Household and Community Systems for Groundwater Remediation in Bihar, India: Arsenic and Inorganic Contaminant Removal, Controls and Implications for Remediation Selection

Laura A. Richards[1*], Neha Parashar[2,3], Rupa Kumari[2], Arun Kumar[2], Debapriya Mondal[4], Ashok Ghosh[2], David A. Polya[1]

[1]Department of Earth and Environmental Sciences and Williamson Research Centre for Molecular Environmental Science, The University of Manchester, Williamson Building, Oxford Road, Manchester, M13 9PL, UK

[2]Mahavir Cancer Sansthan and Research Centre, Phulwarisharif, Patna, 801505, Bihar, India

[3]now at Indian Institute of Technology Patna, Patna, 801106, Bihar, India

[4]Institute of Medical and Biomedical Education, St George's University of London, SW17 0RE, UK

*Corresponding author: laura.richards@manchester.ac.uk

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Abstract

The presence of arsenic (As) and other inorganic contaminants in groundwater is a key public health issue in India and many other parts of the world. Whilst a broad range of remediation technologies exist, performance can be highly variable, and appropriate selection and management of remediation approaches remains challenging. Here, we have identified and tested the performance of a range of small-scale remediation technologies (*e.g.* sand filters, multi-stage filtration and reverse osmosis (RO)-based systems; n = 38) which have been implemented in Bihar, India. We have undertaken spot-assessments of system performance under typical operating conditions in household and non-household (*e.g.* community, hospital, hostel/hotel) settings. The removal of As and other inorganic contaminants varied widely (ranging from ~ 0 – 100 %), with some solutes generally more challenging to remove than others. We have evaluated the relative importance of technology type (*e.g.* RO-based versus non-RO systems), implementation setting (*e.g.* household versus non-household) and source water geochemistry (particularly concentrations and ratios of As, Fe, P, Si and Ca), as potential controls on remediation effectiveness. Source water composition, particularly the ratio ([Fe] -1.8[P])/[As], is a statistically significant control on As removal ($p < 0.01$), with higher ratios associated with higher removal, regardless of technology type. This ratio provides a theoretical input which could be used to identify the extent to which natural groundwater composition may be geochemically compatible with higher levels of As removal. In Bihar, we illustrate how this ratio could be used to identify spatial patterns in theoretical geochemical compatibility for As removal, and to identify where additional Fe may theoretically facilitate improved remediation. This geochemical approach could be used to inform optimal selection of groundwater remediation approaches, when considered alongside other important considerations (*e.g.* technical, managerial and socio-economic) known to impact the effective implementation and sustainability of successful groundwater remediation approaches.

Keywords: arsenic, inorganic contaminants, remediation, water treatment, groundwater, decision-support

1. Introduction

Groundwater contamination, and particularly the presence of naturally-occurring arsenic (As), amongst other inorganic pollutants, is a major public health challenge [1, 2]. In the major floodplains and deltas of South and Southeast Asia, disproportionately high populations are exposed to As especially via the consumption of As-contaminated water supplies [3-7]. Geogenic groundwater As has been widely reported in the Ganga Basin of India [8], particularly in the States of Bihar [9-17], West Bengal [18-38] and Uttar Pradesh [39-42], with impacted populations estimated to be ~ 1.2 – 4.6 million, ~ 7.4 – 10.1 million and ~ 1.2 – 2.5 million, respectively [43]. A widespread public health target, including as identified as part of the United Nations' Sustainable Development Goal (SDG) 6, is to reduce As concentrations in water used for drinking to below the World Health Organization (WHO) provisional guideline value of 10 $\mu g.L^{-1}$ [6] (see also Indian drinking water standards [44]), whilst noting that detrimental health impacts may still occur at lower concentrations [45] or exposures [46], and in some cases efforts are being made to reach lower targets in drinking water [47]. Whilst there are a number of inorganic groundwater contaminants that may be of concern, we focus here primarily (but not exclusively) on As due to its priority for public health protection.

A number of technical strategies for As remediation exist and have been reviewed extensively elsewhere [48-56]. Additionally, the importance of holistic approaches to promote sustainability has been well-documented [57-61]. In areas which do not have access to centralized treated water supplies, especially in rural or peri-urban areas, or in parts of cities which do not have treated supplies, household or community scale interventions to reduce As concentrations in drinking supplies are sometimes applied [62]. Small scale technology-based interventions may include filtration systems enhanced with sorption media such as activated alumina, granular ferric hydroxide, or hybrid anion exchange media [59, 62, 63] (particularly at community-scale) or via membrane technologies such as reverse osmosis (RO) or nanofiltration (either at household or community-scale) [64, 65]. Other strategies, such as those based on zero-valent iron (ZVI) [66], *in-situ* approaches [63] and electrolytic technologies (*e.g.* electro-coagulation, electro-chemical arsenic remediation) [58], also exist but tend to be relatively less commonly implemented [62]. Small scale, household point-of-use systems have been recommended to be most suitable for short term use (*e.g.* disaster situations) but often fail in the medium-term [67] and previously have not been recommended for long term As mitigation [68], whilst recognizing that suitable alternative options are limited in areas not served by centralized treatment systems. In Bihar, typical small-scale groundwater remediation systems are commonly either sand-based filters (sometimes in tandem with sorption media) or commercial RO systems. Piped supplies of treated surface water are also an important mitigation option in some locations in Bihar. The Department of Drinking Water & Sanitation, Ministry of Jalshakti, Government of India provides resources regarding remediation strategies including through the advice of technical committees and a knowledge hub [69, 70].

Source water composition (including As, P, Fe) is known to have a significant influence on the performance of various groundwater remediation technologies for As removal (particularly adsorption-based removal technologies) [71]. For example, groundwaters characterized by low natural Fe and high P, such as in Bangladesh, require an additional source of Fe to support As removal, whereas high natural Fe concentrations (such as in the Red River Basin, Vietnam) facilitate As removal [71], noting this is technology-dependent and other parameters as well (particularly pH and redox conditions) also may impact removal. In shallow, reducing aquifers typical to South/Southeast Asia [4, 72], another challenge is that relatively high concentrations of As(III) may require oxidation to As(V) in order to enhance removal efficiency in part due to the stronger sorption capacity of As(V) [71]. The presence of P, Si (typically as H_4SiO_4 in dissolved form) and HCO_3^- can also significantly impact As reduction [73]. Although it is very important to consider geochemical composition during remediation selection [71], this often does not occur in practice in such settings. Further, manufacturer specifications on system performance for commercial products often refer to controlled laboratory studies which do not necessarily reflect typical operating conditions and/or real-world implementations, and many systems may not be tested using real environmental samples or representative groundwater matrices.

In addition to geochemical suitability, there are a number of other important aspects which impact the effectiveness or long-term sustainability of remediation interventions; this includes factors such as operation and maintenance requirements, cost, amenability to automation and broader acceptability and sustainability aspects [62, 74]. In As-impacted areas of Bihar, remediation initiatives at the community scale have been implemented by the State government, academic institutions and non-governmental organizations. However, many community-scale filtration units have not been successful and have been left non-functional, particularly due to issues regarding maintenance and monitoring, improper installation, lack of acceptability (including for economic or convenience reasons), lack of clarity around responsibility, lack of awareness and socio-economic exclusion [75]. The reasons behind implementation failures are often complex and depend on the specific locality. For example, in some cases maintenance issues may cause failures, whereas in other areas, even nearby, socio-economic exclusion or social conflict may be the limiting barrier. These issues can collectively create major challenges for the sustainability of such community-scale mitigation approaches. Small household scale, point-of-use remediation units are also relatively common in some areas, particularly in more affluent or urban settings. Whilst private household systems may reduce some of these barriers, issues around maintenance, cost and equitable access can still be prevalent.

Given the wide variety of technical and non-technical factors which can influence the implementation of remediation initiatives, it remains unclear how effective various small-scale remediation systems are in reducing concentrations of As and other (inorganic) chemical solutes in groundwater in Bihar. To our knowledge, a knowledge gap remains in evaluating the performance of a variety of remediation systems actually being implemented under typical "on-the-ground" conditions in Bihar. As such, the aim of this paper is to assess the effectiveness and dominant performance controls on a range of commonly used household and community-scale local groundwater remediation systems in Bihar, India, under typical operating conditions, and to consider the implications on remediation selection in similar settings. The objectives are to: (i) evaluate the removal of As and other inorganic solutes with commonly locally-used remediation approaches and typical operating conditions; (ii) compare the effectiveness of various remediation technologies (*e.g.* RO-based systems, sand filtration) and implementation settings/scales (*e.g.* household versus non-household); (iii) evaluate theoretical geochemical controls on system performance in comparison with actual performance data; and (iv) consider the implications on the selection and management of groundwater arsenic remediation systems in Bihar and elsewhere.

2. Methods

2.1 Study Area & Sampling Strategy

Water samples were collected in 2019 from remediation systems (n = 38) located in the State of Bihar in the Middle Gangetic Plain, India, within the following districts: Patna (n = 21); Buxar (n = 4); Aurangabad, Gaya and Katihar (n = 2 each); and East Champaran, Nawada, Munger, Rohtas, Saran and Vaishali (n = 1 each). Further samples collected from one system located in Ballia District (Uttar Pradesh) were also included in this study because of the proximity of the study site to Bihar and their co-existence in the Mid Ganga Plain. Samples from remediation systems were collected opportunistically within the framework of a larger stratified random groundwater sampling campaign systematically encompassing all districts of Bihar [17]. The wider groundwater sampling campaign involved sampling of ~ 300 tubewells distributed across Bihar [17] and the identified mitigation units reported in this current study are generally indicative of the frequency and types of mitigation systems encountered during random groundwater sampling (noting that more units were more commonly encountered in urban areas such as Patna). Upon arrival at a particular location the field team asked locally if there were any remediation systems present in the surrounding households or community. Sampling was then undertaken if remediation systems were identified and access was granted. Importantly this meant that all samples were collected under “spot check” conditions, under typical operating conditions for that particular system/setting, and where owners or overseers had no prior knowledge that sampling was to take place. In a limited number of cases in Buxar and Patna, sampling was carried out on remediation systems already known to exist by members of the research team. The higher proportion of samples collected in Patna district reflected both the higher density of groundwater sampling points in Patna as well as Patna being an urban area where household point of use water treatment systems are more prevalent.

In this manuscript, we use the term “remediation” as a broad term to encompass multiple potential approaches for the mitigation of one of more groundwater contaminants to minimize risks for human health. This may include the implementation of point-of-use water treatment systems (as largely reported here) as well as other options, for example switching to a less-contaminated source (which potentially may not require any water treatment technology). Although variations in terminology are used in the literature, “remediation” is commonly used in the context of geogenic arsenic mitigation and in source-pathway-receptor models reported in environmental risk assessment [1, 50, 76, 77].

2.2 Remediation System Sampling & Characterization

For each remediation system identified (n = 38, see **Section 3.1**), subsamples of (i) untreated groundwater sources used as the system feed/inlet and (ii) corresponding finished product/outlet water were sought to be collected. Some additional packaged water from local suppliers was also sampled, as packaged water supplies can also be considered a remediation approach, although the corresponding inlet groundwater was not possible to sample. Inlet water samples were typically collected either directly from corresponding handpumps, using methods previously published [17], or from household taps connected directly to the untreated groundwater source. Outlet water samples were collected directly from system outlets or from the nearest point of access (in some cases this was from the outlet of a connected storage vessel). All samples were collected in plastic beakers which were thoroughly sample rinsed between samples.

Measurements of *in-situ* parameters pH, electrical conductivity (EC) and temperature were undertaken directly at the time of sampling using Hanna handheld meters. Subsamples for subsequent laboratory analysis of major and trace cations and anions were filtered (0.45 µm cellulose/polypropylene syringe filers) upon collection and stored in glass bottles. Samples for cation and trace metal(loid) analysis were acidified (2 % trace grade HNO_3) after transport and arrival at the University of Manchester laboratories (Manchester Analytical Geochemistry Unit), due to HNO_3 transport restrictions. Further sampling details are provided elsewhere [17].

Further information was obtained about the remediation systems sampled and their typical usage, both through discussion with the local owner/caretaker and observationally. The following types of information were ideally sought, directly or indirectly: (i) technology type and brand, if applicable; (ii) typical maintenance undertaken and/or the nature of any existing maintenance agreements; (iii) why the system was installed/purchased; (iv) age of system; (v) location of purchase, if applicable; (vi) approximate cost of system and maintenance, if applicable; (vii) who is typically responsible for water-related decisions; and (viii) the general upkeep/cleanliness of the surrounding area. It was not always possible to collect all this information, depending highly on the person(s) present. As much information was obtained as possible and as situationally appropriate.

2.3 Chemical Analysis (Laboratory)

Chemical analysis of major and trace elements was undertaken at the Manchester Analytical Geochemistry Unit (MAGU). Inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500cx) was used for the analysis of As, U and Zn. Inductively coupled plasma atomic emission spectrometry (ICP-AES, Perkin-Elmer Optima 5300 dual view) was used for the analysis of Fe, P, Ca, Mg, Mn, Na, K and Si. Ion chromatography (IC, Dionex ICS5000 Dual Channel Ion Chromatograph) was used for the analysis of F^-, Cl^-, NO_3^- and SO_4. Further analytical method details and information on quality assurance/quality control are provided elsewhere [17].

2.4 Data Analysis

Removal (R_i, as %) was calculated as:

$$R_i = \left(1 - \frac{C_{outlet,i}}{C_{inlet,i}}\right) \cdot 100 \qquad \text{Eqn 1}$$

where $C_{outlet,i}$ and $C_{inlet,i}$ are outlet and inlet concentrations, respectively, of component *i*. For the purpose of calculating molar ratios and removal, measured solute concentrations that were below detection limits were input as 0.1 µg.L^{-1} for As and 0.1 mg.L^{-1} for Fe and P (representing 10 % of lowest calibration standard and near instrumental detection limits) to enable calculations without divide-by-zero errors which are thus maximum or minimum expected values depending on the ratio calculated. The terminology "removal" and "retention" are often used interchangeably particularly in the context of membrane systems and here "removal" is used to be more broadly applicable across technology types.

OriginPro 2017 was used for basic statistical analysis (*e.g.* simple linear regression, Mann Whitney U test, descriptive statistics) with 95 % confidence on reported p values unless otherwise stated. Multivariate generalised linear model (GLM) was used to determine the factors potentially associated with As removal. Appropriate functions were used to generate the best fitting model predicting the outcome (*e.g.* As removal). Firstly, the contribution of the independent variable ([Fe] -1.8[P])/[As] was calculated (crude model), based on results from simple linear regression. The additional geochemical factors (*e.g.* Fe, As, P, Ca, Mg, Na, Si, [Fe]/[As] and [Fe]/[P]) and system characteristics (*e.g.* technology type and setting) were then included (adjusted) in order to assess the improvement to the overall prediction, and association of each of those factors to the best fitting model. Data was included in multivariate analysis if inlet As concentrations were above detection and removal values were positive (n = 20 observations met this criteria). Akaike's Information Criterion (AIC) values were used to select the best-fitted model. STATA 11.2 [78] was used for multivariate analysis. QGIS (version 3.12.2 București) was used for mapping.

3. Results and Discussion

3.1 Remediation System Characterization

The 38 remediation systems identified and sampled were initially characterized by system type/technology, scale and setting. A selection of the range in types of remediation systems encountered are shown in **Figure 1**. Most systems were commercial RO-based units (n = 27, mostly small-scale point of use systems), followed by multi-stage filtration systems (n = 4, all community-scale) and a single-stage homemade sand filtration system made in a ~ 50 L bucket (n = 1, household scale). The systems were installed and used in various settings, including in private households (n = 18) and in non-household settings (n = 13) such as communities, hospitals and hotels/hostels. Some of the sampled systems were implemented specifically for arsenic removal, although many were used for general or other specific water quality concerns (including high iron leading to red/orange residue and unsatisfactory taste/smell). Many owners of household systems reported that the composition of their inlet water had never been tested (although some still had concerns about the possible presence of arsenic). There were additional samples of packaged water (n = 6) with unknown remediation system type; these were typically from small local businesses providing a private paid water delivery service (cost reported to be ~ 15 to 20 INR, or ~ 0.20 to 0.30 USD, for a ~ 15 L large plastic bottle) and it was generally not possible to sample the associated inlet water for packaged water suppliers.

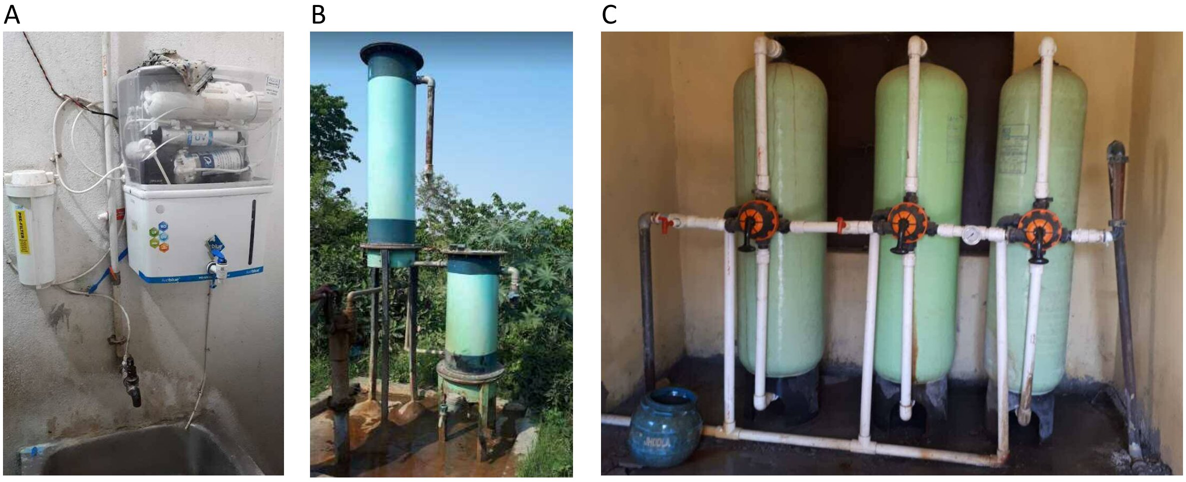

Figure 1. Example types of small-scale remediation units in Bihar: (A) Household scale commercial reverse osmosis (RO) based unit; (B) Multi-family scale gravity fed multi-stage sand filtration unit; (C) Community-scale commercial filtration unit.

The commercial RO systems were from a range of Indian and international brands and were typically bought from Patna or shops in district headquarters in Bihar. Most owners of the household RO systems reported maintenance arrangements ranging from paid service contracts (visits ~ 1 – 6 months) to occasional filter changes at the discretion of the owner (~ 6 months on average); in some cases maintenance practices were not apparent or unknown. Household RO systems were usually specified up to ~ 20 L/hour production rate using mains electricity (power consumption ~ 40 – 60 W) with a storage capacity of ~ 8 – 15 L. Details of specific RO membrane type was usually not available. Some RO systems were equipped with other remediation elements (e.g. pre-filter, UV disinfection stage) in series. The multi-stage filtration units were all community scale systems with filter media ranging from sand to proprietary commercial products. Commercial product labels on larger-scale commercial systems were all nearly worn off and unreadable, presumably due to long-term wear and tear, and caretakers were often not able to provide detailed technical product information. The household-scale handmade bucket sand filtration system was built from low cost and highly local materials (*e.g.* nearby gravel and sand) by a household personally concerned with orange residue in water obtained from their household tubewell; the sand in this system was reported to be changed monthly. In many cases reject/concentrate water from RO and other systems was reported to be simply discarded in a sink/drain (for household scale systems) or behind the system facilities (for community scale systems); management of waste products from water treatment systems is an important consideration for operation & maintenance and longer term sustainability [79]. Comprehensive information on all technologies used was usually not available during site visits and information was supplemented, where possible, through visits to local shops supplying various mitigation units. It is important to note that there are also likely variations between the same type of systems used in the study, so this context should only be considered indicative.

3.2 Characterization of Inlet Water Composition

In brief, groundwater in Bihar has been previously characterized as generally circum-neutral (pH ranging from 5.7 – 8.3) and typically of the $Ca\text{-}HCO_3^-$ and $Na\text{-}HCO_3^-$ water type [17]. The geochemical composition of the subset of groundwater samples reported here and used as inlet water for remediation units was broadly similar to the wider dataset across Bihar [17] (**Table 1**). Remediation system source water ranged in pH from 6.8 – 7.9, and arsenic concentrations ranged from < 1 – 200 $\mu g.L^{-1}$ (median ~ 1 $\mu g.L^{-1}$), with ~ 20 % exceeding the WHO provisional guideline value of 10 $\mu g.L^{-1}$. Other parameters which exceeded WHO guideline values for a sub-set of samples included NO_3^- (~ 16 % exceedance of 50 $mg.L^{-1}$); Mn (~ 10 % exceedance with regard to previous WHO guideline, noting that the WHO previous guideline of 0.4 $mg.L^{-1}$ has been discontinued; Indian Drinking Water guidelines stipulate 0.1 $mg.L^{-1}$ [44] of which ~ 50 % are in exceednace); and U (~ 10 % exceedance of provisional guideline of 30 $\mu g.L^{-1}$). In general there was a narrower distribution of concentrations of trace and major elements in the subset of remediation samples as compared with a Bihar-wide dataset [17], consistent with the smaller sample numbers and uneven distribution of sampling locations corresponding to where remediation units were present.

Table 1. Composition of groundwater sources used as inlet water for remediation units (this study; n = 31), as compared with representative groundwater samples across all districts of Bihar (n = 273) [17], and arsenic impacted groundwater from Bangladesh (B-GW) and New Hampshire (NH) as reported by other authors and used in co-precipitation tests [80]. Data shown for Bihar groundwater are reported as a range with median value in parentheses; 'n/a' indicates data not available. [Fe]/[As] molar ratios are shown for natural groundwaters and as required for As removal based on co-precipitation tests published elsewhere (* = required to reduce As to < 50 $\mu g.L^{-1}$; ** sufficient for ~ 100 % removal) [80].

	Bihar, Remediation Units (this study)	Bihar, All [17]	B-GW [80]	NH [80]
As ($\mu g.L^{-1}$)	< 1 – 200 (1)	< 1 – 870 (1)	280 - 600	70
Fe ($mg.L^{-1}$)	< 0.1 – 10 (< 0.1)	< 0.1 – 10 (< 0.1)	4.7 – 7.7	0.7
P ($mg.L^{-1}$)	< 0.1 – 1.8 (0.1)	< 0.1 – 1.8 (0.1)	1.6 – 2.7	0.02
Na ($mg.L^{-1}$)	10 – 110 (30)	3 – 250 (30)	15 – 78	13
Ca ($mg.L^{-1}$)	50 – 220 (80)	10 – 240 (70)	65 – 151	16
Mg ($mg.L^{-1}$)	10 – 100 (20)	2 – 150 (20)	14 - 42	2.9
Si ($mg.L^{-1}$)	12 – 18 (16)	6 – 30 (15)	14 - 20	6.6
K ($mg.L^{-1}$)	1 – 50 (3)	0 – 100 (3)	n/a	n/a
Mn ($mg.L^{-1}$)	< 0.1 – 1 (0.1)	< 0.1 – 4 (0.1)	n/a	n/a
Zn ($\mu g.L^{-1}$)	3 – 1600 (20)	2 – 5400 (15)	n/a	n/a
U ($\mu g.L^{-1}$)	< 1 – 50 (3)	0 – 80 (2)	n/a	n/a
Cl^- ($mg.L^{-1}$)	0.1 – 220 (4)	0.1 – 450 (10)	n/a	n/a
SO_4 ($mg.L^{-1}$)	< 0.1 – 180 (2)	< 0.1 – 230 (10)	n/a	n/a
NO_3^- ($mg.L^{-1}$)	< 0.1 – 130 (0.4)	< 0.1 – 220 (0.4)	n/a	n/a
F^- ($mg.L^{-1}$)	< 0.1 – 0.7 (< 0.1)	< 0.1 – 1.3 (0.1)	n/a	n/a
EC ($\mu S.cm^{-1}$)	500 – 2000 (600)	30 – 3000 (650)	n/a	n/a
pH (--)	6.8 – 7.9 (7.3)	5.7 – 8.3 (7.2)		
Fe / As (M:M); natural	~ 0 – 330 (~ 1)	~ 0 – 6800 (~ 17)	~ 17 - 30	~ 13
Fe / As (M:M); required	--	--	~ ≥ 54 *	~ < 16 **

3.3 Performance of Community and Household Systems under Typical Operation

3.3.1 Reduction of Arsenic with Community and Household Systems

Arsenic removal varied widely, ranging from ~ 0 % to 100 % (median 87 %), with removal usually > ~ 40 % for As inlet concentrations > 1 $\mu g.L^{-1}$ (**Figure 2** and **Table 2**). Most systems were reasonably effective, noting that reported removals ~ 0 % usually corresponded to inlet concentrations near detection. In most cases (with the exception of two), permeate As concentrations were well below the WHO guideline value of 10 $\mu g.L^{-1}$, although source water As concentrations only exceeded guideline values in a limited number of cases. Importantly, the highest raw groundwater As (~ 200 $\mu g.L^{-1}$) was encountered with the homemade bucket sand filter system – so although a low technology solution, the ~ 90 % reduction achieved under the site-specific conditions still substantially reduced As concentrations (to ~ 20 $\mu g.L^{-1}$) , even though the final product water still exceeded WHO guideline values. The relatively high removal of the bucket sand filter is noteworthy as it demonstrates that simple, homemade solutions can be comparatively as effective as commercial products in some circumstances. For the avoidance of doubt please note that this does not imply that such simple technologies are likely to be effective under *all* circumstances. All of the packaged water sampled (n = 6) had As concentrations < 1 $\mu g.L^{-1}$, although corresponding removal values cannot be calculated as source water composition was unknown. Controls on performance are discussed in **Section 3.4**.

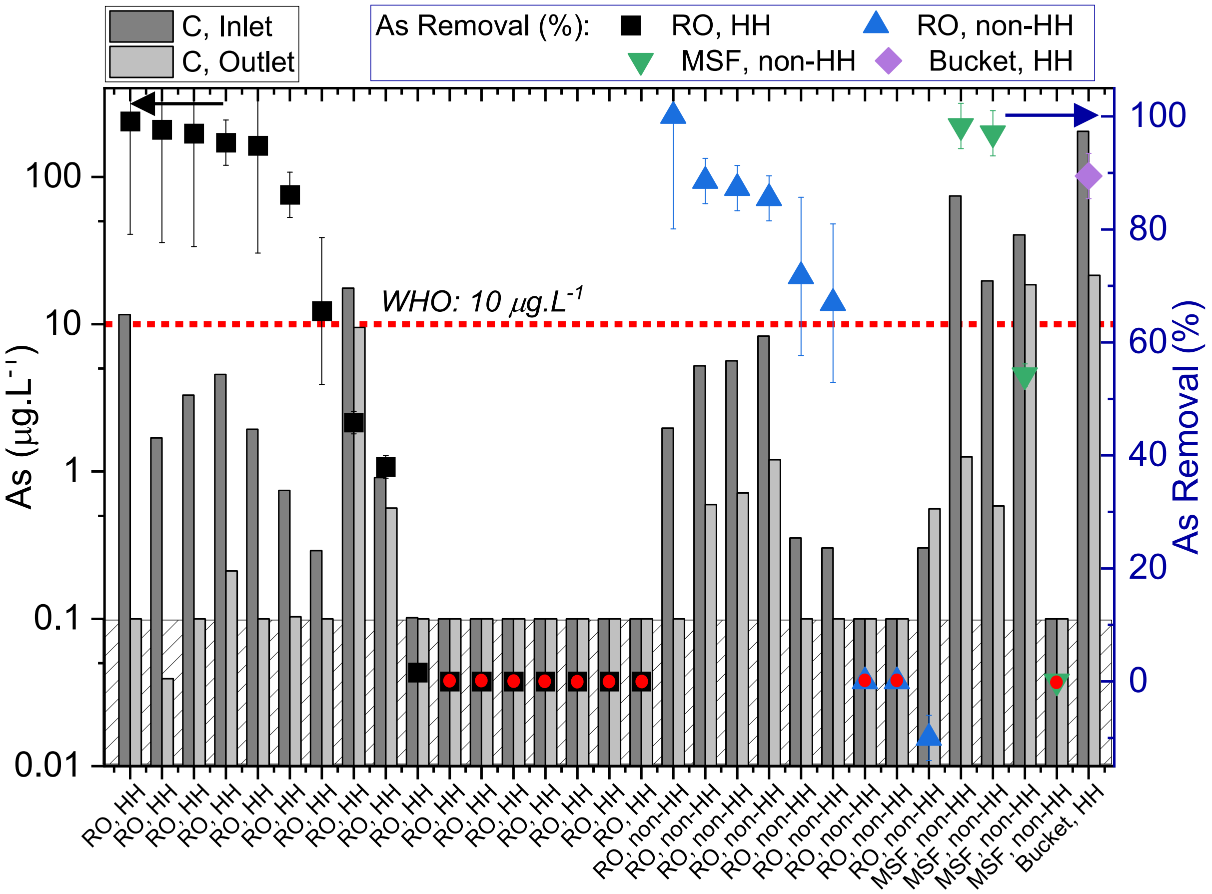

Figure 2. Concentrations of arsenic in in inlet (dark grey) and outlet (light grey) water (left axis, as µg.L^{-1} in log-scale) and arsenic removal (right axis, as %) for all remediation systems sampled under "typical" operation where paired inlet-outlet samples were available (n = 31). Symbol shape/color indicates system type: grey square = reverse osmosis (RO)-based technology in a household (HH) setting; blue up-facing triangle = RO technology in a non-HH setting (*e.g.* community, hospital, hostel/hotel); purple diamond = homemade bucket filtration system (non-RO) in HH setting; green down-facing triangle = multi-stage filtration system (non-RO) in a non-HH setting. The grey diagonally dashed box indicates concentrations below approximate instrumental detection (estimated to be ~ 0.1 µg.L^{-1}, noting in some cases indicative peaks were identified below this limit); concentrations not detected are shown at a conservative estimate of the maximum value. Estimated errors on removal were conservatively estimated on the basis of estimated uncertainties of 20 % and 3 % for analytical uncertainties for As measurements < 1 µg.L^{-1} and > 1 µg.L^{-1}, respectively, and propagated for the removal calculation. Propagated removal errors are therefore relatively high when permeate concentrations are low. A red dot in some symbols indicates where apparent removal is calculated to be 0 % due to concentrations in both inlet and outlet being below detection (this also means that apparently calculated propagated errors for these sample sets are 0 %); these datapoints are included to visualize the full dataset although over-interpretation of these particular datapoints should be avoided.

Table 2. Removal (R) values (%) reported as "Range (Median)" for (i) all paired samples (n = 31; note difference in calculation basis marked by § or ‡), by technology: (ii) reverse osmosis (RO; n = 26) and (iii) non-RO (n = 5), comprising of multistage filtration (n = 4) and simple sand filtration (n = 1), and by system setting: (iv) household (HH; n = 18) and (v) non-HH (*e.g.* community, hospital, hostel/hotel setting; n = 13). Negative removals indicate that concentrations in outlet are reported to be higher than the inlet; in some cases this is when concentrations are very near detection and errors are large. Removals less than -10 % have been input as -10 % as a maximum value in the limited cases where this was applicable. The columns RO versus non-RO and HH versus non-HH show asymptotic p-values reported within the 95 % confidence level as determined by the Mann-Whitney non-parametric statistical test; these determine if the distributions of removals between technology type and setting type, respectively, are significantly different (if $p < 0.05$ the distributions are statistically different and marked in bold). § Statistics calculated based on the parameter-specific exclusion of below detection values in inlet concentrations (n = 21, 23, 27, 29 and 14 for As, Fe, P, NO_3^- and F^-, respectively, where not all inlet water had detectable levels present); ‡ Statistics calculated on the basis of full dataset of paired samples (n = 31) where all inlet samples had detectable concentrations.

Parameter	**R (%), All**	**R (%), RO**	**R (%), non-RO**	**R (%), HH**	**R (%), non-HH**	**p, RO vs non-RO**	**p, HH vs non-HH**
Arsenic and Directly Related							
As §	< -10 – 100 (87)	< -10 – 100 (86)	0 – 98 (89)	0 – 99 (89)	< -10 – 100 (86)	> 0.05 (0.39)	> 0.05 (1)
Fe §	< -10 – 100 (97)	< -10 – 100 (94)	52 – 100 (100)	< -10 – 100 (84)	48 – 100 (99)	> 0.05 (0.07)	**< 0.05 (0.03)**
P §	16 – 99 (93)	16 – 99 (92)	71 – 98 (93)	16 – 98 (88)	65 – 99 (95)	> 0.05 (0.56)	> 0.05 (0.10)
Other Trace Inorganics							
U ‡	< -10 – 100 (97)	< -10 – 100 (98)	-5 – 97 (28)	< -10 – 100 (97)	-5 – 100 (97)	**< 0.05 (0.02)**	> 0.05 (0.51)
Zn ‡	< -10 – 99 (56)	< -10 – 99 (56)	< -10 – 99 (< -10)	< -10 – 99 (67)	< -10 – 99 (< -10)	> 0.05 (0.44)	**< 0.05 (0.02)**
Multi-valent Cations							
Mn ‡	< -10 – 100 (99)	<-10 – 100 (99)	< -10 – 99 (97)	53 – 100 (99)	< -10 – 100 (97)	> 0.05 (0.28)	> 0.05 (0.73)
Mg ‡	-2 – 100 (90)	6 – 100 (92)	-2 – 96 (6)	6 – 100 (93)	-2 – 100 (84)	**< 0.05 (0.01)**	> 0.05 (0.56)
Ca ‡	2 – 100 (84)	19 – 100 (89)	2 – 96 (23)	11 – 99 (92)	2 – 100 (82)	> 0.05 (0.06)	> 0.05 (0.62)
Mono-valent Cations							
K ‡	-7 – 100 (79)	3 – 100 (79)	-7 – 90 (-2)	-1 – 94 (78)	-7 – 100 (79)	**< 0.05 (0.01)**	> 0.05 (0.98)
Na ‡	< -10 – 100 (77)	4 – 100 (78)	< -10 – 88 (-3)	4 – 92 (79)	< -10 – 100 (97)	**< 0.05 (0.02)**	> 0.05 (0.59)
Anions							
NO_3^- §	< -10 – 100 (71)	< -10 – 100 (86)	< -10 – 56 (< -10)	< -10 – 99 (55)	< -10 – 100 (99)	> 0.05 (0.09)	> 0.05 (0.05)
F^- §	< -10 – 99 (63)	< -10 – 99 (79)	< -10 – 22 (14)	-7 – 99 (78)	< -10 – 99 (44)	> 0.05 (0.16)	> 0.05 (0.62)
SO_4 ‡	< -10 – 100 (59)	< -10 – 100 (72)	< -10 – -4 (-7)	< -10 – 100 (57)	< -10 – 100 (71)	> 0.05 (0.06)	> 0.05 (0.90)
Cl^- ‡	< -10 – 100 (56)	< -10 – 100 (81)	< -10 – 1 (-3)	< -10 – 97 (67)	< -10 – 100 (34)	**< 0.05 (0.02)**	> 0.05 (0.82)
Other							
Si ‡	< 10 – 95 (72)	4 – 95 (75)	< -10 – 92 (5)	-3 – 95 (89)	< -10 – 92 (68)	**< 0.05 (0.02)**	> 0.05 (0.07)

3.3.2 Reduction of Other Inorganic Solutes with Community and Household Systems

In addition to As, the reduction of other inorganic solutes with the various remediation systems was also evaluated (**Figure 3** and **Table 2**). The overall selectivity sequence, based solely on the median removal of all paired samples (excluding those where inlet concentrations were below detection) in decreasing order, follows: Mn (99 %) > Fe (97 %) ≈ U (97 %) > P (93 %) > Mg (90 %) > As (87 %) > Ca (84 %) > K (79 %) > Na (77 %) > Si (72 %) > NO_3^- (71 %) > F^- (63 %) > Cl^- (56 %) ≈ Zn (56 %) > SO_4 (56 %). When the distributions of removals (rather than median values) are considered, three broad groupings where a significant difference (at the 0.05 level) in distributions were observed: Mn, Fe, U > P, Mg, As, Ca > K, Na, Si, NO_3^-, F^-, Cl^-, Zn and SO_4. Due to the wide distributions in removal observed, these groupings, rather than median removal, values are more likely to be indicative of general selectivity trends. This indicates which contaminants were generally easier (*e.g.* Mn, Fe, U) and more challenging (*e.g.* K, Na, Si, NO_3^-, F^-, Cl^-, Zn and SO_4) to remove in the studied systems, noting there remains a wide range of variability in the removal of all of these parameters across the remediation systems sampled. The relatively high median removals for Mn and U (which exceeded guideline values in some cases) suggests that many of the remediation systems were reasonably effective for these higher priority contaminants as well as As. Similarly, some of these systems also removed F^- to some extent, although the lower removals observed for F^- suggest the systems were not optimally designed for this pollutant (whilst noting that inlet concentrations were below guideline values). Concentrations of Mn, U and NO_3 (parameters which exceeded guideline values in some of the inlet waters sampled) in outlet waters were always less than the corresponding WHO guideline values for the systems sampled (noting Mn exceeded the Indian Drinking Water Standard [44] in one outlet sample).

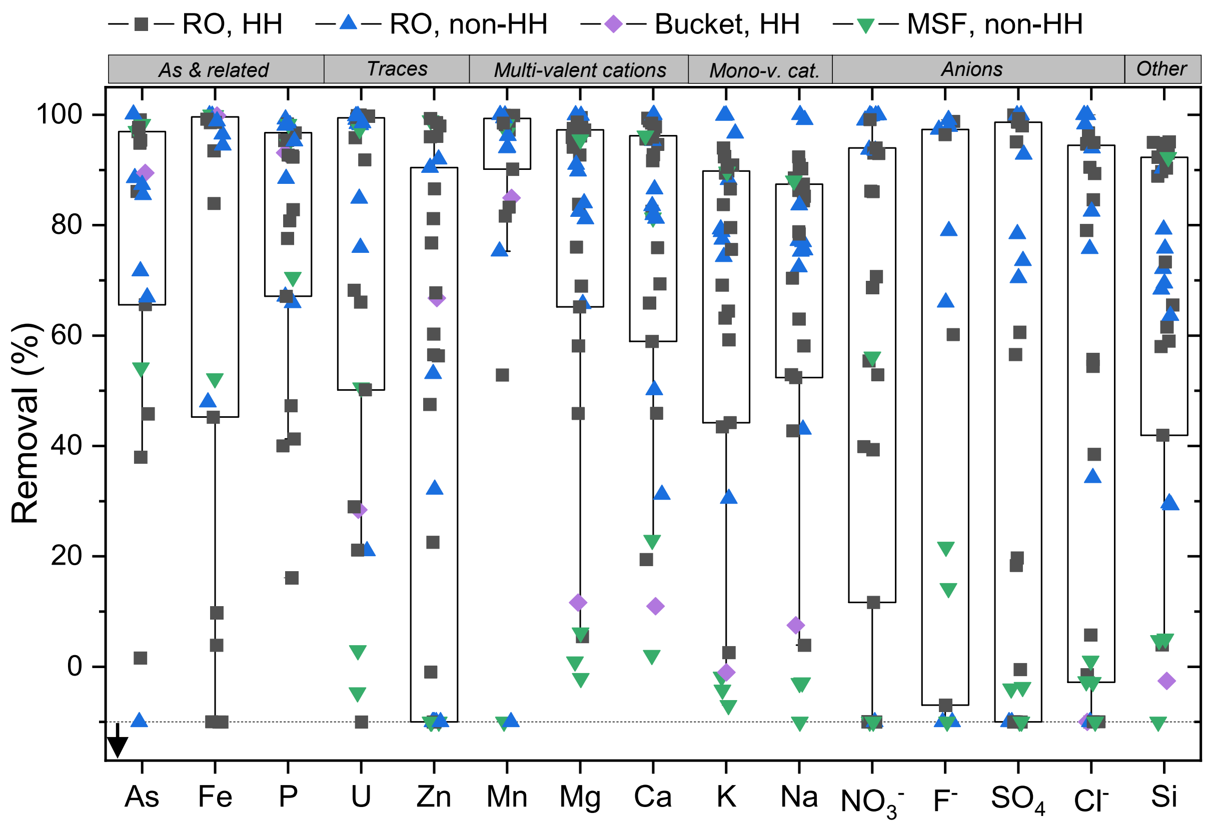

Figure 3. Box plot of the removal (%) of various chemical components including arsenic and related (As, Fe, P) and in decreasing order of median value per category as categorized by trace elements (U, Zn), multi-valent cations (Mn, Mg, Ca), mono-valent cations (K, Na), anions (NO_3^-, F^-, SO_4, Cl^-) and other (Si). Negative removals indicate that concentrations in outlet were reported to be higher than the inlet; in some cases this is when concentrations were near detection and errors are large. Removals less than -10 % were input as -10 % as a maximum value in the limited cases where this was applicable (indicated by down arrow at -10 %). Bold line in box indicates median value; whiskers represent the 10 – 90 % distribution. Symbol shape/color indicates system type: grey square = reverse osmosis (RO)-based technology in a household (HH) setting; blue up-facing triangle = RO technology in a non-HH setting (*e.g.* community, hospital, hostel/hotel); purple diamond = homemade bucket filtration system (non-RO) in HH setting; green down-facing triangle = multi-stage filtration system (non-RO) in a non-HH setting. Data excluded if inlet concentrations are below detection.

3.3.3 **Ion selectivity in RO systems**

When only membrane-based RO systems are considered, general selectivity trends based on significance of differences in removal distributions again showed three broad groupings: Mn, U, Fe > P, Mg, Ca, As, NO_3^-, Cl^-, F^-, K, Na, Si, SO_4 > Zn. The selectivity sequence based solely on median removal values was: Mn (99 %) > U (98 %) > Fe (94 %) > P (92 %) ≈ Mg (92 %) > Ca (89 %) > As (86 %) ≈ NO_3^- (86 %) > Cl^- (81 %) > F^- (79 %) ≈ K (79 %) > Na (78 %) > Si (75 %) > SO_4 (72 %) > Zn (56 %), noting that, as above, this should not be over-interpreted given the broad distributions encountered. The relatively high removal of multivalent ions (*e.g.* Mn, Mg, Ca) as compared to monovalent ions (*e.g.* NO_3^-, Cl^-, F^-, K, Na) was generally consistent with charge exclusion mechanisms well-demonstrated in membrane literature [81]. Whilst pH has been demonstrated to substantially impact the removal of some solutes in RO [81, 82], the variation of natural pH in the source waters in this study was relatively small (range 6.8 – 7.9) and thus pH is not expected to be a dominant control on removal or selectivity in this case. High variability in the removal of As specifically using membrane technology has been reported in other settings ranging from near negligible to near complete depending on membrane type, operating conditions and groundwater composition [83-88]. More detailed mechanistic evaluation of ion selectivity trends in the RO-based systems is difficult given the variability in source water chemistry, membrane type and operating conditions encountered in this study, although this would be an interesting topic for future work. Systematic investigations of a range of available RO-based systems under a range of typical groundwater matrices that may be encountered would be a future recommendation for more detailed understanding of dominant removal mechanisms and matrix influences.

3.4 Controls of the Effectiveness of Community and Household Remediation Systems

3.4.1 **Technology Type and System Setting**

The statistical significance of differences in the distributions of removal values observed for the various parameters was considered with respect to groupings according to technology type and system setting (**Figure 4**). Interestingly, there was no significant difference (at the 95 % confidence level) in the removal distributions for RO- versus non-RO technology for As, as well as Fe, P, Zn, Mn, Ca, NO_3^-, F^-, and SO_4 (**Table 2**). The implication is that, at least for these parameters, and under these site-specific conditions, the RO-based systems were comparatively effective as (and as variable as) the non-RO systems. Given the similarities in removal and the range of variability across system type, this data suggests that the remediation technology itself was likely not the dominant control on performance for As and the other parameters with similar trends, suggesting that the technologies implemented were reasonably well-suited for the contaminants and groundwater conditions observed in our study. Relatively high degrees of removal have been reported for some of these parameters in other non-RO systems and under different conditions, for example ~ 95 % removal of nitrate has been reported by other authors in a slow sand filter due to biological denitrification [89], and calcium removal ranging from ~ 20 – 95 % has been reported using flocculation and sand filtration which is highly dependent on water composition [90].

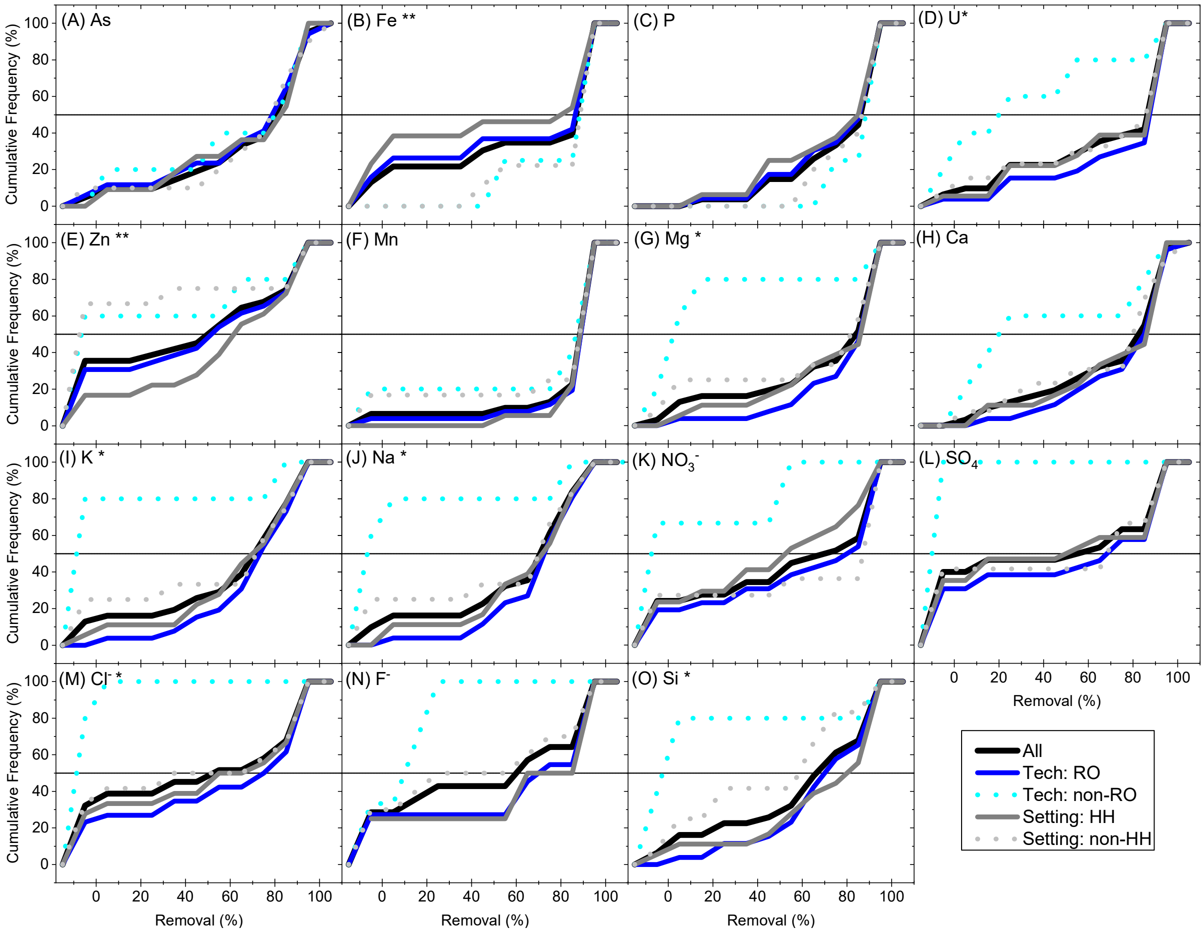

Figure 4. Cumulative frequency distribution of removal (bin size 10 % plotted at bin center) for parameters (A) As; (B) Fe; (C) P; (D) U; (E) Zn; (F) Mn; (G) Mg; (H) Ca; (I) K; (J) Na; (K) NO_3^-; (L) SO_4; (M) Cl^-; (N) F^-; and (O) Si. Line type represents grouping: black solid line is all data; blue solid line is reverse osmosis (RO)-based technology (n = 26); cyan dashed line is non-RO technology (n = 5); grey solid line is household (HH) setting (n = 18); light grey dashed line is non-household setting (*e.g.* community, hospital, hostel/hotel) (n = 13). * indicates that the distributions of removals from RO versus non-RO technologies were significantly different; ** indicates that the distributions of removals from HH versus non-HH settings were significantly different; both are reported within the 95 % confidence level as determined by the Mann-Whitney non-parametric statistical test. Negative removals less than -10 % have been input as -10 % as a maximum value in statistical analysis in the limited cases where this was applicable. Removal data excluded if inlet concentrations are below detection.

However, even though there was not a significant difference in the distributions between RO- and non-RO based technology for the reduction of As (as well as Fe, P, Zn, Mn, Ca, NO_3^-, F^-, and SO_4), this was not necessarily the case for all parameters, especially those which are more challenging to remove. In particular, the distributions of removal for U, Mg, K, Na, Cl^- and Si were observed to be dependent on technology type. Notably, better performance of RO-based technologies was observed for each of these elements, with distributions trending towards generally higher removals achieved in RO rather than non-RO systems. This suggests whilst technology type does not appear to be a major control on the effectiveness of As removal based on the systems spot-evaluated in this study, this could be a more important consideration for the removal of particular types of other contaminants depending on remediation priorities. A detailed mechanistic comparison of the removal of specific ions (e.g. Mg^{2+} versus Ca^{2+}) is beyond the scope of this study given the numerous co-variants inherent in this study and due to the experimental or computational requirements for a systematic mechanistic investigation, noting that ion mobility and interactions are also associated with relative dehydration energies [91] which has been shown to impact ion transport in modelled nanopores [92]. We emphasize that the observations made in this study, particularly the similar performance between RO- and non-RO based systems under the site-specific conditions, may not be applicable to all groundwater types (especially those where concentrations exceed or greatly exceed drinking water standards). Matrix-specific investigations should be conducted to help inform optimal remediation selection for particular conditions.

Further, there does not appear to be a systematic difference across parameters with regard to the type of setting (*e.g.* household versus non-household) where the remediation unit is installed (**Table 2**). This suggests that the dominant control on system performance was also likely not something that was specific to those types of settings (*e.g.* the type of person that may be responsible for system care would likely be different in a household as compared to a community or business setting), whilst noting management/maintenance arrangements are highly variable. Further, this indicates that one type of installation setting was not necessarily inherently more susceptible to remediation successes (or failures), and that both household and non-household systems can, and do, both perform effectively under the right conditions. A statistically significant difference in distributions of removal was only observed for two parameters only when HH vs non-HH settings were compared: Fe (generally higher removal in non-HH systems) and Zn (generally higher removal in HH systems). It is likely that these observations relate to confounding factors rather than systematic trends given that they were not observed across parameters.

There was no apparent trend observed in co-variance of technology type and setting type; for example both RO-based and non-RO systems were found in both HH and non-HH settings alike. A detailed analysis of potential co-variance may be limited, in part, by the relatively small sample numbers and particularly the relatively small number of non-RO systems encountered.

3.4.2 Geochemical Controls of Arsenic Remediation System Performance

The influence of inlet water composition, particularly concentrations of As, Fe, P, Ca and Si, were considered (**Figure 5**) as these parameters have been observed to impact the performance of various As remediation technologies [71, 80, 93]. Here, no systematic relationship was observed between As removal and source water concentrations of As (*p = 0.46*, **Figure 5A**), Fe (*p = 0.54*, **Figure 5B**) nor P (*p = 0.51*, **Figure 5C**), suggesting that geochemical controls were more complex than simply the inlet concentration of these parameters. Notwithstanding, the highest concentrations of As, Fe and P were all clearly observed with the bucket system which may, in part, be related to the effective performance of even the simplest technology. In previous studies the wide variability of As removal using a household co-precipitation and filtration system was attributed in part to variations in PO_4 and silicate concentrations [80], both of which are associated with decreased As removal due to increased competition for Fe-(oxy)hydroxide sorption sites [93]. Theoretically, higher concentrations of silicates and carbonates negatively impact As removal due to competitive sorption, whereas Ca positively impacts As removal due to increased PO_4 and Fe sorption and precipitation [71]. Competitive sorption is still useful to consider in the context of RO systems as interactions between dissolved solutes in water and/or between dissolved solutes and charged membrane surfaces can impact both the steric hindrance and charge mechanisms known to impact ion transport in membrane systems (which is dependent on water composition, membrane material and operating conditions) [81, 94].

Here there was no statistically significant relationship observed between As removal and Ca (*p = 0.58*, **Figure 5D**) nor Si (*p = 0.42*, **Figure 5E**) although this is perhaps unsurprising given the confounding geochemical and operational variables in the real-world systems sampled. Although some broad general relationships may be hinted within the dataset, particularly within specific groups (for example trending towards a positive relationship between As removal and Si for the RO-HH subset) these are not statistically significant. The potential impacts of Si and Ca are difficult to systematically quantify in this study, particularly due to the limited range of groundwater matrices encountered.

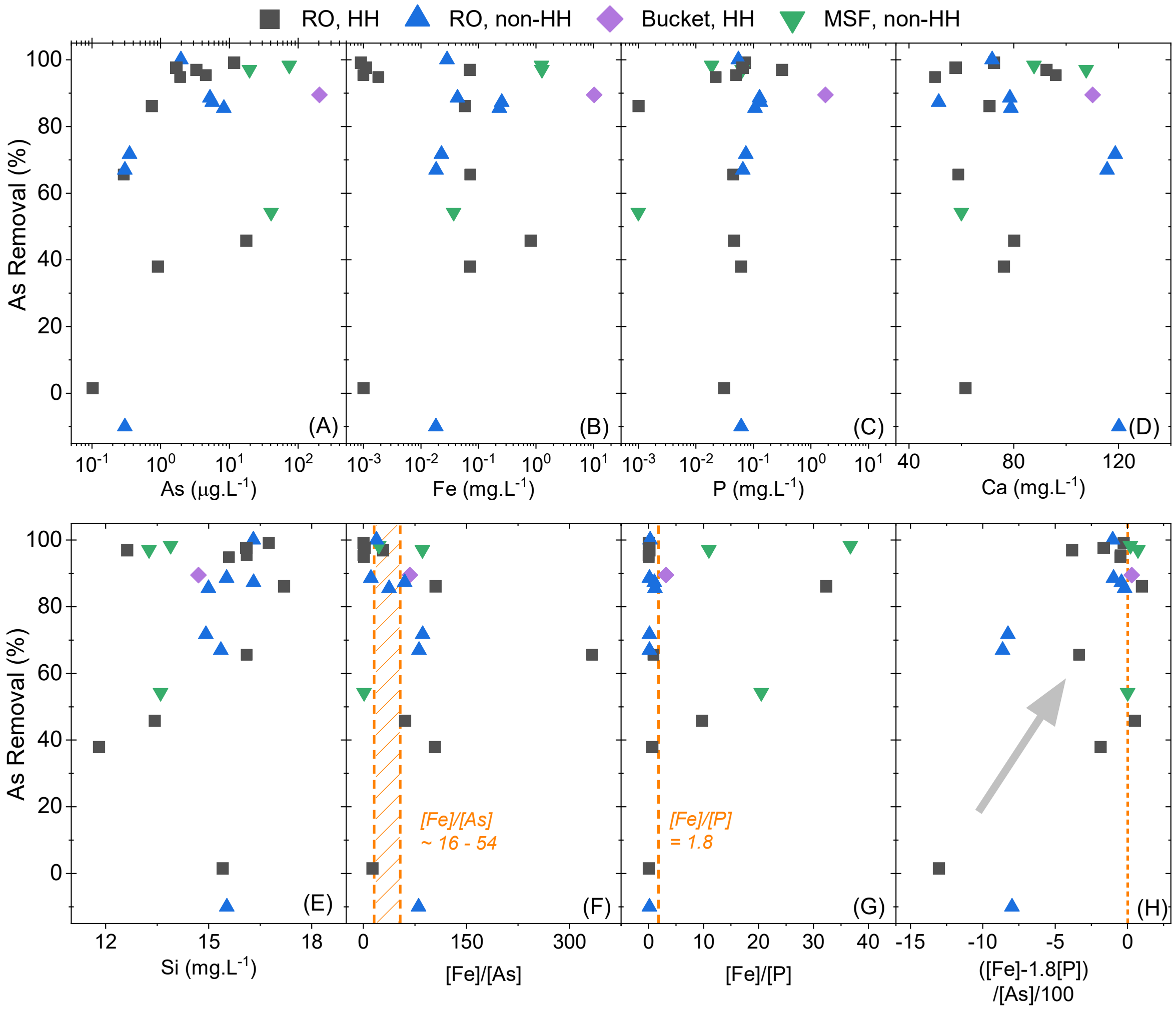

Figure 5. Arsenic removal (%) versus inlet water characteristics (A) As, (B) Fe, (C) P, (D) Ca, (E) Si, (F) the molar ratio [Fe]/[As], (G) the molar ratio [Fe]/[P], and (H) the molar ratio of ([Fe] – 1.8[P])/[As]/100 for various remediation systems sampled under "typical" operation. The orange dashed box on (F) represents [Fe]/[As] ranging from ~ 16 – 54 for comparison to approximate [Fe]/[As] ratios previously reported in other studies/settings to be sufficient for As removal under certain geochemical conditions [71, 80]. The dashed line on (G) represents [Fe]/[P] ~ 1.8, on the basis that a molar ratio of ~ 1.5 to 2.0 [Fe]/[P] is required for optimal removal of PO_4 at neutral pH [71]. The ratios shown on (H) represent the remaining [Fe]/[As] available after PO_4 removal on that same basis [71] (the dotted line here is at 0 and thus indicates if this ratio is positive or negative). The grey arrow on (H) indicates the statistical significance of a positive trend between As removal and ([Fe] – 1.8[P])/[As] (*t value = 3.8, degrees of freedom = 19, $p < 0.01$*). Removal data is shown at - 10 % as a maximum value in the single case where negative removal was calculated (this is likely due to high errors near instrumental detection limits); data excluded if inlet As concentrations are below detection. Note that inlet As, Fe and P are shown on a log-scale. Symbol shape/color indicates system type: grey square = reverse osmosis (RO)-based technology in a household (HH) setting; blue up-facing triangle = RO technology in a non-HH setting (*e.g.* community, hospital, hostel/hotel); purple diamond = homemade bucket filtration system (non-RO) in HH setting; green down-facing triangle = multi-stage filtration system (non-RO) in a non-HH setting.

The source water ratios of [Fe]/[As] and [Fe]/[P] have been reported to be more important geochemical determinants for As removal, rather than absolute values of individual concentrations, in other settings [71]. For example, high natural Fe concentrations in the Red River Basin (Vietnam) were shown to facilitate As removal, whereas Bangladeshi groundwaters characterized by low Fe and high PO_4 required additional Fe to support As removal [71]. It is known that high As concentrations, particularly As(III), combined with low Fe and high P and Si are particularly challenging, especially given that As(III) sorbs relatively weakly to precipitating Fe-(oxy)hydroxides as compared to As(V) which is oxidized and more strongly sorbing [71]. Whilst the impacts of this may be technology-dependent (for example in RO systems solute-solute interactions may lead to changing steric and charge interactions which impact retention mechanisms and/or may lead to membrane fouling), the underpinning principles of geochemical interactions remain similar. For example, in RO systems, Fe-As interactions in relative high Fe waters could plausibly improve As retention due to increased steric hindrance arising from larger apparent solute sizes, whereas in sand filtration Fe-As interactions and surface sorption on the sand itself may enhance As removal.

Based on co-precipitation tests using exemplar groundwaters from Bangladesh (representing a comparatively high As, Fe, P groundwater) and New Hampshire, USA (representing a mid-As, lower Fe and lower P groundwater) (**Table 1**), a previous study estimated that a molar ratio of > ~ 54 [Fe]/[As] (or ~ 40 on a mass basis) was required to achieve reduction of As to < 50 $\mu g.L^{-1}$ in the high As Bangladeshi groundwaters, whereas a molar ratio of < 16 [Fe]/[As] was sufficient to achieve ~ 100 % removal of As in the New Hampshire groundwater using a household co-precipitation and filtration system [80]. The high [Fe]/[As] ratios required for As removal in Bangladesh [80] were attributed to the high concentrations of PO_4 and silicates, leading to competition for sorption sites and thus the requirement for higher levels of Fe to support As removal. Given that the feedwater composition of the Bihar set presented here falls generally between these two exemplar groundwaters (with closer similarity to the Bangladeshi water), it might be reasonable to expect that [Fe]/[As] molar ratio broadly between the range of ~ 16 to 54 might plausibly be a similar range for effective As removal in Bihar (noting this would be dependent on local geochemical conditions and remediation targets). Importantly, there is likely to be continuum of [Fe]/[As] ratios, and values reported elsewhere should not necessarily be considered to be a target range in Bihar; rather this provides general comparison and site-specific investigation would be necessary to confirm what [Fe]/[As] ratios were sufficient to meet As removal targets in local conditions. Although there was not an overall trend observed of higher As removal with increasing [Fe]/[As] ($p = 0.40$, **Figure 5F**) nor [Fe]/[P] ($p = 0.56$, **Figure 5G**), it can be observed that many of the systems with high levels of removal indeed corresponded to source water with [Fe]/[As] within, or exceeding, the range of ~ 16 – 54 (**Figure 5F**). Particularly within MSF systems, higher levels of As removal appeared to be associated with higher [Fe]/[P] (**Figure 5G**) although this is not clear across the whole dataset.

Interestingly, however, there was a clear and statistically significant relationship between As removal and source water ([Fe] -1.8[P])/[As] (*t value = 3.8, degrees of freedom = 19,* $p < 0.01$; **Figure 5H**), with the highest removals systematically associated with higher values of this coupled Fe-P-As relationship. This can be explained theoretically because PO_4 strongly binds to Fe-(oxy)hydroxides, resulting in minimal As removal if free PO_4 is present [71]. Thus, only the Fe remaining after the removal of PO_4 is available for As removal [71]. A molar ratio of ~ 1.5 to 2 [Fe]/[P] is required for PO_4 removal at neutral pH, so Fe remains available for As removal only in excess of this [Fe]/[P] ratio [71]. The molar ratio of ~ 1.8 [Fe]/[P] is used here on the simple basis of a molecular weight conversion for the optimal removal of PO_4^{3-} by precipitation with $FeCl_3$, although the chemical reactions between Fe and P in aqueous solutions are complex, dependent on source water chemistry and can involve numerous complexes and precipitates [95]. Other studies have also reported that [Fe]/[P] ratios ~ 2 were necessary for sufficient P removal using membrane bioreactor systems, whilst noting negative impacts such as severe membrane fouling have been reported at higher [Fe]/[P] ratios ~ 4 [96]. The consistency of observed trends in As removal with this theoretical Fe-P-As relationship is noteworthy and suggests that this was a key control (both theoretically and practically) on the performance of As remediation systems in Bihar. Whilst increased ([Fe] -1.8[P])/[As] was associated with higher As removal, the value of this ratio in most cases is still negative, suggesting that the presence of P may inhibit effective As removal. This means that theoretically As removal may be further enhanced with the addition of additional Fe in cases where this might be necessary. Notably, the source water used for the homemade bucket filter had both [Fe]/[As] > 54 and a positive value of ([Fe] -1.8[P])/[As], indicating the natural water conditions in this case were geochemically favorable for As removal, achieved even with a very simple technological intervention. Thus, source water composition, and particularly ([Fe] -1.8[P])/[As], was a key control on the relatively high levels of As removal observed, regardless of technology type implemented. Although beyond the scope of this current study, potential long term operational concerns (*e.g.* membrane fouling at high [Fe]/[P] ratios [96]) should be systematically considered and could impact optimal technology selection.

Whilst the trends in As removal were broadly consistent with what might be expected theoretically from source water chemistry, and particularly ([Fe] – 1.8[P])/[As] (consistent with molar ratios of Fe required for PO_4 removal [71]), there remained high variability in system performance. This suggests that whilst the source water composition was a very important control on remediation performance, the geochemical basis alone was not necessarily sufficient to predict overall effectiveness of all system types in practice, which was likely attributed to numerous other factors impacting system performance in real-world implementations. Importantly, as well, the geochemical controls discussed here are specific to As; geochemical controls impacting the removal of other target contaminants for remediation may be different and parameter-specific.

3.4.3 Multivariate Analysis of Arsenic Removal

Multivariate analysis to explain As removal as the independent variable with all available parameters (([Fe] -1.8[P])/[As], Fe, As, P, Ca, Mg, Na, Si, [Fe]/[As], [Fe]/[P], technology type and setting) suggests that ([Fe] -1.8[P])/[As] is the only significant predictor variable ($p < 0.05$) in the best fit model. The best fit model predicts that a one unit increase in ([Fe] -1.8[P])/[As] leads to an increase in As removal of 0.24 % (confidence interval 0.03 % to 0.46 %). Although none of the other explanatory variables individually are significant in the best fit model, their cumulative impact is important as a model based on ([Fe] -1.8[P])/[As] alone becomes insignificant, likely influenced by the complexity of the systems and relatively low sample numbers. In the case that only the subset of data for RO-HH systems is considered, the ([Fe] -1.8[P])/[As] ratio is significant both with and without the inclusion of Si, indicating that ([Fe] -1.8[P])/[As] is still the most important control within a comparison of relatively similar system types.

3.5 Other Factors Impacting Performance

As has been well-documented in the literature, the importance of social, socio-economic, behavioral and regulatory factors [60, 61, 97, 98] can heavily influence remediation uptake and usage in South and Southeast Asian settings including in Bihar [74, 75], Vietnam [57] and Bangladesh [99, 100]. Our site visits revealed substantial variability in how the remediation systems were maintained and managed even at individual household level, which is very difficult to account for in a survey intended to evaluate spot checks of system performance under normal operating conditions. Our site visits also revealed a very wide range of attitudes towards water quality and remediation, and complex social interactions between and within households with influence on water-related behaviours at a local level.

Although the purpose of this study was not to systematically evaluate socio-economic and/or managerial factors, we acknowledge that ultimately these factors may be as important, or in some cases perhaps even more important, than technical and geochemical aspects in terms of overall sustainability. Indeed, the observation that the data presented in this study cannot be fully explained by geochemical aspects alone suggests that there are confounding factors which can and do influence remediation performance. Analysis of stakeholder networks and local attitudes towards water quality and water remediation in Bihar is the subject of ongoing work by co-authors.

From the perspective of cost, and for additional context in the case of Bihar, untreated groundwater is usually provided free of cost through handpumps (either private or governmental; noting private handpumps would incur installation costs); whereas it was locally reported that most packaged and treated water was sold for ~ 15 to 20 INR (~ 0.20 to 0.30 USD), for a ~ 15 L large plastic bottle, and household-scale RO units were available at local shops at prices that were observed to range from ~ 8,000 to 25,000 INR (~ 110 to 340 USD). Although comprehensive economic information was not available for all systems studied, the relative magnitude of costs indicates that affordability and willingness-to-pay is likely an important practical factor on remediation selection on the individual level from the end-user perspective. In the case that untreated groundwater is not of adequate quality in a particular location, the most affordable option may be identifying suitable alternative supplies (including an alternative source of treated water). However, in all cases, site-specific testing and/or information should be evaluated to inform optimal local remediation recommendations.

3.6 Implications for Remediation Selection

Given the evidence showing that source water chemistry, and specifically ([Fe] – 1.8[P])/[As], was strongly related to the performance of remediation systems for As removal in Bihar, this Fe-P-As relationship could be used as a decision-support input to help inform remediation selection. Using data from groundwater characterization (*e.g.* Fe, P and As concentrations), the Fe-P-As relationship could be calculated to theoretically determine, for example: (i) areas where natural groundwater conditions are more or less likely to be geochemically compatible for As removal; and/or (ii) areas where additional Fe inputs might help improve remediation performance. Importantly, however, this would not account for all factors which can impact remediation performance (*e.g.* complex groundwater matrices, technical properties of remediation systems, operation/maintenance, socio-economic factors), nor would it necessarily be applicable to other solutes which may also be targeted for remediation. Notwithstanding these limitations, however, ([Fe] – 1.8[P])/[As] is valuable as a key theoretical input to assist selection of appropriate As remediation approaches.

The molar ratio of ([Fe] – 1.8[P])/[As] has been calculated for groundwater across Bihar (**Figure 6**) based on geochemical data published elsewhere [17]. This indicates that values of ([Fe] – 1.8[P])/[As] are generally lowest in areas south of the Ganges (Ganga) River, and thus natural geochemical conditions in these areas are likely to be less favorable for As removal by the technologies studied here. In these cases, in particular, additional Fe might help to facilitate As remediation. In areas to the north of the Ganga, and particularly between the Gandak and Koshi Tributaries, natural conditions are generally more favorable for As removal. There are several groupings of points, for example near the lower stretches of the Gandak and in the eastern part of Bihar where the ([Fe] – 1.8[P])/[As] ratios are very high and thus highly favorable for As removal. Whilst this spatial distribution is useful in identifying broad spatial patterns (with implications on the selection of appropriate remediation approaches), importantly, these patterns are heterogeneous and would need to be verified on a site-specific basis.

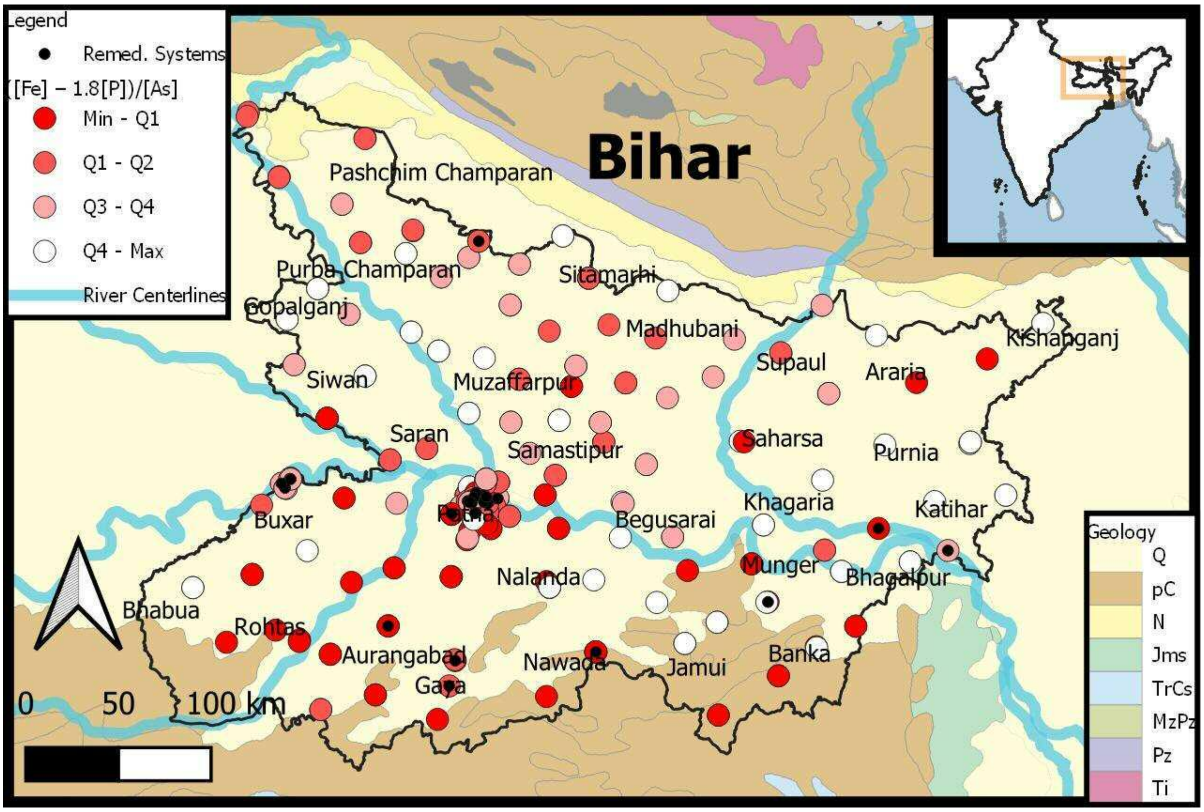

Figure 6. Map of Bihar with values of calculated values of the molar ratio of ([Fe] – 1.8[P])/[As]/100 based on geochemical data from representative sampling across Bihar published elsewhere [17] and the sites where remediation systems were sampled (this study). Colored dots represent calculated values of ([Fe] – 1.8[P])/[As]/100 for Min - Q1 (~ -180 to -3.7), Q1 - Q2 (~ -3.7 to 0.0); Q2 - Q3 (0.0 to 1.9); and Q3 - Q4 (1.9 to 580). Higher values of this ratio are associated with conditions that are more geochemically favorable for As removal. In general the lower values and quartiles suggest that lower efficiency of As removal may be encountered in these areas and that the addition of Fe may make conditions more geochemically favorable for higher As removal efficiency; in contrast higher efficiency of As removal might be expected in areas with higher values and quartiles of ([Fe] – 1.8[P])/[As]. Importantly site-specific verification of local conditions is necessary and highly recommended. Underpinning geology [101] with Q = Quaternary; pC = Precambrian; N = Neogene; Jms = Jurassic metamorphic & sedimentary; TrCs = Lower Triassic to Upper Carboniferous; MzPz = Mesozoic and Paleozoic; Pz =Paleozoic; Ti = Tertiary igneous dark grey = other. Boundaries and exaggerated river centrelines are from Natural Earth (https://www.naturalearthdata.com/).

A similar approach could be considered for other As-impacted areas particularly where there is the availability of spatially distributed data of groundwater As, Fe and P. This theoretical input is not necessarily indicative that As remediation technologies will perform well, but simply a tool to identify areas where the geochemical conditions are generally favorable for As removal. Ultimately, to gain a well-rounded perspective on the likelihood of remediation success in a particular area, this geochemical indicator would need to be considered holistically alongside other selection factors including remediation priorities and targets, technology availability, technical considerations, operation, maintenance and management, socio-economic conditions and cost effectiveness [57, 74].

4. **Conclusions**

We have evaluated the performance of household and community scale systems for groundwater remediation (and particularly As removal) in Bihar, India via spot-assessments under typical operating conditions. A wide range of remediation systems were identified ranging from simple homemade bucket sand filters to multi-stage commercial filtration systems to RO-based systems at various scales. The performance of remediation systems under these conditions varies widely, with removal of As and other inorganic contaminants ranging from ~ 0 to 100 %. The removal of As varied widely, with high removal achieved (usually to below the WHO provisional guideline value whilst noting that most inlet water was also below 10 $\mu g.L^{-1}$) using a variety of technologies. Generally, removals of Mn, Fe, U, P, Mg, As and Ca were relatively high (*e.g.* median value > 80 %) whereas more challenging solutes included K, Na, Si, NO_3^-, F^-, Cl^-, Zn and SO_4 (*e.g.* median values < 80 %). A comparison between RO and non-RO based technologies indicated that the remediation technology itself was likely not the dominant control on the removal of many inorganic solutes including As, Fe, P, Zn, Mn, Ca, NO_3^-, F^- and SO_4, for appropriately selected technologies and the groundwater conditions observed in this study Importantly in the context of As, this shows that even relatively simple technologies can achieve high levels of As removal in certain circumstances such as the conditions studied. RO systems, however, did achieve generally higher degrees of removal for some solutes (U, Mg, K, Na, Cl^- and Si) indicating that technology type was a more important factor for these particular solutes. The type of implementation setting (*e.g.* household or non-household) did not appear to significantly impact performance in most cases. Source water composition, and particularly the ratio of ([Fe] -1.8[P])/[As], was a significant control on As removal, with a statistically significant relationship between higher levels of As removal in groundwaters with higher values of ([Fe] -1.8[P])/[As], consistent with theoretical As-Fe-P interactions. The ratio of ([Fe] -1.8[P])/[As] thus provides an important theoretical input relevant for remediation selection, as it provides information regarding the degree to which groundwater composition may be geochemically compatible with high levels of As removal (regardless of type of remediation technology). As illustrated for Bihar, this could be used to identify spatial patterns in geochemical compatibility and to identify where, for example additional Fe may be needed to facilitate higher levels of As removal. This geochemical approach could be used as a decision-support tool, alongside other important considerations (*e.g.* technical, operation/maintenance, managerial and socio-economic), to help inform optimal selection of groundwater remediation approaches in Bihar and more widely.

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Author Contributions

LAR: formal analysis; investigation; data curation; visualization; supervision; project administration; methodology; funding acquisition; writing – original draft. **NK:** investigation; methodology; writing - review & editing. **RK:** investigation; writing - review & editing. **AK:** investigation; writing - review & editing. **AG:** conceptualization, funding acquisition; writing - review & editing. **PM**: conceptualization, methodology; formal analysis; funding acquisition; writing - review & editing. **DAP:** conceptualization; methodology: resources; supervision; project administration; funding acquisition; writing - review & editing.

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