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Location-specific strategies for eliminating US national racial-ethnic PM_{2.5} exposure inequality

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Air pollution levels in the United States have decreased dramatically over the past decades, yet national racial-ethnic exposure disparities persist. For ambient fine particulate matter (PM_{2.5}), we investigate three emission-reduction approaches and compare their optimal ability to address two goals: 1) reduce the overall population average exposure (“overall average”) and 2) reduce the difference in the average exposure for the most exposed racial-ethnic group versus for the overall population (“national inequalities”). We show that national inequalities in exposure can be eliminated with minor emission reductions (optimal: ~1% of total emissions) if they target specific locations. In contrast, achieving that outcome using existing regulatory strategies would require eliminating essentially all emissions (if targeting specific economic sectors) or is not possible (if requiring urban regions to meet concentration standards). Lastly, we do not find a trade-off between the two goals (i.e., reducing overall average and reducing national inequalities); rather, the approach that does the best for reducing national inequalities (i.e., location-specific strategies) also does as well as or better than the other two approaches (i.e., sector-specific and meeting concentration standards) for reducing overall averages. Overall, our findings suggest that incorporating location-specific emissions reductions into the US air quality regulatory framework 1) is crucial for eliminating long-standing national average exposure disparities by race-ethnicity and 2) can benefit overall average exposures as much as or more than the sector-specific and concentration-standards approaches.

air pollution | environmental justice | fine particulate matter | air quality regulatory

The Clean Air Act has dramatically reduced outdoor air pollution levels in the United States, with (during 1990 through 2020) aggregate benefits exceeding costs 30-to-1 (\$2 trillion versus \$65 billion) (1). Important regulatory strategies include the National Ambient Air Quality Standards (NAAQS) and sector-specific emission-reduction technology requirements (e.g., Best Achievable Control Technology [BACT] standards). However, exposure inequalities persist (2–7). Disparities by race-ethnicity are larger than, and distinct from, those by income (4–6, 8, 9). Racial-ethnic inequalities in US ambient air pollution and subsequent exposures are attributable in part to racist planning, including historical, race-based housing segregation and land-use practices (10–18). Environmental racism scholars have suggested that strategies and policies for eliminating disparities will be most effective when racial-ethnic injustices are centered and directly addressed (19–22).

The existing literature documents exposure inequities (3–7, 9, 23–26) and investigates the impacts on inequities of emission changes for specific sources (e.g., refs. 27–36) or locations (37–43). However, the scientific literature has not investigated how to eliminate national racial-ethnic inequalities in air pollution or what level of emission reduction would be required to do so (44).

We examine three potential approaches to reduce or eliminate national exposure inequalities: 1) location-specific emission reductions (hereafter, “location”), 2) sector-specific emission reductions (“sector”; analogous to BACT-type approaches), and 3) requiring regions to meet a concentration standard (“NAAQS-like”). Approaches 2 and 3 mirror aspects of current regulations; approach 1 would be a new regulatory approach. We find that the location approach is by far the most effective (can eliminate national disparities with only small absolute emission reductions); the sector approach is poor (can reduce disparities, but requires substantially larger emission reductions; cannot eliminate disparities except by eliminating nearly all emissions); and NAAQS-like is the least effective (does not eliminate disparities). The location approach is also the strongest of the three for reducing population-average exposures.

To quantitatively compare the three approaches, we use the publicly available InMAP (Intervention Model for Air Pollution) source-receptor matrix (ISRM) (45) to estimate long-term average ambient fine particulate matter (PM_{2.5}) concentrations across the

Significance

National racial-ethnic exposure disparities in air pollution and their persistence across time are well-documented. It is unknown, however, how to systematically eliminate these disparities. Our results suggest that two main current regulatory strategies are ineffective at addressing national average racial-ethnic inequalities. In contrast, those inequalities can be eliminated with modest emission reduction using a location-specific approach. That approach is not included in current national regulatory frameworks (but it is being discussed, and some states have implemented it); also, that approach does as well as or better than the two other approaches at the goal of reducing overall population average. Our article informs the active national conversation about addressing environmental injustice, by providing a scientifically grounded approach for eliminating disparities.

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contiguous United States caused by anthropogenic emissions in 2014. Disparity here refers to the difference between population-weighted average PM_{2.5} concentrations for the most exposed racial-ethnic group minus the overall population (in sensitivity analyses, we instead investigate government-designated “high vulnerability” [HV] locations; *Materials and Methods*). ISRM predicts the concentration of primary PM_{2.5} and secondary PM_{2.5} formed from nitrogen oxides (NO_x), sulfur oxides (SO_x), ammonia (NH₃), and volatile organic compounds (VOCs). We employ the 2014 US Environmental Protection Agency (EPA) National Emission Inventory, grouped into 14 source sectors (see Fig. 2B, *SI Appendix*, and below). ISRM contains 52,411 grid cells (locations) with size (i.e., spatial resolution) ranging from 1 km in densely populated urban centers to 48 km in sparsely populated rural areas; the average spatial resolution is 2.6 km in Urban Areas and 22.6 km in non-Urban Areas (13.2 km overall). Emissions reductions for location and sector are an optimization to maximally reduce disparities for the most exposed group relative to the overall population average. They use the spatial resolution of the simulation grid and the 14 source sectors, respectively. Thus, our results inform what that method could optimally do to reduce or eliminate racial-ethnic exposure disparities. The NAAQS-like approach simulates successive, proportional emission reductions in each region violating the hypothetical NAAQS (e.g., 6 μg/m³), until the NAAQS-like standard is met.

Results

The base (no emission reductions) model predicts that the population-average PM_{2.5} concentration (units: μg/m³) is 7.0; for racial-ethnic subpopulations, it is 6.5 (White), 8.5 (Black), 7.7 (Hispanic), 8.0 (Asian), and 6.6 (other). The concentration disparity (which the location and sector approaches aim to optimally reduce) is 1.4 μg/m³ (20%) for non-Hispanic Black (herein, “Black”); a value that is consistent with empirical analyses (4, 5).

The results reveal that the location approach is substantially more effective and more efficient in reducing concentration

disparities for racial-ethnic groups and population average concentration than sector or NAAQS-like approaches (Fig. 1). “Effective” refers to successfully eliminating disparities; “efficient” refers to the reduction in disparity per unit reduction in emissions. The location approach is the most effective of the three approaches, in part because it is so much more efficient. For example, it requires 28-fold less emission reduction to achieve a 50% reduction in racial-ethnic concentration disparities compared to the sector approach (0.04 MT/y [relative value: 0.1% of total national emissions] for location versus 1.2 [4%] for sector). To reduce the disparity by 90% and 99%, respectively, the analogous emission reductions are 54-fold different (0.2 [0.7%] for location versus 12 [40%] for sector) and 83-fold different (0.4 [1.2%] versus 30 [99%]).

If optimizing to reduce overall population-average concentrations (rather than disparities), the location and sector approaches provide similar improvements initially (Fig. 1: For the first ~3 MT/y in emission reductions, both approaches reduce the average concentration to ~4 μg/m³), after which the location approach is comparatively more efficient (reduces the concentration quicker). Historically, total emissions of the five pollutants declined ~13 MT (27%) from 2010 (46 MT) to 2020 (33 MT), an annual change of ~1.3 MT (3.2%) (46).

NAAQS-like does not eliminate disparities and is much less efficient than location and sector (Fig. 1); i.e., it requires dramatically greater emission reductions to achieve comparable reductions in concentration disparities. For example, having all US urban areas meet an NAAQS-like concentration standard of 6 μg/m³ (much lower than the current NAAQS [12 μg/m³]) would require 9.1 MT/y of emission reduction (30% of total national emissions). This would reduce the population average concentration dramatically, to 3.5 μg/m³, but it would only lower, not eliminate, racial-ethnic disparities (to 0.6 μg/m³ [17%]). Our finding that the NAAQS-like and sector approaches cannot eliminate racial-ethnic disparities, even after substantial emission reductions, is consistent with recent historical analyses (3–5, 47).

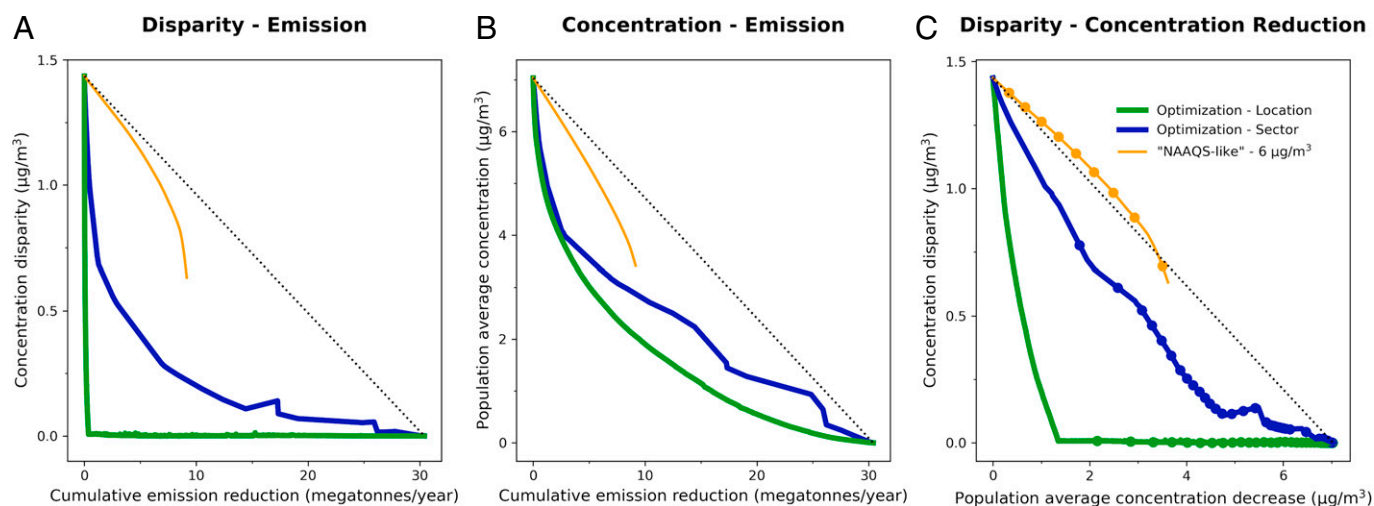


Fig. 1. PM_{2.5} exposure-disparity and concentration-reduction curves. (A) Concentration disparity between the most exposed racial-ethnic group (Asian, Black, Hispanic, White, or Other) and the population average (y axis) versus cumulative emission reduction (x axis). (B) Population average concentration versus cumulative emission reduction. (C) Concentration disparity versus population averaged concentration (i.e., the y axis values from A and B). For each panel, current conditions are the left side (i.e., “do nothing” at x = 0), and a complete (100%) emission reduction is the right side (i.e., achieving zero emissions: lower right, at x = 30.4 MT/y in A and B, at x = 7.0 μg/m³ in C). Each panel compares three approaches to emission reduction: location (green line), sector (blue line), and NAAQS-like (i.e., employing a concentration standard; here, 6 μg/m³; orange line). An “equal reduction” approach, where all emissions are reduced proportionately, would be a straight line (black dotted line). The location approach (green line) can eliminate national disparities with modest total emission reductions, whereas with the other two approaches, national disparities remain, even after substantial emission reductions (A and C). The location approach also does as well as or better than the other two approaches, for population average concentration (B and C).

As described in *SI Appendix*, we conducted several sensitivity analyses, such as considering relative, rather than absolute, inequality; urban or regional, rather than national, disparities; disparities for government-defined high-vulnerability locations, rather than racial-ethnic disparities; and average concentrations, rather than disparities (*SI Appendix*, section S1 and Figs. S1–S7). For NAAQS-like, we considered several values for the standard (*SI Appendix*, Fig. S8). In each case, the conclusion still holds, that the location approach is by far the most efficient approach. None of the NAAQS-like scenarios eliminate disparities.

The location approach (Fig. 2A) prioritizes urban emission reductions in the Midwest and Southeast, where there are clusters of 1) emission sources and 2) the most exposed racial-ethnic group. For the national-level optimization, higher prioritization for emission reductions is consistently correlated in univariate

(*SI Appendix*, Fig. S9) and multivariate (*SI Appendix*, Table S1) analyses with lower income, higher density of people and of emissions, higher percentage of non-White, higher percentage of Black, and greater level of segregation. In contrast, the relationship with percentage of Hispanic and Asian varies across models (*SI Appendix*, section S2). The spatial extent of high-priority locations varies by urban area (Fig. 2A and *SI Appendix*, Figs. S10 and S11; e.g., it is comparatively large in Los Angeles and Atlanta [$>50\%$ of the area is in the top 50% reduction priorities] and comparatively small in New York, Boston, Miami, Dallas, and Philadelphia [$<20\%$ of the area is in the top 50% reduction priorities]). In addition, the high-priority location(s) in an urban area can be in one cluster (e.g., Philadelphia; Chicago; Washington, DC; and Atlanta) or many clusters (e.g., New York, Houston, Dallas, and Boston). These between-urban differences

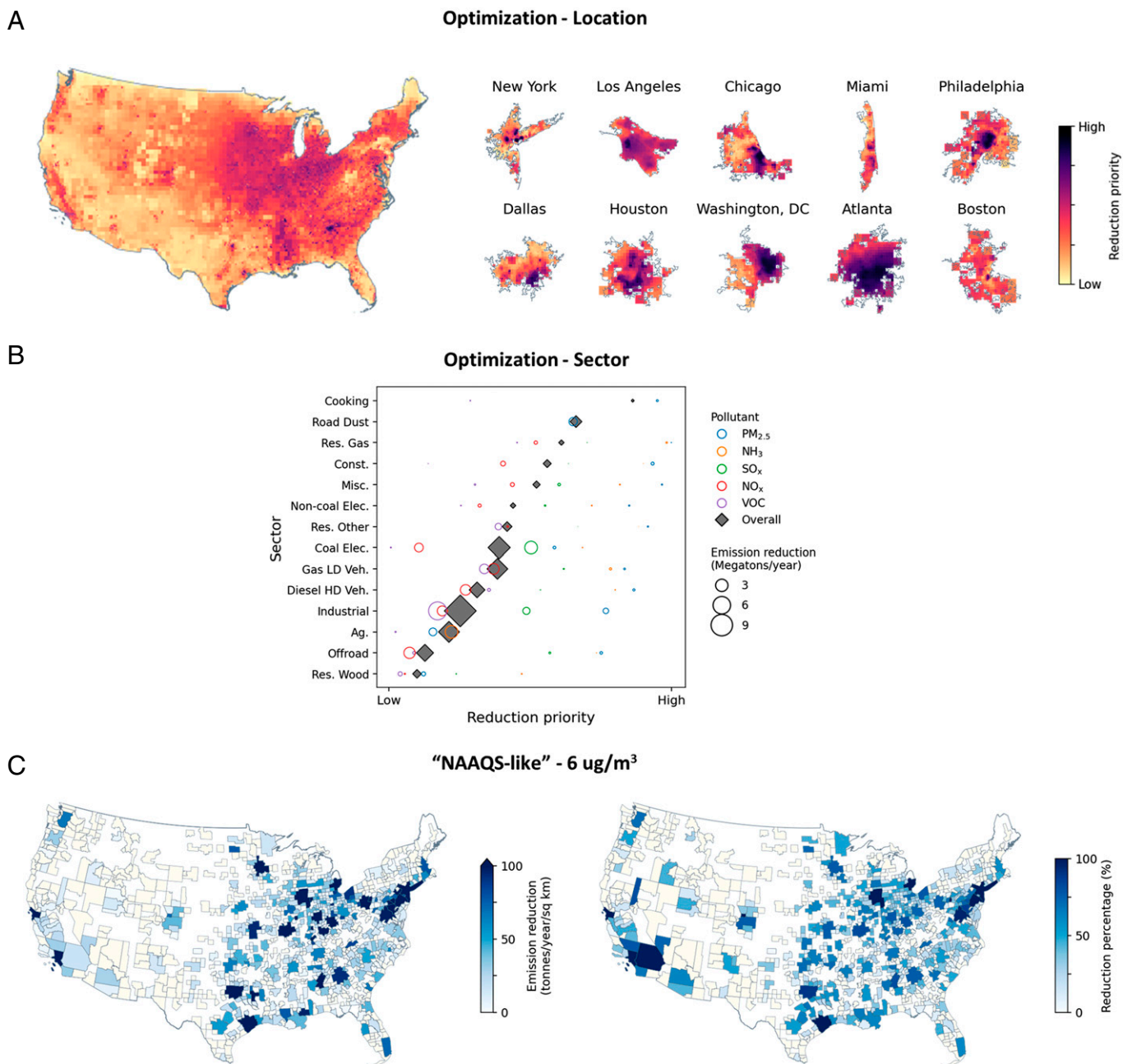


Fig. 2. Emission reductions for the three approaches: by location (i.e., corresponding to the green lines, Fig. 1) (A), by sector (corresponding to blue lines, Fig. 1) (B), and NAAQS-like (orange lines, Fig. 1) (C). A displays national results (Left) and zoomed-in results for 10 large areas (Right). Spatial units displayed in C are CBSAs, the geographic unit for NAAQS evaluation. The three approaches offer fundamentally different ways of formulating and prioritizing emission reductions. Ag., agriculture; Const., construction; Elec., electricity; HD, heavy duty; LD, light duty; Misc., miscellaneous; Res., residential; Veh., vehicle.

likely reflect differences in within-urban patterns in emissions and racial-ethnic segregation (48–50).

For the location approach, comparing the base case with the sensitivity analysis optimizing reduction of within-urban (rather than national) disparities (*SI Appendix, Figs. S12–S14*), the latter has more spatial variability than the former, but both approaches identify similar locations for emission reduction. This suggests that identifying and reducing emissions in those high-impact locations would, in many cases, reduce both urban-scale and national-scale disparities.

The sector approach (Fig. 2*B* and *SI Appendix, Figs. S15–S17*) prioritizes emission reduction from commercial cooking, road dust, residential gas combustion, and construction. Those sources have the largest marginal benefits, and so are ranked highly by our optimization. However, those four sectors have relatively small absolute emissions and, therefore, only modestly reduce the total disparities (reduce $0.4 \mu\text{g}/\text{m}^3$ [31%] of disparity in total; *SI Appendix, Fig. S16*). Industrial has the largest total emissions (9 MT/y [30%]) and also the largest contribution to inequality ($0.3 \mu\text{g}/\text{m}^3$ [25%]). However, the marginal improvement per MT/y is comparatively low, emphasizing that for industrial emissions, large emissions reductions are needed to achieve large benefits to disparities. Industry contains many types of sources; exposure disparities may be more sensitive to specific types of industrial sources than the industrial sector as a whole. Overall, of the five species that contribute to $\text{PM}_{2.5}$, for most sectors, reduction of primary $\text{PM}_{2.5}$ emissions causes the largest reduction in inequality (*SI Appendix, section S3*).

Discussion

Our analysis provides insight for general, archetypal emission-reduction approaches. Limitations of our approach include the following. We do not consider several important factors, including emission-reduction costs, technologies, and enforcement. Additionally, this study focused on ambient concentrations, which are related to, but distinct from, individual-level exposures and risks; indoor air pollution, microenvironments, and individuals' mobility can also contribute to exposure disparities (39, 41, 51–53), and background rates of disease can modulate disparities by concentration (54). We focused on disparities in $\text{PM}_{2.5}$ because most of the health damages from air pollution are attributed to $\text{PM}_{2.5}$ (55, 56). We do not explore other air pollutants, climate effects, and toxicities. Our approach employs national-level source-receptor matrix and emission inventories, which include documented uncertainties (44, 45, 57). Our results are robust at the national and aggregate levels; for investigating specific locations, we recommend additional (local) data and analyses, including local air quality data, local emission sources, and a source-receptor matrix at finer resolution. Our location analysis was performed at, on average, a 3.2-km scale in Urban Areas; future work can investigate whether greater spatial resolution provides additional efficiencies (58). A more recent emission inventory is available (2017 is available; we used 2014); we do not expect that our core conclusions would change for an inventory that is 3 y different. Finally, we grouped sources into 14 sectors; there may be additional insight from considering more granular categorization, especially for the industrial sector.

Exposure disparities are a legacy of race-based planning (8, 11, 12, 15, 16, 20, 47). They reflect systematic discrimination, housing segregation, and segregation in the proximity to pollution sources: for almost every sector of the economy, at various spatial scales, and persisting for decades (8, 44, 59). Our study highlights

the need for a fundamentally different framework for national air quality regulation in the United States that involves location-focused emission reductions in order to address national racial-ethnic exposure disparities. That framework would help accelerate efforts to redress the harms caused by environmental racism.

Our findings can inform national action [e.g., implementation of the Biden–Harris Administration's Justice40 Initiative (60, 61)] and emerging state and local environmental justice laws to identify overburdened communities and develop community emission-reduction plans (62–64). Our study supports the long-standing request from environmental justice communities and local organizations for location-specific solutions that center overburdened communities (11, 65–68). Our results also support putting in safeguards to address the potential for pollution trading (e.g., greenhouse gas-focused cap-and-trade) to exacerbate pollution inequities, especially for already overburdened communities (69–71).

Our results can help inform where to specifically target emission reductions, but more research is needed. Future work can further explore how various location-specific emission-reduction strategies—framed at a specific spatial scale (e.g., regional, state, urban, or neighborhood) and incorporating hyperlocal emission sources, community characteristics, and context (e.g., historic and contemporary zoning, planning policies, or engagement of community groups)—can swiftly achieve benefits across heavily impacted communities. Because $\text{PM}_{2.5}$ includes primary and secondary components (i.e., is emitted and also can form in the atmosphere), the most efficient locations to reduce emissions might be in a community or might be upwind. The present work considers national average inequalities (i.e., the difference in the average exposure for the most exposed racial-ethnic group versus for the overall population); future work should investigate other aspects of inequalities (also considering, e.g., exposure distributions, not just averages; other demographic attributes, not just race-ethnicity; and other geographic units), as well as broader aspects of achieving environmental justice (e.g., remedies for past harms). Tools are urgently needed that can 1) connect local and national decisions (e.g., planning and zoning changes, infrastructure investments, provision of public services such as mass transit, and household installation of solar panels) with benefits to highly impacted communities, and 2) support new and innovative approaches to environmental improvement (e.g., low-emission zones, reflecting cumulative burdens).

Materials and Methods

Emission Data and the Source-Receptor Matrix. We use the ISRM (45) to estimate, for the contiguous United States, concentrations caused by anthropogenic emission. Specifically, the ISRM, which is simulated by using InMAP (57), provides an estimate of the isolated impact of a 1-t emission change at any source location upon $\text{PM}_{2.5}$ concentration at each receptor location.

Emission data, which are from the 2014 US EPA National Emission Inventory v1, are grouped into 14 source sectors, 5 pollutants, and 3 emission stack heights (ground level: <57 m; low stack height: 57 to 379 m; high stack height: >379 m) and are allocated to the individual grid cells (52,411 in total) of the ISRM; see ref. 44 for details. The 14 sectors are: 1) agriculture; 2) coal electricity utility; 3) noncoal electricity utility; 4) commercial cooking; 5) construction; 6) diesel heavy-duty vehicle; 7) gasoline light-duty vehicle; 8) industrial; 9) road dust; 10) residential gas combustion; 11) residential wood combustion; 12) residential others; 13) off-highway vehicle and equipment; and 14) miscellaneous. Emissions from biogenic, wildfire, and international sources are not investigated. The five pollutants are: primary $\text{PM}_{2.5}$ and four precursors of secondary $\text{PM}_{2.5}$: NO_x , SO_x , NH_3 , and VOCs. The spatial resolution of ISRM grid cells ranges from $1 \text{ km} \times 1 \text{ km}$ (e.g., in densely populated urban centers) to $48 \text{ km} \times 48 \text{ km}$ (e.g., in remote, sparsely populated regions). The average

(i.e., unweighted average) spatial resolution is 2.6 km in Urban Areas, 22.6 km in non-Urban Areas (13.2 km overall); the population-weighted average spatial resolution is 3.2 km [Urban], 20.9 km [non-Urban] (12.2 km overall). The ISRM has separate matrices for the five pollutant types and three stack heights. The ISRM and emission data are freely available from <https://zenodo.org/record/2589760> and <https://zenodo.org/record/5831940>, respectively.

Calculating National Exposure Inequalities by Race-Ethnicity and Social Vulnerability. We calculate the total PM_{2.5} concentration from all source-locations for each grid cell. National population-weighted average concentrations overall and for the race-ethnicity and social vulnerability subgroups are calculated as straightforward weighted averages: the sum of the multiplied values of population count and concentration for each grid cell divided by the total corresponding population. Race-ethnicity is classified according to the 2010 Census at block level. We employ five racial-ethnic groups: non-Hispanic White (63% of the total population; herein, "White"); Latino or Hispanic of any race (17%; herein, "Hispanic"); non-Hispanic Black or African American (12%; herein, Black); non-Hispanic Asian and Pacific Islander (5%; herein, "Asian"); and populations who identify as American Indian, as another race, or as multiracial (3%; herein, "Other").

To identify vulnerable populations, we employ the Centers for Disease Control and Prevention (CDC)/Agency for Toxic Substances and Disease Registry's (ATSDR's) Social Vulnerability Index (SVI; <https://www.atsdr.cdc.gov/placeandhealth/svi/index.html>). SVI indicates, by Census tract, the population ability to prevent human suffering and economic loss in a natural or human-caused disaster or disease outbreak. SVI is calculated by CDC/ATSDR using 15 social factors across 4 themes: socioeconomic status, household composition, race/ethnicity/language, and housing/transportation. The CDC defines the 10% of Census tracts (excluding zero-population tracts) with the highest SVIs as HV and the remaining tracts as non-HV; here, we label grid cells overlapping (ArcGIS command: INTERSECT) those tracts as "HV" (9.5% of the total population; *SI Appendix, Fig. S4*) and remaining grid cells as "non-HV" (90.5%). For additional sensitivity analyses, we also employ two alternative definitions for HV locations: 1) the Census tracts in the highest 20% CDC SVIs (19.1% of the total population; in contrast, the main definition employs the top 10% of Census tracts); and 2) the Census tracts in the highest 10% of the PM_{2.5} Environmental Justice (EJ) index defined by the EPA's EJScreen (EJ Screening and Mapping Tool, <https://www.epa.gov/ejscreen/environmental-justice-indexes-ejscreen>). PM_{2.5} exposure disparity by race-ethnicity is calculated as the population-weighted average concentration for the most exposed racial-ethnic group minus the population-weighted average concentration for the total population. In the modeled "initial conditions" (i.e., without any hypothetical/simulated emission reduction), the most exposed racial-ethnic group is Black. During the emission-reduction procedure (see below), the most exposed racial-ethnic group can be any of the five groups. PM_{2.5} exposure inequality for HV is calculated as the population-weighted average concentration for the HV or the non-HV (the higher concentration between the two groups) minus the population average for the total population. We mainly focus on the absolute term of inequality ($\mu\text{g}/\text{m}^3$); we also report the relative (i.e., percent) change in inequality during the reduction procedure.

Calculating Within-Region and Within-Urban Exposure Inequalities. To calculate the within-region exposure inequalities, we use the geographic region classification by regional offices of the EPA. There are 10 EPA regions (<https://www.epa.gov/aboutepa/regional-and-geographic-offices>). We calculate the exposure inequalities within each of the 10 regions first and then calculate the national average (population-weighted) of within-region inequalities.

The within-urban exposure inequalities are calculated based on the same idea. We use Core-Based Statistical Area (CBSA), as defined by the US Census (<https://www2.census.gov/geo/tiger/TIGER2019/CBSA/>). CBSA is the geographic unit that the EPA uses for the NAAQS and refers to both metropolitan statistical areas and micropolitan statistical areas. There are 894 CBSAs in the contiguous United States, including 379 metropolitan statistical areas and 515 micropolitan areas.

Emission-Reduction Scenarios and the Optimization Methods. In order to perform optimization, we first calculate the PM_{2.5} concentrations from every location (i.e., each grid cell; $n = 52,411$ in total) and pollutant (location approach, 239,348 combinations in total [i.e., 52,411 locations; 5 pollutants;

of the 262,055 maximum possible location-pollutant pairings, 22,707 have zero emissions and so are not considered here as an opportunity for emission reduction]) and from every sector and pollutant (sector approach, 61 combinations in total [i.e., 14 sectors, 5 pollutants; of the 70 maximum possible sector-pollutant pairings, 9 have zero emissions and are excluded here]) for the total population and for each subpopulation (race-ethnicity groups and HV). We then calculate the marginal concentration difference between each subpopulation and total population, per 1-t emission change of each by-sector or by-location combination: $MCD_{ij} = (C_{ij} - C_{i,P})/E_i$. Here, i represents a certain combination of sector and pollutant or location and pollutant (e.g., NO_x from road dust, SO_x from grid cell #100, etc.); j represents a certain subpopulation (e.g., Hispanic population, HV, etc.); P represents the total population; C_{ij} is the population-weighted average PM_{2.5} concentration from source/location i for group j ; $C_{i,P}$ is the population-weighted average PM_{2.5} concentration from source/location i for the total population; E_i is the total emission from source/location i ; and MCD_{ij} is the marginal concentration difference between group i and the population average per 1-t emission from source/location j .

The optimization determines the order in which emissions will be reduced and models the exposure impacts as those emission reductions occur. The optimization is performed by using the following algorithm:

1. Determine the most exposed racial-ethnic group or the more exposed social vulnerability group (j_0) at the initial (i.e., zero emission reduction) condition, and then determine the combination (i_0) with the largest marginal concentration difference (MCD) for that subgroup.
2. Reduce all emissions from i_0 , and then calculate the remaining concentrations for the total population and for all subgroups.
3. Repeat step 1 for the new conditions: Determine the most exposed racial-ethnic group or the most exposed social vulnerability group (j_1) for the updated concentrations, and then find out the combination (i_1 ; from the remaining available options) with the largest MCD for that subgroup.
4. Repeat step 2, for the new conditions: Remove all emissions from i_1 , and then calculate the remaining concentrations for the total population and for all subgroups.
5. Repeat steps 3 and 4 until the emissions from all combinations have been reduced to zero. The concentrations for the total population and all subgroups also reach zero at this point, and the optimization ends.

Via the steps above, we simulate the local optimum solution to minimize the concentration inequality.

Regional and Urban-Level Optimizations. In addition to optimization to reduce the overall national disparities (see above), we also explore the optimization methods for reducing disparities at regional and urban scales. We assume that each region or Urban Area reduces emissions in order to optimally reduce the inequalities within that region/Urban Area. We use the 10 EPA regions and US Urban Areas defined by the US Census (<https://www2.census.gov/geo/tiger/TIGER2018/UAC/>). We consider only Urban Areas (as defined in the US Census: population: 50,000 or more) with more than 20 ISRM grid cells within their geographic boundaries ($n = 171$). (If Urban Areas are divided into small/medium/large based on national population tertiles, as described in ref. 72, our approach provides 10 large, 44 medium, and 117 small Urban Areas.) We explore both optimization approaches (location and sector).

NAAQS-Like Methods. For the NAAQS-like approach, we set CBSAs as the emission-reduction unit and set one concentration target for all CBSAs. The NAAQS-like goal is that the concentration in each grid cell in a CBSA doesn't exceed the concentration target (i.e., the NAAQS). We separately employ multiple concentration targets, including 10, 9, 8, 7, 6, and 5 $\mu\text{g}/\text{m}^3$ (the recently updated World Health Organization guideline for annual average PM_{2.5} concentration is 5 $\mu\text{g}/\text{m}^3$). For this approach, the emission-reduction algorithm is:

1. Check each CBSA to see whether the maximum concentration within the CBSA is lower than the concentration target.
2. For those CBSAs that exceed the concentration target, reduce the emissions for all sources and locations within the CBSA by 1% of the total emission

(i.e., proportional to sources and locations), and calculate the updated grid-cell concentrations for each CBSA.

3. Repeat step 1: Check each CBSA to see whether the updated concentrations reach the target.
4. Repeat step 2: For those CBSAs that still exceed the target, reduce all emissions within the CBSA by 1% of the remaining emission or by 1 t/y (use the larger value between the two), and calculate the updated grid-cell concentrations for each CBSA. The minimum reduction unit is 1 t/y.
5. Repeat steps 3 and 4 until all CBSAs have reached the target (the NAAQS) or reached zero emission.

The rationale for some of the specific steps in this optimization are as follows: For some CBSAs, even if we reduce all the emission within their boundary, the maximum concentration in that CBSA will still exceed the target (the NAAQS). For those cases, we avoid the optimization entering an infinite loop via these steps described above: 1) having a small emission reduction (1 t/y) as the minimum reduction per optimization step, and 2) halting the optimization if all emissions being considered have become zero.

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Data, Materials, and Software Availability. ISRM and emission data have been deposited in Zenodo (<https://zenodo.org/record/2589760> and <https://zenodo.org/record/5831940>) (73, 74). All study data are included in the article and/or *SI Appendix*.

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