Supplementary Information

Removal of Pb-based compounds mediated by graphene oxide-like materials obtained from Sargassum: Unravelling key features of their interaction using density functional theory and spectroscopic methods.

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The optimization curves for the cutoff energy, k-points and graphene cell parameter are depicted in **Figure S1**. Notice that at 480 eV of energy cutoff, a linear tendency is observed denoting that this energy is good enough to describe the system. The k-points graph shows that for a grid of 7×7×1, the energy is converged.
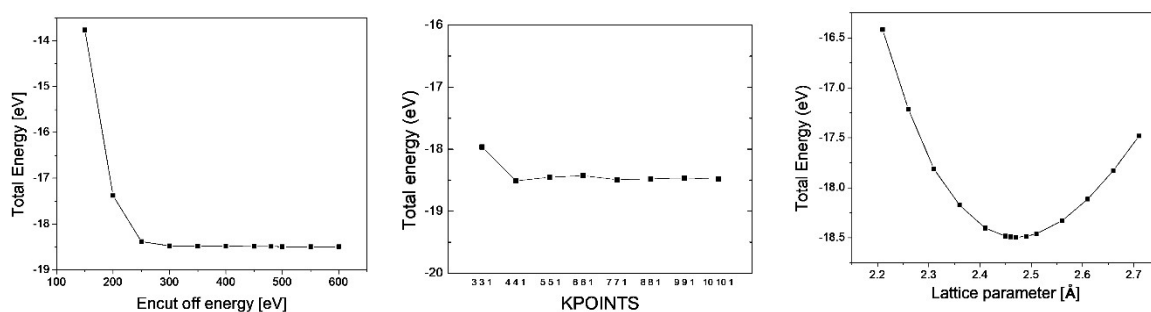

Figure S1. From left to right are the optimization curves for energy cutoff, k-points, and cell parameters.

In the hydrocerussite mineral, the $PbCO_3$ is a laminar compound composed of Pb^{2+} ions three-fold coordinated with CO_3^{2-} functional groups by six ionic bonds of 2.62 Å in length. To perform a realistic interaction between GOs and cerussite, we isolated the $Pb(CO_3)_3$ molecule to maintain the chemical environment of Pb in the mineral.

Figure S2 shows the electron localization function (ELF) isosurfaces for the $PbCO_3$ crystal and the $Pb(CO_3)_3$ molecule, which are equivalent. Also, **Figure S2c** depicts the line profiles for the Pb-O bonds in the crystal and the molecule. Notice a similar ELF distribution in both, crystal and molecule; the line profiles also show similar bond lengths and ionic behavior.
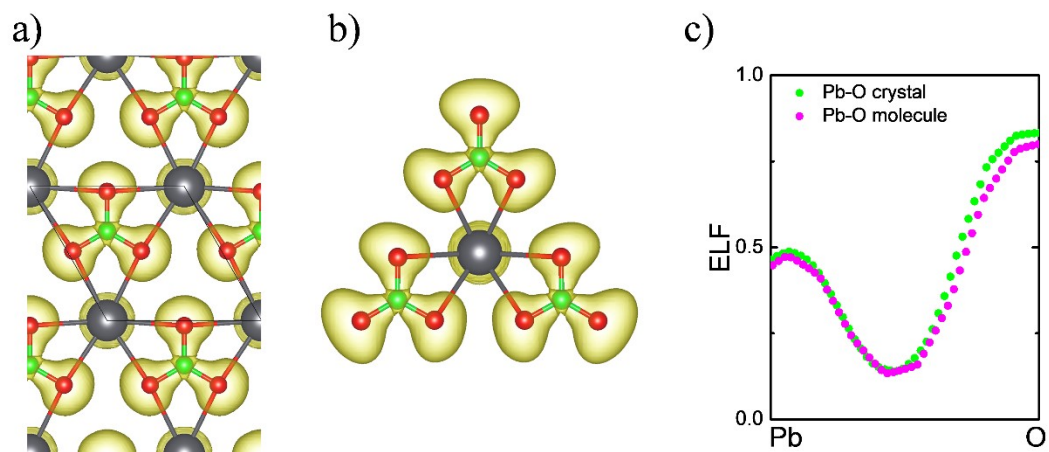
Figure S2. ELF isosurfaces (with isovalue of 0.5) for a) the crystal $PbCO_3$ and b) the $Pb(CO_3)_3$ molecule, and c) depicts the line profile along the Pb-O bond present in both systems.
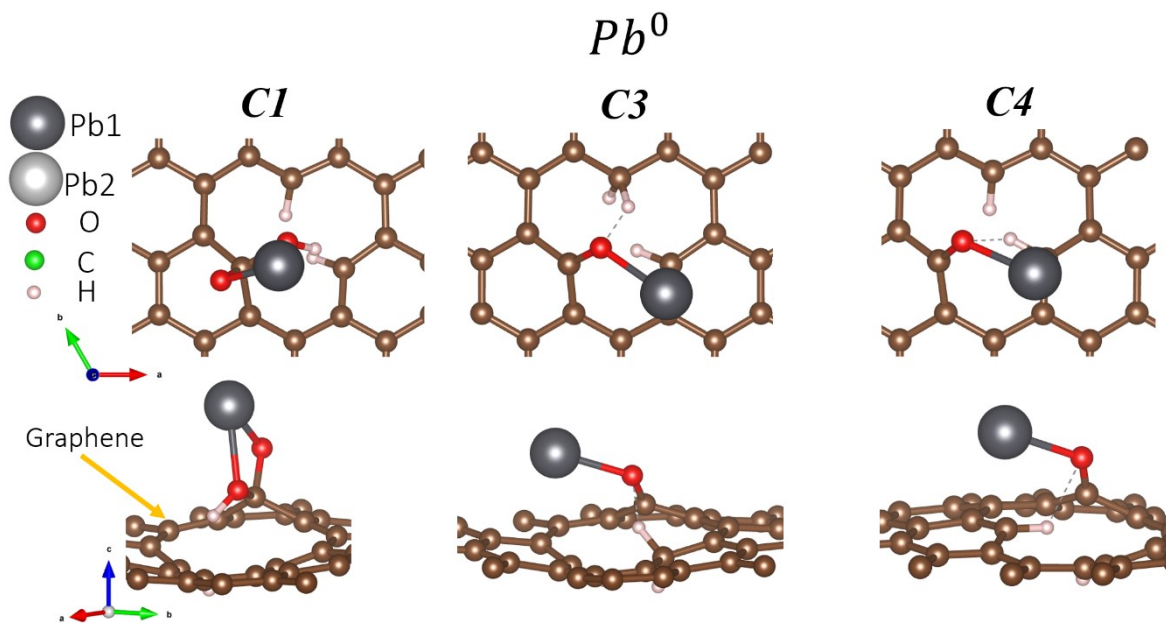

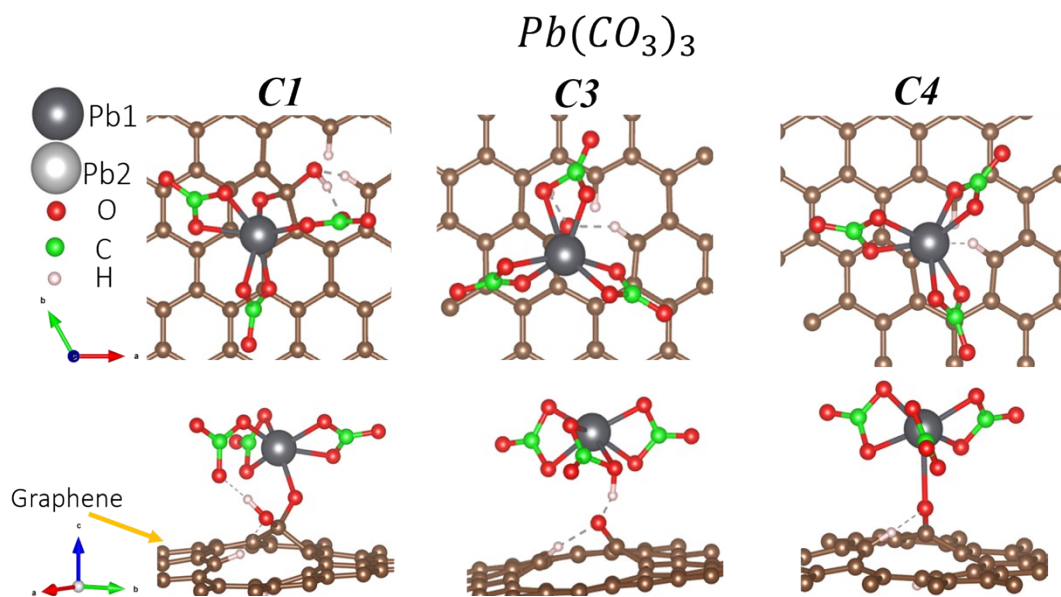**Figure S3**. Top and side views of several models for the Pb^{2+} adsorption.

$Pb(CO_3)_3$

Figure S4. Top and side views of several models for $Pb(CO_3)_3$ molecule adsorption onto GOs.
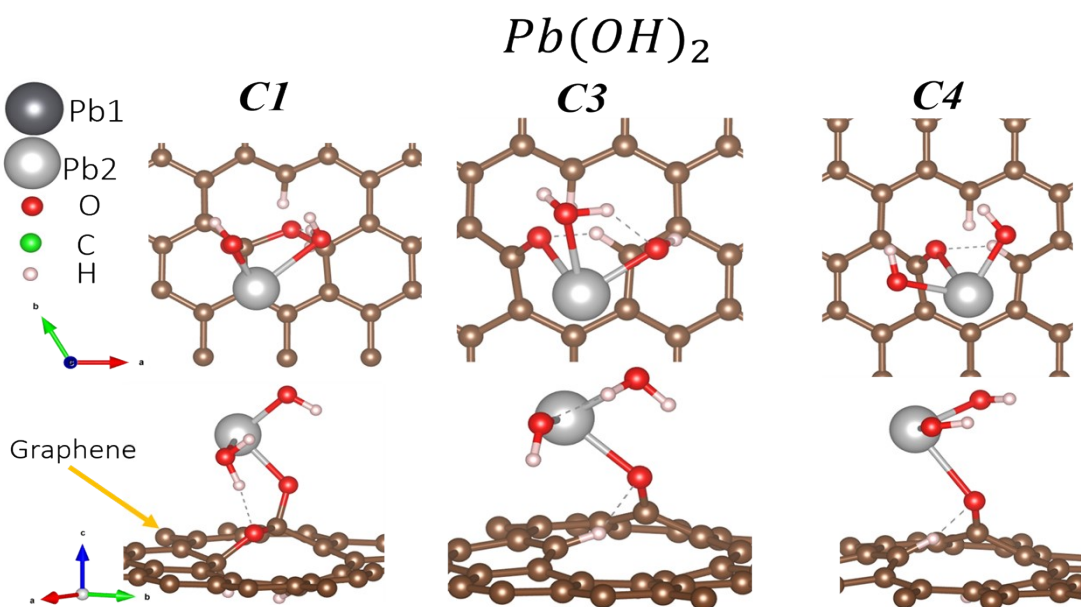
$Pb(OH)_2$

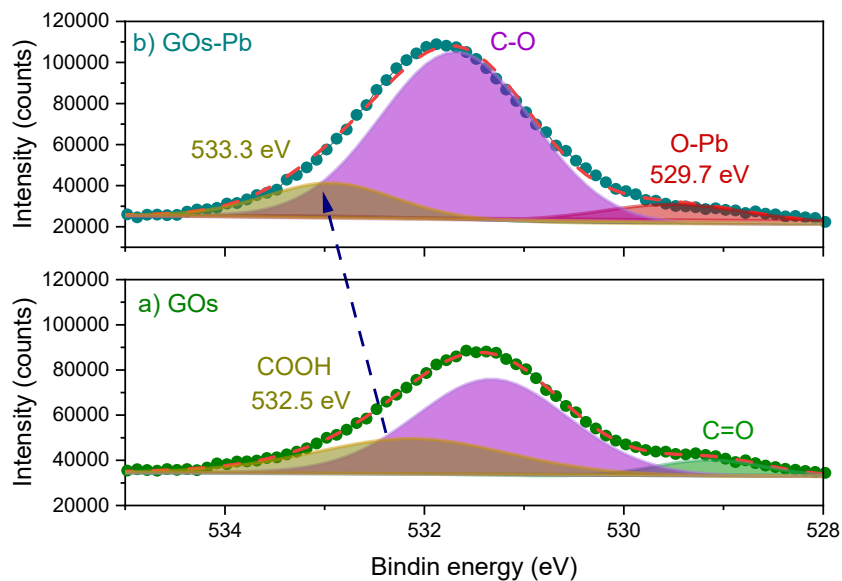**Figure S5**. Top and side views of the less stable $Pb(OH)_2$ adsorption models.

Figure S6. Deconvolution of O_{1s} peak for GOs a) after the interaction with Pb^{2+} and b) before the interaction.

The EDS map likely shows a spatial distribution of elements, and in this case, it reveals the presence of metallic lead (Pb) on the surface of the GOs-Pb material. The map would display regions where the Pb signal is detected, indicating the localization of metallic lead within the material.
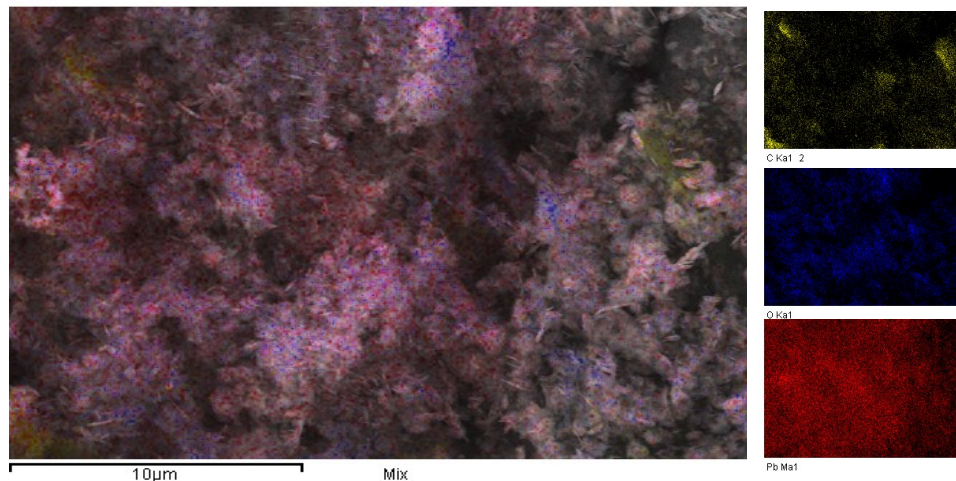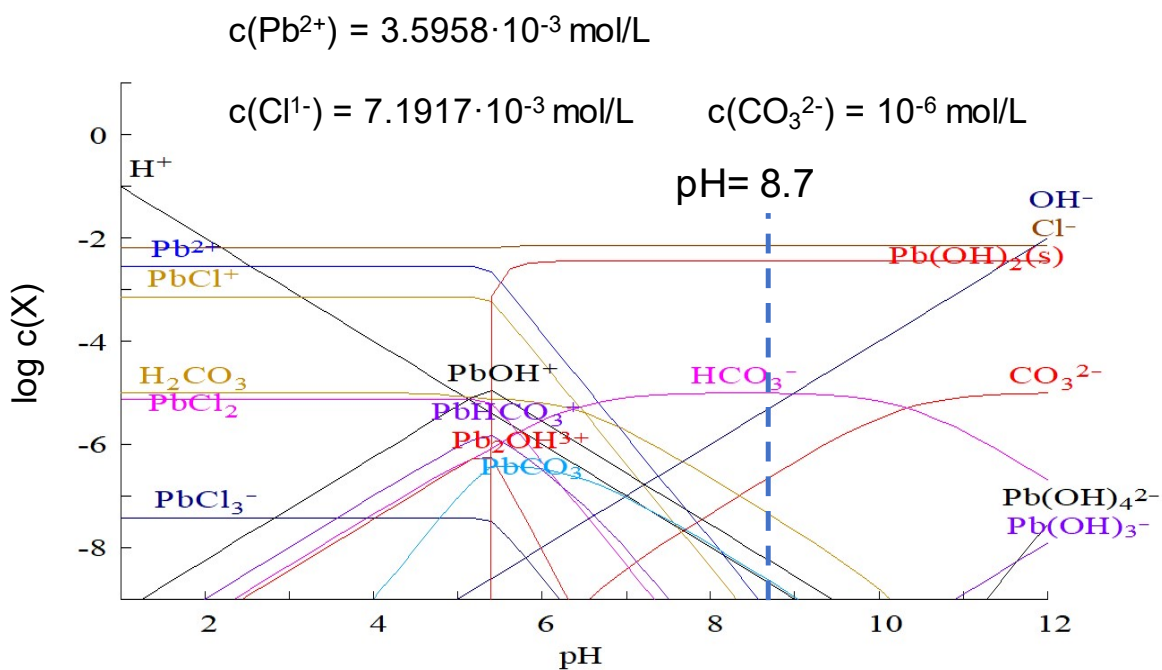
Figure S7. EDS mapping of GOs-Pb^{2+} material

$c(Pb^{2+}) = 3.5958 \cdot 10^{-3}$ mol/L

$c(Cl^{1-}) = 7.1917 \cdot 10^{-3}$ mol/L $c(CO_3^{2-}) = 10^{-6}$ mol/L

pH= 8.7

Figure S8. Logarithmic distribution diagram of the species presents during the GOS-Pb^{2+}
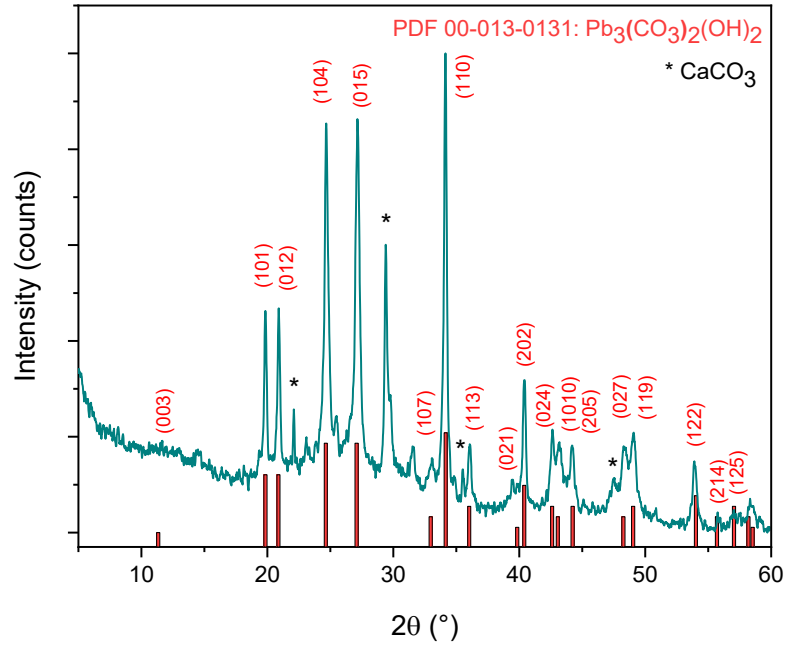
interaction.

Figure S9. XRD of GOs-Pb^{2+} system.

Guide of terms

Electron density gradient $\nabla\rho(r)$

$$\rho\nabla(r) = \begin{bmatrix} \dfrac{\partial\rho(r)}{\partial x} \\ \dfrac{\partial\rho(r)}{\partial y} \\ \dfrac{\partial\rho(r)}{\partial y} \end{bmatrix}$$

Electron density Gradient Norm $|\nabla\rho(r)|$

$$|\nabla\rho(r)| = \sqrt{\nabla\rho(r)\cdot\nabla\rho(r)} = \sqrt{\left(\dfrac{\partial\rho(r)}{\partial x}\right)^2 + \left(\dfrac{\partial\rho(r)}{\partial y}\right)^2 + \left(\dfrac{\partial\rho(r)}{\partial z}\right)^2}$$

Reduced density gradient $s(\rho)$

$$s(\rho) = \dfrac{1}{2(3\pi^2)^{\frac{1}{3}}}\dfrac{|\nabla\rho|}{\rho^{\frac{4}{3}}}$$

Electron density Hessian $\nabla\nabla^T\rho(r)$

$$\nabla\nabla^T\rho(r) = \begin{bmatrix} \dfrac{\partial^2\rho(r)}{\partial x^2} & \dfrac{\partial^2\rho(r)}{\partial x\partial y} & \dfrac{\partial^2\rho(r)}{\partial x\partial z} \\ \dfrac{\partial^2\rho(r)}{\partial y\partial x} & \dfrac{\partial^2\rho(r)}{\partial y^2} & \dfrac{\partial^2\rho(r)}{\partial y\partial z} \\ \dfrac{\partial^2\rho(r)}{\partial z\partial x} & \dfrac{\partial^2\rho(r)}{\partial z\partial y} & \dfrac{\partial^2\rho(r)}{\partial z^2} \end{bmatrix}$$

Electron density Laplacian $\nabla^2\rho(r)$

$$\nabla^2\rho(r) = \nabla\cdot\nabla\rho(r) = \dfrac{\partial^2\rho(r)}{\partial x^2} + \dfrac{\partial^2\rho(r)}{\partial y^2} + \dfrac{\partial^2\rho(r)}{\partial z^2} = \lambda_1 + \lambda_2 + \lambda_3$$