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Accounting of the Use and Emissions of Polychlorinated Biphenyl Compounds (PCBs) in India, 1951−2100

Jayesh Parmar and Asif Qureshi*

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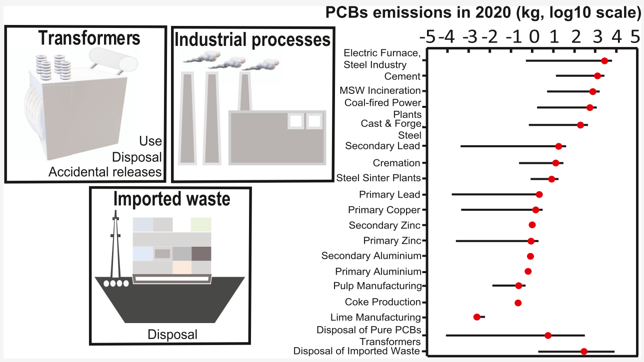
ABSTRACT: Polychlorinated biphenyl compounds (PCBs) are highly toxic organic chemicals still prevalent in the environment. While global inventories of the use and emissions of PCBs have been developed, estimates for individual countries determined using bottom-up approaches are few and often show different trends from the global inventory. Here, we determine the past, present, and future consumption and emissions of PCBs in India. A mass balance model was used to estimate middle (low−high) emissions in the period 1950−2100. Up to 7296 tonnes of PCBs have been used in transformers. PCBs imported as wastes are estimated to be approximately 5000 (2400−9100) tonnes. Total emissions from the use and disposal of transformers, industrial processes, and imported waste disposal are estimated to become 13 (0.1−537) tonnes, 89.26 (0.5−178) tonnes, 63 (3−910) tonnes, respectively, in the period 1950−2100. Congener-specific emissions are relatively high for low-chlorinated PCBs (−8, 18, 28, 31, 52, 101, 110, 118, 153, range: 0.1−118 tonnes). We find that industrial emissions are becoming important sources of PCBs and may become predominant, depending on emission scenarios.



KEYWORDS: *PCBs, India, consumption, emissions, mass balance*

1. INTRODUCTION

Polychlorinated biphenyl compounds (PCBs) are highly toxic organochlorine compounds classified as legacy persistent organic pollutants (POPs),[1] one of the "dirty dozen". PCBs have traditionally been used extensively in a variety of products, including paints, capacitors, transformers, and hydraulic equipment due to their high heat resistance, low electrical conductivity, and moderate rates of degradation.[2,3] PCBs can be introduced into the environment after the use and disposal of such products where they have been added intentionally. PCBs are also produced "unintentionally" by thermally driven de novo synthesis during industrial activities,[4,5] such as sintering, casting, forging, and incineration facilities in copper, aluminum, zinc, steel, and cement production industries and waste incineration.[6,7] Once emitted, PCBs can be transported to and deposited in different environmental compartments, air, water, soil, and vegetation, where they can persist for years or decades.[8,9] PCBs bioaccumulate in food chains and cause toxicity to humans upon exposure.[10]

Global production of PCBs between the years 1930 and 1990 was estimated to be about 1.3 million tonnes.[11] Earliest projections of total global emissions of 22 PCBs (PCB-5, 8, 18, 28, 31, 52, 70, 90, 101, 105, 110, 118, 123, 132, 138, 149, 153, 158, 160, 180, 194, 199) between 1930 to 2000 ranged from 440 tonnes to 91722 tonnes (with a middle value of 7709 tonnes). Peak emissions were projected to occur between 1970 and 1975 and then decline rapidly to a low value, 17.70 tonnes/y (range: 0.37−463.83 tonnes/y), by 2000.[12,13] Global emissions were later updated to account for emissions from Poland and the global e-waste transport.[14,15] The amount of PCBs not consumed by OECD countries was distributed to non-OECD countries based on their gross domestic product (GDP), and emissions were estimated.[12] The updated inventory[14] predicted total emissions of PCBs to fall to a low value, 0.1−1 tonnes/y or lower by 2040−50. However, some country-specific studies have shown different trends. For Switzerland and other countries by extrapolation, it was suggested that emissions from largely overlooked "open applications" such as use in paints will continue to persist, and emissions from Switzerland alone can be more than 0.1 tonnes/y beyond 2030; the peak in emissions was also delayed to after 1980.[16] In China, an increasing trend of atmospheric PCB concentrations was reported in rural sites and was explained by an increase in the unintentional thermally driven

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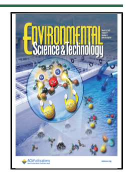

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industrial emissions previously not accounted for in the global emission inventory.[17] The decline in Chinese emissions was also more gradual than the global decline.[14] It has been acknowledged[12,13] that the validity of emissions and consumption estimates for any specific country suggested in the global inventory is uncertain and may be different from that obtained using a bottom-up country-specific approach. Further, a recent study[18] highlights that a majority of the signatories to the Stockholm Convention on POPs have incomplete inventories and are uncertain on current stocks and the extent of environmentally sound management within their borders. The use of PCBs in open applications such as paints and joint sealants is poorly documented, if at all.[18]

India is one of the signatories of the Stockholm Convention on POPs. PCBs were used in transformers until 2006.[19] The National Implementation Plan (NIP) on the Stockholm Convention on POPs (year 2011) and the PCB-specific NIP (year 2010)[20,21] have aimed at eradicating the use of PCBs and ensured their safe disposal, by 2025. PCB wastes were included in the Hazardous Waste (Management, Handling & Transboundary Movement) Rules[22] of 2008. The ″Regulation of Polychlorinated Biphenyls Order, 2016″[23] under the umbrella of Hazardous Waste Rules of 2016[24] implemented a ban on the manufacture, import, and export of PCBs and PCB-containing equipment; but it permitted the use of PCB-containing equipment for their certified lifetime or until 31 December 2025 (whichever is earlier) while "minimizing the possibility of leakage or release of PCBs into the environment".[23] However, the NIP report has also noted that PCB management was weak, with no dedicated support to implement the rules.[21] Data management was also poor.[21] A "significant import" and use of PCBs was reported in the NIP documents of 2010−2011.[20,21] This could possibly be because of the ambiguity over the import of PCB-containing equipment and manufacturing. PCBs containing oils were still being imported during 2011−2018.[25]

According to the NIP for India, overall PCB management in the country is hindered by unaccountability of use in transformers and open and partially open applications.[21] PCB emissions from India are presently available only from the global emission inventory scaled to India's GDP.[13] The total consumption of PCBs in India until the year 2000 was reported as 9264 tonnes.[13] The NIP on the Stockholm Convention on POPs[21] evaluated voluntary disclosure by 10 industries on their inventories of PCBs containing transformers. It estimated an existence of 3000 tonnes of pure PCBs, 6717.7 tonnes of oil-containing PCBs at a concentration above 500 ppm, including "pure" PCB transformers, and 120 tonnes of PCB waste stored in drums.[20,21] Temporal trends of PCBs used in and emissions from products such as transformers in the entire country have not been quantified. Additionally, while thermally driven industrial processes have been shown to contribute substantially in a major industrial nation such as China,[17] their contribution to PCB emissions in India has not yet been quantified.

In this work, we seek to better understand and quantify the use of PCBs in India and the associated emissions. We develop a bottom-up inventory of the use and emissions of PCBs in India from intentional use in transformers, imports of PCBs containing waste, and unintentional releases of PCBs from industrial activities, using primary historical and projected data. We identify the main sources of historical and future emissions of PCBs. Finally, we perform a sensitivity analysis to identify priority research areas for improving PCBs accounting in India.

2. MATERIALS AND METHODS

2.1. Scope of the Study. A preliminary literature review was conducted to identify the activities for which primary data were available and the corresponding time periods. Consequently, this work quantifies the use of PCBs in and emissions from transformers. Sufficient primary data on other uses such as paints, capacitors, and joint sealants were not available (Supporting Information, SI Section S1). Unintentional emissions were calculated for 12 activities: cement, steel, coke, lime, zinc, lead, aluminum, copper, and pulp production; coal-based thermal power generation; municipal solid waste production/incineration; and human cremation. PCBs containing waste imports from OECD countries and the associated emissions were quantified. The time period of analysis was 1951 to 2100 for products and industrial processes and 1980 to 2100 for waste imports.

2.2. PCB Use in Transformers and Resulting Emissions. *2.2.1. PCB Use in Transformers.* Data on the production of transformers in use were obtained from national statistics (Supporting Information, SI-Figure S1).[26,27] Missing production data were estimated from past or projected trends (Table S1). All assumptions and calculations on the use and emissions of PCBs are provided in the Supporting Information.

Data on in-use transformers were available from the year 1985.[26,27] Electrification was done at an "exponential pace" post independence (starting in year 1950) through adoption of various ″five-year development plans″ for the expansion of electrification in India.[28] We considered two rates of addition of transformers between 1950 and 1984: (i) an exponential rate, in accordance to the "exponential pace" of electrification (exponential addition scenario) and (ii) a more modest linear rate of addition (linear addition scenario). The number of transformers in 1985 and thereafter was as per actual data[26,27] (SI Section S1.2). New transformers introduced each year were calculated as the difference in the number of in-use transformers in a particular year with the number in the previous year. We stopped the addition of PCBs containing transformers to society in 2006 based on a report by the Central Power Research Institute (CPRI),[19] which records 2006 as the last manufacturing date of PCBs containing transformers.

We used the data on the total number of step-up, step-down, and distribution transformers present in any year and their aggregate power rating[26] to estimate the average power rating, which was then used to estimate the total amount of oil present in these three categories of transformers (SI Section S1.2 and Table S2−S7).[29,30] The CPRI report[19] also noted that most PCB-containing equipment were found in thermal power stations and that there were 1098 transformers containing pure PCBs. To account for these and to assess the relative importance of pure PCB transformers on the overall use and emissions of transformer-associated PCBs, we considered an additional scenario where we considered these transformers to be askarel transformers[31] and calculated their PCB contents following the procedure from recent works,[31] which was based on the assumption that approximately 30% of transformer weight is oil and 60% (range 40−80%) of this weight is attributable to PCBs.[31]

The CPRI report[19] provides information on the percentage of total transformers produced in each decade that contained

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PCBs (Table S5). Briefly, 40 and 30% of transformers contained PCBs in the period 1970−80 and 1980−90, respectively.[19] Only 1.4% of produced transformers had PCBs after year 2000. Accordingly, we calculated the number of PCB-containing transformers added each year within each decade, until 2006. PCBs introduced each year through the transformer were estimated using the information on PCB contents in transformer oil. More details are provided in the SI (Section S1, Tables S2−S10, and Figure S2).

2.2.2. Emissions of PCBs from Transformers. Emissions from transformers were estimated using the mass flow models of Breivik et al. and Glüge et al.[13,16] In these models, the PCBs used in products may be released accidentally or recovered and finally be disposed of (SI-Figure S3). The mass flow and emissions of PCBs, as a sum of the 209 PCB congeners,[32] are estimated using accidental releases, disposal and emission factors, and degradation rates. Further details and an example calculation are provided in the SI (Section S2).

Emissions from transformers were estimated from the mass flow model assuming that they were "closed" systems[13] and included fugitive emissions of PCBs during use and capture and disposal of remaining PCBs in disposal category landfill, open burning, waste incineration, and hazardous waste incineration. The disposal factors for transfer to each category are provided in SI-Table S13, based on Glüge et al.[16] The technical mixture (Aroclor) used in transformer oil globally was also found in transformers in India.[33] This validated our use of the mass flow model, which was based on similar assumptions.[16]

The mass balance model also provides congener-specific emissions for some PCBs (PCB-28, 52, 101, 138, 153, 180, 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189), which are also reported in this work. Three emission scenarios, middle, low, and high, were generated using the middle, minimum, and maximum PCB content values, respectively.

2.3. Unintentional Emissions of PCBs. Emissions from the 12 activities listed in Section 2.1 were determined as the product of respective activity rates and the corresponding emission factors (PCBs emitted per unit activity). Activity rates were determined from governmental and technical reports (Table S1).[34−52] Emission factors (EFs) for each activity are presented in Table S25, and example calculations are provided in SI Section S3. Primary EFs for emissions of PCBs from any of these activities do not exist for India. Therefore, we use the range of activity-specific EFs available in the literature, from the European Union,[53] which was a review of information from other works[54,55] and reports,[56−59] the United Kingdom [National Atmospheric Emissions Inventory (NAEI)], which had created inventory from sources,[53,57,60] and China[17] to derive a range of possible emissions. These reported EFs are for the sum of all possible congeners, and no information was given on the emissions of a single congener. Therefore, our estimates also correspond to the sum of all congeners.

Three emission scenarios, middle, low, and high were generated using the range of literature-derived emission factors (Table S25). We have classified emissions from these sources as unintentional, as PCBs are not intentionally used. Therefore, PCBs accidentally released from transformers, where they were added intentionally, are not added to the unintentional emissions; this also allows us to separate the primary sources of calculated emissions for further analysis.

2.4. Import of PCBs Containing Waste. Three scenarios of consumption of 22 PCBs within OECD countries, middle, low, and high, have been reported in the study of Breivik et al.[12] We used these scenarios to calculate the import and disposal of PCBs containing waste generated in OECD countries into India between 1980 and 2100. Our import estimates of PCBs consider transformers and capacitors only, as primary production data for OECD countries were available for them.[12] Consumption of PCBs in OECD countries has been reported until year 2000. Sixty percent of it has been reported to be used in transformers and capacitors.[12] We assumed that these would attain end-of-life according to a Weibull density function and be disposed thereafter. Incremental collection efficiency (0 to 50%) of disposed of products within the OECD countries was implemented from 1980 to 2000 and was at 50% post-2000.[13,14] Nine percent to 11% of the waste collected in OECD countries was exported into India.[14] We took 9, 10, and 11% as a low, middle, and high import scenarios, respectively. Imported waste was assumed to be disposed of within the same year. A waste disposal strategy was modeled based on literature-defined parameters by Breivik et al.[14] and included different proportions for open burning, landfilling, or disposal at dumpsites. Emissions from the disposed waste were estimated for 22 PCBs using the same approach as the Glüge et al. model.[16] Further explanation is provided in SI section S4 and Figure S5. The model then computed PCB congener-specific emissions for PCB-5, 8, 18, 28, 31, 52, 70, 90, 101, 105, 110, 118, 123, 132, 138, 149, 153, 158, 160, 180, 194, and 199. The range of emission factors and the three import scenarios were used to calculate the low, middle, and high PCB emission scenarios.

2.5. Uncertainty and Sensitivity Analysis. To account for the uncertainty in data on PCB contents and emission factors, we estimated the middle, lower, and upper range values for PCB contents and emissions.

For sensitivity analysis, we have considered parameters that influence the total emissions and the contribution of transformers to these emissions: PCB contents, emission factors, and the final methods of disposal. When evaluating the sensitivity of PCB emissions to PCB contents, we computed the overall change in PCB emissions for the period 1951−2100 when PCB contents were changed by +0.1% for the entire 1951−2006 duration. The impacts of each disposal method on emissions are assessed by increasing the factor signifying transfer to any single disposal method by +0.1% and evenly reducing the disposal factors for transfer to all of the other disposal methods so that overall disposal is maintained at 100%, throughout the time period of 1951−2100. Emission factors for use and disposal were collectively changed by +0.1% for the entire 1951−2100 period, and sensitivity was calculated.

Calculation for unintentional emissions involves the product of activity rates with the emission factors. A change in either activity rates or emission factors will enforce an equally proportional change in emissions. Thus, sensitivity analysis for unintentional emission is not presented.

3. RESULTS AND DISCUSSION

3.1. Intentional Use in Transformers. *3.1.1. PCB Consumption in and Emissions from Transformers.* A total of 1196 (44−2436) tonnes in the exponential scenario and 894 (33−1820) tonnes in the linear scenario are used in transformers. When we account for the 1098 pure PCB transformers, we estimate that a total of 4282 (2061−7296) tonnes of PCBs have been used in transformers (Figure 1).

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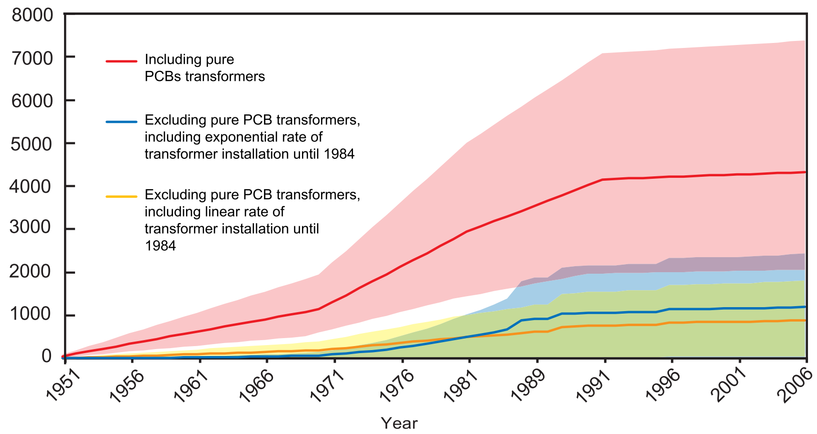

Figure 1. Cumulative use of PCBs in transformers. The dark line is the middle value estimate. Shaded bands represent the lower and upper limits of the estimate.

These numbers are in agreement with the report of 6717 tonnes of PCBs present in transformers.[21] By 2011, total PCBs present in the in-use transformers were estimated to be 320 (range: 7−873) tonnes, 222 (range: 5−582) tonnes, and 679 (range: 174−1691) tonnes in the exponential addition, linear addition, and pure PCB transformers scenario, respectively (Table S10).

Annual emissions with associated uncertainties are presented in Figure 2. The total emissions were 13 (0.1−138) tonnes for middle (low−high) scenarios in the case of exponential use, while 11 (0.1−117) tonnes for middle (low−high) scenarios in linear use. The values for the exponential scenario are approximately 1.2 to 1.5 times compared to the linear scenario. Hereafter, unless stated, the exponential addition scenario values will be discussed. A total of 60 tonnes (6−537) are expected to be emitted by the use of pure PCB transformers. Thus, the use of PCBs in and their emissions from transformers are dominated by the PCBs existing in pure PCB transformers that were used historically.

The sudden drop in emissions from transformers from 2012 is because of the considered removal and safe disposal in model after 2011, as suggested in the Indian National Implementation Plan for POPs (also refer to SI Section S2.2.4).[19−21] Emissions from transformers are expected to be over by 2020 in all scenarios.

3.1.2. Use and Emission Comparisons. Breivik et al.[13] suggest that peak emissions due to the use of products (including transformers, paints, and other sources) in India occurred between 1971 and 1976 with a middle value of approximately 4.6 (range, 0.2−48) tonnes/y. A downward trend was observed post-1976, leading to emissions of 0.4 (0−6.7) tonnes in the year 2000.[13] Estimates from our presented work on India suggest that emissions from transformers would peak later compared to that calculated by Breivik et al.,[13] between 1991 and 1996 depending on the scenario considered, with the middle value of the peak emission rate as 2 (0.4−19) tonnes/y. Emissions from products including open applications in Switzerland were in the range of 1−14 tonnes/y between 1950 and 2030.[16] Emissions peaked in 1980 and started decreasing, with values reaching below 1 tonne/y by 2020. In Germany, emission rates from open systems were 7−12 tonnes/y.[61] In China, emission of indicator PCBs (PCB-28, 52, 101, 118, 138, 153, 180) from products including open applications varied between 1 and 4 tonnes/y between 1965 and 2010.[62] Emissions peaked around 1990 and decreased gradually till 2010, with a value of 3.1 tonnes/y. Our estimated peak emission period for India is similar to that for China.[62]

Overall, when we consider the publicly available data of PCBs in transformers, our bottom-up estimates are in agreement with the nationally reported numbers.[19,21] The use of PCBs (and emissions) from open applications (that included paints) is high in countries that maintained a log.[18,63] Therefore, the use of PCBs (and emissions) from paints in India is also a possibility,[18,63] one that could potentially add an order of hundreds to thousands of tonnes of PCBs to the whole inventory.

The National Implementation Plan[21] has reported that PCB imported via products have been recycled into other products. Additionally, data are absent on the use of PCBs in applications such as paints, capacitors, and joint sealants. When these applications are considered, the overall use and emissions can surpass those from Breivik et al.[13] Unavailability of primary use data hinders further constraining of India's PCB inventory, and steps should be taken scientifically and administratively to generate new data on present use of PCBs in paints and other open applications and to make public any records on recycling and reuse of PCBs.

3.2. Unintentional Emissions. Cumulative emissions generated from industrial processes between 1950 and 2020 were 89 (0.5−178) tonnes. These are approximately 8−33% of the cumulative emissions from transformers by the year 2020. However, as activity data on steel processing, zinc metal processing, secondary lead and aluminum processing, cremation, and municipal waste generation were available only after the 1970s or 2000s (SI-Table S1), actual total unintentional emissions could be higher. After 1995, the yearly emissions keep on increasing, from approximately 1−11 tonnes/y by 2020 (Figure 3). Unintentional emissions in China for the steel and cement industry became prevalent after 1980 and remained in the range of 1−10 tonnes/year from 1980 to 2010, reaching almost 15 tonnes/year by 2010.[17] This suggests that similar to China,[17] unintentional emissions from India are becoming major sources of PCBs.

India is one of the highest cement-producing countries, and the activity rates for cement manufacturing are high (Figure S4). Even so, the steel industry's contribution to total unintentional PCB emissions was higher. The higher contribution of the steel industry is because of the high emission factors associated with the steel industry, specifically for electric arc furnaces (Table S25). Post-1995, the use of

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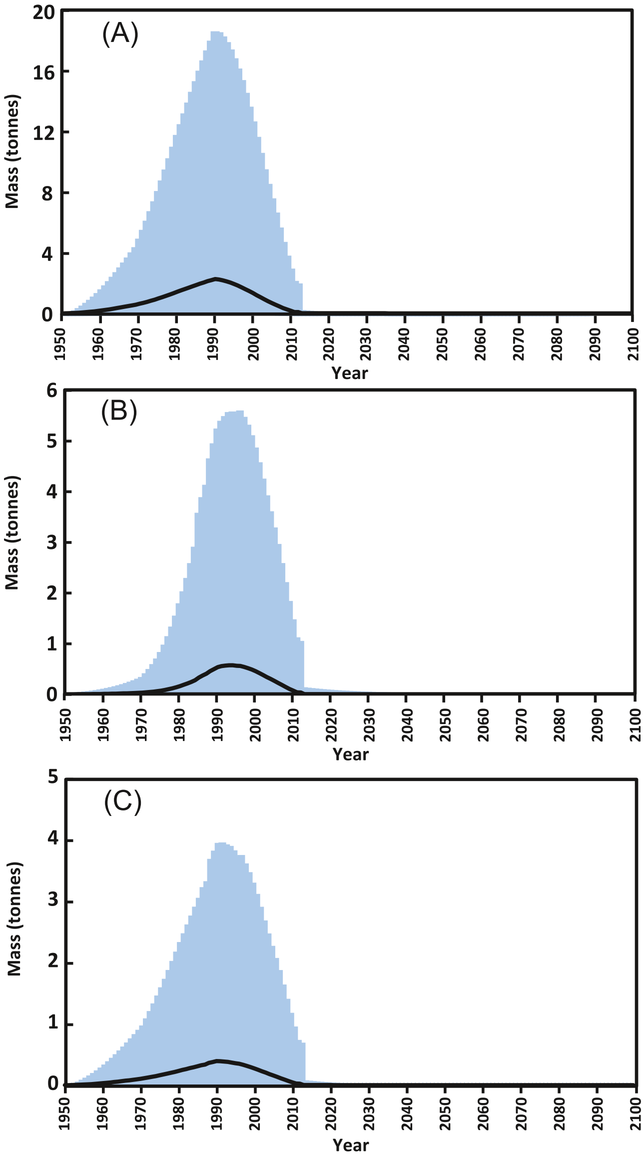

Figure 2. (A) Total emissions from the use of pure PCB transformer. (B) Total emissions from the use of transformers in the exponential scenario. (C) Total emissions from the use of transformers in the linear scenario. The dark black line is the middle value emission. Blue shaded bands represent the lower and upper limits of the estimate.

electric arcs and electric induction facilities in the steel industry has increased,[46] making them the primary source of PCB emissions from the steel industry. Thus, the steel industry shows a steep growth in emissions with middle value increasing from 0.01 tonnes/y in 1970 to 3 tonnes/y in 2020 (Figure S6). In comparison, cement shows a near linear growth in PCB emissions since 1950, approaching 1.25 (0–2.5) tonnes/y in 2020.

For total unintentional PCB emissions throughout the time period of 1951–2020 in different scenarios, cement industry and steel contributed approximately 65–79%. Municipal solid waste incineration contributed 10–15%, whereas thermal power generation contributed 6–10%, and the other activities contributed 1–10% to the total unintentional PCB emissions.

3.3. Import of PCB-Containing Wastes and Corresponding Emissions. We estimate an import of 5006 (2397–9084) tonnes of PCBs between 1980 and 2100 (Figure 4). These import values are higher than the PCBs that can be attributed to the import of POP-containing waste oil in the period 2011–2019,[25] 1280 tonnes (no information on the specific POP was given) even after we consider a high PCB content in waste oil, 26631 mg/kg (Table S8, Figure 4). The total emissions of 22 PCBs associated with imported wastes in the period 1980–2100 are 63 (3–909) tonnes. The emissions peaked in the year 2000 with a value of 3 (0.1–38) tonnes/y and are projected to be prevalent until 2035.

3.4. PCBs Stored in Landfills and Soil. From the domestic use of transformers and imported waste, a total of 2533 (1156–4136) tonnes of PCBs are disposed into landfills and a total of 2237 (1138–3838) tonnes are disposed of onto soil during 1950–2100. When we consider the 1098 pure PCB transformers that were disposed, along with imported waste, a total of 4001 tonnes (2246–5999) and 2364 tonnes (1139–4284) of PCB are expected to be disposed in landfill and soil. Thus, again, overall PCB transport to landfills is substantially influenced by the pure PCB transformers. The disposal of PCBs to soil is dominated by imported waste compared to transformers as the accidental release of PCBs from transformer onto soil is less; therefore, the values on PCB disposal to soil are not appreciably influenced by the quantification of PCBs in transformers. The storage of PCBs in soil and landfill peaked between 1996 and 2006 and began to decline after 2006 (Figure S7). This is because the use of PCBs in transformers was stopped in 2006,[21] and their disposal was accomplished by safe methods after 2012 in our model as per the NIP and CPRI reports.[21,64] Also, the imports of PCB waste started to decline by the year 2000.

We note that landfilling and soil contamination only delay PCB emissions and releases.[16] Higher temperatures and higher degradation rates could help in faster degradation.[13,65] Our model estimates that the PCBs stored within landfills and soils will be degraded by up to 95% by the year 2100. In 2100, the amount of PCB stored in landfills and soil is expected to be below 50–115 tonnes.

3.5. Congener Analysis. Congener-specific emissions from the intentional use of transformers and waste import models were estimated for PCB congeners (PCB-5, 8, 18, 28, 31, 52, 70, 77, 81, 90, 101, 105, 110, 114, 118, 123, 126, 132, 138, 149, 153, 156, 157, 158, 160, 167, 169, 180, 189, 194, 199). A total of 49 (3–609) tonnes of emissions were contributed by these congeners within the period 1950–2100 (pure PCB transformer use scenario). When these pure PCB transformers are excluded, emissions are 32 (1.2–470) tonnes.

Total emissions of indicator PCBs (PCB-28, 52, 101, 138, 153, 180) till 2100 were 35 (3–657) tonnes [without pure PCB transformer case; 25 (1–360) tonnes otherwise]. Total emissions for dioxin-like PCBs (PCB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189) were 12 (0.8–149) tonnes till 2100 [without pure PCB transformer case; 7 (0.2–110) tonnes otherwise]. Emission rates of congeners were below 1 tonne/y in the low scenario, while they varied up to 5 tonnes/y in the middle and high scenarios.

The range of total congener-specific emissions that could be emitted into the atmosphere between 1950/1980 and 2100 are presented in Figure 5. Total emissions of PCB-8, 18, 28, 31, and 52 were in the range of 0.2–118 tonnes (low–high-emission scenarios) and of PCB-70, 101, 105, 110, 118, 138, 149, 153, and 180 were 0.1–92 tonnes (low–high-emission scenarios). Emissions of the rest of the PCB congeners are

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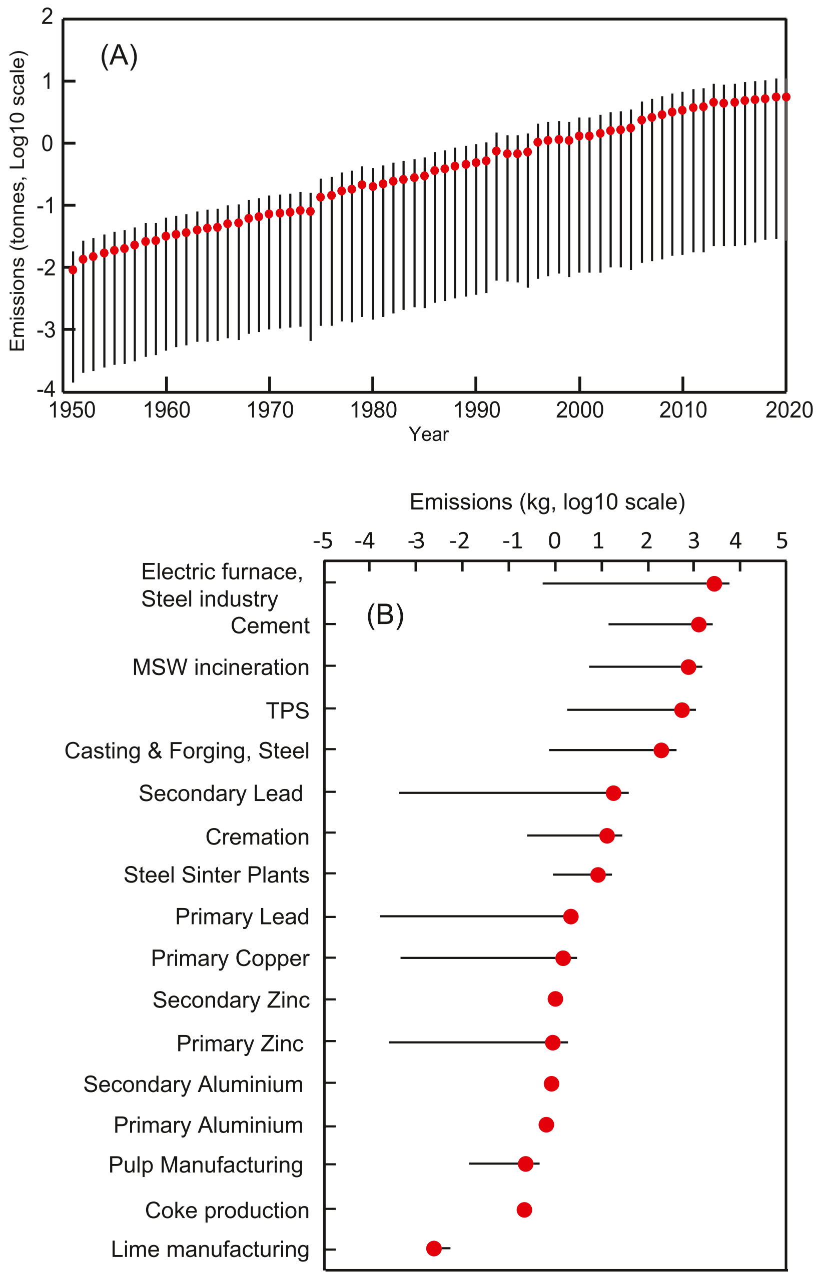

Figure 3. (A) Unintentional emissions (industrial) during 1951–2020. (B) Activitywise unintentional industrial emissions in 2020 [TPS: coal-fired thermal power stations]. The red dot is the middle value estimate, and the dark black line represents the lower and upper limits of the estimate.

lower than 23 tonnes. In general, low-chlorinated PCBs had high emissions compared to high-chlorinated PCBs.

Emissions with or without considering the use of pure PCB transformers together with imported waste appear to generate almost similar congener-specific emissions. This is because the higher emissions are dominated by the imported waste, and calculated emissions increases only by a factor of 1.5–2, while Figure 5 is plotted at a log10 scale. PCB congeners PCB-81 and -169 are used in paints only, and hence, their emissions are not included in Figure 5, which does not include the potential emission from paints.

For estimation of congener emissions from unintentional emissions, more information is required on congener use or congener-specific emission factors.

3.6. Uncertainty and Sensitivity Analyses. The three (middle, low, high) emission scenarios considered deal with the uncertainty in emissions and provide us with a broader picture of the overall PCB emissions in India. Emissions from products (intentional emissions) were 10 to 100 times higher in the high-emission scenario than the middle-emission scenario, while the low scenario was a factor of 10^{-3} to 10^{-10} of the middle scenario. The domestic literature provides little information on PCB emissions in India, and the assumptions required to estimate the data increase the uncertainty of the calculations. The industrial emissions rates (unintentional emissions) in low-emission scenarios were 100x lower and 2x higher in high-emission scenarios, relative to the middle scenario. The rates of emissions from waste import in low- and high-emission scenarios were 10x lower or higher than the middle-emission scenario, respectively.

Sensitivity analysis suggests that the disposal method of landfilling is the most sensitive parameter that influences

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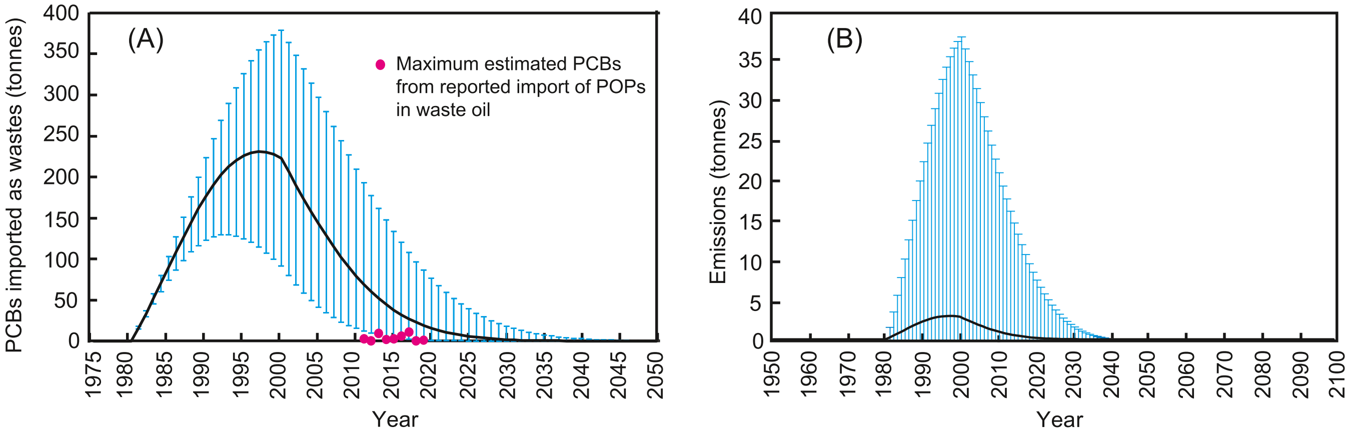

Figure 4. (A) Total PCB-containing waste imported. (B) Total PCB emissions through the disposal of imported waste during 1980−2100. The dark black line is the middle estimate. Blue bands represent the lower and upper limits of the estimate.

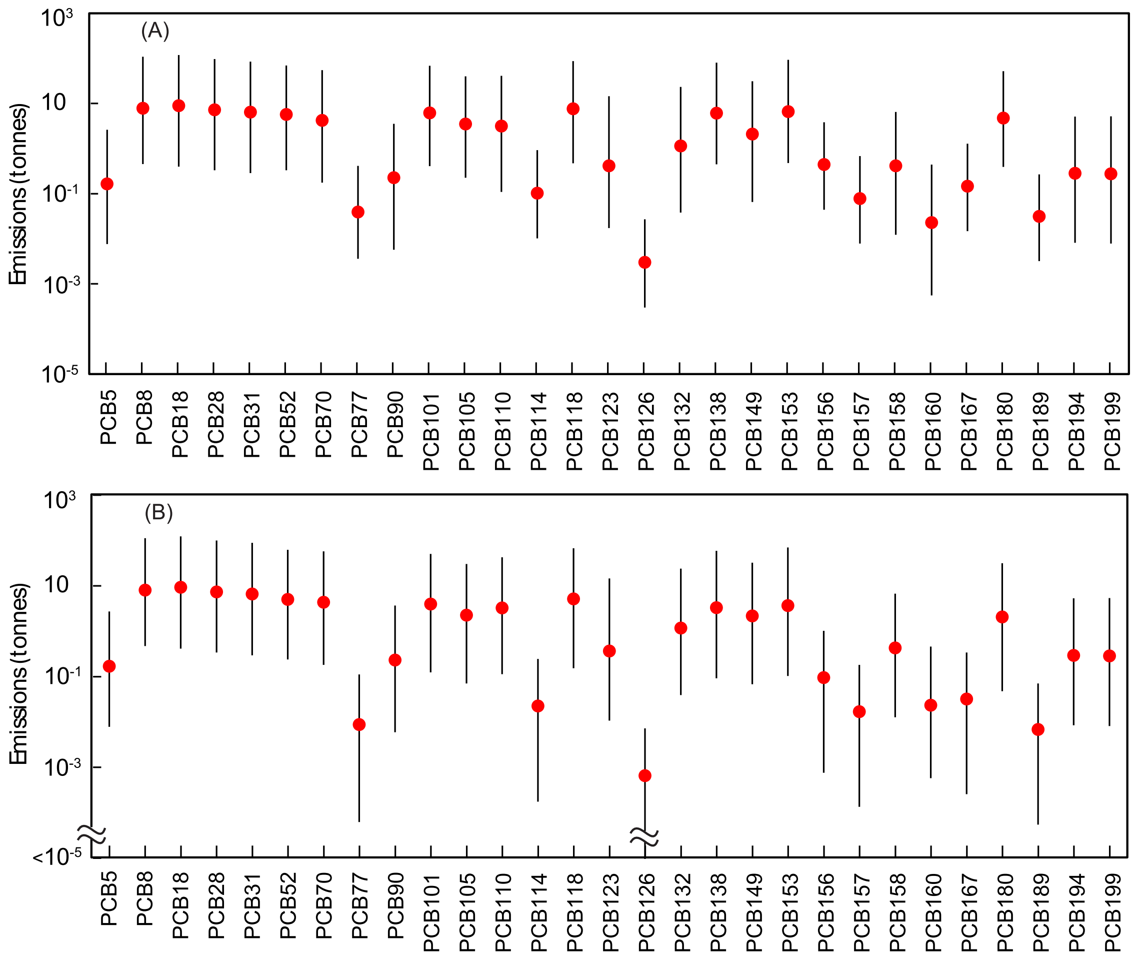

Figure 5. (A) Congener-specific emissions from pure PCB transformer use and PCBs containing imported waste during 1951−2100. (B) Congener-specific emission from transformer use in the exponential scenario and PCBs containing imported waste during 1950−2100. The red dot is the middle value estimate, and the dark black line represents the lower and upper limits of the estimates.

emissions from the transformers to the atmosphere (Table 1). This is because a major part of the use of PCBs in transformers goes to landfilling, where the degradation rates are comparatively high compared to emission factors.[13,16] The higher sensitivity in the low scenario toward the landfilling is due to the small lifetime of transformers (Table S11) that leads to early and higher disposal (Table S13) in landfills. In general, a decrease in sensitivity is observed from low to high scenarios because at high emissions, other parameters such as PCB contents start influencing the atmospheric emissions more.

Emissions to the atmosphere increase if the proportion of open burning as the final disposal method increases because the emission factors associated with open burning are higher than other disposal methods. Increases in the proportion of PCBs going to landfill, waste incineration, and hazardous waste incineration influence the atmospheric emissions negatively

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Table 1. Sensitivity Analysis for the Product Emission Model (Total Emissions = Y, Parameter Manipulated = X)

0.1% change adopted in (X)	sensitivity $[(\Delta Y/Y)/(\Delta X/X)]$		
	middle	high	low
PCB content of transformer oil	1.000	1.000	1.000
disposal factors			
landfill	−1.645	−1.071	−2.231
open burning	0.768	0.520	0.985
waste incineration	−0.697	−0.526	−0.745
hazardous waste incineration	−0.263	−0.214	−0.252
emission factors	1.000	1.000	1.000

because of the simultaneous decrease in the proportion of waste going to open burning, which has the highest emission factor; thus, the overall emissions decrease. All disposal methods except landfilling have a lower influence on atmospheric emissions compared to PCB contents and emission factors.

4. ANALYSIS OF PER CAPITA EMISSIONS OF PCBs IN INDIA WITH RESPECT TO OTHER REGIONS

Per capita emissions of PCBs attributed to their use in transformers are 4 mg/capita/y (range: 1−16 mg/capita/y) derived using the 2011 census population data.[66] Unintentional emissions would be 5 (1−9) mg/capita/y after 2000 and below 2 mg/capita/y before 2000. Emissions associated with imported wastes would be in the range of 1−32 mg/capita/y. In comparison, emissions of PCBs associated with products in China would be in the range of 1−4 mg/capita/y, derived using the 2010 census data for China.[67] Unintentional emission would be approximately 1 mg/capita/y. Emissions in the city of Zurich in Switzerland are approximately equal to 2−116 mg/capita/y.[13,68,69] Emissions for the city of Toronto in Canada have been reported in the range of 6−350 mg/capita/y.[70] Thus, overall per capita emissions estimated for India appear to be somewhat higher than that for China but somewhat lower than that for the two cities in Europe and North America. Also, the overall magnitude of per capita emissions of PCBs in India appears to be dominated by those associated with imported wastes.

5. SIGNIFICANCE AND FUTURE DIRECTIONS

Emission estimates made for India were previously limited to the year 2000.[13] Our work uses a bottom-up approach to provide inventory and emissions until the year 2100.

Our work shows that the overall emissions of PCBs in India (Figure 6) can be as high as 38 tonnes/y. The informal sector and the unreported industries remain unexplored. Open systems, such as applications of paints, could be a major contributor to emissions. Yearly production data from the monthly abstract of statistics by the Ministry of Statistics Production Implementation department[27] show that up to 3.4 million tonnes of paints were produced between 1980 and 1990 and 19 million tonnes between 1991 and 2020 (see Table S1, Figure S1, and SI Section 1 for more details). In paints where PCBs are used intentionally, PCB contents elsewhere have been reported in the range of 0.05−480 mg/kg.[71] PCBs have been detected in some paint pigments in the US until year 2016.[72] Lack of data on intentional use of PCBs in paints and their presumably unintentional existence at present in India hampers a quantitative assessment of PCBs associated with paints in India.

Small capacitors and similar nominally closed systems were not incorporated in our study. Despite the planned determination of PCB use in capacitors, no preliminary data is depicted in NIP except their manufacturing date.[19−21] Awareness and self-reporting surveys, as conducted for inventorying PCB use in transformers, could be conducted for capacitors. The use of PCBs in inks, adhesives, pesticide extenders, and microencapsulation of dyes for carbonless copy paper is also reported in other countries.[2,73] These sources have not been explored in India. Quantification of PCB emissions from these sources would help in better estimating PCB emissions. More data on the stored or discarded PCBs products would also help in a better constraining of PCBs in stocks, which may act as future sources to the environment.

Based on past trends, unintentional emissions have overtaken the emissions from transformers and wastes (intentional emissions) in the period 2010−2020 under the middle- or high-emission scenarios. This dominance may be delayed once information on other products such as paints, joint sealants, and capacitors is available to be considered. Nevertheless, industrial activity rates are increasing. For example, the global demand and production of cement and steel is ever-increasing,[74−76] and India is one of the major producers of steel and cement. Thus, unintentional emissions from India will likely become more important sources of PCBs. Future studies could focus more on quantifying and controlling unintentional emissions of PCBs.

In cement industries, reductions in emissions of dioxin-like PCBs have been observed when cement production is done

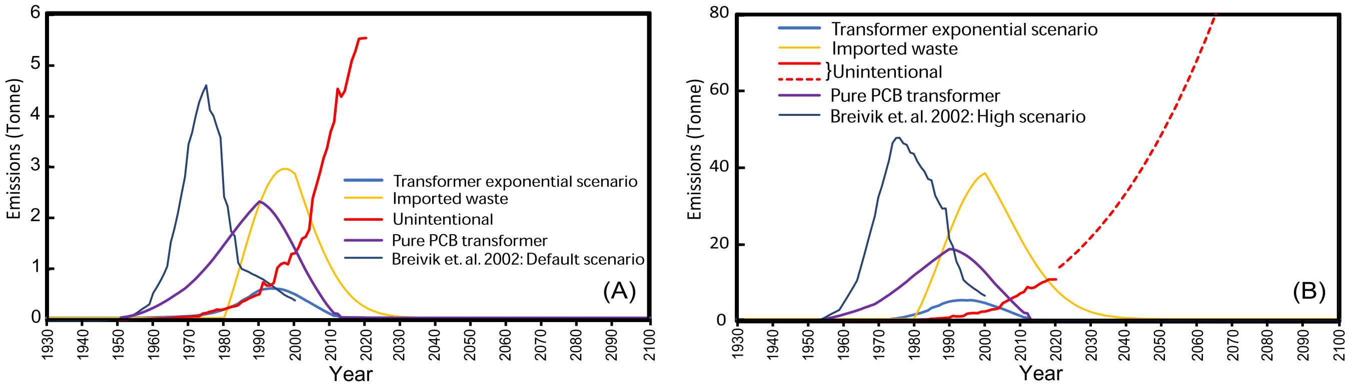

Figure 6. (A) Projected PCB emissions (middle value estimate). (B) Projected PCB emissions (upper limit estimate).

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with the coincineration of waste- or refuse-derived fuels.[77] Such measures may be evaluated for the Indian scenario, especially because the generation of municipal solid waste (MSW) is high in India and refuse-derived fuels can be potentially readily produced.[39]

Emissions of low-chlorinated biphenyls generally occur in the vapor phase and emissions of the high-chlorinated biphenyls typically occur in the particulate phase.[78] Flue gas desulfurization (FGD) systems could remove the solid-phase PCBs (high-chlorinated PCBs) more effectively than vapor-phase PCBs (low-chlorinated PCBs).[79] Memory effects where delayed emissions and concentrations increase due to PCB entrapment on a particulate matter and feeding material have been observed in control devices (fabric filter, wet scrubber).[80,81] These memory effects can lead to delayed increased emissions and calculation of negative removal efficiencies for both particulate and vapor-phase PCBs.[80−84] As such, further research in this area is required.

Unintentional emissions depend upon the activity rates and emission factors, and uncertainties in these rates and factors lead to uncertainty in calculated emissions for certain processes. For example, emissions factors for MSW incineration in closed environments have been defined, but such factors for informal open environment incineration[39,85,86] are highly uncertain, especially for India. Similarly, the activity rates and emission factors for open/controlled human cremation are not available. Certain landfills in India have management issues or are uncontrolled;[86] assessment of PCB emission factors for such landfills will lead to improved emission estimates. A domestic database of detailed activity rates and emission factors for PCB emissions would substantially reduce the uncertainty in PCB emissions from India and lead to a better reporting of data.

It has been noted that concentrations of PCBs in dust collected near urban recycling facilities can be 10 times higher than industrial dust[87] and higher in the soil at e-waste recycling sites compared to open dumpsites.[88] The inclusion of formal and informal recycling industry, along with well-defined activity rates and PCB emission factors for these activities will help in improving the overall emission estimates.

Industrial, municipal, and agricultural wastes in India are found to be PCB contaminated.[89] PCBs are lipophilic, and their transfer in food chains has been observed in India. They have been detected in human tissues, fish, and meat[90−92] and at high levels in breast milk.[93−95] Our reported emission estimates and trends can be used to determine the inventory of PCBs in different compartments of the environment (air, water, soil, sediment, and biota) and their intermedia fluxes and concentrations, for example, through multimedia modeling.[8,96] This will lead to better health risk assessments and may better help inform policy decisions.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c09438.

> Description of calculation approaches; tables on production data, emission factors and model parameters; and figures on production estimates, model schematics, and calculated emissions (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Asif Qureshi − *Department of Civil Engineering, Indian Institute of Technology (IIT) Hyderabad, Sangareddy, TS 502285, India; Department of Climate Change, Indian Institute of Technology (IIT) Hyderabad, Sangareddy, TS 502285, India;* orcid.org/0000-0003-3329-0166; Email: asif@ce.iith.ac.in

Author

Jayesh Parmar − *Department of Civil Engineering, Indian Institute of Technology (IIT) Hyderabad, Sangareddy, TS 502285, India;* orcid.org/0000-0001-6992-3549

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.2c09438

Notes

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