Recent nanofluidic experiments revealed strongly different surface charge measurements for boron-nitride (BN) and carbon nanotubes when in contact with saline and alkaline water.¹,² These observations contrast with the similar reactivity of a graphene layer and its BN counterpart, using density functional theory (DFT) framework, for intact and dissociative adsorption of gaseous water molecules. We investigate, by DFT in implicit water, single and multiple adsorption of anionic hydroxide on single layers.³ A differential adsorption strength is found in vacuum for the first ionic adsorption on the two materials - chemisorbed on BN while physisorbed on graphene. The effect of implicit solvation reduces all adsorption values, resulting in a favorable (nonfavorable) adsorption on BN (graphene). We also calculate a pKa ≈ 6 for BN in water, in good agreement with experiments. Comparatively, the unfavorable results for graphene in water echo the weaker surface charge measurements but point to an alternative scenario. Various carbon and boron nitride allotropes are currently tested to probe possible effects of stacking, chirality and curvature. The adsorption of alternative ions are also investigated to determine the charging mechanism of the carbon surface.