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Long term trend of particulate matter in Philadelphia, Pennsylvania and its association with introduction of environmental policies

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Abstract
Since the 1970s, air quality has improved at the national level in the United States, coincident with the introduction of the Clean Air Act and other air pollution regulations at a greater frequency. We present a case study from Philadelphia, Pennsylvania—the sixth most populous city in the United States. The main objectives of this study are to analyze long-term trends of particulate matter (PM) from 1986 to 2021 in Philadelphia and to examine their association with the introduction of environmental policies relevant to air pollution at the federal, state, and local levels. We find that annual PM_{10} concentration decreased by 47% from 1986 to 2021 and annual $PM_{2.5}$ concentration decreased by 31% from 2000 to 2021 in Philadelphia. We find that carbonaceous content (both elemental and organic carbon) has declined over the same period of 2000 to 2021, demonstrating its contribution to overall $PM_{2.5}$ reduction in Philadelphia. In Philadelphia, high PM concentrations occur in the summer months; however, seasonal patterns have changed for PM_{10} in the last decade (2011–2020). Overall, PM reductions occurred over all seasons, with the greatest reductions occurring for $PM_{2.5}$ during summer months and for PM_{10} during winter months. The Clean Air Act contributed to the creation of many regulatory policies that address unhealthy levels of PM. The introduction of various environmental policies that target the transportation sector has contributed to the reduction of PM levels in Philadelphia. Air quality would continue being improved by implementing such environmental policies specific to the emissions sectors.

Keywords Particulate matter · Organic carbon · Elemental carbon · Philadelphia · Clean air act

1 Introduction

Particulate matter (PM) pollution is a major concern for global air quality. Various studies on the effects of PM pollution have been carried out globally, with studies done in both developed and developing countries [1–4]. Exposure to PM is associated with mortality and morbidity; both long and short term respiratory, cardiovascular, and cardiopulmonary diseases [2, 5–9]. The United States Environmental Protection Agency (US EPA) has adopted the National Ambient Air Quality Standards (NAAQS) for PM of two sizes: $PM_{2.5}$ (particles smaller than 2.5 μm in aerodynamic diameter) and PM_{10} (particles smaller than 10 μm in aerodynamic diameter). The US EPA's NAAQS are important in improving the air quality of states, as these standards lead to further policy implementation within states. For example, Pennsylvania adopted the

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NAAQS and additionally began regulating ambient concentrations of beryllium, fluorides, and hydrogen sulfide [10]. A study analyzing long-term PM trends across all 50 U.S. states, found negative time trends with adequate measures to reduce PM [2]. This study found that eastern states are seeing steeper decreasing trends in PM compared to western states, supporting the effectiveness of NAAQS implemented in some areas [2]. In 2019, ambient $PM_{2.5}$ caused 47,800 deaths in the US and over 4 million deaths globally [11]. Air pollution was the fourth-leading cause of global mortality in 2020 [12, 13]. About a 1% increase in mortality and 2–4% increase in mortality has been associated with 10 μg/m^3 increase in PM_{10} and 5 μg/m^3 increase in $PM_{2.5}$, respectively [14]. Several epidemiological studies have showed the increased risk of low birthweight and preterm birth with per 10 μg/m^3 increase in $PM_{2.5}$ exposure [12].

PM is composed of various chemical components, which is highly variable; its environmental and health effects also depend on its chemical composition and size. Carbonaceous components consisting of elemental carbon (EC) and organic carbon (OC) contribute to large amount of PM mass and are highly variable spatially and temporally [4, 15, 16]. Carbonaceous components can contribute up to 40% of $PM_{2.5}$, which are distinguished by their sources, water solubility, light, and heat [17]. Carbonaceous aerosol are emitted from various sources including vehicle exhaust, vegetative burning, cooking, and atmospheric reactions [18]. Carbonaceous aerosol affects climate by scattering insolation, and affecting condensation nuclei formation. Human health effects of carbonaceous aerosols include cancer, developmental effects, and cardiovascular mortality [1, 18].

PM concentrations have declined in the US over the last two decades. From 2000 to 2021, the US EPA reported that there has been a 37% decrease in $PM_{2.5}$ on a national scale and a 7% decrease in PM_{10} concentrations [19–21]. However, between 2016 and 2018, a 5.5% increase in $PM_{2.5}$ was associated with 9,700 additional premature deaths [22]. An increase in wildfires, weakened enforcement of Clean Air Act (CAA) regulations, economic growth, and increased emissions are likely to be the main contributors to the recent increase of PM in the United States [2]. From 2000 to 2021, $PM_{2.5}$ declined by about 42% and PM_{10} declined by about 32% in the Northeast region [19, 20].

Philadelphia, Pennsylvania is the sixth populous city in the US; as of 2020, Philadelphia has a population of 1.62 million and an annual growth rate of 0.48% [23]. Philadelphia also has a large minority community with 40.1% of residents being African American [23]. About a quarter of Philadelphia's population lives below the poverty line, which is $19,700 for a family with one adult and two children [24]. Philadelphia is the 17th out of 199 most polluted city – in annual PM pollution – in the US [25]. A case study from Philadelphia found that high levels of $PM_{2.5}$ are associated with increased risk of childhood asthma, making $PM_{2.5}$ especially dangerous to populations with fragile respiratory systems [26]. A mobile monitoring study in Philadelphia found that $PM_{2.5}$ constitutes 83.6% of PM_{10}, indicating that anthropogenic sources such as traffic or increased urban development are major contributors to Philadelphia's air pollution [27]. $PM_{2.5}$ pollution also varies highly across Philadelphia neighborhoods, with some neighborhoods having about three times higher $PM_{2.5}$ concentration (Chestnut Hill: ~ 13 μg/m^3) than others (Mill Creek: ~ 4 μg/m^3) [28].

Beginning in the 1970s with the creation of the US EPA and the passage of the CAA, the US has created more stringent policies regarding air pollution. The passage of the CAA provided the United States an estimated $22 trillion in human health and reduced mortality benefits, while only losing $500 billion in implementation costs (monetary values not adjusted for inflation from 1990) [29]. Aside from economic value, the CAA has successfully decreased the risk of mortality from ambient particulate pollution. Just one year of implementation was believed to prevent 1,300 premature deaths [30]. Further amendments to the CAA and consequent policies can have significant economic and health benefits [31].

The US EPA created the NAAQS to regulate six criteria air pollutants: carbon monoxide, nitrogen dioxide, sulfur dioxide, ozone, lead, and particulate matter ($PM_{2.5}$ and PM_{10}). Initially, the CAA regulated only "total suspended particles", without distinguishing between $PM_{2.5}$ and PM_{10} [32]. It was not until 1987 that the US EPA revised particle pollution standards by targeting PM_{10} through the creation of a 24-h standard of 150 μg/m^3 and an annual standard of 50 μg/m^3. $PM_{2.5}$ regulation was not added until 1997 and was set at a 24-h standard of 65 μg/m^3 and an annual standard of 15 μg/m^3. Since 2010, $PM_{2.5}$ has had a primary annual standard of 12 μg/m^3 (averaged over 3 years) and a 24-h standard of 35 μg/m^3, while PM_{10} has had a 24-h standard of 150 μg/m^3. In 2024, the US EPA has reduced annual $PM_{2.5}$ standard from 12 to 9 μg/m^3. While these are the current enforced standards, the World Health Organization (WHO) recommends that the annual mean should not exceed 5 μg/m^3 and the 24-h mean should not exceed 15 μg/m^3 for PM_{10} [33]. Along with new standards, several federal policies have been passed with the goal of improving air quality as a result of the CAA and its subsequent revisions of 1977 and 1990, which are discussed in detail in the environmental policies section below. Several environmental policies addressing air quality concerns are likely to be responsible for the overall declining trends of PM in the United States [34, 35].

This study analyzes PM_{10} trends from 1986 to 2021, $PM_{2.5}$ trends from 1999 to 2021, and elemental and organic carbon content of $PM_{2.5}$ from 2000 to 2021 in Philadelphia, Pennsylvania. Introduction of various environmental policies relevant to air pollution on a federal, state, and local level are reviewed and their association with particulate matter reduction are examined.

2 Method

2.1 Air quality data

This study was conducted using publicly available PM observations (24-h measurement) downloaded from the US EPA's Air Quality System database (https://www.epa.gov/aqs). We downloaded PM data from all the sites in Philadelphia, and selected the monitoring site that had the PM measurement from the earliest year. Air Monitoring Site 42,101,004 (1501 E. Lycoming Ave, Philadelphia) had PM_{10} data starting from the year 1986. The same site started to have $PM_{2.5}$ measurements starting from 1999, and therefore, we selected this site for $PM_{2.5}$. To fill in any missing periods, we selected another available site that was in proximity to the same site within Philadelphia. PM_{10} data at this station was not available after 2012. We downloaded PM_{10} data from another site, Site 421,010,048 (3000 Lewis St, Philadelphia) that was within 2.1 miles from Site 4 for the years 2013 to 2021. These sites were selected because they were closer to each other and after merging, PM data was available from the beginning of the measurement i.e. 1986. The locations of sampling sites within Philadelphia are shown in Figure S1 in supplementary section. Number of observations for each pollutant type with site information is given in Table S1 in Supplementary Information.

$PM_{2.5}$ data was available from 15 different sites in Philadelphia. We selected the site that has $PM_{2.5}$ data for the longest period, which is Site 421,010,004, where $PM_{2.5}$ data was available from 1999 to 2018. $PM_{2.5}$ data was not available after the year 2018 at this site, and we selected the site that is closest to this site, Site 421,010,048. $PM_{2.5}$ data was downloaded from the years 2019 to 2021 for Site 421,010,048. We checked for negative and zero values and removed one PM_{10} and three $PM_{2.5}$ data. $PM_{2.5}$ was measured by more than one method and for several months, more than one $PM_{2.5}$ data was available for the same day because of different measurement methods. We included all available $PM_{2.5}$ data. We prepared a figure for yearly averages (Fig. 1). We prepared boxplots to show variation within a year (Figures S2 and S3).

We also downloaded $PM_{2.5}$ data from the additional sites that had continuous measurements for 10 years or more. There were three sites meeting such criteria: Site 421,010,024 (Philadelphia NE Airport), 421,010,055 (25th and Ritner Street), and 421,010,057 (240 Spring Garden Street). Results for these additional sites are given in Figure S4.

Elemental (EC) and organic (OC) carbon content of $PM_{2.5}$ was obtained from the Site 421,010,004 for the years 2000 to 2010, Site 421,011,002 (5200 Pennypack Park) for the years 2011 to 2012, and from Site 421,010,048 for the years 2013 to 2021. For the years 2000 to 2007, total EC and total OC data of $PM_{2.5}$ were downloaded from chemical speciation network (CSN) that was labelled as EC-CSN-PM2.5-LC-TOT, and OC-CSN-Unadjusted-PM2.5-LC-TOT, respectively. From the years 2008 to 2021, total EC and OC data were downloaded from the chemical speciation network that were labelled on the database as EC-CSN-Rev-Unadjusted-PM2.5-LC-TOT and OC-CSN-Rev-Unadjusted-PM2.5-LC-TOT, respectively. We checked for data and removed one unusually high OC value and 16 zero or negative EC values. EC and OC data were also available from other sites in Philadelphia. We downloaded EC and OC data from Site 421,010,136 (Amtrak, 5917 Elmwood Avenue)

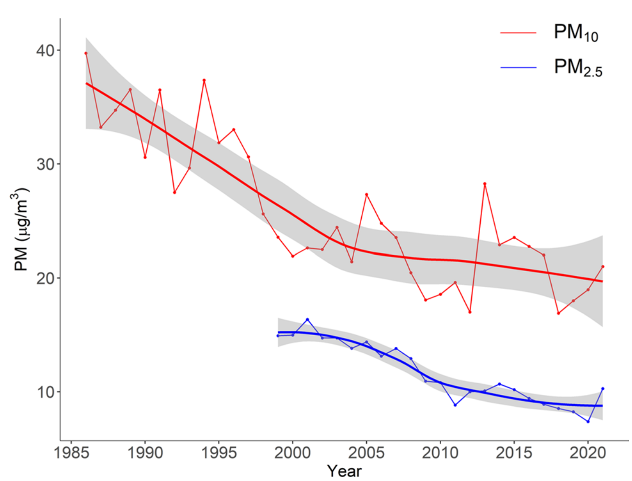

Fig. 1 Trend of yearly average PM_{10} (red) concentrations from 1986 to 2021 and $PM_{2.5}$ (blue) concentrations from 1999 to 2021. A smoothed analysis from ggplot2 package in r is placed over the data in gray in this figure and subsequent plots. Boxplots showing variation within a year for PM_{10} and $PM_{2.5}$ are given in Figure S2 and S3, respectively. Yearly trend of $PM_{2.5}$ concentrations for additional sites are given in Figure S4

for the years 2002 to 2004 and from Site 421,010,055 (24th & Ritner Streets) for the years 2005 to 2021. Yearly trend and monthly trends for this data are shown in Figures S5 and S6.

All data analysis was conducted in RStudio using R packages openair and ggplot2. We computed yearly means and analyzed yearly trends. We divided our data into different decades (except for the period 1986–2000, which is longer than a decade) and computed monthly means for each decade. We then analyzed monthly means to assess the seasonal variety in different decades. We used the geom_smooth function from ggplot package to show the pattern of trend on the plots. Besides the yearly changes, we investigated the decadal changes, and the trends during events such as July 4th, where there may be large PM emission from firework activities [36].

2.2 Emission inventories

PM emissions in Philadelphia were obtained from the US EPA National Emissions Inventory (NEI) [37]. PM emissions in Philadelphia, Pennsylvania were downloaded for five different years (2008, 2011, 2014, 2017, and 2020). The emission data were available for various emission sectors and are given in tons. We analyzed emission data for five major sources (commercial, dust, fuel combustion, industrial processes, mobile-off road and mobile-on road). A table listing the PM emission inventories is given in Table S2 in supplementary section. Documentation and further information on emission inventories for various pollutants are available on the website of US EPA (https://www.epa.gov/air-emissions-inventories). Besides identifying these key sources based on emission inventories, we considered the influence of various sources around Philadelphia such as refinery, coal emission sources, traffic, and tourism activities [1, 36, 38, 39].

2.3 Environmental policies

We obtained federal environmental policies that were introduced by the US EPA by reviewing the Federal Register (https://www.federalregister.gov/). State policies were listed by Pennsylvania's Department of Environmental Protection and were in the Pennsylvania Code and Bulletin. Regulatory actions were placed on a timeline to identify periods of high government action, though this list is not extensive. We prepared a graph listing the environmental policies that are relevant to air quality based at federal, state, and local level. A list of the policies is given in Table S3 in the supplementary section. We analyzed the association between reduction in yearly PM levels with the introduction of any relevant environmental policies. We mainly investigated environmental policies based on the transportation sector and energy sector. This is a qualitative analysis, and we did not use any statistical tests for seeking the association of PM trends with the introduction of environmental policies. We reviewed the studies conducted in other cities in the U.S. to learn about the effectiveness of environmental policies in reducing air pollution, and how it can serve as a model for future air pollution reductions [6, 40–42].

3 Results

3.1 Particulate matter yearly trend

Overall PM concentrations have decreased in the Philadelphia region (Fig. 1). The yearly average PM_{10} decreased by 47% from 1986 to 2021. The yearly average PM_{10} decreased by about 11% and $PM_{2.5}$ decreased by 31% from 2000 to 2021. While the overall decreasing trend for both pollutants are similar, spikes and drops for PM_{10} and $PM_{2.5}$ occurred in different years. Largest declines for PM_{10} were observed for the periods 1995–2000 (31%) and 2005–2009 (34%); and during 2007–2011 (36%) and 2015–2020 (27%) for $PM_{2.5}$. Large increases occurred in 1994, 2005, and 2013 for PM_{10}; and 2001, 2012, and 2021 for $PM_{2.5}$ (Fig. 1). Besides looking at yearly averages, we also looked at overall trend by making the boxplots of PM_{10} and $PM_{2.5}$ for each year (Figures S2 and S3). This shows an increase in PM_{10} concentrations from the year 2013. Overall, there is a decreasing trend for $PM_{2.5}$ concentrations as shown by other air quality monitoring sites within Philadelphia (Figure S4).

3.2 Particulate matter seasonal trend

Both $PM_{2.5}$ and PM_{10} showed high seasonal variability in Philadelphia (Figs. 2a and b), with a shifting seasonal pattern for PM_{10} in recent years (Fig. 2b). For the first two periods (1986–2000 and 2001–2010), PM_{10} was higher during summer months (June, July, August) (Fig. 2b). However, the trend shifted for 2011–2020, when the peak PM_{10} concentrations appeared during winter months (December-January). For $PM_{2.5}$, there appears to be two peaks, a large peak during summer months (June–August), and a small peak during winter months (December-January) (Fig. 2a). $PM_{2.5}$ decreased during all months from the first period (1999–2010) to the second period (2011–2020). A large reduction for $PM_{2.5}$ was observed in the summer months from the first (1999–2010) to the second period (2011–2020). In contrast, PM_{10} observed a large reduction in the winter months period from the first period (1986–2000) to the second period (2001–2010). The last decade (2011–2020) showed a contrasting pattern with no summer peak of PM_{10}, and the largest decline observed during summer period.

3.3 Carbonaceous aerosol trends

EC and OC trends in Philadelphia began to decline mainly after 2005 (Fig. 3). From 2005 to 2009, there was a greatest decline in carbonaceous content. During this period, EC concentrations decreased by about 59% and OC concentrations decreased by about 51%. But EC concentration started to increase from 2017 to 2019. EC concentration increased by about two times from 2016 to 2019, but it started to decrease after 2019. In contrast, OC concentrations have steadily decreased over the last two decades, with slight increases during 2013 and 2015. Yearly trend of EC and OC follows a similar pattern until about 2012, then they seem to deviate (Fig. 3). When EC and OC concentration yearly trend was analyzed for additional sites (Figure S5), both EC and OC concentrations showed a decreasing trend except a slight increase in EC from 2017.

OC concentrations followed a similar pattern to $PM_{2.5}$ in seasonality and a decreasing trend (Fig. 4). OC concentrations showed two large peaks during summer and winter months. EC concentration seasonal profiles were different than OC. EC concentrations were higher during winter months compared to summer months. EC concentration decreased in 2011–2020 compared to 2000–2010. The greatest decline in EC concentration occurred during winter months (Fig. 4).

3.4 Environmental policy

Several regulatory policies were developed over the last three decades to address air pollution concerns throughout the country and to establish, update, and meet changing NAAQS. Here, we list relevant policies and regulations at federal,

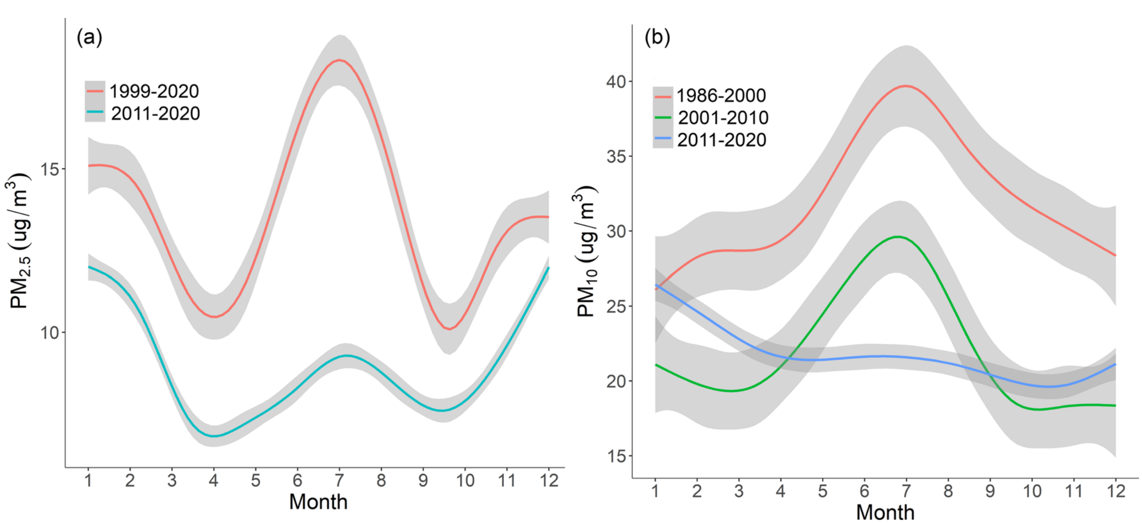

Fig. 2 **a** Monthly averages for $PM_{2.5}$ concentrations in μg/m^3 from two periods: 1999–2010 and 2011–2010; **b** Monthly averages for PM_{10} concentrations in μg/m^3 from three periods: 1986–2000, 2001–2010 and 2011–2020. A smoothed analysis from ggplot2 package in r program is placed over the data in gray. Month numbers are in order from January as month 1 and December as month 12

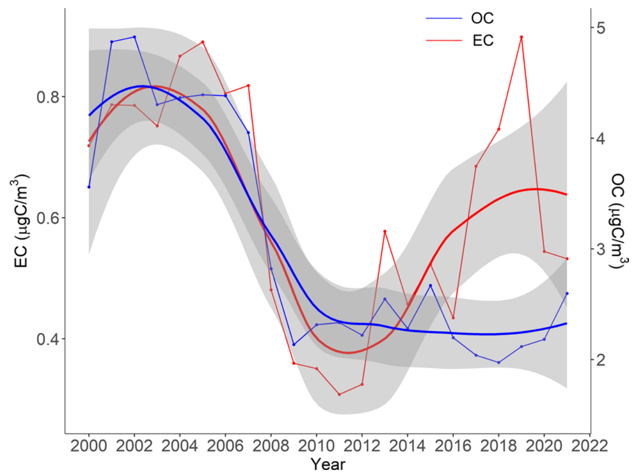

Fig. 3 Yearly trend of EC and OC concentrations from 2000 to 2021 at Philadelphia. Yearly trend of EC and OC concentrations for additional sites are given in Figure S5

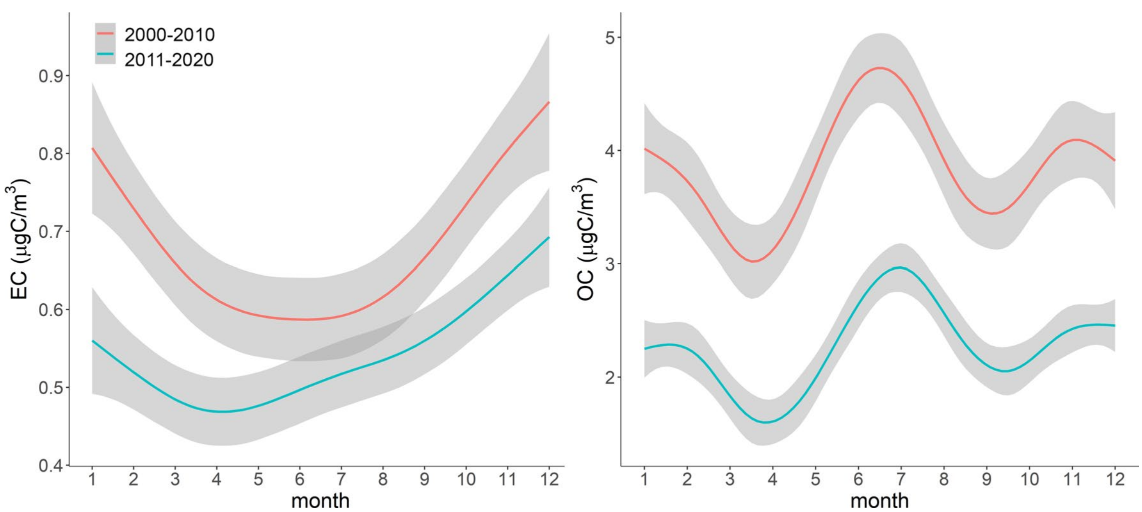

Fig. 4 Monthly variation of (a) EC and (b) OC concentrations from 2000 to 2020 at Philadelphia for two decades. Months 1 to 12 are January to December. Monthly variation for additional sites is given in Figure S6

state, and local levels benefiting Philadelphia's air quality. A timeline of introduction of major environmental policies and regulations relevant to particulate pollution is shown in Fig. 5 and Table S3.

3.4.1 Transportation sector

A large portion of national policies worked to combat PM emissions from the transportation sector. Programs such as the National Low Emissions Vehicle Program (1999), the Reformulated Gasoline Program (1995), and stringent on and off-road diesel emissions standards were thought to be especially effective during this period. The Clean Air Non-Road Diesel Rule (2004) that was phased in with the 2008 model year, when non-road diesel engines were introduced to reduce PM emissions. Many other environmental policies promised similar reductions and have been successful, as PM has experienced significant reductions since their implementation [6, 35, 35]. Many of these programs were created as a result of the 1970 CAA and its subsequent revisions in 1977 and 1990 that called for cleaner vehicles, more stringent standards for non-road engines and industrial processes, state implementation plans (SIPs), and nationwide standards. In 2005, the Energy Policy Act was passed which led to the creation of many grant programs, such as the Diesel Emissions Reductions Act of 2008 that allocated money to states to create diesel reductions programs.

On the state level, policies such as the Pennsylvania Clean Vehicles Program (1998) and the Pennsylvania Heavy Duty Diesel Emissions Control Program (2004) have reduced emissions from vehicles. The Pennsylvania State Clean Diesel Grant Program (2010) offered grants for projects that would replace, repower, or retrofit fleet diesel-powered highway

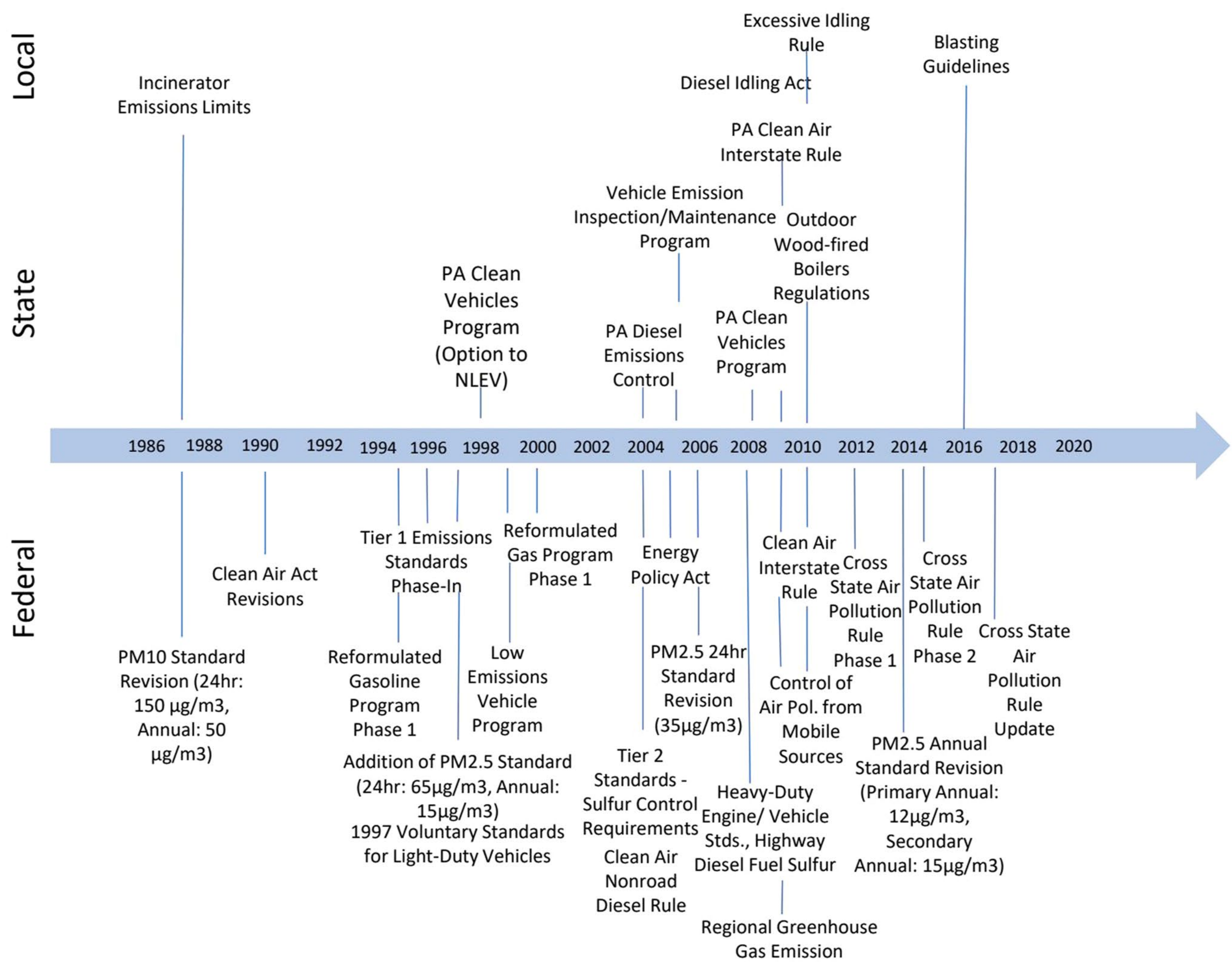

Fig. 5 A non-exhaustive list of historical environmental policies at federal, state, and local level. Dates largely based on when policies were phased-in, rather than legally passed

and nonroad vehicles, engines and equipment. Programs like the Pennsylvania Clean Vehicle Program go beyond federal regulations and create stricter standards that must be followed within the state. This started as an option to the National Low Vehicles Program for vehicle manufactures; however, the option to follow the national program expired in 2006 and manufacturers were required to comply with the state program by 2008. Many of these programs were implemented as part of Pennsylvania's SIP to meet NAAQS and were inspired by other state SIP actions. Pennsylvania's Clean Vehicles Program was largely based on California's Low Emissions Vehicle Program. On the local level, Philadelphia has set excessive idling rules [43].

3.4.2 Energy sector

On the national level, the US has set forth policies regarding energy and energy emissions. The Energy Policy Act (2005) addresses energy efficiency, renewable energy, oil and gas, coal, vehicle and motor fuels, and more [44]. The US has regulated oil and gas industries, which has in turn lent to attaining air quality standards. The Energy Independent and Security Act (2007) increased the use of clean fuels and increased vehicle efficiency [45]. On a local level, the City of Philadelphia has set limits on incinerator emissions and has created strict blasting guidelines. Also, local limits on incinerations emissions (1987) were implemented in Philadelphia. The Energy Policy Act (2005) additionally plans that energy in the US will change towards more renewables, lowering greenhouse gas emissions from fuel combustion. Due to interstate air travel of pollutants, the Regional Greenhouse Gas Initiative (RGGI) (2009) was adopted to cap and reduce emissions from the power sector across 10 New England States, including Pennsylvania. The RGGI will lower emissions of carbon dioxide, nitrogen oxides, sulfur dioxide, and PM [10]. The US EPA implemented the Cross-State Air Pollution Rule (CSAPR) to target the transport of air pollutants from upwind states to downwind states. The CSAPR rule requires to reduce power plant emission in unwind states because the pollutants emitted can travel to 100s of miles downwind and can form the soot and smog during the transport affecting air quality and public health locally and regionally.

3.5 Association of introduction of environmental policies with PM trend

The first major period of decline in PM_{10} began after 1995 alongside the passage of several federal vehicle emissions reduction policies. During the first period of decline in PM_{10} (1995–2000), the Tier 1 Emissions Standards were phased in from 1994 to 2003 which included the Reformulated Gasoline Program (phased in from 1995 to 1999), new $PM_{2.5}$ standards (1997), and Voluntary Standards for Light Duty Vehicles (1997). Additionally, the National Low Emissions Vehicle Program was enacted in 1999 and the Pennsylvania Clean Vehicles Program was enforced starting in 1998. The next period of decline for PM_{10} began in 2005 lasting until 2009, followed by the first period of decline in $PM_{2.5}$ which occurred from 2007 to 2011. From 2005 to 2011, national policies such as the Energy Policy Act (2005), the Heavy-Duty Engine and Vehicle Standards (2007), Highway Diesel Fuel Sulfur Control Requirement (2007), the decrease in the $PM_{2.5}$ 24-h standard from 65 μg/m^3 to 35 μg/m^3, the Clean Air Interstate Rule for NO_x (2009), and the Clean Air Interstate Rule for sulfur dioxide (SO_2) (2010) were all phased in. On the state level, Model Year 2008 vehicles are required to comply with the stricter Pennsylvania Clean Vehicles Program, rather than the National Low Emissions Vehicle Program. The 2015–2020 $PM_{2.5}$ period of decline aligned with the national passage of the Cross-State Air Pollution Rule (2015), the Tier 3 Vehicle Standards (2017–2025), and new lowering of the $PM_{2.5}$ Primary Annual Standard Revisions from 15 μg/m^3 to 12 μg/m^3. The passage of these vehicle control standards brought large reductions in PM levels in the Philadelphia region and throughout the country. There was about 11% decline in PM_{10} in Philadelphia from 2000 to 2021, which was slightly greater than national PM_{10} decline of 7% from 2000 to 2021 [20, 21]. Philadelphia also experienced a 31% $PM_{2.5}$ decline from 2000 to 2021, while the US EPA reported national $PM_{2.5}$ declines of 37% from 2000 to 2021 [19, 21]. For the northeast region, the US EPA reported a 32% decrease for PM_{10} and a 42% decrease in $PM_{2.5}$ from 2000 to 2021 [19–21]. The major changes in standards for vehicles has allowed for a 99% reduction in diesel PM from a 2012 diesel truck compared to a 1970 diesel truck that has not experienced any of the control standards prior to 1970 [46]. Diesel PM reductions are closely associated with EC reductions as EC forms through incomplete combustion of carbon-containing materials. Changing vehicle policies and resulting lower vehicle emissions may be largely responsible for the large decline in EC emissions that occurred from 2007–2011; though, lag time in policy enaction and actual reductions could be a factor. This decline also aligns with the first $PM_{2.5}$ decline, implying that the dominance of primary combustion sources of PM at the time. However, $PM_{2.5}$ continues an overall decreasing trend after this period, indicating a decrease in secondary PM from precursor gases such as SO_2, NH_3, and other volatile organic compounds (VOCs), whereas EC begins to rise again after 2011. Like EC, OC also declined greatly from 2007–2011.

4 Discussion

4.1 Yearly trend of PM in Philadelphia

The 11% decline in PM_{10} in Philadelphia from 2000 to 2021 is comparable to the reported US EPA national PM_{10} decline of 7% from 2000 to 2021 [20, 21]. The 31% $PM_{2.5}$ decline in Philadelphia from 2000 to 2021 was slightly lower than the US EPA reported national $PM_{2.5}$ declines of 37% from 2000 to 2020 [19, 21]. Average annual PM_{10} concentrations in Philadelphia have been under the NAAQS since 2011 while the average annual $PM_{2.5}$ concentrations have been under the NAAQS since 2009 [43].

Until 2019, there were two large oil refineries located in the Philadelphia area. On June 21, 2019, the Philadelphia Energy Solutions (PES) Refinery (located 9 miles from collection sites), the largest crude oil refinery on the east coast, burned down resulting in a permanent closure. PES was a major sources of air pollution in Philadelphia, creating 762,000 pounds of chemical releases in 2012 and ranking 23 out 134 facilities for chemical release according to the Toxic Release Inventory [47]. Both $PM_{2.5}$ and PM_{10} monthly and yearly averages reflect changes during and after 2019. With PES no longer servicing Philadelphia, the region experienced a decrease in the yearly averages for $PM_{2.5}$, from 8.5 μg/m^3 in 2018, to 8.2 μg/m^3 in 2019, and to 7.3 μg/m^3 in 2020. PM_{10}, unlike $PM_{2.5}$, experienced a spike in its yearly average in 2019: 16.9 μg/m^3 in 2018, 18 μg/m^3 in 2019, and 18.9 μg/m^3 in 2020. Decrease of $PM_{2.5}$ in 2020 is also influenced by the reduced emission from various activities due to COVID-19. Monthly average for $PM_{2.5}$ and PM_{10} decreased after the June incident in Philadelphia. There was a decrease of 24% PM_{10} and 27% $PM_{2.5}$ from June to October in 2019. Such a decline was not observed in the previous year.

The Monroe Energy Refinery located in Trainer, Pennsylvania (about 25 miles from Philadelphia) remains open, processing about 190,000 barrels of crude oil each day [48]. Emissions from nearby refineries may contribute to PM pollution in Philadelphia. Other studies have found that refineries contribute to local PM concentrations. A study completed in Houston, Texas found that particulates can be released directly during startup and shutdown operations, equipment failures, routine maintenance, and from the Fluid Catalytic Cracking Unit [38].

Although following a general downward trend, PM_{10} spikes were seen in 1994, 2005, and 2013. However, according to the EPA's national PM trends, no spikes occurred nationally or in the northeast region in these years [19]. These spikes signal local sources of pollution. Each spike was followed by an immediate decrease, perhaps caused by an event-driven emission like the 2019 refinery fire. The most significant increase occurred in 2013, however, this increase was not supported by findings from the Philadelphia Air Management Services (AMS) Report for 2013 [49]. The Philadelphia AMS report uses data from several monitoring sites throughout the city; The significant 2013 increase could be specific to the study site. Policies restricting emissions by the transportation sector have been most prominent and effective. Philadelphia's spikes in PM that are inconsistent with national trends, are most likely due to localized event-driven emissions. This could suggest the need for the Philadelphia region to adopt stronger controls over the industrial sector. Though, other factors, like long-range transport from other poorly controlled upwind sources, and other factors including the older age and slower turnover of the vehicle fleet, or growth of offsetting controls, could be the cause.

4.2 Seasonal trend of PM in Philadelphia

Up until 2010, summer months accounted for the highest PM concentrations during the year. There are several possibilities for the rise in PM during summer months. Studies during 1986–2000 have shown that seasonality of PM_{10} concentrations vary by region, but that it is typical for the northeast region of the United States to have PM_{10} peaks in the summer [50]. Similar patterns of high $PM_{2.5}$ concentrations during summer months were observed in Beijing where high temperature and humidity was considered to promote secondary $PM_{2.5}$ formation [51]. PM associated with primary emissions such as local heating or industrial emission are dominant during winter, and secondary formation are dominant during summer and other seasons [16]. Photochemical formation can play a great role in seasonality of PM, especially the PM chemical components such as sulfates and organics [52]. A study in Edmonton, Alberta, Canada measured $PM_{2.5}$ over the course of 8 years and found the main contributors to secondary particulate formation were secondary organic aerosols, secondary nitrate, and secondary sulfate, respectively [53, 54]. Increased energy demand from air conditioning operations during summer can increase primary PM emission, and increased biogenic emissions in the Eastern US can promote secondary PM formation during summer. Summertime tourism and elevated driving activity can increase transportation sector PM emissions. According to Philadelphia's Tourism Indicators and Impacts Report [55], leisure travel peaks during summer months. The decline of summer particulate concentrations in the last decade indicates that the series of vehicle emissions reduction strategies implemented leading up to 2010 were successful, assuming the high concentrations from summer were mainly caused by the increase in traffic emissions. Changes in seasonal PM spikes in different decades suggest targeting season specific PM emission sources, for example formulating energy policies, not only vehicle policies. Increased firework shows were also considered as a factor in the summer concentration increase. After a firework show in Albany, New York, aerosol emissions were 10 times higher than the expected hourly vehicular emissions rate [36]. PM_{10} and $PM_{2.5}$ data from around the time of July 4th of each year from our study proved inconclusive, however. A likely cause is that the sampling site is far from the locations of the fireworks. Despite not showing a high immediate PM increase after July 4th fireworks activity, July was often the month with the highest $PM_{2.5}$, PM_{10}, and OC concentrations.

There was a summer peak of $PM_{2.5}$ in Philadelphia, but the trend differed for PM_{10} in the last decade (2011–2020) with the peak only in winter. After 2010, winter PM_{10} surpassed summer PM_{10} concentrations, which may insinuate another source of PM_{10} became more prominent or the PM_{10} emissions reduction in the recent decade occurred mainly in summer months. It is more likely that another source is present because winter concentrations in the decade 2011–2020 are higher than previous decade (Fig. 2b). In the previous decades, peak concentrations occurred in July and did not experience any other rise throughout the year. Furthermore, all PM_{10} monthly averages decreased from the first period (1986–2000) to the second period (2001–2010); however, in the third period (2011–2020), January average rises back to its average concentration in the first decade. For $PM_{2.5}$, the summer concentrations remained as peaks in the second decade (2011–2020) as they did in the first decade (1999–2010); however, these summer increases in the second period (2011–2020) were slightly lower than in winter averages unlike in the first period (1999–2000) (Fig. 2a). The summer peak in July from the first period (1999–2010) decreased significantly, while the smaller winter peak in January was less impacted compared to summer $PM_{2.5}$ reductions. Decreased spring and fall averages were observed in both decades. This pattern can be explained by looking at yearly carbonaceous aerosol trends (Fig. 3). After 2012, OC continues to decline but EC started to increase. This shows the increasing influence of primary emission sources, and whose contribution is likely to be greater in winter months. These trends correspond with findings from one study analyzing long-term seasonality across the United States from 2007–2015, which found that $PM_{2.5}$ was highest in July and January in the Philadelphia region [56]. Winter peaks are often associated with coal-powered electricity to power heating sources in the region [51]. However, according to the Philadelphia Office of Sustainability, electricity generation began transitioning from coal to natural gas in the early 2000s [57]. A recent study that reviewed data from the Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) aircraft campaign suggests that sunlight and colder temperatures can affect SO_2 and nitrogen oxides (NO_x) reactions and thus affecting secondary PM formation [58]. Although air quality regulations have reduced emissions of SO_2 and NO_x, these emissions are now more easily converted to particulates in wintertime conditions. The study suggests that when emissions reductions outpace the canceling effect occurring in the winter, the winter trends in PM will reach reductions achieved in the summer [58].

4.3 Trends of carbonaceous aerosol

OC concentrations in Philadelphia have declined since 2007 albeit at a slower rate from 2010 (Fig. 3). Before 2009, summer OC concentrations were greater than all other seasons, suggesting a source prominent during the summertime. There has been a greater decline of OC concentrations in summer from the first decade (2000–2010) to the second decade (2011–2020) similar to $PM_{2.5}$ (Fig. 4). The summer peak of OC was larger than the winter peak in the first decade (2000–2010), but the summer decrease has been greater from the first decade (2000–2010) to the second decade (2011–2020). OC follows the consistent downward trends in $PM_{2.5}$ indicating that regulations targeting total PM are also effective specifically against its OC component. However, carbonaceous concentrations are expected to increase as Philadelphia transitions to alternative energy sources such as biomass burning [57]. Studies have found that biomass burning significantly increase urban OC and $PM_{2.5}$ emissions, especially during the months of June to August [2, 59]. Carbonaceous content at the down-wind site in New Hampshire showed a high seasonal variation with high concentrations during winter and small concentrations during summer [16].

Large EC declines also began in 2007. But EC started to show an increasing trend from 2016 until 2019. EC decreases in earlier years align with a nationally observed decrease in EC from 2005 to 2015 [60]. There was also a large decline of EC from 2019 to 2020 (Fig. 3). This showed that the large increase after 2016 has been corrected as EC is continuing to decrease after 2019.

4.4 Environmental policy

In general, air quality policy should set strict permitted levels for substances in the air that states and emitters must obey, and act to lower those levels when such levels are exceeded [42]. Major policies in the US, like the CAA of 1970, set forward a precedent for both the federal government and state governments to continue enacting policies for bettering air quality (Table S3). Pennsylvania demonstrates some action to lower large emission sectors for the state, mainly transportation and energy.

Vehicle emissions are the major contributor to particulate matter and NO_x in urban locations; however, air quality can be improved with better fuel-efficiency and policies controlling transportation related emissions [34]. National and statewide policy implementations to lower vehicle standards were effective in lowering those emissions, and Pennsylvania further lowered emissions by offering grants to retrofit diesel fleets. Studies show that the introduction and growing prominence of electric vehicles, especially when they replace diesel vehicles, reduces PM emissions, and improves air quality [35]. South Korean policies incentivizes electric vehicle ownership with the goal of improving air quality [35]; similar US policy can help reducing vehicle emissions beyond improving standards. A study done in Kathmandu Valley, Nepal also found vehicle emissions to be the most prominent source [4]. Pennsylvania has focused on vehicle standards. With EC and OC being largely emitted from light-duty gasoline vehicles, continuing to implement standards on vehicle emissions can help control EC and OC. PM emissions in urban areas are often attributed to traffic, industries, natural gas, and combustion, and the environmental policies targeting such emissions can help to reduce the air pollutants.

Environmental policies were found to be instrumental in reducing air pollution in Rochester, New York [6]. Several studies have attributed the environmental policies for improving air quality in the US [6, 41, 61]. While reviewing the impacts of environmental policy on air quality, it is important to acknowledge the lag time that occurs from the passage date of a policy to its effects. Despite the lag times, trends can still be identified in years following the passage of environmental policies.

The decline of summer PM indicates that the vehicle emissions reduction strategies implemented leading up to 2010 were successful, assuming high summer concentrations were due to increased traffic. After 2010, winter surpassed summer PM_{10} concentrations likely caused by increased use of coal powered electricity to power heating sources. Carbonaceous aerosols, especially EC, are products of winter heating systems—further regulations should be focused on those winter emission sources in hopes to reduce carbonaceous aerosols. Air quality standards and policies address existing sources of emitted particulate matter, but emerging sources of PM must continue to be monitored and addressed.

Various environmental policies targeting fuel switching, renewable energy, building and transportation were attributed for the PM reductions in New York City [62]. Decline in sulfate concentrations and cleaner mobile sources reduce OC and EC concentrations by 1 $\mu g/m^3$ between 2008 to 2018 in New York city [62]. A study by [1] analyzed carbonaceous aerosols throughout 14 cities in China over 2003 to 2013. Similar to this study in Philadelphia, seasonality of carbonaceous aerosols examined emissions sources and China's emissions policies implementations. China's Five-Year Plan (2006–2010) aimed to lower sulfur dioxide emissions, conserve energy, and decrease ambient sulfur dioxide and PM_{10} [1]. They found that EC, OC, and $PM_{2.5}$ were reduced over the decade despite a 93% increase in coal consumption and 470% increase in vehicles [1]. This study conducted in China supports our study's importance of identifying seasonal trends and attributing them to a diverse set of sources, which need a diverse set of environmental regulations. A policy like the Five-Year Plan would diversify the Philadelphia region's policy profile beyond vehicular emissions.

Alongside several vehicle emissions standards, Pennsylvania experienced changes in its energy production profile. After the passage of the Energy Policy Act in 2005, states were more motivated by guaranteed loans provided by the federal government to develop technologies that avoid the by-production of greenhouse gasses which often lead to the secondary formation of PM. The City of Philadelphia's Office of Sustainability reported that since 2007, the carbon intensity of the grid has declined more than 26% [57]. This is largely due to electricity generation switching from coal to natural gas. This is also the period with a large PM decline in Philadelphia. Although Pennsylvania ranks 3rd in coal production in the United States, the state has been seeing declines in coal production. In 1986, Pennsylvania was estimated to produce 71,648 short thousand tons of coal energy, in 2019 estimates declined to 50,078 short thousand tons. As a state, and as a city, coal consumption continues to be phased out.

National policies passed have encouraged state and local improvements; for example, the national Diesel Emissions Reductions Act (2008) inspired the Pennsylvania Clean Diesel Grant Program (2010) that has been sponsoring the transition of state vehicles from diesel to electric. The push to switch from diesel to electric has also been seen on the public transportation level in Philadelphia. Beginning in 2016, the Southeastern Pennsylvania Transportation Authority (SEPTA) began replacing diesel buses with hybrid-electric buses; this transition will be fully completed by 2021. A study in the Netherlands found that particle number count concentrations were highest in diesel buses and lowest in electric buses [63]. Promoting transitions like these in the transportation sector can reduce PM emissions.

The transition from coal to natural gas or alternative clean energy sources will help to reduce PM emissions. Since 2007, Philadelphia has decreased the carbon intensity of the grid by 26% [57]. To continue lowering the carbon intensity of the grid, Philadelphia has outlined a plan up to 2050 that includes biomass burning in its renewable energy profile [57]. As of 2020, Pennsylvania is among the top dozen producers of electricity by biomass [48]. Though coal usage has decreased in the region, it is important to monitor how using biomass burning as a renewable replacement will impact EC and OC, and overall PM concentrations going forward.

While regulation on sources like vehicles and coal have been proven effective, further studies on specific sources, like commercial cooking, in the region would help to target sources that need to be more regulated. Analysis on specific air pollutants such as EC, OC, SO_x, and NO_x may help to identify local sources [60].

4.5 PM emission sources

The NEI released sector and county organized data for PM emissions every three years from 2008–2020. Sectors include mobile (non-road and on-road), agriculture, dust, fuel combustion, industrial processes, miscellaneous, solvent, and waste disposal [37]. Mobile non-road and on-road, industrial processes, fuel combustion, and dust experienced decreasing trends from 2008 to 2020. Over the 12-year period, major reduction in PM emissions occurred for dust, fuel, industrial and mobile sources (Fig. 6). These decreases in dust, fuel, industrial, and mobile sources follow from policies focusing on these sectors leading up to and during this period (Clean Air Interstate Rule, PA Clean Vehicles Program, Regional Greenhouse Gas Initiative, etc.). However, there was an increase in PM emissions for commercial and agricultural sources. The commercial sector, which only includes commercial cooking, increased significantly from 2008. With the limited amount of policy regarding commercial cooking, this sector is an unexplored opportunity to reduce PM emissions. PM emissions from cooking depends on the type of cooking process (i.e. frying, roasting, broiling, etc.), ingredients, fuel types, temperature, and ventilation equipment [64]. PM from cooking is considered the possible causes for higher PM emissions in another urban city in Pennsylvania (Pittsburgh), where meat charbroiling was found to contribute to carbonaceous PM emissions [65]. A study in New York City found cooking and traffic to be two distinct organic aerosol source contributing 30% of the total organic aerosol mass collected [39]. With the main emission sectors showing decreasing trends after targeted policies were enforced, it may be time to shift policy direction towards more prominent and unaddressed sources like commercial cooking. Another interesting observation was the large increase in the agriculture sector (Table S2).

4.6 Trends and policies in other geographical regions

Urban environments typically have many emissions sources, including motor, maritime, airport, industry, incineration, and biomass burning [6]. Similar to Philadelphia, Rochester, New York has a typical urban emissions profile, and attributes some of its air quality improvements to the US EPA Tier 1 and 2 Emissions standards [6].

An air pollution study conducted in the state of Maryland [3], showed the influence of regulations on reducing sulfate pollution. The Maryland Healthy Air Act (2006) reduced sulfates from power plants and reduced PM concentrations especially in the summer months compared to other neighboring states, including Pennsylvania [3]. The benefits of energy regulations in Maryland should suggest policies for the Philadelphia region. Adding more energy regulation in proportion to vehicle regulations will target gaps in Philadelphia's policy profile that led to PM, EC, and OC spikes.

The State of California has some of the highest concentrations of PM in the US and highest levels of PM-related deaths [5]. The study by [5] reported that the PM regulations target mass concentration largely; so, this study is suggesting that specific emission sources are important considerations when designing emissions standards. Like Philadelphia, vehicular emissions contribute largely to PM in California in most areas, while in other areas biomass burning dominates $PM_{2.5}$ concentrations, especially in winter [5]. The same study found that vehicle emissions are higher in fall and are shown by EC and OC. These findings are similar to our conclusions.

There has been great improvement in air quality in the US due to the reduction in emissions produced by electric power plants and industries [6]. Air pollution regulations helped to reduce SO_2, black carbon and primary organic aerosol emissions by 80%, 40%, and 30%, respectively in the continental US from 1980 to 2014 [61]. Air pollution regulations helped to reduce $PM_{2.5}$ mass and sulfur concentrations in Boston, Massachusetts from 1999 to 2018 [41]. Secondary sulfate and secondary nitrate of PM were reduced in Rochester, New York due to the various policies related to curb

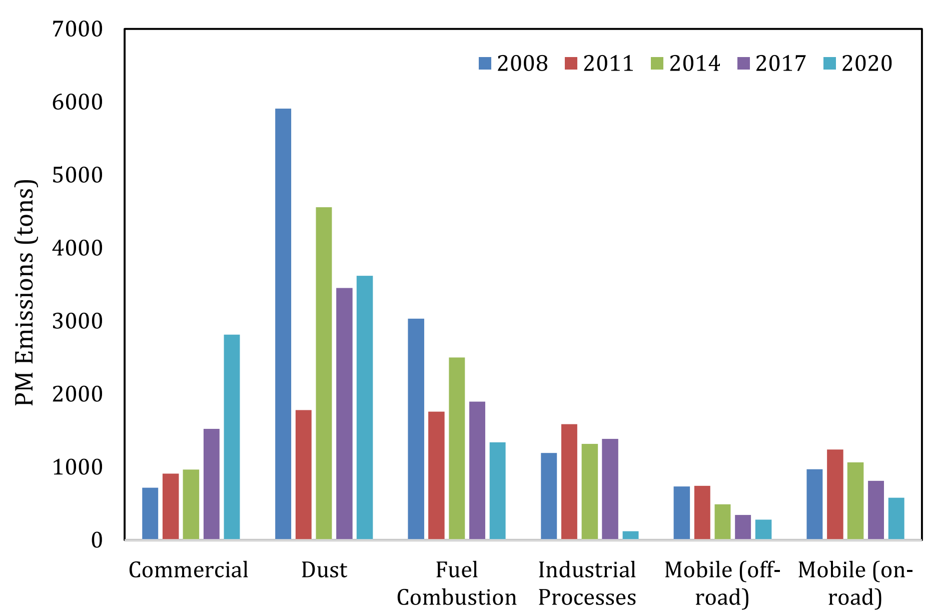

Fig. 6 PM emissions in Philadelphia based on the National Emissions Inventory. Sources fall under 10 main sectors: agriculture, commercial, dust, fuel combustion, industrial processes, miscellaneous, mobile (on and off road), solvent, and waste disposal. Only six major sources are shown here. Please see supplementary section for the complete table (Table S2)

emissions from power plants and vehicles [6]. In the same city of Rochester, New York, monthly $PM_{2.5}$ concentrations decreased by 41% from 2001 to 2015 [40].

4.7 Limitations

Although we attributed the success in PM reductions in Philadelphia to environmental policies on various levels, these could also be contributed by various technological advancements. Therefore, this study does not distinguish the difference in the effectiveness of environmental policies and technological advances for PM reductions. The list of environmental policies may also not be a complete list. Another limitation of this study is that statistical tests were not conducted to determine the effectiveness of any specific environmental policies in reducing PM. In addition, meteorology and long-range transport also plays an important role in PM formation and these are not discussed in this study. Future papers can work on these limitations and also on which level the environmental policies may be the most effective: state level, federal level, or local level. The number of observations vary around the years, which could skew the yearly and seasonal averages we described in this manuscript. For $PM_{2.5}$, we included all observations regardless of the methods. Future studies can study if there is the influence of measurement methods on $PM_{2.5}$. This manuscript relies on one-year averages, and it may be subject to atmospheric conditions. Ideally, in future studies, the moving yearly average can be used to analyze the trends.

5 Conclusions and recommendations

In Philadelphia, the annual mean PM_{10} has decreased by 47% from 1986 to 2021 and annual mean $PM_{2.5}$ has decreased by 31% from 2000 to 2021. PM_{10} and $PM_{2.5}$ showed seasonal variability with high PM occurring in summer months except for the recent decade of 2011–2020, when PM_{10} showed high concentration during winter months. EC and OC also show a decreasing trend although EC shows a brief increase in recent years. Several national policies such as the National Low Emissions Vehicle Program (1999), the Reformulated Gasoline Program (1995), and the Clean Air Non-Road Diesel Rule (2004) that relates to transportation sector has helped to reduce PM concentrations in Philadelphia. Declines in PM concentrations were observed after passage of such policies.

As drawn from the decreased average monthly concentrations of air pollutants examined in this study—PM, EC, and OC—the early- to mid- 2000s appears to be the period of maximum effectiveness in Philadelphia (Figs. 2 and 4). This period coincides with many transportation-related policies as noted above. Policies slightly before the 2000s and during the 2000s most likely led to the lowest concentrations seen beginning in the 2010s like the Reformulated Gasoline Program (1995), Low Emissions Vehicle Program (1999), revised PM standards, and others (Fig. 5). It is important to remember that the effects of policies happen over time, and so lag time from implementation should be considered. Compared to transportation (> 30% reduction for both on- and off-road mobile PM emissions), there is less reduction for industrial sector (10%) and increase in agriculture and commercial sectors. Shifting of PM_{10} peaks from summer to winter in the recent decade suggests the need for regulations targeting winter emission sources.

To further reduce particle pollution in the Philadelphia region, we recommend that vehicle emissions continue to be regulated and their standards continue to be stricter because this has proven effective in lowering PM concentrations overall. Promoting the transition of gasoline or diesel-powered vehicles to electric vehicles, especially in public transportation sectors, could greatly reduce PM emissions and can also help to reduce greenhouse gas emissions. Besides the transportation sector, the transition from coal to renewable energy sources can also help to reduce PM emissions and can also benefit climate change mitigation goals.

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Declarations

Competing interests The authors declare no conflict of interest.

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