

Article

Trends in PM_{2.5} Concentration in Nagoya, Japan, from 2003 to 2018 and Impacts of PM_{2.5} Countermeasures

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Abstract: In Japan, various countermeasures have been undertaken to reduce the atmospheric concentration of fine particulate matter (PM_{2.5}). We evaluated the extent to which these countermeasures were effective in reducing PM_{2.5} concentrations by analyzing the long-term concentration trends of the major components of PM_{2.5} and their emissions in Nagoya City. PM_{2.5} concentrations decreased by 53% over the 16-year period from fiscal years 2003 to 2018 in Nagoya City. Elemental carbon (EC) was the component of PM_{2.5} with the greatest decrease in concentration over the 16 years, decreasing by 4.3 µg/m³, followed by SO₄²⁻ (3.0 µg/m³), organic carbon (OC) (2.0 µg/m³), NH₄⁺ (1.6 µg/m³), and NO₃⁻ (1.3 µg/m³). The decrease in EC concentration was found to be caused largely by the effect of diesel emission control. OC concentrations decreased because of the effects of volatile organic compound (VOC) emission regulations for stationary sources and reductions in VOCs emitted by vehicles and construction machinery. NO₃⁻ concentrations decreased alongside decreased contributions from vehicles, construction machinery, and stationary sources, in descending order of the magnitude of decrease. Although these findings identify some source control measures that have been effective in reducing PM_{2.5}, they also reveal the ineffectiveness of some recent countermeasures for various components, such as those targeting OC concentrations.

Keywords: PM_{2.5}; elemental carbon; organic carbon; urban air pollution; long-term trend; pollution countermeasure



Citation: Yamagami, M.; Ikemori, F.; Nakashima, H.; Hisatsune, K.; Ueda, K.; Wakamatsu, S.; Osada, K. Trends in PM_{2.5} Concentration in Nagoya, Japan, from 2003 to 2018 and Impacts of PM_{2.5} Countermeasures. *Atmosphere* **2021**, *12*, 590. <https://doi.org/10.3390/atmos12050590>

Academic Editor: Ilias Kavouras

Received: 29 March 2021

Accepted: 22 April 2021

Published: 1 May 2021

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1. Introduction

Atmospheric aerosol particles are solid particles or liquid droplets of various sizes suspended in the air. Among aerosol particles, fine particles can be inhaled by the human body [1]; fine particles less than 2.5 µm in diameter are defined collectively as PM_{2.5}. PM_{2.5} can reach deep into the lungs and is known to cause health problems [2]. In Japan, the atmospheric PM_{2.5} concentration has a strong effect on mortality [3,4]. PM_{2.5} is composed of a variety of chemicals, including carbonaceous components, such as elemental carbon (EC) and organic carbon (OC), and inorganic components, such as sulfate and nitrate as major components. EC is produced by the incomplete combustion of fossil fuels, such as oil and coal, and of biomass, such as wood. OC is produced by the combustion of fossil fuels and biomass and by the photooxidation of volatile organic compounds (VOCs) in the atmosphere. These include anthropogenic VOCs used in paints, printing inks, adhesives, and cleaning agents, as well as biogenic VOCs, such as isoprene released from plant leaves. Sulfate is formed by the oxidation of sulfur dioxide (SO₂), which is produced by the combustion of fossil fuels; some SO₂ is also released from volcanoes. Nitrate is formed by the oxidation of nitrogen oxides (NO_x), which are produced by the combustion of fuels. To

reduce the atmospheric PM_{2.5} concentration, various regulations are needed to decrease emissions of particles and their precursor gases.

Various countermeasures for pollutants in Japan are described in Wakamatsu [5]. The major source of EC in Japan was reported to be diesel exhaust [6]. The 1993 regulations introduced by the Japanese government set an upper limit on particle emissions from new diesel vehicles, and they have lowered the limit gradually since then [7]. For stationary sources that emit VOCs into the atmosphere, emission regulations were initiated in 2006 to set the maximum VOC concentrations at emission outlets for each type of facility. For stationary sources of SO₂, SO₂ emission limits have been set for each facility. The sulfur content of diesel oil and gasoline used in automobiles has been reduced gradually. For stationary sources of NO_x, emission concentration limits for each type of facility and total limits in areas of significant pollution have been implemented. The upper limit of NO_x concentration in automobile exhaust was specified in 1973 and has been reduced stepwise since then [7]. For machinery used off the road, such as forklifts for industrial use, tractors for agricultural use, and bulldozers and hydraulic excavators for construction (hereinafter collectively called "construction machinery, etc."), the upper limit for the NO_x concentration in exhaust gas was set in 2006, and the limit was lowered in 2014. In this way, various source control measures have been implemented in Japan.

The annual mean atmospheric concentration of PM_{2.5} in Tokyo exceeded 25 µg/m³ in FY 2001 but decreased to approximately 15 µg/m³ in FY 2012 [5]. In Nagoya City, PM_{2.5} concentrations also showed a decreasing trend from FY 2003 to FY 2007, reflecting the effects of vehicle emission regulations [8]. Since then, the atmospheric concentration of PM_{2.5} in Japan has continued to decrease. On the other hand, PM_{2.5} concentrations in Japan are known to be affected by transboundary pollution from the Asian continent [9], and the reduction in Japanese emissions may therefore not be fully reflected in PM_{2.5} concentrations in Japan. Therefore, it is important to evaluate the extent to which the various source control measures in Japan have contributed to the observed reduction in PM_{2.5} concentration. To evaluate the effectiveness of each source measure, it is necessary to examine each component of PM_{2.5}.

Nagoya, located in the middle of Honshu Island in Japan (Figure 1), is the third-largest metropolitan area in Japan. Nagoya City, with a population of approximately 2.3 million, covers approximately 326 km². The Nagoya area, including Nagoya City, is a region with a thriving manufacturing industry. In addition, roads with heavy automobile traffic are located in Nagoya City, and the area has been designated as a countermeasure area under the Automobile NO_x/PM Law, which designates areas with high levels of NO_x and particulate matter emitted from automobiles. We collected daily PM_{2.5} and measured the major components of PM_{2.5} in Nagoya City from FY 2003 to FY 2018. We estimated data for various emission sources of PM_{2.5} in Nagoya City during this period. This study revealed the long-term trends in the PM_{2.5} concentration and its major components in Nagoya over the 16-year period, while identifying the major components that contributed to the long-term decrease in PM_{2.5} concentration. For each major component, the relationship between its atmospheric concentration and its emissions or precursor gas emissions is examined herein, and the effects of various source control measures are discussed.

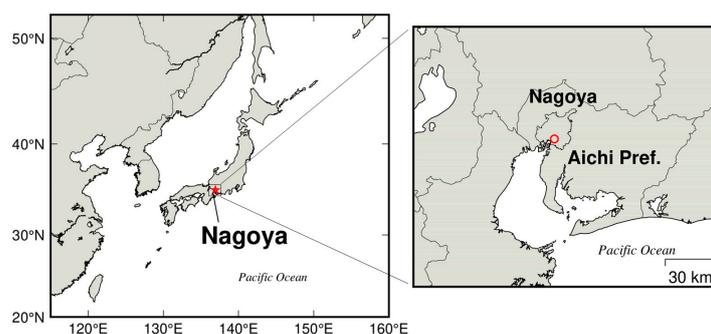


Figure 1. Location map of the PM_{2.5} observation site. Nagoya City Institute for Environmental Sciences (NCIES) at Minami-ku, Nagoya City.

2. Methods

2.1. Sampling

PM_{2.5} samples were collected daily on the roof of the Nagoya City Institute for Environmental Sciences (NCIES; 0.8 m asl, 35.10° N, 136.92° E) located in southern Nagoya City, Aichi Prefecture, central Japan (Figure 1). The sampling site was surrounded by a busy industrial area and a residential area. One possible strong EC source, a busy road (national route 23), was located 110 m south of the NCIES. These aspects and components of the surrounding environment did not change during the study period.

We collected PM_{2.5} samples through an EPA well-type impactor ninety-six (WINS; [10]) inlet at a flow rate of 16.7 L/min, using Partisol Model 2000 (Thermo Scientific, Waltham, MA, U.S.) from April 2003 to March 2014, and Partisol Model 2025 (Thermo Scientific, Waltham, MA, U.S.) from April 2014 to March 2019. From April 2003 to May 2013, aerosol sampling began at 09:30 each day and ran for 23.5 h per day. From June 2013 to March 2019, aerosol sampling began at 10:00 and ran for 23.5 or 24 h per day. From FY 2003 to FY 2013, the number of days sampled per year ranged from 184 to 337, sampling from Sunday morning to Saturday morning. From FY 2014 to FY 2018, the number of days sampled per year was about 360.

PM_{2.5} samples were collected on a PTFE filter (Teflo, 47 mm diameter, 2 µm pore size, R2PJ047; Pall Corp., New York, U.S. or 46.2 mm diameter, 2 µm pore size, 7592-104; Whatman, Maidstone, U.K.) and a quartz-fiber filter (47 mm diameter, 2500 QAT-UP; Pall Corp., New York, U.S.). PTFE filters were used for analyzing mass concentrations of PM_{2.5} and major ions. The PTFE filters were weighed before and after sampling using a microbalance (ME5-F; Sartorius AG, Göttingen, Germany) under constant temperature (21.5 °C ± 1.5 °C) and relative humidity (35% ± 5%) for 24 h. Quartz-fiber filters were used for analyzing EC and OC.

2.2. Chemical Analyses

The procedure for analyzing PM_{2.5} was the same as that used in Yamagami et al. [11]. After weighing for the PM_{2.5} mass concentration, the PTFE filters were cut into halves for additional chemical analyses. One half was placed in a pre-cleaned polypropylene tube (15 mL; Iwaki Glass Co. Ltd., Shizuoka, Japan) with 10 mL ultrapure water (18.2 MΩ, PURELAB Ultra, ELGA/ORGANO, High Wycombe, U.K.) for 20 min in an ultrasonic bath. After the extract was filtered using a 0.2-µm pore size PTFE filter (DISMIC-25HP; Advantec, Tokyo, Japan), it was analyzed for the major ions (Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺, and Ca²⁺) using an ion chromatograph (ICS-1000; Dionex Corp., Sunnyvale, CA, U.S.). Commercially available standard solutions were used for ionic analyses (Anion Mixture Standard Solution 1 and Multication Standard Solution; Wako Pure Chemical Industries Ltd., Osaka, Japan). EC and OC concentrations were measured in terms of thermal optical reflectance following the IMPROVE protocol [12] using a carbon analyzer (Sunset Laboratory Inc., New York, NY, USA). The OC fractions were measured at four temperature steps in 100% helium: OC1 at 120 °C, OC2 at 250 °C, OC3 at 450 °C, and OC4 at 550 °C. The EC fractions were measured at three temperature steps in a mixture of 2% oxygen and 98% helium: EC1 at 550 °C, EC2 at 700 °C, and EC3 at 850 °C. The carbon evolved at each temperature was oxidized to carbon dioxide (CO₂). The CO₂ was then reduced to methane (CH₄); it was then quantified using a flame ionization detector.

2.3. Emission Data

For some sources, emissions were estimated every few years. For FYs 2003, 2004, and 2006–2011, for which no aggregated data are available, emissions in FYs 2001 [13], 2005 [14], and 2012 [15] were supplemented by linear regression and calculated by scaling the estimate according to the respective parameters described below. Data for FYs 2013–2018, for which no aggregated data are available, were based on the projected emissions for FY 2018, which were calculated by scaling the estimate according to the respective parameters and aggregated.

2.3.1. Mobile Sources

EC

The method used to calculate EC emissions from vehicles is described in detail by Yamagami et al [11]. Annual particulate matter emissions from vehicles in the Nagoya urban area were reported by Ministry of Environment, Japan (MOE) [16], based on diesel traffic data and particle emission factors. Traffic data for diesel vehicles were recorded based on the vehicle type, holiday and weekdays, and time zones. Vehicles were classified as cars, buses, light-duty diesel trucks, medium-duty diesel trucks, heavy-duty diesel trucks, or “construction machinery, etc.” Note that these emissions were calculated assuming zero particulate matter emissions from gasoline vehicles. According to a report from MOE [17], the ratio of EC in particles emitted from diesel vehicles was 70%. Annual EC emissions in the Nagoya urban area were calculated by multiplying the particle emissions from vehicles by 0.7.

VOC

VOC emissions from fuel evaporation from vehicles in Nagoya City were calculated based on emission factors and traffic volumes [13–15]. For the years for which no data were collected, emissions were calculated by scaling the estimate according to the vehicle traffic [18]. VOC emissions from construction machinery, etc., were calculated based on the total VOC emissions of construction and industrial machinery in Japan via proportional allocation to the city of Nagoya according to the construction cost and the value of manufactured goods shipped [13–15]. VOC emissions from agricultural machinery were less than 1 ton (t) per year, so they were not included in the total. Emissions for years with no data were calculated by scaling the estimate according to energy consumption by industry [19].

SO₂

SO₂ emissions from vehicles in Nagoya City were calculated based on the sulfur content of diesel and gasoline, fuel consumption by vehicle type, and traffic volume [15]. Emissions for years for which no data were available were calculated by scaling the estimate according to the sales volume of diesel and gasoline in Aichi Prefecture [20,21] and the sulfur content in diesel and gasoline at the time.

NO_x

NO_x emissions from vehicles in Nagoya City were calculated based on emission factors and traffic volumes by vehicle type [13–15]. Emissions for years for which no data were available were calculated by scaling the estimate according to the traffic volumes [18]. NO_x emissions from construction machinery, etc., were calculated based on the total estimated NO_x emissions in Japan via proportional allocation to Nagoya City [13–15]. Emissions for years with no available data were calculated by scaling the estimate according to the industrial energy consumption [19].

2.3.2. Stationary Sources

VOC

VOC emissions from stationary sources were compiled from data in the Pollutant Release and Transfer Register [22]. The emissions of the top 20 substances with the largest emission masses into the atmosphere in Nagoya City from FYs 2003 to 2018 were tabulated by industry. These 20 substances were acrylonitrile, ethylbenzene, ethylene glycol, ethylene glycol monoethyl ether, propylene oxide, xylene, chloroform, ethylene glycol monoethyl ether acetate, vinyl acetate, dichlorobenzene, methylene chloride, tetrachloroethylene, normal-dodecyl alcohol, trichloroethylene, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene, toluene, normal-hexane, benzene, and methyl methacrylate. The emissions of these 20 substances accounted for 97–99% of the total VOC emissions from stationary sources in each year.

SO₂

SO₂ emissions from large-scale industrial facilities in Nagoya City were calculated annually based on flue gas SO₂ concentrations and either flue gas volume or fuel consumption [20,21,23–36]. SO₂ emissions from small-scale industrial facilities were calculated based on the fuel consumption of tertiary industries and sulfur content by fuel type [13–15]. Emissions for years with no data were calculated by scaling the estimate according to the population of Nagoya City at the time [37]. SO₂ emissions from households were calculated based on kerosene consumption and the sulfur content in kerosene [13–15]. Emissions for years with no data were calculated by scaling the estimate according to the number of households in Nagoya City at the time [37].

NO_x

NO_x emissions from large-scale industrial facilities in Nagoya City were calculated annually based on flue gas NO_x concentrations and flue gas volumes [20,21,23–36]. NO_x emissions from small-scale industrial facilities were calculated based on the fuel consumption of tertiary industries and NO_x emission factors by fuel type [13–15]. Emissions in years for which no data were available were calculated by scaling the estimate according to the population of Nagoya City at the time [37]. NO_x emissions from households were calculated based on the fuel consumption of households and NO_x emission factors for each fuel type [13–15]. Emissions for years with no data were calculated by scaling the estimate according to the number of households in Nagoya City at the time [37].

2.4. Backward Trajectory Analysis

We calculated the backward trajectories using the hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model version 4 [38]. The trajectories were calculated using the model vertical velocity, and the trajectory duration was 5 days. The starting height of the trajectories was set at 1500 m above ground level over NCIES, and the starting time was set at 0300 UTC (1200 JST).

3. Results and Discussion

3.1. Long-Term Trends in PM_{2.5} Concentration and Chemical Composition

Figure 2a presents the proportion of the daily average wind velocity at the local meteorological observatory in Nagoya during the sample collection of PM_{2.5}, separated into five categories from FYs 2003 to 2018. No significant change was present in wind velocity over the 16-year period; however, the proportion of low wind velocity (< 2 m/s) was lower in FYs 2004, 2012, and 2013. Because low wind velocity increases PM_{2.5} concentrations, it is possible that PM_{2.5} concentrations were lower in these years. Figure 2b presents the proportion of backward trajectories over the Asian continent and over the ocean and Japan from FYs 2005 to 2018. In FYs 2009 and 2013, the proportion over the Asian continent was high. Therefore, it is possible that long-distance transport had a significant impact in these years.

Figure 2c shows the annual mean concentrations of PM_{2.5} and of the major components of PM_{2.5} (EC, OC, NH₄⁺, NO₃[−], SO₄^{2−}) from FYs 2003 to 2018 at the NCIES. PM_{2.5} concentrations decreased at an annual rate of −3.5% from FYs 2003 to 2018. The annual mean PM_{2.5} concentration decreased from 25.9 μg/m³ in FY 2003 to 12.3 μg/m³ in FY 2018, a decrease of 13.6 μg/m³ (53%) over 16 years. Five chemical components accounted for 89% of this 16-year decrease in PM_{2.5} concentration: EC (4.3 μg/m³ decrease), SO₄^{2−} (3.0 μg/m³ decrease), OC (2.0 μg/m³ decrease), NH₄⁺ (1.6 μg/m³ decrease), and NO₃[−] (1.3 μg/m³ decrease).

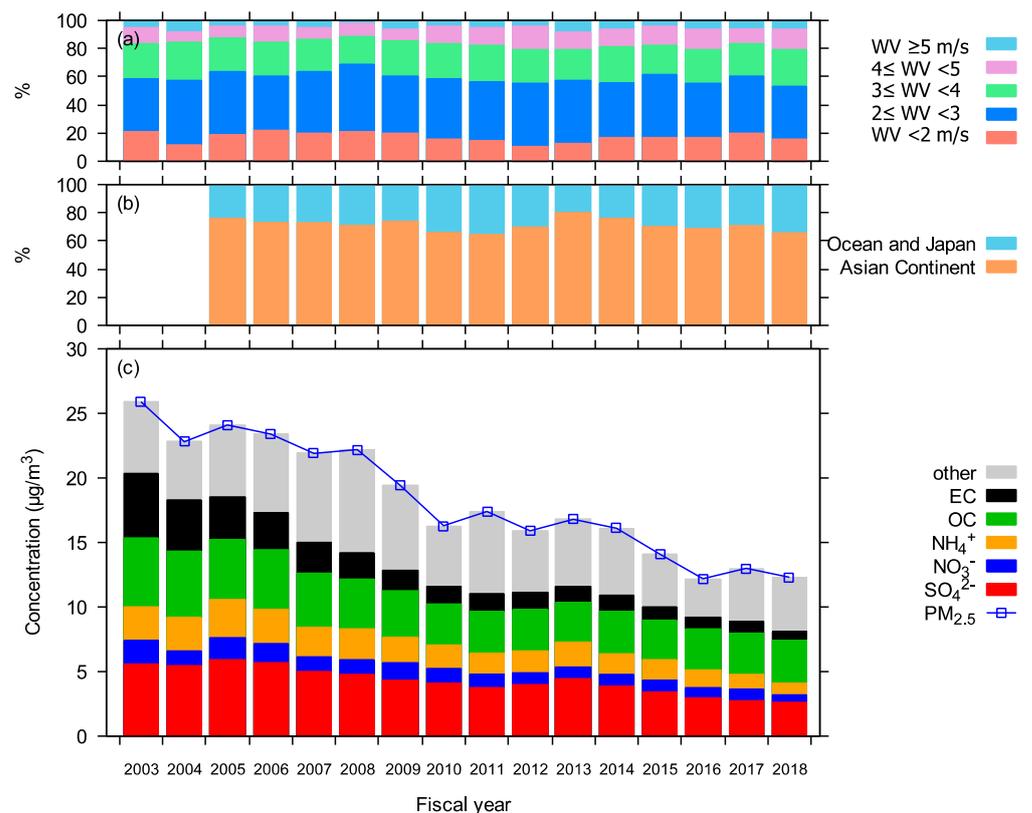


Figure 2. (a) Proportion of daily average wind velocity (WV) at the local meteorological observatory in Nagoya during the sample collection of PM_{2.5} from fiscal years (FYs) 2003 to 2018. (b) Proportion of backward trajectories over the Asian continent and over the ocean and Japan from FYs 2005 to 2018. (c) Annual mean concentrations of PM_{2.5} and of SO₄²⁻, NO₃⁻, NH₄⁺, OC, and EC in PM_{2.5} at the Nagoya City Institute of Environmental Sciences from FYs 2003 to 2018.

3.2. Elemental Carbon in PM_{2.5}

Figure 3a displays the annual and seasonal mean concentrations of EC in PM_{2.5} between FYs 2003 and 2018 at the NCIES. EC was the PM_{2.5} component that showed the greatest reduction in concentration over the 16-year study period, accounting for 31% of the 13.6 μg/m³ long-term PM_{2.5} concentration reduction. EC decreased from an annual mean concentration of 5.0 μg/m³ in FY 2003 to 0.7 μg/m³ in FY 2018, a decrease of 4.3 μg/m³ (86%), or 4.8%/year on average. The annual mean concentration of EC decreased monotonically during this period. EC concentrations decreased considerably through FY 2010 and more moderately after FY 2010.

Figure 3b shows EC emissions from diesel vehicles by type, estimated for the Nagoya urban area. Annual EC emissions decreased from 1600 t in FY 2003 to 184 t in FY 2018, a decrease of 88% over 16 years, that is, 6.0%/year on average. The declines in EC concentration and EC emissions over the 16-year period were about the same. EC emissions decreased considerably through FY 2010 and more moderately after FY 2010. The vehicle type with the highest EC emissions was heavy-duty diesel trucks, which accounted for a fairly constant proportion of total EC emissions over the 16-year study period: 76% in FY 2003 and 73% in FY 2018. EC emissions from cars accounted for 6% of total emissions in FY 2003, but dropped to 1% by FY 2018.

Figure 4 presents the relationship between annual EC emissions from diesel vehicles and the annual mean EC concentration. A linear regression analysis was conducted with the annual mean concentration as the dependent variable and emissions as the independent variable. These parameters were extremely strongly correlated ($r = 0.97$) over the period from FYs 2003 to 2018. These high correlations suggest that regulations on diesel vehicle

emissions were effective in reducing EC concentrations. The intercept of the regression line is interpreted to represent the EC concentration from sources other than diesel vehicles. Sources other than diesel vehicles include biomass combustion; combustion of fossil fuels, such as stationary sources and ships; and transboundary pollution. The contribution to EC concentration of these sources was estimated to be $0.24 \pm 0.13 \mu\text{g}/\text{m}^3$, based on the intercept value. The impact of these sources was relatively small when EC concentrations were high. However, in 2018, when EC concentrations were low, this “other sources” value corresponded to one third of the annual mean EC concentration of $0.69 \mu\text{g}/\text{m}^3$; such EC sources are becoming increasingly influential. In recent years, the influence of sources other than diesel vehicles has begun to be observed at NCIES, as EC concentrations have been observed to increase depending on the wind direction, because of the combustion of heavy oil from ships and stationary sources in the bay [39].

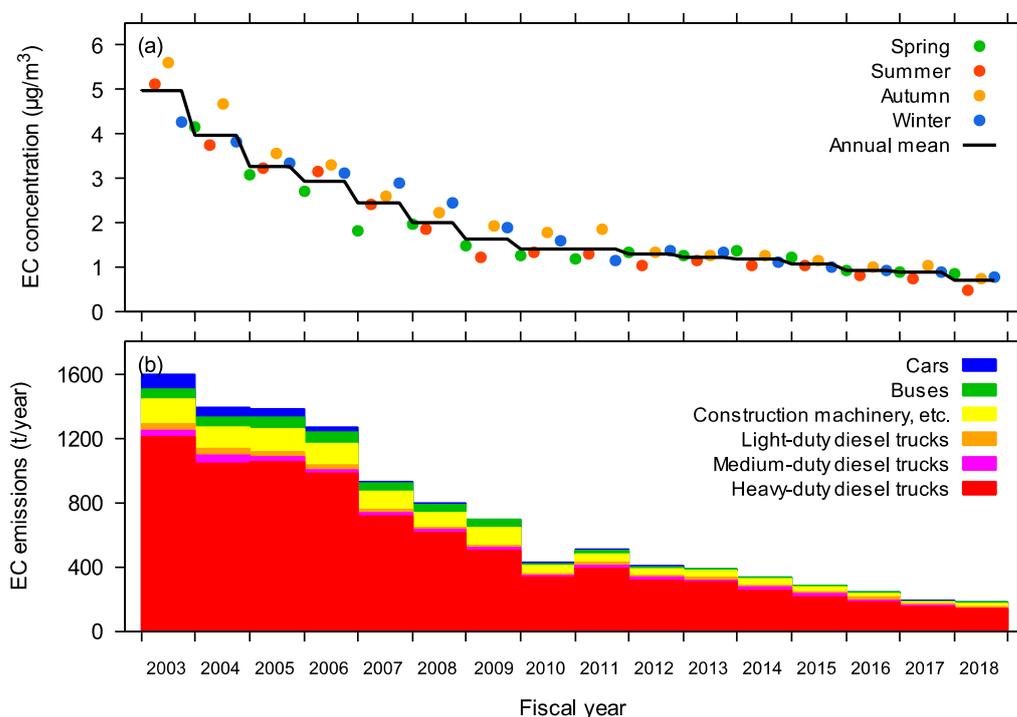


Figure 3. (a) Annual and seasonal mean concentrations of elemental carbon (EC) in $\text{PM}_{2.5}$ at the Nagoya City Institute for Environmental Sciences in Nagoya from fiscal years 2003 to 2018. (b) EC emissions from diesel vehicles by type in the Nagoya urban area from fiscal years 2003 to 2018.

3.3. Organic Carbon in $\text{PM}_{2.5}$

Figure 5a displays the annual and seasonal mean concentrations of OC in $\text{PM}_{2.5}$ between FYs 2003 and 2018 at the NCIES. Of the $13.6 \mu\text{g}/\text{m}^3$ reduction in $\text{PM}_{2.5}$ over 16 years, the contribution of OC to this reduction was 15%. OC decreased from an annual mean concentration of $5.3 \mu\text{g}/\text{m}^3$ in FY 2003 to $3.3 \mu\text{g}/\text{m}^3$ in FY 2018, a decrease of $2.0 \mu\text{g}/\text{m}^3$ (38%), or $2.6\%/$ year on average. After FY 2010, the OC concentration remained unchanged at about $3.2 \mu\text{g}/\text{m}^3$.

Figure 5b shows VOC emissions in Nagoya City. The annual VOC emissions decreased from 5185 t in FY 2003 to 1523 t in FY 2018, a decrease of 3663 t (71%) over 16 years. The decrease in OC concentration (38%) was smaller than this 71% decrease in VOC emissions. VOC emissions decreased significantly from FYs 2003 to 2009, then remained stable from FYs 2009 to 2014, and finally decreased again starting in FY 2015. VOC emissions from vehicles decreased by 1549 t (84%) over 16 years, from 1851 t in FY 2003 to 301 t in FY 2018. This reduction was almost the same as that of EC emissions. VOC emissions from vehicles accounted for 36% of total emissions in FY 2003, but only 20% in FY 2018. VOC emissions from construction machinery, etc., decreased by 183 t (42%) over 16 years. The regulation of

VOC emissions from stationary sources began in 2006, with the goal of reducing emissions by 30% from the FY 2000 level by FY 2010. VOC emissions from stationary sources decreased steadily until FY 2010. After that, emissions from stationary sources increased slightly, peaked in FY 2014, and then continued to decrease. VOC emissions from stationary sources decreased from 2898 t in FY 2003 to 967 t in FY 2018, a decrease of 1931 t (67%) over 16 years. The top five emitters were the manufacture of ceramic, stone, and clay products; transportation equipment; fabricated metal products; non-ferrous metals and products; and food. Together, these five emitters accounted for 70–80% of stationary source emissions. VOC emissions from the manufacture of ceramic, stone, and clay products accounted for 31% (1598 t) of total emissions in FY 2003, but decreased year by year to less than 1% (10 t) in FY 2018. On the other hand, VOC emissions from the manufacture of food increased from FY 2010, accounted for 22% (330 t) of total emissions in FY 2018. In sum, the industry-by-industry proportions of emissions from stationary sources changed significantly during the 16-year study period.

Figure 6 presents the relationship between the annual VOC emissions and the annual mean OC concentration. These parameters were extremely strongly correlated ($r = 0.97$) over the period from FYs 2003 to 2018. However, the OC concentration did not decrease after FY 2016 even as VOC emissions decreased. The VOCs that decreased after FY 2016 were found not to contribute to OC production. The intercept of the regression line from FYs 2003 to 2018 is $1.9 \pm 0.14 \mu\text{g}/\text{m}^3$, which corresponds to 58% of the FY 2018 OC concentration of $3.3 \mu\text{g}/\text{m}^3$. This finding indicates that the contribution from sources other than anthropogenic VOC sources was 58%. One such “other source” is biomass burning. The contribution of biomass burning to OC was 40% in autumn in Nagoya City [40]. Other sources include anthropogenic primary OC, condensable particle matter from stationary sources, OC oxidized from biogenic VOCs and unidentified anthropogenic VOCs, and long-range transport. Biogenic VOC emissions in Japan were estimated to be 1303 Gg/year in 2016 [41], which was higher than anthropogenic VOC emissions (671 Gg/year) [42]. To further reduce OC concentrations, these source contributions need to be clarified.

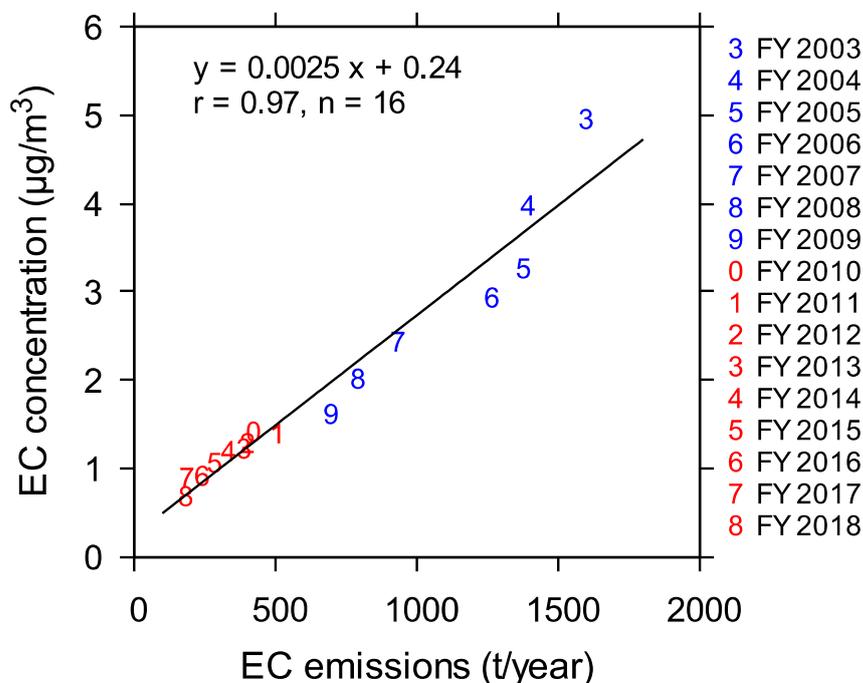


Figure 4. Scatter plot of annual EC emissions from diesel vehicles in the Nagoya urban area versus the annual mean concentration of EC in $\text{PM}_{2.5}$ at the Nagoya City Institute for Environmental Sciences in Nagoya from FYs 2003 to 2018. The black line shows the results of linear fitting for the period from FYs 2003 to 2018.

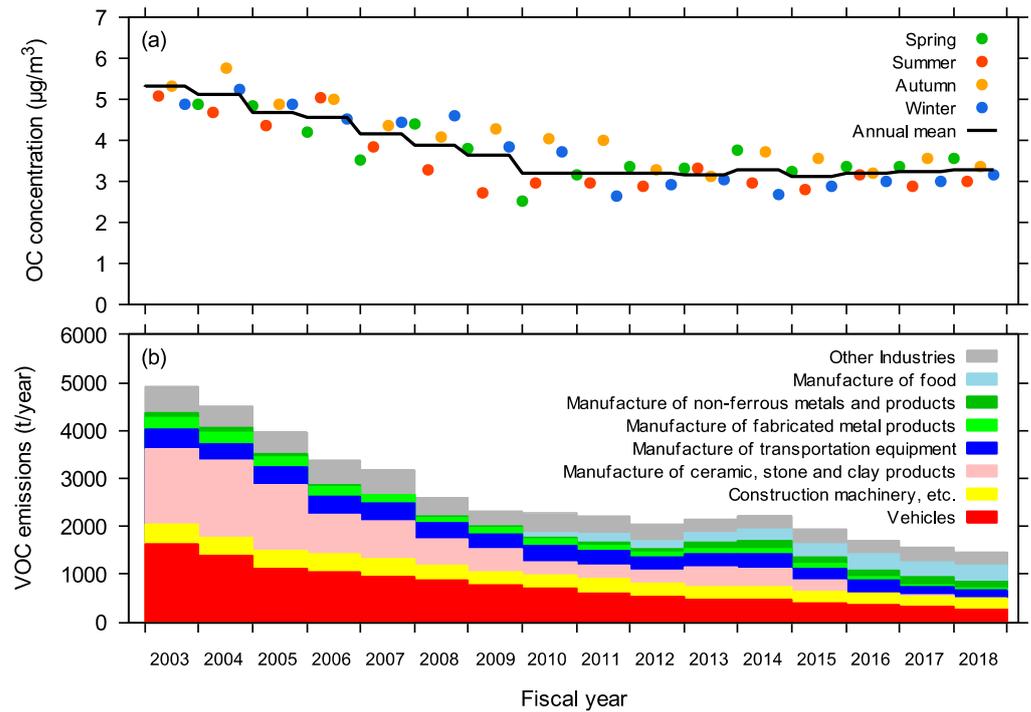


Figure 5. (a) Annual and seasonal mean concentrations of organic carbon (OC) in PM_{2.5} at the Nagoya City Institute for Environmental Sciences in Nagoya from fiscal years 2003 to 2018. (b) Volatile organic compound (VOC) emissions in Nagoya City from fiscal years 2003 to 2018.

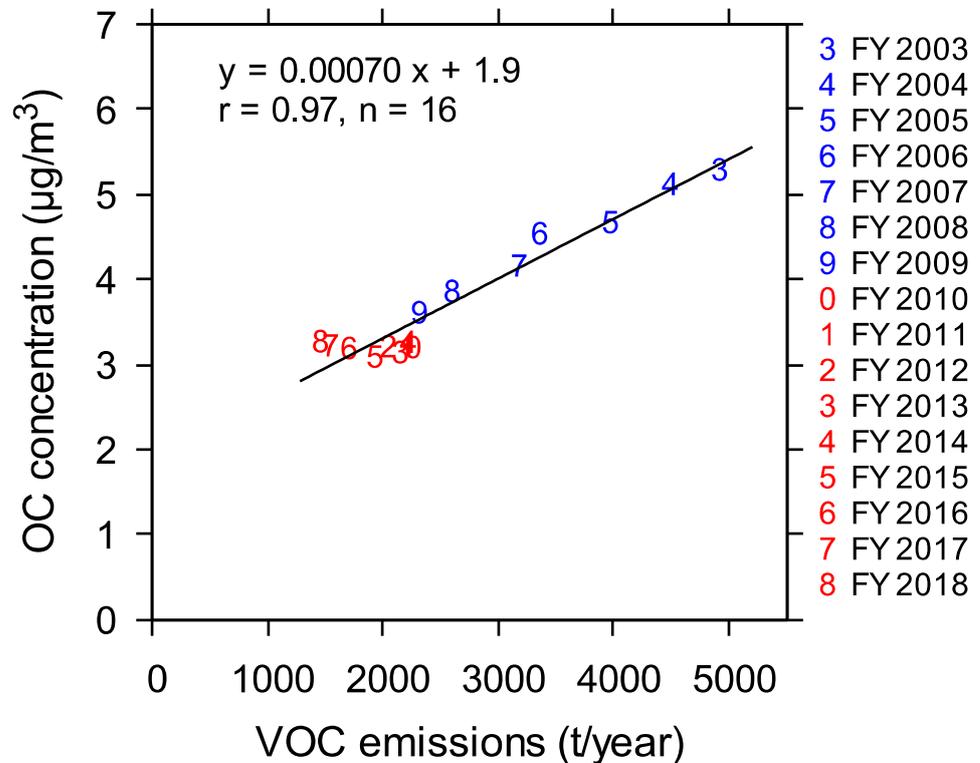


Figure 6. Scatter plot of VOC emissions in the Nagoya urban area versus the annual mean concentration of OC in PM_{2.5} at the Nagoya City Institute for Environmental Sciences in Nagoya from FYs 2003 to 2018. The black line shows the results of linear fitting for period from FYs 2003 to 2018.

3.4. Sulfate (SO_4^{2-}) in $\text{PM}_{2.5}$

Figure 7a presents the annual and seasonal mean concentrations of SO_4^{2-} in $\text{PM}_{2.5}$ between FYs 2003 and 2018 at the NCIES. SO_4^{2-} is the component of $\text{PM}_{2.5}$ that showed the second-largest decrease in concentration over the 16-year study period. Of the $13.6 \mu\text{g}/\text{m}^3$ reduction in $\text{PM}_{2.5}$, SO_4^{2-} reduction contributed 22%. There was an overall downward trend in SO_4^{2-} concentration, despite the local maxima in FYs 2005 and 2013. The annual mean SO_4^{2-} concentration decreased from $5.6 \mu\text{g}/\text{m}^3$ in FY 2003 to $2.6 \mu\text{g}/\text{m}^3$ in FY 2018, a decrease of $3.0 \mu\text{g}/\text{m}^3$ (54%) over 16 years, or $3.8\%/$ year on average.

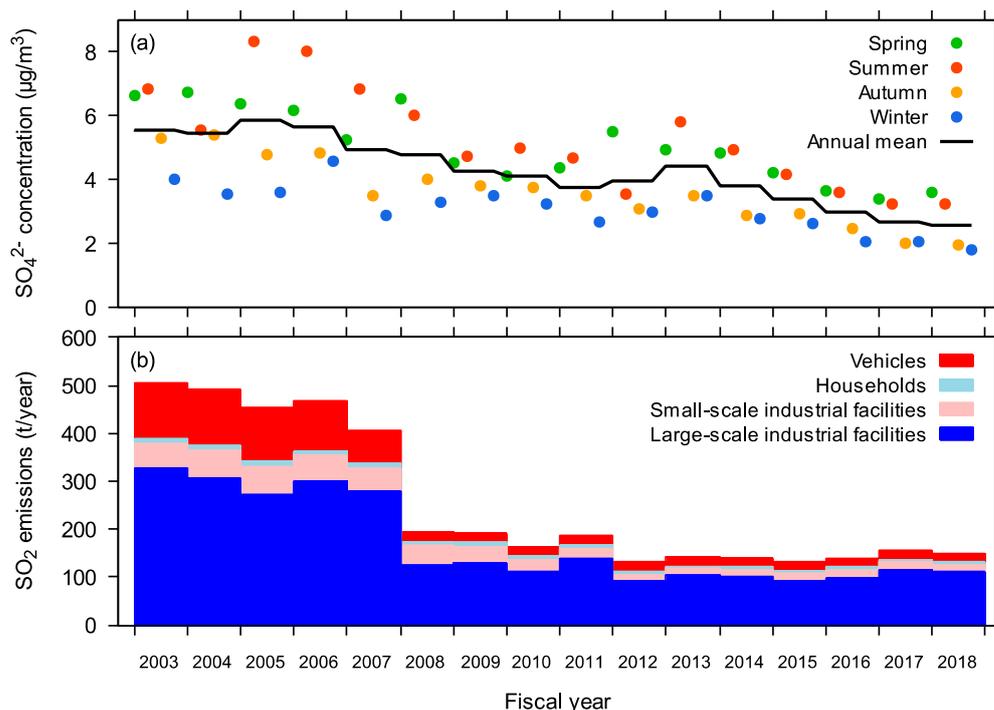


Figure 7. (a) Annual and seasonal mean concentrations of SO_4^{2-} in $\text{PM}_{2.5}$ at the Nagoya City Institute for Environmental Sciences in Nagoya for the period from fiscal years 2003 to 2018. (b) SO_2 emissions in Nagoya City from fiscal years 2003 to 2018.

Figure 7b shows SO_2 emissions in Nagoya City. Annual SO_2 emissions decreased from 504 t in FY 2003 to 148 t in FY 2018, a decrease of 71% over 16 years. As in the case of OC, the reduction in SO_4^{2-} concentration (54%) was smaller than the reduction in SO_2 emissions. SO_2 emissions decreased gradually beginning in FY 2003, but dropped significantly in FY 2008 and remained low thereafter. In particular, SO_2 emissions from vehicles decreased significantly in FYs 2007 and 2008. This drop was due to the tightening of regulations on sulfur content in diesel oil from 50 to 10 ppm in 2007 and in gasoline from 50 to 10 ppm in 2008. SO_2 emissions from stationary sources also decreased significantly in FY 2008. This drop was due to a significant decrease in fuel consumption of heavy oil at stationary sources in Nagoya City and a switch to LNG. However, the SO_4^{2-} concentration did not decrease significantly in FY 2008.

Figure 8 presents the relationship between annual SO_2 emissions and the annual mean SO_4^{2-} concentration. Overall, the SO_4^{2-} concentration was lower at lower SO_2 emissions levels ($r = 0.85$), but the association between SO_4^{2-} concentration and SO_2 emissions prior to FY 2008 was weak ($r = 0.60$, $n = 5$). Moreover, a negligible association was found between the SO_4^{2-} concentration and SO_2 emissions after FY 2008. The intercept of the regression line assumed a high value of $2.7 \pm 0.29 \mu\text{g}/\text{m}^3$. This value was almost as high as the annual mean value in FY 2018, suggesting a large contribution from sources other than SO_2 emissions in Nagoya City. SO_4^{2-} concentrations in Japan are known to be affected by

transboundary pollution [9]. SO₂ emissions in China peaked in 2006 and then showed a moderate downward trend through 2015, followed by a significant single-year decrease of 40% in 2016 [43]. SO₄²⁻ concentrations in Nagoya also showed a downward trend through 2015 and may have been influenced by emissions originating in China. However, there was no peak in China’s SO₂ emissions in FY 2013 to correspond to the 2013 peak in the SO₄²⁻ concentration at the NCIES. In addition, the single-year 13% decrease in SO₄²⁻ concentration in FY 2016 was not as large as the decrease in China’s SO₂ emissions. Thus, SO₄²⁻ concentrations in Nagoya and emissions in China did not match particularly well from year to year. The impact of transboundary pollution on the SO₄²⁻ concentration is expected to be related largely to meteorological factors.

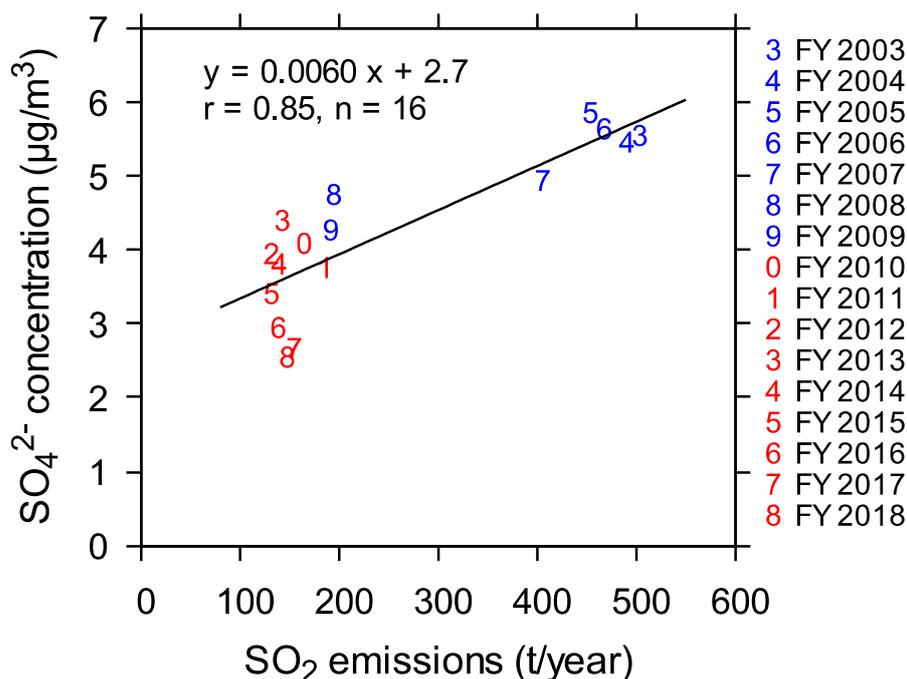


Figure 8. Scatter plot of SO₂ emissions in Nagoya City versus the annual mean concentration of SO₄²⁻ in PM_{2.5} at the Nagoya City Institute for Environmental Sciences in Nagoya from FYs 2003 to 2018. The black line shows the results of linear fitting for the period from FYs 2003 to 2018.

In Japan, SO₂ emissions from volcanoes have been observed frequently, and SO₄²⁻ measurement values at observation sites are affected by the wind direction. Because the observation site is close to the ocean, it may be affected by dimethyl sulfide (DMS) emitted from the ocean. Phytoplankton in the ocean are more active in summer, and SO₄²⁻ derived from DMS is expected to affect them in summer when there are stronger southerly winds from the ocean. However, as illustrated in Figure 7, the same decreasing trend is observed in all seasons (spring, summer, autumn, and winter), suggesting that changes in factors other than DMS contributed to the decreasing trend. SO₄²⁻ concentrations in Nagoya City may have been affected substantially by transboundary pollution, volcanoes, and meteorological factors, rather than primarily by sources in Nagoya City.

3.5. Nitrate (NO₃⁻) in PM_{2.5}

Figure 9a presents the annual and seasonal mean concentrations of NO₃⁻ in PM_{2.5} between FYs 2003 and 2018 at the NCIES. The decrease in NO₃⁻ constituted 9% of the 13.6 µg/m³ PM_{2.5} concentration decrease over the 16-year study period. The annual mean NO₃⁻ concentration decreased from 1.9 µg/m³ in FY 2003 to 0.6 µg/m³ in FY 2018, a decrease of 1.3 µg/m³ (67%) over 16 years, or 3.3%/year on average.

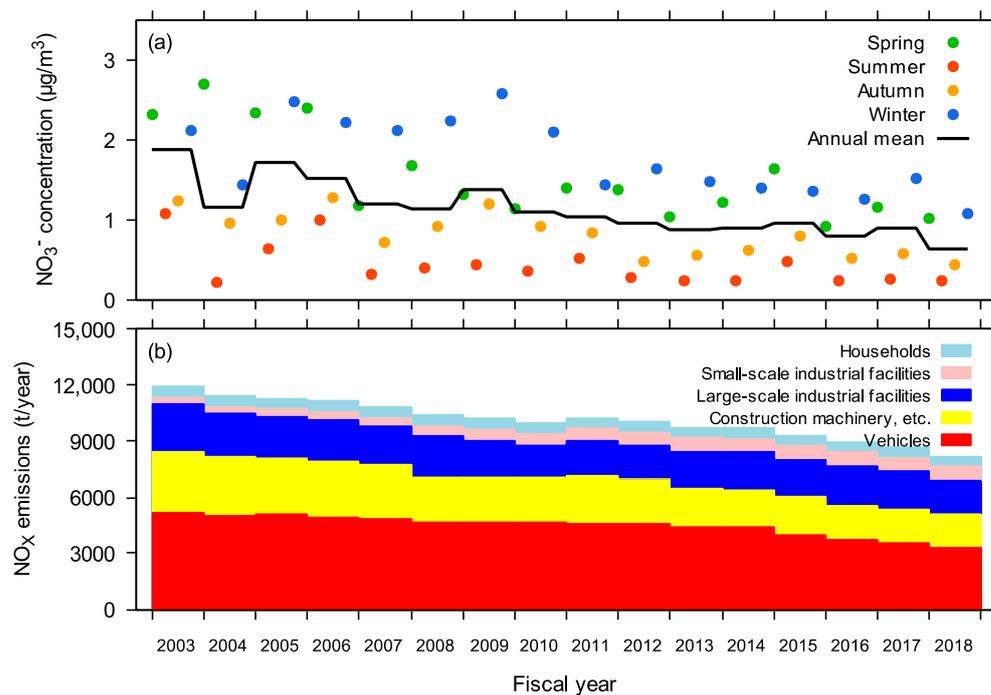


Figure 9. (a) Annual and seasonal mean concentrations of NO_3^- in $\text{PM}_{2.5}$ at the Nagoya City Institute for Environmental Sciences in Nagoya from fiscal years 2003 to 2018. (b) NO_x emissions in Nagoya City for the period from fiscal years 2003 to 2018.

Figure 9b shows NO_x emissions in Nagoya City. The annual NO_x emissions decreased from 11,921 t in FY 2003 to 8210 t in FY 2018, a decrease of 31% over the 16-year study period. Unlike OC and SO_4^{2-} , NO_3^- showed a concentration decrease (67%) that was larger than the corresponding decrease in emissions. NO_x emissions trended downward from FYs 2003 to 2018, decreasing at an average rate of 1.8%/year. NO_x emissions from vehicles showed the largest decrease among the major sources, decreasing by 1890 t (36%) over the 16-year study period, from 5202 t in FY 2003 to 3312 t in FY 2018. NO_x emissions from vehicles accounted for 44% of total emissions in FY 2003, but decreased slightly to 40% in FY 2018. NO_x emissions from construction machinery, etc., decreased by 1492 t (46%) over the 16-year study period, from 3263 t in FY 2003 to 1771 t in FY 2018. NO_x emissions from construction machinery, etc., accounted for 27% of the total emissions in FY 2003, but only to 22% in FY 2018. Annual NO_x emissions from stationary sources decreased by 736 t (29%) over 16 years, from 2530 tons in FY 2003 to 1794 t in FY 2018. The proportion of total NO_x emissions that originated from stationary sources remained almost constant, at about 20%.

Figure 10 presents the relationship between annual NO_x emissions and the annual mean NO_3^- concentrations. The NO_3^- concentration was lower at lower NO_x emissions levels ($r = 0.87$), indicating that the reduction in NO_x emissions in Nagoya City was effective in reducing nitrate in $\text{PM}_{2.5}$. On the other hand, the intercept of the regression line was negative. This finding indicates that particulate NO_3^- would not be observed for NO_x emissions below 6100 t, assuming that the linear relationship is maintained as NO_x emissions decrease. As NO_x emissions continue to decrease in the future, it will be necessary to closely monitor changes in the concentration of NO_3^- in $\text{PM}_{2.5}$.

3.6. Ammonium (NH_4^+) in $\text{PM}_{2.5}$

NH_4^+ in $\text{PM}_{2.5}$ is typically present as ammonium sulfate or ammonium nitrate. Figure 11 presents the relationship between the sum of SO_4^{2-} and NO_3^- concentrations and the NH_4^+ concentration over the period from FYs 2003 to 2018. The correlation between the NH_4^+ concentration and the $\{\text{SO}_4^{2-} + \text{NO}_3^-\}$ concentration is very high ($r = 0.99$),

indicating that the NH_4^+ concentration is determined by SO_4^{2-} and NO_3^- concentrations. The annual mean NH_4^+ concentration decreased from $2.6 \mu\text{g}/\text{m}^3$ in FY 2003 to $0.96 \mu\text{g}/\text{m}^3$ in FY 2018, a decrease of $1.6 \mu\text{g}/\text{m}^3$ (63%) over 16 years. The decrease in NH_4^+ constituted 12% of the $13.6 \mu\text{g}/\text{m}^3$ $\text{PM}_{2.5}$ concentration decrease over the 16-year study period. NH_4^+ in $\text{PM}_{2.5}$ was probably incidentally reduced by the reductions in SO_2 and NO_x emissions.

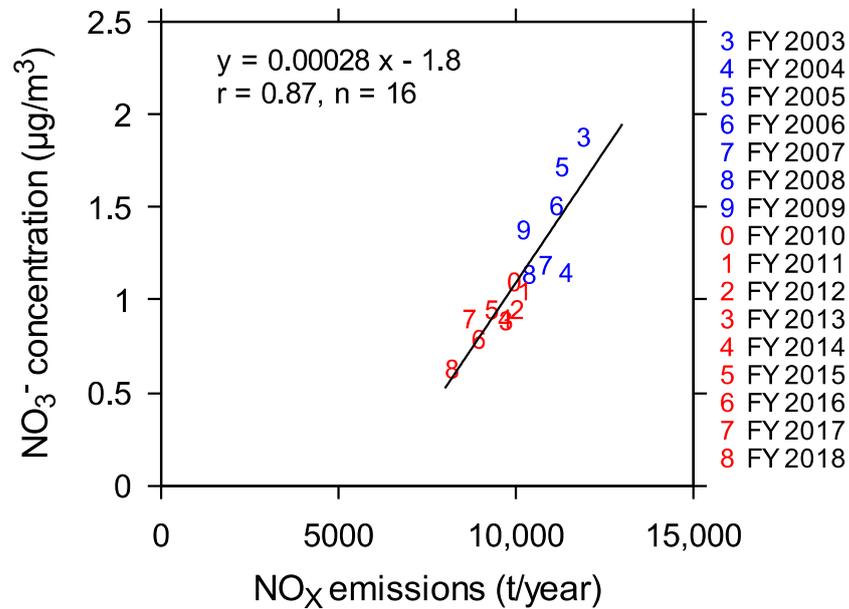


Figure 10. Scatter plot of NO_x emissions in Nagoya City versus the annual mean concentration of NO_3^- in $\text{PM}_{2.5}$ at the Nagoya City Institute for Environmental Sciences in Nagoya from FYs 2003 to 2018. The black line shows the results of linear fitting for the period from FYs 2003 to 2018.

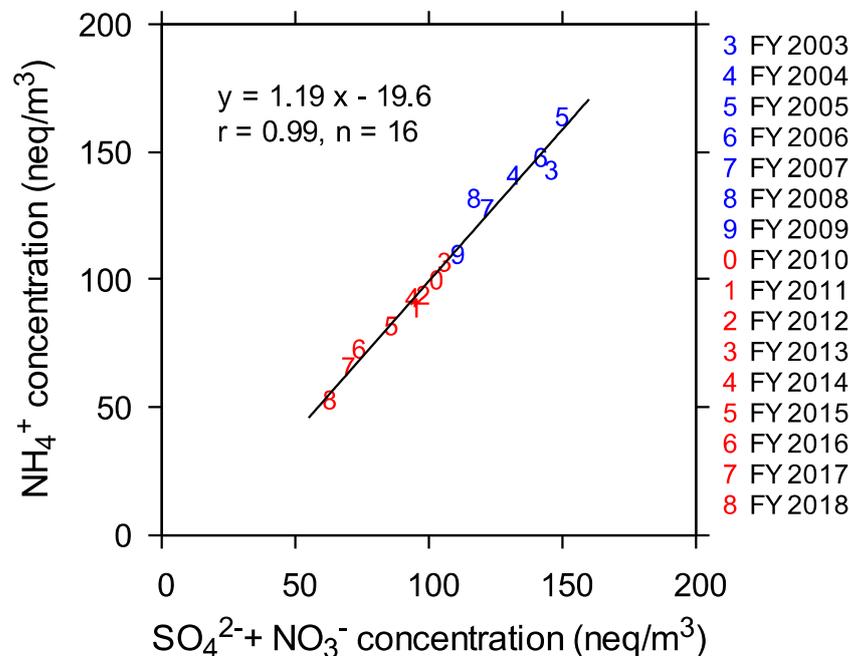


Figure 11. Scatter plot of the $\text{SO}_4^{2-} + \text{NO}_3^-$ concentration versus the NH_4^+ concentration in $\text{PM}_{2.5}$ at the Nagoya City Institute for Environmental Sciences in Nagoya from FYs 2003 to 2018. The black line shows the results of linear fitting for the period from FYs 2003 to 2018.

4. Conclusions

We evaluated the extent to which various countermeasures were effective in reducing PM_{2.5} concentrations in Japan, by analyzing the long-term trends in the concentrations of the major components of PM_{2.5} and their corresponding emissions in Nagoya City. PM_{2.5} concentrations decreased by 53% (13.6 µg/m³) over the 16-year period from FYs 2003 to 2018 in Nagoya City. Among the components of PM_{2.5}, EC showed the greatest decrease (4.3 µg/m³) over the 16-year period, followed by SO₄²⁻ (3.0 µg/m³), OC (2.0 µg/m³), NH₄⁺ (1.6 µg/m³), and NO₃⁻ (1.3 µg/m³). The decrease in EC concentration was found to be due largely to the effect of diesel emission control. Patterns in SO₄²⁻ concentration indicated that domestic measures had a limited effect and that the effects of transboundary pollution from China, volcanoes, and meteorological factors were significant. OC concentrations were affected by domestic measures, such as VOC emission regulations for stationary sources and the reduction of VOCs from vehicles, construction equipment, etc. NO₃⁻ concentrations decreased; vehicles contributed most strongly to this decrease, followed by construction machinery etc., and finally stationary sources. NH₄⁺ concentrations decreased incidentally because of reductions in SO₂ and NO_x emissions. In sum, the relationship between the concentrations of the major PM_{2.5} components and their corresponding emissions revealed which source measures in Japan were effective in reducing PM_{2.5} concentrations. These data relationships also revealed the ineffectiveness of some recent countermeasures for some components, such as those targeting OC concentrations.

Author Contributions: Conceptualization, S.W., M.Y., and K.O.; methodology, M.Y. and K.O.; software, M.Y. and K.H.; validation, F.I. and K.H.; formal analysis, M.Y., F.I., and K.H.; investigation, M.Y., F.I., K.H., and H.N.; resources, K.U.; data curation, M.Y.; writing—original draft preparation, M.Y.; writing—review and editing, M.Y., K.O., F.I., K.H., H.N., K.U., and S.W.; visualization, M.Y. and K.H.; supervision, M.Y.; project administration, M.Y.; funding acquisition, K.O., M.Y., K.U., and S.W. All authors have read and agreed to the published version of the manuscript.

Funding: This research was partly supported by the Environment Research and Technology Development Fund (JPMEERF20165004) of the Environmental Restoration and Conservation Agency of Japan. This research was also partly supported by Grants-in-Aid for Scientific Research from the Japanese Ministry of Education, Culture, Sports, Science and Technology (16K09097).

Data Availability Statement: The data reported in this study will be available on request from the corresponding author.

Acknowledgments: Maps were drawn using the Generic Mapping Tools (GMT) 5.4.3 [44].

Conflicts of Interest: The authors declare no conflict of interest.

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