Summary

The investigation of the fate and transport of waterborne particles is an evolving field within research and engineering, driven by advancements in transport analysis methodologies and the development of innovative engineered nano- and micromaterials. Waterborne particles can be classified into two categories based on their origin source: natural particles or anthropogenic particles. Both categories can diversly impact on the environment and humans. On one hand, they can be specifically engineered to remove contaminants from water bodies. In such case, engineered particles are introduced on purpose into the environment, and it is therefore crucial that the materials they are composed of are not harmful for humans and the ecosystem. On the other hand, waterborne particles can also act as contaminants, potentially posing toxicity or carcinogenic risks to humans. Groundwater serves as a critical pathway for human exposure to hazardous particles, being one of the primary water sources for various anthropogenic activities, including agriculture, industry, and human consumption. The experimental and modelling study of particle transport in saturated porous media is therefore essential to implement effective particle-based groundwater remediation measures, and to understand the environmental fate of hazardous particles. In this study, four particle transport-related research topics were explored. These themes aimed to address aspects pertaining to both the fate of hazardous particles in aquifer systems (i.e. asbestos fibres) and in water treatment plants (i.e. nanoplastics) and the enhancement of subsurface delivery of engineered nanomaterials (i.e. iron-based nanoparticles) for environmental remediation.

The first research topic deals with nanoplastics (NPs), widespread anthropogenic particles acting as water contaminants. The detection of microscale plastics in potable water have led to concerns about the efficacy of current drinking water treatment plants (DWTPs) to remove even smaller particles. Synthetic plastics (≈ 200 nm), were used in this study to investigate for the first time the behaviour and removal of NPs in conventional DWTPs. Starting from lab-scale and pilot-scale experiments, the removal of NPs through multiple filtration steps in a real municipal full-scale DWTP was simulated. Model predictions indicated that a filtration system composed of rapid sand, activated carbon and slow sand filtration steps would guarantee a global removal efficiency greater than 99.9%. This result suggests that conventional treatment systems, employing the filtration trains of this study, are likely to effectively remove NPs from water without requiring specific upgrades.

Not only anthropogenic, but also natural particles can act as water contaminants. For instance, asbestos is widely recognized as a carcinogen agent when dispersed in air, but very little is known about its migration and health effects in water. Several studies highlighted asbestos presence in groundwater but failed to assess its mobility in aquifer systems. To fill this gap, a lab-scale study was conducted to investigate the transport of crocidolite, an amphibole asbestos, through sandy porous media mimicking different aquifer systems. The results showed for the first time that crocidolite is potentially mobile in quartz sand due to repulsive interactions between asbestos fibres and sand grains. Specifically, 5-to-10-μm-long fibres flowed through all the tested sands while fibres longer than 10 μm were mobile only in the coarser medium. This study therefore confirmed that the exploitation of crocidolite-reach aquifers could expose humans to asbestos and highlighted the importance of including groundwater as a potential exposure pathway in the assessment of the human health risk in asbestos contaminated sites.

The second part of this work deals with particles specifically engineered to remove contaminants from water. For example, zero-valent iron nanoparticles (nZVI) are extensively studied for in-situ remediation of aquifer systems, to the extent that many types of nZVI particles are nowadays available on the market. However, in addition to the high nZVI reactivity towards numerous persistent contaminants, a successful remediation application cannot do without effective delivery of these particles into the subsurface. In this study, the effect of five different surface modifications (e.g. sulfidation, passivation, organic coating) on nZVI transport in sandy porous media was investigated by performing several column tests. The findings from column tests were then scaled up through numerical transport modelling to simulate a realistic field injection. Comparing the simulated particle distribution and migration distance revealed that a slight sulfidation degree appears to be the optimal method for nZVI surface modification, ensuring enhanced mobility and more homogeneous distribution within the aquifer.

Despite surface modification or the use of viscous stabilizers showed effective in improving the colloidal stability and subsurface delivery of nZVI, the creation of a uniform reactive zone within contaminated aquifers still remains an open challenge, particularly in heterogeneous and low-permeability horizons. To cope with this issue, in this research an innovative strategy was proposed to synthesize nZVI directly within a contaminated aquifer through injection of environmentally friendly precursor solutions. The protocol was validated through a 3D medium-scale homogeneous permeable aquifer, demonstrating the effectiveness of nZVI production in realistic conditions, and in a 2D laboratory setup replicating three different heterogeneous aquifers. This novel strategy shifts the paradigm of particle delivery in porous media, potentially enabling effective remediation of heterogeneous aquifers with mid- and low-permeability inclusions.

The findings of this study, achieved through a combined experimental and modelling approach, provide a comprehensive understanding of the transport and the fate of both contaminant and engineered particles within saturated porous media, such as aquifer and filtration systems. This enables to assess whether current technologies are adequate to safeguard or remediate the environment, and if not, how they should be improved, all aimed towards the mitigation of human exposure to natural or anthropogenic contaminants.