communications earth & environment

ARTICLE

https://doi.org/10.1038/s43247-023-01135-3 OPEN

A lingering legacy of leaded gasoline in Southeast Asia

Iravati Ray[1] & Reshmi Das[1,2]✉

Recent reports of 1 in 3 children globally having blood lead levels ≥ 5microgram decilitre^{-1} demands thorough understanding of lead (Pb) sources of the present century and the fate of legacy Pb from the past use of leaded gasoline. The present hotspot of pollution is South and Southeast Asia. To investigate this issue, here we compile Pb isotopic compositions of aerosols ($n = 341$) along with established and previously excluded sources for Singapore, Thailand, Vietnam, and India. The data was subjected to Bayesian 3D isotope mixing model simulation. Model estimates reveal consistent contributions from natural background. Leaded gasoline is the largest contributor in Southeast Asia (39%). Tertiary coal/fuelwood combustion and ore processing dominate in India, while ship emission contribute up to 15%. Thus, along with Pb from present sources, the historic use of leaded gasoline left a legacy of Pb in soil which is remobilised to the atmosphere after more than two decades of its phase-out.

[1] School of Environmental Studies, Jadavpur University, Kolkata, India. [2] Earth Observatory of Singapore, Nanyang Technological University, Singapore, Singapore. ✉email: reshmidas.sest@jadavpuruniversity.in

The use of leaded gasoline dates back to the early 20th century when tetraethyl lead (TEL) was first introduced as a petroleum additive to improve the performance of internal combustion engines. During the combustion process, gasoline lead (Pb) was partially oxidised and PbO deposits caused engine fouling. To overcome this issue, dichloroethane and dibromoethane were added to leaded gasoline to act as Pb scavengers. The Pb oxides were converted to volatile Pb species such as lead chloride and lead bromide that were emitted into the atmosphere. Once emitted, Pb has an atmospheric residence time of 5–10 days[1] before getting deposited. The Pb compounds emitted from the vehicle exhaust spread to the remote locations of the planet including Antarctica, Greenland, Alaska, and the middle of the oceans[2,3]. Patterson's identification of the automobile tailpipe as the environmental Pb source causing elevated blood lead level (BLL), endemic in the United States led to his fight against the petroleum lobby to ban TEL additive in gasoline[4]. His efforts met with success in 1972 when USEPA proposed to phase out leaded gasoline. US leaded gasoline phasing out was completed in 1986 and it took 20 years for another 120 countries including Indonesia, Australia and South Africa to achieve the target. Post 2006 only five countries (Algeria, Yemen, Afghanistan, Iraq, and Myanmar) were using leaded gasoline, and in 2021, Algeria became the last country to ban leaded gasoline marking the success story of a global collaboration. However, along with the feat, a grave concern about the fate of the already released Pb compounds from automobile exhaust loomed.

The atmosphere is the first recipient of the gasoline exhaust in the form of volatile Pb halogenides[5]. After several days the emitted Pb compounds settle on the top soil. Residence time of Pb in soil is 100–200 years[6]. Hence it was presumed that the legacy Pb from the global use of leaded gasoline will prevail in the environment. Indeed, a strong evidence of recirculating legacy lead in the atmosphere from soil resuspension were reported from different corners of the world. The fingerprinting of the source was done by the immutable Pb isotopic signature[7–10]. Pb has four naturally occurring isotopes out of which ^{206}Pb, ^{207}Pb, and ^{208}Pb are radiogenic. ^{204}Pb is the least abundant and the only non-radiogenic Pb isotope. Hence the difference in isotopic signature of Pb bearing minerals arises from the relative proportion of initial U–Th–Pb in the system and radioactive decays of ^{238}U, ^{235}U, and ^{232}Th. The relative atomic weight differences between the isotopes of Pb are minimal resulting in minimal mass-dependent Pb isotope fractionation in natural physical, chemical, and biological processes[11–13]. Pb isotopes are an effective tool for tracing Pb pollution as they do not fractionate during industrial or environmental processes and preserve the source signature even after degradation, processing, and transportation.[14].

South and South East Asian countries have recently come under scrutiny of Pb pollution due to several reasons. In India, Thailand, and Vietnam, more than 250 million children have blood lead level > 5 microgram.decilitre^{-1} (ug dL^{-1})[15]. Additionally, present day Pb emissions have surpassed past century emission from leaded gasoline used in these countries[16]. Lastly, several countries including Indonesia and Myanmar in the region were using leaded gasoline until this century.

Recent source apportionment studies of atmospheric Pb over Singapore, Thailand, Vietnam (referred to as SEA countries herein), and India by utilising Pb isotopic compositions and elemental ratios of aerosols[7,10,17–23] have identified the potential sources to be crust, sea spray, coal combustion, high temperature industrial activities, vehicular and ship traffic, solid waste incineration and biomass burning. In addition to these modern sources, historic Pb deposited on top soil from leaded gasoline emissions still recirculates in the atmosphere[7,9,17]. As Indonesia used leaded gasoline until 2006[24], and Myanmar until 2016[25], historic Pb recirculation may contribute substantially towards atmospheric Pb in the region. All past studies considered linear 2 or 3 endmember mixing models that may lead to over-simplification of the dynamics of atmospheric Pb pollution by exclusion of potential contributors. To overcome the issue of multiple end members, Bayesian stable isotope mixing models such as MixSIR, SIAR, and MixSIAR has been widely used in the field of ecology to study food webs and infer the diets of consumers based on stable isotope systems such as δ^{13}C, δ^{15}N and δ^{34}S[26,27]. Recently, MixSIAR has also been used in few source apportionment studies concerning Pb pollution that utilises Pb isotopic compositions of pollution sources[28–32]. Isotopic systems such as δ^{13}C, δ^{15}N, and δ^{34}S permit the integration of concentration data within the MixSIAR framework, owing to the distinct concentrations associated with each element. For Pb isotopic system also the concentration-dependent model can be utilised where including concentration data results in identical concentrations for each ratio However, it may not be effective to execute concentration-dependent mixing models by utilising Pb concentration data solely, as it has large orders of magnitude variations across sources. Some of the sources may have concentration in percentage range compared to ppm or ppb range of other sources, that will introduce bias in model results. Thus, to avoid such biases, techniques, such as normalisation is useful. Normalisation of Pb concentration of the sources with a crustal element such as Al, Fe, Ti, etc can be an effective strategy. This study aims to perform a quantitative retrospective analysis of Pb isotope data of aerosols from the past two decades and their potential endmembers using mixing polygon simulation followed by MixSIAR analysis to distinctly understand the sources of atmospheric Pb. Utilising MixSIAR for source apportionment of atmospheric Pb over SEA countries and India will lead to discerning the sources that require revisiting and identification of any probable underdetermined source that should be prioritised in future studies.

Results and discussion

Identifying contributing sources. Despite some overlaps, the aerosols of (a) India, (b) Singapore & Thailand and (c) Vietnam plot in 3 distinct regions, with Singapore and Thailand overlapping with each other in ^{206}Pb/^{207}Pb vs ^{208}Pb/^{207}Pb space (Fig. 1a). The aerosols also exhibit a similar arrangement in ^{206}Pb/^{204}Pb vs ^{208}Pb/^{204}Pb space (Supplementary Fig. 1). This indicates that atmospheric Pb is either sourced from different endmembers in different regions or common sources have variable contributions in different countries. The sources are distributed along the diagonal of the three-isotope space, with certain sources bracketing the aerosol samples and others positioned at intermediate points.

When analysing in ^{206}Pb, ^{207}Pb, and ^{208}Pb space, the median probabilities of the aerosols to fall inside the mixing envelope formed by these sources were 32%, 36%, 44%, and 40% for Singapore, Thailand, Vietnam, and India respectively when the conventional endmembers (coal from India, Indonesia, Vietnam and south China, SEA and Indian ore, unleaded and leaded fuel, solid waste and biomass burning in SEA and crust) were considered[7,17,18,33–43](Fig. 1b). Analogous probabilities were obtained in ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb space, which is provided in Supplementary Fig. 2. The data for probabilities of all the aerosols are provided in Supplementary Data 1 [44]. These low median probabilities observed in both the isotope spaces for all the four countries indicates that there must be sources that were not considered in previous studies.

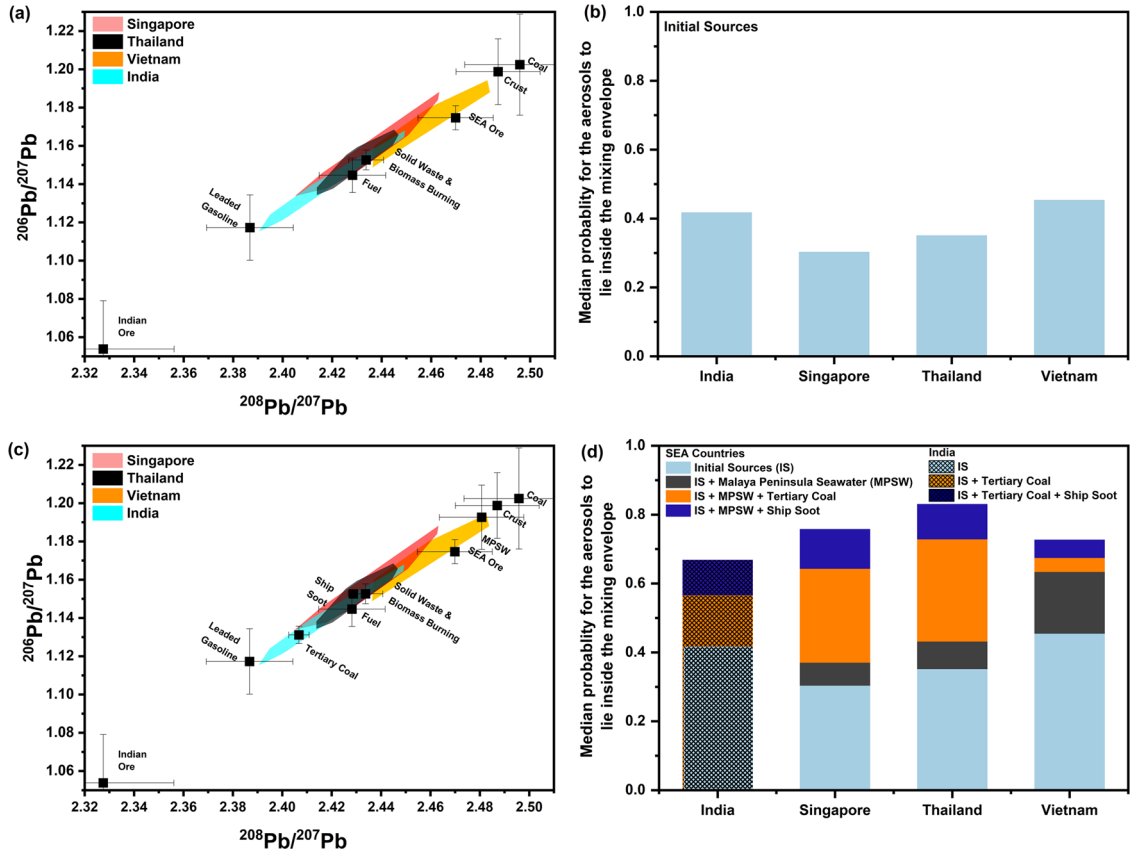

Fig. 1 Aerosol mixture and sources in three isotope space and their probability to lie inside the source defined mixing envelope. a Three—isotope plots of the aerosol mixtures from India, Singapore, Thailand, Vietnam, and their initial sources from previous literatures in $^{208}Pb/^{207}Pb$ vs $^{206}Pb/^{207}Pb$ space. The error bars signify 1 SDs of the sources and **b** Bar plots depicting the median probability of the aerosol mixtures to fall inside the mixing envelope constructed by the initial sources, determined with the help of 3 - dimensional mixing polygon simulation in $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ space **c** Three—isotope plots of the aerosol mixtures from India, Singapore, Thailand, Vietnam and their initial sources along with the additional sources in $^{208}Pb/^{207}Pb$ vs $^{206}Pb/^{207}Pb$ space and **d** Bar plots depicting the median probability of the aerosol mixtures to fall inside the mixing envelope constructed by the initial and additional sources, determined with the help of 3—dimensional mixing polygon simulation in $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ space. The aerosol mixture data for respective countries are depicted as fields, encompassing all the data points.

Based on elemental ratios calculated from elemental concentration data provided in previous investigations, it is observed that aerosols in Singapore, Thailand, and Vietnam closely align with the typical Na/Mg value of seawater (~8) (Fig. 2a)[20,21,35,45]. The coastal waters around Malaya Peninsula have a wide range of Na/Mg values that overlap with that of the aerosols from Singapore, Thailand, and Vietnam (Fig. 2a)[46–48]. Moreover, the MPSW has higher concentration of Pb (range = 24.8–368 pmol kg^{-1}) compared to the average Pb concentration in South China seawater (67.2 pmol kg^{-1}) and Bay of Bengal (68.1 ± 10.52 pmol kg^{-1})[16,49]. Seawaters in the northern (North Atlantic, Pacific, and Antarctic surface waters) and southern hemisphere (East Australian coastal shelf waters) also have lower Pb concentration ranging from 12 to 160 pmol.kg^{-1}[50]. This suggests that the aerosols may exhibit some degree of influence from seawater in addition to other predominant sources. Similarly, the V/Ni ratios in aerosols from India, Singapore, and Vietnam demonstrate characteristics of heavy oil and ship emissions[18,23,35,36,51–54] (Fig. 2b). Tertiary coal from India exhibit a distinctively lower $^{206}Pb/^{207}Pb$[7] composition and has not been used as an end member in previous studies. The comparable isotopic composition of tertiary coal, wood, and wood charcoal[7], coupled with India's substantial dependence on fuelwood[55], makes it necessary to consider this as an end member. Thus, it is imperative that all these additional sources are considered while calculating the contribution of each source towards the aerosol of these countries. All the initial sources and additional sources plotted in $^{206}Pb/^{207}Pb$ vs $^{208}Pb/^{207}Pb$ three—isotope space are presented in Fig. 1c. The three—isotope plot in $^{206}Pb/^{204}Pb$ vs $^{208}Pb/^{204}Pb$ space is presented in Supplementary Fig. 3. Incorporation of these previously excluded sources (sea spray, tertiary coal and ship soot) increased the area of the mixing envelope and consequently the median probabilities for the aerosols to lie inside the mixing envelope in $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$ space to 76%, 83%, 73% and 67% for Singapore, Thailand, Vietnam, and India respectively (Fig. 1d). The results obtained from the polygon simulation performed in $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ space are discussed in Supplementary Note 1 (Supplementary Fig. 4). Data is provided in Supplementary Data 1 [44].

On performing Analysis of Variance (ANOVA) at 0.05 significance level, on the probability results, it was observed that the addition of the MPSW resulted in significant increase in probabilities for Singapore, Thailand, and Vietnam. Significant increases for India, Singapore, and Thailand were also observed when tertiary coal and ship soot were added as sources. However, tertiary coal and ship soot had no significant effect on Vietnam aerosols.

Insights from linear mixing models. From the three—isotope plot (Fig. 3), it can be observed, that the aerosols do not fall on

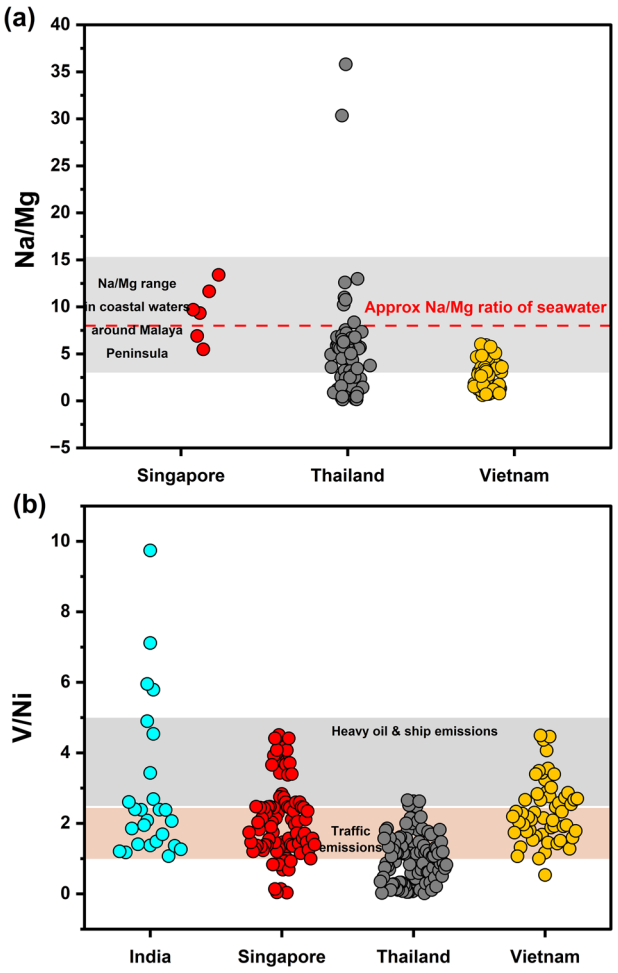

Fig. 2 Elemental ratios of aerosols demonstrating influences of sea spray and ship emissions. Trends in **a** Na/Mg ratios in aerosol mixtures from Singapore, Thailand, and Vietnam; **b** V/Ni ratios in India, Singapore, Thailand, and Vietnam. The shaded regions illustrate the ranges observed in sources for which these ratios serve as representative indicators.

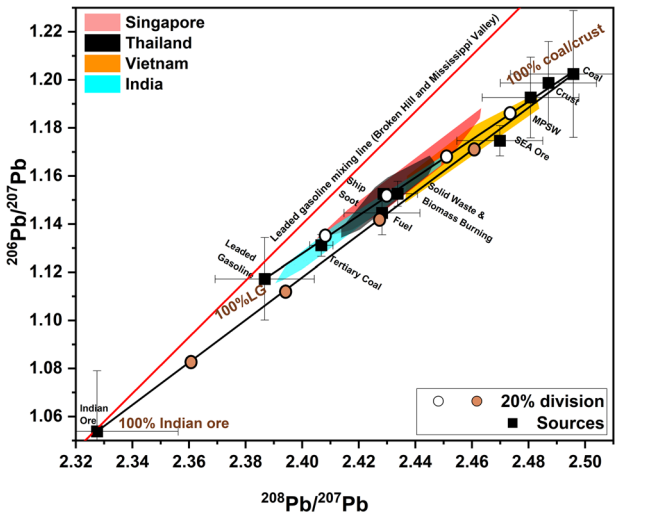

Fig. 3 Geometric mixing of sources. Geometric mixing lines constructed in three—isotope space between two extreme sources for SEA country aerosols and Indian aerosols segregated into divisions of 20%. The sources considered for SEA countries are leaded gasoline on one side and coal/crust on the other. For India, the sources are Indian ore on one side and coal/crust on the other. The traditional mixing line for leaded gasoline is depicted in red, constructed with the isotopic compositions of Broken Hill and Mississippi Valley ores. The aerosol mixture data for respective countries are depicted as fields, encompassing all the data points.

the traditional leaded gasoline mixing line constructed using Australian Broken Hill ore and USA's Mississippi Valley ore[56]. While the mean of the regional leaded gasoline data also deviates marginally from this traditional mixing line, the standard deviations exhibit alignment with it. Thus, in the three-isotope space ($^{206}Pb/^{207}Pb$ vs $^{208}Pb/^{207}Pb$), the aerosols from India are positioned between regional leaded gasoline and Indian ore on one end, and crust and coal on the other end. On the other hand, the SEA aerosols fall between local leaded gasoline and tertiary coal on one side, and crust and coal on the other side. Crust and coal are very similar in isotopic compositions and it is difficult to distinguish between their individual contribution towards aerosol Pb.

A simple two-source geometric mixing line between leaded gasoline and crust/coal for the SEA countries (Fig. 3) reveals that coal/crust contributes from 20 to 60% towards the Singapore and Thailand aerosols while the rest is from leaded gasoline. In the case of Vietnam, the contribution range for coal is 40–90% and the rest of the aerosol Pb stems from leaded gasoline. Similarly for Indian aerosols, a two-source mixing line between Indian ore and coal/crust indicates that coal/crust contributes 25–60% towards the aerosol Pb and the rest is from Indian ore. In such a binary mixing system, the contribution of each source is actually weighted based on its proximity to the composition of the mixture[57,58]. However, in this study, as there are more than two sources, their relative contributions cannot be accurately represented by this binary weightage method based on proximity. Thus, for gaining primary insight into the contributions of multiple sources towards the aerosol mixtures a simple two-isotope ratio linear mixing model having >3 sources is beneficial. The optimum results derived from the iterative procedure in such an underdetermined mixing system are presented in Fig. 4. The entire dataset is presented in Supplementary Data 2 [44] The model outcomes indicate considerable variability in the contributions of leaded gasoline (0–75%), natural sources (1–70%), coal (0–67%), intermediate sources (0–91%), for SEA countries. In India the contributions of leaded gasoline, natural sources, coal, intermediate sources, and Indian ore range from 18–44%, 1–62%, 0–30%, 2–37%, and 0–39%, respectively. In fact, the extreme values of these ranges seem to be impractical. For example, zero percent contribution obtained for different sources, especially crust is not possible. Further, a contribution of up to 75% for leaded gasoline in the case of Singapore aerosols also is highly unlikely. In Singapore, where only 1.2% of the fuel mix used for electricity generation is coal[59], it is highly unlikely that local and transboundary coal emissions will contribute up to 53%. Such unlikely extremities may indicate overemphasis on one source, causing it to dominate the mixture, while suppressing other sources. However, some useful primary observations from this linear model can be drawn:

A. Sources demonstrating the highest contribution percentages for all the four countries are leaded gasoline and natural sources (treating a spike of 91% from intermediate sources in case of Vietnam as an outlier). Thus, these two sources can be the dominating factors controlling the Pb isotopic compositions of the aerosol mixtures.
B. Singapore demonstrating the highest contribution percentage from leaded gasoline might have greater influence from the source compared to other countries.

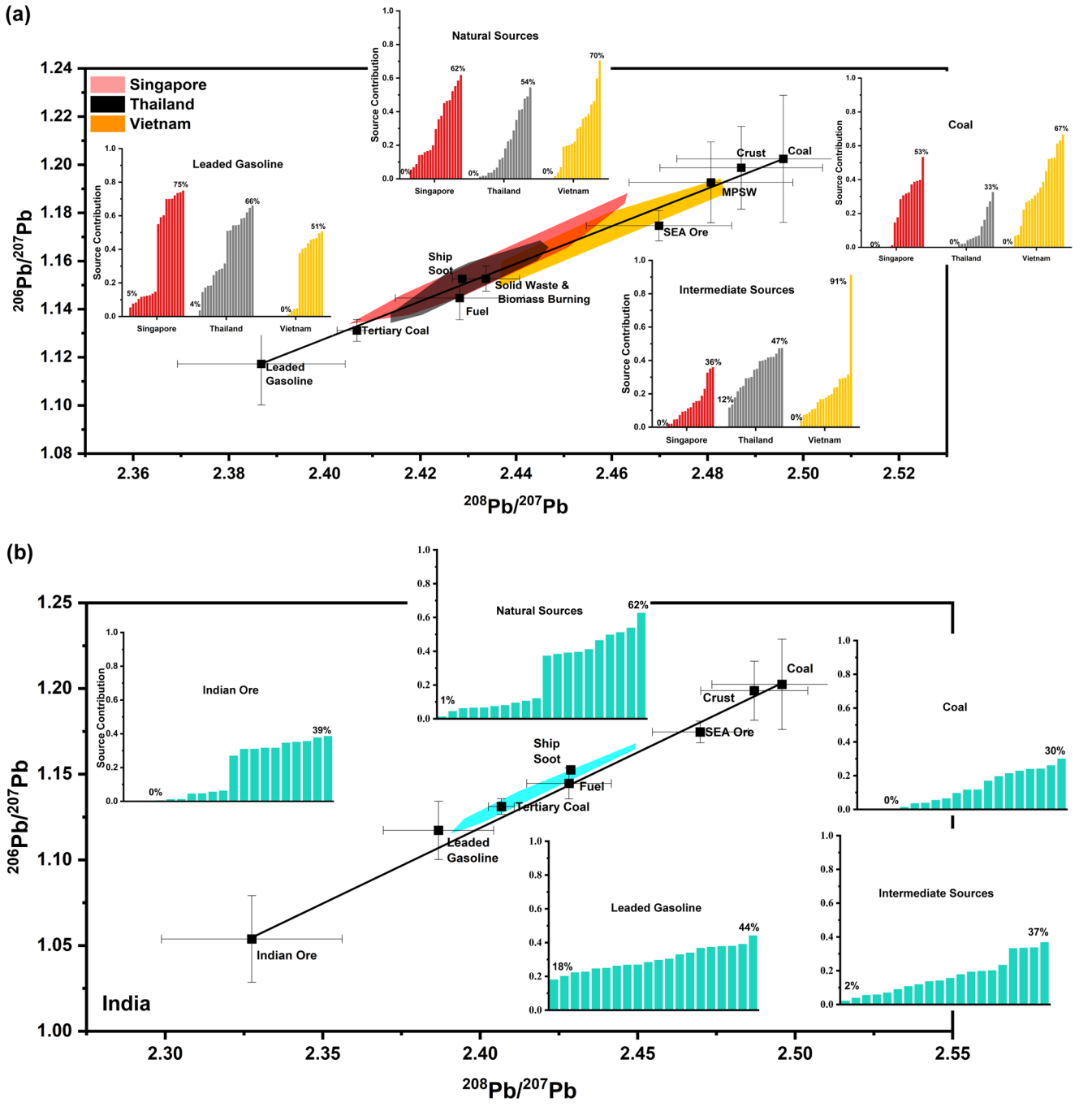

Fig. 4 Source contributions determined from simple linear mixing model. Bar plots for each source within insets in the three—isotope plots of aerosols and sources representing source contributions for **a** SEA countries and **b** India in $^{208}Pb/^{207}Pb$ vs $^{206}Pb/^{207}Pb$ space. Sources considered are 5 for India (coal, crust, Indian ore, leaded gasoline, and intermediate) and 4 for SEA countries (coal, crust, leaded gasoline, and intermediate). The intermediate source represents the mean of tertiary coal, unleaded fuel, ship soot, solid waste & biomass burning, and SEA ore for SEA countries, and tertiary coal, unleaded fuel, ship soot, and SEA ore for India. Percentages at the end of each bar depict the range of contributions, from the lowest to the highest, for each source.

C. Vietnam can have a greater influence from coal.
D. Among the 4 countries, India receives the least contributions from leaded gasoline.
E. The intermediate sources are the least to moderate contributing factors towards the aerosol mixtures.

However, as this linear model lacks the probabilistic framework of Bayesian models such as MixSIAR, which allows for incorporating uncertainties, the results obtained from the linear model cannot be fully relied upon. The broad variation of source contributions obtained from the linear model has to be narrowed down for a more accurate estimation of the relative proportions of different sources leading to a better understanding of the underlying source dynamics and their potential impacts. The results of the linear mixing model thus, were cross-verified using MixSIAR, where overlapping sources can be better resolved with the incorporation of standard deviations.

Insights from MixSIAR analysis. Similar trends between source contribution results obtained from both linear model and MixSIAR were observed. Both approaches identify natural sources

and leaded gasoline consistently as dominant contributors, indicating their substantial impact on the aerosol mixtures. Leaded gasoline contributed the highest towards Singapore aerosol Pb as was observed from the linear model outcome. Similarly, coal has a greater influence on Vietnam than towards other SEA countries. Contrarily, tertiary coal emerges as a substantial contributor to aerosol Pb levels in Thailand and India, an observation not evident from the linear model.

Thus, the basic observations from the MixSIAR analysis (Fig. 5; Supplementary Data 2)[44] performed on aerosol Pb samples from India and SEA countries are:

A. Natural background (UCC) is a prominent aerosol Pb source for India and the SEA countries.
B. Tertiary Coal combustion and ore processing dominate the anthropogenic Pb emissions in India.
C. Ship emission is probably an underdetermined source in India and to some extent in Thailand.
D. In the SEA countries, the largest anthropogenic source of aerosol Pb is the persistence of legacy Pb from the use of leaded gasoline in the last century.

The roughly analogous results observed between the two distinct analytical approaches (linear model and Bayesian model) lend credibility to the findings. Thus, the identified sources and their contributions are likely to be representative of the actual atmospheric Pb pollution dynamics of India and SEA countries.

In detail, the resultant contributions of the individual sources obtained from the MixSIAR model (Fig. 5) reveal a consistent contribution of background crustal material to all 4 countries. This is consistent with the linear model results as well as previous investigations that demonstrated a mixture of crustal and anthropogenic contributions towards atmospheric Pb[17,18,20,21,35,36]. Among other natural sources in SEA, MPSW made median contribution up to 14% in Vietnam. In Singapore and Thailand, the median contribution of Malaya Peninsula seawater was up to 10% and 9% respectively. Surprisingly the major anthropogenic contributor towards Pb in SEA was found to be leaded gasoline (median contribution up to 39%), even after approximately quarter century of leaded gasoline phase out in the three countries. Tertiary coal contributed substantially to Thailand (median contribution up to 20%). Tertiary coal fields are found in north-eastern India and there is proof of long-range transport of particulate matter from northeast India to Thailand[60]. Some of the endmembers such as fuel, and solid waste & biomass burning did not appear to individually contribute much (median contribution ≤9%). Vietnam atmosphere receives the least contribution from ship soot, which corroborates with finding from previous literature where insignificant impact of ship soot was observed[19].

In India, ore processing (median contribution up to 26%) and tertiary coal combustion (median contribution up to 37%) are the major anthropogenic sources. Indian atmospheric Pb was derived from ship emissions to some extent (median contribution of up to 15%) which was not considered in any of the previous studies. Leaded gasoline resuspension contributes relatively small (up to 9%) proportion of atmospheric Pb in India. To validate these results, the model was run in ^{204}Pb space for Singapore aerosols having the largest spread in three—isotope space, the outcomes of which are provided in Supplementary Note 2 and Supplementary Fig. 5. Further validations have been performed using concentration dependent Bayesian and linear models as discussed in Supplementary Note 3. The results for the concentration dependent model align with the concentration-independent ones as demonstrated in Supplementary Figs. 6 and 7.

Tertiary coal, wood, and wood charcoal from India exhibit comparable Pb isotopic compositions, which prompted their a priori grouping for MixSIAR modelling. In rural, semi-urban areas, and brick kilns, of India, fuelwood still remains a predominant energy source, with consumption reaching a staggering 216.4 million tonnes annually, as of 2011[55]. As radiogenic Gondwana coal dominates Indian coal reserves over less radiogenic tertiary coal[61], emissions from tertiary coal are likely to be less prominent. With analogous Pb isotopic composition[7], open burning of wood and wood charcoal could potentially be the most substantial contributor to atmospheric Pb in India only second to ore processing.

Pb emission estimates in the region show coal combustion in India emitted ~3500 tonnes of Pb in 2010 as compared to ~150 tonnes from Thailand[16]. Thus, probably emissions from coal combustion, fuelwood burning, and high-temperature metallurgy in India have overwhelmed the leaded gasoline Pb signature which is not the case for the SEA countries.

The average concentration of Pb in UCC is 17 ppm. Thus, aerosolisation of soil produced by weathering of the upper crust contains substantial Pb. Crustal dust as the natural background

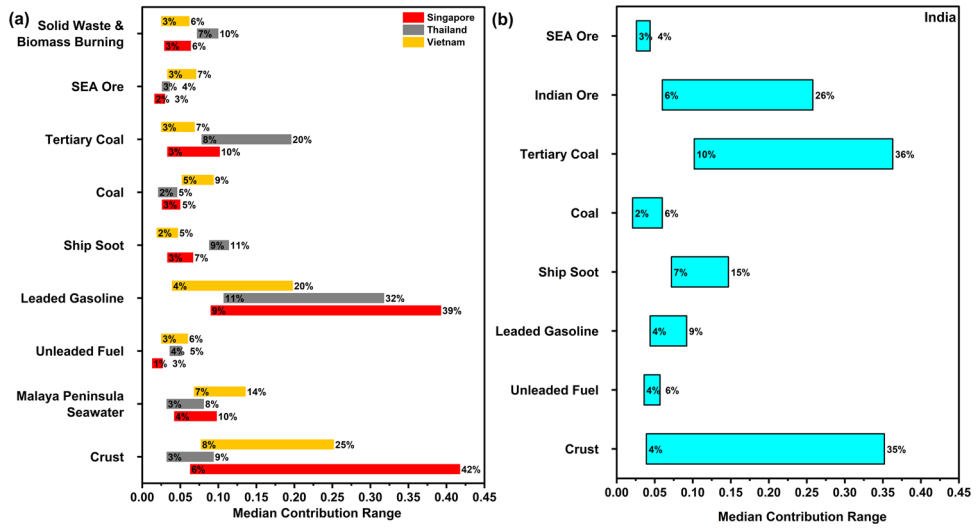

Fig. 5 MixSIAR results depicting median contributions of sources. Median contribution of individual sources towards atmospheric Pb in **a** SEA countries and **b** India determined from MixSIAR simulation. The percentages on left-hand side and right-hand side of the bars represent the minimum and maximum contributions from each source respectively.

has been hypothesised in almost all the previous studies. MixSIAR analysis suggests, that in addition to the crustal dust, sea spray also contributed towards aerosol Pb in the SEA countries (up to 8–14%).

Previous studies in India showed that post-leaded gasoline phase-out, the atmospheric Pb ratios plot away from the leaded gasoline mixing line and closer to the mixing line between coal combustion and ore processing[10,18]. The results obtained from MixSIAR analysis are in line with this observation. The Indian peninsula has a long coastline. Since 1992, the Asian ship traffic has increased by 200%[62]. Trace element ratios (V/Ni) indicated ship emission to be a plausible source of heavy metals in Singapore and Vietnam aerosols[23,35] (Fig. 2b). However, Pb isotopic composition of this source was not measured. None of the studies on Indian aerosol postulated ship emission as a plausible source of aerosol Pb. Insufficient Pb isotope data of Heavy Fuel Oil used by ships may have led to underestimation of source contributions towards atmospheric Pb in India.

Migration rate experiments and field observations suggest that gasoline Pb is retained in soil on generational time-span. Thus, resuspension of such soil could be an important source of atmospheric Pb. The contaminated roadside soils and dusts are often re-suspended by the turbulence created by the urban traffic[63]. Aerosols collected from the SEA countries in the past decade shows the largest anthropogenic source of aerosol Pb is leaded gasoline. Whether it is the result of transboundary transport from Myanmar and Indonesia using TEL additive till the 21st century or they are remobilisation of local top soil contaminated with legacy Pb remains a moot question.

Conclusion and future perspective. Pb wreaks its havoc silently and insidiously and hence often goes unrecognised. As atmosphere is the first recipient of the pollutant, identification of atmospheric Pb sources is of utmost importance. From a public health standpoint, inhalation and/or ingestion of atmospheric particles can be an important exposure pathway. Concentration of Pb bound to PM_{10} can contribute towards elevated BLL[64].

In the case of topsoil resuspension, simple yet effective abatement strategies such as covering the contaminated soils with clean soil have proved to be effective[65]. Regular and continuous monitoring of soil Pb could be performed with field portable XRF instrumentation. Future directions for research and policy to mitigate legacy Pb will require constant monitoring of soil Pb and thorough understanding of urban pedogenesis.

Previous research has typically focused on a limited set of end members, which may exclude important sources of Pb. In fact, it is evident from this analysis, that previously underdetermined sources such as and ship emissions contribute substantially towards atmospheric Pb. Additionally, non-combustible vehicular sources like tyre and brake wear have been found to be important contributors towards atmospheric Pb in other countries as they contain high concentration of Pb (10–55 ppm).[9,66,67] In fact, evidence of the presence of brake wear in atmospheric aerosols can be found in Vietnam from Cu/Sb ratios[35]. However, unfortunately, no local Pb isotopic data is available for these sources. Other non-combustible vehicular sources having even higher Pb concentrations include asphalt (738 ppm) and road paint (88 ppm) dusts generated from abrasion due to traffic[67]. Subsequently, these sources must be sampled locally and analysed for Pb isotopic compositions. Further Pb isotope data of sources such as Indian ore which is different from SEA ore is three decades old and hence is in urgent need of reanalysis. Extensive Pb isotope data of coal having high $^{206}Pb/^{207}Pb$ ratios is available globally. However, it can be observed from MixSIAR results that tertiary coal and wood charcoal having low $^{206}Pb/^{207}Pb$ ratio (from Assam, India) contributes heavily towards atmospheric Pb of Thailand and India. There are no Pb isotope data of coal from Thailand which may possess low $^{206}Pb/^{207}Pb$ ratio and contributes towards atmospheric Pb of SEA countries. Thus, to gain a comprehensive understanding of the sources and pathways of Pb in natural systems, it is important to quantify the isotopic composition of Pb in all potential end members.

Methods

Source and mixture data compilation. A comprehensive literature review was conducted to gather Pb isotope and relevant elemental concentration data of aerosol samples collected from South and Southeast Asian countries over the past decade. The studied regions encompass India, and SEA countries from where a total of 341 aerosol Pb isotope data points were acquired[7,10,16–18,20–22,35,36]. These aerosol data were compiled into 4 mixture datasets, one for each country, ready, to be fed to the mixing polygon and MixSIAR modelling frameworks. The Pb isotope data of the potential sources of these aerosols available from the same and cross-referenced literatures were compiled as source datasets for India and SEA countries. The isotope data was visualised in three isotope space ($^{206}Pb/^{207}Pb$ vs $^{208}Pb/^{207}Pb$). Further details about the source data are provided in the following section.

Constructing the mixing envelope. The ability of Pb to travel long distances due to its high atmospheric residence time makes it crucial that, all natural and anthropogenic sources from the regional and local emissions are taken into consideration. Natural background contribution from upper continental crust (UCC) has been consistently observed in SEA countries[17,20,36] and India[18,22] Among the regional sources, coal combustion emissions (both Gondwana and Tertiary coal) from India[7,18], China[20], Vietnam[20,35], Indonesia[23], and Thailand[20] have been proven to influence the SEA nations. Local sources include traffic emissions, high temperature metallurgy, historic leaded gasoline recirculation, and solid waste incineration[17,20,21,23,36].

The means and 1SDs of $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$, and $^{208}Pb/^{206}Pb$ of the well-documented sources were computed to perform a mixing polygon simulation (Monte Carlo) in 3-dimensional space in R. The simulation on the same dataset was also performed in $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ space which is discussed in Supplementary Note 1. The script for the simulation is available at http://www.famer.unsw.edu.au/downloads.html. Each iteration of the simulation generates a mixing envelope. The proportion of iterations in which the mixing envelope includes the isotopic composition of the aerosols within it, represents the probability of the aerosols ($n = 341$) for lying inside the envelope[68]. To increase this probability, the sources mentioned in previous literatures as potential sources, although lacking Pb isotope data were tried to be incorporated by finding proper Pb isotope data through literature survey.

Ship emission, and sea spray aerosols were indicated to be probable sources in previous investigations based on elemental ratios such as V/Ni and Na/Mg respectively[18,20]. As Pb isotopic compositions of these sources were not measured in those studies, the sources could not be considered in linear mixing models used in the studies. From literature survey, only one Pb isotopic data representative of regional ship emissions (ship soot) in Vietnam[19] was found. Since there was only one data point for regional ship soot, ship emission data available from Germany having only 10‰ and 4‰ difference in $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ spaces respectively was also combined[69]. Owing to relatively high concentration of Pb in Malaya Peninsula seawater (MPSW) compared to global average sea water, we further considered Pb

isotopic compositions of seawater around Malaya Peninsula as a natural contributor (sea spray aerosol) to SEA aerosol Pb[49]. Thus, as anthropogenic contributors for all 4 countries, we included ship soot[19,69], leaded and unleaded gasoline, coal from Vietnam, India, Indonesia south China (referred to as coal herein), and tertiary coal & wood charcoal combustion emissions from India[7,17,18,33–35,37,39]. We performed a priori grouping of Indian tertiary coal & wood charcoal isotopic compositions (referred to as tertiary coal herein) as both the sources demonstrated similar ratios[7,70]. As transboundary Pb contaminator for India we considered ores of Indonesia, Thailand, and South China (referred to as SEA ore herein)[40,42,71,72]. The sources were separately selected for India and SEA countries, which are listed in Table 1. We added the sources one at a time and ran the mixing polygon simulation to observe the resultant changes in probabilities. The probabilities obtained per aerosol were subjected to Analysis of Variance (ANOVA) at 0.05 significance level.

Linear mixing model. The aerosol mixture was primarily visualised along a two-end member geometric mixing line, partitioned into intervals of 20%. The proximity of the aerosol mixture to each source on the line provides the relative contribution between the two considered sources[57]. Since, such an approach cannot consider more than 2 sources, following this, the aerosol mixture and the source Pb isotopic data were subjected to conventional mass balance based linear mixing model based on the principle of IsoSource by employing the following equations[73]:

$$\left(\frac{206}{207}Pb\right)_{mix} = \left[f_a\left(\frac{206}{207}Pb\right)_a + f_b\left(\frac{206}{207}Pb\right)_b + \ldots + f_n\left(\frac{206}{207}Pb\right)_n\right] \quad (1)$$

$$\left(\frac{208}{207}Pb\right)_{mix} = \left[f_a\left(\frac{208}{207}Pb\right)_a + f_b\left(\frac{208}{207}Pb\right)_b + \ldots + f_n\left(\frac{208}{207}Pb\right)_n\right] \quad (2)$$

$$f_a + f_b + \ldots + f_n = 1 \quad (3)$$

where $\left(\frac{206}{207}Pb\right)_{mix}$ and $\left(\frac{208}{207}Pb\right)_{mix}$ are isotopic compositions of the aerosol mixtures; $\left(\frac{206}{207}Pb\right)_{a,b,\ldots,n}$ and $\left(\frac{208}{207}Pb\right)_{a,b,\ldots,n}$ are isotopic compositions of the sources and $f_{a,b,\ldots,n}$ is the unknown proportional contribution for each source towards the aerosol mixture. It is worth noting that these equations are approximations that work because of the limited fractional variance of Pb isotope data. This system using two isotope ratios can provide unique solution for each f, up to $n = 3$. However, when $n > 3$, this becomes an underdetermined system with no unique solution[57]. This linear model was computed for each country by considering the mean isotopic composition of the sources. For India, the considered sources consist of leaded gasoline and Indian ore as less radiogenic sources; coal and crust as the most radiogenic sources that bracketed all the Indian aerosols. An intermediate source was derived as the mean of unleaded fuel, tertiary coal, ship soot, and SEA ore that falls in the field of the aerosol mixtures. In case of SEA countries, the less radiogenic source was leaded gasoline. MPSW and upper continental crust were merged as natural sources due to their closely related isotopic compositions. Consequently, natural sources and coal constituted the radiogenic end members, while the intermediate end member was the mean of tertiary coal, unleaded fuel, solid waste & biomass burning, ship soot, and SEA ore. Thus, the total number of sources considered for India are 5 and SEA countries are 4, making the linear model framework an underdetermined system. To estimate the source proportions ($f_{a,b,\ldots,n}$) from such an underdetermined system, iterative approach as followed in traditional models like IsoSource was employed[57]. The linear model was made to perform an optimisation process to find the optimal value for mixture of sources that matches given target aerosol mixture values. The optimisation process performs ten restarts, each starting with a set of randomly assigned initial values for source proportions for the five sources. This process of conducting 10 restarts in the model was repeated 10 times to yield a set of 10 distinct sets of best results. The goal is to achieve an average deviation of 0.5‰ or less between the predicted and target mixture values. Two target aerosol mixtures, one with most radiogenic signature and the other with least, were selected. This selection is based on the fact that the source contribution towards the aerosols, lying between these extremes, will fall within the range of source contributions obtained for these extreme target aerosols. The model then identifies the best solution based on the lowest deviation. The potential combinations of source proportions sum up to 100%.[57].

Limitations of a linear model. While these linear systems can yield initial insights, their accuracy and precision are limited for a system having multiple end members. Primarily, they fail

Table 1 Individual sources of Atmospheric Pb in SEA countries and India.

India		South East Asia	
Individual Sources	**References**	**Individual Sources**	**References**
Crust	7,36,41	Crust	7,36,41
Gondwana Coal	7,18,33,35,37,39		
Indian Tertiary Coal & Wood Charcoal	7	Malaya Peninsula Seawater	49
Unleaded Fuel	7,33,35,43	Gondwana Coal	7,18,33,35,37,39
		Indian Tertiary Coal & Wood Charcoal	7
		Unleaded Fuel	7,33,35,43
Leaded Gasoline	17,34		
Indian Ore	38		
Transboundary SEA Ore	40,42,71,72	Leaded Gasoline	17,34
Ship Emissions	19,69	SEA Ore	33,40,42,71
		Solid Waste Incineration & Biomass Burning	17
		Ship Emissions	19,69
Aerosol Mixture	7,10,18,22	Aerosol Mixture	7,10,16,19–23,35–37

to yield distinct solutions when sources outnumber equations, a common occurrence in pollution apportionment studies. Furthermore, they solely account for source means without incorporating associated variability (standard deviations). Given the multiplicity and inherent heterogeneity of sources, it becomes imperative to consider both means and standard deviations in the modelling framework to achieve enhanced accuracy. Thus, the necessity of using Bayesian statistics based mixing models arises, that integrates the mean and uncertainty of the data into its framework. It is based on Markov Chain Monte Carlo (MCMC) simulation that produces plausible source proportion results based upon probability densities of the fed data[74]. This is particularly useful for analysing complex mixing problems where there are many more sources compared to isotope systems. Source apportionment of atmospheric Pb over a region requires consideration of isotopic compositions of multiple endmembers in which the Pb isotopic composition of each endmember may be spread over a large range. Thus, it is preferable to apply robust Bayesian models such as MixSIAR to these kinds of data set where we can consider multiple endmembers along with their uncertainty for determination of the proportion of contribution by the sources.

Bayesian statistics based mixing model. The means and 1SDs of $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$, and $^{208}Pb/^{206}Pb$ of the sources, listed in Table 1 and the isotopic composition of the aerosols were fed to the model framework in R[75]. Due to limited number of aerosol and source data, with respect to ^{204}Pb, the comprehensive modelling was conducted using ^{206}Pb, ^{207}Pb, and ^{208}Pb isotopes. With inadequate data, the standard deviations may not accurately represent the true heterogeneity in sources. Consequently, it can compromise the accuracy and reliability of MixSIAR results by generating higher uncertainties in source contributions and ambiguous source identifications. However, as the ratios relative to ^{204}Pb stem from distinct decay chains and possess the ability to discern source ages, the utilisation of ^{204}Pb contributes to improved source apportionment accuracy[32,76]. Thus, in order to validate the reliability of the modelling results derived from ^{206}Pb, ^{207}Pb, and ^{208}Pb isotopes, the aerosol mixture from Singapore and its potential end members were subjected to MixSIAR analysis specifically considering the ^{204}Pb isotope as well (Supplementary Note 2). The challenges for performing the analysis in ^{204}Pb space for this particular dataset are also discussed in Supplementary Note 2. While compiling the source data, the values that were greater/less than 2 SD of the mean were treated as outliers and eliminated. This reduces the overlapping of sources and increases model accuracy[31]. The mixture data was combined every 5‰ with respect to $^{206}Pb/^{207}Pb$ ratios. The model was run using 3 parallel chains with a "long" MCMC chain length of 300,000 iterations. To ensure that the chains reached equilibrium, the first 200,000 iterated values were discarded/"burned". The remaining samples were "thinned" by keeping every 100th iterated value[77]. The model convergence was checked using Gelman Rubin diagnostics.

Concentration-dependent mixing models. Concentration-dependent stable isotope mixing models are important in source apportionment studies, particularly when dealing with multiple sources having differences in elemental concentrations[78,79]. Accurate concentration data for the sources is a necessity for effectively utilising concentration-dependent stable isotope mixing models. This accuracy can only be achieved when isotope and elemental concentration data of the same sample set are available. While compiling the Pb isotope data of the south Asian and SEA aerosols along with their end members, this study identified substantial gaps in the availability of their corresponding elemental concentration data that are discussed in Supplementary Note 3.1. Hence, we could test the concentration dependent linear model for Indian aerosols only. MixSIAR model with concentration input was tested for Singapore and Thailand aerosols as they have the largest and the smallest variability in aerosol Pb isotope ratios in three isotope spaces respectively (Fig. 1a). The overarching conclusions drawn from the concentration independent model align with that of the concentration dependent model. The approaches followed for this process and the final outcomes are presented in Supplementary Notes 3.1 to 3.3. There were several challenges (discussed in Supplementary Notes 3.1) due to the unavailability of elemental concentration data in several literatures due to which concentration-dependent model outcomes cannot be entirely relied upon for this study. The dataset adopted for the concentration-dependent Bayesian and linear models are presented in Supplementary Tables 1 and 2, respectively.

Data availability

The input data for the models are adopted from previously published literature, as listed in Table 1. The probabilities from mixing polygon simulation and contribution fractions generated from linear & Bayesian model in this study are available in Supplementary Data 1 and Supplementary Data 2 respectively and also in Zenodo (https://doi.org/10.5281/zenodo.10101283).

Code availability

The R codes used for running the mixing polygon simulation and MixSIAR model are available publicly at http://www.famer.unsw.edu.au/software/polygon.html and https://zenodo.org/records/1209993, respectively.

Received: 30 May 2023; Accepted: 20 November 2023;
Published online: 12 December 2023

References

1. Sturges, W. T. & Barrie, L. A. Lead 206/207 isotope ratios in the atmosphere of North America as tracers of US and Canadian emissions. *Nature* **329**, 144–146 (1987).
2. Planchon, F. A. M. et al. One hundred fifty–year record of lead isotopes in Antarctic snow from coats land. *Geochim. Cosmochim. Acta* **67**, 693–708 (2003).
3. Sturges, W. T. & Barrie, L. A. Stable lead isotope ratios in arctic aerosols: evidence for the origin of arctic air pollution. *Atmospheric Environ.* **23**, 2513–2519 (1989).
4. Patterson, C. C. Contaminated and natural lead environments of man. *Arch. Environ. Health Int. J.* **11**, 344–360 (1965).
5. Radojevic, M. & Harrison, R. M. Concentrations and pathways of organolead compounds in the environment: a review. *Sci. Total Environ.* **59**, 157–180 (1987).
6. Erel, Y. Mechanisms and velocities of anthropogenic Pb migration in Mediterranean soils. *Environ. Res.* **78**, 112–117 (1998).
7. Mitra, A. et al. Lead isotope evidence for enhanced anthropogenic particle transport to the himalayas during summer months. *Environ. Sci. Technol.* **55**, 13697–13708 (2021).
8. Morton-Bermea, O., Rodríguez-Salazar, M. T., Hernández-Alvarez, E., García-Arreola, M. E. & Lozano-Santacruz, R. Lead isotopes as tracers of anthropogenic pollution in urban topsoils of mexico city. *Geochemistry* **71**, 189–195 (2011).
9. Resongles, E. et al. Strong evidence for the continued contribution of lead deposited during the 20th century to the atmospheric environment in London of today. *Proc. Natl. Acad. Sci. USA* **118**, e2102791118 (2021).
10. Sen, I. S., Bizimis, M., Tripathi, S. N. & Paul, D. Lead isotopic fingerprinting of aerosols to characterize the sources of atmospheric lead in an industrial city of India. *Atmos. Environ.* **129**, 27–33 (2016).
11. Doe, B. R. *Lead Isotopes*; Springer Science & Business Media (1970).
12. Flegal, A. R.; Smith, D. R. Measurements of environmental lead contamination and human exposure. In *Reviews of Environmental Contamination and Toxicology. Continuation of Residue Reviews*; Ware, G. W., Ed.; Reviews of

Environmental Contamination and Toxicology; Springer. 1–45 (Springer, New York, NY, 1995).
13. O'Nions, R. K., Frank, M., von Blanckenburg, F. & Ling, H.-F. Secular variation of Nd and Pb isotopes in ferromanganese crusts from the Atlantic, Indian and Pacific Oceans. *Earth Planet. Sci. Lett.* **155**, 15–28 (1998).
14. Komárek, M., Ettler, V., Chrastný, V. & Mihaljevič, M. Lead isotopes in environmental sciences: a review. *Environ. Int.* **34**, 562–577 (2008).
15. Ericson, B. et al. Blood lead levels in low-income and middle-income countries: a systematic review. *Lancet Planet. Health* **5**, e145–e153 (2021).
16. Lee, J.-M. et al. Coral-based history of lead and lead isotopes of the surface Indian Ocean since the mid-20th century. *Earth Planet. Sci. Lett.* **398**, 37–47 (2014).
17. Carrasco, G. et al. An update of the Pb isotope inventory in post leaded-petrol Singapore environments. *Environ. Pollut.* **233**, 925–932 (2018).
18. Das, R., Bin Mohamed Mohtar, A. T., Rakshit, D., Shome, D. & Wang, X. Sources of atmospheric lead (Pb) in and around an Indian Megacity. *Atmos. Environ.* **193**, 57–65 (2018).
19. Hoàng-Hòa, T. B. et al. Pb, Sr and Nd isotopic composition and trace element characteristics of coarse airborne particles collected with passive samplers. *Comptes Rendus Geosci.* **347**, 267–276 (2015).
20. Kayee, J., Bureekul, S., Sompongchaiyakul, P., Wang, X. & Das, R. Sources of atmospheric lead (Pb) after quarter century of phasing out of leaded gasoline in Bangkok, Thailand. *Atmos. Environ.* **253**, 118355 (2021).
21. Kayee, J. et al. Metal concentrations and source apportionment of PM $_{2.5}$ in Chiang Rai and Bangkok, Thailand during a biomass burning season. *ACS Earth Space Chem.* **4**, 1213–1226 (2020).
22. Kumar, S. et al. Tracing dust transport from middle-East over Delhi in march 2012 using metal and lead isotope composition. *Atmos. Environ.* **132**, 179–187 (2016).
23. Ray, I., Das, R., Chua, S. L. & Wang, X. Seasonal variation of atmospheric Pb sources in Singapore - elemental and lead isotopic compositions of PM10 as source tracer. *Chemosphere* **307**, 136029 (2022).
24. Walsh, M. P. The global experience with lead in gasoline and the lessons we should apply to the use of MMT. *Am. J. Ind. Med.* **50**, 853–860 (2007).
25. Dumitrescu, E. Global Efforts to Promote Cleaner Used Vehicles: Focus on Solutions (2017).
26. Drucker, D. G. et al. Aquatic resources in human diet in the late mesolithic in Northern France and Luxembourg: insights from carbon, nitrogen and sulphur isotope ratios. *Archaeol. Anthropol. Sci.* **10**, 351–368 (2018).
27. Parnell, A. C. et al. Bayesian stable isotope mixing models. *Environmetrics* **24**, 387–399 (2013).
28. Chen, H., Yan, Y., Hu, D., Peng, L. & Wang, C. High contribution of vehicular exhaust and coal combustion to PM2.5-bound Pb pollution in an industrial City in North China: an insight from isotope. *Atmos. Environ.* **294**, 119503 (2023).
29. Dietrich, M., Krekeler, M. P. S., Kousehlar, M. & Widom, E. Quantification of Pb pollution sources in complex urban environments through a multi-source isotope mixing model based on Pb isotopes in lichens and road sediment. *Environ. Pollut.* **288**, 117815 (2021).
30. Koffman, B. G. et al. Provenance of anthropogenic Pb and atmospheric dust to northwestern North America. *Environ. Sci. Technol.* **56**, 13107–13118 (2022).
31. Longman, J., Struve, T. & Pahnke, K. Spatial and temporal trends in mineral dust provenance in the South Pacific—evidence from mixing models. *Paleoceanogr. Paleoclimatology* **37**, e2021PA004356 (2022).
32. Longman, J. et al. Quantitative assessment of Pb sources in isotopic mixtures using a Bayesian mixing model. *Sci. Rep.* **8**, 6154 (2018).
33. Bi, X.-Y. et al. Lead isotopic compositions of selected coals, Pb/Zn Ores and fuels in China and the application for source tracing. *Environ. Sci. Technol.* **51**, 13502–13508 (2017).
34. Bollhöfer, A. & Rosman, K. J. R. Isotopic source signatures for atmospheric lead: the southern Hemisphere. *Geochim. Cosmochim. Acta* **64**, 3251–3262 (2000).
35. Chifflet, S. et al. Origins and discrimination between local and regional atmospheric pollution in Haiphong (Vietnam), Based on Metal(Loid) concentrations and lead isotopic ratios in PM10. *Environ. Sci. Pollut. Res. Int.* **25**, 26653–26668 (2018).
36. Das, R. et al. Suspension of crustal materials from wildfire in Indonesia as revealed by Pb isotope analysis. *ACS Earth Space Chem* **7**, 379–387 (2023).
37. Das, A. et al. Tracing lead contamination in foods in the city of Kolkata, India. *Environ. Sci. Pollut. Res.* **23**, 22454–22466 (2016).
38. Deb, M., Thorpe, R. I., Cumming, G. L. & Wagner, P. A. Age, source and stratigraphic implications of Pb isotope data for conformable, sediment-hosted, base metal deposits in the proterozoic Aravalli-Delhi orogenic belt, Northwestern India. *Precambrian Res* **43**, 1–22 (1989).
39. Díaz-Somoano, M. et al. Stable lead isotope compositions in selected coals from around the World and implications for present day aerosol source tracing. *Environ. Sci. Technol.* **43**, 1078–1085 (2009).
40. Huang, C., Li, H. & Lai, C.-K. Genesis of the Binh do Pb-Zn deposit in Northern Vietnam: evidence from H-O-S-Pb isotope geochemistry. *J. Earth Sci.* **30**, 679–688 (2019).
41. Millot, R., Allègre, C.-J., Gaillardet, J. & Roy, S. Lead isotopic systematics of major river sediments: a new estimate of the pb isotopic composition of the upper continental crust. *Chem. Geol.* **203**, 75–90 (2004).
42. Xu, J. et al. Mineralogy, fluid inclusions, and S–Pb isotope geochemistry study of the Tuboh Pb–Zn–Ag Polymetallic Deposit, Lubuklinggau, Sumatra, Indonesia. *Ore Geol. Rev.* **112**, 103032 (2019).
43. Yao, P.-H. et al. Lead isotope characterization of petroleum fuels in Taipei, Taiwan. *Int. J. Environ. Res. Public. Health* **12**, 4602–4616 (2015).
44. Ray, I.; Das, R. Dataset for: A Lingering Legacy of Leaded Gasoline in Southeast Asia (2023). https://doi.org/10.5281/zenodo.10101283.
45. See, S. W.; Balasubramanian, R.; Wang, W. A study of the physical, chemical, and optical properties of ambient aerosol particles in Southeast Asia during Hazy and Nonhazy Days. *J. Geophys. Res. Atmospher.* 111 (2006). https://doi.org/10.1029/2005JD006180.
46. Gasim, M., Khalid, N. A. & Muhamad, H. The influence of tidal activities on water quality of Paka River Terengganu. *Malaysia* **19**, 979–990 (2015).
47. Lim, W. Y., Aris, A. Z., Ismail, T. H. T. & Zakaria, M. P. Elemental hydrochemistry assessment on its variation and quality status in Langat River, Western Peninsular Malaysia. *Environ. Earth Sci.* **70**, 993–1004 (2013).
48. Looi, L. J., Aris, A. Z., Wan Johari, W. L., Md. Yusoff, F. & Hashim, Z. Baseline metals pollution profile of tropical Estuaries and Coastal waters of the straits of Malacca. *Mar. Pollut. Bull.* **74**, 471–476 (2013).
49. Chen, M. et al. Boundary exchange completes the marine Pb cycle jigsaw. *Proc. Natl. Acad. Sci. USA* **120**, e2213163120 (2023).
50. Apte, S. C. et al. Baseline trace metal concentrations in New South Wales Coastal waters. *Mar. Freshw. Res.* **49**, 203–214 (1998).
51. Wu, P.-C. & Huang, K.-F. Tracing local sources and long-range transport of PM10 in central Taiwan by using chemical characteristics and Pb isotope ratios. *Sci. Rep.* **11**, 7593 (2021).
52. Rabha, S. et al. Year-long evaluation of aerosol chemistry and meteorological implications of PM2.5 in an urban area of the Brahmaputra valley, India. *Environ. Sci. Atmospher.* **3**, 196–206 (2023).
53. Police, S., Sahu, S. K., Tiwari, M. & Pandit, G. G. Chemical composition and source apportionment of PM2.5 and PM2.5–10 in Trombay (Mumbai, India), a COASTAL INDUSTRIAL AREa. *Particuology* **37**, 143–153 (2018).
54. Khare, P. & Baruah, B. P. Elemental characterization and source identification of PM2.5 using multivariate analysis at the suburban site of North-East India. *Atmospher. Res.* **98**, 148–162 (2010).
55. Sharma, J. V. *Impact of wood-based energy on Forests in India | Wood Energy Catalogue | Food and Agriculture Organization of the United Nations*. https://www.fao.org/forestry/energy/catalogue/search/detail/en/c/1380965/ (accessed 2023-08-22).
56. Sangster, D. F., Outridge, P. M. & Davis, W. J. Stable lead isotope characteristics of lead ore deposits of environmental significance. *Environ. Rev.* **8**, 115–147 (2000).
57. Phillips, D. L. & Gregg, J. W. Source partitioning using stable isotopes: coping with too many sources. *Oecologia* **136**, 261–269 (2003).
58. Phillips, D. L. & Gregg, J. W. Uncertainty in source partitioning using stable isotopes. *Oecologia* **127**, 171–179 (2001).
59. Energy Market Authority Singapore. *Energy Transformation*. https://www.checkfirst.gov.sg/c/1ac4306f-4df9-4a7c-8759-00fda969d6a5 (accessed 2023-08-11).
60. Amnuaylojaroen, T., Inkom, J., Janta, R. & Surapipith, V. Long range transport of southeast Asian PM2.5 pollution to Northern Thailand during high biomass burning episodes. *Sustainability* **12**, 10049 (2020).
61. INDIAN BUREAU OF MINES. Indian Minerals Yearbook. (2014).
62. Tournadre, J. Anthropogenic pressure on the open ocean: the growth of ship traffic revealed by altimeter data analysis. *Geophys. Res. Lett.* **41**, 7924–7932 (2014).
63. Laidlaw, M. A. S., Zahran, S., Mielke, H. W., Taylor, M. P. & Filippelli, G. M. Re-suspension of lead contaminated urban soil as a dominant source of atmospheric lead in Birmingham, Chicago, Detroit and Pittsburgh, USA. *Atmos. Environ.* **49**, 302–310 (2012).
64. Zahran, S., Laidlaw, M. A. S., McElmurry, S. P., Filippelli, G. M. & Taylor, M. Linking source and effect: resuspended soil lead, air lead, and children's blood lead levels in Detroit, Michigan. *Environ. Sci. Technol.* **47**, 2839–2845 (2013).
65. Mielke, H. W. et al. The concurrent decline of soil lead and Children's blood lead in New Orleans. *Proc. Natl. Acad. Sci. USA* **116**, 22058–22064 (2019).
66. Jeong, H. Toxic metal concentrations and Cu–Zn–Pb isotopic compositions in tires. *J. Anal. Sci. Technol.* **13**, 2, https://doi.org/10.1186/s40543-021-00312-3 (2022).
67. Jeong, H., Ryu, J.-S. & Ra, K. Characteristics of potentially toxic elements and multi-isotope signatures (Cu, Zn, Pb) in non-exhaust traffic emission sources. *Environ. Pollut.* **292**, 118339 (2022).

68. Smith, J. A., Mazumder, D., Suthers, I. M. & Taylor, M. D. To fit or not to fit: evaluating stable isotope mixing models using simulated mixing polygons. *Methods Ecol. Evol.* **4**, 612–618 (2013).
69. Lahd Geagea, M., Stille, P., Gauthier-Lafaye, F. & Millet, M. Tracing of industrial aerosol sources in an urban environment using Pb, Sr, and Nd isotopes. *Environ. Sci. Technol.* **42**, 692–698 (2008).
70. Phillips, D. L. et al. Best practices for use of stable isotope mixing models in food-web studies. *Can. J. Zool.* **92**, 823–835 (2014).
71. Dongsheng 2. 越南东北部佐田铅锌矿床硫化物S、Pb 同位素特征及其地质意义. 地质通报, **34**, 757–768 (2015).
72. Hsu, Y.-K. Chinese Lead Isotope Database, (2019). https://doi.org/10.7910/DVN/VID3WR.
73. Phillips, D. L., Newsome, S. D. & Gregg, J. W. Combining sources in stable isotope mixing models: alternative methods. *Oecologia* **144**, 520–527 (2005).
74. Parnell, A. C., Inger, R., Bearhop, S. & Jackson, A. L. Source partitioning using stable isotopes: coping with too much variation. *PLoS ONE* **5**, e9672 (2010).
75. Stock, B.; Jackson, A.; Ward, E.; Venkiteswaran, J. Brianstock/MixSIAR 3.1.9, 2018. https://doi.org/10.5281/zenodo.1209993.
76. Baron, S., Tămaş, C. G. & Le Carlier, C. How mineralogy and geochemistry can improve the significance of Pb isotopes in metal provenance studies. *Archaeometry* **56**, 665–680 (2014).
77. Stock, B.; Semmens, B. MixSIAR GUI User Manual v3.1. (2016).
78. Phillips, D. L. & Koch, P. L. Incorporating concentration dependence in stable isotope mixing models. *Oecologia* **130**, 114–125 (2002).
79. Cox, T., Laceby, J. P., Roth, T. & Alewell, C. Less is more? A novel method for identifying and evaluating non-informative tracers in sediment source mixing models. *J. Soils Sediments* **23**, 3241–3261 (2023).

Acknowledgements

We extend their heartfelt appreciation to the three reviewers for their invaluable viewpoints in enhancing the quality of our manuscript. Reviewer 1's fundamental insights and comments have helped us provide more clarity to our article. The comprehensive overview provided by Reviewer 2 has ensured a smooth progression of the manuscript. We are grateful to Dr. Jack Longman, whose extensive work on MixSIAR analyses has significantly influenced and shaped the article. His thorough evaluation, has greatly enhanced the modelling aspects of the study. Lastly, we extend our heartfelt gratitude to the editorial board members for their constant support, positive attitude, and valuable input. We would like to thank Prof. Xianfeng Wang of Nanyang Technological University for supporting extensive atmospheric lead (Pb) research work in India and Southeast Asia for the past several years. This article would not have been possible without the insights gained from the previous work in the region by the authors. This research was supported by the Singapore Ministry of Education (MOE) Tier 1 grant (MOE-NTU_RG125/16-(S)), Department of Environment, Government of West Bengal (Grant no. ENV-29014(11)/1/2022-ACS (ENV)) and Science and Engineering Research Board (Grant no. SPG/2021/002652). RD's position is supported by UGC- Faculty Recharge Program and IR is supported by AICTE.

Author contributions

R.D. conceived, analysed, and supervised the entire study. I.R. compiled the data, performed the modelling studies, and prepared figures. Both authors wrote the manuscript.

Competing interests

The authors declare no competing interests

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s43247-023-01135-3.

Correspondence and requests for materials should be addressed to Reshmi Das.

Peer review information *Communications Earth & Environment* thanks Yigal Erel, Jack Longman, and the other, anonymous, reviewer(s) for their contribution to the peer review of this work. Primary Handling Editors: Yinon Rudich and Clare Davis. Peer reviewer reports are available.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.