Persistent Organic Pollutants in Dust From Older Homes: Learning From Lead

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Settled dust found indoors is a mixture of biologically derived materials, particles deposited from indoor aerosols, particles deposited from building materials (e.g., deteriorated paint), and soil particles that infiltrate from outdoors (e.g., soil tracked indoors on shoes).[1] Because typical cleaning removes only a portion of dust from indoor environments (e.g., when vacuuming a carpet), indoor dust acts as a reservoir for chemical contamination.[2] Settled dust can be an important source of chemical exposures, especially for young children, who have frequent hand-to-mouth contact.[3] As early as 1904, Gibson suggested the importance of dust as a route of exposure to lead, hypothesizing that "painted surfaces . . . become a dry easily detachable powder . . . carried to the mouths of children . . . who bite their nails, suck their fingers or eat with unwashed hands."[4(p302)]

More than a century after dust was first implicated in childhood lead poisoning, children's exposure to lead-contaminated dust remains a major public health issue, especially for children living in older homes.[5,6] Lead-based paint, the major source of lead in the residential environment,[7] was banned in the United States in 1978. Thus, homes that predate the ban are more likely to contain lead-based paint, and these older homes have greater lead contamination than do newer homes.[8] Numerous investigators have reported a relation between home age and lead levels in dust (Table A, available as a supplement to the online version of this article at http://www.ajph.org). In addition to lead, several organic chemicals that were used extensively worldwide have been banned, including polychlorinated biphenyls (PCBs) and organochlorine insecticides; and these contaminants likewise have been found at higher levels in dust from older homes compared with newer homes (Table B, available as a supplement to the online version of this article at http://www.ajph.org).

Objectives. We aimed to (1) evaluate the relation between home age and concentrations of multiple chemical contaminants in settled dust and (2) discuss the feasibility of using lead hazard controls to reduce children's exposure to persistent organic pollutants.

Methods. As part of the California Childhood Leukemia Study, from 2001 to 2007, we used a high-volume small surface sampler and household vacuum cleaners to collect dust samples from 583 homes and analyzed the samples for 94 chemicals with gas chromatography–mass spectrometry and inductively coupled plasma mass spectrometry. We evaluated relations between chemical concentrations in dust and home age with Spearman rank correlation coefficients.

Results. Dust concentrations of lead, polychlorinated biphenyls, organochlorine insecticides, and polycyclic aromatic hydrocarbons were correlated with home age ($\rho > 0.2$; $P < .001$), whereas concentrations of pyrethroid insecticides and polybrominated diphenyl ethers were not.

Conclusions. Dust in older homes contains higher levels of multiple, persistent chemicals than does dust in newer homes. Further development of strategies to reduce chemical exposures for children living in older homes is warranted. (*Am J Public Health.* 2014;104:1320–1326. doi:10.2105/AJPH.2013.301835)

As part of the California Childhood Leukemia Study (CCLS), we collected dust samples from 583 homes. We showed in previous multivariable models that home age was positively associated with levels of PCBs,[9] polycyclic aromatic hydrocarbons (PAHs),[10,11] and the organochlorine insecticides chlordane and dichlorodiphenyltrichloroethane (DDT)[12] in dust samples from CCLS homes. We present the first systematic evaluation of the relation between home age and concentrations of a broad suite of chemical contaminants in dust samples from CCLS homes. Because no established protocol exists to control persistent organic pollutants in homes, we describe mitigation strategies that have been used successfully for lead hazard control and discuss their potential utility in reducing children's exposure to organic chemicals.

METHODS

The CCLS is a population-based case–control study that seeks to identify genetic and environmental risk factors for childhood leukemia.

We collected dust samples from homes of case participants and control participants younger than 8 years at diagnosis (similar reference date for control participants) who were enrolled in the study from December 1999 to November 2007 and who were still living at the diagnosis home. Sampled homes were composed of a wide range of construction dates (pre-1900 to post-2000), types (single-family homes to multifamily apartments), sizes (< 1000 sq ft to > 5000 sq ft), and income levels (annual income: < $15 000 to > $75 000).

Dust Sampling

We collected dust samples from 583 CCLS homes with the high-volume small surface sampler (n = 415) and household vacuum cleaners (n = 499) from 2001 to 2007, as previously described.[13] Initially, dust was collected with both sampling methods, and samples that were collected with the standardized high-volume small surface sampler protocol[14] were used in chemical analyses. However, because chemical concentrations in dust

samples collected by high-volume small surface sampler and household vacuum from the same homes were found to be comparable,[13] starting in 2006, household vacuum sampling was used in place of the more labor-intensive high-volume small surface sampler protocol. Dust samples were collected by high-volume small surface sampler from a carpet or rug in the room where the child spent the most time while awake (commonly the family room). Vacuum dust samples were collected by removing the used bag or by emptying the loose dust from the household vacuum cleaner into a sealable polyethylene bag. Dust samples were stored in the dark at 4°C for up to 10 years prior to chemical analysis. When dust was collected with the high-volume small surface sampler, the sampling area was measured; thus, chemical concentrations (mass of chemical per mass of dust) and chemical loadings (mass of chemical per area sampled) were calculated. In contrast, when dust was collected with a household vacuum cleaner, the sampling area was unknown, and only chemical concentrations were calculated.

Chemical Analysis

We used a multiresidue analysis scheme to analyze 74 chemicals from several compound classes (organochlorine insecticides, organophosphate insecticides, carbamate insecticides, pyrethroid insecticides, phenoxy herbicides, amide herbicides, dinitroaniline herbicides, PAHs, PCBs, metals, and tobacco markers) at Battelle Memorial Institute (Columbus, OH), as previously described.[13] A complete list of analytes is available in Table C (available as a supplement to the online version of this article at http://www.ajph.org). To accommodate the diverse group of organic analytes, 3 different extraction methods were used (hexane:acetone, dichloromethane, and acetonitrile:sodium acid phosphate) in combination with gas chromatography–mass spectrometry in the multiple ion detection mode. In addition, metals were analyzed with microwave-assisted acid digestion in combination with inductively coupled plasma mass spectrometry. These initial chemical analyses used 168 dust samples (29%) collected from household vacuum cleaners and 415 dust samples (71%) collected via high-volume small surface sampler. Most of the 415 homes sampled with the high-volume small surface sampler

also provided a household vacuum sample that was not initially analyzed.

Subsequently, 292 of the available dust samples collected from household vacuum cleaners were analyzed for 20 polybrominated diphenyl ethers at the California Department of Toxic Substances Control (Berkeley), as previously described.[15] Of the 292 homes with a vacuum dust sample analyzed for polybrominated diphenyl ethers, 106 homes had a sample from the same vacuum bag that was previously analyzed for the other 74 chemicals, whereas 186 homes had a dust sample collected by high-volume small surface sampler that was previously analyzed for the other 74 chemicals. Briefly, dust samples were extracted with a 95:5 hexane:methylene chloride mixture and analyzed for polybrominated diphenyl ethers with high-resolution gas chromatography–high-resolution mass spectrometry in the multiple ion detection mode.

Questionnaire Responses

At the time of dust collection, participants were asked to identify their residence's date of construction (pre-1940, 1940–1949, 1950–1959, 1960–1969, 1970–1979, 1980–1984, 1985–1989, or post-1989) and type, their child's race/ethnicity, and their household's annual income. Of 583 respondents, 513 were able to estimate their residence's construction date; 70 respondents did not know the age of their home and were therefore excluded from statistical analyses that used home age. In the 415 homes that were sampled with the high-volume small surface sampler, participants were also asked to identify the age (in months) of the rug or carpet that was sampled (413 participants responded; carpet ages ranged from < 1 month to 30 years).

Statistical Analysis

We used nonparametric Spearman rank correlation coefficients to identify relations between chemicals, relations between chemical levels and home age (an ordinal variable), and relations between chemical levels and carpet age. Our primary statistical analysis included dust samples collected by both high-volume small surface sampler and household vacuum cleaners; thus, chemical levels were characterized by concentration. We conducted secondary statistical analyses of both chemical

concentrations and loadings for dust samples collected by high-volume small surface sampler. We also conducted analyses stratified by household annual income, child's race/ethnicity, and residence type. We set all values that were below the analytical limit of detection to a value of zero (equivalent to the lowest rank when estimating Spearman rank correlation).

RESULTS

Figure 1 shows a matrix of Spearman rank correlation coefficients between concentrations of 94 chemicals measured in 583 dust samples collected from CCLS homes. As expected, the chemicals with the strongest correlation were found within chemical classes—these relations are characterized as areas of red near the diagonal line in Figure 1. For example, several of the PAHs were highly correlated ($\rho > 0.6$) with each other (e.g., $\rho = 0.90$ for benzo[a]anthracene, chrysene), as were some of the organochlorine insecticides, pyrethroid insecticides, PCBs, and polybrominated diphenyl ethers. More interestingly, there was also moderate correlation ($\rho = 0.2$–0.6) between individual chemicals across some of the chemical classes (e.g., between some organochlorine insecticides, PAHs, PCBs, and metals)—these relations are shown in orange. For example, lead is correlated with DDT, chrysene, and PCB-180 ($\rho = 0.40, 0.37, 0.36$, respectively; $P < .001$ for each).

Table C shows the detection frequency, median, and interquartile range of each of 94 chemicals measured in 513 dust samples collected from CCLS homes, stratified by the residence construction date. Of the 94 chemicals analyzed, 21 chemicals from various classes were detected in fewer than 20% of the participating homes. We generally did not observe a significant correlation between home age and concentrations of these 21 infrequently detected chemicals.

Figure 2 shows Spearman rank correlation coefficients between home age and chemical concentrations for each of 94 chemicals measured in 513 dust samples collected from CCLS homes. Of the 94 chemicals analyzed, dust concentrations of DDT (and its metabolite dichlorodiphenyldichloroethylene) and chlordane (both the α and the γ isomers) were most positively correlated with home age ($\rho > 0.5$;
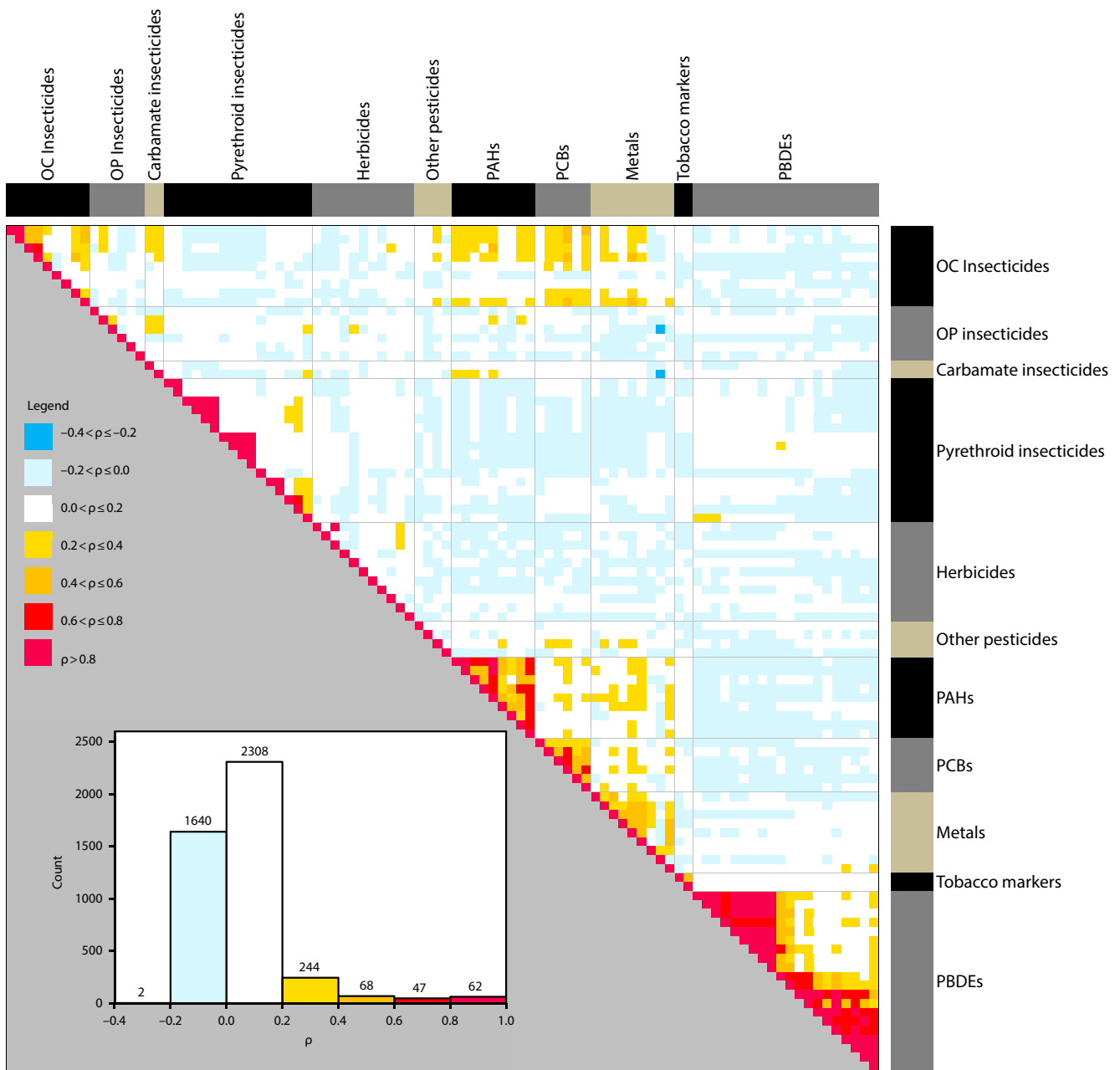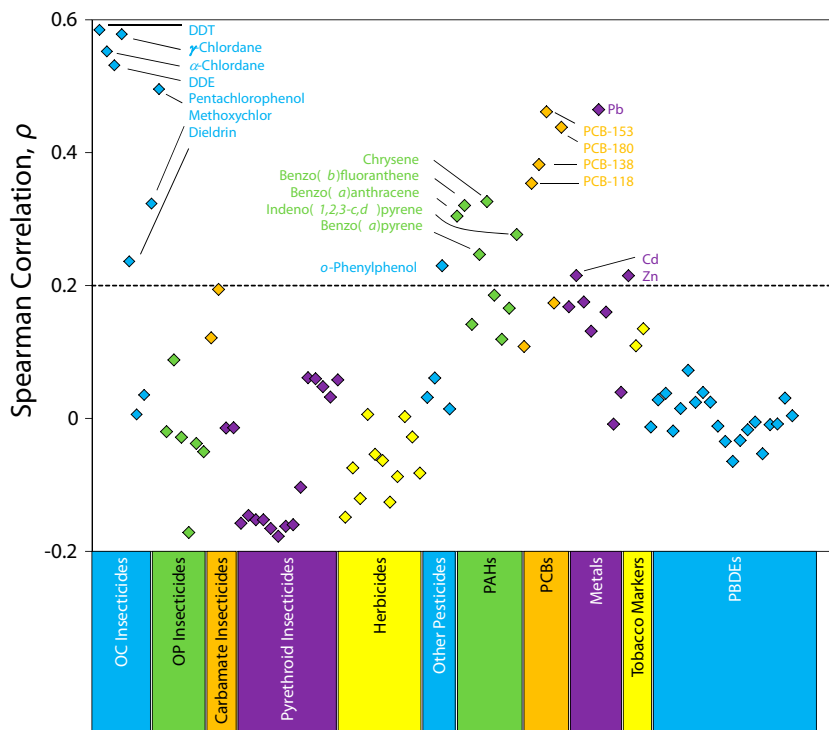
Note. OC = organochlorine; OP = organophosphate; PAHs = polycyclic aromatic hydrocarbons; PBDEs = polybrominated diphenyl ethers; PCBs = polychlorinated biphenyls. Chemicals are grouped by class, and groups are shown along the top and right borders (in black, gray, and gold). Inset is a histogram of correlation coefficients for the 94 chemicals.

FIGURE 1—Spearman rank correlation coefficients between concentrations of 94 chemicals measured in 583 dust samples collected from California Childhood Leukemia Study homes: 2001–2007.

$P<.001$), indicating that these chemicals were found at higher concentrations in dust from older homes than in dust from newer homes. Likewise, dust concentrations of 3 other organochlorine insecticides (dieldrin, methoxychlor, and pentachlorophenol), 4 PCBs, and 5 PAHs were positively correlated with home age ($\rho>0.2$; $P<.001$). We also observed significant positive correlations between home age and concentrations of lead ($\rho>0.46$; $P<.001$) and other metals (cadmium and zinc; $\rho>0.2$; $P<.001$). By contrast, concentrations of chemicals that were first produced at high volumes within the past 30 years, such as polybrominated diphenyl ethers and pyrethroid insecticides, were either unrelated to, or negatively correlated with, home age.

Note. DDE = dichlorodiphenyldichloroethylene; DDT = dichlorodiphenyltrichloroethane; OC = organochlorine; OP = organophosphate; PAHs = polycyclic aromatic hydrocarbons; PBDEs = polybrominated diphenyl ethers; PCBs = polychlorinated biphenyls. Chemicals are grouped by class and groups are shown by color along the bottom border (blue, green, orange, purple, yellow). Chemicals highly correlated with home age are labeled (if ρ > 0.2)

FIGURE 2—Spearman rank correlation coefficients between home age and chemical levels for each of 94 chemicals measured in 513 dust samples collected from California Childhood Leukemia Study homes: 2001–2007.

Compared with the relations observed in the complete data set (Figure 2), the relations between home age and chemical concentrations were similar in the subset of 415 homes sampled with the high-volume small surface sampler (Table D, available as a supplement to the online version of this article at http://www.ajph.org). For example, for the 20 chemicals that were most positively correlated with home age (those with ρ > 0.2, highlighted in Figure 2), the correlation coefficients were generally at least as large in the subset of samples collected via high-volume small surface sampler. Likewise, in the subset of 415 homes that were sampled with the high-volume small surface sampler, most of the relations between home age and chemical loadings were similar to relations between home age and chemical concentrations (Table D). One exception was lead, for which concentrations were more strongly correlated with home age

than were loadings (ρ = 0.44 and 0.29, respectively). In stratified analyses, the positive correlations between home age and chemical concentrations were observed in homes from all strata of income, race/ethnicity, and residence type (Table E, available as a supplement to the online version of this article at http://www.ajph.org).

For most chemicals, the age of the carpet or rug sampled was less strongly correlated with the chemical concentrations and loadings than was home age. For the 20 chemicals that were most positively correlated with home age, none were as strongly correlated with carpet age (ρ < 0.2 for each chemical; Table D).

DISCUSSION

We observed moderate correlations between contaminants with disparate sources,

such as lead, PCBs, organochlorine insecticides, and PAHs. Likewise, we observed relations between concentrations of these contaminants in dust and home age. On the basis of these observations, we suggest that older homes are likely to contain dust contaminated by multiple persistent chemicals from various classes.

In the United States, the production and distribution of PCBs were halted in 1979. Thus, PCB-contaminated construction materials, such as ceiling tiles, insulation, building joint caulk, floor finish, paint, and roofing material, are more likely to be present in homes built before 1980 than in more recently constructed residences.[9] Moreover, homes built before 1980 may have contained PCB-contaminated consumer products, such as fluorescent light ballasts, refrigerators, televisions, carpet pads, and air conditioners, which may have contaminated household dust or surfaces.[9] Several investigators have reported a relation between home age and PCB levels in dust (Table B). Like PCBs, many of the insecticides that were positively associated with home age in our analysis (DDT, chlordane, dieldrin, methoxychlor, and pentachlorophenol also have been restricted or banned in the United States. In support of our findings, multiple investigators have reported a relation between home age and levels of DDT or chlordane in dust (Table B).

Importantly, in addition to confirming previous reports that banned organic contaminants are found at higher levels in dust from older homes than in dust from newer homes, we discovered that PAHs—chemicals that are still being generated—are also found at higher levels in older homes. As evidenced by the decreasing trends in ambient air PAH concentrations in California,[16] many PAH sources that contributed to dust-PAH contamination in CCLS homes, including emissions from vehicular traffic, cigarettes, and fireplaces, likely diminished over time. We hypothesize that older homes were initially contaminated by these PAH sources in the past and that long-lasting PAH residues contribute to current PAH concentrations in dust.

Our systematic evaluation of the relation between home age and a broad suite of chemical contaminants complements prior findings from the CCLS, which suggested that home age was predictive of levels of PCBs,[9]

participants' homes and carpets may have been misclassified. Underlying factors not included in this analysis, such as the presence of indoor and outdoor chemical sources and the condition of the home, also may explain the observed relation between home age and chemical levels in dust. Another limitation of our analysis was that some of the chemicals we measured were infrequently detected in dust samples from participating homes, which reduced our power to identify relations between home age and levels of these chemicals.

To maximize the size of our primary statistical analysis, we included dust samples collected with both the high-volume small surface sampler and household vacuum cleaners. The major drawback of using dust samples collected from household vacuum cleaners is the absence of a consistent sampling protocol across study homes. Each model of vacuum cleaner has a unique combination of flow rate, sampling efficiency, and range of particle sizes collected. Likewise, each vacuum cleaner may have been used in a different combination of rooms and at different proximity to chemical sources. It was reassuring that our results were similar whether we included or excluded dust samples collected from household vacuum cleaners in our statistical analysis.

Because the area sampled by a household vacuum cleaner is not well defined, chemical loadings could not be calculated for vacuum samples, and, as a result, our primary statistical analysis used chemical concentrations. It is not known whether dust loadings or dust concentrations are more representative of children's exposure to organic pollutants. It has been reported that blood-lead levels are more highly correlated with dust-lead loadings than with dust-lead concentrations, suggesting that dust-lead loadings are more useful markers of childhood lead exposures.[49] In the subset of samples we collected with the high-volume small surface sampler, our results were similar for chemical concentrations and loadings.

In summary, we showed that lead, PCBs, organochlorine pesticides, and PAHs are found at higher levels in dust collected from older homes than in dust collected from newer homes. We conclude that residents living in older homes potentially experience coexposures to many of these persistent chemicals, which may result in adverse health effects,

especially for children. Whether strategies that have been successful for controlling lead hazards in older homes will be helpful in controlling organic pollutants is uncertain. Mitigation strategies for older homes with dust contaminated by organic pollutants should be developed. ■

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Contributors

T. P. Whitehead conducted the statistical analyses and prepared the article. C. Metayer co-directed the study's implementation, interpreted the data, and provided critical review of the article. M. H. Ward co-directed the study's implementation, planned the chemical analyses, supervised the laboratory analyses, interpreted the data, and provided critical review of the article. J. S. Colt supervised the laboratory analyses, interpreted the data, and provided critical review of the article. R. B. Gunier planned the chemical analyses, interpreted the data, and provided critical review of the article. N. C. Deziel interpreted the data and provided critical review of the article. S. M. Rappaport supervised the statistical analyses, interpreted the data, and provided critical review of the article. P. A. Buffler designed the study and co-directed its implementation, planned the chemical analyses, interpreted the data, and provided critical review of the article.

Human Participant Protection

We obtained written informed consent from the children's parents, and study protocols involving research with human participants were approved by the institutional review boards at the University of California, Berkeley, and all other participating institutions.

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