

Experimental study of environmental (pH, T, salinity, concentration) control on ammonium adsorption on clay minerals

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Adsorption of ammonium (NH_4^+) by clay minerals in sediments and altered oceanic crust plays an important role in Earth's nitrogen cycle by transferring nitrogen from the biosphere and hydrosphere into the lithosphere. In addition, the nitrogen in clay minerals can be a potential proxy for reconstructing marine environments of ancient oceans and searching for life on extraterrestrial planets. However, how different environmental conditions (e.g., ambient pH, temperature, salinity and NH_4^+ concentration) affect NH_4^+ adsorption onto clay minerals is still poorly understood. In this study, we carried out laboratory experiments to examine the NH_4^+ adsorption behavior of a number of clay minerals (montmorillonite, vermiculite, illite, chlorite, kaolinite) under conditions varying in aqueous NH_4^+ concentration (20, 50, 100, 200, 500, 1000 mg/L), pH (2, 5, 7), salinity (fresh water vs. artificial seawater), and temperature (23, 50, 70 °C). Our results show that NH_4^+ adsorption is very fast (reaching equilibrium within a few minutes) for all studied minerals. In contrast, NH_4^+ adsorption capacity is strongly mineral-dependent and follows the order of vermiculite \approx montmorillonite \gg illite $>$ kaolinite \approx chlorite. Vermiculite and montmorillonite have much higher NH_4^+ adsorption capacities because of the exchange between their interlayer cations and NH_4^+ . For individual clay mineral, its NH_4^+ adsorption capacity is highly susceptible to environmental conditions (e.g., aqueous NH_4^+ concentration, pH, temperature, and salinity). For example, NH_4^+ adsorption on montmorillonite and vermiculite significantly decreases with salinity increase from fresh water to artificial seawater, pH decrease from 7 to 2, and/or decreasing aqueous NH_4^+ concentration. Our data best fit the Freundlich Model of adsorption, and the output of this model suggests that NH_4^+ adsorption on clay minerals likely occurs in energetically heterogeneous surface sites. Our results provide new insights into the understanding of NH_4^+ transfer from seawater to sediments and to altered oceanic crust, preservation capability of the environmental nitrogen signature in clay minerals, and optimal conditions for clay to catalyze organic synthesis towards the origin of life on the early Earth.