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# Large-Scale Uranium Contamination of Groundwater Resources in India

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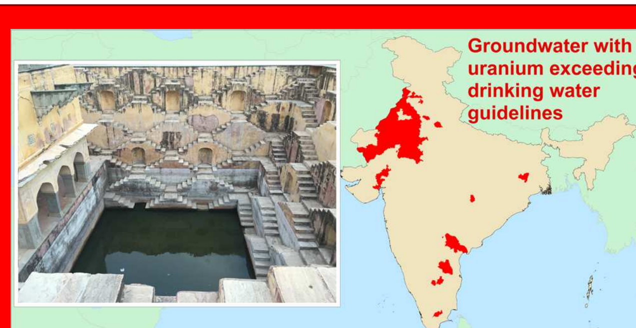
*Supporting Information*




Groundwater overexploitation has caused massive groundwater depletion and raised concerns for water and food security in India. Groundwater in India also suffers from multiple water quality issues such as arsenic and fluoride contamination that pose human health risks. Here we report new data showing that the occurrence in uranium in Indian groundwater is an emerging and widespread phenomenon. We present compiled data on groundwater uranium from 16 Indian states and new data from 324 wells in the states of Rajasthan and Gujarat that show a high prevalence of uranium concentrations above the World Health Organization provisional guideline value of 30 μg/L across India. Using geochemical and uranium isotope data, we suggest factors that may drive high uranium concentrations in groundwater, including uranium content in aquifer rocks, oxidation state, and groundwater chemistry that promotes the formation of soluble uranyl carbonate complexes. While the primary source of uranium is geogenic, anthropogenic factors such as groundwater table decline and nitrate pollution may further enhance uranium mobilization. These findings suggest the need for revision of the current water quality monitoring program in India, evaluation of human health risks in areas of high uranium prevalence, development of adequate remediation technologies, and, above all, implementation of preventive management practices to address this problem.

## INTRODUCTION

India, the world's second most populous country, extracts more than a third of worldwide groundwater resources, more than 90% of which is used for irrigation.[1] Intense abstraction has led to severe groundwater table declines in many parts of the country, especially in the northwestern Indian states of Punjab, Haryana, and Rajasthan.[2−5] In 2013, the Indian Central Groundwater Board estimated that groundwater in the majority (66−70%) of blocks (Indian administrative division above village) in these three states was either critically exploited or overexploited.[5] At the same time, parts of northwestern India that import surface water through canals are dealing with water logging issues, even in arid, previously groundwater-deficient areas.[2,4−6] Overexploitation of groundwater and the use of imported surface water, combined with reported changes in precipitation patterns induced by climate change, have raised concerns about future water sustainability in India,[2−4] yet water quality issues are perhaps even more pressing. High concentrations of salinity, fluoride, and nitrate are widespread in groundwater resources throughout the country.[6−8] Groundwater arsenic problems have been reported in the delta aquifers of West Bengal and Bangladesh, as well as along the Indo-Gangetic Basin aquifer in Pakistan.[2,9] There are also reports of high levels of uranium in groundwater, particularly in northwestern India, which is the focus of this study.

Uranium's threat to human health comes from its chemical rather than it's radiological properties. Epidemiological and toxicological studies have examined the link between the prevalence of uranium in water and chronic kidney disease (CKD) and demonstrated that exposure to uranium through drinking water is associated with nephrotoxic effects.[10−12]

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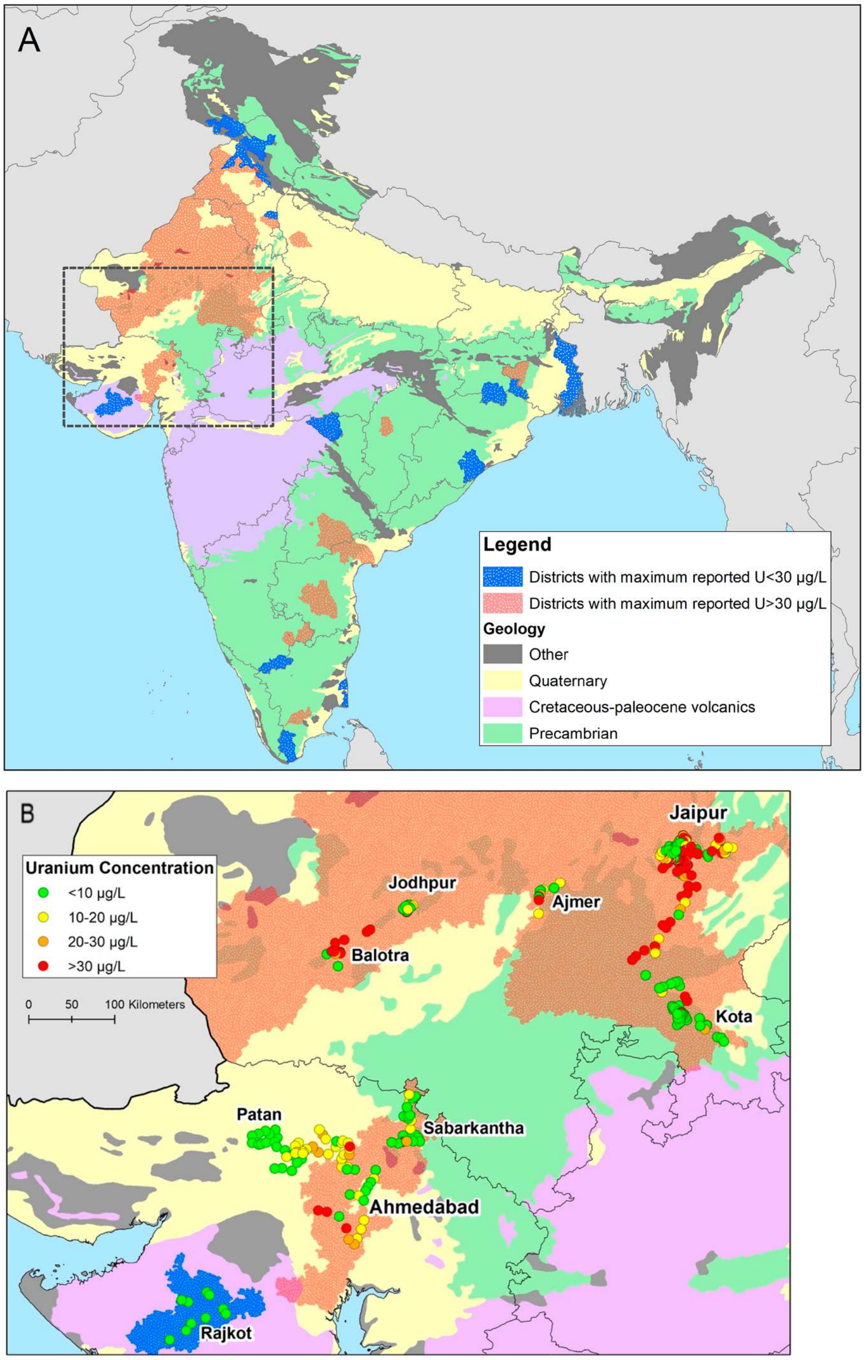


**Figure 1.** (A) Distribution of major geological formations in India that compose local aquifers, combined with identified districts in India where uranium in groundwater has been reported to exceed (red zone) or not to exceed (blue zone) the World Health Organization provisional drinking water guideline value of 30 $\mu$g/L. (B) Distribution of uranium concentrations in groundwater collected in this study, together with the major geological formations and identified districts in Rajasthan and Gujarat, where uranium content in groundwater has been reported to exceed (red zone) or not to exceed (blue zone) 30 $\mu$g/L.

Consequently, the World Health Organization (WHO) has set a provisional guideline value of 30 $\mu$g/L for uranium concentrations in drinking water,[13] which is consistent with the U.S. Environmental Protection Agency[14] drinking water standards. Despite this, uranium is not included in the Bureau of Indian Standards' Drinking Water Specification, though the Atomic Energy Regulatory Board has set a radiologically based limit of 60 $\mu$g/L for uranium in drinking water.[15] Uranium also bioaccumulates in crops irrigated with water containing uranium, though the magnitude of enrichment depends on multiple factors, such as soil and crop type.[16]

Elevated concentrations of uranium have been observed in groundwater worldwide from both anthropogenic (e.g., uranium mining and nuclear disposal sites) and geogenic sources (e.g., high-uranium source rocks, like granites).[17−20] In addition to the uranium sources, other factors such as oxidation state, water−rock interactions, and the formation of soluble aqueous complexes are important factors controlling uranium


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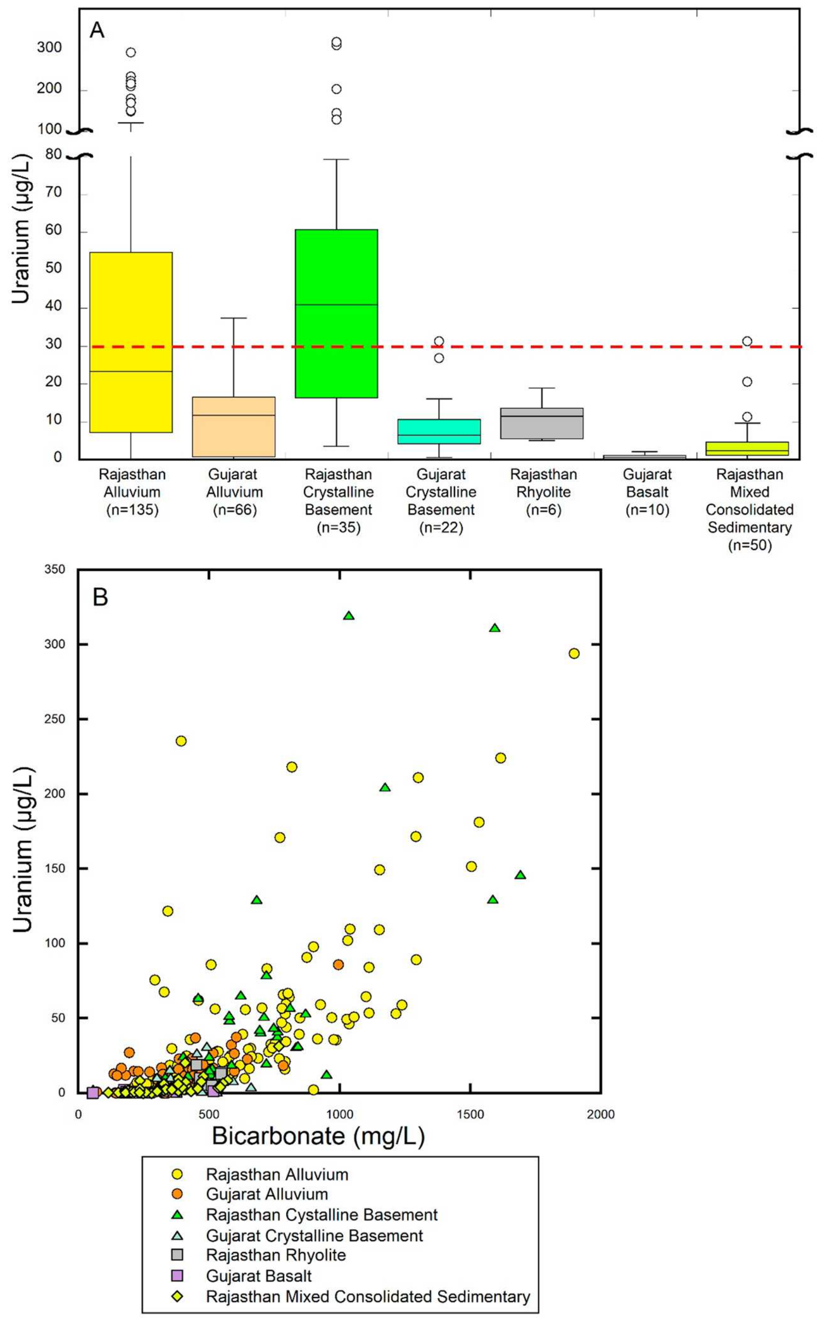


**Figure 2.** (A) Box plots of uranium concentrations of groundwater from different aquifers in Rajasthan and Gujarat investigated in this study. Red lines represent the WHO's provisional guideline values for uranium in drinking water. For statistical analysis of the differences in U distribution by aquifer, see Table S4. (B) Uranium vs bicarbonate concentrations in groundwater sorted by aquifer lithology. See Table S6 for Spearman correlations sorted by aquifer.

occurrence, speciation, and mobility in groundwater. Though uranium has two primary oxidation states, +4 and +6, it is most soluble as uranyl ion ($UO_2^{2+}$) in the oxidized +6 form. Uranium may be mobilized from aquifer rocks through uranium-bearing mineral dissolution and adsorption/desorption processes with clay minerals, iron oxides, and organic matter.[20−24] Water chemistry also plays an important role, as uranium forms stable complexes with several different ligands.[25] Uranyl carbonate complexes, including ternary species, are particularly important as these highly stable and mobile complexes promote desorption of uranium from the solid phases.[20,26,27]

While previous studies of uranium in Indian groundwater have focused on relatively small geographic areas and regions, here we present, for the first time, evidence that uranium contamination in groundwater is widespread in India and occurs at different magnitudes in many of India's aquifers. Our analysis is based on our sampling and chemical analysis of well water in Rajasthan ($n$ = 226) and Gujarat ($n$ = 98), combined with compiled data from previous studies to show the wide extent of uranium in India's groundwater (Figure 1 and Table S1). The new dataset represents a variety of hydrological settings, aquifer types, and land uses. With these tools, we show the known and likely uranium distribution in Indian aquifers



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and suggest mechanisms that may control the occurrence of uranium in these groundwater settings.

## ■ MATERIALS AND METHODS

We examined the inorganic water chemistry of 324 wells in the states of Rajasthan and Gujarat (Table S2). Water samples from wells were collected prior to any treatment systems and were immediately filtered and preserved in high-density polyethylene, airtight bottles following U.S. Geological Survey (USGS) protocols.[28] Temperature, pH, and specific conductivity were measured in the field. Samples were filtered in the field through 0.45 $\mu$m filters for dissolved anions. Trace metal and cation samples were similarly filtered and preserved with Optima nitric acid to pH <2. Samples were kept in coolers and shipped to Duke University for analysis. Major anions were measured by ion chromatography using Dionex IC DX-2100, and major cations were measured by direct current plasma optical emission spectrometry. Bicarbonate was determined via titration to pH 4.5 in duplicate. Nitrate was analyzed via QuickChem Method 10-107-04-2-D (Nitrate/Nitrite in Waters by Hydrazine Reduction). Trace elements, including uranium, were analyzed by a VG PlasmaQuad-3 inductively coupled plasma mass spectrometer (ICP-MS) calibrated to the NIST 1643e standard.

A subset of groundwater samples was analyzed for $^{234}U/^{238}U$ activity ratios at the University of Texas at El Paso U-series Isotope Analytical Laboratory. Thirty grams of each water sample was evaporated in a Class-100 clean room for measuring $^{234}U/^{238}U$ ratios. Samples were then processed using conventional cation exchange chromatography. Uranium isotopic ratios were measured on a Nu-plasma MC-ICP-MS. Measured $^{234}U/^{238}U$ isotope ratios were converted to $(^{234}U/^{238}U)$ activity ratios using the decay constants of $^{234}U$ and $^{238}U$.[29,30]

Complied data on uranium concentrations in groundwater in India were generated using search engines to identify peer-reviewed or other published reports. All statistical analyses were performed using R version 3.3.1 (The R Foundation) using nonparametric methods that did not assume normally distributed data (Spearman's rank correlation and Wilcoxon rank sum test). Uranium speciation indices were calculated using the USGS PHREEQC software and Lawrence Livermore National Laboratory Thermodynamic Database with formation constants for ternary complexes of uranyl carbonate with Ca, Mg, and Sr added manually, and assuming a pE value of 4.[26]

## ■ RESULTS AND DISCUSSION

**Uranium Distribution in Groundwater Resources.** We find that levels of uranium in 75 of the 226 wells tested in Rajasthan and five of the 98 wells tested in Gujarat exceeded the WHO provisional health guideline (Figure 1). Integration of our new data and results from previous studies shows a wide distribution of areas throughout India in which groundwater with a level of uranium exceeding 30 $\mu$g/L occurs (Figure 1 and Table S1). Our list of previous studies includes 68 studies covering 16 states and 61 districts. Of these, nine districts with >30 $\mu$g/L U are located in southern and southeastern parts of India, which are primarily composed of aquifers associated with crystalline basement rocks (Archean granites and early Proterozoic metamorphics)[6] (Table S1). The other 26 districts reporting >30 $\mu$g/L U are located in northwestern India (Figure. 1) and are associated primarily with alluvial aquifers, which were formed from the erosion of Himalayan rocks[31] (Table S1).

Findings from these reports are consistent with our new results (Figure 2 and Table S2), which find instances of >30 $\mu$g/L U almost exclusively in groundwater from alluvial aquifers and aquifers associated with crystalline basement rocks (metamorphic rocks in Rajasthan sites and mixed metamorphic and granitic rocks in Gujarat sites) (Figures 1 and 2A). Other aquifers sampled during this campaign, including Paleozoic sandstone and limestone aquifers (termed consolidated sedimentary aquifers), and Deccan basalt aquifers, had significantly lower uranium concentrations (Figure 2A; see statistical analysis in Table S4).

Though the alluvial aquifers from our study sites in Rajasthan and Gujarat are composed of similar rock materials, there are key differences between their hydrogeological settings. The alluvial aquifers of Rajasthan are primarily unconfined, similar to other alluvial aquifers in Northwest India, such as Punjab.[31] In contrast, the alluvial aquifers of Gujarat often have clay confining to semiconfining layers, which retards direct recharge to deeper parts of the aquifer in some areas. We were able to obtain specific depth information for wells from Gujarat, which was unavailable for wells in Rajasthan. Well depth in Rajasthan may be qualitatively inferred from the types of wells. The Rajasthan dataset contains 87 hand pumps, which are limited to ∼90 m in depth, and 13 dug wells, which are similarly shallow. Thus, we conclude that most of the investigated wells from Gujarat are deeper than those from Rajasthan. Both of these factors likely contribute to the concentrations of uranium in the Gujarat alluvium groundwater samples being lower than those in the Rajasthan data set (Figures 1 and 2A), because of redox controls on U solubility. A large number of studies have reported elevated levels of uranium in groundwater in Punjab (Table S1), where the alluvium aquifers are similarly unconfined.[31]

**Controls on the Occurrence of Uranium.** Bicarbonate complexation and oxidizing conditions are two of the most important chemical factors controlling uranium concentrations in groundwater[20,25,26,32−34] and appear to be the key factors for the alluvial aquifers in northwestern India. The accumulation of bicarbonate in groundwater enhances the formation of highly soluble uranyl carbonate complexes, which results in elevated uranium concentrations in groundwater. This process is evidenced by the correlation between bicarbonate and uranium in groundwater from most of the aquifers in Rajasthan and Gujarat (Figure 2B and Table S6). This is consistent with speciation modeling conducted with PHREEQC, which predicted that uranyl carbonate species, especially ternary complexes with Ca, are the predominant uranium complexes in groundwater from the alluvium aquifers (Table S7).

Additionally, previous studies have observed massive groundwater table declines in many areas in the unconfined alluvial aquifers of northwestern India.[5] This hydrogeological condition likely promotes oxic conditions, which favor the occurrence of uranium as a soluble complex and migration into deeper parts of the aquifer. Although neither oxidation−reduction potential nor dissolved oxygen concentration was directly measured, relatively low concentrations of both iron and manganese and high nitrate concentrations further support our hypothesis of oxidizing conditions in the shallow U-rich groundwater (Table S2). We observe a statistically significant decrease in U concentration with depth in the Gujarat alluvial aquifers.


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Some of the high U values at depth may be explained by recharge from shallower parts of the aquifer (Figure S3).

In the crystalline basement aquifers, high-uranium source rock may play a role in uranium concentrations as previous reports have shown high uranium in a variety of crystalline basement formations throughout India.[35,36] Although the alluvial aquifers are not composed of unusually high uranium material (~3 ppm, comparable with the global crustal average),[37,38] they consist primarily of silicate minerals, which are subject to hydrolysis under the influence of carbonic acid to produce groundwater of the Na−Ca bicarbonate chemical type, thus promoting the formation of soluble uranyl carbonate complexes.

Other studies have linked U mobilization to nitrate. It has been suggested that agricultural activities that may lead to nitrate contamination (e.g., fertilizer application and soil cultivation) also increase $pCO_2$ through increased biological activity and/or that nitrate itself participates in redox reactions, which in turn lead to the biotic or abiotic oxidation of U(IV) to U(VI).[19,37−40] Nitrate concentrations were high in our studied groundwater, with 42% of tested wells being above the WHO drinking water guideline of 45 mg/L for nitrate[13] (Figure S1). Nonetheless, correlations between uranium and nitrate were either weak or not significant (Table S5).

**Uranium Isotope Variations.** The uranium isotope data suggest multiple end members and evolution pathways for U. Most of the studied groundwater has a $^{234}U/^{238}U$ activity ratio (AR) of >1.0 (Figure 3 and Table S3), which indicates disequilibrium between the dissolved $^{234}U$ and $^{238}U$ nuclides.[39] It has been shown that such disequilibrium is derived from either direct recoil processes, in which $^{234}U$ is displaced to the mineral surface or directly into the ambient groundwater. In addition, preferential release of $^{234}U$ from damaged mineral lattice sites can cause selective enrichment of $^{234}U$ in the aqueous phase over $^{238}U$.[40,41] In the Gujarat alluvium, the data show a decrease in U concentration and an increase in $^{234}U/^{238}U$ AR with depth (Figure S3), which reflects decreasing U solubility in the transition from oxidizing to reducing conditions with depth. Deep groundwater with lower U contents would be more affected by recoil, resulting in a higher $^{234}U/^{238}U$ AR.[42,43] In contrast, we observed groundwater with both high U concentrations and high AR in Rajasthan (Figure 3). It is possible that this is due to oxic leaching, with loosely bound $^{234}U$ on aquifer rocks being preferentially released. This is consistent with a water table drop in the shallow wells of Rajasthan, in which newly oxic conditions would preferentially mobilize $^{234}U$ during the early stages of chemical weathering to produce high AR and high U concentrations.[41] In this case, further weathering would result in bulk U mobilization from the rocks and decrease the AR in associated groundwater. The combined high U concentration and AR could therefore reflect the water table decline observed in Rajasthan's aquifers.

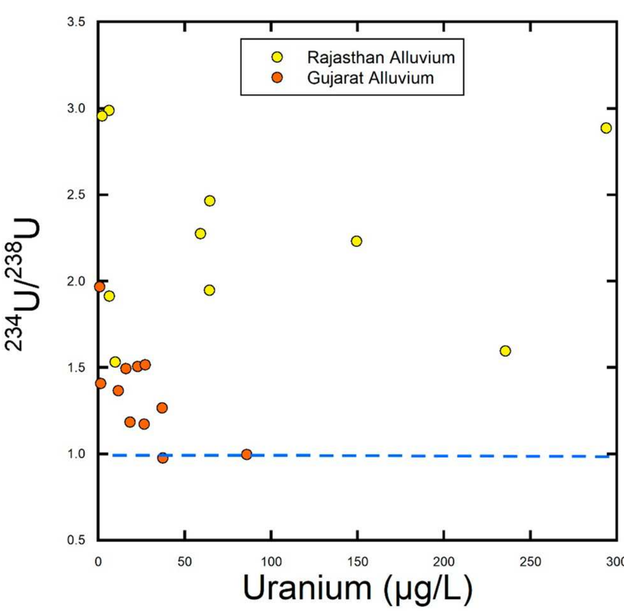


**Figure 3.** $^{234}U/^{238}U$ activity ratios vs uranium concentration in groundwater from the alluvial aquifers in Rajasthan and Gujarat. The blue line represents secular equilibrium in which the $^{234}U/^{238}U$ activity ratio is ~1. $^{234}U/^{238}U$ activity ratios of >1 observed in most of the investigated groundwater indicate selective $^{234}U$ chemical mobilization and/or physical recoil from the aquifer rocks.

**Broader Implications.** Much of the high-uranium groundwater we tested also suffered from other water quality problems, such as high salinity, fluoride, and nitrate, which make them unsuitable for human consumption. Still, a great number of these wells are being used as primary drinking water sources (defined here as sources of water that serve as the exclusive or nearly exclusive source of drinking water for more than one household) because of the lack of alternative water sources. In Rajasthan's alluvium and crystalline basement aquifers, 45 of 121 primary drinking water wells we tested had uranium concentrations that exceeded the EPA and provisional WHO guideline of 30 μg/L for uranium. In Gujarat, where people have greater access to alternative sources of drinking water such as canal water or RO desalted water, only three of 68 tested primary drinking water wells had uranium concentrations exceeding 30 μg/L. Alluvial aquifers and aquifers associated with crystalline basement together serve more people and land area in India than all other aquifer types combined.[6] On the basis of the correlations we observed between uranium and bicarbonate (Figure 2B and Table S6), and the fact that ~50% of wells we sampled in these aquifers with bicarbonate concentrations above ~410 mg/L had >30 μg/L U, we suggest that groundwater reliant areas with bicarbonate concentrations above this threshold should be targeted for future uranium monitoring. Bicarbonate is easy to measure and is already a part of regular groundwater monitoring efforts. Additionally, areas with aquifers composed of known high-uranium rocks should also be targeted for monitoring.

Some studies have reported high rates of CKD in India,[44] and the possible link between uranium in drinking water and CKD in India is worthy of future research. Additional research should investigate the possible role of human activities such as irrigation and fertilizer application on the concentration of dissolved U in Indian aquifers. As groundwater becomes an increasingly important source of water around the world,[1] it is important to understand how human activities can affect and limit its suitability for future use. We are hopeful the findings reported in this study will lead to measures such as including a health-based uranium standard in the Bureau of Indian Standards Drinking Water Specification, establishing adequate and systematic monitoring systems for identifying at-risk areas for uranium exposure, and exploring prevention and treatment options to mitigate the large-scale uranium problem in India.


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## ■ ASSOCIATED CONTENT

### Supporting Information

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Seven tables and three figures (PDF)

## ■ AUTHOR INFORMATION


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### Notes

The authors declare no competing financial interest.

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