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研究課題名（和文）The effect of polycyclic aromatic hydrocarbons, nitro-PAHs and meteorological conditions on atherosclerotic cardiovascular disease under influence of climate change and Asian Sand Dust

研究課題名（英文）The effect of polycyclic aromatic hydrocarbons, nitro-PAHs and meteorological conditions on atherosclerotic cardiovascular disease under influence of climate change and Asian Sand Dust

研究代表者

Pham Oanh (Pham, Oanh)

一般財団法人日本環境衛生センターアジア大気汚染研究センター・情報管理部・研究員

研究者番号：10895816

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研究成果の概要（和文）：西日本における黄砂と多環芳香族炭化水素（PAHs）およびニトロPAHsの長距離輸送と健康影響を調査した。黄砂は西日本の環境中粒子状物質濃度を上昇させたが、総PAHおよびニトロPAH濃度は上昇させなかった。アジア大陸からの長距離輸送が2年間にわたって検出され、特に冬季に強かった。西日本における黄砂とPAHsとニトロPAHsの長距離輸送は、輸送ルートと起源が異なることがわかった。PAHsは鼻症状、成人日本人の喘息、慢性咳嗽患者のリスク増加と正の関係を示した。都市部では、PAHsに起因する生涯追加がん症例数も農村部より多かった。アジア大陸に近い農村では、PM2.5に起因する症例が多かった。

研究成果の学術的意義や社会的意義

西日本における黄砂と多環芳香族炭化水素（PAHs）およびニトロPAHsの長距離輸送と健康影響を調査した。黄砂は西日本の環境中粒子状物質濃度を上昇させたが、総PAHおよびニトロPAH濃度は上昇させなかった。アジア大陸からの長距離輸送が2年間にわたって検出され、特に冬季に強かった。西日本における黄砂とPAHsとニトロPAHsの長距離輸送は、輸送ルートと起源が異なることがわかった。PAHsは鼻症状、成人日本人の喘息、慢性咳嗽患者のリスク増加と正の関係を示した。都市部では、PAHsに起因する生涯追加がん症例数も農村部より多かった。アジア大陸に近い農村では、PM2.5に起因する症例が多かった。

研究成果の概要（英文）：In this project, the principal investor investigated the health effect of Asian Dust and long-range transport of polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs in Western Japan. Asian Dust elevated ambient particulate matter concentration in Western Japan, while it did not increase total PAH and nitro-PAH concentrations. Long-range transport from the Asian Continent was detected over the two years and especially strong during wintertime. Asian Dust and long-range transport of PAHs and nitro-PAHs in Western Japan were found to have different transport routes and origin. The results showed that PAHs had positive relationship with increased risk of nasal symptoms, asthma in Adult Japanese, and Japanese patients with chronic cough. Urban areas had higher cases attributable to PM2.5 and additional lifetime cancer cases due to PAHs than rural areas. Rural area near Asian Continent had high attributable cases per 100,000 population due to PM2.5.

研究分野：Atmospheric Chemistry

キーワード：Long-range transport PM2.5 Asian Sand Dust PAHs and nitro-PAHs Health effect Asthma Chronic Cough

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様式 C-19、F-19-1、Z-19 (共通)

1. 研究開始当初の背景

Originally, the project theme is “The effect of polycyclic aromatic hydrocarbons, nitro-PAHs and meteorological conditions on atherosclerotic cardiovascular disease under influence of climate change and Asian Sand Dust”. However, due to changing workplace (from Kanazawa Univ. to Asia Center for Air Pollution Research) during the second year of the project, PI was unable to access the health data and urine samples under policy of Kanazawa Univ. Therefore, the project had to change direction to “The effect of polycyclic aromatic hydrocarbons, nitro-PAHs and meteorological conditions on health under influence of Asian Sand Dust”.

2. 研究の目的

Due to theme changing, the aim of the study is changed **to investigate the effect of meteorological conditions and PAHs, N-PAHs on human health under influence of ASD**. The changed objectives are following:

1. To investigate the effect of Asian Sand Dust on PM, PAHs, N-PAHs at urban and rural areas
2. To elucidate the mechanism of the effect of ASD, PAHs and N-PAHs on human health

3. 研究の方法

To achieve the goal of the study, total suspended particles (TSP) are collected daily using high volume air sampler Kanazawa, Noto, Fukushima, and Fukue. The filters are weighted before and after the collection filter and stored at -30° C until analysis. PAHs and nitro-PAHs are obtained using liquid-liquid extraction (benzene:ethanol) and base/acid clean up. PAHs are analyzed by high performance liquid chromatography (HPLC) coupled with fluorescence detector (FD). Nitro-PAHs are analyzed by HPLC-Reduction/chemiluminescence detection (CL) (Hayakawa et al, *Anal Sci*, 1991). The analysis of nitro-PAHs involves complicated transformation to selectively quantify nitro-PAHs. We have a **collaboration with one of the nitro-PAH analysis's pioneers** (Dr. Hayakawa, Kanazawa University) to analyze nitro-PAHs from our samples.

4. 研究成果

Pham K-O, Hara A, Zhao J, Suzuki K, Matsuki A, Inomata Y, Matsuzaki H, Odajima H, Hayakawa K, Nakamura H. **Different Transport Behaviors between Asian Dust and Polycyclic Aromatic Hydrocarbons in Urban Areas: Monitoring in Fukuoka and Kanazawa, Japan.** *Applied Sciences*. 2022; 12(11):5404.

To clarify different effects of Asian Sand Dust (ASD), long-range transported from Asian continent, on total suspended particles (TSP) and polycyclic aromatic hydrocarbons (PAHs) in Japan, TSP were simultaneously collected during ASD periods (from 1 March to 31 May 2020 and 2021) in Fukuoka and Kanazawa. During ASD days, decided by Light Detection and Ranging and Japan Meteorological Agency, TSP concentrations increased significantly ($p < 0.001$) at two sampling sites. PAH concentrations increased in Kanazawa ($p < 0.001$) but not in Fukuoka on ASD days. Correlation coefficients (r) between daily TSP and total PAHs concentrations were weak in Kanazawa: 0.521 (non-ASD) and 0.526 (ASD) ($p < 0.01$), and in Fukuoka: 0.321 (non-ASD) and 0.059 (ASD). However, correlation between seasonal (average monthly) TSP and total PAH concentrations were stronger: 0.680 (Kanazawa) and 0.751 (Fukuoka). The reasons might be that seasonal variations of TSP and total PAHs in two cities depend equally on planetary scale westerly, while daily TSP and total PAHs variations in each city varied by different transportation distances from ASD and PAHs sources in the Asian continent to Japan. Different local sources and meteorological conditions were considered. These results are important for elucidating the causes of chronic and acute respiratory diseases.

Pham K-O, Hara A, Zhao J, Suzuki K, Matsuzaki H, Odajima H, Hayakawa H, Nakamura H, **Effect of long-range transport on air pollution in rural and urban areas on the western side of Japan**, 第 63 回大気環境学会年会講演要旨集, 160(2022)

Figure 1 shows daily concentrations of PM, PAHs and 1-NP in 4 sites. The orders of concentrations were

PM: Kanazawa ($44.1 \mu\text{g}/\text{m}^3$) > Fukue ($28.9 \mu\text{g}/\text{m}^3$) > Fukuoka ($28.4 \mu\text{g}/\text{m}^3$) > Noto ($12.6 \mu\text{g}/\text{m}^3$);

PAHs: Fukuoka ($0.96 \text{ ng}/\text{m}^3$) > Fukue ($0.67 \text{ ng}/\text{m}^3$) > Kanazawa ($0.39 \text{ ng}/\text{m}^3$) > Noto ($0.33 \text{ ng}/\text{m}^3$);

1-NP: Fukue ($1.73 \text{ pg}/\text{m}^3$) > Fukuoka ($1.48 \text{ pg}/\text{m}^3$) > Kanazawa ($1.33 \text{ pg}/\text{m}^3$) > Noto ($0.34 \text{ pg}/\text{m}^3$).

The south-west Japan had higher concentrations compared to north-west Japan. Noto (rural) had the lowest air pollution, while air pollution in Fukue (rural) was comparable to Kanazawa and Fukuoka (urban). PM was not correlated with PAHs in all places, implying different sources. PM was correlated with 1-NP (Fukue: $r=0.7, p<0.01$; Fukuoka: $r=0.5, p<0.01$) in south-west Japan but not in north-west Japan ($r<0.5$). Unexpectedly, PAHs was not associated with 1-NP in Fukue ($r=0.2$), while their correlations were strong in Fukuoka ($r=0.7, p<0.01$), Kanazawa ($r=0.8, p<0.01$), Noto ($r=0.8, p<0.01$).

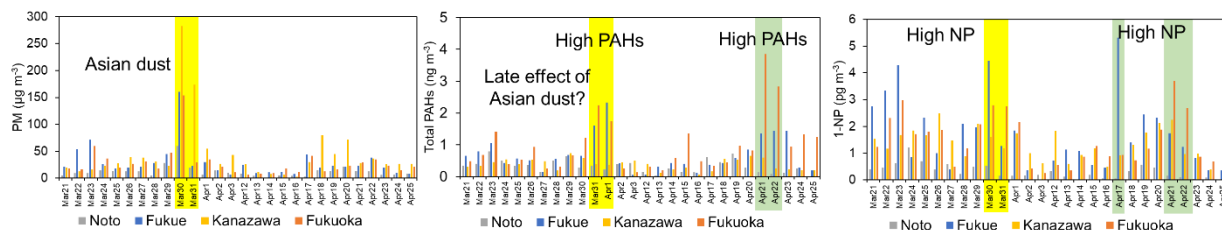


Fig. 1 Concentrations of PM, PAHs and 1-NP in rural and urban areas during AD period.

Kanazawa and Noto had similar backward-trajectories and their correlations between air pollutants were also high ($r>0.6, p<0.01$), which implied they are closely related in sources. The relationships between Fukuoka and Fukue were strong in PM ($r=0.9, p<0.01$) and PAHs ($r=0.7, p<0.01$) but weak in 1-NP ($r=0.4, p<0.05$). Their backward trajectories also showed dissimilarity.

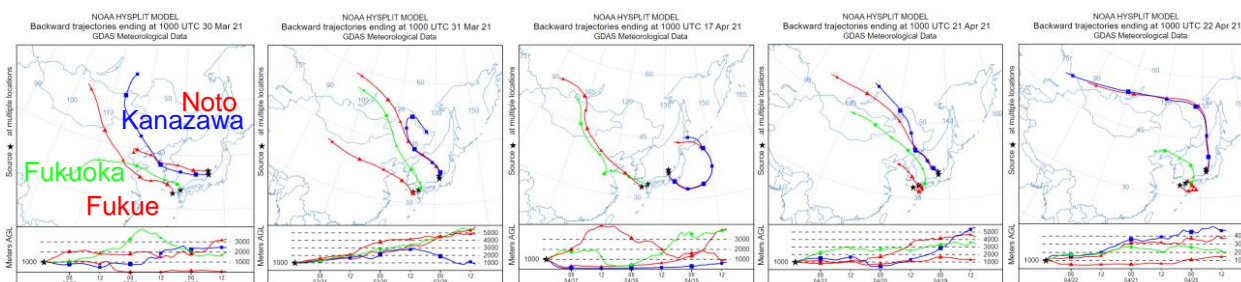


Fig. 2 Backward-trajectories during AD episodes, and days with high PAHs or high 1-NP concentrations.

AD had a significant effect on the concentrations of PM, but not PAHs due to different transportation routes. Air pollution in rural areas was affected by nearby urban areas and LRT; however, the effect of LRT may depend on locations. LRT from east China had strong influence on air pollution in Fukue, but not in Noto.

Pham K-O, Hayakawa H, Hara A, Zhao J, Suzuki K, Matsuzaki H, Odajima H, Nakamura H, **Combustion source apportionment and risk assessment in western side of Japan**, 第 64 回大気環境学会年会講演要旨集, 190(2023)

In the present study, the NP method, using PAHs and nitro-PAHs as markers, was applied to identify their combustion sources in rural (Noto and Fukue) and urban (Kanazawa and Fukuoka) areas in Western Japan for four seasons in two years (2020-2021). PM mass, PAHs and nitro-PAHs were measured to compare the effect of LRT at the four sites. Benzo[*a*]pyrene (BaP) is the toxicity indicator for PAHs because it's well-studied among carcinogenic PAH compounds (ASTDR, 2022). The toxicity of PAHs was calculated via BaP equivalence (BaP_{eq}), which is the sum of carcinogen PAH compound concentrations multiplying with their potency equivalency factors (PEFs). The health risk assessment due to long-term exposure to PM_{2.5} and PAHs were estimated using AirQ+, a software tool for quantifying the health impact of air pollution developed by WHO Regional Office for Europe.

The average concentrations of APM were the highest in Kanazawa ($23.8\pm 34.1 \mu\text{g m}^{-3}$). Fukuoka had the highest average total PAH concentrations ($0.76\pm 0.84 \text{ ng m}^{-3}$). Noto had the lowest concentrations for all the compounds, while Fukue had equal or higher concentrations than urban area (Fukuoka). Moreover, during winter, the concentrations of air pollutants increased in Noto and Fukue. Back-ward trajectory from the Asian Continent was detected over the two years except summer season. LRT was especially strong during wintertime in Fukue and Noto.

As shown in Table 1, the BaP_{eq} concentrations in Western Japan were much lower than the established annual limit of 1.0 ng m^{-3} for BaP (European Environment Agency, 2018) and comparable to concentrations of BaP in European remote

areas (0.02 ng/m³) (European Commission, 2001). Urban areas (Kanazawa and Fukuoka) had higher cases attributable to PM_{2.5} and additional lifetime cancer cases (ALCC) due to PAHs than rural areas. However, Fukuoka had the highest attributable cases per 100,000 population at risk of PM_{2.5} among the four sites posing a health threat of LRT in rural areas.

These results indicate the air pollution and health risk at each site is strongly dependent on differences in routes and distances of LRT of pollutants from Asian Continent.

Table 1. Risk assessment for long-term exposure to PM_{2.5} and PAHs in sampling sites

Health risk	Noto	Kanazawa	Fukue	Fukuoka
Attributable Proportion due to PM _{2.5} (%)	2.4 (1.82-2.68)	13.49 (10.39-14.98)	9.44 (7.24-10.51)	9.31 (7.13-70.37)
No. of death per 100,000 population at risk due to PM _{2.5}	48.08 (36.51-53.76)	130.81 (100.76-145.25)	183.99 (140.97-204.82)	72.70 (55.69-80.93)
No. of death attributable to PM _{2.5}	12 (9-13)	606 (467-673)	63 (48-70)	1172 (898-1305)
ALCC due to PAHs	0.06	1.61	0.21	11.22

In collaboration with other researchers, the effect of meteorological conditions and PAHs, N-PAHs on human health under influence of ASD is also investigated.

Yoshino et al. (2022) indicated that during 2020 and 2021, most PAHs (93%) in Kanazawa were transported from the Asian continent by westerlies and that the main source was coal and biomass combustion. The presence of NPAHs in Kanazawa was caused by a mixture of transport from the Asian continent (53%) and local emissions (47%), with the main source of the latter being from vehicles. Although the content of combustion-derived particulates (Pc) was <2.4% of PM_{2.5} in Kanazawa, this showed a similar seasonal variation (winter > summer) to that of PAHs. The contribution of Pc transported from the Asian continent exceeded that of locally emitted Pc. The current situation of Kanazawa is considerably different from that of 1997, when local vehicles were the main source of pollution.

Hayakawa et al. (2024) focused on the components of fine particulate matter, i.e., PM_{2.5}, during spring 2020 in Fukuoka, Japan. The backward trajectory showed that air masses reached Fukuoka via the Yellow Sea and the southern part of South Korea when PM_{2.5} and each chemical component were high in concentrations. On the other hand, diurnal variations in ozone were also observed, suggesting that both transboundary and local air pollution are involved. Air masses reached the southern part of the Kyushu region when only sulfate concentrations were high. A volcano eruption led the high sulfate concentration. When only polycyclic aromatic hydrocarbons (PAHs) concentrations were high, air masses often reached the northern part of Kyushu, indicating that there may be a specific local source for PAHs.

Zhao et al. (2022) using the shared data from this project, and examined the relationship in 51 adults living in Ishikawa prefecture, Japan, and conducted a 2-month follow-up survey on these participants. All participants were asked to record daily nasal symptoms in an allergy diary during the study period between 1 April to 31 May 2020. After adjustment for confounding factors, the B values of fluoranthene, pyrene, and Benzo[k]fluoranthene were 2.389 (p = 0.026), 3.744 (p = 0.022) and 9.604 (p = 0.041), respectively, with a one-day lag. In contrast, the B value of indeno[1,2,3-cd] pyrene was 6.664 (p = 0.013) with no lag. Collectively, these results suggest ambient PAHs such as Flt, Pyr, and BkF were associated with nasal symptoms in adults. Further studies are needed to elucidate the mechanisms contributing to the relationships between specific PAHs and nasal symptoms.

Hara et al. (2022) investigated airborne PAH exposure's effects on cough symptoms in children and adults with

chronic coughs in Kanazawa and Fukuoka, Japan, were investigated in this longitudinal study. A total of 98 patients with chronic coughs were enrolled and followed up between 1 April and 31 May 2020. Among nine PAHs measured, benz[a]anthracene (BaA) was significantly associated with cough symptoms for both lag4 and lag5 PAH exposure. These findings suggest that airborne specific PAHs, especially BaA, affect cough symptoms in children and adults with chronic cough. Further studies are needed to develop effective measures to prevent respiratory diseases against specific PAHs.

Kress et al. (2022) compared the role of polygenic susceptibility on air pollution-associated asthma between German and Japanese women. We found an increased risk of asthma in Japanese women after exposure to low pollutant levels (PM_{2.5}: median = 12.7µg/m³, p-value < 0.001, NO₂: median = 8.5µg/m³, p-value < 0.001) and in German women protective polygenic effects (p-value = 0.008). While we found no significant G × E effects, the direction in both groups was that the PRS increased the effect of PM_{2.5} and decreased the effect of NO₂ on asthma. Our study confirms that exposure to low air pollution levels increases the risk of asthma in Japanese women and indicates polygenic effects in German women; however, there was no evidence of G × E effects. Future genome-wide G × E studies should further explore the role of ethnic-specific polygenic susceptibility to asthma.

Reference:

Yoshino A, Takami A, Shimizu A, Sato K, Hayakawa K, Tang N, Pham K-O, Hara A, Nakamura H, Odajima H. **Analysis of Chemical Components of Fine Particulate Matter Observed at Fukuoka, Japan, in Spring 2020 and Their Transport Paths.** *Appl. Sci.* 2022; 12(22) 11400-11400.

Hayakawa K, Pham K-O, Takami A, Odajima H, Nagato E. G., Hara A, Nakamura H. **Contributions of long-range transport from the Asian continent and local emissions on atmospheric PM_{2.5}, polycyclic aromatic hydrocarbons, and their nitro-derivatives in Kanazawa and Noto Peninsula, Japan.** *Heliyon* 2024; 10(5) e27320-e27320.

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Hara A, Odajima H, Matsuzaki H, Fujimura M, Toma T, Wada T, Ohkura N, Zhao J, Pham K-O, Suzuki K, Tsujiguchi H, Takami A, Hayakawa K, Nakamura H. **Association between Cough and Ambient Polycyclic Aromatic Hydrocarbons in Patients with Chronic Cough: An Observational Study in Two Regions of Japan.** *Appl. Sci.* 2022; 12(24) 12505-12505.

Kress S, Hara A, Wigmann C, Sato T, Suzuki K, Pham K-O, Zhao Q, Areal A, Tajima A, Schwender H, Nakamura H, Schikowski T. **The Role of Polygenic Susceptibility on Air Pollution-Associated Asthma between German and Japanese Elderly Women.** *Int J Environ Res Public Health.* 2022; 19(16) 9869-9869.

5. 主な発表論文等

〔雑誌論文〕 計6件（うち査読付論文 6件/うち国際共著 6件/うちオープンアクセス 6件）

1. 著者名 Pham Kim-Oanh, Hara Akinori, Zhao Jiaye, Suzuki Keita, Matsuki Atsushi, Inomata Yayoi, Matsuzaki Hiroshi, Odajima Hiroshi, Hayakawa Kazuichi, Nakamura Hiroyuki	4. 巻 12
2. 論文標題 Different Transport Behaviors between Asian Dust and Polycyclic Aromatic Hydrocarbons in Urban Areas: Monitoring in Fukuoka and Kanazawa, Japan	5. 発行年 2022年
3. 雑誌名 Applied Sciences	6. 最初と最後の頁 5404 ~ 5404
掲載論文のDOI (デジタルオブジェクト識別子) 10.3390/app12115404	査読の有無 有
オープンアクセス オープンアクセスとしている (また、その予定である)	国際共著 該当する
1. 著者名 Zhao Jiaye, Hara Akinori, Hayakawa Kazuichi, Pham Kim-Oanh, Suzuki Keita, Tsujiguchi Hiromasa, Matsuzaki Hiroshi, Odajima Hiroshi, Takami Akinori, Nakamura Hiroyuki	4. 巻 12
2. 論文標題 Longitudinal Study on the Association between Ambient Polycyclic Aromatic Hydrocarbons and Nasal Symptoms in Adult Japanese	5. 発行年 2022年
3. 雑誌名 Applied Sciences	6. 最初と最後の頁 12544 ~ 12544
掲載論文のDOI (デジタルオブジェクト識別子) 10.3390/app122412544	査読の有無 有
オープンアクセス オープンアクセスとしている (また、その予定である)	国際共著 該当する
1. 著者名 Hara Akinori, Odajima Hiroshi, Matsuzaki Hiroshi, Fujimura Masaki, Toma Tomoko, Wada Taizo, Ohkura Noriyuki, Zhao Jiaye, Pham Kim-Oanh, Suzuki Keita, Tsujiguchi Hiromasa, Takami Akinori, Hayakawa Kazuichi, Nakamura Hiroyuki	4. 巻 12
2. 論文標題 Association between Cough and Ambient Polycyclic Aromatic Hydrocarbons in Patients with Chronic Cough: An Observational Study in Two Regions of Japan	5. 発行年 2022年
3. 雑誌名 Applied Sciences	6. 最初と最後の頁 12505 ~ 12505
掲載論文のDOI (デジタルオブジェクト識別子) 10.3390/app122412505	査読の有無 有
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1. 著者名 Yoshino Ayako, Takami Akinori, Shimizu Atsushi, Sato Kei, Hayakawa Kazuichi, Tang Ning, Pham Kim-Oanh, Hara Akinori, Nakamura Hiroyuki, Odajima Hiroshi	4. 巻 12
2. 論文標題 Analysis of Chemical Components of Fine Particulate Matter Observed at Fukuoka, Japan, in Spring 2020 and Their Transport Paths	5. 発行年 2022年
3. 雑誌名 Applied Sciences	6. 最初と最後の頁 11400 ~ 11400
掲載論文のDOI (デジタルオブジェクト識別子) 10.3390/app122211400	査読の有無 有
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1. 著者名 Kress Sara, Hara Akinori, Wigmann Claudia, Sato Takehiro, Suzuki Keita, Pham Kim-Oanh, Zhao Qi, Areal Ashtyn, Tajima Atsushi, Schwender Holger, Nakamura Hiroyuki, Schikowski Tamara	4. 巻 19
2. 論文標題 The Role of Polygenic Susceptibility on Air Pollution-Associated Asthma between German and Japanese Elderly Women	5. 発行年 2022年
3. 雑誌名 International Journal of Environmental Research and Public Health	6. 最初と最後の頁 9869 ~ 9869
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オープンアクセス オープンアクセスとしている (また、その予定である)	国際共著 該当する

1. 著者名 Hayakawa Kazuichi, Kim-Oanh Pham, Takami Akinori, Odajima Hiroshi, Nagato Edward G., Hara Akinori, Nakamura Hiroyuki	4. 巻 10
2. 論文標題 Contributions of long-range transport from the Asian continent and local emissions on atmospheric PM2.5, polycyclic aromatic hydrocarbons, and their nitro-derivatives in Kanazawa and Noto Peninsula, Japan	5. 発行年 2024年
3. 雑誌名 Heliyon	6. 最初と最後の頁 e27320 ~ e27320
掲載論文のDOI (デジタルオブジェクト識別子) 10.1016/j.heliyon.2024.e27320	査読の有無 有
オープンアクセス オープンアクセスとしている (また、その予定である)	国際共著 該当する

〔学会発表〕 計5件 (うち招待講演 0件 / うち国際学会 2件)

1. 発表者名 Oanh Pham
2. 発表標題 Lifetime lung cancer risk due to polycyclic aromatic hydrocarbons during Asian Sand Dust season in Fukuoka and Kanazawa
3. 学会等名 18th Annual Meeting of the Association for Preventive Medicine of Japan
4. 発表年 2021年

1. 発表者名 Kim-Oanh Pham
2. 発表標題 Asian Sand Dust and its effect on polycyclic aromatic hydrocarbons in Fukuoka and Kanazawa
3. 学会等名 4th Japan-Germany Symposium on Advanced Preventive Medicine (国際学会)
4. 発表年 2021年 ~ 2022年

1. 発表者名 Pham KO
2. 発表標題 Long-range transport in rural and urban areas on the western side of Japan
3. 学会等名 10th International Conference on Acid Deposition (国際学会)
4. 発表年 2022年

1. 発表者名 Kim-Oanh Pham
2. 発表標題 Effect of long-range transport on air pollution in rural and urban areas on the western side of Japan
3. 学会等名 The 63rd Annual Meeting of Japan Society for Atmospheric Environment
4. 発表年 2022年

1. 発表者名 Kim-Oanh Pham
2. 発表標題 Combustion source apportionment and risk assessment in western side of Japan
3. 学会等名 The 64rd Annual Meeting of Japan Society for Atmospheric Environment
4. 発表年 2023年

〔図書〕 計0件

〔産業財産権〕

〔その他〕

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6. 研究組織

	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考
研究協力者	中村 裕之 (Nakamura Hiroyuki) (30231476)	金沢大学・医学系・教授 (13301)	

6. 研究組織（つづき）

	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考
研究協力者	早川 和一 (Hayakawa Kazuichi) (40115267)	金沢大学・環日本海域環境研究センター・連携研究員 (13301)	
研究協力者	原 章規 (Hara Akinori)	金沢大学・医学系・准教授 (13301)	

7. 科研費を使用して開催した国際研究集会

〔国際研究集会〕 計0件

8. 本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関