



Contents lists available at ScienceDirect

## Journal of Environmental Management

journal homepage: [www.elsevier.com/locate/jenvman](http://www.elsevier.com/locate/jenvman)

## Research article

## Impact of wastewater derived dissolved organic carbon on reduction, mobility, and bioavailability of As(V) and Cr(VI) in contaminated soils

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## ARTICLE INFO

## Article history:

Received 19 February 2016

Received in revised form

4 August 2016

Accepted 8 August 2016

Available online xxx

## Keywords:

Arsenate

Chromate

Dissolved organic carbon

Reduction

Adsorption

Bioavailability

## ABSTRACT

In this work, the effects of various wastewater sources (storm water, sewage effluent, piggery effluent, and dairy effluent) on the reduction, and subsequent mobility and bioavailability of arsenate [As(V)] and chromate [Cr(VI)] were compared using both spiked and field contaminated soils. Wastewater addition to soil can increase the supply of carbon, nutrients, and stimulation of microorganisms which are considered to be important factors enhancing the reduction of metal(loid)s including As and Cr. The wastewater-induced mobility and bioavailability of As(V) and Cr(VI) were examined using leaching, earthworm, and soil microbial activity tests. The rate of reduction of As(V) was much less than that of Cr(VI) both in the presence and absence of wastewater addition. Wastewater addition increased the reduction of both As(V) and Cr(VI) compared to the control (Milli-Q water) and the effect was more pronounced in the case of Cr(VI). The leaching experiment indicated that Cr(VI) was more mobile than As(V). Wastewater addition increased the mobility and bioavailability of As(V), but had an opposite effect on Cr(VI). The difference in the mobility and bioavailability of Cr(VI) and As(V) between wastewater sources can be attributed to the difference in their dissolved organic carbon (DOC) content. The DOC provides carbon as an electron donor for the reduction of As(V) and Cr(VI) and also serves as a complexing agent thereby impacting their mobility and bioavailability. The DOC-induced reduction increased both the mobility and bioavailability of As, but it caused an opposite effect in the case of Cr.

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## 1. Introduction

Increasing incidents of human and ecosystem toxicity resulting from the accumulation of heavy metal(loid)s in soil and water have been reported in many countries. Mining and industrial activities release heavy metal(loid)s in the environment, resulting in the buildup of toxic elements including arsenic (As) and chromium (Cr). They reach both terrestrial and aquatic environments thereby entering the food chain. For example, As reaches food chain through the use of As-rich potable water for cooking and irrigation of edible crops such as rice (*Oryza sativa* L.) and vegetables

(Bhattacharya et al., 2012). Garelick et al. (2008) listed important sources of anthropogenic As sources on human exposure such as coal burning, pesticide production and usage, and wood preservation. While As is largely from geogenic sources, Cr is predominantly due to waste disposal from industries such as electroplating, pulp production, petroleum refining, leather tanning [trivalent – Cr(III)], and timber treatment [hexavalent – Cr(VI) – chromate] (Zhitkovich, 2011; El-Sherif et al., 2013).

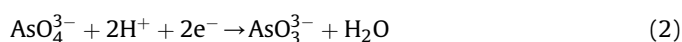
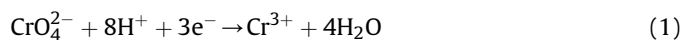
In soil, As is subject to chemical and biological transformations resulting in the formation of various inorganic and organic species (Mahimairaja et al., 2005). The main inorganic forms of As are arsenite [As(III)] and arsenate [As(V)] with the former being more mobile and toxic compared to Cr(VI) (Smith et al., 1998; Bolan et al., 2015). Several researchers studied the toxicity profiles of these two species and related them to their adsorption behaviour in soils

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(Lafferty and Loeppert, 2005; Ohe et al., 2005; Bolan et al., 2015). Arsenate is found in low concentration in soil due to their strong adsorption capacity compared to As(III) (Smith et al., 1998). In the case of Cr, Cr(III) is strongly adsorbed in soil compared to Cr(VI), thereby the former being less toxic (Kosolapov et al., 2004; Choppala et al., 2013, 2016). Hence, the reduction of As(V) and Cr(VI) to their trivalent forms can result in contrasting toxicity effects. Therefore, increasing efforts have been made to control the oxidation and reduction reactions of these two metal(loid)s, thereby reducing their bioavailability and ecotoxicity (Gadd, 2010; Bolan et al., 2013a).

Wastewater irrigation can act both as a sink and source for heavy metal(loid)s including As and Cr (Kunhikrishnan et al., 2012; Ungureanu et al., 2015). For example, irrigation of rice crops is a major source of As contamination in many countries including Bangladesh and India (Ahsan and Del Valls, 2011). Wastewater contains dissolved organic carbon (DOC), and the addition of carbon-rich organic amendments such as wastewater, manures, biosolids, and biochars to soils has been shown to act as an electron donor for the reduction of metal(loid)s, such as As(V) and Cr(VI), and non-metals, such as nitrate ( $\text{NO}_3^-$ ) (Kunhikrishnan et al., 2012). The supply of carbon, nutrients, and stimulation of microorganisms are considered to be important factors enhancing the reduction of metal(loid)s including As and Cr. The DOC also forms soluble complexes with metal(loid)s thereby influencing their mobility and bioavailability (Herngren et al., 2005; Bolan et al., 2011; Kunhikrishnan et al., 2012). In the case of Cr, reduction of Cr(VI) to Cr(III) decreases its bioavailability because Cr(III) is more strongly retained and therefore becomes less mobile (Eq. (1)). However, reduction of As(V) to As(III) (Eq. (2)) increases its bioavailability because As(III) is less strongly retained and hence becomes more mobile (Fig. 1). While irrigation with As-containing wastewater enters the food chain, enhancing pollutant phyto-availability becomes a key to effective phytoremediation.



Although a number of studies have examined the phytotoxicity of metal(loid)s derived from wastewater sources (Amiri et al., 2008; Ahmed and Al-Hajri, 2009), and several studies showed the effect of DOC on reduction of Cr (Bolan et al., 2003) and As (Cao et al., 2003), there is a notable dearth of information on the relative effect of

wastewater-derived DOC on the reduction and subsequent immobilization and bioavailability of these metal(loid)s. In this study, the effects of a number of wastewater sources on the reduction, mobility, and bioavailability of As(V) and Cr(VI) in both spiked and field contaminated soils were examined. The (im)mobilization of As(III) and Cr(III) following the reduction of As(V) and Cr(VI), respectively is discussed in relation to DOC-induced changes in the adsorption reactions of these two heavy metal(loid) species.

## 2. Materials and methods

### 2.1. Soil and wastewater sources

An Alfisol (red sandy loam soil with 18% clay content) with low organic matter content was collected from a non-contaminated source at Northern Adelaide Plains, South Australia. The soil was treated individually with two levels of As(V) and Cr(VI) (0 and 600 mg  $\text{kg}^{-1}$  soil) using freshly prepared  $\text{K}_2\text{HAsO}_4$  and  $\text{K}_2\text{Cr}_2\text{O}_7$  solutions, respectively, and incubated at field capacity for four weeks. In this study, an As-contaminated surface soil sample (0–15 cm depth), resulting from the historical use of As herbicide was collected from a railway corridor at Kapunda in South Australia (Smith et al., 2006). A Cr-contaminated surface soil (0–15 cm depth) from a long-term tannery waste contaminated site at Mount Barker, South Australia was also collected and the contamination of Cr resulted from the disposal of tannery sludge.

Four wastewater sources [storm water (SW), sewage effluent (SE), piggery effluent (PE), and dairy effluent (DE)] were used in this study. Natural storm water was sourced from the Bennett Road storm water drain, located in Mawson Lakes, Adelaide, South Australia; the tertiary-treated sewage effluent was collected from Bolivar wastewater treatment plant, South Australia; the piggery effluent was collected from Pig Production Institute (Roseworthy Campus, University of Adelaide); and the dairy effluent was collected from Flaxley Agricultural Centre, SARDI (South Australian Research and Development Institute), South Australia. Milli-Q (MQ) water was used as a control.

### 2.2. Characterization of soil and wastewater samples

All the soil samples (non-contaminated, spiked, and field contaminated) were air-dried and sieved to <2 mm before carrying out the characterization analyses such as pH, electrical conductivity (EC), total metal(loid) content, cation exchange capacity (CEC), and total organic carbon (TOC). The pH and EC of soil were determined by equilibration of soil with water (1:5 ratio) using end-over-end shaker and measuring the solution with a pH/EC meter (smartCHEM-LAB, TPS, Australia) after one hour. Dichromate oxidation method was employed for determining soil organic matter (Walkley and Black, 1934). The CEC was estimated using Hendershot and Duquette (1986) methods, but ammonium chloride was used instead of barium chloride and the cation concentration was analyzed using inductively coupled plasma mass spectroscopy (ICP-MS, Agilent). For total metal(loid) analysis, 0.5 g of soil was suspended in 5 mL aqua regia directly into a Teflon digestion vessel and digested in a micro-wave digestion oven (MARS5, CEM, USA). Method 3051A (USEPA, 1997) was employed for digestion parameters and the concentration of digested metal(loid)s was measured using ICP-MS. Standard reference material (Montana Soil SRM2711, certified by the National Institute of Standards and Technology, USA) and digestion blanks were used for each digestion batch to validate the digestion operation.

The wastewater samples were analyzed for pH, EC, dissolved solids, total, particulate and dissolved OC. To measure DOC, the samples were passed through 0.45  $\mu\text{m}$  membrane filters and

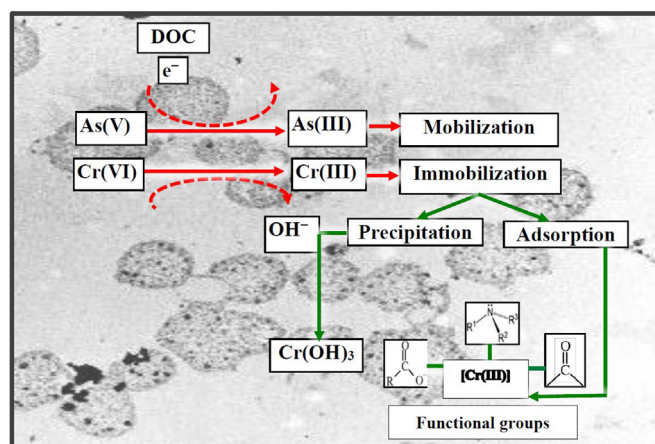


Fig. 1. Differential effect of dissolved organic carbon (DOC) on the reduction-induced mobility and bioavailability of arsenate (As(V)) and chromate (Cr(VI)).

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