



# Ranking traditional and nano-enabled sorbents for simultaneous removal of arsenic and chromium from simulated groundwater



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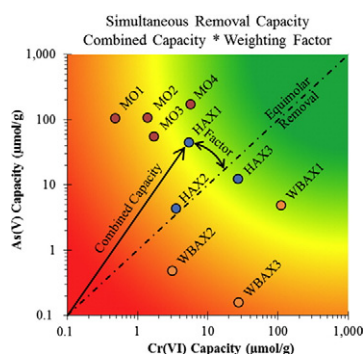
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## HIGHLIGHTS

- We search for sorbents that simultaneously remove arsenic and chromium from water.
- A new quantitative ranking index considers absolute and relative removal capacity.
- It applies to simultaneous removal of any set of pollutants in water or air.
- Metal (hydr)oxide nanoparticles are formed inside weak base anion exchange resins.
- Nano-enabled sorbents rank more highly than metal oxides and anion exchangers.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Water from many municipal and private wells are treated to meet arsenic (As(V)) regulations, and new regulations for hexavalent chromium (Cr(VI)) are soon possible. Rather than adding costly capital infrastructure, we explored many types of sorbents' ability to remove both oxygenated anions simultaneously. In laboratory pseudo-equilibrium tests, metal (hydr)oxide sorbents demonstrated high affinity for As(V) but exhibited 30 to 100-fold lower capacities to remove Cr(VI). WBAX resins had some ability to sorb both Cr(VI) and As(V), but competing anions lowered their sorption capacity for As(V) by >90% compared to in a deionized water matrix. Nano-enabled sorbents with iron or titanium nanoparticles embedded inside the porous structure of an anion exchange resin demonstrated high ability to remove both pollutants simultaneously despite competition, with the tested sorbents showing 11 µmol/g capacity for Cr(VI) and 19 µmol/g for As(V) in simulated groundwater on average. To quantitatively rank sorbents' ability to remove multiple pollutants, a Simultaneous Removal Capacity scoring tool is proposed. This index uses both absolute removal capacity for each contaminant and relative capacity for multiple contaminants, and may be applied to simultaneous removal of any set of aqueous or atmospheric contaminants. Here, the five nano-enabled sorbents ranked in the top seven out of twelve sorbents tested for simultaneous Cr(VI) and As(V) removal. This work demonstrated traditional and novel nano-enabled sorbents can reduce multiple contaminants of health concern, resulting in groundwater treated to drinking water standards.

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## 1. Introduction

For centuries, groundwater has served as the primary source for supplying potable water to the populous of many urbanized and non-urbanized communities across the world. Much of this water frequently receives no additional treatment beyond disinfection before it is distributed to consumers. Even in the United States, where the groundwater serves as the primary source for 95% of drinking water systems, <40% of the groundwater is treated to remove organic and inorganic contaminants before distribution (Impellitteri et al., 2007). With identification of new risks stemming from common groundwater contaminants, however, this problem has been gaining conspicuity. Research conducted over the last few decades has demonstrated that many commonly occurring groundwater contaminants, such as arsenic, chromium, fluoride, and nitrate, have the potential to cause serious health issues among the populous who is chronically exposed at low concentrations for a long period of time (USEPA, 1998a, 1998b). Many groundwater resources that were disregarded because of high concentration of inorganic contaminants are being reexamined to determine whether they could be employed to mitigate ever-increasing water demands associated with climate change-induced concerns about water scarcity.

Advancements in nanotechnology applications for water treatment offer a new promise in exploiting these excluded groundwater resources for potable water use. Development of nano-enabled water treatment technologies has the potential of providing potable water to communities at a fraction of today's treatment costs. Benefiting from the unique properties of nanomaterials, new point-of-use or well-head water treatment systems could be developed to employ sorption, disinfection, or photocatalytic redox processes (Qu et al., 2013). The small sizes allow nanomaterials to be incorporated into the porous structure of other materials, which serve as a support or base media, which by itself may exhibit water treatment properties. This process of nano-enabling the sorbent allows development of smaller and more-compact bulk water-treatment platforms, while concurrently maintaining the high specific surface area-to-mass ratios and unique properties of the incorporated nanomaterials. For example, hybrid sorbents that could simultaneously remove or reduce organic and inorganic contaminants have been fabricated by combining nano-sized zero-valent iron and polymers (Du et al., 2013), or incorporating nano-scale metal (hydr)oxides in biochar (Hu et al., 2015), chitosan (Yamani et al., 2012), or activated carbon (Sandoval et al., 2011).

Recent studies have demonstrated the feasibility of using hybrid-ion exchange media for simultaneous removal of arsenic and other strong oxyanions by nano-enabling commercially available strong base ion-exchange resin with titanium or ferric (hydr)oxide nanoparticles (Athanasaki et al., 2015; Elton et al., 2013; Hristovski et al., 2008). Nitrate and perchlorate are suspected carcinogens that are frequently found in groundwater because of anthropogenic contamination. As strong acid oxyanions, they do not have an affinity to sorb to metal (hydr)oxide and are typically removed from water via a strong-base anion-exchange (SBAX) mechanism (Elton et al., 2013). Arsenic, a known human carcinogen in drinking water, is commonly found in groundwater as pentavalent arsenic (As(V)) (USEPA, 2010b). It is a weak acid oxyanion that can be effectively removed with metal (hydr)oxides because it forms inner-sphere complexes (Mohan and Pittman, 2007; Speitel et al., 2010; Westerhoff et al., 2005). In contrast, hexavalent chromium (Cr(VI)), which is also a weak acid oxyanion, does not form the same inner-sphere complexes with metal (hydr)oxides, but necessitates the use of an ion-exchange mechanism to remove it from groundwater. Leading treatment technologies for Cr(VI) employ strong-base anion-exchange, weak-base anion-exchange (WBAX), or reduction to trivalent chromium followed by coagulation and filtration (Brandhuber et al., 2004; McGuire et al., 2007). However, sulfate and other strong acid anions, which are often present in high groundwater concentrations together with Cr(VI), reduce the effectiveness of Cr(VI) removal by SBAX. Furthermore, reduction, coagulation, and filtration are operationally

complicated to employ in a wellhead treatment scenario. This often renders WBAX to be the only viable technology for removal of Cr(VI) from groundwater and necessitates development of new technologies that could simultaneously remove multiple oxyanion contaminants from groundwater matrices. The importance of addressing this challenge is especially amplified in light of the recent promulgation of a new 10 µg/L Cr(VI) maximum contaminant level (MCL) standard by the State of California (California Code of Regulations, 2014).

Led by the State of California's example, the USEPA has initiated a process to review its current MCL of 100 µg/L as total chromium with possibility to lower the MCL or add an MCL specifically for Cr(VI). In 2006, arsenic underwent a similar review process that lowered its federal MCL from 50 µg/L to 10 µg/L (USEPA, 2010b). This regulatory pressure caused a significant economic stress on small utilities, which heavily rely on groundwater as a potable water source and employ well-head treatment systems (McGavisk et al., 2013). To illustrate, about 67% of the non-compliant systems in the United States serve fewer than 500 people (McGavisk et al., 2013), indicating a disproportionate risk to customers served by small systems. Similarly, with 31% of community groundwater sources exhibiting occurrence of Cr(VI) (USEPA, 2015) along with upcoming new regulatory and scientific pressures, it is expected that compliance costs could increase water fees by \$30/yr to \$50/yr per household as exemplified by the California precedence (Coachella Valley Water District, 2014). Consequently, development and testing of simple to operate and effective groundwater treatment technologies that could simultaneously remove Cr(VI) and co-contaminant oxyanions, like As(V), becomes an imperative for small potable water providers, which rely on groundwater and do not have the financial or technical capabilities of large utilities. Sorbent based systems are particularly applicable in context of small, groundwater systems as they are more simple to operate than coagulation based treatment and may not produce high brine volumes that are difficult to dispose of like membrane based treatment.

A small utility's ability to select and implement effective water treatment requires a screening tool for comparing multiple technology efficacies and determining the best performing technology under given conditions. In context of sorption-based technologies, the efficacy is often expressed in terms of equilibrium sorption capacity ( $q$ ), but this absolute quantity only communicates the capacity for one pollutant. In contrast, comparative capacities for multiple pollutants are often expressed by a selectivity coefficient or separation factor ( $\alpha$ ); however, this only communicates relative preference and not absolute capacity. For hybrid media, which have the ability to simultaneously remove multiple contaminants, the existing screening tools are frequently rendered inapplicable because both absolute and relative descriptors are synchronously required to discern the best performing media. Consequently, this study aims to develop a new analysis tool that incorporates both absolute and relative descriptors to quantify sorbents' abilities to treat multiple pollutants, while concurrently reporting the fabrication of novel nano-enabled hybrid weak-based anion-exchange resin for simultaneous removal of Cr(VI) and As(V) from groundwater. To address the goal of this study, (1) a set of eight commercially available sorbents capable of removing As(V) or Cr(VI) were obtained and four hybrid media were synthesized under different conditions using two of the commercial weak base anion-exchange resin as a base media; (2) a new tool for comparison of media's performance was developed; (3) isotherms were developed from batch sorption tests conducted in a realistic groundwater matrix containing As(V) and Cr(VI); and (4) comparison between the commercial and the synthesized media was conducted using the newly developed tool to assess its feasibility for discernment of the best performing hybrid media for simultaneous removal of As(V) and Cr(VI). The goals of this study are particularly applicable to the total environment because the developed ranking tool may be useful for treatment of hydrospheric and atmospheric contaminants, and providing that treatment is the primary method of providing environmental remediation as well as improvement to human health.

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