

Adsorption-desorption of arsenic species in river sediment, water and associated microplastics

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Thesis submitted in fulfilment of the requirements for the degree of **Doctor of Philosophy**

under the supervision of Professor John Zhou

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June 2022

CERTIFICATE OF ORIGINAL AUTHORSHIP

I, Kien Thanh Nguyen declare that this thesis, is submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the School of Civil and Environmental Engineering/Faculty of Engineering and Information Technology at the University of Technology Sydney.

This thesis is wholly my own work unless otherwise referenced or acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

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This research is supported by the Australian Government Research Training Program.

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Date: 06/06/2022

ACKNOWLEDGEMENT

First and foremost, I would like to thank my principal supervisor, Professor John Zhou, for his constant guidance, encouragement, and patience throughout my PhD study. His guidance helped me in the research, publications and this thesis. This thesis would not be possible without his help, exceptional supervision, and valuable contributions. In the last semester, I appreciated his support to obtain the tuition fee scholarship so that I could fully focus on study. I also would like to thank my co-supervisor, Doctor Yuhan Huang for his support, insightful comments and revisions on my publications and thesis. He provided significantly useful and practical help whenever I needed.

I would like to thank Doctor Mohammed Johir and Nirenkumar Pathak for their help in various laboratory work, including the interruption period due to COVID-19 pandemic. I also would like to thank Ms Van Le and UTS Graduate Research School for their help in administration matters. I would like to thank Professor David McGloin and GRS for their support to obtain the UTS Scholarship for the Research Session 2 2021. I would like to thank all my friends, who encouraged and helped me over the time. I also appreciate Dr. Youngwoo Choo at University of Technology Sydney, Dr. Bin Gong and Dr. Songyan Yin at the University of New South Wales to help in analysing samples. Finally, I would like to acknowledge my family (my parents and, especially my wife Nguyet Tran and my daughter Chi Nguyen), who have worked hard and stayed with me through one of the toughest times of my life.

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ABBREVIATIONS

α:	the Elovich initial adsorption rate
β:	the Elovich desorption constant
As:	Arsenic
As(III):	Inorganic arsenite
As(V):	Inorganic arsenate
BET:	Brunauer-Emmett-Teller
C _e :	the concentration in solution at the equilibrium time
C _o :	the concentration in solution at the initial time
C _t :	the concentration in solution at time t
DI water:	Deionized water
DOM:	Dissolved organic matter
Eh:	Redox potential
FTIR:	Fourier Transform Infrared Spectroscopy
HDPE:	High-density polyethylene
K _F :	the Freundlich constant
K _L :	the Langmuir bonding energy constant
K _p :	the solid-solution partition coefficient
Ks:	the Sips energy constant
LDPE:	Low-density polyethylene
LOI:	Loss-on-ignition
MPs:	Microplastics
OM:	organic matter
n:	the Freundlich exponent

n _s :	the Sips exponential factor
PA:	Polyamide
PCBs:	Polychlorinated biphenyls
PE:	Polyester
PET:	Polyethylene terephthalate
PFO:	Pseudo first-order
PP:	Polyprolylene
PS:	Polystyrene
PSO:	Pseudo second-order
PTFE:	Polytetraflouroethylene
PUR:	Polyurethane
PVC:	Polyvinyl chloride
q _e :	the concentration in the adsorbent at the equilibrium time
q _o :	the concentration in the adsorbent at the initial time of desorption
q _m :	the maximum concentration in the adsorbent at the equilibrium time
q _t :	the concentration in the adsorbent at time t
R _L :	the equilibrium parameter
RS:	River sediment
RW:	River water
RS-NOM:	River sediment without organic matter
SEM-EDS:	Scanning electron microscopy ad energy dispersive X-ray spectroscopy
SOM:	Soil organic matter
SSA:	Specific surface area
TOC:	Total organic carbon
UV:	Ultraviolet

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- XPS: X-Ray Photoelectron Spectroscopy
- XRD: X-ray powder diffraction

LIST OF PUBLICATIONS

Kien Thanh Nguyen, Hung Manh Nguyen, Cuong Kim Truong, Mohammad Boshir Ahmed, Yuhan Huang, John L. Zhou (2019) Chemical and microbiological risk assessment of urban river water quality in Vietnam. *Environmental Geochemistry and Health*, 41: 2559-2575. https://doi.org/10.1007/s10653-019-00302-w

Kien Thanh Nguyen, Mohammad Boshir Ahmed, Amin Mojiri, Yuhan Huang, John L. Zhou, Donghao Li (2021) Advances in Arsenic contamination and adsorption in soil for effective management. *Journal of Environmental Management*, 296: 113274. <u>https://doi.org/10.1016/j.jenvman.2021.113274</u>

Kien Thanh Nguyen, Yuhan Huang, John L. Zhou. Behaviour and mechanism of arsenite and arsenate interactions with river sediment and microplastics. *Journal of Environmental Management* (Accepted on 6 June 2022).

ABSTRACT

Arsenic (As) is a ubiquitous toxic metalloid, and its pollution has been reported in soil, surface water, groundwater and sediment worldwide. Additionally, micro-plastics (MPs) are an emerging organic pollutant widely detected in different environments. So far, studies on the interfacial behaviour of As in the river sediment-water were limited. This PhD research, therefore, aims to explore the adsorption and desorption processes and interfacial behaviour of As in the contaminated water-sediment system. The kinetic and isotherm sorption models are used to estimate the sorption behaviours of As(III) and As(V), while various surface characterisation methods are applied to understand the interactions between As species with adsorbent surface and the transformation among As species.

Firstly, the adsorption and desorption of As(III) on river sediment (RS) were investigated under various environmental conditions and sediment characteristics. Higher As(III) and As(V) adsorption on RS was found in acidic to neutral conditions and on smaller size fractions of sediment. The monolayer maximum surface adsorption (q_m) of As(V) (210.0 mg/kg) was higher than that of As(III) (201.7 mg/kg). The FTIR results showed the changes in surface functional groups of river sediment before and after adsorption, indicating that Fe–O/Fe–OH, Si(Al)–O, –OH and –COOH functional groups were predominantly involved in As(III) and As(V) adsorption on sediment surface.

Secondly, the adsorption of As(III) and As(V) on polystyrene (PS) and low-density polyethylene (LDPE) in deionized (DI) water and simulated river water (RW) conditions were investigated by using bead MPs. Physisorption was the main mechanism involved in the adsorption processes based on the isotherm modelling. Moreover, the interactions between As species and PS and LDPE mainly occurred on the carboxyl and hydroxyl groups of adsorbent surfaces, whilst electrostatic force and non-covalent interaction played an important role in the adsorption mechanism of As(III) and As(V) on PS and LDPE.

Furthermore, the sorption behaviour of As(III) and As(V) was evaluated by using mixtures of sediment with PS or LDPE and DI water or RW. The amounts of As(III) and As(V) adsorbed in RW solution with the presence of PS and LDPE were lower than those in sediment only, suggesting that PS and LDPE may inhibit sediment adsorption of As(III) and As(V). The desorption process showed a positive impact of RW in the release of As(III) and As(V) into the water phase. This study provided valuable information on the sorption behaviour and mechanism of inorganic As species in the simulated river system.