Temporal trends of particle gross β-activity and PM$_{2.5}$ mass concentrations in the USA during 2001–2017

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Abstract

Recent studies conducted in the USA have shown that adverse health effects of fine particulate matter (PM$_{2.5}$) persist at levels below the national air quality standards. More recently, particle radioactivity has also been associated with adverse health effects. However, the importance of particle radioactivity at low PM$_{2.5}$ levels has not been thoroughly explored. The present paper investigates the temporal trends and the relationship of particle gross β-activity (PM-β) and PM$_{2.5}$ mass in the 48 states of the contiguous USA during the period of 2001–2017. With the implementation of stringent air pollution control policies, national ambient PM$_{2.5}$ concentrations decreased by 38.5% during this period. However, a smaller decrease of 9.4% was observed for PM-β, while the mean PM-β/PM$_{2.5}$ ratio increased by 49.1%. PM-β is mostly associated with radon emissions and its progeny, which do not change much with time. The largest PM$_{2.5}$ and PM-β reductions were observed in the Southeast, while the smallest were found in the West. When the aggregated PM-β to PM$_{2.5}$ ratio is stratified by PM$_{2.5}$ levels, the ratio was found to be highest when PM$_{2.5}$ is <3 μg/m^3, with a median PM-β to PM$_{2.5}$ ratio of 0.77 (0.64–0.88; 25th–75th percentiles). Overall, when not stratified by PM$_{2.5}$ levels, the greatest state-wide overall PM-β /PM$_{2.5}$ ratios were found in Wyoming (0.69) and South Dakota (0.51), areas with higher radon, while the lowest (0.17) were in Delaware followed by New Jersey (0.18). These results indicate that the ratio of ambient particle radioactivity to particle mass concentration typically is higher at low PM$_{2.5}$ levels, and consequently, the toxicity per unit mass is expected to be higher.

Keywords Air quality · Ambient PM$_{2.5}$ · Particle radioactivity · Toxicity · Trend analysis

Introduction

Exposure to PM$_{2.5}$ (concentration of airborne particles with diameters less than 2.5 μm) is an important environmental risk. According to the Global Burden of Diseases study (Cohen et al. 2017), it ranks quite high among the avoidable causes of non-communicable diseases and presents a critical exposure risk factor (Murray et al. 2020). PM$_{2.5}$ air pollution is classified as a group 1 human carcinogen by the World Health Organization International Agency for Research on Cancer (IARC 2013) and is arguably the largest environmental risk to public health, responsible for over 7 million pre-mature deaths annually (WHO 2021). PM$_{2.5}$ is a complex mixture with varying particle size and chemical composition, which varies across space and time. Ambient PM$_{2.5}$ can be both directly emitted by sources (primary) or formed from photochemical reactions (secondary) (Harrison 2020). Particle radioactivity (PR) is a radiometric characteristic of airborne particles and is attributed to radionuclides bound on their surfaces.

Radionuclides are present in the atmosphere due to anthropogenic, cosmogenic, and terrestrial sources (Papastefanou 2010; Dlugosz-Lisiecka 2016). In the absence of anthropogenic sources, radioactive radon decay products are the most important contributors to PM-β. Radon decay produces radioactive nanoparticles, which rapidly attach to particles in the accumulation mode (Liu et al. 2020). In turn, these radionuclides gain the properties of the carrier particles (Reineking and Porstendorfer 1990; Papastefanou 2012) and may contribute to the overall toxicity of PM$_{2.5}$.

Inhaled particle-bound radionuclides deposit onto the lung airways and, subsequently are circulated through the

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body, where they continue to decay and emit radiation, generating an internal dose of α, β, and γ radiation, causing cell damage and chronic inflammation (Nie et al. 2012). Recent studies have reported associations between PM-α, PM-β, PM-γ, and telomere length (Scherthan et al. 2016), neurodegenerative diseases, inflammation, impaired lung function, oxidative stress, and elevated blood pressure (Nyhan et al. 2018, 2019; Santos et al. 2020). While emission control policies and regulations have been effective at reducing ambient $PM_{2.5}$, radon is a naturally occurring gas whose emission rates are geologically determined so its ambient concentrations have remained relatively stable. This study explores the temporal trends of PM-β and $PM_{2.5}$ concentrations across the 48 contiguous states of America during 2001–2017 and focuses on the spatiotemporal patterns of the PM-β/$PM_{2.5}$ ratio under changeable $PM_{2.5}$ levels.

Materials and methods

$PM_{2.5}$ data

Daily $PM_{2.5}$ data were obtained from the Environmental Protection Agency (EPA) Air Quality System (AQS) (https://www.epa.gov/aqs) that reports ambient air pollution data collected by EPA, state, local, and tribal air pollution control agencies from over a thousand monitoring sites. Information about monitoring stations including geographic location, classification, monitoring methods, and data quality assurance/quality procedures is also reported by AQS (https://www.epa.gov/aqs/aqs-manuals-and-guides). The $PM_{2.5}$ data that we included was collected using a variety of methods, and data were selected based on the county of each city. The first option was to include hourly $PM_{2.5}$ measurements from continuous monitors (normally beta attenuation, light scattering, or laser absorption instruments). In the absence of hourly measurements, we included $PM_{2.5}$ integrated daily averages that were measured using gravimetric techniques that determine $PM_{2.5}$ levels from collected air filters. For cities with more than one sampling site, the following procedure was followed to standardize daily measurements for all monitors within a city boundary and prevent missing days from one monitor from adding false variability to the daily value (Zanobetti and Schwartz 2009): (1) daily deviations from the annual mean for each monitor were calculated; (2) each monitor's daily deviations were standardized by dividing by its annual standard deviation; (3) mean daily standardized deviations were calculated for each city by averaging the daily standardized deviations for all monitors assigned to the city; (4) multiplied this mean daily deviation value by the standard deviation of all monitors within the city, and added back the annual mean of all monitors within the city.

PM-β data

The US EPA RadNet is a national monitoring network for particle-phase environmental radiation in ambient air (Fraass 2015). We obtained PM-β data from 129 monitors with more than 265 days for every year (Blomberg et al. 2019) or at least 1 year of monitoring data (when the city had 2 or more monitors) during the study period of 2001–2017, and the same procedure as described above (see "$PM_{2.5}$ data") was carried out to calculate city values. The monitors at each site use high volume air samplers equipped with polyester fiber filters to collect total suspended particles (TSP) over a 24-h period. The filters are sent twice a week to the National Analytical Radiation Environmental Laboratory for analysis to measure PM-β (USEPA 2012). The gross beta analysis was performed using a gas flow proportional detector. This type of detector measures beta radiation by counting the number of ionizing events that occur within a gas-filled chamber. As beta radiation released from the sample passes through the chamber, it ionizes the gas molecules, creating a photon that can be detected and counted by the instrument. Despite using TSP and not $PM_{2.5}$ in PM-β (in this study), Liu et al. (2020) showed that measurements of PM-β from the RadNet TSP samples showed only slightly smaller concentrations than co-located filter samples of coarse and fine particles, indicating that most beta emissions come from the accumulation mode (particle sizes below 2.5 microns). It should be noted that the filters are analyzed offline, 1 to 2 weeks after collection, so that practically almost all the short-lived radon progeny have decayed. Therefore, most of the measured PM-β is related to longer-lived radon progeny. In addition, while PM-β is also associated with other radionuclides such as Potassium-40 and Uranium-238, and Berrilum-7, their contributions to the measured values are expected to be negligibly low.

Data analysis

Data were processed and displayed using R version 4.2.0 (R Core Team 2022) using R packages ggplot2 (Wickham 2011), openair (Carslaw and Ropkins 2012), and mcgv (Wood 2003). The statistical method followed for trend calculation is a non-parametric Mann-Kendall approach. The trend slope was calculated with the Theil-Sen (Theil 1950; Sen 1968) method available in the R-openair package. Briefly, for a given set of n x, y pairs, the slopes between all pairs of points are calculated and the median was taken as an estimate of the most probable slope (trend). This method is robust to outliers and

can be used in both non-normal and heteroscedastic (non-constant error variance) data series. For the trend analysis, the data was de-seasonalized by applying the smoothing locally estimated scatterplot smoothing (LOESS) function which is a non-parametric regression method used to fit a smooth curve to a scatterplot of data (Cleveland et al. 1990). LOESS is a popular technique for exploratory data analysis and can be used to identify trends and relationships between variables (Matthaios et al. 2019). The LOESS function works by fitting a series of local regression models to subsets of the data, where each subset consists of a small number of nearby data points. We also applied bootstrap re-sampling for the calculation of confidence intervals at the 95% level and p-values. A trend was statistically significant when $p < 0.05$ (represented with a "*" symbol), meaning that the trend was not random at a 95% probability; $p < 0.01$ and $p < 0.001$, marked by "**" and "***," respectively, indicate very highly significant trends, while $p > 0.05$ and $p < 0.1$ indicate partly significant trends.

Results and discussion

Figure 1 shows the aggregated (across the 48 contiguous states) normalized trend of PM-β, PM$_{2.5}$, and PM-β/PM$_{2.5}$ annual ratios for 2000–2017. Figures S1–S4 show the trends in West, Midwest, South, and Southeast USA, while

Tables S1–S3 show the state trends of PM$_{2.5}$, PM-β, and PM-β/PM$_{2.5}$ ratio for different states. On average, PM$_{2.5}$ levels decreased by 38.5% over the study period. This is consistent with EPA assessment that the national annual PM$_{2.5}$ concentrations declined on average by 42% between 2000 and 2016 (USEPA 2022). This decline was due to the stringent air quality standards implemented over the last 20 years. Note the sharp decrease (~15%) in PM$_{2.5}$ from 2007 to 2009. This sharp reduction is most likely caused by the 1997 EPA NAAQS revisions to apply both annual and 24-h standards for PM$_{2.5}$. A further sharp decrease of 20% in PM$_{2.5}$ levels was observed between 2011 and 2017. EPA again revised the 24-h NAAQS from 65 to 35 μg/m^3 in 2006, which resulted in reductions across the USA since more counties followed national pollution standards as of 2007 compared with previous years (USEPA 2010). We observed the largest PM$_{2.5}$ decreases (50%) in the southeastern USA (Fig. S1) most likely due to stricter emission reduction policies (Zhang et al. 2018), while a smaller PM$_{2.5}$ reduction (28.3%) observed in the Western USA (Fig. S3) most likely due to the impact that wildfires have on the ambient PM$_{2.5}$ (McClure and Jaffe 2018).

In contrast to the large reductions in PM$_{2.5}$ levels across the USA, PM-β levels exhibited only a modest decline. From 2001 to 2017, PM-β dropped by 9.4% on average across the USA. During the study period, while the PM-β/PM$_{2.5}$ ratio increased by 49.1%. The smallest and largest increases in the PM- β/PM$_{2.5}$ ratio were observed for the Southern

Fig. 1 US aggregated times series of PM-β, PM$_{2.5}$ mass, and their ratio. The times series were normalized using 2001 as the base year and were set equal to 100. The brackets indicate the confidence interval of the slope. ** indicate trend significance at $p < 0.01$.* indicates significance at $p < 0.05$

3.32** (2.91,3.70)% y^{-1}

-0.91* (-1.02,-0.79)% y^{-1}

-2.52** (-2.63, -2.37)% y^{-1}
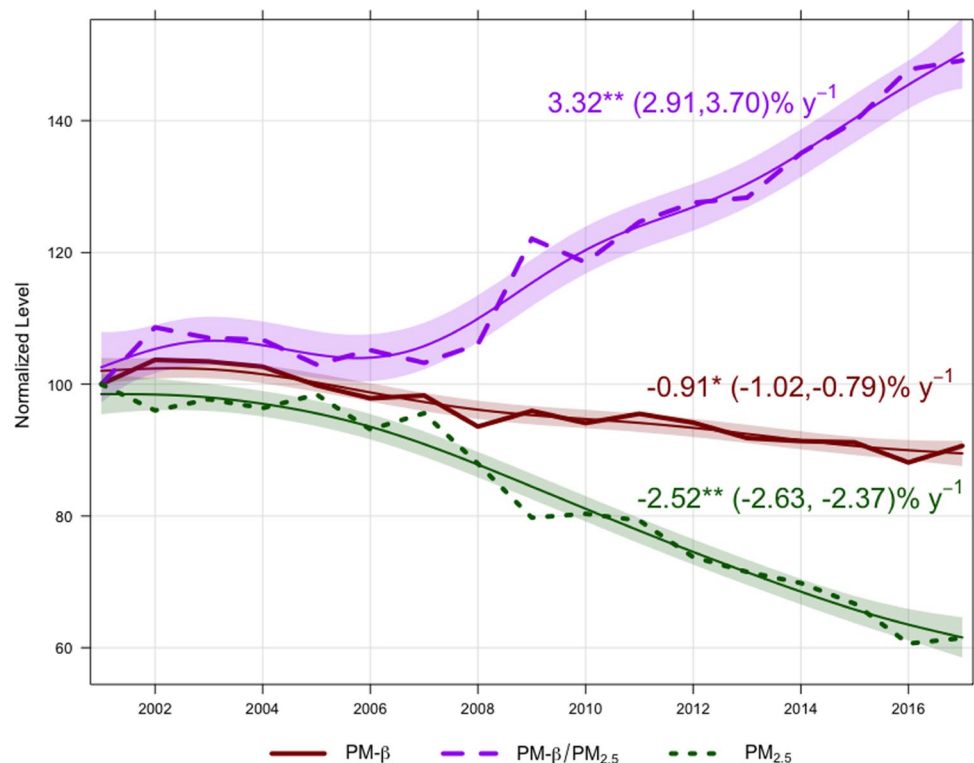
PM-β   PM-β/PM$_{2.5}$ PM$_{2.5}$

Fig. 2 Distribution of monthly PM-β/PM$_{2.5}$ ratios corresponding to different PM$_{2.5}$ level ranges aggregated across the USA during 2001–2017. The figure shows interquartile ranges (IQR), the horizontal line is the median, and whiskers extend to 1.5 times the IQR above the 75th and below the 25th percentiles. Dots are values beyond 1.5 times the IQR
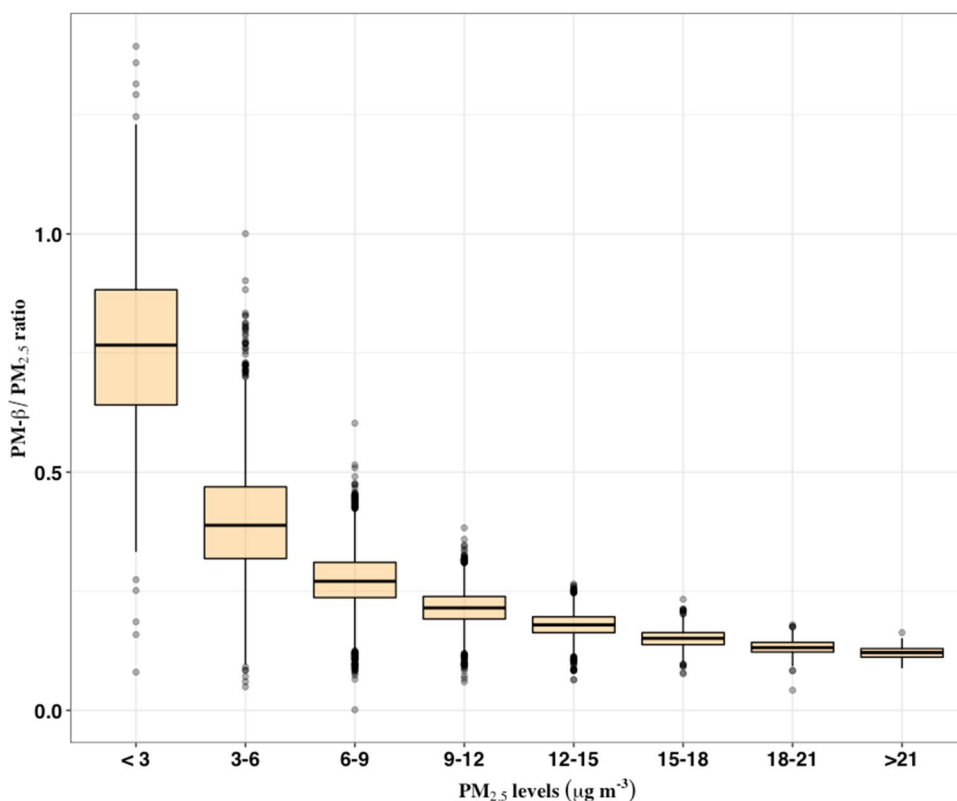
(35.5%) and Midwestern (56.6%) regions, respectively. The Midwest includes states with relatively high background radon concentrations, such as South Dakota, Ohio, Iowa, Wisconsin, and Illinois. Consequently, despite having the second highest PM$_{2.5}$ concentration decreases in this region, PM-β activity only dropped by 8%, which was the second lowest. In contrast, the Southern region, despite having only the second lowest PM$_{2.5}$ decreases, had the second largest decrease in PM-β. Overall, the trend analysis showed that PM$_{2.5}$ levels dropped significantly ($p < 0.01$) by 2.5% (CI: 2.63, 2.37) per year, while PM-β levels decreased by only 0.90% (CI: 1.02,0.79) per year, as shown in Fig. 1. Therefore, PM$_{2.5}$ dropped 2.8 times faster than PM-β activity, most likely due to the actions taken to control PM$_{2.5}$ emissions.

Figure 2 shows the aggregated monthly PM-β/PM$_{2.5}$ ratios for different PM$_{2.5}$ levels. The ratio is greater for lower PM$_{2.5}$ levels, e.g., PM$_{2.5}$ concentrations <3 μg/m^3. The median ratio is 0.77 (25th–75th: 0.64–0.88) and is two times greater than that for the 3–6 μg/m^3 PM$_{2.5}$ range of 0.39 (25th–75th: 0.32–0.47). Table S4 shows the ratio for each state. The greater PM$_{2.5}$ reduction compared to that of PM-β is understandable, since pollution control policies do not impact radon emissions. As a result, the amount of

PM-β per PM$_{2.5}$ mass has increased. Based on our health effects studies reported above, increasing the amount of radioactivity per particle mass may enhance particle toxicity. However, PM$_{2.5}$ controls have also helped to reduce PM-β to some extent. As PM$_{2.5}$ surface concentration decreases, more of the unattached radon progeny can be removed from the atmosphere through dry deposition and to some extent by wet deposition. A similar phenomenon takes place in indoor environments where more unattached radon progeny diffuses to the walls and furniture (plate out) when indoor PM$_{2.5}$ are low (Matthaios et al. 2021).

The steady decline in PM$_{2.5}$ concentrations in the USA has helped to improve public health (Schraufnagel et al. 2019). However, recent studies have found adverse PM$_{2.5}$ effects even below the NAAQS standards with no threshold level (Di et al. 2017; Yazdi et al. 2021; Shi et al. 2022). This suggests that there might be toxic components which are still present at low PM$_{2.5}$ levels. As shown in Fig. 2, the higher PM-β/PM$_{2.5}$ ratios observed at lower PM$_{2.5}$ concentrations suggest that these particles have relatively more radioactivity per unit mass. Therefore, future studies should test the hypothesis that particle radioactivity enhances PM$_{2.5}$ toxicity.

Declarations

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