components. 3. Results and discussion

The vertical profile measurements clearly showed the difference in dry deposition process between NO_3^- and SO_4^{2-} in $PM_{2.5}$. Concentration gradients of NO_3^- in $PM_{2.5}$ from over to under the canopy were significantly higher than those of SO_4^{2-} in $PM_{2.5}$ at all sites (Fig. 1). In the REA measurements at FM Tama (October to December 2016), representative values of deposition

velocity were estimated to be 0.80 cm/s for SO42- in PM_{2.5}, 1.4 cm/s for NO₃⁻ in PM_{2.5} and 1.9 cm/s for HNO₃ (Sakamoto et al., 2018). The differences of vertical profiles and deposition velocities indicated that NO3⁻ in PM_{2.5} was more efficiently deposited and removed in the forest than SO42- in PM2.5. It was considered that the efficient removal of NO3- in PM2.5 was associated with the process of the conversion between NH₄NO₃ and HNO₃/NH₃ (Sakamoto et al., 2018; Nakahara et al., 2019). Bi-directional exchange of NH₃ were found at FM Tama and Teshio. NH₃ fluxes calculated by using default input parameters for forest surface suggested by Zhang et al. (2010) were not in accordance with the observations. Trends of emission or deposition were influenced by some environmental factors such as wet or dry conditions of surface, ammonia emission potential of stomata and soil. These deposition processes above could influence the behavior of relevant reactive nitrogen compounds in the atmosphere significantly. Reference

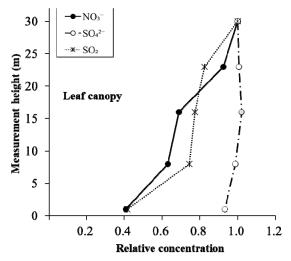


Fig. 1 Vertical profiles of relative concentration of NO₃ in $PM_{2.5}$, SO_4^{-2} in $PM_{2.5}$, and SO_2 during the daytime from 27 September to 11 October in 2016 at FM Tama site.

Flechard et al., Atmospheric Chemistry and Physics 11, 2703–2728 (2011); Shimadera et al., Journal of the Air & Waste Management Association 64, 374–387 (2014); Matsuda et al., Atmospheric Environment 107, 255–261 (2015); Sakamoto et al., Journal of Japan Society for Atmospheric Environment 53, 136–143 (2018) (in Japanese).; Nakahara et al., Atmospheric Environment 212, 136–141 (2019)

Boreal climate change and fires: drivers of atmospheric composition change and impact on the Earth surface system

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In this presentation, our recent studies are introduced, particularly those over the boreal zone including Russia and Alaska, with relevance to the atmospheric chemistry- terrestrial ecosystems interaction. The region is characterized with recent rapid warming and large fire events. The frequent fires may emit larger amount of black carbon particles into the atmosphere contributing to warming, and thereby form a positive feedback loop. On the other hand, higher temperature may result in larger emission of biogenic volatile organic compounds (BVOCs) facilitating production of secondary organic aerosol (SOA), to form a negative feedback loop. While individual studies to be exemplified here are on very basic processes and yet fragmentary, recognition of such overarching feedback loops among systems is important.

First, we introduce results of MAX-DOAS (Multi-Axis Differential Optical Absorption Spectroscopy) observations targeting formaldehyde (HCHO) at Zvenigorod Scientific Station (55.70N, 36.78E, 208 m asl), IAP/RAS, ~50 km west of Moscow, since October 2008. The tropospheric vertical column density of HCHO commonly showed summertime peaks in multiple years. The peak reached 5×10^{16} cm⁻² in 2010, under the strong influence of forest fires, which was ~3 times larger than those in the adjacent two years. For the cases without influence from forest fires or urban plumes, clear positive temperature dependence of HCHO was found, suggesting formation of HCHO via oxidation of natural VOCs. The observations are fundamental to evaluate satellite observations essential to scaling up.

Second topic is on the black carbon emission from boreal forest fires. Using higher resolution (better than 30 m) satellite products (Landsat 7 ETM+, RapidEye etc.), we found that small fires are significantly missed from burned area product (MCD64A1) on which a frequently-used emission inventory (e.g., GFED) of black carbon is based. At Poker Flat Research Range (PFRR, 65.12N, 147.43W, 500 m asl), central Alaska, USA, we found that a large variation of Δ BC/ Δ CO emission ratios from fires could be interpreted in a consistent manner in terms of fire radiative power, enabling better estimation of BC emission.

When time permitting, yet more studies and topics encompassing oceanic regions are introduced, including a case where a giant forest fire plume from Siberia resulted in elevated number densities of bioaerosols and ice nucleating particles over the Bering Sea, after long-range transport.

Interannual variation of summer precipitation and atmospheric water circulation over and around Alaska

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Arctic warming is more than twice as fast as the global warming. Water cycle in the region is changing due to the climate warming. Variations of atmospheric water circulation influence pan-Arctic terrestrial ecosystems through precipitation and soil water content. Ohta et al. (2014) indicated that summer precipitation increased in eastern Siberia from 2005 to 2008, resulting in waterlogging and damaged conditions of a larch forest in the region. Hiyama et al. (2016) revealed that a drastic shift of the atmospheric circulation pattern in Siberian pan-Arctic region resulted in the increase in summer precipitation in eastern Siberia. For interior Alaska, a recent observation showed that an increase in summer precipitation stimulated methane emissions at a black spruce forest. This study investigated interannual variabilities of summer precipitation and associated atmospheric water circulation in interior Alaska using the ERA-Interim reanalysis data and rain gauge observation data in Fairbanks from 2003 to 2017.

We extracted dry and wet years based on summer (June, July, and August) precipitation time series, and then conducted composite analyses. Geopotential height and horizontal wind at 850 hPa as well as vertically integrated moisture flux were composited. We found that the change in low-level westerly wind and moisture flux blowing into interior Alaska was a primary factor to induce the interannual variation of the summer precipitation in this region. This westerly moisture flux was associated with the atmospheric circulation pattern with cyclonic and anticyclonic anomalies over and around Alaska. Moreover, the anomaly patterns were related to the numbers and tracks of cyclones. More cyclones were detected in the cyclonic anomaly, while less cyclones in the anticyclonic anomaly over the region. Interestingly, it was found that the interannual variation of the atmospheric circulation pattern over the region was modulated for the period from 1979 to 2017 on decadal time scales. The running correlation between the summer precipitation and PDO (Pacific Decadal Oscillation) index suggests a regime shift occurred in the atmospheric circulation over and around Alaska in late 1990s.

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Reconstructing and predicting global potential natural vegetation with a deep neural network model

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Potential natural vegetation (PNV) is the vegetation cover equilibrium with environmental condition, which would exist at a given location without human land-conversion. For operational mapping of PNV, we developed an empirical model using a deep neural network (DNN), which was trained by an observation based PNV map (Figure 1) and graphical images of global air temperature and precipitation at 0.5 degree resolution. The trained model well reconstructs an observation based global PNV map, demonstrating that this way of DNN application can capture empirical relationships between PNV and climate. Then, the trained model was applied to projected climate at the end of the 21st century, predicting significant shift of global PNV distribution with rapid warming trends (Figure 2).

Figure 1.

An observation based potential-natural-vegetation (PNV) map of the ISLSCP2.

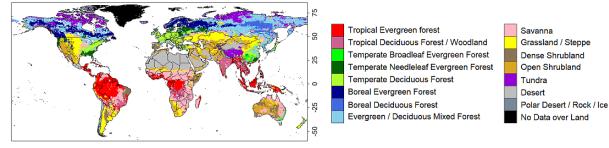
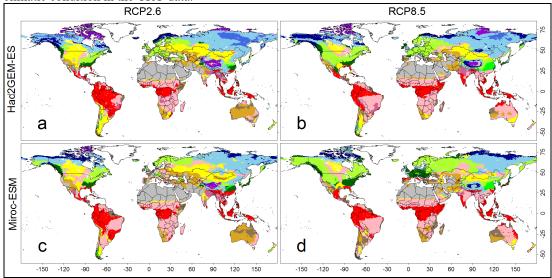


Figure 2.

Predictions of PNV map under the forecasted climate during 2091 to 2100. Monthly means of four forecasted climatic conditions, those are the combinations of two climate models (Had2GEM-ES and Miroc-ESM) and two RCP scenarios (RCP2.6 and RCP8.5), were applied for the DNN model that was trained by present climatic condition in the CRU data.



Bidirectional exchange of Biogenic VOCs between terrestrial ecosystems and atmosphere

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Biogenic VOCs including isoprene and monoterpenes are emitted by many plant species. Because of high reactivity and large amounts of emissions of these compounds, they might to significantly contribute to production of photochemical oxidants and secondary organic aerosols.

We have measured BVOC emissions by using leaf cuvette, blanch enclosure and micrometeorological flux measurement methods, to qualify the emission strength of various tree species in Japan. The isoprene emission is affected by temperature and light intensity. Monoterpene emission is governed by temperature if the compounds are stored in special storage organs of plants. If monoterpenes are immediately emitted after being produced by some plants, the emission is controlled by temperature and light intensity. For tree species grown in Japan, isoprene emission rate of broad-leaved tree species is generally ten times higher than monoterpene emission rate of coniferous tree species. We have revealed that isoprene and monoterpenes emitted by several tree species are also affected by water stress, integrated temperature and CO_2 and ozone concentrations.

On the other hand, isoprene emitted into the atmosphere is easily oxidized to produce methacrolein and methyl vinyl ketone. We have shown that these compounds are readily absorbed through stomata of plant leaves. The uptake rates are affected by the degree of the stomatal opening, i.e. proportional to stomatal conductance. Using many kinds of VOCs, we examined what kinds of VOCs were quickly absorbed through stomata and, in some cases, how these compounds were converted inside plant leaves. We show that some VOCs absorbed by plants are reduced inside leaves and emitted into the atmosphere after being modified.

In this presentation, we will show the recent progress of study on bidirectional exchange of Biogenic VOCs between terrestrial ecosystems and atmosphere.

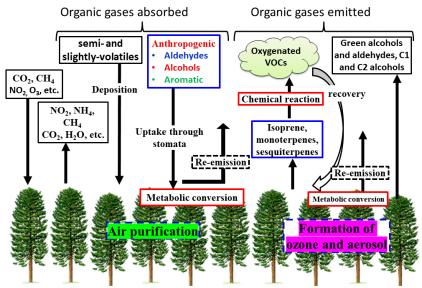


Figure 1 Trace gas species exchanged between plants and atmosphere

Observational study on emission of biogenic volatile organic compounds (BVOCs) from vegetation in terms of ozone reactivity

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Volatile organic compounds (VOCs) have been focused on in atmospheric chemistry as critical precursors of photochemical oxidants (tropospheric ozone) and secondary organic aerosols (SOAs). VOCs are emitted into the atmosphere from biogenic sources such as vegetation and plants, as well as from anthropogenic sources. It has been estimated that the annual global VOC flux is 1150 TgC [1], consisting of 44% isoprene and 11% monoterpenes, both of which are biogenic VOCs (BVOCs). To investigate the impact of BVOCs on air quality and pollution, atmospheric BVOCs have been one of the most discussed topics in atmospheric chemistry in recent decades. Contributions of BVOCs to photochemical oxidants and SOAs, observations and estimation of emission factors of BVOCs such as isoprene and monoterpenes, and laboratory experiments to establish the mechanisms of SOA formation from BVOCs have all been investigated. From the standpoint of air pollution and air quality, it is important to investigate detailed emission characteristics of BVOCs from plants. It has been reported that these emissions depend on meteorological conditions (e.g., air temperature, intensity of solar radiation, wind speed, relative humidity) and canopy environment (e.g., broadleaf trees and needle trees, leaf temperature). Thus, to study the emission of BVOCs from plants in detail, it is essential to observe the emission flux based on sensitive and fast-response measurements of various BVOC compounds. Gas chromatography (GC) is widely used to analyze gaseous VOCs. GC-based analyzers can quantify concentrations of multiple VOC compounds simultaneously. However, monitoring of VOCs by GC is based on batch analysis and is unsuitable for fast-response, continuous, real-time analysis. Proton transfer reaction-mass spectrometry (PTR-MS) has been developed as a real-time analyzer of VOCs. However, isomers of monoterpenes cannot be separated and PTR-MS cannot characterize certain reactive alkenes. Furthermore, VOCs and BVOCs include a vast number of compounds and it is difficult to measure all of these by individual analyses.

Most BVOCs have C=C double bonds and can react with O₃. In this study, a new system that uses the total reactivity with ozone (R_{O3}) was developed for measuring BVOCs emitted from vegetation into the atmosphere [2,3]. The decrease in ozone caused by the reaction with BVOCs was monitored at the ppbv level by fast-response dual chemiluminescence detectors (CLDs). A glass double-tube was adopted as the reactor. Gaseous cyclohexane was added to the sample in order to scavenge secondary OH radicals. From the characterization of the R_{O3} analyzer using the standard VOC sample, the dependence of ozone reduction on the reaction time and reactivity were shown to agree with theoretical predictions. A calibration procedure for determining the reaction time was also established. The

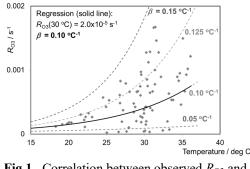


Fig.1 Correlation between observed R_{O3} and ambient temperature.

detection limit of the analyzer reached $2x10^{-5}$ s (S/N = 3, 60-s average, 50-s reaction). It was confirmed that the R_{O3} analyzer was capable of measuring BVOC levels. A practical trial was conducted in which BVOCs emitted from a real needle-leaf tree were monitored. BVOC emissions from the tree were detected and a significant increase in R_{O3} was observed when the tree was irradiated with light. To demonstrate the validity and usefulness of the analyzer, observation tests of R_{O3} in the ambient air were conducted at a suburban site in Japan (Tokorozawa campus, Waseda University) [4]. R_{O3} was significantly captured when the temperature was high during daytime in summer. Fig. 1 shows an example of correlation plot between the observed R_{O3} and the ambient temperature. It was found that observed R_{O3} increased with temperature increasing. Observed R_{O3} could be explained by temperature dependence of BVOCs emission from plants.

As future subjects of our studies on ozone reactivity, followings are important: (1) Practical examples of R_{O3} observations in various forest environments should be accumulated; (2) Relationship between ambient and leaf temperatures and its impact on BVOCs emission should be explored further.

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Field Measurements of Biogenic Organic Aerosols in the Forest Atmosphere

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It is crucial to quantitatively understand the origin of biogenic organic aerosols (OAs) in the atmosphere, because organic matter in atmospheric aerosol particles are closely linked to climate impact of aerosol as well as biogeochemical cycle of bioelements, such as carbon and nitrogen. Since 2010, to investigate origins and formation processes of terrestrial biogenic OAs, we have conducted both long-term and intensive field measurements of atmospheric OAs at several research forest sites in Japan, which have representative vegetation of temperate and boreal forests: the Sapporo forest meteorology research site of the Forestry and Forest Products Research Institute, the Tomakomai Experimental Forest of Hokkaido University, and Fujiyoshida Forest Meteorological Research Site. At each site, aerosol samples were continuously collected on quartz fiber filters. The filter samples were used for off-line analyses of molecular tracers of biogenic secondary OAs as well as primary biological aerosol particles (PBAPs), bulk organic carbon and nitrogen, and stable carbon isotope ratios.

In this presentation, an overview on our field research will be given, which includes topics on the origin of biogenic OAs and their possible effects on the formation of cloud particles. From the field measurements of ambient OAs, we have obtained the following major findings:

- a) Elucidating origins and their quantitative contributions to biogenic OA mass at the forest sites, which are linked to the phenology at each site.
- b) Identification of five secondary fatty alcohols (SFAs) in atmospheric aerosols as new tracers for PBAPs originated mostly from plant wax.
- c) Multiple sources and formation processes of organic nitrogen aerosols dominated in summer and autumn.
- d) Cloud-forming potential of aerosol particles suppressed by biogenic organic matter emitted from the forest floor, such as soil and litters.
- e) Possible impact of biogenic OA on aerosol optical properties above the forest canopy.

The following key questions have been raised by our field studies regarding potential links between IGAC and iLEAPS toward quantitative understanding of atmosphere-terrestrial biosphere interactions via atmospheric aerosols:

- 1) How does biogenic aerosol production respond to changes in ambient temperature, cloudiness/rain/snow, and carbon fluxes?
- 2) How will cloud-forming potential of aerosols respond to changes in the formation of biogenic OA particularly from the forest floor?
- 3) How will aerosol formation respond to long-term system disturbance, such as nitrogen deposition, carbon uptake, and forest thinning?

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Contribution of Biogenic Secondary Organic Aerosols to Cloud Condensation Nuclei Concentrations at a Forest Site in the Kii Peninsula, Japan

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Objectives: Aerosol particles in the atmosphere are indispensable in cloud formation as they provide necessary surface areas for water vapors to condense. Moreover, cloudiness is found to complement the global climate change due to greenhouse gas emissions yet constituents the largest uncertainty in the prediction of global climate change. While the presence of biogenic secondary organic aerosol (BSOA), the oxidation reaction products of biogenic volatile organic vapor (BVOC), is estimated to enhance 3.6–45.2% of the global annual mean cloud condensation nuclei (CCN) number concentrations at a water vapor supersaturation (SS) condition of 0.2% (Scott et al., 2014), the observation based estimation of the contribution is scarce. On the other hand, the emission of BVOC from vegetations and the formation of BSOA are affected by the climate change. Hence, it is important to study the contribution of BSOA to CCN concentrations from the view point of BSOA formation. Furthermore, BSOA is formed either by the condensation of oxidized BVOC onto preexisting particles or by participating new particle formation (NPF). This presentation reviews studies based on field observations in a forest in the Kii peninsula, Japan, in which the contribution of BSOA to the CCN number concentration was estimated and the role of NPF in this contribution was assessed.

Methodologies: The chemical compositions, CCN activities, and hygroscopic growth of submicrometer aerosol particles were observed using respectively a high-resolution time-of-flight aerosol mass spectrometer (AMS), a CCN counter coupled with a differential mobility analyzer (DMA) and a condensation particle counter (CPC), and a hygroscopic tandem DMA during two intensive field observations (from 28 July to 28 August, 2014 and from 31 August to 22 September, 2015). The contribution of BSOA to CCN number concentrations was derived either from the measured CCN concentrations combined with aerosol chemical compositions or from the contribution of BSOA to total aerosol water uptake.

Results and discussions: Organic aerosol (OA) matter dominated the aerosol mass in the afternoon hours when the formation of BSOA was intensive. The increases of CCN number concentrations per 1 μ g m⁻³ BSOA at 0.11%, 0.24%, 0.42%, and 0.80% SS were, on average, 23, 84, 136, and 299 cm⁻³, respectively. Considering the total super-30 nm particle number concentration of 200 cm⁻³ in Amazon (Gunthe et al., 2009), the contribution of BSOA to the CCN number concentration was significant. This contribution at high SS conditions (0.42% and 0.80%) may be enhanced by NPF. In the afternoon hours, the contribution of BSOA to CCN concentrations can reach 28%. And this contribution can increase substantially if aging of BSOA occurs.

Future perspectives: First, the influence of NPF on the contribution of BSOA to CCN concentrations in the current forest needs to be confirmed with a larger dataset. Next, the quantification and CCN activity of BSOA under the influence of wide spread anthropogenic pollution needs to be studied. Besides, similar studies should be performed in other forest environments in and out of East Asia, for example to understand the influence of the influence of maritime and terrestrial air masses to coastal forests on the formation of new particles and CCN.

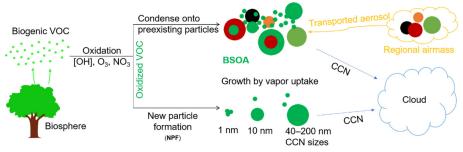


Figure 1: Sketch on the formation of BSOA from BVOC and the relationship between BSOA and cloud. References: Gunthe et al., *Atmos. Chem. Phys.*, 9, 7551–7575, 2009; Scott et al., *Atmos. Chem. Phys.*, 14, 447–470, 2014; Deng et al., *J. Geophys. Res.-Atmos.*, 123, 9703–9723, 2018; Deng et al., *Atmos. Chem. Phys.*, 19, 5889–5903, 2019.

Estimation of BVOC emissions with a terrestrial ecosystem model

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Terrestrial plants release a variety of biogenic volatile organic compounds (BVOCs) to the atmosphere. Scrutinizing the BVOCs emissions is important, because they affect a series of atmospheric chemical reactions and the atmospheric composition (e.g., tropospheric O₃, one of the GHGs) and because the emission also affects mass balance of terrestrial ecosystems. Formation of secondary organic aerosols induced by vegetation-generated BVOCs can, in turn, affect vegetation productivity by altering surface radiation budget and bio-physical conditions. Mainly for the former (atmospheric chemistry) perspective, vegetation BVOC emission has been parameterized as functions of light (isoprene), temperature, productivity, and leaf age: e.g., MEGAN scheme by Guenther et al. (2012). Plant-specific activity is mainly characterized by the emission factor, unit BVOC emission per unit leaf area / mass per unit time.

The BVOCs emissions scheme has been incorporated into terrestrial ecosystem models, which simulate atmosphere–land surface gas exchange and carbon cycle in ecosystems. A former version of MEGAN was introduced into the Vegetation Integrative SImulator for Trace gases (VISIT), in which leaf area index and its age composition and productivity are simulated during the period from 1901 to 2018. Using a standard set of forcing data, total BVOCs emissions from the global land vegetation in 2000–2018 was estimated as 776 Tg C yr⁻¹, of which 516 Tg C yr⁻¹ was by isoprene. Monoterpenes and sesquiterpenes accounted for 70.2 and 67.3 Tg C yr⁻¹, respectively. As expected by environmental conditions and biological activity, tropical rain forests were the largest source of BVOC. The long-term simulation showed that total BVOC emission has increased by +16% since the 1900s. The sensitivity simulations, in which driving factors were individually fixed or activated, indicated that the historical BVOC emission increase was mainly attributed to the fertilization effect due to elevated atmospheric CO₂ concentration.

A recent meta-analysis study (Feng et al., 2019) indicates that isoprene emission is more sensitive than monoterpenes to climate drivers, including elevated CO₂. Observations show that doubling of ambient CO₂ concentration inhibits isoprene emission by about 20%, while net photosynthesis rate increases and stomatal gas conductance decreases. Elevated CO₂ could affect the isoprene emission in several inconsistent ways: e.g., expansion of leaf area and biochemical inhibition. When atmospheric CO₂ concentration was fixed to the level in 1901 (297 ppmv), the simulated global isoprene emission showed little change through the experimental period. When including the biochemical inhibition effect in a simplified manner, the historical increase from the 1900s to the 2000s was reduced to +14% (mainly due to canopy leaf-area expansion). Although the inhibition effect seems tiny in the historical period, it can become more important under the future high CO₂ conditions.

Recent atmospheric chemical modeling and field observational studies imply that the present model overestimates isoprene emissions particularly from Asian tropical forests. The original model used the emission factor of 24 μ g g⁻¹ dw h⁻¹ for isoprene from broad-leaved forest, while the observation at Pasoh, Malaysia (Saito et al., 2008) obtained it as 1.2 mg m⁻² h⁻¹ (corresponding to 9 μ g g⁻¹ dw h⁻¹ based on specific leaf area of 170 cm² g dw⁻¹). When using the observation-based emission factor, a lower total isoprene emission, 356 Tg C yr⁻¹, was obtained. However, we don't have enough confidence to adopt the emission factor obtained at a single site in Southeast Asia as a representative value of global tropical forests.

There remain issues in the present BVOC schemes. First, few schemes consider mutual exchange or BVOC uptake by vegetation. Second, few models account for BVOC emissions from non-leaf, i.e. stem, root, and litter components. Constraining emission factors is the largest issue of the present model-based estimations, and further bioinformatic, experimental, remote-sensing, and modeling studies are required.

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3-D distributions of atmospheric greenhouse gases over Siberia

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For a better understanding of the role for the terrestrial biosphere in the global cycle of greenhouse gases, atmospheric mixing ratios have been observed by air sampling method using airplanes over 3 sites with different vegetation in Siberia since July 1993. We also conduct continuous measurement of CO_2 and CH_4 using tower network in taiga, steppe, and wetland regions over Siberia since 2004 (JR-STATION: Japan–Russia Siberian Tall Tower Inland Observation Network).

Air sampling has been carried out almost once per month using a chartered aircraft at 8 different altitudes between 0.5 and 7 km over the wetland near Surgut (61°N, 73°E) and over the forest near Novosibirsk (55°N, 83°E), West Siberia and between 0.1 and 3 km over the forest near Yakutsk (62°N, 130°E), East Siberia. Collected air samples are transported to National Institute for Environmental Studies to measure the mixing ratios of CO₂, CH₄, N₂O, SF₆, CO and H₂ and stable isotope ratio of CO₂.

JR-STATION project started by 9-tower network in Siberia. In 2019, we operate the measurement system at Berezorechka (56°N, 84°E), Karasevoe (58°N, 82°E), Noyabrsk (63°N, 76°E), Demyanskoe (60°N, 71°E), Azovo (55°N, 73°E) and Vaganovo (54°N, 62°E), all sites being located in West Siberia. Air inlet in each site is installed at 40m or higher on the tower. CO₂ and CH₄ mixing ratios are measured by NDIR (LI-820 LI-COR) and originally developed CH₄ semiconductor sensor, respectively. Cavity Ring-Down Spectroscopies (CRDS) have been used for measuring CO₂ and CH₄ at Karasevoe, Noyabrsk and Demyanskoe since 2015.

Figure 1 shows the averaged vertical profiles of CO_2 mixing ratio over Surgut. Vertical gradient of CO_2 in each month indicates that land surface acts as source or sink for atmosphere in corresponding period.

The amplitudes in seasonal variation of CO_2 mixing ratio observed both by aircraft (especially in lower altitude) and towers show extremely larger compared to those observed in coastal sites, which indicates strong influence by activities of Siberian vegetation.

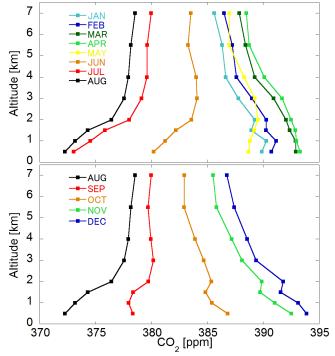


Figure 1. Vertical profiles of CO2 mixing ratio over Surgut

Uncertainty in recent enhancement of terrestrial CO₂ uptake

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In contrast to increasing anthropogenic CO₂ emissions, CO₂ uptake to the land and ocean has been steadily increasing during recent decades. Between the decades of 1990s and 2000s, global CO₂ uptake was enhanced to the level where the amount of uptake doubled with respect to the past 50 years, slowed the atmospheric CO₂ growth despite of rapidly increased CO₂ emissions during the 2000s. In the land, multiple evidences suggest that the Northern Hemisphere is the dominant contribution to the uptake enhancement, confirmed by measurements of the interhemispheric gradient of atmospheric CO₂. In the ocean, studies based on observations of the surface partial pressure of CO₂ (pCO₂) and ocean CO₂ concentrations demonstrated that the Southern ocean is the large contribution to the uptake enhancement between the 1990s and 2000s.

Abundant studies and evidences support validity of the CO₂ uptake enhancement in both the land and ocean. Meanwhile, in the circumstance where increasing anthropogenic CO₂ emissions aggravate climate change, it is a major concern for how long the CO₂ uptake enhancement continues to mitigate CO₂ emissions even after the 2000s. Furthermore, question still remains on whether processes implemented in current biogeochemical models are sophisticated enough to simulate future projections of the CO₂ uptake. To answer this question, here we gathered the latest Global Dynamic Vegetation Models (DGVMs) and Global Ocean Biogeochemical Models (GOBMs) from the Global Carbon Budget 2018), and re-evaluate the pattern of recent trends and CO₂ budget changes from 1980 to 2017. We discuss agreement and also discrepancy between process models (DGVMs and GOBMs) and CO₂ uptake simulated by atmospheric inversions and that inferred from atmospheric growth rate and fossil fuel emissions on continued increase in CO₂ uptake for the study period.

Carbon stock in Japanese forests has been outrageously underestimated

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An accurate estimation of total forest carbon (C) stock and C uptake in Japan is crucial in terms of securing viable resource for the economies and the mitigation of global warming. Forest inventory is among forest biomass estimations, based on the field measurements of individual tree size, and is thought to give the most accurate estimation of the C stock. National Forest Inventory (NFI) provides data of the forest stand scale stem volume throughout Japan, and there exist two types of NFI, i.e., the NFI based on prediction using the yield tables (p-NFI) and direct field measurements (m-NFI). This study found that Japanese national and local forestry agencies and even previous academically-valued studies have exceedingly underestimated total Japanese forest biomass, and C stock and uptake, because they failed to update appropriately the yield tables in p-NFI or mistook p-NFI for m-NFI. Such previous reports might have misled policymakers for Japanese forest management policy. Further, we suggested the most accurate estimations of the C stock and uptake so far. For the accurate estimation of the forest C stock, we require updating the yield tables used in p-NFI or ideally, continuing to carry out the field measurement campaign for m-NFI.

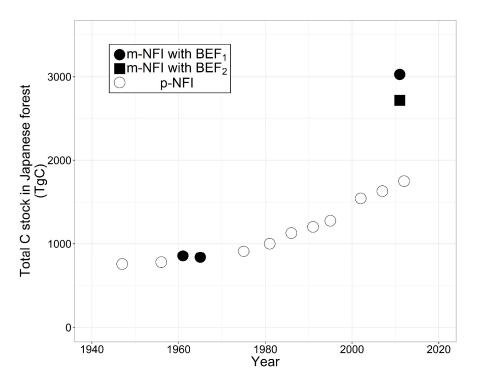


Figure. Temporal changes in total forest carbon stock in Japan. p-NFI and m-NFI denote National Forest Inventory (NFI) based on the predictions from yield tables and the direct field measurements, respectively. BEF1 and BEF2 represent two different types of Biomass Expansion Factor (BEF)

Estimation of Regional-Global Methane Emissions and Refinement of Its Estimate by GOSAT-2 and Surface Observations

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Methane (CH₄) is one of the most important greenhouse gases in the atmosphere. Methane has both natural and anthropogenic sources, such as wetlands, termites, coal mining, livestock, landfills, and rice paddies. Significant amounts of methane are emitted from the surface in South Asia and uncertainties in the estimate of the emission there remain large [Patra et al., 2016]. In addition, the CH₄-rich air is transported from the surface to the upper troposphere due to convective transports associated with Asian summer monsoons [Xiong et al., 2009; Chandra et al., 2017]. The purpose of our study is to elucidate the emission sources and processes of methane in South Asia for the reduction of uncertainties in the regional methane estimates there and to estimate global methane emissions based on "top-down" approach based on a chemical transport model, surface observations, and satellite observations.

We have conducted continuous measurements of methane fluxes and concentrations through several different rice cropping seasons by laser absorption spectroscopy in rice paddies in Tamil Nadu in South India. We have also conducted air sampling by flask in Nainital in India and Comilla in Bangladesh, and analyzed concentrations of methane and carbon monoxide (CO) and carbon stable isotope ratio of methane (δ^{13} C-CH₄) in the sampled air to reveal detailed methane emission sources there. We have validated vertical profiles of methane concentrations retrieved from the thermal infrared (TIR) band of the Thermal and Near Infrared Sensor for Carbon Observation (TANSO)-Fourier Transform Spectrometer (FTS) on board Greenhouse Gases Observing Satellite (GOSAT) by aircraft observations globally and then assessed the impact of vertical transports on the seasonal cycle of columnar methane in India by the validated GOSAT/TANSO-FTS TIR methane data. To conduct methane inversion analysis globally, we have newly developed MIROC4.0-based Atmospheric Chemistry-Transport Model (MIROC4-ACTM) [Patra et al., 2018] and evaluated its vertical transport processes and several methane emission schemes by comparing the model outputs with aircraft, Atmospheric Chemistry Experiment (ACE)-FTS, and GOSAT/TANSO-FTS TIR data.

For future work, we plan to utilize huge amounts of data for columnar methane obtained by the Tropospheric Monitoring Instrument (TROPOMI) on board the Copernicus Sentinel-5 Precursor satellite in addition to GOSAT/TANSO-FTS and GOSAT-2/TANSO-FTS-2 vertical and columnar methane data. We will also use columnar carbon monoxide data newly obtained by GOSAT-2/TANSO-FTS-2 to estimate the emission sources of the observed methane enhancement by GOSAT-2/TANSO-FTS-2. One of our challenging tasks is to establish an appropriate way to compare data with different time and horizontal scales from surface observations, satellite observations, and model simulations for evaluating the methane inversion analysis.

Acknowledgements: This research has been supported by the Environmental Research and Technology Development Fund (2-1802) of the Environmental Restoration and Conservation Agency of Japan.

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Potential links between iLEAPS and IGAC toward integrated analysis of atmosphere-land interactions using remote sensing data

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Linking atmospheric chemistry community (e.g., IGAC) and land surface community (e.g., iLEAPS) is an essential step toward a better and integrated understanding of global or regional environmental changes. In particular, extreme anomalies at large scales, such as heatwave and biomass burning, have been occurring across global in recent years. Responding to ongoing climate and anthropogenic changes, these anomalous environment changes will occur more frequently in future. Therefore, understandings of carbon cycle and its interaction with climate, including land-atmosphere exchanges of multiple gases (e.g., CO₂, CH₄, CO, BVOC) and their chemical reaction in the atmosphere, are highly required. However, comprehensive analysis across multiple chemical species are lacking.

Recently, more and more satellite-based observations of multiple chemical species in the atmosphere and land surface status are becoming available to the community. Therefore, we believe now is the right timing to start with an integrated analysis of these data sets across atmospheric chemistry group and land surface group, in particular among remote sensing communities. As an initial step for collaboration, we will introduce our research plan for integrated analysis of global and continental-scale environmental changes. Our research target is to understand biomass burning and its impact on regional to continental-scale atmospheric and climate changes using multiple remote sensing products and network of in-situ measurements.