An Overview of $PM_{2.5}$ Pollution Research Conducted in ERTDF Projects since 2011

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Abstract

Urban and regional air pollution caused by $PM_{2.5}$ and other air pollutants is a major issue worldwide. Air pollution in the developing counties of Asia is especially serious. In Japan, environmental standards for $PM_{2.5}$ were established in 2009. After severe air pollution occurred around Beijing in 2013, long range transboundary air pollution of $PM_{2.5}$ became a public concern in Japan. Since then, much research on $PM_{2.5}$ pollution has been conducted under the Environment Research and Technology Development Fund (ERTDF) funded by the Ministry of the Environment, Japan (MOEJ). This paper gives a comprehensive overview of the outcomes of these projects and discusses issues needing research to promote domestic and international $PM_{2.5}$ pollution policies. These research efforts need to be further strengthened toward solving issues of $PM_{2.5}$ and other air pollutants.

Key words: Environment Research and Technology Development Fund (ERTDF), $PM_{2.5}$ pollution

1. Introduction

The Environment Research and Technology Development Fund (ERTDF) aims to contribute to preservation of the environment in Japan and around the world to establish a sustainable society. It will achieve this by assembling the collective strength of researchers within Japan from a range of fields to promote survey research and technology development in a comprehensive manner from an international academic perspective. The ERTDF website can be accessed here: http://www.env.go.jp/policy/kenkyu/suishin/english/gaiyou/index.html

Particulate matter (PM) with an aerodynamic diameter of less than 2.5 μm ($PM_{2.5}$) is an atmospheric pollutant consisting mainly of several major components such as sulfate, nitrate, ammonium, organics and others. Based on accumulated epidemiological evidence, the World Health Organization (WHO) has issued air quality guidelines for $PM_{2.5}$ of 10 $\mu g/m^3$ for long-term (annual mean) exposure and 25 $\mu g/m^3$ for short-term (24-hour) exposure (WHO, 2005). The rapid growth in economic activity and energy consumption in recent decades has resulted in increased anthropogenic emissions of air pollutants in Asian countries, particularly China (Ohara *et al*., 2007). In the winter of 2013, China faced extreme $PM_{2.5}$ pollution, and the hourly $PM_{2.5}$ concentration reached 886 $\mu g/m^3$ on 12 January in Beijing (https://twitter.com/BeijingAir). This extreme $PM_{2.5}$ pollution was widely reported in the mass media and attracted much public attention in Japan.

Environmental standards for $PM_{2.5}$ were established in Japan in 2009, and since then, many kinds of measures including monitoring of $PM_{2.5}$ have been taken. These actions accelerated after the $PM_{2.5}$ episode of January 2013. In March 2014, the Expert Committee for $PM_{2.5}$ and others of the Central Environmental Council were set up and began discussing $PM_{2.5}$ measures, publishing *Principles of Domestic Emission Control for $PM_{2.5}$ Mitigation (Interim Report)* (MOEJ, 2015) in March, 2015. They suggested that more scientific knowledge about $PM_{2.5}$ needed to be gathered to help promote measures. In this context, research projects related to $PM_{2.5}$ have increased under the ERTDF since around 2011, and a variety of studies are being carried out.

This paper summarizes the research projects related to $PM_{2.5}$ that have been carried out under the ERTDF since 2011 (including ongoing projects), reviewing them from the viewpoint of the policy needs shown in the MOEJ's Interim Report (MOEJ, 2015), and indicates research subjects that should be more emphasis in the future.

2. Review of ERTDF Projects since 2011

2.1 Overview

National Air Quality Standards for fine particulate

matter ($PM_{2.5}$) were established in Japan in 2009. After that, many research projects for $PM_{2.5}$ funded by the ERTDF MOEJ were conducted. This section focuses on and reviews 13 research projects on formation mechanisms of $PM_{2.5}$ and their modeling and 10 research projects on health effects of $PM_{2.5}$ conducted since 2011. Additionally, three topics from these research projects are covered in this special issue: "Characterization and Source Apportionment Studies of $PM_{2.5}$ Using Organic Maker-based Positive Matrix Factorization" by Kumagai and Iijima, "Establishing a Reference Modeling for Source Apportionment and Effective Strategy-Making for Suppressing Secondary Air Pollutants" by Chatani *et al*. and "Epidemiological Studies on Health Effects of Fine Particulate Matter ($PM_{2.5}$) in Japan" by Shima.

2.2 $PM_{2.5}$ Formation Mechanisms and their Modeling

This section gives a summary of the outlines and outcomes of 13 research projects related to $PM_{2.5}$ formation mechanisms and their modeling, funded by the ERTDF since 2011 (including ongoing projects) shown in Table 1, summarized referring to the Ministry of the Environment's Environment Research and Technology Comprehensive Information webpage (MOEJ, 2018).

2.2.1 Brief Summaries and Outcomes of the Research Projects

(1) Current Status Elucidation and Source Contribution Assessment of $PM_{2.5}$ Pollution in Collaboration with Environmental Research Institutes across Japan (5B-1101)

The purpose of this project is to gain a better understanding of the current status of $PM_{2.5}$ across Japan, update $PM_{2.5}$ source profiles and estimate $PM_{2.5}$ source apportionments. The monitoring network has shown some new findings, such as a seasonal mean $PM_{2.5}$ concentration profile that is higher in the west and lower in the east among remote sites, except in summer. Long-range transport and local source fractions largely depend on the season, year and weather conditions. $PM_{2.5}$ emission inventories for industrial soot and heating facilities have been updated.

(2) Integrated Observational and Modeling Study on *Kosa* Impacts throughout Japan for the Japanese $PM_{2.5}$ Regulations (5B-1202)

The three-dimensional distribution of Asian dust and air pollution particles was continuously observed using a ground-based lidar network (AD-Net). At the same time, continuous observation was performed in Seoul with an in-situ polarization particle counter (POPC) that could measure the size and non-sphericity of single particles. A method of characterizing states of mixing using lidar-derived particle depolarization ratios and backscattering color ratios was validated by comparison with the POPC measurement. Also, empirical conversion factors were determined for estimating dust $PM_{2.5}$ and air pollution $PM_{2.5}$ from lidar-derived dust extinction coefficients and spherical aerosol extinction coefficients.

The chemical transport model was improved to calculate $PM_{2.5}$ for dust and other major aerosol components. A dust emission model for the Gobi Desert was improved by an inverse method using observation data. The model estimated the dust emissions from the Gobi desert to 300–600 Tg/year, demonstrating that approximately 5% was deposited in the area surrounding Japan.

(3) Investigation of Un-measured VOCs that might Contribute for Photochemical Oxidant Formation (5-1301)

OH reactivity was measured using a laser instrument and concentrations of 100 kinds of VOC species were measured by chemical analysis. "Missing OH reactivity" was defined as the difference found in reactivity between the laser instrument and chemical analyses. The missing reactivity in the summer trial in a semi-urban area in Tokyo were detected to be 36%. A series of chamber experiments on the photooxidation of VOCs was conducted to evaluate the contributions of unmeasured products to total product OH reactivity. Propene, isoprene, toluene, p-xylene, 1, 3, 5-trimethylbenzene, and several monoterpenes were used as the precursors of secondary products. The contributions of unmeasured products to total product reactivity were found to be 55–72% and were higher than those of unmeasured products to total product reactivity for the other classes of VOCs. Product OH reactivity was simulated using an MCM model that approximated the experimental results for total product OH reactivity. The results suggest that models taking into account products similar to MCM can be applied for predicting OH reactivity of unmeasured products after adjusting product yields. Effects of unmeasured and unidentified VOCs on OH reactivity and photochemical ozone creation potential were evaluated using a regional air quality model. The sum of OH reactivity by unmeasured species using simulated concentrations was much smaller than the observed value. Ignoring unidentified secondary VOCs in air quality models might result in overestimation of the amount of ozone that would be decreased by reducing anthropogenic VOCs emissions.

(4) Characterization and Source Apportionment Studies of $PM_{2.5}$ Using Organic Marker-based Positive Matrix Factorization (5-1403) (Kumagai & Iijima, 2018)

Organic marker compounds in $PM_{2.5}$ were observed over four seasons at urban, suburban, and forested sites in the Kanto region of Japan, and their source contributions to $PM_{2.5}$ were evaluated. At both urban and suburban sites, the concentrations of levoglucosan (a biomass burning marker) increased in winter and fall, whereas dicarboxylic acids (photo-oxidation products) increased during warm seasons. At a forested site, the concentrations of 2-methyltetrols (a biogenic secondary organic aerosol marker from isoprene) tended to increase

Table 1 Summary of research projects related to $PM_{2.5}$ formation mechanisms and their modeling, funded by ERTDF since 2011 (including ongoing projects).

Subject No.	Principal Investigator	Period	Subject name	Key Words	Category						
					1	2	3	4	5	6	7
5B-1101	Seiji SUGATA	2011-2013	Current Status Elucidation and Source Contribution Assessment of $PM_{2.5}$ Pollution in Collaboration with Environmental Research Institutes across Japan	$PM_{2.5}$, Transboundary pollution, Local pollution, Source attribution, Receptor analysis, Remote site observation, Environmental standard	◎						
5B-1202	Nobuo SUGIMOTO	2012-2014	Integrated Observational and Modeling Study on Kosa Impacts throughout Japan for the Japanese $PM_{2.5}$ Regulations	Asian dust, Aerosol, Internal mixing, External mixing, Dust $PM_{2.5}$, Polluted dust, Lidar observation network, Dust forecast model, Transport, Deposition	◎	○					
5-1301	Yoshizumi KAJII	2013-2015	Investigation of Un-measured VOCs that might Contribute for Photochemical Oxidant Formation	OH reactivity, Photochemical oxidant, Unknown reactivity, Unmeasured VOC, Smog chamber, Unidentified secondary VOC	○	○	○			◎	
5-1403	Kimiyo KUMAGAI	2014-2016	Characterization and Source Apportionment Studies of $PM_{2.5}$ using Organic Marker-based Positive Matrix Factorization	Organic carbon, Levoglucosan, Secondary formation, Biogenic source, Biomass burning, Primary organic aerosol, Secondary organic aerosol	○				◎		
5-1408	Seiji SUGATA	2014-2016	Improvement of a Simulation Model and Emission Data and Evaluation of the Aerosol Volatilization Characteristic for the Improvement of the Accuracy of $PM_{2.5}$ Forecast	Organic aerosol, Volatility, Volatility Basis-Set (VBS) model, Oligomer, Emission inventory, Condensed dust, Secondary formation, Photochemical aging, Heterogeneous reaction		◎			○	○	
5-1502	Atsushi SHIMIZU	2015-2017	Development of an Advisory and Assessment System for the Environmental Impacts of Aeolian Dust	Aerosol transport model, Asian dust, Lidar, Data assimilation, Epidemiology		◎					○
5-1506	Yuji FUJITANI	2015-2017	Development of Measurement Method of Semi-volatile Primary Aerosols by Isothermal Dilution at Combustion Sources	$PM_{2.5}$, Condensable particle, Volatility distribution, Volatility basis set, Combustion source, Organic aerosols			○	◎	○	○	
5-1601	Satoru CHATANI	2016-2018	Establishment of a Reference Modeling for Source Apportionment and Effective Strategy Making to Suppress Secondary Air Pollutants	Air quality modeling, Model inter-comparison, Secondary air pollutants, Source apportionment		◎					
5-1604	Kazuo OSADA	2016-2018	Factors Controlling Enhancement of Urban $PM_{2.5}$ and Development of a Supporting Method for Administrative Monitoring Data	Urban $PM_{2.5}$, Ammonia, Organic marker, Open burning, Monitoring data	○				◎		
5-1605	Mizuo KAJINO	2016-2018	Studies on $PM_{2.5}$ Composition, Oxidative Potential, Health Hazard and Their Model Prediction	$PM_{2.5}$, Chemical composition, Oxidative potential, Health hazard, Exposure model, Chemical transport model							◎
5-1708	Masayoshi KIMOTO	2017-2019	Investigation on Removal Characteristics of $PM_{2.5}$ in Flue Gas Treatment Equipments of Large Scale Plant	Fine particle, Fuel gas treatment, Removal characteristic			◎				
5-1801	Yu MORINO	2018-2020	Model, Field, and Laboratory Studies on Source Apportionment of Anthropogenic and Biogenic Organic Aerosols	Organic aerosols, Source apportionment, Condensable particle, Organic maker, Receptor model, Organic aerosol model, Secondary organic aerosol		○		○	◎	○	
5-1802	Tatsuya SAKURAI	2018-2020	Assessment Study for Air Quality Improvement obtained from the 2020 Global Sulphur Limit in Marine Fuels	Marine Fuels Sulphur, Ship emission inventory, Regulation effect, Air quality, Assessment			○				◎

(Note) These research projects are classified into categories (1) - (7): (1) better understanding of formation mechanisms; (2) improvement of chemical transport models; (3) development of emission inventories; (4) clarification of condensable dust emissions from stationary sources; (5) better understanding of formation mechanisms of secondary PM; (6) accumulation of scientific knowledge on VOC; and (7) others. The marks ◎ and ○ indicate primary and secondary focus, respectively.

during warm seasons, especially in summer. The levels of organic markers for cooking and vehicle emissions were higher at the urban site than the other sites. As a result of a positive matrix factorization analysis using a $PM_{2.5}$ component data set, it was possible to apportion 80 to 90% of $PM_{2.5}$ mass concentration to 12 sources, including sources of organic particles such as those of biogenic primary/secondary origin, biomass burning and cooking. The contribution ratios of these sources involved in organic aerosols in $PM_{2.5}$ were estimated to be 41% at urban, 39% at suburban, and 46% at forested sites.

(5) Improvement of a Simulation Model and Emission Data and Evaluation of the Aerosol Volatilization Characteristic for the Improvement of the Accuracy of $PM_{2.5}$ Forecast (5-1408)

To improve the performance of chemical transport models at simulating organic aerosols (OA), the volatility of primary OA (POA) and secondary OA (SOA) were determined. An emission inventory for semi-volatile organic compounds (SVOC) emitted from stationary combustion sources was developed by analyzing measured PM concentrations from stationary combustion sources before and after dilution. Total POA emissions increased by a factor of five and, accordingly, simulated OA concentrations increased greatly in both winter and summer.

(6) Development of an Advisory and Assessment System for the Environmental Impacts of Aeolian Dust (5-1502)

Continuous observations by the East Asian AD-Net lidar network of the vertical distribution of Asian dust were evaluated. The data confirmed a negative trend in Asian dust density over Japan over last 10 years and that intensity was dependent on altitude. The AD-Net data were provided to Japan's Ministry of the Environment for their Dust and Sand-Storm information web page. An analytical method (LETKF; Local Ensemble Transform Kalman Filter) for assimilating satellite observation data (Himawari-8) in the aerosol transport model was developed. Also, a data assimilation method (2D-Var) capable of high-speed computation was developed and a daily data assimilation and prediction experiment was started. In addition, it has become possible to conduct health risk assessments in areas where lidar observations have not been available and epidemiologic studies have not been performed due to lack of current information on concentrations of Asian dust. Furthermore, we propose a method for providing information to vulnerable people and provided it, demonstrating that the provision of information reduced the risk of unscheduled clinic visits by vulnerable people on days with high Asian dust concentrations.

(7) Development of Measurement Method of Semi-volatile Primary Aerosols by Isothermal Dilution at Combustion Sources (5-1506)

For improving $PM_{2.5}$ emission inventories and predictivity of air quality models, it is important to understand emission factors of condensable organic compounds including semi/intermediate volatile organic compounds emitted from combustion sources. For this purpose, we developed a measurement method applied it to diesel automobile exhaust, incineration plants and sludge dryer combustion using bunker A oil. The emission factors were presented as volatility distribution, which is a function of saturation concentrations of the material emitted using two sophisticated methods and one simplified method. We confirmed that the volatility distribution based on each method was similar. Finally, we found it recommendable to apply sophisticated methods to combustion sources using different fuel types and simplified methods to combustion sources as much as possible to estimate the representativeness and variability of combustion sources within the same fuel type in order to develop $PM_{2.5}$ emission inventories that include condensable organic compounds in Japan.

(8) Establishment of a Reference Modeling for Source Apportionment and Effective Strategy-Making to Suppress Secondary Air Pollutants (5-1601) (Chatani, *et al.,* 2018)

Regional air quality modeling is a powerful tool for considering effective strategies to suppress ambient $PM_{2.5}$ concentrations. It is particularly able to evaluate source apportionments, which are critical information for designing effective strategies, by simulating complex photochemical reactions and aerosol formation in the atmosphere. Its performance at reproducing $PM_{2.5}$ concentrations, however, needs improvement.

There are many factors that could alter simulated pollutant concentrations. It is not an easy task for a single model user to get a grasp of the uncertainties arising from these factors overall. The aim of this project, called "Japan's Study for Reference Air Quality Modeling" (J-STREAM), is to promote research in Japan that can provide an assemblage of references on air quality modeling. The variabilities and uncertainties in pollutant concentrations simulated by currently available models are being evaluated using model inter-comparisons. The project is also seeking suitable model configurations among the models participating in these inter-comparisons. Such information could serve as a reference for model users in Japan. A detailed analysis of model performance and source apportionments is being conducted.

(9) Factors Controlling Enhancement of Urban $PM_{2.5}$ and Development of a Supporting Method for Administrative Monitoring Data (5-1604)

The purpose of this project is to gain a clearer picture of the actual emission sources contributing to high $PM_{2.5}$ concentrations in urban areas and propose a new auxiliary methodology for interpreting hourly $PM_{2.5}$ concentration data from monitoring stations using a new sophisticated method of measuring and a conventional

measurement method. For that purpose, field measurements, data analysis and method development are being conducted on the Kanto and Nobi plains, aiming at the three following goals: (1) obtaining scientific knowledge on the formation mechanisms of secondary particles in winter based on field measurements and analysis of ammonia, nitric acid and other gaseous matters; (2) obtaining scientific knowledge on emission sources causing high concentrations of $PM_{2.5}$ based on an analysis of organic and elemental markers using time-dependent or wind direction-dependent air sampling in coastal and inland areas; and (3) proposing an auxiliary methodology for interpreting hourly $PM_{2.5}$ concentration data from monitoring stations based on the relationship between humidity, volume concentration of particles and chemical composition measured by a light scattering particle detection sensor or other methods not relying on particulate collection.

(10) Studies on $PM_{2.5}$ Composition, Oxidative Potential, Health Hazard and Their Model Prediction (5-1605)

The purposes of these studies are to grasp the relationship between $PM_{2.5}$ and respiratory illness based on atmospheric field observations focusing on the oxidative ability of particulate matter as an index of harmfulness to health, to understand the spatiotemporal variations of that ability using an air quality model, and to evaluate the validity of air quality policies based on the weight concentration of particulate matter. For this purpose, automatic instrumental measurement of the oxidation ability of $PM_{2.5}$ was newly developed and simultaneous measurement of oxidation ability, metals and organic matters was conducted. In addition, the quantitative relationship among emission sources, chemical matter, oxidation ability and oxidation stress were determined through a cell exposure experiment and factor analysis. As a result, we newly propose "respiratory illness hazard" for oxidation stress caused by chemical matter deposited in the respiratory tract. Additionally, a combined model was created incorporating a three-dimensional chemical transport model and a respiratory tract deposition model based on this knowledge, and used to clarify the source contributions and spatiotemporal variations of respiratory illness hazard. Finally, the difference between risk assessments based on the hazard in this study and conventional method based on $PM_{2.5}$ will be clarified through both observation and modeling.

(11) Investigation on Removal Characteristics of $PM_{2.5}$ in Flue Gas Treatment Equipments of Large Scale Plant (5-1708)

The electric dust collecting equipment, bag filters and wet removal equipment are widely adopted as flue gas treatment equipment in large-scale plants. However, the solid-fine-particle dust collecting characteristics have not been sufficiently clarified and depend highly on the operating condition of the equipment. The purpose of this study is to grasp the solid-fine-particle dust collecting characteristics of each kind of flue gas treatment equipment and the influence of operating conditions. In the first year, the solid-fine-particle dust collecting characteristics of electric dust collecting equipment and the influence of electric charge conditions were investigated, along with characteristics by particulate size when bag filters with unused filter cloth were used.

(12) Model, Field, and Laboratory Studies on Source Apportionment of Anthropogenic and Biogenic Organic Aerosols (5-1801)

Focusing on organic aerosols, of which the dynamics and emission sources are poorly understood and are difficult to predict using models, an innovative organic aerosol model is being developed to facilitate elucidation of the formation mechanisms and origins of organic aerosols using laboratory experiments and field observations. Firstly, a kinetics model considering production and loss processes of SOA is being developed. Secondly, the emission and transformation process of condensable dust is being modeled and validated based on emission estimations using vapor pressure and dust oxidation experiments in a chamber. Finally, atmospheric observations of the organic markers of anthropogenic SOA are being conducted, and the sources of the organic aerosols are being estimated by a receptor model based on the observed data. These results are being used for validation and improvement of the organic aerosol module in the chemical transport model. Through this study, it is expected that the contributions of evaporative sources of VOC and stationary combustion sources to organic aerosols can be estimated.

(13) Assessment Study for Air Quality Improvement Obtained from the 2020 Global Sulphur Limit in Marine Fuels (5-1802)

In this study, improvement in $PM_{2.5}$ pollution from ships as a result of stronger regulation of sulphur in ship fuel oil (hereafter, regulation reinforcement) in 2020 is quantitatively estimated. Long-term/regional-scale estimation including secondary particles and short-term/local-scale estimation of primary particles is being conducted using observation and numerical simulation. In the long-term/regional-scale estimation, observations of $PM_{2.5}$ concentration and boundary layer height will be conducted each year before and after regulation reinforcement on the Seto Inland Sea coast. The effect of regulation reinforcement will be estimated through a combined analysis of these observation data with other weather, air quality and ship traffic data and through use of an air quality model. In addition, the process by which ship emissions on the Seto Inland Sea influence $PM_{2.5}$ in the coastal area will be clarified and the contribution of ship emissions to $PM_{2.5}$ after the regulation reinforcement will be estimated using a model. In the short-term/local-scale estimation, the change in ships' contribution caused

by regulation reinforcement will be estimated by observing primary pollutants (SO_2, NO_x, particulate matter) on both sides of the Kanmon channel, close to sea lines. In addition, using a diffusion model, the ships' emissions will be inversely estimated quantitatively from the concentration contributed by ships and the change in the ships' emissions due to regulation reinforcement.

2.2.2 Summary and Discussion

In Table 1, the research projects are classified into categories (1) – (7) based on the policy needs indicated in the "Interim Report" (MOEJ, 2015) : (1) better understanding of formation mechanisms, (2) improvement of chemical transport models, (3) development of emission inventories, (4) clarification of condensable dust emissions from stationary sources, (5) better understanding of formation mechanisms of secondary PM, (6) accumulation of scientific knowledge on VOC, and (7) other projects. As can be seen in Table 1, some features of the $PM_{2.5}$ research projects conducted by ERTDF at present or in the past are as follows:

- There have been few studies in category 3 (emission inventories). Emission-related research such as developing an emission inventory system and reducing uncertainties in emission inventories based on inversion modeling have not been carried out so far.
- There are few projects primarily corresponding to category 6 (VOCs).

Based on these weaknesses in previous research, strengthening of this research field to contribute to policy making in the future is expected. In addition, it will be necessary to conduct research on assessment of technology and measures for $PM_{2.5}$ reduction, develop and evaluate $PM_{2.5}$ measurement technology, and conduct future prediction and mitigation scenario studies for $PM_{2.5}$ reduction. Furthermore, with regard to both climate change and air pollution, studies of co-benefit/co-control policies, interaction among phenomena, and co-effects on human health and ecosystems will be needed.

2.3 Health Effects

In this section, outlines and outcomes of 10 research projects related to the health effects of $PM_{2.5}$, funded by ERTDF since 2011 (including ongoing projects) shown in Table 2 are summarized, referring to the Ministry of the Environment's Environment Research and Technology Comprehensive Information webpage (MOE, 2018).

2.3.1 Brief Summaries and Outcomes of Research Projects

(1) Quantitative Evaluation of Desert Dust (Asian Dust) on Respiratory/Allergy Risk, Taking into Consideration Times Spent Outside (5C-1152)

The purpose of this project is to examine possible health effects of desert dust exposure on pregnant women and their children, as an adjunct study of the Japan Environment and Children's Study (JECS) involving three regions: Kyoto, Toyama and Tottori. Light Detection and Ranging (LIDAR) was used with a polarization analyzer for exposure measurements. The outcomes were allergic symptoms among the mothers and development of asthma and other allergic or respiratory diseases among their children. The data were acquired in a time-coordinated manner by connecting local LIDAR equipment to an online questionnaire system. The participants answered the online questionnaire using their mobile phones or personal computers.

The pregnant women were more likely to develop allergic symptoms on Asian dust days compared with other days. Those who had a past history of allergic rhinitis or pollinosis and who spent times outside on the day had a significantly higher risk of developing symptoms than those who did not. Children with higher exposure to Asian dust during the first year in their lives developed wheezing or asthma/asthmatic bronchitis earlier than children with lower exposure. This was more apparent among boys than girls.

(2) Research to Reveal Association of Human Health and Asian Dust by the Clinical and Basic Study (5C-1154)

The purpose of this project is to investigate the effects of Asian dust storms (ADS) on the respiratory system. A telephone survey of the effects on respiratory tract symptoms was conducted for patients with asthma and chronic obstructive pulmonary disease (COPD). The adverse effects of ADS on patients with asthma were greater than on the patients with COPD. Peak expiratory flow (PEF) testing was conducted with school children in spring for two years. The PEF values in these children were significantly reduced upon exposure to ADS. The decrease in PEF was greater in the children with asthma. Substances attached to airborne ADS particles may increase airway inflammation by elevating interleukin-8.

Exposure to ADS aggravates respiratory tract symptoms in adult patients with asthma and COPD, and in schoolchildren. Substances attached to airborne ADS particles may increase airway inflammation by elevating interleukin-8 (IL-8).

(3) Studies on Biological Effects and Clarification of the Mechanisms of Asian Dust Aerosol, Attached Microorganisms and Chemical Substances (5C-1155)

Asian sand dust (ASD) contains various air pollutants. In this project, the aggravating effects and mechanism of microbial elements and chemical products on allergic diseases were investigated. Direct sampling and bioanalysis over Suzu City on the Noto Peninsula using a tethered balloon and an airplane were conducted in order to reveal the health effects of *kosa* bioaerosol. Gram-positive and gram-negative bacteria and *Bjerkandera adusta* fungus increased in the atmosphere during dust storm events. Two kinds of ASD were collected using a high-volume air sampler from the atmosphere in Kitakyushu, Fukuoka on 1–3 May, 2011

Table 2 Summary of research projects related to health effects of $PM_{2.5}$, funded by ERTDF since 2011（including ongoing projects）.

				key words	methods	subjects	outcomes
5C−1152	Takeo NAKAYAMA	2011−2013	Quantitative Evaluation of Desert Dust（Asian Dust）on Respiratory/Allergy Risk, taking into consideration times spent outside	Asian dust, Kosa, transboundary air pollution, PM2.5, Asthma, Allergy, Maternal and Child Health	online questionaire	pregnant women and their children	asthma, allergic or respiratory diseases
5C−1154	Masanari WATANABE	2011−2013	Research to Reveal Association of Human Health and Asian Dust by the Clinical and Basic Study	Asian dust, Airborne particles, Asthma, COPD, School children, Pulmonary function, Airway inflammation, Interleukin-8, PM2.5	telephone survey	school children and their parents	asthma, COPD, respiratory synptoms, daily recording survey, peak expiratory flow
5C−1155	Takamichi ICHINOSE	2011−2013	Studies on Biological Effects and Clarification of the Mechanisms of Asian Dust Aerosol, Attached Microorganisms and Chemical Substances	Asian sand dust, Allergic diseases, Airway epithelial cells, Immune cells, Splenocytes, Male reproductive function, Kosa bioaerosol	cell exposure experiments,		asthma, allergic rhinitis, airway epithelial cells, Immune cells, Splenocytes, reproductive function
5−1452	Akinori TAKAMI	2014−2016	Research on Health Effects of Short-term Exposure to PM2.5, Composition, and Asian Dust Particles on Cardiovascular and Respiratory Diseases	PM2.5, Asian dust, Transboundary air pollution, chemical composition, Health effects, Emergency transportation, Acute myocardial infarction	Emergency transportation, hospitalization database	Emergency transportation, hospitalization	acute myocardial infarction
5−1453	Masanari WATANABE	2014−2016	A Study to Determine the Toxicity of Substances Contained in Asian Dust and PM2.5, and Monitor Their Effect on Health	Air pollution monitoring, Allergy, Cancer, Component analysis, Epidemiological study, Inflammation, Asian dust, PM2.5, Schoolchildren	questionaire	school children	respiratoy system, peak expiratoy flow, allergy, carcinogenisity, inflammation
5−1456	Masayuki SHIMA	2014−2016	An Epidemiological Study on Effects of Fine Particulate Matter（PM2.5）and Ozone on Respiratory Health in Areas with Different Air Pollution Levels	Fine particulate matter, Chemical composition, Ozone, Asthma attacks, Pulmonary fuction, Positive Matrix Factorization（PMF）analysis, Chemical transport model, Epidemiology	hospital and clinic database	hospital and clinic database, school children	asthma attacks, peak expiratory flow, forced expiratory volume in 1 second
5−1457	Takamichi ICHINOSE	2014−2016	Aggravating Effects of the Combined Air Pollution by Asian Dust and PM2.5 on Lung Inflammation and Allergy Disease, and Elucidation of the Mechanism	PM2.5, Asian sand dust, Combined exposure, Inflammation, Immune response, Lung allergy, Asthma, Allergic conjunctivitis, Quinone, PAHs , Microbial elements, LPS（Lipopolysaccharide）	cell exposure experiments,		Inflammation, Immune response, Lung allergy, Asthma, Allergic conjunctivitis
5−1555	Takeo NAKAYAMA	2015−2017	Short-Term Health Effects on Infants of Asian Dust：Considering Fossil Fuel Related Air Pollution as an Effect Modifier	PM2.5, Asian dust, PAH, PAH-quinone, cross-border air pollution, source apportionment, infants, pregnant women, allergy, JECS	questionaire	pregnant women, infants	allergy
5−1651	Tomoaki OKUDA	2016−2018	Identification of the Factors Responsible for the Health Effects of PM2.5 by Newly Developed Sampling Methods and Exposure Experiments	physical and chemical characteristics, cyclone based particle sampler, cell exposure experiment	cell exposure experiments,		respiratory system, immunological system
5−1751	Akinori TAKAMI	2017−2019	A Study for Acute Effects of Stroke and Mortality Caused by PM2.5 and Coarse Particle	Stroke, mortality, emergency transportation, out-hospital cardiac arrest, PM2.5 monitoring databse, chemical components database, receipt data	hospital database	hospital database	stroke, cardiac arrest

(ASD1) and 12–14 May, 2011 (ASD2), after dust storm events. ASD2 had higher levels of PAHs, LPS and β-glucan, compared with ASD1, but ASD1 was richer in SiO_2 than ASD2. The effects of ASD on splenocytes were also investigated. Western blotting demonstrated that nuclear factor kB (NF-kB) was activated in splenocytes. These novel findings may serve as a warning about the deleterious effects of airborne desert dust on the human respiratory system.

(4) Research on Health Effects of Short-term Exposure to $PM_{2.5}$, Composition, and Asian Dust Particles on Cardiovascular and Respiratory Diseases (5-1452)

This project consists of epidemiological studies to elucidate PM effects on human health and chemical analyses of PM to investigate the influence of transboundary air pollution on local air quality. PM sampling was performed in Kumamoto and Fukuoka, Kyushu, where transboundary air pollution is severe. In the epidemiological studies, emergency transportation data from seven major cities throughout Japan provided by the Fire and Disaster Management Agency, and data on hospitalization due to acute myocardial infarction (AMI) in Kumamoto Prefecture collected by the Kumamoto Acute Coronary Events (KACE) Study Group were used. The concentrations of sulfate ions and highly oxidized organics were higher in the daytime with high $PM_{2.5}$ concentrations, indicating that transboundary air pollution was influencing the local air quality of Fukuoka and Kumamoto, Kyushu. The epidemiological studies suggested that short-term exposure to $PM_{2.5}$ increased emergency transportation due to acute illnesses.

(5) A Study to Determine the Toxicity of Substances Contained in Asian Dust and $PM_{2.5}$, and Monitor Their Effect on Health (5-1453)

This study aimed to investigate the effects of $PM_{2.5}$, PM_{10} and Asian dust (AD) on the respiratory system in schoolchildren and the inflammatory potential of PM. The schoolchildren's morning peak expiratory flow was measured daily as a respiratory function and respiratory symptoms were recorded. Inflammatory potential was assessed to measure production of cytokines such as IL-8 in THP1 cells stimulated by PM_{10} and $PM_{2.5}$ collected daily during the epidemiology study. These IL-8 concentrations were determined as the daily inflammatory potential of PM_{10} and $PM_{2.5}$. PM_{10}, $PM_{2.5}$ and AD particle levels were not significantly associated with respiratory function. By contrast, this study found a significant negative association between respiratory function and daily levels of IL-8 induced by PM_{10} and $PM_{2.5}$. The association between respiratory symptoms and exposure to PM_{10} and $PM_{2.5}$ was found to be similar. These findings suggest that the effects of airborne PM exposure on the respiratory system of schoolchildren differ with the type and source of PM_{10} and $PM_{2.5}$ and might depend more on pro-inflammatory reactions among the responses to the PM composition rather than the PM mass concentration. According to the analysis, among the components of PM collected, endotoxins constitute one of the important factors in harm to the respiratory system due to exposure to PM_{10}, $PM_{2.5}$ and AD particles.

(6) An Epidemiological Study on Effects of Fine Particulate Matter ($PM_{2.5}$) and Ozone on Respiratory Health in Areas with Different Air Pollution Levels (5-1456) (Shima, 2018)

In order to evaluate the effects of fine particulate matter ($PM_{2.5}$) and ozone on respiratory health, epidemiological studies were conducted in two regions located around the Seto Inland Sea.

In Himeji City, Hyogo, weekly data on asthma attacks from hospitals and clinics and daily numbers of primary care visits were collected. The mass concentrations and chemical components of airborne $PM_{2.5}$ were continuously measured. The association between the average concentration of $PM_{2.5}$ and weekly asthma attacks was significant in subjects aged 15–64 years. The concentrations of water soluble organic carbon and particle acidity were significantly associated with weekly asthma attacks. These associations were marked in spring and summer.

On Yuge Island, Ehime, peak expiratory flow (PEF) and forced expiratory volume in one second (FEV1) were measured every morning in spring and autumn among 48 healthy students. The concentration of $PM_{2.5}$ was continuously measured, and the chemical components of $PM_{2.5}$ were analyzed at the school. PEF and FEV1 significantly decreased in relation to increased $PM_{2.5}$ concentrations. Decreases were also significantly associated with various components of $PM_{2.5}$, including carbon and sulfate ion. In addition, pulmonary function was associated with sulfate suggesting transboundary advection.

(7) Aggravating Effects of the Combined Air Pollution by Asian Dust and $PM_{2.5}$ on Lung Inflammation and Allergy Disease, and Elucidation of the Mechanism (5-1457)

This project tried to elucidate the effects of *kosa* and $PM_{2.5}$ on pulmonary inflammation and allergic asthma, find the components of those particles affecting inverse effects on pulmonary inflammation and allergic diseases, and evaluate the combined exposure effects of these components.

An epidemiologic survey demonstrated a significant association between the number of outpatient visits for allergic conjunctivitis and the $PM_{2.5}$ level. In experimental studies, ASD acted as an adjuvant to promote allergic disease induced by allergens such as pollen and fungi. Also, $PM_{2.5}$ and ASD promoted pollen-induced allergic inflammation through conjunctival epithelial damage. Furthermore, particle-bound microbial elements exacerbated pollen-induced allergic conjunctivitis *via* a TLRs/MyD88-signaling pathway. In an in *vitro* study, $PM_{2.5}$ increased the level of

inflammatory molecules from cultured human conjunctival epithelial cells.

(8) Short-Term Health Effects on Infants of Asian Dust: Considering Fossil Fuel Related Air Pollution as an Effect Modifier (5-1555)

Asian dust (AD) exposure is associated with increased risk of exacerbation of allergic symptoms in pregnant women. There is limited evidence, however, regarding the influence on infants, who are theoretically anticipated to be vulnerable to air pollution, whether within Japan or overseas. Also, how much of an effect AD and $PM_{2.5}$ actually exert, and on the other hand, what must be reduced to effectively reduce their effect, are not yet understood. AD particles contain anthropogenic pollutants such as PAHs. Further, their catalytic effects promote secondary formation of chemicals, one of which, PAH-quinone, is known to generate ROS in the human body, which may contribute to allergic diseases.

How much of the effects of AD can be attributed to PAH-quinone and the effects of $PM_{2.5}$, AD and PAH-quinone on infants who had never wheezed were estimated. Both AD and PAH-quinone showed significant associations with allergic symptoms both in pregnant women and infants who had never wheezed. In pregnant women, the term of interaction of AD and PAH-quinone was significantly larger than 0 ($P<0.1$), and it was thought that the effects of AD were attributable to PAH-quinone. In infants who had never wheezed, AD and PAH-quinone were independently associated with symptom development.

(9) Identification of the Factors Responsible for the Health Effects of $PM_{2.5}$ by Newly Developed Sampling Methods and Exposure Experiments (5-1651)

The adverse effects of $PM_{2.5}$ depend on its physical and chemical characteristics. Thus research on its health effects needs to be conducted, focusing on differences in the particles' components. In this project, $PM_{2.5}$ collection using a newly developed cyclone-based instrument, and a chemical components analysis and exposure experiment using collected particles were conducted. The final aim was to define the factors and chemical components that contribute to health effects.

Two approaches were used: 1) development of a new particle sampler that can collect large amounts of fine and coarse particles, 2) cell exposure experiments using the particles collected.

In the first project, sampling fine and coarse particles at three points in Japan and analyzing the components of the particles collected began. In the second project, cell exposure experiments using fine and coarse particles collected in Yokohama and two standard particles (fine particles from automobile exhaust and total particles from city air pollutants) were started.

(10) A Study for Acute Effects of Stroke and Mortality Caused by $PM_{2.5}$ and Coarse Particle (5-1751)

The purpose of this project is to elucidate the acute effects of stroke and mortality caused by $PM_{2.5}$ and coarse particles using the mortality and emergency transportation cases in Japan and stroke database of Fukuoka Stroke Registry (FSR). The following three matters were investigated: 1) the effects of $PM_{2.5}$ on mortality and out hospital cardiac arrest in Japan, 2) acute health effects of $PM_{2.5}$ and coarse particle on stroke in Fukuoka, 3) the effects of $PM_{2.5}$ on emergency transportation cases and outpatients in Japan.

The present situations is as follows: 1) Data were collected via $PM_{2.5}$ monitoring data in Japan, a chemical components database of $PM_{2.5}$ in four seasons compiled by MEJ, a chemical components database of three cities, a mortality database in Japan and an outpatient cardiac arrest database in Japan, and analyses of those data were started. 2) FSR data collection was continued, and an analysis of the relationship between FSR data and concentration and chemical components of $PM_{2.5}$ collected in Fukuoka City was started. 3) Emergency transportation and reimbursement specification (receipt) data in Japan were collected.

2.3.2 Summary and Discussion

Table 2 provides an outline of the subjects, methods and outcomes of the projects. Seven are epidemiological studies and the other three are experimental studies.

Many of the epidemiological studies focus on allergic diseases, respiratory symptoms and immune responses, for instance asthma, PEF and FEV1. Other studies focus on strokes and myocardial infarctions. Among the epidemiologic studies underway, there are questionnaire studies; online surveys (by telephone), interview surveys and studies using hospital databases. The subjects of the questionnaire studies are mainly pregnant women and their children (infants), and school children. The studies using hospital databases cover all age groups.

In the three experimental studies, experimental cells are exposed to PM2.5, particles and components of particles. In these studies, respiratory systems, immunological systems, splenocytes, reproductive function, inflammation, etc. are being tested.

Regarding the previous research overall, including ongoing research, almost none focuses directly on future prediction. In addition to research using high quality air pollution (PM2.5) datasets acquired using newly developed measurement and estimation methods, research focusing on future prediction of air pollution (PM2.5) taking climate change into account is needed.

3. Conclusions

$PM_{2.5}$ research projects funded by ERTDF since 2011 are summarized and reviewed herein from the viewpoint of policy needs. Additionally, research subjects that should receive more emphasis in the future are suggested. ERTDF is expected to promote research that contributes to mitigation of $PM_{2.5}$ and other air pollutants, based on strong collaboration between policy

makers and scientists.

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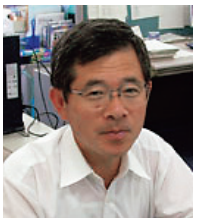

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