



# Chemical vs bio-mediated reduction of hexavalent chromium. An in-vitro study for soil and deep waters remediation



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## ABSTRACT

The removal of hexavalent chromium, Cr(VI), from a water-sediment system has been studied in vitro in different condition of soil water saturation, in order to simulate different possible occurring real scenarios. Two different approaches have been compared: the bio-mediated and the chemical Cr(VI) reduction. In the first technique three organic nutrients have been tested: glucose, trehalose and  $\beta$ -cyclodextrin. For the chemical remediation reducing agents such as sodium sulphite, sodium metabisulphite and ascorbic acid were considered.

Both bio-mediated and chemical remediation approach yielded to the total abatement of Cr(VI) from a contaminated soil. No relevant drawbacks were observed in term of release of metal ions in solution or pH perturbation. Among the organic nutrients glucose showed the best performances while the best chemical reducing agent was ascorbic acid. Ascorbic acid can be considered more advantageous because its reaction with Cr(VI) is very fast (total Cr(VI) abatement in 24 h) and allows a better control of the process parameters, not involving the action of microorganisms; moreover its higher cost compared to glucose, is compensated by the much lower amount necessary to attain 100% of Cr(VI) reduction.

In case of in-situ remediation of groundwater, the spreading on soil of ascorbic acid would contribute to reduce the leaching of Cr(VI), helping the overall site remediation.

## 1. Introduction

The contamination of natural ecosystems by hexavalent chromium, Cr(VI), is of major concern worldwide, since Cr(VI) has been recognized as highly toxic, mutagenic and cancerogenic for living organisms. It has been classified among the 20 most hazardous environmental pollutants of the last 15 years (Chrysochoou et al., 2012). The highest risk of release of Cr(VI) in the environment is due to industrial activities, such as plating, painting processes, metallurgy and tanning industry. An evaluation of the European Pollutants Emission Register referred to 2012, estimates the contribution of different sources to the release of Cr into waters and soils and also reports the contribution of each country (<http://prtr.ec.europa.eu>, 2017).

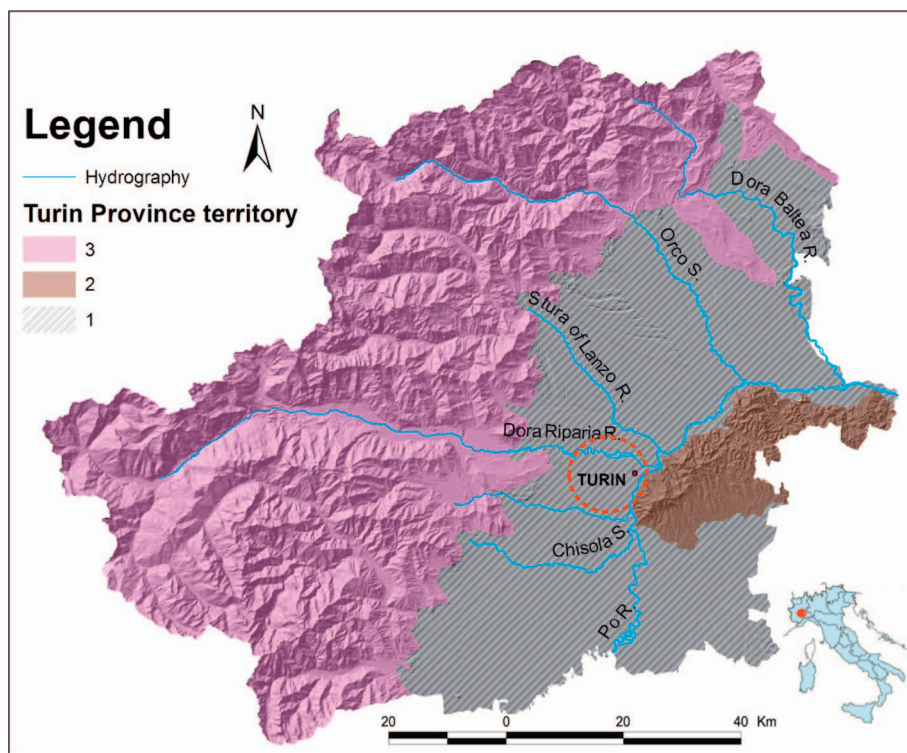
When present in soil-water systems Cr(VI) could be naturally reduced to trivalent chromium, Cr(III), being the latter specie much less toxic and with a very low solubility in the typical natural pH and redox potential ranges (Zayed and Terry, 2003). Chromate reduction in soils is mainly due to the action of iron, vanadium, sulfides and organic matter; the reduction efficiency increases when the pH decreases (Zayed and Terry, 2003). Nevertheless the capacity of soil is not enough

to operate this reduction in reasonable times; indeed Cr(VI) can persist in soils and because of its high mobility, it can easily leach and contaminate ground waters (Fendorf, 1995).

The transformation of Cr(VI) into Cr(III) is controlled by many factors, both biotic and abiotic; as for the abiotic ones, the main physical parameters influencing the reduction on soil are pH and redox potential (Dhal et al., 2013 and references therein). On the other hand, from the biotic point of view, the Cr(VI) conversion to Cr(III) depends from the microorganisms present in soil, able to mediate the process (Valls and De Lorenzo, 2002). The microbial reduction of Cr(VI) to Cr(III) has been deeply studied and reviewed (Cervantes et al., 2001; Cheung and Gu, 2007); it can follow two different paths, i.e. direct or indirect reduction. The indirect mechanism involves reducing agents produced by the bacterial metabolism; the main microorganisms involved in such a mechanism are iron and sulfate reducing bacteria, that transform  $\text{Fe}^{3+}$  and  $\text{SO}_4^{2-}$  into  $\text{Fe}^{2+}$  and  $\text{HS}^-$ . These latter compounds can react with Cr(VI) yielding to its conversion to Cr(III) (Arias and Tebo, 2003). On the other hand the direct process can be operated by a variety of reductase bacteria taking electrons from reduced organic substrates (amino acids, nucleotides, sugars, vitamin, organic acids or

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**Fig. 1.** The Turin Province territory. The Turin Po Plain (1), consisting of fluvial sediments, is interposed between the Alps (2) formed by metamorphic and Turin Hill (3), formed by sedimentary marine rocks. Red circle: area of study. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

glutathione) (Martha et al., 2007).

The increasing diffuse contamination of anthropogenic origin poses the need of developing remediation technologies for soil and groundwater treatment. Starting from 80-ies many approaches have been followed, physic-chemical as well as biological, operating in-situ or ex-situ (Higgins et al., 1997), aiming in most cases to the transformation of Cr(VI) into Cr(III), not harmful. In the remediation of contaminated sites, microbial processes are of potential application for the removal of toxic metal species, by varying their oxidation state and therefore transforming toxic species into less harmful ones or less soluble ones, thus allowing their precipitation (Mulligan et al., 2001).

The bio-reduction of Cr(VI) represents an interesting alternative to the traditional approaches, mainly based on physic-chemical processes of soil washing or the use of chemical reducing agents. Such processes are expensive and energy consuming and can severely modify the soil characteristics (Jing et al., 2007). Bio-remediation is cheaper, and has a lower impact on soil properties (physical, chemical and biological); its kinetic could be favored by inoculation of selected bacteria resistant to Cr(VI) and with high capability to reduce Cr(VI) to Cr(III) (bioaugmentation) (Viti and Giovannetti, 2007).

Recently the use of organic nutrients has been proposed as a possible innovative bio-mediated approach (Leita et al., 2011; Smith et al., 2002; Tokunaga et al., 2003; Michail et al., 2015); in particular the use of glucose, cheap and non-toxic substance, could be of great interest. Indeed glucose is able to promote the growth of microorganisms naturally present in soils and stimulate their reductive capability. The microorganisms action mechanism is rather complicate and can include a direct involvement, if bacteria can uptake Cr(VI) and transform it into Cr(III) enzymatically, as well as an indirect mechanism, if the Cr reduction is mediated by bacteria metabolism or decomposition (Gadd, 1992).

Among the drawbacks of the bio-mediated approach the slow kinetic and the difficulty of keep the system well controlled can be cited. In this direction, an alternative could be represented by the addition of reducing chemical substances; their advantage is represented by the usually very fast and complete reaction while a drawback could be the reagent cost and safety (Dhal et al., 2013).

When considering a real case of contamination, soil remediation is more difficult compared to groundwater remediation; in the latter case the redox agent can diffuse rather fast and efficiently, whereas the dispersion in a solid medium is much more difficult. Moreover it is of fundamental importance to plan both soil and water remediation; it is meaningless to remove Cr(VI) from groundwater if the soil is contaminated, because of the risk of leaching and re-contamination of the aqueous phase.

In any case, before applying a remediation process in field, a preliminary evaluation of its performances at laboratory scale would provide useful information on the effect of the experimental parameters on the process efficiency; it will allow also to evidence possible side effects, due to the eventually pH and redox condition modification.

The present work deals with the in vitro study of Cr(VI) removal from a water-sediment system in different condition of soil water saturation %.

The Cr(VI) removal was addressed following two different approaches: i) bio-mediated and ii) chemical Cr(VI) reduction. In the first case three organic nutrients have been tested: glucose, trehalose and  $\beta$ -cyclodextrin. For the chemical remediation reducing agents such as sodium sulphite, sodium metabisulphite, and ascorbic acid were considered.

Since both biotic and abiotic process could affect water physical-chemical properties, a cascade effect on the chemistry of metal oxides present in the soil cannot be disregarded; a specific attention has therefore been devoted to the concentration of naturally occurring element such as Fe, Ni and Mn, whose content in solution has to be kept under control in order to guarantee the environmental suitability of the overall Cr(VI) abatement procedure.

## 2. Materials and methods

The study area is located in the plain in the urban area of Turin city. The Turin Province consists of three distinct sectors: a plain (Turin Po Plain), an alpine area and a hilly area (Fig. 1). The lowlands form the central section and are bordered by mountains and hills to the west and east, respectively.

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