様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科研費 KAKENHI

科学研究費助成事業　　研究成果報告書

令和　４ 年　 ８ 月 ２９ 日現在

機関番号：１０１０１
研究種目：研究活動スタート支援
研究期間：2020～2021
課題番号：２０Ｋ２３３５９

研究課題名（和文）Analysis of hazardous components(PAHs) and their unknown derivatives in atmospheric PM 2.5 organic aerosols collected over Hokkaido

研究課題名（英文）Analysis of hazardous components(PAHs) and their unknown derivatives in atmospheric PM 2.5 organic aerosols collected over Hokkaido

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交付決定額（研究期間全体）：（直接経費）　　1,600,000円

研究成果の概要（和文）：エアロゾルは、気体中に浮遊する微小な粒子から構成されています。近年、粒子状物質（PM）は世界的な健康上の関心事となっています。大気中のPMは気候と健康の両方に影響を与え、PMは喘息や呼吸器疾患など、人間の健康に大きな悪影響を与える可能性があります。したがって、PAHの多様な影響を検討し、制御することができるように、その存在量、化学種、分布、潜在的な発生源を理解することが重要である。

研究成果の学術的意義や社会的意義
Seasonal variation of PAHs and related compounds have been examined in the Sapporo aerosols. These results will help to understand the air quality of the Sapporo, Sapporo is the cold area of Japan where heating activity is more in autumn-winter and spring.

研究成果の概要（英文）：PAHs are a group of organic compounds of multiple aromatic rings. PAHs and related compounds have toxic, mutagenic, and carcinogenic properties. Hence, it is important to understand the abundance, speciation, distribution, and potential sources, so that the diverse effects of PAHs can be examined and can be controlled. Currently, LC/MS methods have been established with 0.01 ng LOQ and 0.001 ng LOD. Aerosol samples have been collected on the rooftop of health science, Hokkaido University from 2021 and 2022. Benzo perylene is the most abundant species followed by 6-nitro benzo pyrene and Benzo anthracene As the cold season approaches the concentration of PAH and related compounds also increases from October to April. Benzo perylene concentration ranged from 3.5 pg/m3 to 19.4 pg/m3 from October to January.6-nitro benzo pyrene ranges from 2.9 pg/m3 to 7.1 pg/m3 from October until March and 6-nitro benzopyrene is considered a secondary aerosol formed in the atmosphere.

研究分野：Analysis of hazardous components(PAHs)

キーワード：PAH　LCMS　Atmospheric aerosols

１．研究開始当初の背景

In recent years, particulate matter (PM) has become a worldwide health concern. Atmospheric PM affects both climate and health. PM significantly impacts human health, including asthma, respiratory illness, and cardiovascular mortality and morbidity. Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds of multiple aromatic rings. PAHs and their derived oxygenated PAHs (oxy-PAHs) have toxic, mutagenic, and carcinogenic properties. Hence, it is important to understand the abundance, speciation, distribution, and potential sources of PAHs in aerosols, so that the diverse effects of PAHs can be efficiently controlled. PAHs are released from incomplete combustion or pyrolysis of materials containing carbon and hydrogen, such as coal, oil wood, and petroleum products.

２．研究の目的

Understanding the characteristics of PAHs and their derivatives in the atmosphere, the source profiles, and the technology available for controlling PAHs emission is essential to reducing the impact of PAHs. PAHs are mostly formed from saturated hydrocarbon under oxygen-deficient conditions. Pyro synthesis and pyrolysis are the two main mechanisms for PAH formation. The US Environmental production agency (EPA) selected 16 PAHs (Naphthalene, Acenaphthene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Pyrene, Benzo anthracene, Chrysene, Benzo fluoranthene, Benzo pyrene, Dibenzo anthracene, Indeno pyrene, and Benzo perylene) as priority compounds. PAH levels in TSP aerosols over Hokkaido were limited despite diverse anthropogenic and biogenic activities over the region.

３．研究の方法

Sapporo has been selected as a study site for the PAH analysis. Aerosols samples on the rooftop of the Department of health science have been collected**.** The targeted PAH has been extracted with dichloromethane: methanol (2:1) 5ml X3 and filtered using quartz wool until it reaches about 1ml and transferred into an Eppendorf tube and dried completely under nitrogen and redissolve in 100uL of CAN. LC/MS method has been optimized for quantifying PAH, oxy-PAH, and nitro PAH. ZORBAX Eclipse PAH has been selected as a good candidate for the PAH analysis. Solvent A: 10 mM ammonium acetate in Milli-Q water and B: ACN with the flow rate of 200 uL/ min at 30℃ column temperature have been optimized. LOD of most of the PAH is 10 pg, and LOD of oxy-PAH and nitro-PAH is 5 pg. 9 PAHs, 4 oxy-PAH, and 3-Nitro-PAH.

４．研究成果

The PAH compounds detected in the Sapporo aerosol samples are listed in **Table 1**, Most of the species' concentration is max in Dec, Jan, and Feb. Among them, benzo perylene is the most abundant species followed by 6-nitrobenzopyrene and Benzo anthracene**.** As the cold season approaches the concentration of PAH and related compounds also increases. Benzo perylene concentration ranged from 3.5 pg/m^3 to 19.4 pg/m^3 from October to January. Pyrene and fluoranthene are structural isomers, and they can be separated in the current LCMS method. Pyrene shows a significant difference between Oct and Dec. Chrysene and Benzo_a_anthracene are the structural isomers and Benzo_a_anthracene shows a significant difference between Oct and Nov. Benzo_ghi_perylene shows a highly significant difference between Oct to Dec. Interestingly, 6-nitrobenzopyrene concentration increases as the cold season increases ranging from 2.9 pg/m^3 to 7.1 pg/m^3 from October until March, and 6-nitro benzopyrene is considered a secondary aerosol formed in the atmosphere Nitrate-PAHs and oxy-PAH are produced in the atmosphere by secondary processes. In Sapporo aerosols, I detected 4 nitro and 1 oxy PAH but no significant difference between each month. Sample collection is going on and shortly in summer, samples will be analyzed. The photooxidation of these types of molecules is probably high in winter, as shown in **Figure 1**. Even in winter photooxidation is active and secondary aerosol formation is available in the atmosphere. Further, a study will be conducted to know the concentration of PAH and related compounds in the snow samples collected at the Department of Health Science, Hokkaido University.

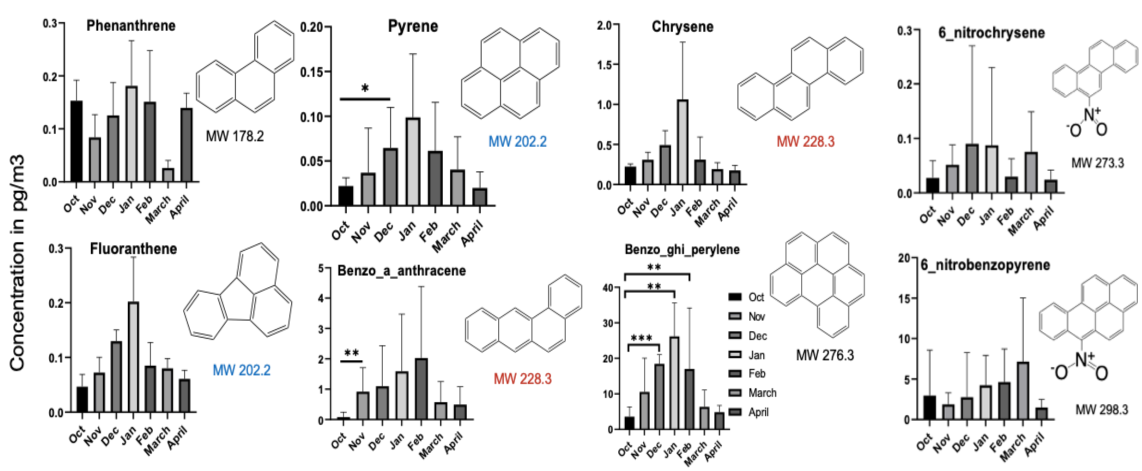

Figure 1. Monthly PAHs and nitrated-PAH concentrations in the Sapporo sampling site.

	October		November		December		January		February		March		April	
	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max	Min	Max
Acenaphthylene	0.000	0.080	0.000	0.024	0.005	0.086	0.000	0.143	0.000	0.107	0.000	0.196	0.000	0.049
Anthraquinone	0.000	0.095	0.003	0.089	0.023	0.053	0.000	0.085	0.000	0.000	0.000	0.091	0.000	0.000
2_nitro_fluorene	0.000	0.436	0.000	0.000	0.000	0.389	0.000	0.404	0.000	0.234	0.000	0.029	0.000	0.205
Acenaphthene	0.323	1.055	0.000	1.035	0.102	1.415	0.000	1.163	0.375	1.003	0.000	2.250	0.301	3.062
Phenanthrene	0.052	0.239	0.016	0.209	0.024	0.153	0.068	0.349	0.000	0.331	0.000	0.054	0.071	0.186
Fluoranthene	0.000	0.103	0.000	0.133	0.122	0.185	0.043	0.311	0.003	0.144	0.040	0.127	0.030	0.089
Pyrene	0.010	0.032	0.000	0.106	0.069	0.104	0.032	0.173	0.006	0.114	0.000	0.072	0.000	0.044
1_nitro_pyrene	0.000	0.031	0.000	0.005	0.000	0.000	0.000	0.000	0.000	0.009	0.016	0.037	0.000	0.017
6-nitrochrysene	0.000	0.057	0.000	0.085	0.000	0.360	0.000	0.252	0.000	0.065	0.000	0.176	0.000	0.041
Benzo_a_anthracene	0.000	0.325	0.000	1.666	0.000	2.715	0.000	3.672	0.275	4.708	0.000	1.329	0.000	1.157
Chrysene	0.126	0.268	0.135	0.567	0.360	0.995	0.066	2.443	0.011	0.874	0.051	0.420	0.000	0.286
6-nitrobenzopyrene	0.000	11.381	0.000	3.146	0.000	11.036	0.000	6.902	0.717	8.845	0.000	18.372	0.000	2.156
Benzo_a_pyrene	0.102	0.418	0.104	0.808	0.720	1.115	0.259	1.195	0.069	1.454	0.233	0.603	0.279	0.345
Benzo_b_fluoranthene	0.000	0.166	0.000	0.187	0.000	0.000	0.000	0.000	0.000	0.019	0.037	0.224	0.074	0.150
Benzo_k_fluoranthene	0.003	0.026	0.000	0.056	0.000	0.174	0.000	0.014	0.001	0.018	0.000	0.139	0.000	0.068
Benzo_ghi_perylene	0.916	7.063	2.742	23.692	15.786	21.278	16.135	34.702	2.074	35.756	0.000	11.393	2.201	6.544

Table 1. The concentration (pg/m^3) of PAH and related compounds were detected in the Sapporo sampling site.

５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　0件／うち国際共著　1件／うちオープンアクセス　0件）

１．著者名 Divyavani Gowda, Marumi Ohno, Siddabasave Gowda B. Gowda, Hitoshi Chiba, Masashi Shingai, Hiroshi Kida & Shu‐Ping Hui	４．巻 11
２．論文標題 Defining the kinetic effects of infection with influenza virus A/PR8/34 (H1N1) on sphingosine‐1‐phosphate signaling in mice by targeted LC/ MS	５．発行年 2021年
３．雑誌名 Divyavani Gowda	６．最初と最後の頁 20161
掲載論文のDOI（デジタルオブジェクト識別子） 10.1038/s41598-021-99765-0	査読の有無 無
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計0件

〔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

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