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Patterns of contamination and burden of lead and arsenic in rooftop harvested rainwater collected in Arizona environmental justice communities

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All survey participants were consented under the University of Arizona Institutional Review Board, which ensures the rights and welfare of human participants in research.

Abstract

As climate change exacerbates water scarcity, rainwater harvesting for household irrigation and gardening becomes an increasingly common practice. However, the use and quality of harvested rainwater are not well studied, and the potential pollutant exposure associated with its use are generally unknown. There are currently no federal standards in the United States to assess metal(loid)s in harvested rainwater. Project X, a community science research project, was created to address this knowledge gap and study the quality of harvested rainwater, primarily used for irrigation, in four environmental justice communities in Arizona, USA. Community scientists collected 577 unique rooftop harvested rainwater samples from 2017-2020, analyzed for metal(loid)s, where arsenic (As) concentrations ranged from 0.108 to 120 ug L^{-1} and lead (Pb) concentrations ranged from 0.013 to 350 ug L^{-1}; with the highest concentrations in the communities of Hayden/Winkelman > Tucson > Globe/Miami > Dewey-Humboldt. Linear mixed models were used to analyze rooftop harvested rainwater data and results indicated that concentrations of As and Pb in the summer monsoon were significantly greater than winter; and contamination was significantly greater closer to extractive industrial sites in three of the four study communities (ASARCO Copper Smelter in Hayden/Winkelman, Davis-Monthan United States Air Force Base in Tucson, and Freeport McMoRan Copper and Gold Mine in Globe/Miami). Based on linear mixed models, rainwater harvesting infrastructure such as roof material, presence of a cistern screen, and first-flush systems were not significant with respect to As and Pb when controlling for relevant spatiotemporal variables. These results however, indicate that concentrations vary seasonally and by proximity to industrial activity, not by decisions made regarding collection system infrastructures at the individual home level. This study shows that individuals are not responsible for environmental contamination of rooftop harvested rainwater, rather activities and decisions of government and corporate industries control contaminant release.

Keywords: environmental justice; mixed model; community science; rainwater harvesting; arsenic; lead

Abbreviations

Project X: PX
University X: (UX)
Rooftop Harvested Rainwater: RHR
National Atmospheric Deposition Program: NADP
Community Organization X: CX
Community Scientists: CS
US Department of Defense Davis-Monthan Air Force Base: AFB
Tucson International Airport Area: TIAA
Arizona Trail System: ATS
Home Description Survey: HDS

1. Introduction

The United Nations Sustainable Development Goal 6 progress report states that 2.3 billion people live in water-stressed countries (United Nations, 2021). Only 14 out of 109 countries (excluding US, much of Europe, India, parts of North Africa, and others) report having high levels of community participation. Water scarcity could force migration for more than 700 million people by 2030 (United Nations, 2021). As climate change worsens water scarcity (Schewe et al., 2014), we will become more reliant on alternative sources of water (Pearson et al., 2015), such as rooftop harvested rainwater (RHR). However, the impacts of environmental pollution on RHR remain understudied.

Environmental pollution was responsible for an estimated nine million premature deaths globally in 2015 (Landrigan et al., 2018). Pollution caused by industrial activity, mechanized agriculture, and mining are on the rise, while household-scale pollution is decreasing (Landrigan et al., 2018). Arizona (AZ), has 380 active mines, and is second after Nevada for non-fuel related mineral production in the US (Conway, 2019; Richardson et al., 2019). Mining is a top contributor of pollutants, including arsenic (As) and lead (Pb), into the environment in terms of quantity, land area, and toxicity (Csavina et al., 2012), which is interconnected with climate change, increased industrialization, and ecosystem destruction (Csavina et al., 2012; Odell et al., 2018; Phillips, 2016). This may impact the quality of RHR in AZ.

In response, community and citizen science efforts strive for community engagement to address environmental injustices (David-Chavez et al., 2020; Author et al., 2021a; Pandya, 2012). Community science is research stemming from the public to contribute to grassroots social action (Cooper et al., 2021; Pandya, 2012; Wilson et al., 2014);

and, citizen science is research led by institutions involving the public in parts of the scientific process (Shirk et al., 2012). Community generated data is commonly considered unreliable; however, multiple studies observed that community generated data are on par with professionally collected data (Bowser et al., 2020; Danielsen et al., 2014; de Sherbinin et al., 2021; Kosmala et al., 2016). Aceves-Bueno et al. (2017) recommend training, investing in long-term data collection, and discuss that when participants have an economic or health stake in the research, they perform 68% better than less engaged participants. Regardless of who collects the samples, generated datasets can suffer from issues of non-independence, noise, and non-standard effort, which can be resolved through statistical methods (Bird et al., 2014; Kosmala et al., 2016). Environmental data are often nested and crossed with several levels of plots and subplots (Schielzeth & Nakagawa, 2013), have missing data, and lack true independence (Bolker et al., 2009; Ogle, 2009). Mixed and hierarchical models provide opportunities to account for and integrate the complexities of these data, especially for community generated data (Bird et al., 2014).

Here, we analyze results from The University X's Project X (PX), a community science research program developed in partnership with the Community Organization X (CX); working with rural communities near active and legacy mining and one urban city to address environmental injustices and answer community-driven research questions (Author et al., 2018a, 2019, 2020, 2022a). In addition to participatory community research methods (Hébert et al., 2015; Shirk et al., 2012), we utilize environmental health justice and informal science education frameworks; our analyses are informed by critical environmental justice (EJ) (Pellow, 2016) and Indigenous EJ (Hernandez, 2019; Spencer et al., 2020).

We focus on As and Pb in RHR because of their toxicity (World Health Organization, 2018, 2019), community concern, and iterative statistical analysis. The US EPA categorizes As is a carcinogen and Pb as a probable carcinogen (US EPA IRIS, n.d.-a, n.d.-b); and, exposure to any concentration of Pb is unsafe (US EPA, 2016; World Health Organization, 2019). Exposure to As has been shown to cause gastrointestinal, endocrine, respiratory, and cardiovascular health issues potentially resulting in death (ATSDR, 2007; Naujokas et al., 2013). Similarly, Pb is a known endocrine disrupter and harms the respiratory, cardiovascular, and neurological systems (ATSDR, 2020; Author et al., 2022b; National Toxicology Program, 2012). Immunocompromised, pregnant, and young people are most at risk.

We hypothesize that: (1) As and Pb concentrations of RHR are elevated in study sites near industrial activity compared to control background levels and their concentrations increase closer to sites of extraction and (2) that rainwater harvesting infrastructure and home design do not have a meaningful impact on RHR quality.

2. Methods

2.1 Project X

Trained community scientists (CS) collected and submitted RHR, along with garden soils and garden plants, from the communities of Dewey-Humboldt, Globe/Miami, Hayden/Winkelman, and Tucson, AZ (Figure 1, SI Table 1 for number of submitted samples per year) (Author et al., 2018a, 2019, 2020). The majority of CS are from EJ communities; over half of PX CS self-identify as people of color (primarily Latinx/Hispanic), are low-income based, and do not have a college degree; see Author et al., 2020 and Author et al., 2022a for further details.

In addition to a co-created community science design, PX employed promotoras (community health workers) (Author et al., 2015a, 2018b), who were trained to support their community across all sampling methods (Author et al., 2020). Promotoras and community members participated in most parts of the research process (Author et al., 2019). Yearly English and Spanish data sharing events were held in each of the partner communities with co-produced results booklets, presentations, focus groups, and an interactive art installation, *Ripple Effect* (Author et al., 2021c). Samples were analyzed for a variety of microbial, organic, and inorganic contaminants (see: https://projectharvest.arizona.edu/). Here we focus on the analysis of As and Pb in the RHR.

2.3 Sample Collection

CS collected RHR samples during four prescribed sampling windows each water year from December 2017 to February 2020, totaling 577 samples. Historical data from the National Weather Service informed the sampling windows, which were designed around the binary rainfall regime of the North American monsoon and winter wet seasons (NWS, 2017). Sample collection were during the first winter (December to January), last winter (February), first monsoon (June to July), and last monsoon (September). First and last collections were operationally defined as sampling period; winter and monsoon collections were named sampling season.

CS collected RHR, after flushing the first 10 seconds of water, in trace metal-free 50 mL tubes (VWR, Cat. Number 89049-17). Field blanks were collected at the rainwater harvesting system by carefully transferring 50 mL of supplied field blank type I laboratory water (>18 MΩ deionized nanopure water, Millipore) using the same trace metal-free tubes. Each sample was sealed, recorded on a chain of custody form, and delivered to the UX where they were stored at 4° C prior to analyses.

2.4 Site Descriptions

2.4.1 Background Sites

PX partnered with the National Atmospheric Deposition Program (NADP) National Trends Network to obtain wet-only deposition rainwater samples (National Atmospheric Deposition Program, 2019). Five NADP sites were used in AZ (Figure 1). The 78 background samples were collected by NADP personnel and shipped to the UX for contaminant analysis. NADP samples were categorized into the PX sampling periods and season. No significant differences were observed between NADP sites, therefore these data were aggregated and defined as AZ background rainwater.

2.4.2 Dewey-Humboldt, AZ USA

Dewey-Humboldt is a rural community (pop. 4123 in 2020) in Yavapai County, AZ, near the US EPA Iron King Mine-Humboldt Smelter Superfund site (Author et al., 2015b, 2015c, 2016; US EPA, n.d.-b). PX sites in Dewey-Humboldt were categorized by proximity and direction from the Superfund site, which includes more than 64 million tons of contaminated tailings (US EPA, n.d.-c). The distances from each site to the tailings sources were calculated to explore the association between proximity from a point source and contamination in RHR.

2.4.3 Globe/Miami, AZ USA

Globe/Miami is a rural community in Gila County, AZ (pop. 9250 in 2020) with the active Freeport McMoRan Copper and Gold Mine (Niemuth et al., 2008), plus other sites of extraction such as the Pinto Valley Mine. Sites in Globe/Miami were geopolitically categorized by sub-locations for comparison (Figure 1). Then, the distance from each collection site within the geopolitical categories to the border of the mine were calculated to explore the association between distance from a point source and contamination in RHR.

2.4.4 Hayden/Winkelman, AZ USA

Hayden/Winkelman is a rural community in Gila and Pinal Counties, AZ (pop. 1178 in 2020) adjacent to an active copper smelter run by ASARCO Grupo Mexico (Author et al., 2021b, 2021d). Hayden (west of smelter) and Winkelman (south of smelter) were compared to assess the differential impacts of the smelter on community contamination (Figure 1). The distance between sampling sites and both the ASARCO smelter and closest distance to tailings were calculated to explore the association between point-source proximity and contamination in RHR. During this study, workers at the smelter went on strike (October 13th, 2019 – July 6th, 2020) to protest unfair labor practices and effectively shut down smelting operations (Broussard, 2019; International

Brotherhood of Boilermakers, 2020), creating a fortuitous experiment to assess the impact of halting smelting operations.

2.4.5 Tucson, AZ USA

The Tucson metropolitan area in Pima County, AZ (pop. ~1 million, 2020), was the urban community in this study. Over the course of PX, 59-75% of the urban CS were from southern metropolitan Tucson area and we consider Tucson to be an EJ community (Author et al., 2020). Households were categorized to their Ward or nearest Ward (Figure 1) (*Ward Maps for Tucson*, 2022). Analyzing contamination by Ward is important due to stakeholder interest, unjust practices of gentrification, displacement, and environmental racism impacting the most marginalized areas of the city (Bristol et al., 2020; Williams & Florez, 2002).

It was challenging to select a contamination point source in the Tucson metropolitan area because of the numerous potential sources. The US EPA Toxic Release Inventory (TRI) data indicated that the US Department of Defense Davis-Monthan Air Force Base (AFB) releases the most Pb and/or Pb compounds to the environment (2.2 tons/year), via “Onsite Other Disposal” (US EPA, 2019). The reported release comes from the use of munitions, known to release Pb to the soil, dust, and air (Laidlaw et al., 2017; National Research Council, 2013; Pain et al., 2019), though the TRI data does not report fugitive air emissions (US EPA, 2019). There is likely an additional contribution of Pb to the environment through the use of aviation gasoline. Based on an EPA technical support document, Tucson International Airport (TIA) was estimated to release 0.5 tons of Pb to the air through leaded gasoline (US EPA, 2008).

These sites provide potential pathways for RHR contamination through wet and dry deposition. The AFB and TIA are located in an industrial area southwest of the city center, near other TRI sites, Raytheon Missiles and Defense, and close to areas highly vulnerable to gentrification and displacement. This is also the most racially diverse area in the city, has the highest poverty rates, and the most industrial activity (Bristol et al., 2020; Author et al., 2015a). The distances between each sampling site and the AFB and TIA were calculated to explore the association between proximity to point-source and contamination of RHR.

2.4.6 Arizona Trail System

The Arizona Trail System (ATS), which supplies harvested water for potable use for hikers and recreators on the cross-state trail system, submitted four RHR samples from a cistern in Pinal County, AZ (Figure 1). These samples provide a novel snapshot comparison to home sites because they were collected in an area with relatively little human activity, similar to the NADP samples, but with rainwater harvesting infrastructure like other PX samples. These samples were not included in statistical analyses due to their uniqueness.

2.5 Home Description Survey

Promotoras administered a home description survey to the CS to gather information about RHR infrastructure and environmental perceptions. All participants consented under the UX Institutional Review Board and survey and consent forms were administered in the participant’s primary language, see Author et al., (2022a). Roof material, home age, Pb paint testing, presence of paint chips, proximity to major roadway, cistern material, cistern age, cistern screen, and presence or absence of a cistern first flush system were considered for this study.

2.6 Sample Analysis

Water samples were analyzed for dissolved metal(loid) (Be, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Mo, Ag, Cd, Sn, Sb, Ba, Pb) concentrations via inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700) at the Arizona Laboratory for Emerging Contaminants (ALEC). Briefly, a 20 mL aliquot of RHR was filtered to 0.45 µm, and acidified with 0.2 mL of 3.2 M nitric acid. Sample pH and electrical conductivity (EC) were analyzed after daily instrument calibration (Fisher XL-20, Accumet electrode).

The quality control/quality assurance (QA/QC) protocol was adapted from U.S. EPA Method 200.8 (U.S. EPA, 1994). Calibration standards for the ICP-MS were prepared from multi-element stock solutions (SPEX Certiprep, Metuchen, NJ). Calibration curves included at least seven points with correlation coefficients >0.995. The QC protocol included a continuing calibration blank (CCB); a continuing calibration verification (CCV) solution; and at least one quality control sample (QCS) to be analyzed just after calibration, again after every 12 samples, and at the completion of the run. The QCS solutions were from an independent source, such as NIST SRM 1643e Trace Elements in Water. Acceptable QC responses were between 90 and 110% of the certified value. Lastly, a suitable internal standard (usually Rh, In, Ga or Ge) was added using in-line addition into the sample line and mixing tee. In addition, reagent blanks, dilution duplicates, and spiked duplicates were run with every 20-25 samples analyzed as further QA/QC.

2.7 Statistical Analysis

2.7.1 Method Limit of Detection and Non-detected analytes

ICP-MS instrument detection limits were multiplied by laboratory preparation factors to obtain method limits of detection (MLOD) (see Data in Brief). Sample concentrations below the MLOD were replaced using (1) as a conservative estimate of the actual concentration.

$$\frac{MLOD}{\sqrt{2}} = \text{substitution} \qquad (1)$$

2.7.2 Exceedances

No U.S. federal regulations exist for RHR, so various federal and AZ State standards and recommendations were used to assess the quality of RHR (Data in Brief). Since the majority of PX CS used their RHR for watering gardens, trees, and/or landscaping (Author et al., 2022a), the USDA's recommended maximum concentrations in irrigation water "for continuous use on all soils" were used for comparisons (Pick, 2011).

2.7.3 Correlation and Multiple Factor Analysis (MFA)

Correlations between metal(loid)s in the RHR were calculated for the whole PX dataset and for each community individually, using Spearman tests due to non-normality of the data. The purpose of a MFA (Abdi et al., 2013) is to quantify the overall variation in the data and assess which variables contributed the most to the observed variation and warrant further analysis. Here, an MFA was conducted on geometric means of the dataset by community, sampling season, and sampling period for a total of 16 values per analyte (SI Table 2). Sampling season and sampling period were included as categorical variables grouped as “time”. Community was included as a categorical variable grouped as “location”. The nineteen inorganic analytes were included as scaled continuous variables grouped as “metals”.

2.7.4 Background Modeling

Linear mixed models, an extension of the linear model that allows for fixed and random variables (Bates et al., 2015; Quinn & Keough, 2002), were used to assess the spatiotemporal variation in As and Pb concentrations and compare PX communities to AZ background levels. The data and residuals were natural log transformed to achieve normality. The variables, community, and site were included as nested random intercept effects to account for the spatiotemporal autocorrelation of repeated sampling from the same sites within communities over the course of the study (Bates et al., 2015; Schielzeth & Nakagawa, 2013). A full model was fit using a maximum likelihood method to compare fixed effects, with all variables and interactions (Quinn & Keough, 2002) of interest (sampling season, sampling period, community, and all interactions) (Hajduk, 2019). Year was not included as a variable because different sites were sampled at different frequencies each year, and there were not enough levels to include it as a random variable.

Variables with variance inflation factors (VIF), a measure of multicollinearity, greater than five were iteratively removed (Akinwande et al., 2015). Then, a combination of Akaike Information Criterion (AIC) comparisons and likelihood ratio tests were used to find the best fit model (Bates et al., 2015; Hajduk, 2019; Zuur et al., 2009). Final models were refit with the restricted maximum likelihood method for more accurate error calculations (Gumedze & Dunne, 2011; Hajduk, 2019). Contrasts were calculated via pairwise least-square means tests with Tukey's honest significant difference test (Tukey's HSD).

2.7.5 Community Modeling

A similar protocol as described above was used to model community-specific contaminant trends with site as a random effect. Sample sizes were lower for community models, so the number of variables and interactions included in maximal models was restricted to the most relevant based on full dataset modeling and researcher knowledge. See Table 1 for all maximal models. Additional models were run on Hayden/Winkelman winter sampling season data to assess the impact of smelter status (i.e., strike shut down) on contamination. The Hayden/Winkelman As models had a singular fit because the variance and standard deviation of the random effect was zero, so these data were refit with linear models, without random effects.

2.7.6 Home Description Survey

Linear mixed models were built to assess the influence of nine participant self-reported home characteristics listed in section 2.5 on As and Pb concentrations in RHR (Table 2). Each characteristic (e.g., roof type) was modeled separately using a linear mixed model method as described above where the maximal models included variables from the final As and Pb background models. For As, the maximal model included community, sampling period, sampling season, survey question, and two-way interactions with survey questions. For Pb, the maximal model included community, sampling season, survey question, and two-way interactions with survey questions. These tests specifically gauge the association between participant home characteristics and contamination in RHR, controlling for significant spatiotemporal variables. Models were re-fit using the restricted maximum likelihood method and contrasts were calculated via pairwise least-square means tests with Tukey's HSD for home description survey variables.

2.7.7 Software and Data

Mapping and distances from point sources of pollution to sites were calculated using Google Earth Pro. The 2019 TRI site data for AZ was downloaded from the US EPA website on May 30th, 2021 (US EPA, 2021). All statistical analyses were conducted using R software in RStudio (R Core Team, 2020). The packages, “Hmisc” (Harrell Jr. & Dupont, 2021), “factoMineR” (Lê et al., 2008), “factoextra” (Kassambara & Mundt, 2020), “lme4” (Bates et al., 2015), “lmerTest” (Kuznetsova et al., 2017), “performance” (Lüdecke et al., 2020), “emmeans” (Lenth et al., 2021), “EnvStats” (Millard, 2013), “dplyr” (Wickham et al., 2021), “ggplot2” (Wickham, 2016), “ggeffects” (Lüdecke, 2018), and “patchwork” (Pedersen, 2020) were used.

3. Results

3.1 As and Pb Detection and Exceedances

Arsenic was detected most frequently in Hayden/Winkelman. Pb was detected in every RHR sample analyzed (N = 577), except for one sample in Tucson from the first winter sampling window. Only one RHR sample, from Hayden/Winkelman, out of 577 samples exceeded the USDA's recommended maximum irrigation concentration for continuous use on all soils of 100 µg L^{-1} for As; and no samples exceeded the Pb standard of 5000 µg L^{-1}. Additionally, none of the AZ background samples (N = 78) exceeded the USDA recommended maximum irrigation concentrations for As or Pb (Pick, 2011). See Data in Brief for additional details on detections and exceedances.

3.2 Correlations and MFA

In PX communities overall, As was moderately positively correlated (rho > 0.50) with V and Ba, Pb was moderately positively correlated with Al, Mn, Co, and Cu (SI Table 3). For individual community correlations see SI Tables 4-7. In the AZ background rainwater, As was moderately positively correlated to V and Se and moderately negatively correlated to Mo, Cd, and Ba, and Pb was moderately positively correlated to Al, Mn, Cu, and Ba (SI Table 8). Note that AZ background results may be confounded due to low concentrations.

The first two dimensions, or principal components (Abdi et al., 2013), of the MFA, which are "time" and "metals", explained 27.8% and 18.2% of the data respectively (SI Figure 1) and are most influential in the analysis (SI Figure 2). Figure 2A displays metal(loid) contributions to variation along dimension one and two. In general, samples varied most by community and sampling season, with sampling season being most important because it varied more strongly along the first dimension (Figures 2B, 2C). Samples did not vary greatly by period as indicated by overlapped ellipses (Figure 2D).

3.3 Summary Statistics

PX samples (n=520) pH ranged from 3.50 to 8.49 with a median of 5.88, and EC ranged from 0.180 to 3116 uS cm^{-1} with a median of 42.27 uS cm^{-1}. The highest geometric mean As concentration was 9.68 µg L^{-1} – almost exceeding the US EPA drinking water standard (US EPA, 2015) and USDA's recommended upper limit for livestock and poultry (Pick, 2011) – and the highest geometric mean Pb concentration was 2.65 µg L^{-1}, both in Hayden/Winkelman from a first monsoon sampling window.

When comparing the ATS concentrations to PX and NADP values by sampling window, ATS As was lower than all the geometric means for the corresponding sampling windows, except for last winter (1.014 µg L^{-1}), which was higher than AZ background, Dewey-Humboldt, Globe/Miami, and Tucson last winter geometric means (0.610 µg L^{-1}, 0.854 µg L^{-1}, 0.655 µg L^{-1}, and 0.713 µg L^{-1} respectively). Concentrations of Pb from ATS were lower than all geometric means except last monsoon (0.6947 µg L^{-1}), which was higher than AZ background and Dewey-Humboldt (0.424 µg L^{-1} and 0.153 µg L^{-1} respectively). Importantly, all ATS samples were well below the drinking water standard for As and the maximum contaminant level for Pb, which is the main use for the RHR from this location. See Data in Brief for additional details on summary statistics and data access.

3.4 Background Modeling

Results for background models are tabulated in Table 3 and visualized via effect plots in Figure 3. Arsenic concentrations varied significantly by community, sampling season, and sampling period. Based on a pairwise least square means test with Tukey HSD p-value adjustment, Dewey-Humboldt, Globe/Miami, Hayden/Winkelman, and Tucson each had significantly greater As than AZ background (each $p < 0.05$ SI Table 9). Additionally, Hayden/Winkelman As concentrations were greater than Dewey-Humboldt, Globe/Miami, and Tucson (each $p < 0.05$). The first sampling period for all locations and years was significantly greater than last ($p < 0.05$) and monsoon sampling season was significantly greater than winter ($p < 0.05$) (SI Table 9). There were no significant interactions between the tested variables indicating that each variable trend was consistent across all levels of other variables. For example, monsoon sampling season was greater than winter for every community.

Lead concentrations varied significantly only by community and season. Based on a pairwise least square means test with Tukey HSD p-value adjustment, Hayden/Winkelman and Tucson were almost significantly greater than AZ background Pb concentrations ($p = 0.0594$ and 0.107 respectively, SI Table 9). Hayden/Winkelman and Tucson each had significantly greater Pb concentrations than Dewey-Humboldt (each $p < 0.05$). The monsoon sampling season was significantly greater than winter for Pb ($p < 0.05$) (SI Table 9).

3.5 Community Modeling

Community specific model results are tabulated in SI Table 10 and visualized via effect plots in Figure 4. For Dewey-Humboldt data, concentrations of As were greater during the monsoon sampling season compared to winter ($p < 0.05$) (SI Table 11). Distance from eastern and western tailings was not significant for either As or Pb. Pb did not significantly vary by any variable tested when location was removed from the maximal model due a VIF > 5. However, when location was kept in the model it was the only significant variable. Higher concentrations northeast of the Superfund site compared to northwest were nearly significant based on a pairwise least square means test with a Tukey HSD p-value adjustment ($p = 0.0710$) (SI Table 11).

In Globe/Miami, distance from the mine was removed due to a VIF greater than five, but was more significant than geopolitical location based on an ANOVA on the maximal model, so distance was added back into the variable pool and remained significant in consecutive models with a VIF < five. Also, the distance from the mine and geopolitical location were generally collinear, with distance being more a significant variable for both As and Pb. Based on final models, ln[As] decreased by 0.0689 µg L^{-1} with every kilometer increase in distance from the mine ($p < 0.05$) and ln[Pb] decreased by 0.201 µg L^{-1} with every kilometer increase ($p < 0.05$) (SI Table 10).

In Hayden/Winkelman, linear modeling indicated that distance from the tailings, distance from the smelter, location, and sampling season were all significant for As, but season and distance from the smelter were most significant. Distance from tailings was removed from the model because generally, an increased distance from the tailings corresponded to a decreased distance to the smelter. Hayden had higher As concentrations than Winkelman ($p < 0.05$) (SI Table 11). For Pb, sampling season and distance from the smelter were significant. For every kilometer away from the smelter, ln[As] decreased by 1.06 µg L^{-1} ($p < 0.05$) and ln[Pb] decreased by 2.07 µg L^{-1} ($p < 0.05$) (SI Table 10). While smelter operation status, as a result of the strike, did not significantly influence As or Pb concentrations (only winter analyzed), there was a decrease in As contamination during the strike and smelter shut down (SI Figure 3).

In Tucson, Pb concentrations varied by season and distance from Davis-Monthan AFB, with ln[Pb] significantly decreasing by 0.0814 µg L^{-1} with every kilometer away from the AFB ($p < 0.05$) (SI Table 10). Ward and distance from TIAA were not significant for either As or Pb.

3.6 Home Description Survey Modeling

See SI Table 12 for summary of answers to HDS questions analyzed. Study wide, linear mixed models indicated that roof material, positive paint lead test, presence of paint chips, proximity to roadway, cistern first flush, and cistern screen were not significantly associated with changes in As or Pb concentrations in RHR (Table 2). Home age was significant for both As and Pb, but the pairwise least square means test with Tukey HSD adjustment indicated no significant differences (SI Tables 13 and 14). Cistern age was also significant for Pb, with cisterns 0-2 years in age significantly lower in Pb concentrations than those 5+ years in age ($p < 0.05$) (SI Tables 13 and 14). See SI Figure 4 for effect plot visualizations of results from home age and cistern material analyses.

4. Discussion

4.1 Implications of Project X

Compared to the USDA recommended irrigation concentrations (Pick, 2011), RHR was safe for irrigating plants with regards to As and Pb. There may be additional exposures not captured by recommendations or regulatory standards. CS may need to treat RHR, depending on use. See Authors et al., 2022 and associated Data in Brief for full summary. It is important to understand the influences on water quality because there is no safe level of human exposure to Pb (US EPA, 2016; World Health Organization, 2019) and the USEPA Maximum Contaminant Level Goal for As in drinking water is zero (US EPA, 2015). Metal(loid) contamination also negatively impacts the whole ecosystem (Briffa et al., 2020).

Data and information standards apply homogenous rules to heterogenous systems, not accounting for the specificity and diversity of local epistemologies or inequities. (Montenegro, 2019). These same critiques apply to environmental standards, which express an allowable level of contamination informed by use, social and economic development, exposure assessments, feasibility of clean up, and detection limits (US EPA, 2014, 2017). They rarely integrate place-based knowledge, local contexts, and power relations, leading to bias or cultural and environmental irrelevance (Ranco & Suagee, 2007; Scammell et al., 2009; Senecah, 2004).

This study observed that sampling season was generally the most significant variable in models, with summer monsoon concentrations greater than winter (Table 3 and SI Table 10), a trend also seen in Newcastle, Australia, which is a humid subtropical city surrounded by mining industry (Martin et al., 2010). In AZ, the seasonal trend could be due to increased storm and dust activity in the summer compared to winter (Moreno-Rodríguez et al., 2015; Reheis & Urban, 2011; Sorooshian et al., 2011) increasing both dry and wet deposition. Vegetation cover reduces dust formation (Li et al., 2007), and in AZ, an arid ecosystem with frequent drought and haboobs, aeolian dispersion of dust is a likely source of contamination. Dust also becomes contaminated from mining, as described by Csavina et al. (2011, 2012), Author et al. in Dewey-Humboldt (2016) and Author et al. in Hayden/Winkelman (2021d).

We observed statistically higher than background concentrations of As in all communities and nearly significantly higher Pb in Hayden/Winkelman and Tucson, indicating that a portion of As and Pb contamination of RHR is anthropogenic (Figure 3, Table 3, SI Table 9). PX values are generally lower than other semi-arid/arid communities impacted by industry (Bolivia and Palestine) (Table 4). However, few studies report As and Pb concentrations in RHR from arid regions and further monitoring or meta-analysis should be done to properly compare contamination globally.

The NADP could invest in community-based monitoring and expand their data sharing efforts of rainwater quality across the country, particularly with tribal nations and rural communities, which are generally located closer to current NADP sites. These communities may be highly dependent on rainwater and therefore, highly impacted by rainwater quality. Having NADP and PX data follow open science FAIR (Findable, Accessible, Interoperable, Reusable) principles (Wilkinson et al., 2016) and Indigenous Data Governance CARE (Collective Benefit, Authority to Control, Responsibility, Ethics) principles (Carroll et al., 2020), can greatly benefit Indigenous, non-Indigenous, scientific, and non-scientific audiences (Carroll et al., 2021).

PX community concentrations also varied. The urban community of Tucson had comparable As and Pb contamination to Hayden/Winkelman, a community with an active smelter; both communities had higher As and Pb concentrations than Dewey-Humboldt, which has a Superfund Site (Figure 3, SI Tables 10 and 11). The community variability may be due to differences in meteorological trends, percent plant cover, federal support and remediation progress, and/or household distances from point sources.

Tucson had numerous potential sources of As and Pb contamination including TRI sites, metal recycling facilities, factories, and other common sources of urban pollution. For consistency, we assessed US EPA TRI “form R” reports and selected the Davis-Monthan AFB as a point source of contamination. We included the TIA in our analysis as there are multiple routes of contamination of RHR in an urban setting. Pb concentrations significantly decreased further away from the AFB, indicating an impact on RHR by military and surrounding industrial activity (Figure 4, Table 3). No significant relationship between As and the AFB or As and Pb and airport was observed.

In Hayden/Winkelman, others observed higher contamination compared to Tucson in settled dust on playground equipment (Author et al., *in prep*) and aerosols (Csavina et al., 2011). Furthermore, a worker’s strike shut down smelter operations starting on October 13th, 2019 and potentially resulted in the observed decrease of average winter As concentrations and a decreased range of Pb concentrations in RHR (SI Figure 3). Indoor/outdoor dust levels also decreased after the smelter shut down (Author et al., 2021d). The corporate response to union organizing to shut down smelting operations potentially reduced contamination. This study builds on decades of academic and EPA evidence of pollution by ASARCO (Csavina et al., 2011, US DOJ, 2015; Sicotte, 2009; US EPA, n.d.-a; US EPA, 2020; Wilson & Thomas, 1998) and affirms that it is important for residents of mining communities to be aware of their potential pollution exposure, have access to data, and be part of the public health decision making process.

When controlling for community and seasonal variables, older homes were somewhat associated with increased Pb and As concentrations in RHR (SI Figure 4, SI Tables 13 and 14). However, in general, participant choices about homes did not greatly impact rainwater quality (Table 2). This contrasts what other studies observed, where significant differences were observed by roof material (Hart & White, 2006; Mendez et al., 2011), presence of first flush (Gikas & Tsihrintzis, 2017), and infrastructure in general (Chubaka et al., 2018; Lee et al., 2010).

It is difficult to conduct a direct comparison to the aforementioned studies due to differences in methodology and sampling locations/times. For example, they did not employ community-based EJ research methods, coupled with statistical modeling controlling for spatiotemporal variables. Here, a mixed statistical methods approach including linear mixed models was necessary to analyze the complex community science dataset (Bird et al., 2014; Hill et al., 2016; Le Féon et al., 2016).

4.2 Linking PX to Global Challenges

With climate change exacerbating drought and water scarcity (Schewe et al., 2014), and increased industrial activity (Behrens et al., 2007), we will become more reliant on alternative sources of water (Pearson et al., 2015), including RHR. While results from this study indicate RHR is currently safe for irrigating gardens in AZ, we should follow the precautionary principle (Hanson, 2018; Mushak, 2011).

Launching from this work, it is critical to discuss the systems that influence global environmental pollution. Capitalist relationships to the Earth via colonial land theft, genocide, patriarchy, and chattel slavery necessitate increased reliance on interconnected extractive industries and the State because these relationships require exploitation of people and the environment for profit, at the cost of socioecological health (Moore, 2017; Pellow, 2016; Velicu, 2020). Pollution and colonialism are inseparably related (Liboiron, 2021) and oppression cannot be omitted from environmental analysis for marginalized peoples (Núñez et al., 2020; Whyte, 2017).

Industrial mining is an extractive industry driven by capitalist economies and has been linked to (a) militarization (Keeling & Sandlos, 2009; Simbulan, 2016; Tsing, 2000); (b) unfair labor practices and prisons (Bezuidenhout & Buhlungu, 2011; Perdue, 2018; Sicotte, 2009); (c) environmental contamination (Csavina et al., 2011; Emmanuel et al., 2018; Phillips, 2016; Razo et al., 2004; Sims et al., 2013); (d) negative human health impacts (Entwistle et al., 2019; Mwaanga et al., 2019; Patra et al., 2016; Phillips, 2016), including emotional/psychological harm (Velicu, 2020); (e) colonization (Curley, 2018; Goldtooth, 2004; Hall, 2013; Keeling & Sandlos, 2009; White, 2013a, 2013b), and (f) disproportionate impacts on already marginalized people (Urkidi & Walter, 2011; White, 2013b) across the globe. Mining is informed by systems of oppression and contributes to the continued exploitation of people and the planet (Bledsoe & Wright, 2019; Curley & Lister, 2020). Even though sustainable practices have been proposed (Author et al., 2018c), land grab mining projects continue to move forward and perpetuate environmental injustices because of the capital at the disposal of corporations, such as the Resolution Copper and Rosemont mines in AZ (Abbott, 2022; Krol, 2022).

PX community-level modeling demonstrated significant associations to point sources of pollution in three out of four study areas, including the ASARCO Copper Smelter (Hayden/Winkelman), Freeport McMoRan Copper and Gold Mine (Globe/Miami), and Davis-Monthan US AFB (Tucson) (Figure 4, SI Tables 10 and 11). The Project X dataset described throughout the paper illuminates that in several cases, the mining industry and military are responsible for environmental contamination (not CS), and should be responsible for remediation. The sociopolitical status of mining and military plays a part in the observed contamination.

The combination of community-based research and mixed statistical methods illuminate the significant role that industrial activity and therefore systems of power play in environmental pollution. Communities called “sacrifice zones”, “throw-away communities”, or “fence-line communities” (e.g., Bullard, 2011; Lerner, 2010) bear the burden of extractive economies, but they are not the ones causing the contamination or injustices. This transdisciplinary project pulls from environmental science, critical theory, and environmental sociology to demonstrate that environmental scientists cannot create the evidence-based connections needed to alleviate injustices without considering sociopolitical and environmental contexts. Future research should pay particular attention to the role of systems of oppression and liberation on environmental pollution, disproportionate pollution exposures, and environmental change.

5. Limitations

Limitations of this work include some inconsistent sampling, which was handled best by linear mixed models. We did not have a direct control to background concentrations because NADP samples were not associated with any rainwater harvesting infrastructure, but the comparison still provides a novel assessment of rainwater quality impacted by anthropogenic activity. In Tucson, it was challenging to select point sources of contamination (described in section 2.4.5). Additionally, the demographics of CS in this study are not representative of the entire state. Limited-income and Hispanic/Latina/o/x populations were overrepresented compared to state demographics. While we were close to statewide representation of Asian/Pacific Islanders and those with two or more race/ethnicities, we did not have any Black/African American or Indigenous participants (see Author et al., 2020 and Author et al., 2022a for recruitment and socio-demographics details). Ultimately, we present one assessment of environmental contamination, using western scientific statistics and data science for justice, but recognize that the methods discussed here may not be the most appropriate analysis for certain communities, data, or epistemologies.

6. Conclusions

This study characterized As and Pb contamination of RHR collected by CS in EJ communities around AZ. While the rainwater was deemed safe for garden irrigation, there were exceedances of US federal and/or AZ state drinking, livestock, and body contact (full and partial) standards/recommendations, posing a potential human/ecological pollution exposure. Concentrations of As and Pb in most communities were greater than AZ background concentrations and significantly related to seasonality and industrial activity. In general, contamination was not related to rainwater harvesting infrastructure. This indicates that governments and corporations are main drivers for environmental contamination, not individuals. Systemic change and cultural paradigm shifts should be implemented to manage contamination and prevent future socioecological harm.

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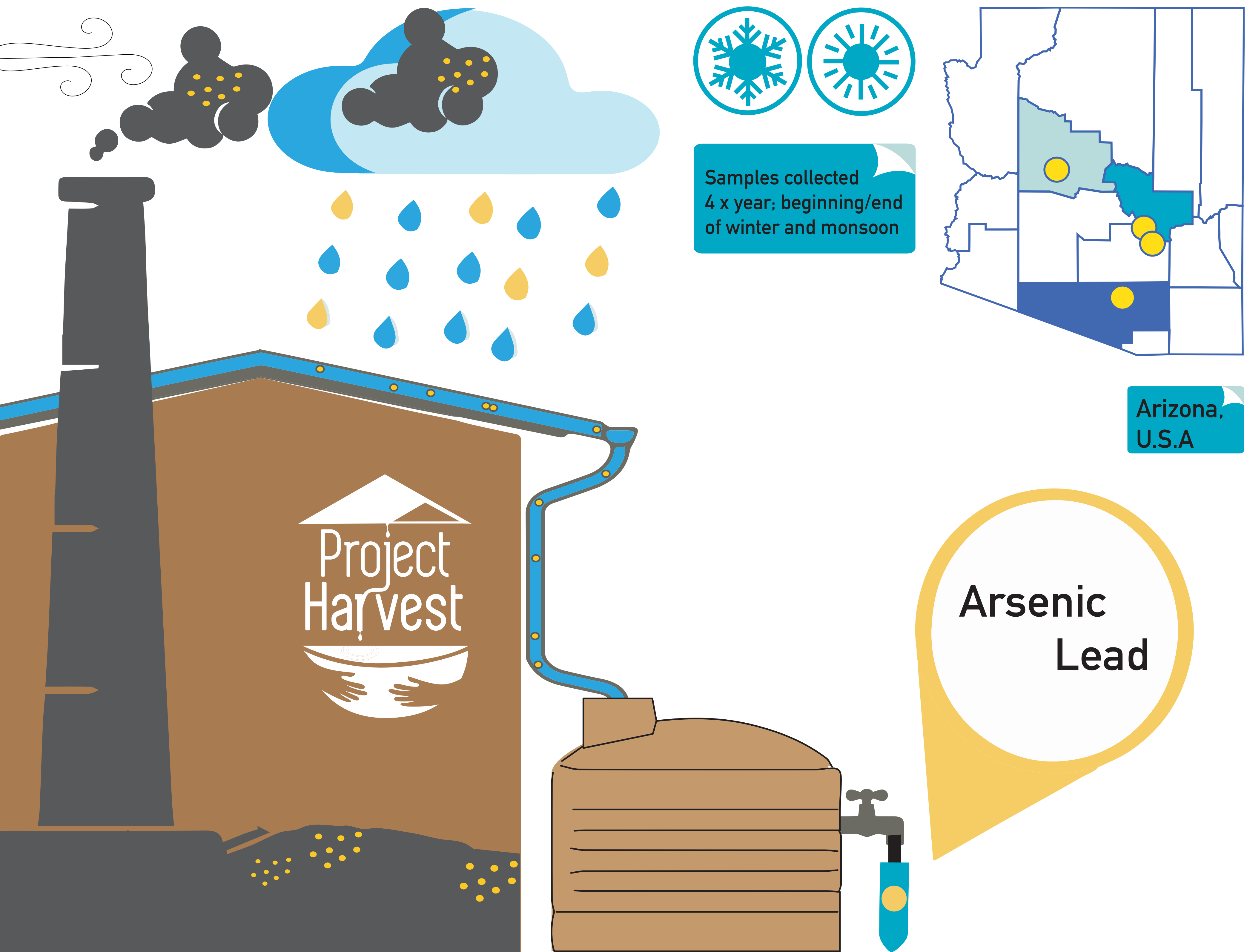
Samples collected
4 x year: beginning/end
of winter and monsoon
Arizona,
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Project
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