Adsorption of Perfluorooctanoic Acid (PFOA) Using Graphene-Based Materials

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Background/Objectives. Due to the persistence of PFOA and related perfluorinated alkyl substances (PFAS) in the environment, novel methods for their remediation are highly desirable. As degradation of these chemicals is highly energy-intensive, adsorption is an effective strategy to manage contamination, in situ. Adsorption of PFOA onto surfaces of carbonaceous materials (like chars and activated carbons) have been demonstrated in the past. Graphene, the latest addition to the nanocarbon family, is an excellent candidate for use as an adsorbent due to its high surface area and versatile surface chemistry. In this study, we synthesised two graphene derivatives, graphene oxide (GO) and an iron-modified graphene composite (FeG), and evaluated them for PFOA sorption. The influence of different pH conditions, ionic strength, and concentrations were investigated to evaluate adsorption efficiency under different environmental conditions, and performance was compared with a commercial adsorbent.

Approach/Activities. Adsorbents GO and FeG were synthesised in the laboratory using graphite. RemBind[™] (which is a mixture of activated carbon, alumina and kaolinite; manufactured by Ziltek Pty. Ltd.) was chosen as the commercial benchmark as it has been used successfully to treat PFAS-contaminated soils in Australia and the USA. All adsorbents were characterised for their surface and chemical properties. Laboratory-scale batch adsorption tests were performed by mixing the adsorbents with ¹⁴C radiolabelled PFOA-spiked solutions (in 10 mM CaCl₂ background electrolyte), under constant agitation on an orbital shaker to attain equilibrium. Sorption of PFOA was investigated under varying conditions of pH (3 − 10), ionic strength (0 − 100 mM CaCl₂), and concentration (10 − 650 µg/L). Supernatants of equilibrium solutions were then analysed for PFOA-derived radioactivity (beta-decay) using a liquid scintillation counter, from which concentrations of PFOA were calculated. All experiments were performed in triplicate. Control experiments were carried out in the absence of any adsorbents, and these data were used to correct for any sorption observed onto lab-ware, which was found to be significant. Experiments involving desorption of sorbed PFOA are currently being undertaken to further understand the nature of binding.

Results/Lessons Learned. Both GO and FeG successfully absorbed PFOA, with FeG exhibiting greater sorption. Performance of FeG was comparable with the commercial adsorbent, RemBind[™]. Considerable sorption of the anionic PFOA molecule by GO, despite its highly negatively charged surface, suggests non-electrostatic interactions. Iron-functionalisation of GO led to enhanced sorption, highlighting the role of goethite minerals in binding PFOA. Increase in pH hindered PFOA sorption by GO due to increased repulsion, however pH did not significantly affect sorption by FeG and RemBind[™]. This could be attributed to stronger interactions associated with the mineral surfaces. Similarly, increases in ionic strength did not affect sorption by FeG and RemBind[™]. At the concentrations tested, adsorption isotherms followed a linear relationship with no indications of saturation. Preliminary data collected indicate the role of polarity of the extracting solvents in desorbing adsorbed PFOA from adsorbents; data are expected to provide further information on strength and nature of binding interactions. Resistance to variations in environmental conditions highlights the possibility of

using graphene-based materials, in situ, for remediation of PFOA and PFAS-contaminated sites. Data highlight the critical role of surface chemistry, rather than surface area in PFOA sorption.