

A COMPARATIVE REVIEW OF CLINOPTILOLITE (natural zeolite), MONTMORILLONITE (bentonite), AND CALCITE-BASED GREEN SORBENTS FOR HEAVY METAL REMOVAL FROM WATER

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Abstract

The continual contamination of aquatic (and terrestrial ecosystems) by heavy metals is a long-lasting, crucial problem that endangers both the environment and public health alike. Due to their toxic nature, non-degradable state, and the tendency to bio-accumulate in the food chain, heavy metals have rightfully earned their ominous reputation as “silent killers.” While traditional (conventional) treatment methods are generally effective in remediating these contaminants, unfortunately, their widespread application is often handicapped and challenged by their time-consuming and operational costs, urging the research community to explore cheaper and more eco-friendly (greener) methods. From this perspective, natural zeolites (clinoptilolite-dominant), bentonites (montmorillonite-rich) and prevalent carbonate minerals such as calcite, are some notable mentions. They present naturally abundant, affordable adsorbents with diverse structures and high pollutant affinity, used as remediation “agents”. However, despite their individual merits, systematic, side-by-side comparative assessments of their characteristics under variable conditions remain insufficient. Consequently, this review attempts to fill the gap in literature by comparing their structural and chemical properties, while dissecting their adsorption mechanisms across different conditions, simultaneously guiding their efficient and informed use in sustainable heavy metal removal, with a focus on water treatment/purification. This comparative evaluation of literature data, highlights that montmorillonite, exhibits the highest adsorption capacity, due to its high surface area and cation exchange capacity, clinoptilolite, provides a balanced combination of efficiency, selectivity and operational stability, owing to its zeolitic framework, structure and charge characteristics, whereas calcite, although influenced by solution chemistry, shows strong efficiency under alkaline conditions, emphasizing the importance on condition-specific adsorbent selection, while supporting the broader implantation of these benign minerals as trajectories toward sustainable heavy metal remediation, aligning both environmental protection objectives and public health safeguarding.

Keywords: natural zeolites, clinoptilolite; bentonites, montmorillonite; carbonate minerals, calcite; heavy metals; water treatment; green chemistry

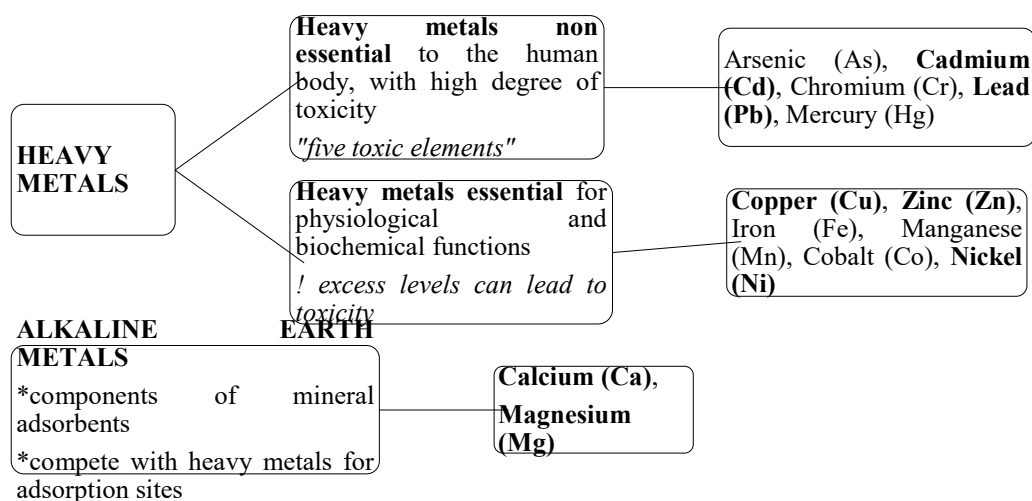
1. Introduction

Water is the heart of life, vital for all biological systems, from the smallest microorganisms to the largest mammals, and is often referred to as “*the nature’s liquid gold*”, thanks to its role in maintaining health and biodiversity [Popkin et al., (2011); Green. Earth, (2023)]. However, despite its importance, it faces serious threats as a dreadful global risk [Ganter, (2015)]. According to the reports of WWC and the joint program of WHO and UNICEF, 1.2 billion people lack reliable

access to safe and clean water; 4 billion people live in regions where water is in short supply; while 1.8 million people die annually from diarrheal diseases¹ (90% of them being children under the age of 5) [WWC, (2021); WHO and UNICEF, (2025); Ritter, (2019)]. And the future? It looks grim. Calculations for 2050, depict that nearly 6 billion people will face severe water shortages [Boretti and Rosa, (2019); Cusick and E&E News, (2022); UNESCO, (2018)]. Contaminated water is also, linked to 80% of illnesses and 50% of deaths, rendering it not just a simple environmental problem, but a deadly one [Anderson, (2025); Liu et al., (2012); Dagdag et al., (2023); Vardhan et al., (2019); Lin et al., (2022)]. Therefore, the question as to why this happens and how to prevent it, arises. A lot of it comes down to the combined effects of urbanization, rapid industrial and population growth, which in turn put a massive pressure on freshwater resources and by extension the ecosystems that depend on them [Kędziora, (2025); Bigas et al., (2012); Dagdag et al., (2023)]. Yet, regrettably, that's not "the whole story"; untreated sewage, industrial spills, and agricultural runoff all pile on, amplifying the negative situation [Vardhan et al., (2019); Bhatnagar and Sillanpää, (2010)], consequently the urgency of this crisis is hard to exaggerate.

The terms "**water** and **soil pollution**" define the presence of unwanted foreign harmful substances (which can be inorganic/organic, biological and radiological in nature), accumulating to critical levels that degrade the quality of aquatic and terrestrial environments [Banaras Hindu University, (2025); Rodríguez-Eugenio et al., (2018); James, (2003); Münzel et al., (2022)]. As such, these substances (whether pharmaceuticals, pesticides, plastic debris, or especially *heavy metals*), disrupt the ecological balance and pose a serious adverse effect for the public health, flora and fauna [Dagdag et al., (2023); Vardhan et al., (2019), Memedi et al., (2016a); Briffa et al., (2020)]. **Heavy metals** is a broad term, encompassing all metallic (including metalloids), non-degradable, inorganic chemical elements that are characterized by a relatively high density (i.e., five times that of water), high atomic mass (>5 and >20 g/cm³, respectively) and having toxic tendencies even at low concentrations [Darwesh and Matter, (2021); Edelstein and Ben-Hur, (2018); Dehari et al., (2022); Briffa et al., (2020); Filipoiu et al., (2022)]. They occur naturally in the environment, when the "parent" material igneous or sedimentary rock, is weathered by processes such as erosion, volcanic eruptions or soil formation [Alengebawy et al., (2021)]. It is crucial to mention that although not classified as heavy metals due to their lower atomic densities, the presence of alkaline earth metals such as calcium and magnesium (Ca-1.55 g/cm³ and Mg-1.738 g/cm³), in the environment and biological systems are critical for maintaining overall health and ecological balance, as imbalances or deficiencies can lead to various health issues [Filipoiu et al., (2022); Witkowska et al., (2021); Hanusa, (2019)].

¹Contaminated water "harbors" bacteria, viruses and infections that can cause gastrointestinal infections leading to diarrhea, which in turn is the third deadly leading cause for vulnerable groups (pediatric and geriatric patients amongst others) [WHO, (2024)].



Scheme 1. General classification of heavy metals, including calcium and magnesium as competitors for adsorption sites [Filipoiu et al., (2022); Witkowska et al., (2021)] *heavy metals of interest along with alkaline earth metals are indicated in bold typeface

Conventional (traditional) **water** treatment (including chemical precipitation, ion-exchange systems, membrane-based processes and electrochemical methods) technologies remove adequately heavy metals from polluted water; however, they are often faced with the disadvantages deriving from their costs, energy consumptions, and the unintended generation of secondary pollutants [Gahrouei et al., (2024); Saravanan et al., (2021); Memedi et al., (2017)]. Similar challenges are encountered in the removal of heavy metals from *soils*, where conventional methods (such as excavation, soil washing, and chemical stabilization) even though often employed, remain expensive, and can permanently damage soil quality [Rajendran et al., (2022); Sharma et al., (2018)]. In light of these flaws, researchers are turning to greener, more economical treatment (remediation) strategies that synchronically lower metal concentrations effectively, without introducing new environmental hazards. As such, the present study critically evaluates naturally occurring alternatives (clinoptilolite-based natural zeolite, montmorillonite-rich bentonite, and common carbonate mineral-calcite), and their capability in removing heavy metals. By synthesizing and bridging current research, it focuses on their physicochemical properties, adsorption mechanisms, and key factors influencing the sorption effects. Because after all safeguarding the purity of water is not just an act of environmental stewardship, but also a necessity to guarantee public health.

Table 1. Maximum permissible limits for selected heavy metals in *water* according to WHO and US-EPA guideline values; their major resources, associated toxicities and ideal targets (including essential minerals Ca and Mg) [ref. for 1-5: Jaleel et al., (2001); Dwivedi et al., (2021); Angon et al., (2024); Jyothi, (2020); Moosavirad et al., (2015); Singh, (2021); Sankhla et al., (2018); US-EPA (2023); WHO (2022); Oziegbe et al., (2025); Sokan-Adeaga et al., (2021); Balali-Mood et al., (2021); ref. for 6-7: Hanusa, (2019); US-EPA (2023); WHO (2022)]

No	WHO GUIDELIN E VALUE IN WATER (mg/L)	US-EPA GUIDELIN E VALUE IN WATER (mg/L)	MAJOR RESOURCE S	KEY TOXIC EFFECTS IN HUMAN BODY	COMMO N IDEAL TARGET IN WATER (mg/L)
SELECTED HEAVY METALS					

1.	Lead (Pb)	0.01 mg/L	0.015 mg/L (action level)	Aircraft engines, batteries, coal combustion, paint industry, traffic emission, smoking	Infants and children: delays in physical or mental development Adults: kidney problems, hypertension	≤ 0.01 mg/L
2.	Cadmium (Cd)	0.003 mg/L	0.005 mg/L	Plastic, fertilizers, pesticides, Ni-Cd batteries, PVC products	Kidney damage, bronchitis, gastrointestinal disorders, cancer	≤ 0.003 mg/L
3.	Zinc (Zn)	3-5 mg/L (aesthetic value)	not established	Oil refining, plumbing, brass manufacturing, galvanization	Zinc fumes have corrosive effect on skin, Cu metabolism interference	≤ 1.0 mg/L
4.	Copper (Cu)	2.0 mg/L	1.3 mg/L	Copper polishing, electronics waste, mining	Failure of brain and kidneys, severe anemia, intestinal irritation	≤ 1.0 mg/L
5.	Nickel (Ni)	0.07 mg/L	not established	Electroplating, zinc base casting, battery industries, fertilizers	Immuno-toxic, neurotoxic, genotoxic, hepatotoxic, rapid hair fall	≤ 0.02 - 0.05 mg/L
SELECTED ALKALINE EARTH METALS (essential minerals → no-health based WHO/EPA limits)						
6.	Calcium (Ca)	no health-based limit (typical 40-80 mg/L)	not regulated	Calcite, Limestone, Gypsum, fertilizers, cement	RARE Excess → water hardness, affecting plumbing or taste	50-200 mg/L

7.	Magnesium (Mg)	no health-based limit (typical 20-30 mg/L)	not regulated	Dolomite, Magnesite, fertilizers, industrial effluents	RARE Excess → mild laxative in sensitive individuals	30-150 mg/L

2. Methodology

This review was conducted using a systematic and structured research by compiling and analyzing existing knowledge from previously published literatures in order to ensure a reliable comparison between clinoptilolite (natural zeolite), montmorillonite (bentonite), and calcite (carbonate mineral) as effective “agents” in removing (adsorbing) heavy metals from water. Scientific literature published between approximately 2000 and 2025 was collected from trusted and well-established sources (databases) like Google Scholar, ScienceDirect, PubMed, and Scopus. In addition to peer-reviewed articles reporting experimental adsorption data (such as the effects of operational parameters on adsorption capacity), relevant books (Manning, (1995); Ciullo, (1954) are worthy mentions), monographs and doctoral dissertations were also included to strengthen the theoretical foundation of the review; while papers analyzing non-aqueous systems were excluded. In total, around 90-200 relevant studies were analyzed, with data gathered in English (mainly), Turkish, Albanian, Spanish and Macedonian language. Keywords like “zeolite”, “natural zeolite”, “clinoptilolite”, “bentonite”, “montmorillonite”, “carbonate minerals”, “calcite”, “heavy metal adsorption by natural minerals”, “water treatment by minerals”, “green chemistry” were used in various combinations. In addition, specific search phrases like “zeolite and carbonate minerals for removal of heavy metals from water”, were also used to zero in on studies that directly addressed water remediation. The extracted data was evaluated and systematically organized to produce a comparative review, with a focus on physicochemical properties, adsorption mechanisms, factors affecting the latter, which in turn enabled a critical assessment of the advantages, disadvantages, and operation differences among targeted minerals in heavy metal remediation of water.

3. Comparative recapitulation of structure, composition and adsorption mechanisms of clinoptilolite (CL), montmorillonite (MTT), calcite (CaCO₃)

3.1. Why were these three minerals chosen?

Clinoptilolite (CL), montmorillonite (MMT), and calcite (CaCO₃) are part of three fundamentally different mineral families. As such, their structures, chemical makeups differ in every aspect and each of them remove heavy metals in their unique ways (which will be thoroughly elaborated in the following sections). In terms of structure, clinoptilolite represents a microporous aluminosilicate, known as “the classic natural zeolite”; montmorillonite, on the other hand, is the predominant bentonite, with a 2:1 layered aluminosilicate structure that is characterized by swelling; while calcite in contrast is the prevalent most stable polymorph of calcium carbonate [Velde, (1977); Zendelska et al., (2018); Kumari and Mohan, (2021); Comadran-Casas et al., (2025)].

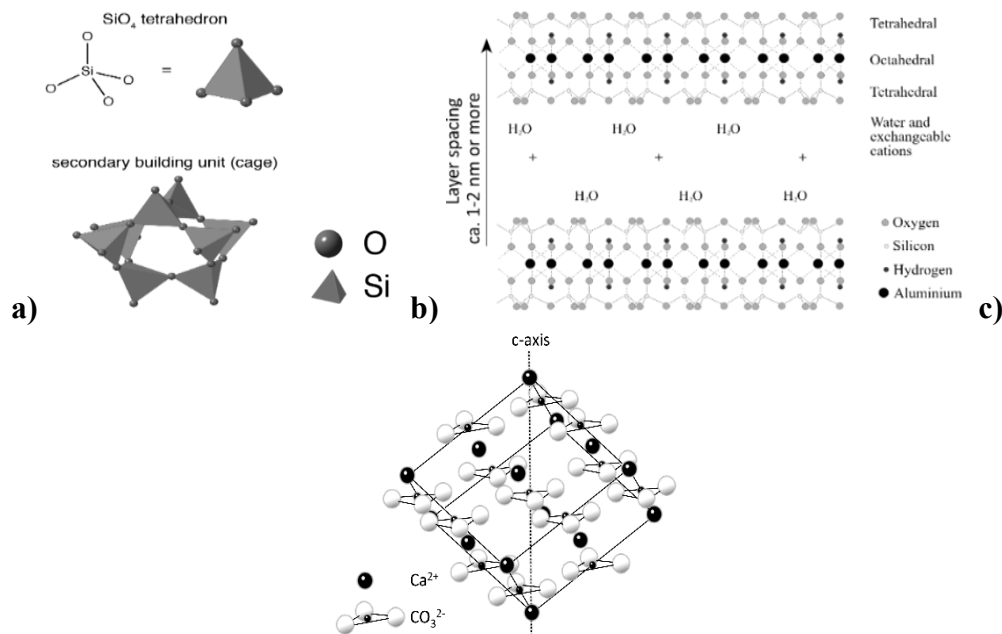


Figure 1. Symbolic representations of the crystal structures of **a)** natural zeolite, **b)** bentonite and **c)** calcite [a) courtesy of Coudert, (2020); b) Tempe and Itub, (2007); c) Tempsu, (2015) licensed under CC BY-SA 4.0²]

3.2. Structure and composition

3.2.1. Natural zeolites: clinoptilolite (CL)

Clinoptilolite (from the Greek words: κλίνω and *περὶ* πλῶν, meaning “oblique feather stone”) represents the most widespread naturally occurring pozzolana among natural zeolites (“*molecular sieves*”) [Ortega et al., (2000); Grifasi et al., (2024)]. It forms when volcanic ash (tuff) transforms under-low temperatures (60-110°C) [Iijima, (2007); Baydemir, (2022)], as well as, in environments with slow sedimentation through the reaction between basaltic glass and water, resulting in “nearly pure” zeolite [Zendelska et al., (2018); Güvenir, (2005); Grifasi et al., (2024); Rodríguez-Iznaga et al., (2022)]. The crystal lattice consists of a maze-like framework, interconnected of channels and cages (with diameters ranging from 3 to 5 Å) [Pabiś-Mazgaj et al., (2021); Cerri et al., (2016)], that are built from corner-sharing $[\text{SiO}_4]^{4-}$ and $[\text{AlO}_4]^{5-}$ tetrahedra units. It is important to note that the substitution of Si^{4+} with Al^{3+} creates a partially negative charge deficiency in the structure, that has to be compensated by the presence of extra-framework cations (like Na^+ , K^+ , and Ca^+) [Grifasi et al., (2024); Kinoti et al., (2022)]. The internal voids of CL, are occupied by water molecules and exchangeable cations, which show significant “flexibility” (mobility) [Bogdanov et al., (2009); Ambrozova et al., (2017); Kowalczyk et al., (2006)]. Consequently, these cations can be easily exchanged with other ions, which is one of the reasons why the mineral has such a high ion-exchange capacity. Additionally, it is porous with rigid structure (unlike *swelling silicates*), and zeolitic water can be reversibly removed by heating without affecting the framework, making it dimensionally stable [Velarde et al., (2023); Król, (2020); Breck, (1974); Zendelska et al., (2018)]; while, in comparison to *carbonate minerals*, clinoptilolite is chemically more resistant to acid dissolution (does not “effervesce”/break down) [Tsitsishvili et al., (2023); Curi et al., (2006)]. Even though morphologically, it closely resembles heulandite (HEU) (“flake”, sheet-like aggregate), they are classified as isostructural minerals,

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because of CL's higher silica content, abundance of alkali elements (*refer to table 2*) and thermal stability (HEU: 400°C; CL: up to 750°C, respectively) [Dimowa and Tzvetanova, (2021); Pabiš-Mazgaj et al., (2021)]. X-ray diffraction of CL, reveal its complex framework through sharp peaks, with some broader reflections hinting at minor amorphous phases [Güvenir, (2005); Cekova et al., (2014); Breck, (1974); IZA Commission of Natural Zeolites (2025); Manning, (1995); Kogel et al., (2006); Ciullo, (1954); Chatterjee, (2009)].

Table 2. Chemical composition of natural zeolite: clinoptilolite [Petrakakis et al., (2007); Markovska et al., (2017); Faghihian et al., (2009); Alexa-Stratulat et al., (2024); mindat.org; handbookofmineralogy.org]

OXIDE	SiO ₂	Al ₂ O ₃	K ₂ O	CaO	Fe ₂ O ₃	Na ₂ O	MgO	Trace	H ₂ O (till 600°C)	Loss on ignition (LoI)
% w/w	68.75- 71.3	11.35- 13.10	3.17- 3.40	2.86- 5.20	1.90- 2.10	0.82- 1.30	1.18- 1.20	4.6	12.90	8.73- 8.86

Si/Al ratio: 5

3.2.2. Bentonites: montmorillonite (MMT)

Montmorillonite (named after the French town where it was first discovered-Montmorillon) [Uddin, (2018)], forms part of the smectite subclass, a soft phyllosilicate mineral [Sejpal, (2025)], which, forms through the hydration and chemical transformations processes of volcanic ash, along with rocks rich in aluminum and magnesium, which in turn, result in microscopic crystals known as clay [Special Chem, (2024); Ross and Hendricks, (1944); Uddin, (2018)]. Being a 2:1 layered mineral, it consists from an octahedral sheet that is placed in a tetrahedral-octahedral-tetrahedral (TOT) arrangement formed by two tetrahedral sheets, known as “sandwich-with-filling like structure” (distances in-between 9.4 and 7 Å) [Babaami, (2024); Silva et al., (2012)]. Unlike *zeolites* that have a constant charge throughout the crystalline structure, MMT exhibits a layer charge which happens thanks to the isomorphic substitutions that occur within the sheets [Wang et al., (2017); Penaloza, (2019)]. The substitutions of Mg²⁺ for Al³⁺ in the octahedral sheet or Al³⁺ for Si⁴⁺ in the tetrahedral sheet cause a net negative charge that is balanced by the presence of Na⁺ or Ca²⁺ cations [Penaloza, (2019); González Rodríguez, (2018)]. Considering that the individual crystals are not tightly bond, the water molecules that exist in the “environment”, can be inserted between the layers and “intervene” [Segad et al., (2010); Bhattacharyya and Gupta (2008)]. This phenomenon, increases the stacking distances, which expand the surface, thus giving rise to the typical swelling behavior of smectites, supporting the high-cation exchange capacity (CEC) of montmorillonite [Harrou et al., (2025); Penaloza, (2019)]. On the contrary, *calcite* is dimensionally stable in aqueous environments, but shows negligible swelling, due to its rigid, three-dimensional crystal lattice [Geissbühler et al., (2004)]. Morphologically, montmorillonite has flake-like crystals [Kumari and Mohan, (2021)], whereas, its X-ray diffraction patterns are majorly characterized by broad and poorly defined peaks, reflecting fine particle size and structural disorder [Nurulla qizi, (2025); Ali-Fil et al, (2014); Manning, (1995); Kogel et al., (2006); Ciullo, (1954); Chatterjee, (2009)].

Table 3. Chemical composition of bentonite: montmorillonite [Halder et al., (2017); Ali-Fil et al., (2013); Jemai et al., (2024), mindat.org; handbookofmineralogy.org]

OXIDE	SiO ₂	Al ₂ O ₃	K ₂ O	CaO	Fe ₂ O ₃	Na ₂ O	MgO	Trace	H ₂ O	Loss on ignition (LoI)
% w/w	49.40-63.02	19.70-21.08	0.04	0.65-1.50	0.30-3.25	1.50-2.57	0.27-2.67	0.72	25.67	5.64

Si/Al ratio: 2-4

3.2.3. Carbonate minerals: calcite

Calcite (from German *Calcit*, meaning lime) [Vale, (2020)], is a substantial carbonate mineral (CaCO₃). It also presents the most abundant mineral on Earth, that forms by precipitation processes from ground and surface waters [WGNHS, (2020); Gao et al., (2017)]. Without calcite, many of Earth's organisms would cease to exist, on account of it constructing the shells and “hard parts” of most invertebrate organisms [University of Minnesota, (2025); Skinner and Jahren, (2007)]. Its crystal lattice is simple ionic, with a “straightforward” rhombohedral system, where planar CO₃²⁻ groups coordinate with Ca²⁺ ions to form a three-dimensional lattice, differing it from aluminosilicate minerals like *zeolite* and *smectite* [Skinner and Jahren, (2007); Aquilano et al., (2016)]. It is non-porous in nature, does not have a charged framework or layers, hence its interactions in water systems are mainly dominated by equilibrium between dissolution and precipitation, or acid-base reactions, instead of ion exchange [Subhas et al., (2017); Woltheras et al., (2012)]. Morphologically, it exhibits well-defined crystalline features, and its X-ray diffraction patterns show sharp, clear reflections that are typical characteristic of simple carbonate lattice [Febrida et al., (2021); Zubovic et al., (2025); Manning, (1995); Kogel et al., (2006); Ciullo, (1954); Chatterjee, (2009)].

Table 4. Chemical composition of carbonate mineral: calcite [Katircioglu-Bayel, (2020); mindat.org; handbookofmineralogy.org]

CONSTITUENT	CaCO ₃	MgCO ₃	Fe ₂ O ₃	SiO ₂	Al ₂ O ₃	Loss on ignition (LoI)
% w/w	99.5	0.2	0.01	0.01	0.02	0.26

The following table covers the most important differences between these three minerals, presenting them “side-by-side” with the purpose to reveal the major differences in their physical and chemical properties which are of interest for their affinity as “agents” for heavy metal removal. The table is a compilation from multitude of different literatures.

Table 5. Comparative recapitulation side-by-side of key properties between clinoptilolite, montmorillonite, and calcite [ref. for clinoptilolite (zeolite): Abdel-Rahim, (2017); Paliulis, (2016); Erdem et al., (2004); Minceva et al., (2008); IZA Commission on Natural Zeolites, (2025); Hakim et al., (2024); Abdelwahab and Thabet, (2023); Velarde et al., (2023); Ahari et al., (2021); Rodríguez-Iznaga et al., (2022); Grifasi et al., (2024); Senila and Cader, (2024); ref. for montmorillonite (bentonite): Yaghmaeiyan et al., (2022); Golubeva et al., (2023); Said et al., (2015); Khalid et al., (2016); González Rodríguez, (2018); Toğay Eskitoros et al., (2024); Randelović et al., (2011); Bonto et al., (2022); ref. for carbonate mineral (calcite): Asghari et al., (2025); Baker et al., (2017); Groenendijk & van Wunnik, (2021); Smithsonian Natural Museum of Natural History, (2025); ref. for all three: Manning, (1995); Kogel et al., (2006); Ciullo, (1954); Chatterjee, (2009); mindat.org; handbookofmineralogy.org]

No	PROPERTY	CLINOPTILOLITE (ZEOLITE)	MONTMORILLONITE (BENTONITE)	CALCITE (CARBONATE MINERAL)
1.	Crystal structure/ Color	Framework zeolite structure composed of SiO ₄ and AlO ₄ tetrahedra forming channels and cavities. Trigonal to monoclinic symmetry, often showing fibrous or tabular crystals. Channels allow water and cation movement/ White	2:1 layered structure (TOT) with weak van der Waals forces between layers. Part of the smectite group, expandable interlayers allow hydration and swelling. Monoclinic symmetry/ Beige, white to gray	Trigonal/hexagonal lattice system. Dense, non-porous crystals/ Typically, white or colorless
2.	Building blocks	SiO ₄ and AlO ₄ tetrahedra form a continuous framework. Exchangeable cations (Na ⁺ , K ⁺ , Ca ²⁺) and water molecules reside in structural channels	Silica tetrahedral sheets sandwich Al ³⁺ /Mg ²⁺ octahedral sheets. Interlayer cations stabilize the structure. Layers can absorb water, causing swelling	Calcium ions are coordinated with carbonate ions (CO ₃ ²⁻) in a dense ionic lattice. Lattice is highly stable and non-expandable
3.	Cation Exchange Capacity (CEC)/ Adsorption properties	High cation exchange capacity (CEC≈1.6–2.0 MEq/gm). Exchanges Na ⁺ , K ⁺ , Ca ²⁺ , Mg ²⁺ efficiently. Adsorbs heavy metals, ammonium, and some organics	Very high CEC (80–150 MEq/100gm). Swelling capacity enhances cation exchange. Exchanges Na ⁺ , Ca ²⁺ , Mg ²⁺ . Adsorbs water and polar molecules	Very low CEC. Minimal exchangeable ions. Can release Ca ²⁺ under acidic conditions. Adsorption negligible
4.	Physical properties	Hardness 3.5–4.0 (Mohs), brittle, dense, porous (34%), low bulk	Soft, plastic, low hardness 1–2 (Mohs), high porosity. Swells up to 2–3 times its dry	Hardness 3 (Mohs), insoluble in water. Moderate thermal stability, decomposes

		density due to channels. Thermally stable up to $\approx 400^{\circ}\text{C}$, retains structure during hydration/dehydration cycles	volume upon hydration. Sensitive to temperature and drying	at $>800^{\circ}\text{C}$ ($\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$)
5.	Functional properties	“ <i>Molecular sieve</i> ”, ion-exchange, selective adsorption of ions and gases. Stabilizes water pH and removes ammonium/heavy metals	Swelling clay. Used for drug carriers, soil stabilization, and catalysts	Filler, pH regulation in soils, cement, and lime production, and biomineralization support
6.	Water incorporation	Zeolitic water in channels	Interlayer water	No bound (structural) water
7.	Water adsorption/ Moisture content	15-25 wt%	20-30 wt%	0-5 wt%
8.	Main exchangeable ions	Na^+ , K^+ , Ca^{2+} , Mg^{2+} in channels	Na^+ , Ca^{2+} , Mg^{2+} interlayer	Ca^{2+} only (lattice cation, not exchangeable except by dissolution/precipitation)
9.	Hydration/ Thermal behavior	Moderate hydration, water can be removed and reabsorbed without structural collapse	High water absorption. Dehydration leads to shrinkage, stable up to $\approx 200\text{--}300^{\circ}\text{C}$	Minimal hydration, highly stable under environmental conditions
10.	Environmental/ Chemical stability	Resistant to neutral and mildly acidic conditions, channels allow reversible cation exchange	Sensitive to acidic environments, easily dispersible in water, prone to structural changes under extreme pH or high temperature	Chemically stable in neutral and basic environments, reacts with acids, releasing CO_2
11.	Typical pore size	Micropores ($\approx 3\text{--}5 \text{ \AA}$ channels)	Interlayer spacing variable ($9\text{--}7 \text{ \AA}$ typical, expands when hydrated)	Non-porous crystal faces; meso/microporosity only if aggregate/porous
12.	Density/ Bulk density	$2.1\text{--}2.3 \text{ g/cm}^3$	$2.2\text{--}2.5 \text{ g/cm}^3$	2.7 g/cm^3

13.	Applications	Water purification, wastewater treatment, heavy-metal removal, ammonia capture, gas adsorption, animal feed additive, environmental remediation	Soil conditioning, pollutant adsorption (heavy metals, dyes), drilling fluids, pharmaceuticals, catalysts, nanocomposite materials	Cement and construction material, lime production, soil amendment (neutralization), carbon sequestration studies, chemical industry filler
14.	Strengths	Highly selective ion exchange for divalent heavy metals (solutions with many competing ions-real wastewater), robust, widely available and shows repeatable regeneration	Very high capacity via interlayer and surface adsorption, versatile to modification (organo-, pillared, cation form)	Can remove metals by precipitation/co-precipitation; low cost, and often acts as a pH buffer
15.	Limitations	Often needs pre-treatment for best capacity; microporosity limits large complexes. Limited pore size for bulky ions	Swelling (reduces stability) and colloidal behavior can complicate use, requiring separation/regeneration (instability in water)	Often removed via precipitation (depends on aqueous chemistry), dissolution in acidic conditions
16.	Implications on adsorption	Ideal for long-term and engineered filter-systems	Best suited for systems where swelling can be tolerated or controlled	Passive remediation of acidic metal-rich waters

4. Heavy-metal absorption mechanisms, selectivity and factors controlling adsorption

Adsorption is the process in which adsorbates from a gas (*g*) or liquid (*l*) phase adhere (“attach”) to the surface of an adsorbent which generally is a solid phase (*s*) [Saleh, (2022)]. Two main types are physical adsorption (physisorption), a reversible process which uses van der Waals forces (i.e. weak chemical bonds); and chemical adsorption (chemisorption), which uses stronger chemical bonds (either covalent or ionic), consuming the material from adsorbent surfaces, making it an irreversible permanent process [Rashed and Palanisamy, (2018)]. Accordingly, the removal of heavy metals from aqueous solutions is mostly controlled by the chemical and structural properties of the sorbent [Memedi et al., (2016b)]. Based on their characteristics, zeolites and clays favor ion exchange and adsorption processes, while carbonates facilitate precipitation-based removal (which will be discussed in detail in the following sections) [Pérez-Botella et al., (2022); Profeta et al., (2025); Orucoglu et al., (2022)].

4.1. Heavy-metal adsorption mechanisms; selectivity (preference order) and factors controlling adsorption

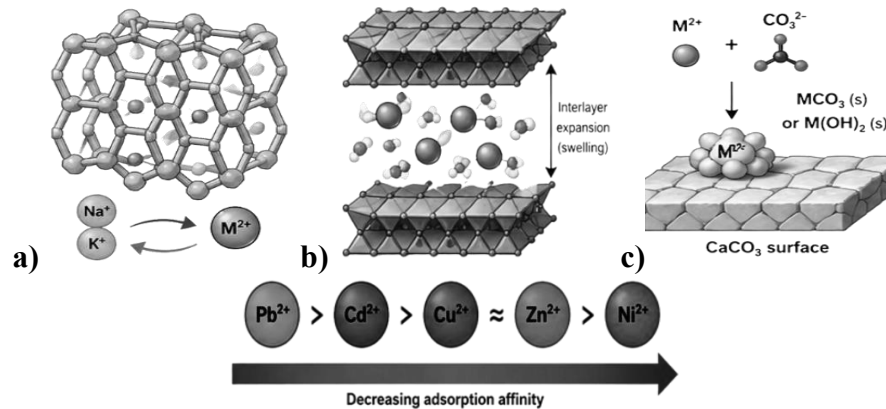
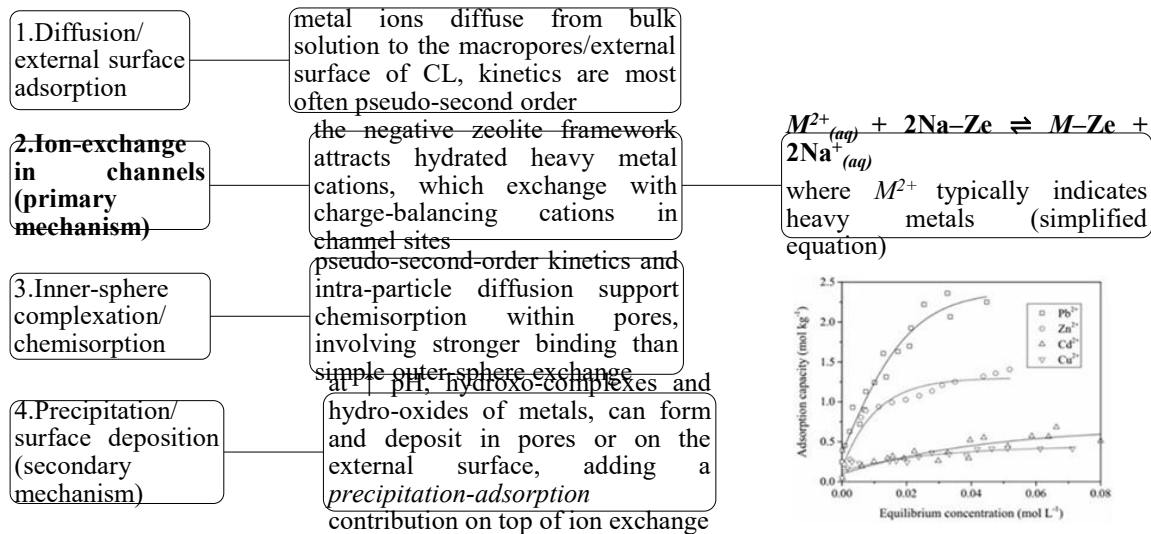


Figure 2. Illustration exhibiting the primary methods used by mineral-based adsorbents to eliminate heavy metals from water sources: a) natural zeolite (clinoptilolite)-selective ion-exchange ; b) bentonite (montmorillonite)-high-selectivity through interlayer uptake and surface complexation of its expandable (swelling) layered structure; c) carbonate mineral (calcite)-surface-induced precipitation, pH-dependent process (up) and general selectivity order with fluctuations depending on different factors (down) [illustration adapted from Velarde et al., (2023); Wang et al., (2024); Fathy et al., (2025) and redefined by authors]

4.1.1. Natural zeolites: clinoptilolite (CL)

a) Heavy-metal adsorption mechanism → ion-exchange



Scheme 2. and Figure 3. Main characteristics of the mechanism and adsorption isotherm of heavy metals on CL at 25.05°C [Velarde et al., (2023); Sprynskyy et al., (2006); Dosa et al., (2022); Mihaly-Cozmuta et al., (2014); Erdem et al., (2004); Restiawaty et al., (2024); Sabadash et al., (2020); the isotherm courtesy of Solinhin et al., (2024) licensed under CC BY-SA 4.0]

b) Heavy metal selectivity (preference order) and factors controlling adsorption

Metal preference depends on various factors including but not limited to: pH, zeolite form (ex. Na-, Ca- ...), solution composition, initial metal concentration, contact time, adsorbent dose,

competing ions, surface modification [Velarde et al., (2023); Mihaly-Cozmuta et al., (2014); Kocaoba et al., (2007); Salih et al., (2021); Ávila et al., (2025)]. However, the typical preference sequence according to various research papers, recapitulates as the following: $Pb^{2+} \gtrsim Cu^{2+} > Cd^{2+} \gtrsim Zn^{2+} > Ni^{2+}$ with fluctuations (variations) depending on minerals pre-treatment, background ions, and pH (amongst others) [Sprynskyy et al., (2006); Mihaly-Cozmuta et al., (2014); Erdem et al., (2004)].

Table 6. Selected factors and their importance in affecting heavy metal removal by CL [Velarde et al., (2023); Mihaly-Cozmuta et al., (2014); Aziz et al., (2021); Salih et al., (2021); Ávila et al., (2025); Zanin et al., (2017); Kocaoba et al., (2007); Galleti et al., (2020); Sprynskyy et al., (2006); Pandey et al., (2020); Lim et al., (2019); Erdem et al., (2004); Munir et al., (2024); Mier et al., (2001)]

No.	FACTOR	EFFECT ON ADSORPTION
1.	pH 1. 1-2 2. 5-7 3. higher	1. competition from H^+ , partial framework dissolution = $\downarrow$ uptake 2. <i>optimal removal, heavy metals largely soluble, but H^+ competition is reduced</i> $\uparrow$ pH (until hydroxide precipitation) = $\uparrow$ selectivity and uptake 3. precipitation becomes important, clinoptilolite acts as ion exchanger and support for hydroxide precipitates
2.	Initial metal concentration	$\uparrow$ concentration = $\uparrow$ capacity until site saturation; affects selectivity patterns
3.	Contact time	fast initial adsorption; equilibrium within 60-180 min in many systems
4.	Clinoptilolite form	synthetic (purified) CL > capacity than impure natural materials because of higher zeolite content and access to channels
5.	Adsorbent dosage	> dose = $\uparrow$ higher % removal
6.	Physicochemical properties of metal ions	1. hydration energy and hydrated radius: ions that dehydrate more easily = $\downarrow$ hydrated radii $\rightarrow$ penetrate channels and bind more strongly 2. charge density and electron configuration: $\uparrow$ field strength and accessible <i>d</i> -orbitals enhance inner-sphere coordination to framework oxygens, contributing to selectivity
7.	Particle size and diffusion	finer particles exhibit $\uparrow$ capacities because of $\uparrow$ external surface area, shorter diffusion paths and $\uparrow$ proportion of zeolitic phase
8.	Co-contaminants	the mineral loses efficiency when Cr (VI) or complexing ligands are present
9.	Temperature	some systems are slightly endothermic (Cd^{2+} , Ni^{2+}), while others are exothermic (Pb^{2+}) overall: most show spontaneous behavior (negative ΔG°)

- **Brief notes on calcium and magnesium on clinoptilolite**

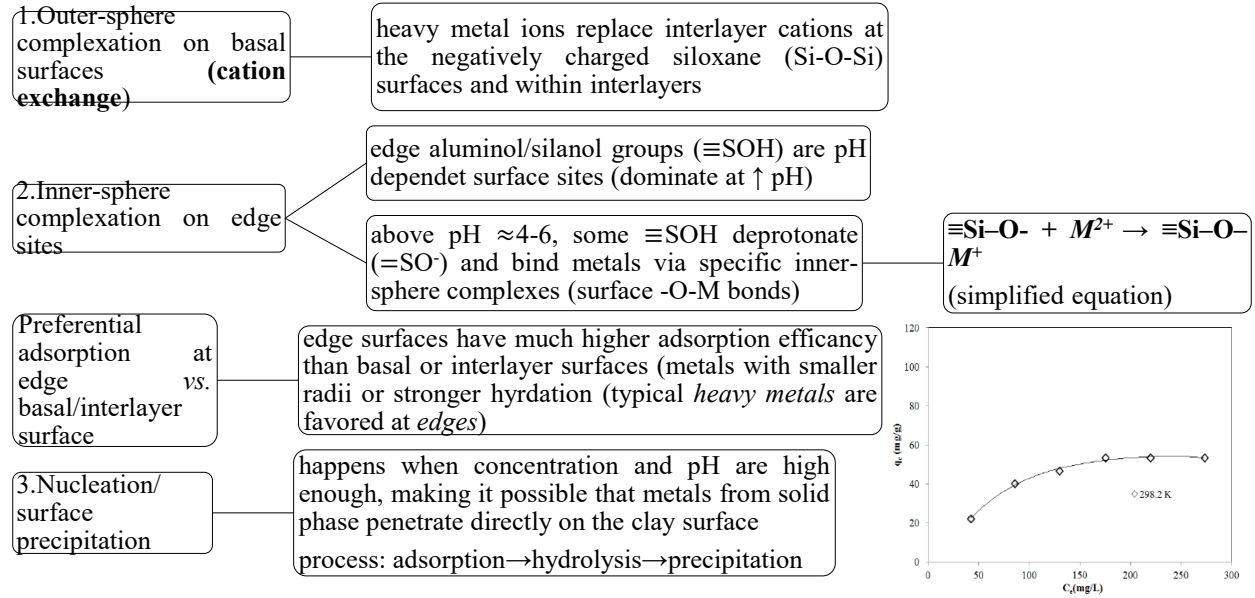
CL reacts with Ca^{2+} and Mg^{2+} for hardness removal and as exchangeable native framework cations. For hardness removal, CL achieves Ca^{2+}/Mg^{2+} uptake up to ≈ 450 mg/g (adsorption is favored at pH-mildly acidic to neutral, approx. ≈ 6) [Nasief et al., (2021)]. On the other hand, when Ca^{2+} and

Mg^{2+} are used for removal of heavy metals, they appear as released counter ions (which typically happens when the mineral adsorbs Cu^{2+}) [Wang et al., (2019)].

4.1.2. Bentonites: montmorillonite (MMT)

a) Heavy-metal adsorption mechanism → dominant: cation exchange

The mechanism of adsorption involves three concurrent pathways: a) cation exchange, b) surface complexation on basal and edge sites and c) nucleation/surface precipitation (given in scheme 3) [Li et al., (2023); Zhu et al., (2016)].



Scheme 3. and Figure 4. Main characteristics of the mechanism and adsorption isotherm of heavy metals on MMT at 25.05°C [Gu et al., (2010); Abollino et al., (2003); Ijagbemi et al., (2008); Liu et al., (2021); Wang et al., (2024); Liu et al., (2022); the isotherm adapted from Umpuch and Sakaew, (2013)]

b) Heavy metal selectivity (preference order) and factors controlling adsorption

The selectivity (preference order) is controlled by various physicochemical factors, with *pH* being the dominant principal parameter because at the same time it governs metal speciation in solution, protonation-deprotonation of edge sites, and competition between protons and metal cations for adsorption sites [Tournassat et al., (2015)]. Nevertheless, the trend of the metal selectivity sequence frequently exhibits the following sequence (approx.): $Pb^{2+} \gtrsim Cu^{2+} \gtrsim Cd^{2+} \gtrsim Zn^{2+} > Ni^{2+}$, even though the order can change depending on ionic strength, competing ions and surface modifications [Gu et al., 2010].

Table 7. Main factors affecting heavy metal removal by MMT [Ma et al., (2019); Shi et al., (2022); Abollino et al., (2003); Gu et al., (2010); de Pablo et al., (2011); Chen et al., (2015); Yang and Yang, (2023); Zhang et al., (2022); Liu et al., (2021); Al-Musawi et al., (2024); Boahen et al. (2023)]

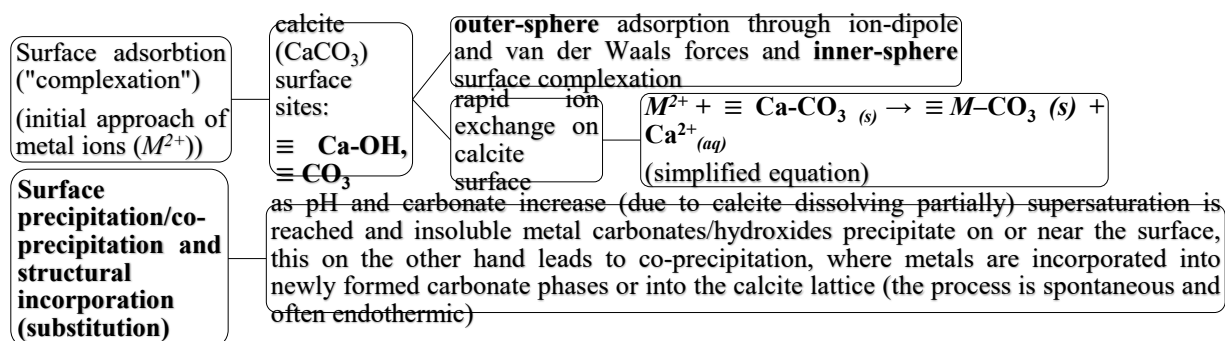
No.	FACTOR	EFFECT ON ADSORPTION
1.	pH (optimal is <i>usually below</i> 7)	↓ pH = reduces adsorption by protonating edges ↑ pH = promotes deprotonation, inner-sphere complexation and sometimes precipitation
2.	Surface sites (basal vs. edge)	basal site favor outer-sphere, ionic-strength-dependent adsorption edge sites favor inner-sphere complexes and much stronger binding
3.	Initial metal ion concentration	↑ in initial metal ion C_0 = ↓ in adsorption percentage but ↑ in adsorption capacity until active sites approach saturation
4.	Contact time	↑ adsorption percentage and capacity with longer contact time between the adsorbent and the solution until equilibrium
5.	Adsorbent dosage	↑ adsorbent dosage = ↓ adsorption capacity, but an increase in the overall removal efficiency of heavy metals
6.	Charge density/heterogeneity	↑ layer charge = ↑ metal uptake
7.	Ionic strength	at ↓ pH, adsorption on $\equiv X^-$ strongly ionic-strength dependent at ↑ pH (edge-site binding) less dependent
8.	Background ions	compete for exchange sites, change apparent capacities
9.	Surface area/porosity	↑ specific surface and enhanced mesoporosity ↑ capacity and mass transfer
10.	MMT modification	↑ surface area, adds functional groups, enhances capacity and selectivity
11.	Temperature	often modest effect, some systems endothermic (capacity increases with T), others exothermic; overall generally negative (ΔG°)

- **Brief notes on calcium and magnesium on montmorillonite**

The water uptake and swelling behavior of montmorillonite and the aggregation of its particles are controlled by Ca^{2+} and Mg^{2+} which at the same time determine the clay's ability to absorb pollutants. The presence of calcium results in stronger particle aggregation while magnesium creates conditions that enable the formation of highly hydrated interlayers which contain water [Li et al., (2024); Ruiz et al., (2024)].

4.1.3. Carbonate minerals: calcite (CaCO_3)

a) Heavy-metal adsorption mechanism → precipitation and co-precipitation



Scheme 4. Main characteristics of the mechanism of adsorption of heavy metals by CaCO_3 [Song et al., (2020); Fathy et al., (2025); Rangel-Porras et al., (2010); Lee, (2024); Wang et al., (2021); Gibert et al., (2021); Azanfire et al., (2025); Jacob et al., (2018); Naseer et al., (2024); Jin et al., (2024)]

b) Heavy metal selectivity (preference order) and factors controlling adsorption

Calcite (CaCO_3) shows adsorption “motifs” that are highly metal-specific. The most accurate selectivity sequence for divalent “target” heavy metals (as reported in literature) is the following: $\text{Pb}^{2+} > \text{Cu}^{2+} \gtrsim \text{Cd}^{2+} > \text{Zn}^{2+} > \text{Ni}^{2+}$ [Zachara et al., (1991); Fathy et al., (2025)]. The data indicates that this happens because Pb^{2+} has low solubility, which leads to strong $\text{Ca}^{2+}/\text{Pb}^{2+}$ exchange and precipitation on CaCO_3 surfaces, while, Cu^{2+} and Cd^{2+} , after their initial adsorption, dehydrate fast [Zachara et al., (1991); Fathy et al., (2025); García-Sánchez and Álvarez-Ayuso, (2002)], whereas Zn^{2+} and especially Ni^{2+} show the highest level of “flexibility”-mobility, as a result of the fact that they stay hydrated as they create surface complexes with less bonding strength, which in turn allows for easy desorption [Jalali and Moradi, (2013); Baghenejad et al., (2016); Elbana et al., (2018)]. Nevertheless, it’s crucial to note that the selectivity order is not fixed and can change depending on various factors, some of which are presented on table 8.

Table 8. Essential factors affecting heavy metal adsorption by calcite [Fathy et al., (2025); Tahervanad and Jalali, (2017); Jalali and Moradi, (2013); Li et al., (2021); Elbana et al., (2018); Li et al., (2023); García-Sánchez and Álvarez-Ayuso, (2002); Miranda et al., (2021)]

No.	FACTOR	EFFECT ON ADSORPTION
1.	pH (speciation, surface charge)	↑ pH increases negative surface charge and carbonate availability, strongly enhancing adsorption/precipitation, often giving nearly complete removal in alkaline conditions
2.	Carbonate content	more calcite provides Ca^{2+} for exchange and CO_3^{2-} which forms metal carbonates/solid solutions
3.	Initial metal concentration	controls driving force and site saturation
4.	Calcite dose/surface area	↑ dose (finer particles) gives more reactive surface, increasing removal efficiencies and apparent capacities for all metals
5.	Metal ionic properties	softer, less strongly hydrated cations bind more strongly

6.	Contact time and kinetics	adsorption is generally fast (minutes-hours), often described by pseudo-second-order kinetics, indicating chemisorption/surface-controlled processes
7.	Temperature	often exothermic or slightly endothermic; lower temperatures can favor adsorption

• **Brief notes on calcium and magnesium on calcite**

Calcium is important because as it was previously mentioned, it builds the framework of calcite, while magnesium functions simultaneously as a growth barrier and as a selector of different crystal forms, which in turn, determines the calcium carbonate mineral types that will form, and their stability in both natural and laboratory conditions [Koskamp et al., (2021)].

5. Recapitulating and discussion

The three minerals of interest show specific benefits and drawbacks that depend on their intended usage conditions. Despite its high selectivity and structural stability, clinoptilolite (CL) shows relatively lower adsorption capacity when compared to swelling (expandable) clays such as montmorillonite (MMT). Moreover, its performance depends on the presence of competing ions in real wastewater system, which in turn may reduce its overall efficiency. On the other hand, although montmorillonite shows superior adsorption capacity because of its high surface area and swelling behavior, its practical application is often limited because of three main operational difficulties, including particle aggregation, solid-liquid separation and instability under varying environmental conditions. Calcite in contrast to aluminosilicate minerals (CL and MMT) operates through precipitation and surface-controlled processes. This fact allows for the system to achieve effective metal removal under alkaline conditions but seeing as its performance depends on pH it can have unpredictable outcomes for complex systems. The proper mineral selection requires strong dependence on three factors which include water chemistry, target contaminants and operational constraints.

Table 9. Quantitative comparison of adsorption performance of clinoptilolite, montmorillonite and calcite [ref. for CL: Velarde et al., (2023); Lim et al., (2019); Şenila et al., (2022); Li et al., (2019); Zahid et al., (2025); for MMT: Chen et al., (2015); Eleraky et al., (2025); Inglezakis et al., (2007); Budianta, (2021); Abdelfattah et al., (2021); for CaCO₃: Fathy et al., (2025) Tahervanad and Jalali, (2017); Miranda et al., (2021)]

No.	MINERAL	METAL ION	ADSORPTION CAPACITY (q _{max} , mg/g)	OPTIMAL pH	CONTACT TIME (min)	NOTES	LIMITATIONS
1.	Clinoptilolite (CL)	Pb ²⁺	50-15	5-6	60-180	High selectivity stable	Competing ions
		Cd ²⁺	20-80	5-6	60-180	Moderate capacity	Limited pore size
		Cu ²⁺	25-100	4-6	60-120	Good ion exchange	Limited pore size
2.	Montmorillonite (MMT)	Pb ²⁺	80-200	5-7	30-120	Very high capacity	Swelling, separation

		Cd ²⁺	40-120	5-7	30-120	Strong adsorption	Instability
		Cu ²⁺	50-150	5-7	30-120	Surface and ion exchange	Instability
3.	Calcite (CaCO ₃)	Pb ²⁺	Up to ~100 (precipitation-based)	>7	10-60	Strong precipitation	Strong pH dependence
		Cd ²⁺	20-80	>7	10-60	Moderate removal	Low versatility
		Cu ²⁺	20-60	>7	10-60	pH dependent	Instability

*Zn²⁺ and Ni²⁺ are excluded because their adsorption from minerals of interest is relatively weaker and less documented

To recapitulate, the following table shows side-by-side comparison of the selectivity trend and main mechanisms of CL, MTT, and CaCO₃ for Pb²⁺-Cd²⁺-Zn²⁺-Cu²⁺-Ni²⁺ (heavy-metals of interest) removal from water, as indicated by various literatures.

Table 10. Recapitulation: comparison of clinoptilolite, montmorillonite and calcite for Pb²⁺-Cd²⁺-Zn²⁺-Cu²⁺-Ni²⁺ removal from water [ref. for CL: Velarde et al., (2023); Lim et al., (2019); Şenila et al., (2022); Li et al., (2019); Zahid et al., (2025); for MMT: Chen et al., (2015); Eleraky et al., (2025); Inglezakis et al., (2007); Budianta, (2021); Abdelfattah et al., (2021); for CaCO₃: Fathy et al., (2025) Tahervanad and Jalali, (2017); Miranda et al., (2021)]

No.	MINERAL	SELECTIVITY TREND	MAIN MECHANISMS
1.	Clinoptilolite (Natural zeolite)	generally: <u>Pb²⁺ ≥ Cu²⁺ > Cd²⁺ ≥ Zn²⁺ > Ni²⁺</u> While for Na-CL selectivity often reported as Pb²⁺ > Cu²⁺ > Zn²⁺ > Cd²⁺ > Ni²⁺ or in multi-metal systems Pb²⁺ > Cd²⁺ > Zn²⁺ > Ni²⁺	Dominantly cation-exchange in zeolite channels; affinity controlled by charge density and hydrated radius. Pb²⁺ and often Cu²⁺ bind strongly, Cd²⁺ intermediate, Zn²⁺ and Ni²⁺ weaker. Uptake is typically ten of mg/g per metal; total capacity in the range 0.1-0.3 mmol/g depending on deposit and conditions. Capacity and selectivity can be enhanced by thermal/acid or salt modification
2.	Montmorillonite (as Na-/Ca-MMT, main phase in many bentonites)	Na-MMT generally surpasses Ca-MMT for Pb ²⁺ , Cu ²⁺ , Cd ²⁺ , Zn ²⁺ ; selectivity within this group is Pb ²⁺ ≥ Cd ²⁺ ≈ Cu ²⁺ ≥ Zn ²⁺ , Ni ²⁺ somewhat lower	Removal dominated by ion exchange , with some precipitation at higher pH. For Na-MMT, capacities (mmol/g) for Pb ²⁺ , Cu ²⁺ , Cd ²⁺ , Zn ²⁺ around 0.262, 0.152, 0.204,

			0.151 mmol/g respectively; slightly lower for Ca-MMT.
3.	Calcite (limestone (CaCO₃), includes calcined CaO)	Selectivity: limestone – Pb²⁺>Cd²⁺>Cu²⁺, calcined limestone – Pb²⁺>Cu²⁺>Cd²⁺	(Surface) precipitation and co-precipitation (pH dependence). Very strong preference for Pb²⁺; weaker but significant for Cu²⁺ and Cd²⁺; Zn²⁺ and Ni²⁺ is weaker due to higher solubility of their carbonates/hydroxides

6. Challenges, future perspectives and conclusions

From the gathered evidence recapitulated in the previous sections and briefly in table 9 and 10, it can be concluded that clinoptilolite (CL) and montmorillonite (MMT) being natural, porous and volcanic-derived silicate minerals, constantly show removal preference via ion exchange mechanisms for Pb²⁺ (followed by Cd²⁺, Cu²⁺, Zn²⁺, Ni²⁺); whereas carbonate mineral calcite (CaCO₃), through pH-dependent precipitation processes, immobilizes Pb²⁺ (and to a lesser extent Cd²⁺), however has a weaker effect or “control” on Zn²⁺, Cu²⁺ and especially Ni²⁺. This, factually makes them diverse “tiny sponges” that “extract” (adsorb) heavy metals from water sources, before they can cause damage to the environment or public health. Nevertheless, despite the potential of these natural sources, they still face multiple challenges, mainly because remediation of real-life wastewater is much more complicated compared to when scientists decontaminate and purify water in laboratory conditions, for various reasons. For example; different metals compete for space on mineral surfaces, which in turn can reduce the efficiency and selectivity of the mineral; the fact that the minerals can become saturated with heavy metals in the process; if there is a possibility to reuse them in those conditions or if not how to safely dispose of the contaminated waste, etc. To navigate these obstacles, researchers are “improving” these materials by combining them with other substances like polymers and metal oxides. With further research and innovation, these natural minerals could become an important, sustainable solution for providing cleaner and safer water worldwide.

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