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Assessing Unconventional Oil and Gas Exposure in the Appalachian Basin: Comparison of Exposure Surrogates and Residential Drinking Water Measurements

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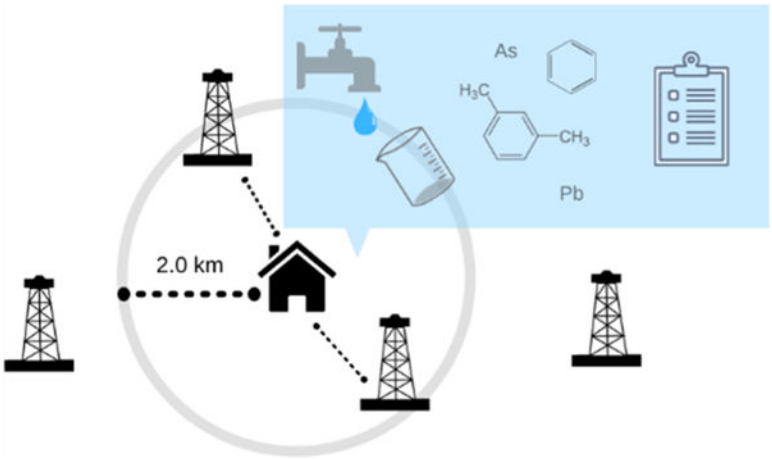
Abstract

Health studies report associations between metrics of residential proximity to unconventional oil and gas (UOG) development and adverse health endpoints. We investigated whether exposure through household groundwater is captured by existing metrics and a newly developed

metric incorporating groundwater flow paths. We compared metrics with detection frequencies/concentrations of 64 organic and inorganic UOG-related chemicals/groups in residential groundwater from 255 homes (Pennsylvania $n = 94$ and Ohio $n = 161$). Twenty-seven chemicals were detected in ≥20% of water samples at concentrations generally below U.S. Environmental Protection Agency standards. In Pennsylvania, two organic chemicals/groups had reduced odds of detection with increasing distance to the nearest well: 1,2-dichloroethene and benzene (Odds Ratio [OR]: 0.46, 95% confidence interval [CI]: 0.23–0.93) and *m*- and *p*-xylene (OR: 0.28, 95% CI: 0.10–0.80); results were consistent across metrics. In Ohio, the odds of detecting toluene increased with increasing distance to the nearest well (OR: 1.48, 95% CI: 1.12–1.95), also consistent across metrics. Correlations between inorganic chemicals and metrics were limited (all $|\rho| \leq 0.28$). Limited associations between metrics and chemicals may indicate that UOG-related water contamination occurs rarely/episodically, more complex metrics may be needed to capture drinking water exposure, and/or spatial metrics in health studies may better reflect exposure to other stressors.

Graphical Abstract



Keywords

unconventional oil and gas development; spatial surrogates; spatial metrics; drinking water; exposure assessment; fracking

INTRODUCTION

Hydrocarbon production from unconventional oil and gas (UOG) resources using horizontal drilling and high-volume hydraulic fracturing has prompted research on the potential for human health impacts.[1] Numerous epidemiologic studies have observed associations between exposure to UOG activity (or combined UOG and conventional development) and adverse health endpoints, including birth outcomes, respiratory symptoms, and cancer.[2–5] Many of these studies used rigorous, high-quality designs and methods, yielding high confidence in the evidence for certain health outcomes (e.g., adverse birth outcomes).[2,6] However, these studies also used spatial surrogates (models that predict exposure potential

based upon proximity to a contaminant source) to estimate exposure to UOG activity rather than environmental or biological measurements. Spatial metrics are useful in health studies because they enable exposure estimation across a large population, can be applied to etiologically relevant time windows occurring in the past, and serve as an aggregate measure when the specific etiologic agent is not known or exposure to multiple hazards is possible. An important knowledge gap is the lack of understanding of which exposures are being captured by these metrics.[7] Examining which specific etiologic agents or stressors are being captured by these metrics in smaller exposure studies can illuminate mechanisms underlying observed epidemiologic associations, inform mitigation strategies, or guide monitoring efforts.

Exposure to contaminated drinking water is one exposure pathway of public health relevance.[8,9] UOG development is a complex, multiphase process with potential for chemical releases to water at several points. Hydraulic fracturing involves pressurized injections of millions of liters of water, chemicals, and proppant into horizontal wells to break and hold open the low-permeability rock, allowing natural gas to flow up the well for capture at the surface.[10] This process generates 1.7–14 million liters of wastewater over the first 5–10 years of production, varying by region and producing formation.[11,12] Hundreds of chemicals have been reportedly used in injection water or detected in wastewater, including metals, volatile organic compounds (VOCs), and polycyclic aromatic hydrocarbons.[13–15] Many of these chemicals are associated with adverse health outcomes in epidemiologic studies, such as reproductive and developmental toxicity and cancer.[16,17] Water contamination has been suggested to occur through improper management or structural failure of wastewater injection wells[18–20] or through surface spills and releases of fracturing fluids or wastewaters that percolate into groundwater.[18,21–26] PA alone experienced 1300 reported spills related to UOG from 2005 to 2014,[27,28] and the PA Department of Environmental Protection (PADEP) received 4099 oil- and gas-related water supply complaints from 2004 to 2016, leading to 215 confirmed instances of UOG-related impairments (“positive determinations”).[29] While specific localized water impairments have been documented, many studies have found no or limited evidence of regional impacts to water quality.[30–33] Furthermore, the human exposure potential from water-related pathways is not well-understood.

While both surface water and groundwater are vulnerable to contamination from UOG operations,[30–33] we focus on groundwater in this study. Approximately 50% of wells hydraulically fractured in Ohio (OH) and PA are located within 2 km of a domestic groundwater well,[34] and approximately 50% of the residents in our predominantly rural study counties rely on groundwater from domestic wells.[35,36] Domestic wells are not subject to federal regulations and monitoring[37] and may be vulnerable to contamination due to their potential shallow depths and lack of continuous casing.[38] Numerous groundwater monitoring studies have been conducted in the Appalachian Basin, primarily with the goal of evaluating UOG impacts to groundwater rather than conducting human exposure assessment.[15,39–42] Several studies applied metrics related to topography, geology, and/or distance to UOG wells to evaluate potential impacts to potable groundwater supplies. However, few have used the density metrics common in human health studies. In addition, while many studies have evaluated methane, noble gases, and anions/cations, comparably

fewer have focused on UOG-related constituents hazardous to human health (e.g., benzene and phthalates).[15,43] A previous pilot study conducted by members of our research team in OH in 2016 reported associations between detections of several health-relevant organic chemicals in drinking water samples and proximity metrics in 66 homes.[44] The current study builds upon this pilot by quantifying 64 organic and inorganic chemicals in 255 homes served by groundwater in PA and OH. Study homes lie over both the Marcellus and Utica shale plays, allowing for comparisons between areas with distinct differences in UOG activity and geology. The objective of this study was to assess the relationship between five spatial surrogates of UOG exposure, including one novel groundwater-specific metric and groundwater detections and concentrations of chemicals that have reportedly been used or produced by UOG. Given the widespread concern over water contamination and the findings from the epidemiologic literature, understanding how and if current exposure surrogates are capturing this potential exposure can inform both interpretation of health studies and more effective policies to protect public health in the face of uncertainty.

METHODS

Study Setting and Population.

This analysis is part of the Yale WATer and Energy Resources (WATER) Study, which focuses on groundwater quality issues in actively drilled areas of the Appalachian Basin (Figure 1). The PA field study was conducted primarily in Bradford County (n = 1529 active UOG wells present in the county between July and September 2018).[45] The county also has the highest number of “water supply determinations” (confirmed instances of drinking water impacts by oil and gas activity), with 63 reported by the PADEP between 2001 and 2020.[46] For OH, we sampled primarily from Belmont and Monroe Counties (n = 598 and 431 active wells per county from May to August 2019, respectively). A few participants living in neighboring counties were included if their zip code intersected with the primary county (PA: n =5 in Tioga County and OH: n = 6 in Noble or Guernsey County).

To reach a broad range of people, multiple methods were used to recruit participants, including informational postcards, flyers posted at local businesses, social media, and newspaper advertisements. Prospective participants who responded to our recruitment methods were screened for eligibility via phone by study staff, and if eligible, scheduled for a home visit (see the Supporting Information, Study Zip Code Selection and Participant Recruitment Criteria). Study eligibility consisted of being an adult household decision-maker (≥21 years of age), English-speaking, and living in our selected counties in a home served by a private groundwater well or spring. The study protocol was approved by the Institutional Review Board of Yale University (HIC #2000021809) and reviewed and approved by the US Environmental Protection Agency (HSR-001162). All participants provided informed consent prior to data collection activities.

Home Visit Overview.

Home visits included administration of informed consent, water sampling, measurement of geocoordinates, and an interview. We completed 94 home visits in PA between July and September 2018 and 161 in OH between May and August 2019. We collected latitude

and longitude at the front door of each home to accurately measure home location (Garmin eTrex 10).[47] Trained interviewers administered a structured questionnaire with three sections: home characteristics, water source characteristics [e.g., treatment system(s) and well depth] and use patterns, and demographic characteristics (e.g., race/ethnicity and household income). Participants were mailed a report comparing their drinking water measurements to health- and aesthetic-based drinking water standards as available and provided state-specific information on water quality and testing.

Water Sample Collection and Analysis.

Water samples were collected upstream of any home water treatment/filtration devices to represent the maximum potential exposure and obtain the best representation of groundwater. Prior to sampling, the well or spring was purged until temperature, pH, conductivity, and dissolved oxygen were stable.[48] Daily field blanks were collected using purified Milli-Q water (18 MOhm, UV-treated for total organic carbon reduction) to identify any contamination introduced by collectors, supplies, or sample transportation. Water samples were analyzed for a broad range of organic and inorganic chemicals reportedly used or produced by UOG activity with the evidence of human health effects or issues related to color, taste, or odor, hereafter referred to as “target chemicals” (Table S1).[16,49] Although linked to UOG processes, some chemicals have natural or other anthropogenic sources. Our VOC analysis followed U.S. Environmental Protection Agency (USEPA) Method 624, with minor modifications previously described by Getzinger et al.[50] Chemicals with peaks that were not able to be differentiated were reported together (e.g., benzene and 1,2-dichloroethene). Samples for inorganic elements were collected following the U.S. Geological Survey groundwater sampling protocol.[51] Major cations and dissolved iron were quantified by inductively coupled atomic plasma emission spectrometry. Major anions and remaining trace elements were quantified using ion chromatography and inductively coupled plasma mass spectrometry, respectively. For details, see the Supporting Information (Water Sampling and Analytical Methods).

UOG Data Sources.

We assembled a database of UOG wells for PA, OH, and West Virginia (WV) that were spud prior to sample collection (May 1, 2018 for PA and May 1, 2019 for OH and WV wells). Locations and activity characteristics of UOG wells were obtained from PADEP’s Office of Oil and Gas Management (2000–2020), OH Department of Natural Resources’ Risk-Based Data Management System (1924–2019), and WV Department of Environmental Protection’s Offce of Oil and Gas (1985–2019). WV wells were included in the OH proximity metrics when the buffer area around OH homes extended across state boundaries. Data were quality-checked and cleaned to remove duplicates, resolve missing data, fix structural errors, and harmonize variables over multiple years and across states.

Calculation of Spatial Metrics.

Using UOG well location data from our database and geocoordinates collected at participant homes, we constructed four previously applied metrics and one newly applied metric capturing proximity and/or density of UOG wells in relation to participant residences.

These included

i. number of UOG wells within a buffer zone,

ii. distance to the nearest UOG well,

iii. inverse distance weighted (IDW) well count [represented by $\sum_{i=1}^{n} \frac{1}{d_i}$ for all UOG wells within a buffer zone (n), where d_i = distance between the ith UOG well and a residence],

iv. inverse distance-squared weighted (ID^2W) well count (represented by $\sum_{i=1}^{n} \frac{1}{d_i^2}$ for all UOG wells within a buffer zone), and

v. a topographically driven groundwater flow-based inverse distance metric ID_{ups} ($\frac{1}{u}$ where u = distance to the nearest upgradient UOG well), determined using the D-infinity flow direction algorithm,[52] first applied within the UOG context in Soriano Jr. et al.[53] This metric is based on the widely accepted conceptual model that groundwater flow in regions of hill-and-valley topography occurs in the downhill direction, parallel to the topographic gradient.[54]

Distance to the nearest UOG well, IDW, and ID^2W metrics was constructed using the Euclidean distance between each participant's home and surrounding eligible UOG wells within 2, 5, and 10 km (ArcGIS 10.8.1). We selected these buffer sizes based on the hydrogeologic literature, which generally supports transport distances of 2 km or less[55–57] and the epidemiologic literature, which incorporates buffer distances up to 10 km.[2] For ID_{ups}, which was specific to the groundwater exposure pathway, we explored buffer sizes of 0.2, 0.3, 0.5, 1, and 2 km.[53,58] Additionally, ID_{ups} buffers are applied around delineated flow paths from UOG well locations and not around participant homes. For all metrics except distance to the nearest UOG well, having no wells within the relevant buffer zone yields a value of zero.

Statistical Analysis.

We summarized the characteristics of the population (e.g., race/ethnicity and gender) and sampled residences (e.g., drinking water well depth) and compared the distribution of these factors between states. We calculated detection frequencies (see the Supporting Information, Water Sampling and Analytical Methods) and summary statistics for chemical concentrations in water samples and compared them to standards from the USEPA and the World Health Organization (WHO). Spatial metrics were evaluated using three approaches: comparing exposure assignments produced by each metric with the other metrics, comparing metrics with chemical detections in drinking water, and comparing metrics with chemical concentrations in drinking water. We assessed the relationships among the metrics using Spearman correlations, calculated separately for each state. We used logistic regression to evaluate the odds of detecting target chemicals (detected in at least 20% of samples) in drinking water in relation to the metrics. We assessed correlations between metrics and concentrations of chemicals in drinking water using scatterplots and Spearman correlations. Finally, we used linear regression to identify associations between the concentrations

(continuous, μg/L) of target chemicals in drinking water and metrics. Correlation and linear regression analyses of chemical concentrations were restricted to chemicals with a detection frequency of at least 50% and/or a twofold difference between the 75th and 25th percentiles to ensure sufficient variability in the data. Measurements below the limit of detection (LOD) were substituted with LOD/$\sqrt{2}$ (see the Supporting Information, Water Sampling and Analytical Methods). Separate models were run for each chemical and metric. For our regression analyses, we considered variables from the home interview as potential covariates. We used metrics with 0.5 (for ID_{ups} only), 1, and 2 km buffer sizes, informed by and to facilitate comparisons with the hydrogeological and epidemiologic literature studies.[55,58] Metrics were used continuously (distance to the nearest UOG well, per km), categorically and high/low (above and below the median level of exposure; ID_{ups}, IDW, and ID^2W), and as discrete data (sum of wells within a buffer).

RESULTS

Study Population and Water Source Characteristics.

The characteristics of our study populations in both PA and OH (Table 1) were similar to the 2019 U.S. Census characteristics for their respective counties with respect to age, race/ethnicity, and income; educational attainment was slightly higher among study participants.[59] The majority of participants had a private well (79%) with few being served by a spring (10%) or both a well and spring (11%) (Table 1). Drinking water wells tended to be shallower in OH compared to those in PA (χ^2 p-value <0.001). Private wells and springs were the primary drinking water source for most households (77%), while 22% relied on bottled water and 1% on another source. A slightly higher percentage of PA versus OH participants reported their water ever having an unnatural color (35 vs 25%) or taste (19 vs 15%) (χ^2 p-values = 0.080 and 0.039, respectively). In PA, the mean distance to the nearest well was 1.15 km (range = 0.15–4.03 km), and on average, homes had 7.80 UOG wells within 2 km and 52.38 within 5 km. In OH, study homes were on average 2.03 km from a UOG well (range = 0.30–7.31 km), with an average of 4.72 UOG wells within 2 km and 31.94 within 5 km.

Distributions of Chemicals in Residential Drinking Water.

Of the 47 organic compounds measured, 10 were detected in at least 20% of PA homes (Table 2) with the following most commonly detected: bromochloromethane (detection frequency = 97%), chloroform (76%), 1,2-dichloroethene and benzene (75%), and trichloroethene (75%). In OH, six organic chemicals were detected in at least 20% of homes (Table 2), where the most frequently detected organics were bromomethane (67%) and vinyl chloride (57%). While some organics (e.g., toluene) were detected more frequently in PA versus OH, the detected concentrations were generally several orders of magnitude below maximum contaminant limits (MCLs) or guidance values (GVs), were often clustered near the LODs and LOQs, and exhibited limited variability with interquartile ranges (IQRs) spanning less than 1 order of magnitude. No organic chemicals exceeded health-based MCLs in either state.

All 17 inorganic chemicals measured were detected in at least 20% of PA homes (Table 2). The most frequently detected inorganic chemicals were barium, chloride, lithium, strontium, sulfate, sodium, and potassium (all 100% of homes), calcium and magnesium (99%), lead (96%), manganese (91%), uranium (85%), and arsenic (81%). In OH, all inorganic chemicals except for arsenic, lead, and uranium were detected in at least 20% of homes (Table 2). Arsenic and lead were much less commonly detected in OH (8 and 12%, respectively) than those in PA (81 and 96%, respectively). Although rare, MCL exceedances were observed in some PA or OH study homes for arsenic, lead, barium, fluoride, and nitrate.

Correlations among Spatial Surrogates.

Patterns in Spearman correlation coefficients among metrics were similar for PA and OH (Figure 2). For both states, IDW and ID^2W metrics were strongly correlated with all buffer sizes ($\rho \geq 0.70$). Distance to the nearest UOG well was inversely correlated with IDW metrics (range $\rho = -0.28$ to -0.75 in PA and -0.62 to -0.71 in OH) and ID^2W metrics ($\rho = -0.66$ to -0.86 in PA and -0.48 to -0.55 in OH) at all buffer sizes. Note that distance to the nearest UOG well is the only metric for which a lower value indicates higher UOG exposure potential. In PA, the strength of the correlations between distance to the nearest well and IDW and ID^2W metrics decreased as buffer size increased. This attenuation was not observed for OH, where UOG wells were farther away from homes on average (mean = 2.03 km in OH vs 1.15 in PA). The novel ID_{ups} metric was most strongly correlated with distance to the nearest UOG well ($\rho = -0.43$ to -0.67) in PA and most strongly correlated with the ID^2W metrics in OH ($\rho = 0.69$–0.72). The UOG well counts in buffer sizes were most strongly correlated with IDW metrics of the same buffer size ($\rho = 0.91$–0.95 in PA and 0.82–0.91 in OH).

The top section shows Spearman correlation coefficients among OH metrics. The bottom section shows Spearman correlation coefficients among PA metrics.

Associations between Organic Chemicals and Spatial Surrogates.

For organic chemicals, regression analyses were limited to modeling detection frequencies due to relatively low concentrations and limited variability within the data. In logistic regression models for PA, the odds of detecting four organics (1,2-dichloroethene and benzene, bromomethane, toluene, and *m*- and *p*-xylene) were positively associated with higher UOG exposure potential based on at least one metric (Table 3). The odds of detecting 1,2-dichloroethene and benzene combined were associated with four of six metrics. The odds of detecting 1,2-dichloroethene and benzene were 0.46 times lower for each additional km between a UOG well and a home (95% confidence interval [CI]: 0.23–0.93), 2.59 times higher in the high exposure group (exposure level above the median) for ID_{ups} and IDW 2 km (95% CI: 1.01–6.67 for both metrics), and 3.29 times higher in the high exposure group for ID^2W 2 km (95% CI: 1.25–8.66). The odds of detecting bromomethane were elevated in the high exposure group for IDups 1 km (odds ratio [OR]: 2.55, 95% CI: 1.06−6.13); associations with other metrics were inconsistent in direction (ID_{ups} 0.5 km, IDW and ID^2W2 km) and statistical significance (distance to the nearest, ID_{ups} 1 and 2 km). The odds of detection of toluene decreased with increasing distance between

UOG well and home (OR: 0.52, 95% CI: 0.27–1.03) and increased in the high exposure group for one metric, ID_{ups} 1 km (OR: 2.67, 95% CI: 1.07–6.45). The odds of detecting toluene were elevated for three other metrics (ID_{ups}, IDW, and ID^2W 2 km). The odds of detection for *m*- and *p*-xylene decreased with increasing distance to the nearest UOG well (OR: 0.28, 95% CI: 0.10–0.80) and were consistently elevated across inverse distance metrics. Spearman correlation coefficients between concentrations of organics with at least 50% detection frequency ranged from −0.02 to−0.27 in PA and were consistent with regression results (see Figure S1), particularly for 1,2-dichloroethene and benzene, toluene, and bromochloromethane.

In contrast to results from PA, in OH, the odds of detection of toluene were higher in homes with lower UOG exposure potential based on four of six metrics (Table 3). The odds of toluene detection were 1.54 times higher for each increasing kilometer of distance between a home and the nearestUOG well (OR: 1.54, 95% CI: 1.17–2.03). Results were consistent in direction across ID_{ups}, IDW, and ID^2W metrics up to the 2 km buffer size. No other organic chemicals were associated with any metric. The concentrations of the two chemicals meeting our criteria for the correlation analysis (bromomethane and vinyl chloride) were not correlated with distance to the nearest UOG well (Figure S1).

Associations between Inorganic Chemicals and Spatial Surrogates.

In PA and OH, most inorganic species were not correlated or weakly correlated with metrics (ρ range: ±0.00–0.28). Concentrations of some inorganics were correlated with increasing UOG proximity and density (e.g., chloride, bromide, and fluoride), while others were inversely correlated with UOG exposure (e.g., calcium, iron, sulfate, magnesium, and manganese) (see Supporting Information, Table 1). Bivariable linear regression results were consistent with the correlation analysis.

DISCUSSION

In this multistate exposure study in Appalachian Basin counties with a high UOG activity, we provide data on detection frequencies and concentrations of 64 organic and inorganic chemicals and compared them with spatial surrogates of UOG exposure commonly used in health studies. We also applied a new spatial surrogate specifically designed to capture exposure via the groundwater pathway.[52,53] Organic chemicals were not commonly detected and were present at concentrations below health-based standards in both states. Nonetheless, some chemicals had greater odds of detection with increasing UOG exposure proxies. Inorganic chemicals, many of which have geologic and anthropogenic sources, were commonly detected at concentrations below MCLs, with a few exceedances observed. The concentrations of inorganic species were generally unrelated to UOG exposure surrogates or were higher in residences with lower exposure potential.

The low detection frequencies and concentrations of organic chemicals are consistent with other groundwater monitoring studies in the area in that, to our knowledge, no study to date has found widespread contamination attributable to UOG activity.[15,39,43,44,60] The results of this study are concordant with the previous pilot study in the Appalachian Basin in regard to low levels of organic contaminants overall.[44] Another study of Northeastern PA

reported benzene and other VOCs in only 10% of samples,[15] while we detected benzene combined with 1,2-dichloroethene more frequently (75% of homes) at comparably low concentrations. A study of 11,156 predrill PA groundwater measurements reported higher benzene concentrations than our study (range: 0.25–7.88 μg/L), although it was detected less frequently (12%).[43] Similarly, a previous OH study detected benzene in 7% of samples (range: 0.3–1.2 μg/L).[44] While we detected benzene and 1,2-dichloroethene in 24% of OH homes, the concentrations were similar (range: <0.09−2.73 μg/L). Our higher detection frequencies may be explained by differences in sample collection, method sensitivity, and reporting of some chemicals as groups when chromatographic peaks were unresolvable. Ultimately, the relatively low concentrations and variability limited the types of statistical analyses we could conduct. Despite low concentrations and detection frequencies, detections of 1,2-dichloroethene and benzene, bromomethane, toluene, and *m*- and *p*-xylene were associated with spatial metrics. Most indicated increased odds of detection with increasing UOG exposure, and associations were consistent across all metrics except for ID_{ups} 0.5 km. Exposure to benzene is of particular public health concern due to its leukemogenic properties.[61,62] In addition to being detected in other groundwater studies, elevated benzene exposures have been observed in communities proximate to oil and gas activities based on air[63,64] and biological samples.[65]

Detection frequencies and concentrations of inorganic chemicals were consistent with existing monitoring data from the area.[35,66] While concentrations of inorganic species in this study were generally below MCLs, exceedances were observed for arsenic (a known carcinogen), barium (a cardiovascular toxicant), nitrate (a probable carcinogen), and lead (a neurotoxicant).[67–69] Lead was detected frequently in PA (96% of samples), and while lead has a health-based MCL, no concentration of lead is thought to be safe.[68] Some inorganic species varied substantially in detection frequency and concentration by state (e.g., arsenic), which may reflect differences in the mineralogic composition of drinking water aquifers and their overlying sediments, as well as differences in groundwater flow patterns and associated differences in water–rock interaction times.[70–72] Our results indicate generally either inverse or no associations between concentrations of inorganic chemicals and UOG exposure metrics, even those accounting for hydrogeological pathways.

The low chemical concentrations and limited associations with spatial metrics could be due to several factors. UOG-related contamination events could occur with low frequency or be transient.[24,73] Thus, individual groundwater samples collected at one point in time may be unlikely to capture a contamination event. In addition to these temporal constraints, physically based transport modeling work conducted under this project demonstrated that the longitudinal extent of the probabilistic capture zones (the area from which a drinking water well likely draws its groundwater) around the PA homes was generally smaller than distances to the nearest UOG wells (<1.6 km with a median of 0.86 km).[55,74] Therefore, the appropriate buffer sizes when considering water quality impacts may be considerably smaller than those traditionally applied in health studies, which are designed to capture a range of possible environmental stressors. Additionally, the multiple natural, industrial, and other sources make it difficult to link the occurrence of particular inorganic chemicals to UOG activity.[75,76] Studies focused on source apportionment use other methods, such

as ratios of inorganic ions (e.g., bromide/chloride), as potential indicators of UOG-related contamination.[77,78]

The limited associations between spatial metrics and chemical concentrations could indicate that to optimize metrics for the water pathway, more complex models incorporating groundwater flow, water source vulnerability, contamination release information, and contaminant transport are needed. Additionally, these results may reflect the need for measurements at multiple times. For example, episodic exposures could occur during phases of hydraulic fracturing or high production. Studies coordinating the timing of sampling events with production stages are rare.[41] They are made difficult by the lack of contemporaneous, publicly available information on UOG activities at specific well pads and complicated further by the temporal lag between chemical release and appearance at receptor locations. Because domestic groundwater is not routinely monitored, there are limited existing data with which to examine issues of temporality.

Although more specific hydrogeological models may be better suited to predicting UOG-related groundwater vulnerability, regulatory protections such as setback distances are based on simple spatial relationships, such as distance from home or school to the UOG site. Therefore, using and understanding these simpler metrics provide value to researchers, government organizations, and community stakeholders. The mandatory setback distance between a UOG well and homes or private drinking water wells is 152 m (500 ft) in PA and 46 m (150 ft) in OH.[79,80] Updates to these distances are the subject of debate and are being informed in some states by exposure and the epidemiologic literature.[6,81–85]

Our survey data indicated areas for consideration for future exposure or health studies. Only 6% of homes had their private wells tested at least once per year, and 28% had never tested their water. Given the lack of regulatory oversight and monitoring of domestic groundwater wells,[37] testing and reporting drinking water results in this research context had the added benefit of informing residents about the safety of their water.[86] Additionally, 22% of participants reported primarily consuming bottled water instead of their well water, underscoring how individual behavior may influence exposures. This also indicates individual and community concern over water quality, which can contribute to negative mental health outcomes, such as psychosocial stress.[87–92]

The UOG metrics we evaluated have yielded significant associations with adverse health outcomes in epidemiologic studies.[2] While we cannot discount the potential importance of the drinking water pathway for public health, our results suggest that these metrics may be reflecting other stressors or a combination of stressors not measured here (e.g., air pollution and noise).[13,22,50,79–81] Given the complexity of UOG exposure, specific models may be needed to explicate distinct exposure pathways. For example, Casey et al. (2016) introduced an activity-based inverse distance-squared weighted metric where the numerator varies by the UOG phase.[93] Allshouse et al. (2017) introduced an activity-based metric to assess exposure to UOG-related air pollution.[94] Furthermore, recent studies have applied exposure models specific to flaring (the intentional burning of natural gas),[95] earthquakes,[96] and radioactivity.[97] As new models and metrics are developed, researchers can consider

the benefits and trade-offs of pathway-specific versus aggregate models for optimizing their exposure assessment approach.

In our study, spatial surrogates exhibited limited associations with detections and concentrations of target chemicals. This may indicate that water contamination by UOG may occur with low frequency and/or be episodic, creating a temporal misalignment between our measurements and exposures, and/or water contamination may be highly localized, and more complex groundwater flow and contaminant fate models or more specific information on spills, leaks, and violations may be needed to accurately capture drinking water exposure. Given the complexities of water contamination and exposure pathways, spatial metrics in epidemiologic studies may be better representing other environmental UOG stressors. More complex models and groundwater monitoring data could provide insights into the drinking water exposure pathway and would have a high value to ensure the protection of public health.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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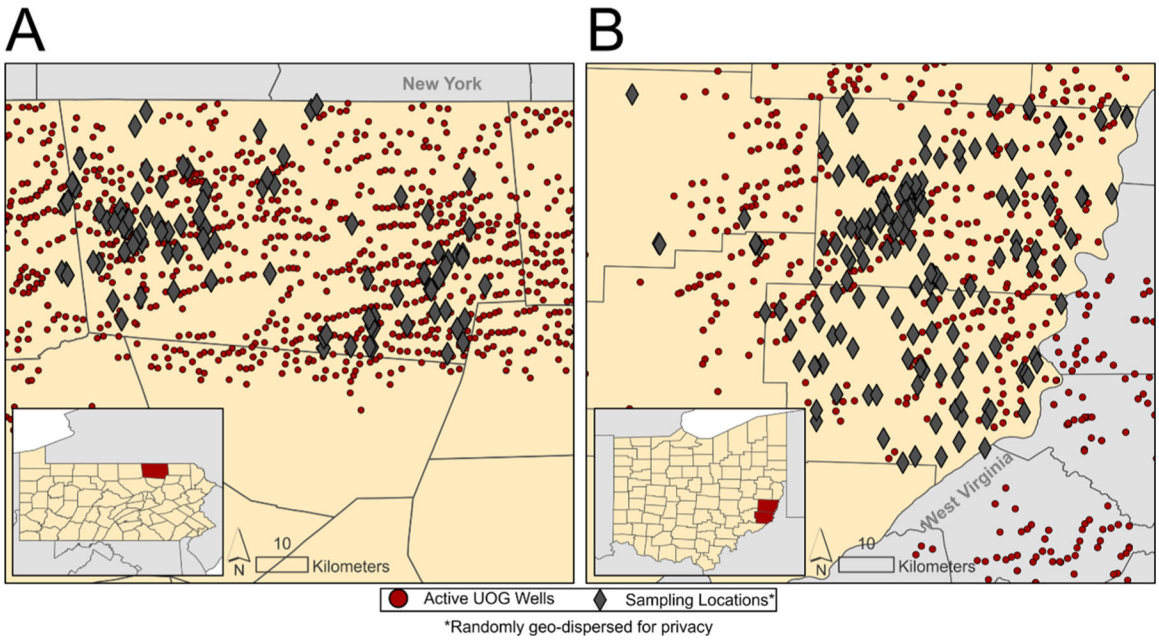

Figure 1.

Sampling locations in Bradford County, PA (A) and Belmont and Monroe Counties, OH (B). Gray diamonds represent sampling locations; red circles represent active UOG wells. Home locations were randomly geodispersed (offset) by 0.1 km for privacy.

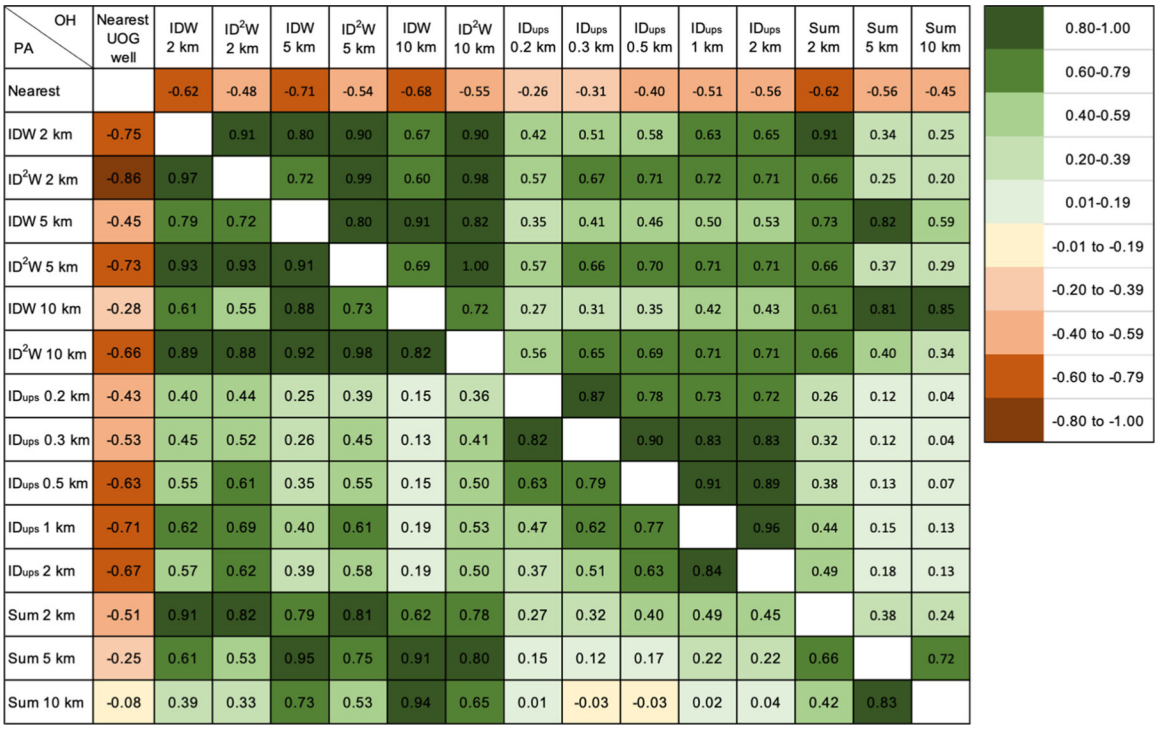

Figure 2.
Spearman correlations between spatial metrics in PA and OH.

Table 1.

Demographic and Residential Characteristics of the 2018 Bradford County, PA Population Homes ($n = 94$) and the 2019 Belmont and Monroe County, OH Participants ($n = 161$)

	PA *N* (%)	OH *N* (%)	Combined ($n = 255$ homes, 250 individuals) *N* (%)	*P*-value[a]
Demographic characteristics				
Age (yrs)				
≤30	1 (1)	4 (3)	5 (2)	
31–50	21 (23)	23 (15)	44 (18)	
51–70	53 (57)	83 (53)	136 (54)	
>70	18 (19)	47 (30)	65 (26)	
Gender				0.096
Male	45 (48)	93 (59)	138 (55)	
Female	48 (52)	64 (41)	112 (45)	
Race				
White	91 (98)	153 (97)	244 (98)	
Other	2 (2)	4 (3)	6 (2)	
Education (yrs)				<0.001
<12	1 (1)	9 (6)	10 (4)	
12–16	47 (51)	114 (73)	161 (64)	
>16	45 (48)	33 (21)	78 (31)	
Refused to answer	0 (0)	1 (1)	1 (0)	
Employment				0.050
Employed	43 (46)	56 (36)	99 (40)	
Retired	40 (42)	89 (57)	129 (52)	
Other	12 (12)	7 (7)	22 (8)	
Household income				0.056
<$49,999	19 (21)	56 (36)	75 (30)	
$50,000–99,999	43 (46)	54 (34)	97 (39)	
$100,000–199,999	21 (23)	25 (16)	46 (18)	
≥$200,000	5 (5)	8 (5)	13 (5)	
Refused to answer	5 (5)	11 (7)	16 (6)	

	PA *N* (%)	OH *N* (%)	Combined (*n* = 255 homes, 250 individuals) *N* (%)	*P*-value[a]
Do not know	0 (0)	3 (2)	3 (1)	
Residential and water characteristics				
Water on property				0.011
Well	82 (87)	119 (74)	201 (79)	
Spring	3 (3)	22 (14)	25 (10)	
Both	9 (10)	20 (13)	29 (11)	
Age of well				0.007
2014 or later	5 (5)	9 (6)	14 (5)	
2000–2013	11 (12)	29 (18)	40 (16)	
1980–1999	30 (31)	32 (20)	62 (24)	
1960–1979	18 (19)	38 (24)	56 (22)	
1940–1959	9 (10)	7 (4)	16 (6)	
1920–1939	2 (2)	3 (2)	5 (2)	
1900–1919	0 (0)	1 (1)	1 (0)	
1899 or earlier	0 (0)	4 (2)	4 (2)	
no private well	3 (3)	25 (16)	28 (11)	
Do not know	16 (17)	13 (8)	29 (11)	
depth of well				<0.001
<50 feet	4 (4)	13 (8)	17 (7)	
50–150 feet	34 (36)	94 (58)	128 (50)	
151–250 feet	23 (24)	7 (4)	30 (12)	
251–500 feet	12 (13)	0 (0)	12 (5)	
>500 feet	3 (3)	0 (0)	3 (1)	
No private well	3 (3)	25 (16)	28 (11)	
Do not know	15 (16)	22 (14)	37 (15)	
Frequency of well-water testing				0.001
Never	18 (19)	53 (33)	71 (28)	
<1/yr	68 (72)	74 (46)	142 (56)	
1/yr	3 (3)	6 (4)	9 (4)	
>1/yr	2 (2)	2 (1)	4 (2)	
No private well	3 (3)	25 (16)	28 (11)	

	PA *N* (%)	OH *N* (%)	Combined (*n* = 255 homes, 250 individuals) *N* (%)	*P*-value[a]
Do not know	0 (0)	1 (1)	1 (0)	
Main drinking water source				0.009
Private well	73 (78)	98 (61)	171 (67)	
Spring	3 (3)	22 (14)	25 (10)	
Municipal (city) water	0 (0)	2 (1)	2 (1)	
Bottled water	18 (19)	38 (24)	56 (22)	
Other	0 (0)	1 (1)	1 (0)	
Main water source for cooking, washing, etc.				
Private well	90 (96)	115 (71)	205 (80)	<0.001
Spring	4 (4)	23 (14)	27 (11)	
Municipal (city) water	0 (0)	9 (6)	9 (4)	
Bottled water	0 (0)	0 (0)	0 (0)	
Rain barrel	0 (0)	3 (2)	3 (1)	
Other	0 (0)	11 (7)	11 (4)	
Water ever had unnatural color	33 (35)	40 (25)	73 (29)	0.080
Water ever had unnatural taste	18 (19)	24 (15)	42 (16)	0.039
	Mean	Median	Range	
UOG activity	PA	OH	PA	OH
Nearest UOG well (km)	1.15	2.03	1.09	1.72
Wells within 2 km	7.80	4.72	6.00	3.00
Wells within 5 km	52.38	31.94	42.00	34.00

[a] χ^2 *p*-value; Fisher's exact test used for cell sizes <5; 2019 census characteristics for study counties: range in percent age 65 and over = 20.1–22.6%; range in percent white = 92.4–96.1%; range in median household income = $45,917–52,358; and range in educational attainment, percent with Bachelor's degree = 14.6–19.1%.[59]

Table 2.

Chemical Distribution at Sampled Homes in Bradford County, PA and Belmont and Monroe Counties, OH

	PA			OH				
Chemical	**LOQ (μg/L)**	**>LOD (%)**	**Median (IQR) (μg/L)**	**LOQ (μg/L)**	**>LOD (%)**	**OH Median (IQR) (μg/L)**	**USEPA MCL[c] (μg/L)**	**WHO GV[c] (μg/L)**
Organic chemicals (PA *n* = 89 homes and OH *n* = 161)								
Bromochloromethane	0.172	97	0.52 (0.42, 0.63)	0.050	46	<LOD (<LOD, 0.08)	NS	NS
Chloroform	0.182	76	0.09 (0.009, 0.19)	0.050	22	<LOD (<LOD, <LOD)	NS	300
1,2-Dichloroethane and benzene[a]	0.059	75	0.02 (<LOD, 0.04)	0.092	24	<LOD (<LOD, <LOD)	5	10
Trichloroethene	0.047	75	0.04 (0.008, 0.06)				5	20
Toluene	0.047	64	0.01 (<LOD, 0.03)	0.046	20	<LOD (<LOD, <LOD)	1000	700
Bromomethane	0.079	58	0.02 (<LOD, 0.06)	0.053	67	0.012 (<LOD, 0.04)	NS	NS
Dibromomethane	0.775	45	<LOD (<LOD, 0.12)				NS	NS
1,1-Dichloroethene and *trans*-1,2-dichloroethene[d]	0.087	42	<LOD (<LOD, 0.02)				100	NS
Vinyl chloride	0.047	26	<LOD (<LOD, 0.0004)	0.046	57	0.003 (<LOD, 0.023)	2	0.3
m-Xylene and *p*-xylene[b]	0.064	24	<LOD (<LOD, <LOD)				10,000	500
Inorganic chemicals (PA *n* = 94 homes and OH *n* = 161)	LOD (μg/L)	>LOD (%)		LOD (μg/L)	>LOD (%)			
Arsenic	0.110	81	0.99 (0.36, 2.44)	0.110	8	<LOD (<LOD, <LOD)	10	10
Barium	0.027	100	166.03 (76.99, 399.46)	0.027	99	88.48 (50.74, 142.80)	2000	1300
Bromide	0.008	34	<LOD (<LOD, 71.29)	0.008	53	27.00 (<LOD, 54.00)	NS	NS
Calcium	0.051	99	34,961 (20,968, 42,863)	0.051	100	72,101.19 (51,144.44, 101,596.12)	NS	NS
Chloride	0.550	100	5831 (3035, 16,128)	0.550	99	6758.00 (3018.00, 19,785.00)	250,000[c]	250,000[c]
Fluoride	0.040	80	82.37 (44.42, 114.2)	0.040	100	110.00 (82.00, 156.00)	4000	1500
Iron	0.017	70	60.37 (<LOD, 139.02)	0.017	51	10.74 (<LOD, 32.70)	300[c]	300[c]
Lead	0.050	96	1.27 (0.72, 2.05)	0.050	12	<LOD (<LOD, <LOD)	15	10
Lithium	0.002	100	23.33 (8.27, 51.95)	0.002	99	10.24 (6.79, 15.22)	NS	NS
Magnesium	0.034	99	6767 (3526, 9845)	0.034	100	16,116.57 (8870.02, 27,149.45)	NS	NS
Manganese	0.004	91	17.1 (0.94, 127.51)	0.004	58	1.84 (<LOD, 19.25)	50[c]	100[c]

	PA			OH				
Chemical	**LOQ (μg/L)**	**>LOD (%)**	**Median (IQR) (μg/L)**	**LOQ (μg/L)**	**>LOD (%)**	**OH Median (IQR) (μg/L)**	**USEPA MCL[c] (μg/L)**	**WHO GV[c] (μg/L)**
Nitrate	0.039	67	334.35 (<LOD, 1009.63)	0.039	99	560.00 (100.00, 1754.00)	10,000	50,000
Potassium	0.012	100	1467.52 (1050.14, 1830.6)	0.012	100	1489.75 (1148.41, 2038.67)	NS	NS
Sodium	0.420	100	16,130 (7282, 46,386)	0.420	100	23,819.14 (16,740.71, 52,714.46)	NS	200,000[c]
Strontium	0.004	100	472.04 (179.83, 1037.06)	0.004	100	526.48 (288.57, 967.63)	NS	250,000[c]
Sulfate	0.085	100	10,063 (6847, 15,648)	0.085	96	30,813.00 (20,117.00, 50,587.00)	250,000[c]	250,000[c]
Uranium	0.001	85	0.87 (0.24, 2.56)	0.001	16	<LOD (<LOD, <LOD)	30	30

[a] Standard listed is for the chemical benzene only.

[b] Standard listed is the sum of standards for total xylenes.

[c] A secondary MCL (related to taste, odor, or other aesthetic qualities) is reported for chemicals with no health-based MCL.

[d] Out of 64 total samples for PA. Samples from 5 (5%) of PA homes were not reported due to the evidence of contamination or other factors, such as leaks or breakage. Twenty-five additional PA samples lack measurements of 1,1-dichloroethene and *trans*-1,2-dichloroethene only.

IQR: interquartile range; LOQ: limit of quantification; LOD: limit of detection; MCL: maximum contaminant level; WHO: World Health Organization; GV: guidance value; and NS: no standard.

Table 3.

Associations between Detection Frequencies of Organic Compounds and Spatial Metrics

Chemical	Nearest (km) OR (95% CI)	ID_{ups} 0.5 km OR (95% CI)	ID_{ups} 1 km[a] OR (95% CI)	ID_{ups} 2 km[a] OR (95% CI)	IDW 2 km[a] OR (95% CI)	ID2W 2 km[a] OR (95% CI)
			PA			
Vinyl chloride	0.71 (0.33, 1.53)	0.92 (0.33, 2.60)	1.87 (0.71, 4.91)	1.87 (0.71, 4.91)	1.87 (0.71, 4.91)	1.47 (0.56, 3.82)
Bromomethane	0.70 (0.37, 1.32)	0.68 (0.26, 1.78)	2.55 (1.06, 6.13)	1.72 (0.73, 4.07)	0.97 (0.42, 2.28)	0.81 (0.34, 1.89)
1,2-Dichloroethene and benzene	0.46 (0.23, 0.93)	0.60 (0.21, 1.72)	1.66 (0.66, 4.14)	2.59 (1.01, 6.67)	2.59 (1.01, 6.67)	3.29 (1.25, 8.66)
Toluene	0.52 (0.27, 1.03)	0.72 (0.27, 1.92)	2.63 (1.07, 6.45)	1.74 (0.73, 4.19)	2.13 (0.88, 5.18)	2.13 (0.88, 5.18)
Chloroform	1.41 (0.63, 3.13)	0.96 (0.33, 2.83)	2.63 (0.32, 2.28)	0.67 (0.25, 1.79)	0.67 (0.25, 1.79)	0.86 (0.32, 2.28)
m-Xylene and *p*-xylene	0.28 (0.10, 0.80)	1.04 (0.35, 3.07)	3.36 (1.16, 9.72)	1.50 (0.56, 4.02)	3.36 (1.16, 9.72)	2.53 (0.91, 7.07)
1,1-Dichloroethene and *trans*-1,2-dichloroethene	0.76 (0.37, 1.57)	0.63 (0.22, 1.83)	2.05 (0.75, 5.63)	2.05 (0.75, 5.63)	1.09 (0.40, 2.96)	1.58 (0.58, 4.30)
Bromochloromethane[b]	0.36 (0.11, 1.19)	0.42 (0.17, 1.06)	1.09 (0.49, 2.45)	1.09 (0.49, 2.45)	0.92 (0.41, 2.06)	1.29 (0.57, 2.91)
Trichloroethene	0.87 (0.44, 1.74)	1.18 (0.42, 3.34)	0.76 (0.29, 2.00)	0.60 (0.23, 1.58)	0.60 (0.23, 1.58)	0.60 (0.23, 1.58)
Dibromomethane	0.91 (0.49, 1.69)	0.75 (0.30, 1.88)	1.80 (0.78, 4.20)	1.25 (0.54, 2.88)	1.04 (0.45, 2.40)	1.25 (0.54, 2.88)
			OH			
Vinyl chloride	1.08 (0.85, 1.37)	0.71 (0.36, 1.39)	0.88 (0.44, 1.77)	0.67 (0.34, 1.33)	0.53 (0.27, 1.05)	0.66 (0.34, 1.28)
Bromomethane	0.91 (0.72, 1.17)	0.89 (0.44, 1.82)	1.99 (0.89, 4.41)	1.48 (0.70, 3.11)	1.12 (0.54, 2.31)	1.16 (0.57, 2.35)
1,2-Dichloroethene and benzene	1.18 (0.91, 1.53)	0.62 (0.27, 1.43)	0.90 (0.40, 2.04)	0.91 (0.41, 2.03)	0.77 (0.34, 1.74)	0.67 (0.30, 1.50)
Toluene	1.54 (1.17, 2.03)	0.33 (0.12, 0.91)	0.64 (0.26, 1.60)	0.44 (0.17, 1.15)	0.25 (0.08, 0.77)	0.30 (0.11, 0.82)
Chloroform	1.05 (0.80, 1.39)	1.95 (0.90, 4.23)	0.71 (0.30, 1.71)	0.61 (0.26, 1.47)	1.06 (0.47, 2.38)	0.92 (0.41, 2.05)
Bromochloromethane[b]	0.99 (0.79, 1.26)	0.97 (0.50, 1.89)	0.89 (0.44, 1.78)	1.02 (0.52, 2.00)	1.02 (0.52, 2.00)	1.45 (0.75, 2.81)

[a] Exposure is defined as a value above the median.

[b] Detection is defined as a value above the median concentration for PA homes only. Compounds marked NA were not detected at a sufficient frequency for analysis.