Understanding Anthropogenic Impacts on Air Quality Through Space and Time

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Despite continued actions to abate harmful air pollutant emissions, air pollution is still a worldwide concern, yet apportioning individual shares of responsibility for pollution is challenging, particularly over extended periods of time. The spatial and temporal changes in particulate matter (PM) emissions related to a steel manufacturing plant in Middletown, Ohio, were investigated using microscopic, elemental, and Pb isotope approaches. For temporal analysis, two sets of 12 deciduous leaves, as passive samplers, collected from a park in Middletown in 1961 and 2022 were compared. For spatial analysis, evergreen leaves were collected at varying distances from the steel plant within a diameter of approximately 70 km.

Regarding temporal changes in elemental pollution in PM over time, the pollution load index (PLI) indicated significantly higher concentrations of pollutants in 1961 (3.49 ± 1.30) compared to those in 2022 (1.92 ± 1.18). Analysis of Pb isotopes revealed that the average relative contributions of PM originating from glacial till, the steel plant, gasoline, and fly ash in 1961 were 38 ± 7 , 21 ± 3 , 32 ± 9 , and $9\pm1\%$, respectively, while the respective values in 2022 were 33 ± 3 , 33 ± 6 , 20 ± 3 and $14\pm2\%$. This suggests a shift in the main anthropogenic source of PM from gasoline in 1961 to the steel plant in 2022. Additionally, PLI exhibited a significant correlation with the estimated relative contributions from Pb isotopes. Overall, the quantity and source of pollution have undergone drastic changes over time.

As for contemporary spatial trends, Pb isotopes indicated that the average PM masses originating from glacial till, steel plant, gasoline, and fly ash were 44 ± 23 , 34 ± 30 , 33 ± 17 , and 18 ± 11 mg m⁻², respectively, highlighting the steel plant and gasoline as the primary anthropogenic PM sources. Strong correlations between steel spherule mass estimated by Pb isotopes and its relative proportion quantified through microscopic investigations and PLI provided support for source apportionment using isotopic methods. The steel spherule quantity decreased exponentially with distance from the plant, with the steel plant's effective PM footprint extending approximately 32 and 40 km upwind and downwind, respectively. These findings underscore the continued environmental impact of the steel plant, despite observed decreasing trend of PM emissions over the last 60 years.