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A Study on Urban Air Pollution Improvement in Asia

Overview of Policy Actions and Observational Data for PM$_{2.5}$ and O$_3$ in Japan: A Study of Urban Air Quality Improvement in Asia

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Overview of Policy Actions and Observational Data for PM$_{2.5}$ and O$_3$ in Japan: A Study of Urban Air Quality Improvement in Asia

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Abstract

Recent topics on PM$_{2.5}$ and O$_3$ in Japan have been briefly discussed in this paper. In the first part, Japan's policy measures for PM$_{2.5}$, including the establishment of Environmental Quality Standards (EQS) and monitoring stations, are described. Additionally, we discuss the monitoring data obtained and the exceedance of EQS in the years 2010-2013. The nationwide averaged data shows that sulfate and EC/OC are the most prominent components of PM$_{2.5}$ and contribute almost equally to it . Secondly, long-term variation of O$_3$ (O$_x$) and its precursors, NO$_2$ and NMHC/VOC (non-methane hydrocarbon/volatile organic compound) are presented. The paradox of the increase in the average concentration of O$_3$ in spite of the decrease in ambient concentrations of NO$_2$ and VOC is discussed. The phenomenon was found to reflect three components: (1) a decrease in the NO titration effect, (2) an increase in transboundary transport, and (3) a decrease in in situ photochemical production. It is proposed that the integrated approach to mitigation measures for PM$_{2.5}$ and O$_3$ pollution should be considered within a framework of the SLCPs (short-lived climate pollutants) co-control policy.

Keywords: PM$_{2.5}$, 0$_3$, Environmental Quality Standards (EQS), SLCPs co-control policy, 0$_3$ paradoxical trend

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1. Introduction

There is a general consensus among atmospheric scientists that from a human health impact viewpoint, $PM_{2.5}$ and O_3 are of most concern for regional air pollution (WHO 2006; Anenberg 2010; Fann et al. 2012; Weinchenthal 2013). O_3 pollution, known as photochemical air pollution, has a more than 60 year history (Finlayson-Pitts and Pitts 2000; Akimoto 2016). Significant studies have been made for elucidating the O_3 formation mechanisms and other oxidants in the urban, suburban, and remote atmosphere (Akimoto 2016). Additionally, numerous policy measures have been taken to mitigate photochemical ozone pollution in most developed countries (see for example, European Commission 2006; UN ECE 2006). Nevertheless, the decrease in ground level O_3 to a level below that which is hazardous to human health has not yet been attained by the United States, Japan or many counties in Europe (Ohizumi et al. 2013).

In contrast to ground level O_3, the issue of $PM_{2.5}$ pollution is relatively new. The air quality standards for $PM_{2.5}$ were first set in the United States in 1997 (USEPA 1997). In Japan, the environmental quality standards (EQS) of $PM_{2.5}$ were set in 2007, and monitoring under the administration of the national and local governments started after that. Compared to ozone pollution, there is still a lot more uncertainty as to the primary and secondary formation of $PM_{2.5}$ and its temporal and spatial variability in the atmosphere.

The purpose of this paper is to introduce policy actions for $PM_{2.5}$ including the establishment of EQS for $PM_{2.5}$, and monitoring and data dissemination within Japan as a way of providing information to environmental and policy scientists concerning the situation of $PM_{2.5}$ control in East Asia. In the second half of the paper, the observational data, which includes trends, seasonal variations of mass concentrations, and the analysis of chemical compositions, are presented together with the present status of O_3 pollution. The importance of simultaneous mitigation of $PM_{2.5}$ and O_3 as a control policy will also be discussed.

2. $PM_{2.5}$

2.1 Policy actions

2.1.1 Establishment of Environmental Standards for $PM_{2.5}$

Serious administrative concern for $PM_{2.5}$ within the Ministry of Environment Japan (MOEJ) began in 1997. New knowledge on the effects that $PM_{2.5}$ has on human health was accumulated mainly in the US in the first half of the 1990s; this then led the USEPA to introduce the National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ in 1997. In 1999, after the initial review of

the methods for measuringPM$_{2.5}$, the MOEJ formally began a survey on the effects of exposure to fine particulate matter on human health and initiated studies on epidemiology and toxicology. The report was published in 2007 (MOEJ 2007). The MOEJ established the "Advisory Committee on Evaluation of Health Effects of Fine Particulate Matters" in 2007, and the report was published in 2008 (MOEJ 2008a). Then, in response to the report of the Expert Panel on the Risk Evaluation Method of Fine Particles (MOEJ 2008b), which was established under the Central Environmental Council (CEC) of Atmospheric Environment, the Minister of the Environment consulted the CEC on the establishment of the EQS for fine particulate matter. Finally, according to the report submitted by CEC, the EQS for PM$_{2.5}$ was officially announced and was effective as of September 9, 2009 (MOEJ 2009).

The EQS of PM$_{2.5}$ that was determined in Japan states that the annual standard for PM$_{2.5}$ should be less than or equal to 15 μg m^{-3}. The 24-hour standard, which employs the annual 98th-percentile values at designated monitoring sites for each area, should be less than or equal to 35 μg m^{-3}. The reference measurement method is stated as a mass measurement with a filter sample collection.

Table 1 shows the EQS of PM$_{2.5}$ in Japan comparing air quality standards (AQSs) in other countries with the WHO guidelines (WHO 2006). As seen in Table 1, the AQSs of PM$_{2.5}$ vary widely by country. The Japanese standards enacted in 2009 are the same as the NAAQS in the US in 2006, while the annual average value of the latter has been lowered to 12 μg m^{-3} since 2013. It should be noted that the WHO has the most stringent guidelines, and that legal frameworks on air quality/environmental standards are different from country to country.

Table 1. Comparison of air quality standards of PM$_{2.5}$ unit: μg m^{-3}

Country and Organization	Annual Average	24-hours average	Notes
Japan	15	35	Enacted in 2009.
China (Grade II for residential)	35	75	Enacted in 2012 as precedent enforcement nationwide enforcement 2016.
Korea	25	50	Enacted in 2011/enforcement 2015.
India	40	60	Enacted in 2009.
USA	12	35	Enacted as 15, 65 in 2016; revised as 15, 35 in 2006; revised as shown in 2013.
EU	20	---	Enacted 2008/1st stage target to be achieved by 2015, 25/2nd stage target to be achieved by 2020.
WHO Guideline	10	25	Set in 2006

Source: Author.

After the establishment of the EQS, in 2013 the Expert Panel on Fine Particulate Matters ($PM_{2.5}$) under the MOEJ, determined the "Interim Guideline for Calling for Attention" as a daily averaged value of 70 μg m^{-3}, which is not based on law.

2.1.2 Monitoring and Data Disseminatio

Since the EQS of $PM_{2.5}$ was established, the number of monitoring sites for the mass concentration of $PM_{2.5}$ which reported data over 250 days had increased to 870 nationwide by 2014 over. Of these sites, 672 are non-roadside stations called "General Environment Air Monitoring Stations" and 198 are roadside stations called "Motor Vehicle Exhaust Monitoring Stations" (MOEJ 2016a). In terms of the EQS of $PM_{2.5}$, the standard method for measuring the mass concentration of $PM_{2.5}$ has been specified as the manual filter-sampling mass concentration measurement, as described above. The automatic measuring instruments, which have been verified to give values equivalent to the standard method, have been accepted for evaluating the environmental standards (MOEJ 2013). As a result of the equivalence test specified by the MOEJ, several automatic instruments have been adopted for the purpose of mass concentration monitoring at the routine monitoring sites.

Although the EQSs of $PM_{2.5}$ have been set as mass concentration, component analysis is necessary for apportioning the sources of $PM_{2.5}$ and taking mitigation action accordingly. Here it is worthwhile noting that $PM_{2.5}$ is composed of primary aerosols that are emitted directly from the sources and secondary aerosols that are formed in the atmosphere from precursor gases. The primary $PM_{2.5}$ is typically emitted from the burning of biomass and fossil fuels, and soil dust, which contains inorganic metals, black carbon, and organics such as levoglucosan. Secondary $PM_{2.5}$ also consists of inorganic salt SO_4^{2-}, NO_3^-, NH_4^+, and organic particles such as succinic acid and pinonic acid, which are formed by the photochemical oxidation of biogenic and anthropogenic hydrocarbons in the atmosphere.

In order to facilitate component analysis, the MOEJ published component measuring manuals for $PM_{2.5}$ on the internet (available from MOEJ 2016c). Manuals for inorganic ions (SO_4^{2-}, NO_3^-, Cl^-, Na^+, K^+, Ca^{2+}, Mg^{2+}, NH_4^+, etc.), inorganic elements (Na, Al, K, Ca, V, Cr. Mn, Fe, CO, Ni, Cu, As, Pb, etc.), carbonaceous components (elemental carbon, EC, and organic carbon, OC), polycyclic aromatic hydrocarbons (PAHs), water-soluble organic carbons (WSOC), levoglucosan, and gaseous species (SO_2, HNO_3, HCl, NH_3) have also been prepared. In addition to these components, a manual on the methods for measuring tracer materials of primary and secondary organic particles has also been released (MOEJ 2016c). Levoglucosan is a good indicator of biomass burning, and succinic acid ($HOOC\text{-}(CH_2)_2\text{-}COOH$) (partly primary) and

pinonic acid, which are formed in the secondary photochemical reactions in the atmosphere, have been selected as secondary organic aerosol (SOA) tracers from anthropogenic and biogenic hydrocarbons, respectively. Among the routine monitoring stations measuring mass concentrations, a substantial number of stations measure $PM_{2.5}$ components for two weeks in every four weather seasons. As of 2014, component analysis for inorganic ions and BC/OC has been reported at 14 sites (MOEJ 2016a).

The collected hourly data of mass concentrations of $PM_{2.5}$ by continuous monitoring is available to the public through the Atmospheric Environmental Regional Observation System or "AEROS" (Japanese name "Soramamekun" <http://soramame.taiki.go.jp>) during one week. Compiled hourly, monthly, and annually, the averaged data since 2010 is accessible to the public on the homepage of the National Institute for Environmental Studies (NIES) (http://www.nies.go.jp/ igreen/index.html).

The $PM_{2,5}$ forecasts are publicized unofficially by the NIES on the Visual Atmospheric Environment Utility System (VENUS) http://envgis6.nies.go.jp/osenyosoku/; by Kyushu University through the Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) http://sprintars. riam.kyushu-u.ac.jp/; and by the Japan Weather Association at http://www.tenki.jp/particulate_ matter/. An official forecast system for $PM_{2.5}$ has not yet been established in Japan.

2.2 Observational Data and Analysis

2.2.1 Trends of mass concentrations and exceedance of EQSs

Figure 1 depicts the recent trends in the annual national averages for $PM_{2.5}$ concentrations at non-roadside and roadside stations. Table 2 shows the total number of stations and the number of stations which attained the annual and 24-hour average EQSs.
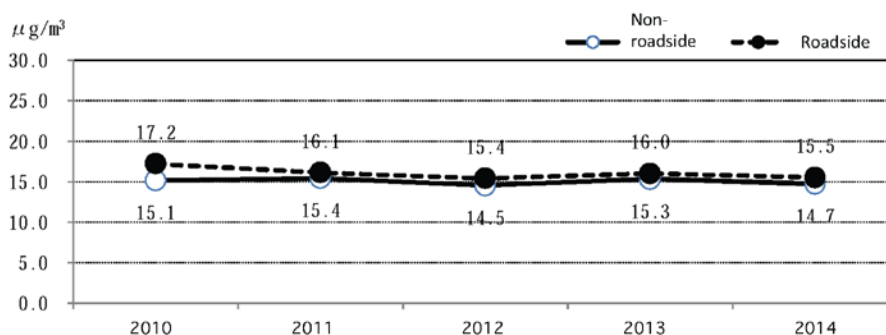

Figure 1. Trends in the annual national averages for $PM_{2.5}$ concentrations at non-roadside and roadside stations in Japan
Source: MOEJ 2015.

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Table 2. Number of effective stations for PM$_{2.5,}$ and percentage of stations that attained annual- and 24-hour average EQS

		2010	2011	2012	2013	2014
Non-roadside stations	Effective number of stations	34	105	312	492	672
	Percentage (%) of stations which attained annual-average EQS	52.9	47.6	61.5	44.3	60.3
	Percentage (%) of stations which attained 24-hour average EQS	32.4	28.6	44.6	16.3	40.6
	Annual-average concentration of PM$_{2.5}$ (μg/m^3)	15.1	15.4	14.5	15.3	14.7
Roadside stations	Effective number of stations	12	51	123	181	198
	Percentage (%) of stations which attained annual-average EQS	16.7	33.3	45.5	32.0	44.4
	Percentage (%) of stations which attained 24-hour average EQS	8.3	29.4	38.2	13.3	28.8
	Annual-average concentration of PM$_{2.5}$ (μg/m^3)	17.2	16.1	15.4	16.0	15.5

Source: MOEJ 2015.

As seen in Figure 1 and Table 2, the annual average concentration range for the total average of all monitoring stations is slightly above the EQS value, and the attainment percentage of annual average EQS varies between 44-62% and 17-46% for non-roadside and roadside stations, respectively. Similarly, the attainment percentage for 24-hour average EQS varies between 16-45% and 8-38% for non-roadside and roadside stations, respectively. The average concentration of PM$_{2.5}$ does not reveal a clear trend in recent years but shows large inter-annual variability across the levels, where EQSs are exceeded at more than half of the monitoring stations.

2.2.2 Seasonal variation of mass concentration

Figure 2 shows the total number of days when the daily average concentration of PM$_{2.5}$ exceeded 35 μg m^{-3} at the non-roadside stations for each year between FY2011-2014 (MOEJ 2015).
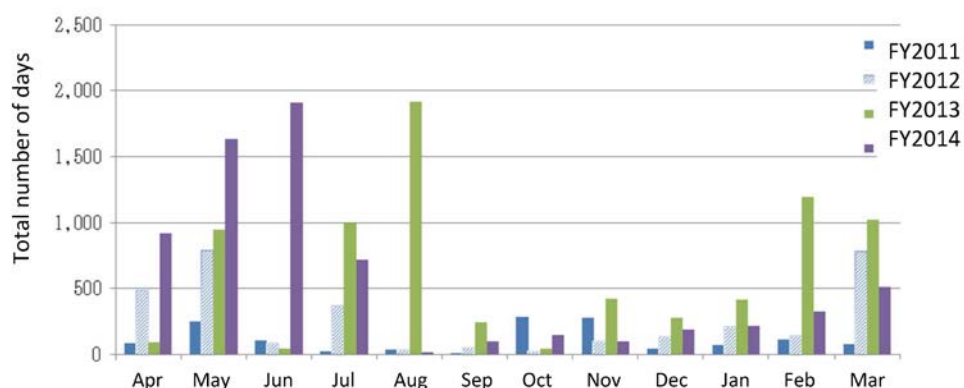

Figure 2. Total number of days for all stations, when the daily average concentration of PM$_{2.5}$ exceeded 35 μg m^{-3} at the non-roadside stations for the years 2011, 2012, and 2013
Source: MOEJ 2015.

As shown in Figure 2, although inter-annual variability is large, it can be noted that the PM$_{2.5}$ concentrations were high in July and August 2013. This can be ascribed to the photochemical formation of secondary aerosols since high photochemical ozone episodes were frequent during those months in 2013. The high PM$_{2.5}$ concentration was also observed in February 2014, due to meteorological conditions such as weak wind speed. Relatively high concentrations were generally observed between March and May.

PM$_{2.5}$ mass concentration was observed at two EANET (Acid Deposition Monitoring Network) monitoring sites that represent remote sites (free from local air pollution) in Japan: Rishiri, a northern island in Hokkaido, and Oki, an island in Shimane Prefecture in the southwestern part of the Sea of Japan. Figure 3 shows two five year averages for seasonal cycles of PM$_{2.5}$ mass concentration at these monitoring sites for FY2003-2007 and FY2008-2012.
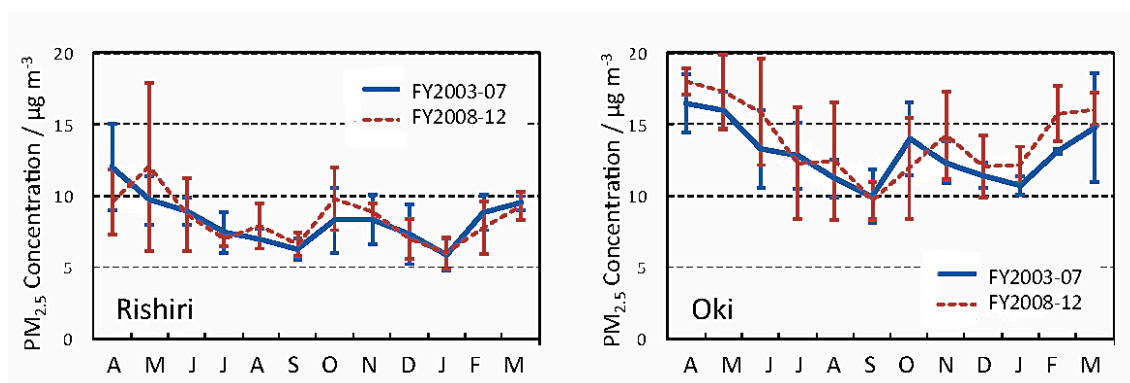

Figure 3. Seasonal variation of PM$_{2.5}$ mass concentration by month at Rishiri and Oki for FY2003-2007 and FY2008-2012

Source: MOEJ 2014.

As shown in Figure 3, at both the remote sites in Rishiri and in Oki, the mass concentration of PM$_{2.5}$ broadly plateaued in late winter/early summer (February-June) with a spring maximum between March and May. It can also be seen that the second maximum occurs in fall (October and November). The cause of seasonal PM$_{2.5}$ maximums at remote sites in Japan may be due to the long-range transport of air pollution and yellow sand from the Eurasian continent, and local primary and secondary production overlapping with the Eurasian background.

2.2.3 Chemical composition

Chemical compositions of PM$_{2.5}$ obtained at routine monitoring sites in 2012 are shown in Figure 4 together with the number of monitoring sites for each site categorization (MOEJ 2013)
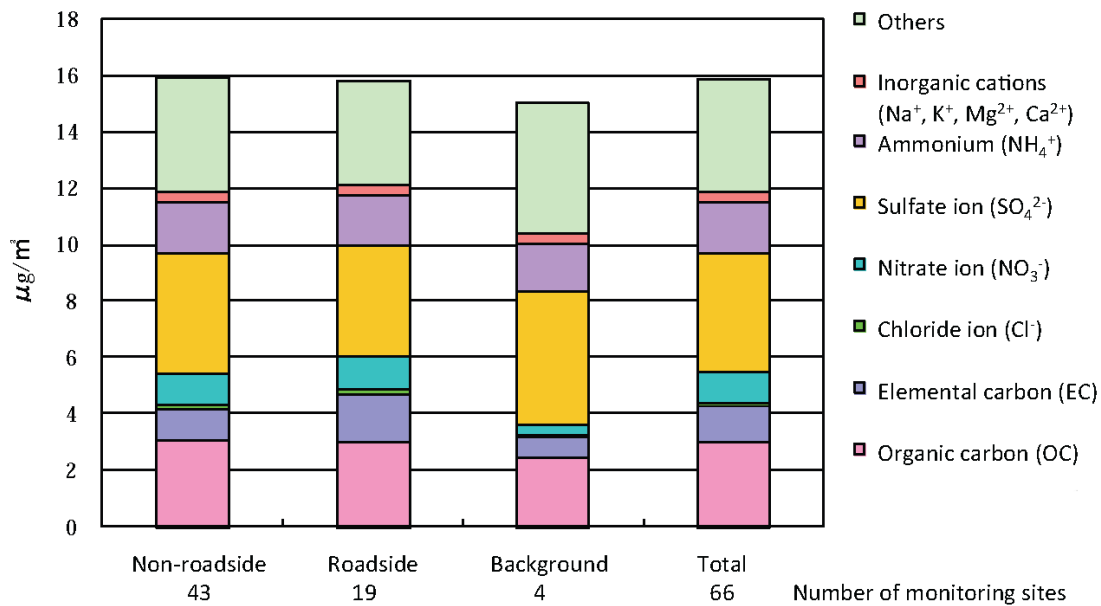
Figure 4. Chemical compositions of PM$_{2.5}$ observed nationwide at categorized monitoring sites in 2012

Source: MOEJ 2014.

For the general non-roadside stations, sulfate ion (SO$_4^{2-}$) and carbonaceous components (EC + OC) are the predominant components with ~25% of the total each, followed by nitrate ion (NO$_3^-$) with 7%. The percentages of EC and OC are 7 and 19%, respectively (the EC:OC ratio is ca. 1:3). At roadside stations, the percentage of EC is higher and those of sulfate and OC are slightly lower than at non-roadside stations. At the background sites, the percentage of sulfate is higher, ca. 32%, and nitrate ion (NO$_3^-$) is much lower than at sites in other categories. The ratio of EC + OC to the total is 21% close to the non-roadside stations and the EC is 5%. (EC:OC ratio is ca. 1:3 nearly the same as the non-roadside stations).

From these results it may be concluded that sulfate from SO$_2$ either from anthropogenic fossil fuel burning and volcanoes, organic carbon from anthropogenic and biogenic VOCs, and elemental carbon from diesel engine exhausts and biomass burning would be the major sources of PM$_{2.5}$. Here the biomass burning in Japan is mainly due to the agricultural waste burning, and forest fires and the domestic use of wood is negligible. The "others" in Figure 4 would include trace metals from combustion sources, and fine soil particles accompanying yellow sand.

2.2.4 Analysis of source apportionment

So far, the analysis of the source apportionment of $PM_{2.5}$ concentration in Japan has been scarce and only a few research papers have been published in scientific journals. Chatani et al. (2011) analyzed the contribution of sources to $PM_{2.5}$ in three large urban areas in Japan - Tokyo metropolitan, Aichi-Mie (including Nagoya), and Osaka-Hyogo (including Kobe) - by using the Community Multi-scale Air Quality (CMAQ), a regional chemical transport model. Comparison of the simulated values with the annual and monthly averaged observed mass concentrations of $PM_{2.5}$ in 2005 at four sites in Miyagi (northeastern prefecture), Hyogo, and Tokyo revealed that the model under-predicted the observation by 30-40% throughout the year. The reason has been ascribed to the under prediction of EC and OC in the model calculation.

Figure 5 shows the calculated annual mean source apportionment of $PM_{2.5}$ in 2005 in Osaka-Hyogo, Aichi-Mie, Tokyo metropolitan (Shuto), and the Tokyo 23-wards area. The sources are attributed to biogenic (Bio), volcano (Vol), ships (Shp), transboundary transport (Fm), biomass burning (Bb), automobile exhaust (Veh), road and tire wear dust (R&T), combustion other than automobiles (Cmb), volatile organic hydrocarbons (VOC) and ammonia (NH_3).
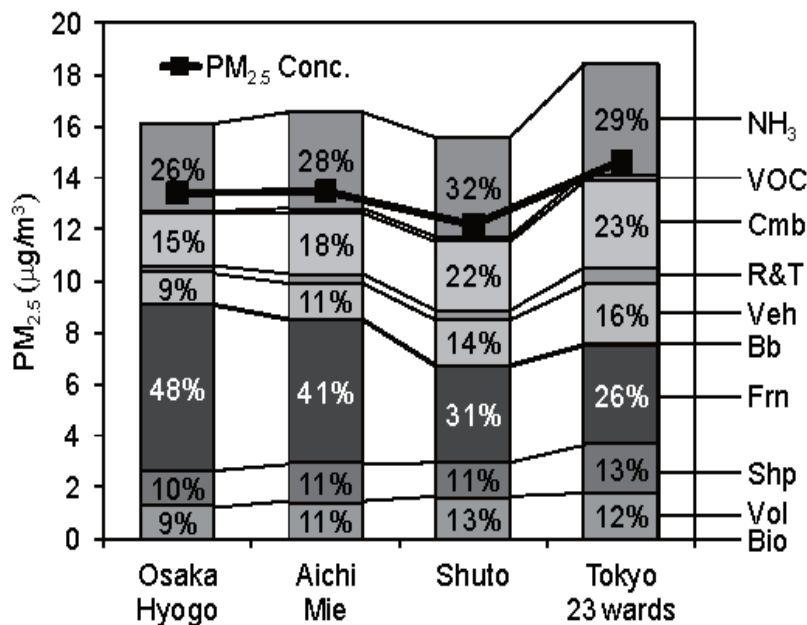

Figure 5. Source apportionment of $PM_{2.5}$ in Osaka-Hyogo, Aichi-Mie, Tokyo Metropolitan (Shuto) and the Tokyo 23-Wards in 2005.

Source: Chatani et al. 2011.

As noted above, OC that contains many of the secondary organic aerosols (SOA) formed by photochemical oxidation reactions of VOC in the atmosphere is substantially underestimated in this calculation; this implies that the contribution of VOC is also greatly underestimated in Figure 5. The percentage of transboundary transport of $PM_{2.5}$ is estimated at 48% in the Osaka-Hyogo area, decreases to the east, and accounted for 26 % in the Tokyo 23-wards.

In another piece of research, Ikeda et al. (2014) used the CMAQ (ver. 4.7.1) model to estimate that the contribution of $PM_{2.5}$ in 2010 originated from different areas in Northeast Asia namely, Fukue Is. (west of Kyushu), Oki Is. (southwestern Japan sea), Mt. Nonodake (Miyagi prefecture), and Rishiri Is. (northern Hokkaido). Figure 6 shows the relative contribution as annual averages from different source areas at the four sites. The observed annual average concentrations at these sites are 18.0, 12.0, 11.5, and 8.6 μg m^{-3} showing the decreasing trend from southwest to northeast. While the model was able to reproduce such spatial tendency, the absolute concentration was underestimated by 30-50%, the same as the study by Chatani et al. (2011). In the western parts of Japan, such as Fukue and Oki, the contribution from the central-north part of China (CHN-CN) is substantial, while in the eastern parts of Japan such as Nonodake, the domestic contribution is predominant; this qualitatively agrees with Chatani et al. (2011). It should be noted, however, that further refinement of the models is necessary in order to obtain more accurate quantitative apportionment of emission sources and originating areas.
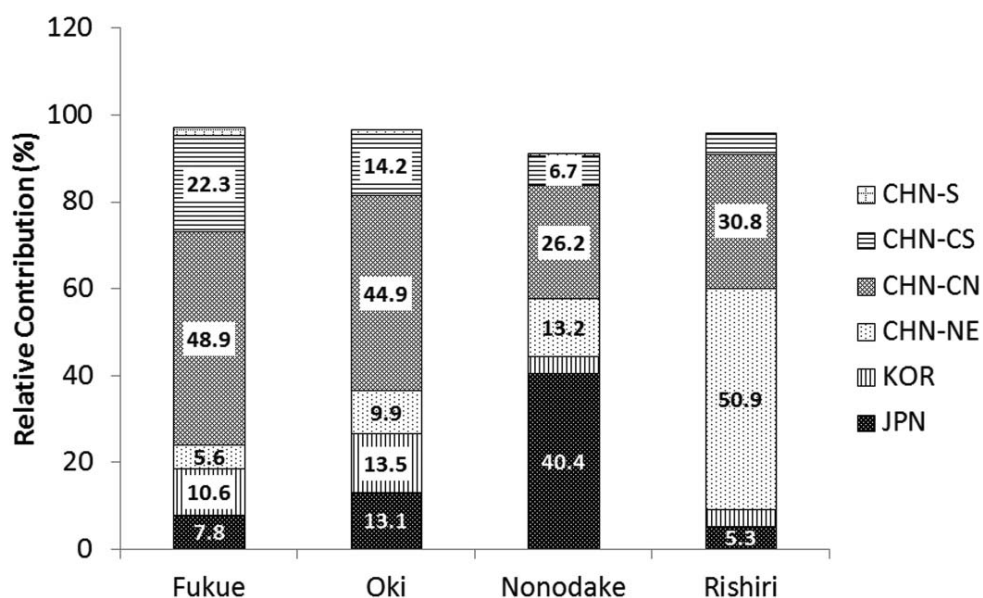

Figure 6. Model results of relative contributions from source regions to annual mean PM2.5 concentration at Fukue Is., Oki is., Mt. Nonodake, and Rishiri Is
Source: Ikeda et al. 2014.

3. Ozone (Oxidants)

3.1 Environmental Standards

Photochemical air pollution episodes in Japan were first publicized in 1970, when many sufferers, mostly school children and high school students, who were supposedly affected by oxidants, were hospitalized. This then gave rise to serious public concerns about photochemical oxidants (Kondo and Akimoto 1975). Environmental standards for photochemical oxidants were set by the MOEJ in 1973 and are shown in Table 3 which compares the AQSs of O_3 in other countries and the WHO guidelines. Together with the environmental standard for O_x (60 ppb 1-hour average), Japan set the warning and alert levels at 120 and 240 ppb, respectively.

Table 3. Comparison of air quality standards of O_3 (O_x): unit: ppbv and $\mu g\ m^{-3}$

Country and Organization	Species	Concentration ppbv	$\mu g\ m^{-3}$	Averaging Time	Notes
Japan	O_x	60	118	1 hour	Effective from 1973.
China (Class 1) (Class 2)	O_3	51 81	100 160	8 hours 8 hours	Effective from 2012. Daily 8-hr maximum. Class 2 standards apply to urban areas.
Korea	O_3	60	118	8 hours	
India	O_3	51	100	8 hours	
USA	O_3	70	138	8 hours	Effective from 2015. Fourth highest annual daily max. 8-hr averaged over 3 years.
EU	O_3	61	120	8 hours	Effective from 2010. Maximum daily 8-hr. mean. 25 days averaged over 3 years.
WHO Guideline	O_3	51	100	8 hours	Set in 2005

Source: Author.

It should be noted that in Table 3 the targeted species of the environmental standard for photochemical air pollution is O_x in Japan, whereas all other countries and organizations specify O_3 for AQSs. In the 1970s, widely-used monitoring instruments for photochemical air pollution were based on the potassium iodide (KI) method, a wet chemical method that responds to "oxidants", a collective term for chemical species which oxidize I^- ions into molecular I_2. The major components of oxidants in photochemical air pollution are O_3 (typically $\geq$ 90-95%), peroxy acetyl nitrate (PAN), hydrogen peroxide (H_2O_2), and organic hydroperoxides. This is the main reason why Japan established its environmental standards for "photochemical oxidants" rather than O_3. Since then, however, instruments responding specifically to O_3 based on UV absorption

have been developed and are universally used worldwide; accordingly, the index of photochemical air pollution has been switched from O_x to O_3 in almost all other countries.

　　　Another point to be noted in Table 3 is that the environmental standard of O_x in Japan has been set for a 1-hour averaged value, whereas the averaging time specified for the present prime AQS values for O_3 is 8 hours in most other countries and for the WHO. This is because epidemiological studies in the last few decades have revealed that 8-hour averaged O_3 concentrations have clear correlations with the impact to human health (WHO 2006).

　　　The present Japanese standard of O_x that has been unchanged since 1973 clearly does not reflect the more recent insight into the appropriateness of alternative O_x metrics described above. A review of the EQSs for photochemical oxidants is under discussion by a committee under the MOEJ.

3.2 Observational Data and Analysis

After the occurrence of the first episode of photochemical air pollution in 1970, monitoring of O_x and its precursors, NO_x (NO and NO_2) and NMHC was initiated, so that records covering more than 45 years are now available in Japan. Figure 7 shows the records of the annual-averaged concentrations of NO_x and NMHC in Japan since 1970 and 1976, respectively (MOEJ 2015a). Figure 8 shows the annual averaged concentration of O_x in Japan since 1976 (MOEJ 2015b).
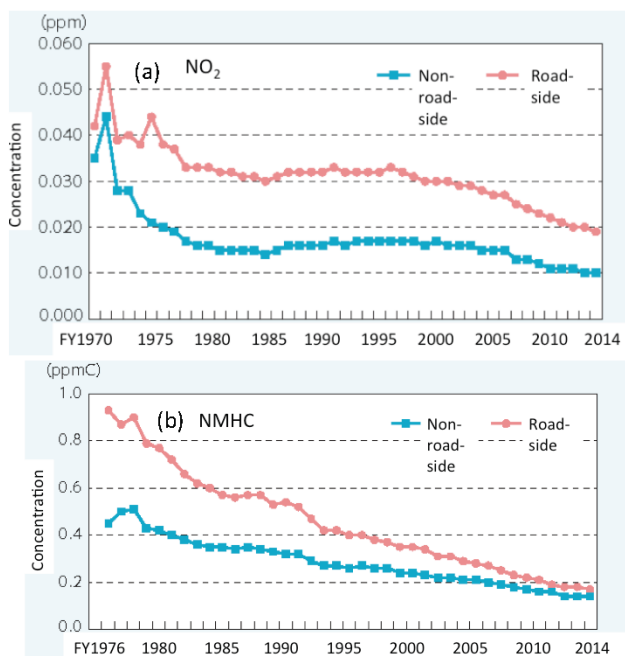

Figure 7. Variations of the annual average concentrations of (a) NO_2, and (b) NMHC (6-9 am) in Japan
Source: MOEJ 2015a.

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As can be clearly seen in Figure 7, historically the early 1970s were the era of serious air pollution in Japan. The most serious situation had been alleviated by the beginning of the 1980s owing to the introduction of what were at that time, the most stringent emission control policy measures for air pollutants in the world.[1]
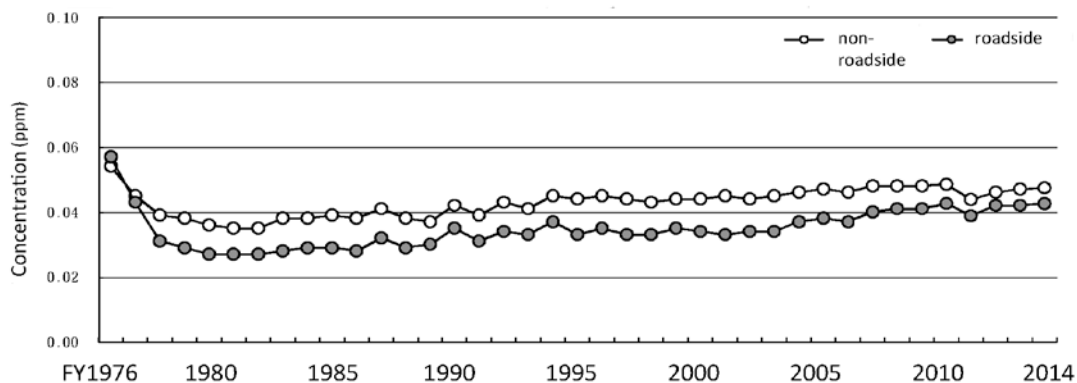

Figure 8. Variation of annual average concentration of O_x in Japan
Sourse: MOEJ 2015b.

Figure 8 shows that the high O_x levels in the 1970s decreased to the lowest values in the early 1980s. It should be noted, however, that the average concentration of O_x started to increase again in the 1990s and continued to increase in the 2000s. As shown in Figure 7 (a), the annual-averaged NO_2 concentrations stayed almost constant for nearly 20 years in the 1980s and 1990s despite the introduction of more and more stringent emission control measures particularly relating to gasoline-derived car exhaust gases. An unsuccessful effort to decrease ambient NO_2 concentrations may be partly due to the compensation by the increase of the number of automobiles and partly due to the titration effect by O_3 ($O_3 + NO \rightarrow NO_2 + O_2$), since NO_x and NO started to decrease slowly in the 1990s. On the other hand, the concentrations of NMHC have decreased monotonically since the 1980s, as can be seen in Figure 7 (b).

After the year 2000, emission control measures for diesel-powered trucks were tightened significantly, resulting in an apparent decrease in ambient NO_2 concentrations as shown in Figure 7. Further, new emission control measures for VOCs from fixed sources were introduced in 2006, which further decreased ambient NMHC (VOC) concentrations. Nevertheless, ambient average concentrations of O_x (O_3) were increasing in the 2000s, which caused serious concern for administration and industries for which VOC-emission regulations were required. In order to

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[1] The details of these control measures are not within the scope of this paper.

understand the contradiction, the Photochemical Oxidant Survey Committee was established under the MOEJ in 2011.

The committee report (Photochemical Oxidant Survey Committee 2014) revealed that the long-term trends of the "average" concentration of O_3 reflect three components: (1) the decrease of the NO titration effect (tends to increase O_3 in urban areas), (2) the increase of transboundary transport (tends to increase O_3 more in the western part of Japan), and (3) the decrease of in situ photochemical production (tends to decrease higher concentrations of O_3) (Akimoto et al. 2015). The suggestion was also made that a more statistically robust index for the evaluation of air quality of photochemical O_3, i.e., the daily maximum 8-hour average values after removing outliers, rather than total averages or 1-hour averaged maximum values, should be used (Ohizumi et al. 2013). In line with this suggestion, the MOEJ proposed the new index of "99-percentile value of daily maximum 8-hour average values in 3-year averages" for the purpose of evaluating the emission control effects of precursors of photochemical oxidants (MOEJ 2016b).

Figure 9 depicts the recent 23-year trends of O_x (O_3) in line with this new index in four typical urban areas in Japan (MOEJ 2015b). In reality, monitoring data in Japan relates to O_x, which is historically measured using the automatic wet chemical KI method; however, the instrument was gradually changed to the UV absorption method specific to O_3 after or around 2010. As shown in Figure 9, it is clearly seen that the daily 8-hour maximum concentrations of O_3 at the 99-percentile have been decreasing since the end of the 2000s. This reflects the decrease in ambient concentrations of NO_x and VOC shown in Figure 7 (MOEJ 2015a).
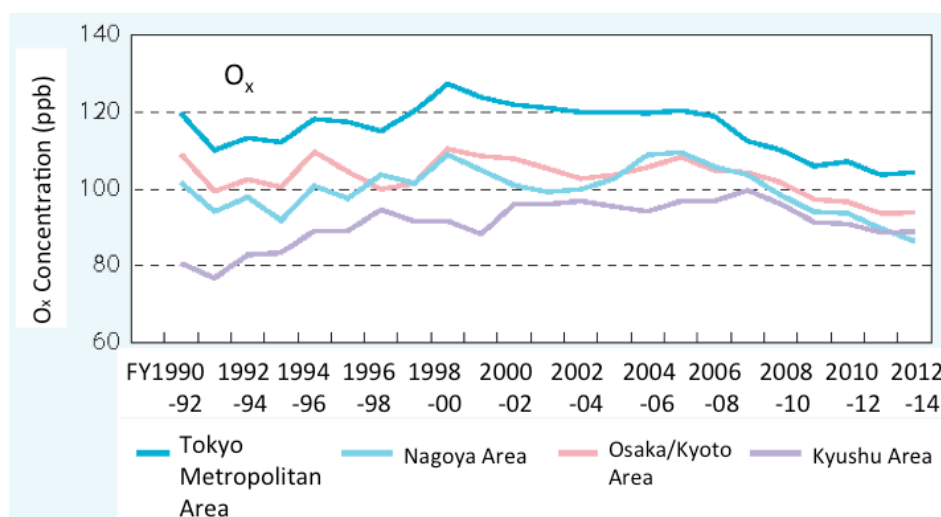

Figure 9. Recent 23 year trends for daily maximum 8-hour O_x (O_3) (99-percentile values) in 3-year averages for four different urban areas in Japan
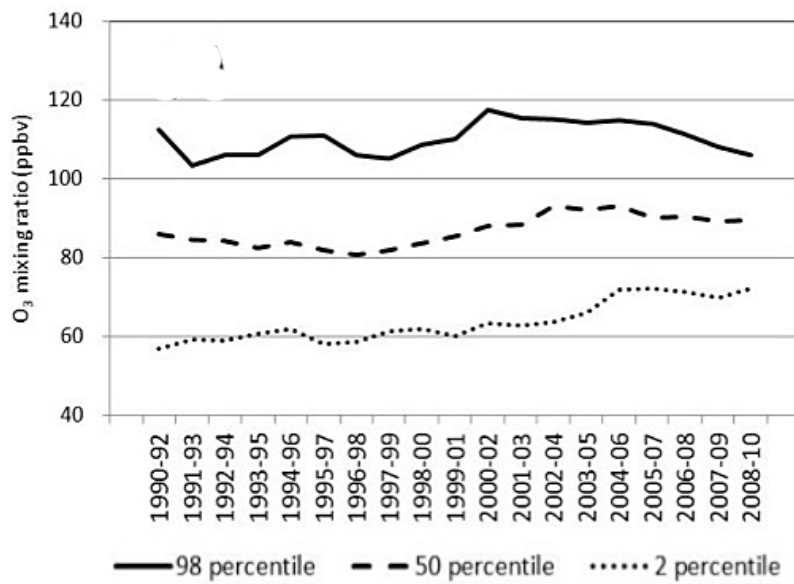
Sourse: MOEJ 2015b.

Figure 10. Trends of 3-year moving average of annual 98-, 50- and 2-percentile values of a daily maximum 8-h average O_3 mixing ratio in TMA
Sourse: Akimoto et al. 2015a.

Figure 10 shows the trends in 3-year moving averages for regional 98-, 50- and 2-percentile values of a daily maximum 8-hour average at each site in TMA (Akimoto et al. 2015a). The figure demonstrates that the trends are not uniform for different mixing ratio ranges, and the annual average at the 98 percentile exhibits a clearly decreasing trend in TMA over the last 10 years in accordance with Figure. 9. Although until recent years the increasing trend of 50- and 2-percentiles was more persistent, a leveling-off of the increasing trend is also discernible for the 50-percentile level. Thus, it can be concluded that that although the O_3 in the higher concentration range has decreased during this period due to the less active photochemical production in urban areas, the average concentration of O_3 still increases as a result of the increase of O_3 in the lower concentration range that arises from the decrease of NO titration effect and the possible increase of "background" O_3 (Akimoto et al. 2015a).

4. SLCPs co-control

Although most air pollutants, particularly the precursors of secondary $PM_{2.5}$ and O_3, are emitted from combustion sources together with CO_2, air pollution control and climate change mitigation have been approached separately in most countries, including Japan. Recently, however, co-controls of so-called short-lived climate pollutants (SLCPs) and CO_2 have been attracting much attention to the mitigation of near-term climate change and to the improvement of air

quality simultaneously in a more cost-effective way (UNEP/WMO 2011; UNEP 2011; Shindell et al. 2012). SLCPs are the air pollutants that have positive radiative forcing causing both global warming and degradation of air quality, and they include tropospheric ozone (O_3), methane (CH_4), and black carbon (BC) (UNEP/WMO 2011; Shindell et al. 2012). Tropospheric O_3 is a secondary pollutant produced by photochemical reactions from NO_x and VOCs in the atmosphere (Finlayson-Pitts and Pitts 2000; Akimoto 2016). BC is a constituent of $PM_{2.5}$ (together with organic carbon (OC), sulfate (SO_4^{2-}), and nitrate (NO_3^-)) which contributes to the adverse impacts on human health but also our ecosystems...

The discussion of SLCPs co-control is beyond the scope of this paper, however it should be remembered that further control of $PM_{2.5}$ and O_3 for air quality, particularly in Asia, has to be promoted in the context of the SLCPs co-control policy (Akimoto et al. 2015b).

5. Summary

As discussed above, $PM_{2.5}$ and O_3 pollution is very complex and deeply inter-related since the NO_x and VOCs are common precursors for O_3, nitrate (NO_3^-), and SOA, which are major components of $PM_{2.5}$. They are all produced by photochemical activities in polluted atmospheres and mitigation policies for $PM_{2.5}$ and O_3 pollution should be implemented simultaneously. Further, recent discussion on the co-control of SLCPs (short-lived climate pollutants) urges us to consider the co-control of air quality and climate change in a more cost-effective manner. Particularly in Asia, further discussions on SLCP co-control policy should consider more practical ways to mitigate $PM_{2.5}$ and O_3.

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Abstract (in Japanese)

要約

本論文では、最近の $PM_{2.5}$ および O_3（光化学オキシダント）に関する日本の状況について論じている。第 1 章では環境品質基準（EQS）の創設とその観測局を含む日本の $PM_{2.5}$ 政策措置の状況を紹介し、さらに、2010 年から 2013 年にかけて得られたモニタリングデータと EQS の超過状況について論じている。ここでは、全国平均の観測データにおいて、硫酸塩と EC/OC は $PM_{2.5}$ の卓越した成分であることが示され、ほぼ同等の影響を与えていることが判明している。第 2 章では $O_3(Ox)$ およびその前駆物質である NO_2 および NMHC/VOC（非メタン炭化水素/揮発性有機化合物）の長期変動を示した。観測上では NO_2 と VOC の濃度が低下しているにもかかわらず O_3 の平均濃度が増加しているパラドックスについて論じている。この現象は、（1）NO 滴定効果の減少、（2）越境輸送の増加 および（3）その場での光化学的生成の減少の 3 つの要因が見出された。 最後に、$PM_{2.5}$ および O_3 汚染の緩和措置を統合したアプローチは、SLCPs（short-lived climate pollutants）協調政策の枠組みの中で考慮すべきであると提案している。