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Review

Inexpensive Organic Materials and Their Applications towards Heavy Metal Attenuation in Waters from Southern Peru

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Abstract: There is interest in using locally available, low cost organic materials to attenuate heavy metals such as Cd, Cr, Cu, Hg, Ni, Pb, and Zn found in surface waters in Peru and other developing regions. Here we mesh Spanish language publications, archived theses, and prior globally available literature to provide a tabulated synthesis of organic materials that hold promise for this application in the developing world. In total, nearly 200 materials were grouped into source categories such as algae and seashells, bacteria and fungi, terrestrial plant-derived materials, and other agricultural and processing materials. This curation was complemented by an assessment of removal potential that can serve as a resource for future studies. We also identified a subset of Peruvian materials that hold particular promise for further investigation, including seashell-based mixed media, fungal blends, lignocellulose-based substrates including sawdust, corn and rice husks, and food residuals including peels from potatoes and avocadoes. Many studies reported percent removal and/or lacked consistent protocols for solid to liquid ratios and defined aqueous concentrations, which limits direct application. However, they hold value as an initial screening methodology informed by local knowledge and insights that could enable adoption for agriculture and other non-potable water reuse applications. While underlying removal mechanisms were presumed to rely on sorptive processes, this should be confirmed in promising materials with subsequent experimentation to quantify active sites and capacities by generating sorption isotherms with a focus on environmental conditions and specific contaminated water properties (pH, temperature, ionic strength, etc.). These organics also hold promise for the pairing of sorption to indirect microbial respiratory processes such as biogenic sulfide complexation. Conversely, there is a need to quantify unwanted contaminant release that could include soluble organic matter and nutrients. In addition to local availability and treatment efficacy, social, technical, economic, and environmental applicability of those materials for large-scale application must be considered to further refine material selection.

Keywords: heavy metals; Peru; remediation; sorption; water treatment

1. Introduction

Heavy metals impair water supplies globally (e.g., [1]). Heavy metals are a group of elements with high density (generally greater than 6 g/cm^3) [2] including Cd, Cr, Cu, Hg, Ni, Pb, and Zn. Common characteristics of heavy metals in the environment are persistence, bioaccumulation, biotransformation [3], and toxic implications for human and environmental health [4]. Once ingested, these metals are not easily eliminated, which can lead to bioaccumulation and damage of internal organs, resulting in chronic and terminal diseases (e.g., [5]).

A wide variety of technologies have been developed and applied globally for the removal of heavy metals from polluted waters, including chemical precipitation, ion exchange, membrane filtration (electrodialysis, reverse osmosis, nanofiltration, ultrafiltration), electrocoagulation, recovery by evaporation, coagulation-flocculation, flotation, solvent extraction, and electrolysis reduction [6]. While effective for contaminant attenuation, these treatment approaches collectively necessitate high operating costs, generate sludge, demand additional chemicals to function properly, and require centralized and skilled operational management by centralized water treatment plants [7]. These limitations necessitate exploring alternatives to remove heavy metals from polluted waters for use in remote locations and developing countries where resources are limited, as well as towards agricultural and other non-potable water applications.

Additional promise for metal attenuation resides in the application of locally abundant, affordable, and renewable organic materials. This has been explored globally in places as diverse as Taiwan [8], Turkey [9], China [10,11]), India [12–15], Nigeria [16], Serbia [17], Saudi Arabia [18,19], Egypt [20], Jordan [21], Poland [22], and Malaysia [23], among many others. Since earlier publications (e.g., [24]), several reviews have synthesized these findings and applications with a particular focus on the developing world, including those conducted by [14,25–27].

With this in mind, it is important to continue expanding the geographic scope of this focus on regionally available and inexpensive materials for heavy metal removal to include additional world regions such as Latin America, where less research has been done. For example, crop irrigation with regional waters generates local food crops in southern Peru that often exceed public and environmental health standards for heavy metals [28]. River water impairment in the area is caused by both natural and anthropogenic causes [29–31]. The volcanoes and high-altitude formations of the Andes mountain range have emitted magmatic products such as ashes, pyroclastic flows, and lahars (in addition to geysers) that contribute metal(loid) elements into the local fluvial system. Metallogenetic deposits in the region (gold, copper, and silver, among others) are created by the contribution of magmatic solutions in existing rocks that in turn affect fluvial and terrestrial ecosystems with heavy metals, which can be enriched when water passes through host rocks [32]. Similarly, anthropogenic activities including agriculture and mining further contribute to heavy metal release into rivers and aquifers [29,33]. Mining has been practiced across the region since pre-colonial times, creating a dispersed landscape of legacy and active sites, ranging from artisanal to large-scale [34]. Precise attribution of contaminant origins is difficult in this context, which means remediation must typically be undertaken at public expense. Given these realities, social, technological, and economic barriers hinder adequate water treatment needed to protect human and ecological health [35].

To this end, this literature-based synthesis and tabulation of materials focuses on combining results of studies conducted with organic materials in southern Peru (Arequipa region) with others from developing countries. In addition to those available in peer-reviewed English-language literature, we curated and contextualized local findings, many of them in Spanish, including theses housed at the Universidad Nacional de San Agustín de Arequipa (UNSA), other local universities, and federal reports from the Autoridad Nacional del Agua (ANA). Performing this type of meta-analysis capitalizes on local knowledge and insights for material screening, which can be difficult to obtain without access to regional data and information. Our analysis specifically focused on treatment technologies and efficiencies related to the removal of the following heavy metals: cadmium, copper, mercury, manganese, nickel, lead, and zinc. While aluminum and iron attenuation have also been investigated, we assigned these metals less relevance due to their ubiquity and comparatively low toxic implications [36]. Collectively, this approach enabled us to synthesize what has been learned about the potential application of locally available and inexpensive materials for heavy metal attenuation, curate these studies based on material class and treatment promise and identify a subset of these materials for further study and application.

2. Discussion

2.1. Screening of Peruvian Materials

We identified 36 studies from southern Peru published within the time period considered (i.e., between 2012 and 2019), in which 29 new (i.e., globally never tested before) organic materials were assessed for the removal of heavy metals. These studies were performed under laboratory batch conditions, with a focus on regional and synthetic waters. The analyses were performed by considering the absorbent used (low-cost organic material) and its efficiency in the removal of the analyzed contaminant from water (only available in percent) using synthetic or regionally harvested waters. Collective results from the synthesis of these materials with additional published reports focused on availability in the developing world resulted in the curation of heavy metal removal performance for a total of approximately 200 different materials. These materials were broadly classified into four groups to coarsely reflect their source: (1) algae and seashells; (2) bacteria and fungi; (3) terrestrial plant-derived materials (fruits, leaves, peels, seeds, stems, shells, moss, and others); and (4) other agricultural and processing materials.

Studies that documented results in equilibrium (q_e) adsorbent loading (mg/g) and removal efficiency (% removal) were included in this international synthesis. While removal efficiencies are experimentally dependent on material mass, aqueous phase volume and contaminant concentrations, this approach enables a coarse screen for potentially promising materials and paths forward within the confines of these experimental caveats. Peruvian results were classified according to the efficiency of the material for metal removal (i.e., removal percent, as previously mentioned), sorting them using to the following criteria: high efficiency ($X \geq 90\%$), medium efficiency ($70\% \leq X < 90\%$), and low efficiency ($X < 70\%$), with "X" being the percentage of metal removal obtained. Peruvian and international studies were combined in the same tables (see Tables 1–4) by curating both equilibrium loading and percent removal, as has been done by others (e.g., [10,14,15,25].

Table 1. Heavy metal removal potential of algae and seashells in the developing world, in mg/g or (% removal) (Peruvian studies in black color). SNS stands for "species not specified".

Absorbant	Cd (II)	Cr (III)	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Algae										
Ascophyllum nodosum	69.7						35.2		41.2	[37]
Chaetomorpha linum	58.6									[38]
Chlorella sp.									(94)	[39]
Chlorella vulgaris	67.0									[40]
Chondracanthus chamissoi				(46)						[41]
Chondrus crispus	65.2						35.2		42.5	[37]
Cladophora sp.				14.5				46.5		[42]
Codium vermilara	21.4						12.9		21.6	[37]
Ecklonia sp.			(60)							[43]
Enterobacter sp.	46.2			32.5				50.0		[44]
Fucus vesiculosus			42.6							[45]
Gracilaria caudata							45.0			[46]
Gracilaria salicornia	19.6									[38]
Laminaria japonica									91.5	[47]
Oedogonium hatei							40.9			[48]
Padina tetrastomatica	64.0									[38]
Palmaria palmate			33.8							[45]
Pelvetia caniculata	75.0									[49]
Sargassum muticum							(70)			[46]
Sargassum natans	115.0									[50]
Scenedesmus obliquus	(99)									[51]
Spirogyra sp.								140.0		[52]
Spirogyra sp.				133.3						[53]

Table 1. *Cont.*

Absorbant	Cd (II)	Cr (III)	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Spirulina platensis				(91)						[54]
				67.9						[55]
Stoechospermum marginatum			32.6							[56]
Ulva lactuca	35.7									[57]
Seashells										
Clam (*Anadara inaequivalvis*)				330.0				621.0		[58]
Crab particles (*Brachyura* sp.)				244.0						[59]
Oyster (*Crassostrea* sp.)	118.0							1591.0	564.0	[60]
Razor clam (*Siliqua patula*)	501.0							657.0	553.0	[60]
Mixed Media										
Algae (*Scenedesmus obliquus*) and fungi (*Wallemia sebi*)								(100)		[61]
Seashells (Mussel *Mytilidae* sp.) and charcoal					(95)					[62]
Seashells (SNS), eggshells, and activated charcoal				(98)						[63]

Table 2. Heavy metal removal potential of bacteria and fungi in the developing world, in mg/g or (% removal) (Peruvian studies in black color). SNS stands for "species not specified".

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Bacteria										
Arthrobacter sp.				32.6						[64]
Bacillus cereus									66.6	[65]
	44.4									[66]
Bacillus circulans			34.5							[67]
Bacillus coagulans		39.9								[68]

Table 2. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Bacillus licheniformis			(95)	(32)						[69]
Bacillus megaterium			32.0							[67]
Bacillus pumilus								28.1		[70]
Bacillus thuringiensis							41.8 (16)			[71]
Escherichia coli	1.5 (79)		1.2 (100)				1.4 (74)			[72]
	(51)							(80)		[73]
Mucor rouxii	8.5									[66]
Pantoea sp.	52.0									[74]
Pseudomonas aeruginosa PU21	57.4									[66]
Pseudomonas fluorescens	66.3									[75]
Pseudomonas putida	53.5								128.0	[76]
								180.4		[77]
Pseudomonas veronii 2E	54 (47)			(35)					(41)	[66]
Rizobium leguminosarum (var. *Viciae*)	135.3									[78]
Trametes versicolor				140.9						[79]
Fungi										
Agaricus bisporus	29.7							33.8		[80]
Aspergillus flavus				93.7						[81]
Aspergillus niger	(58)			9.5 (34)						[82]
								32.6		[83]
								172.3		[81]
Aspergillus terreus				180 (90)						[84]
Auricularia polytricha				6.6	6.0				6.1	[85]
Botrytis cinerea									13.0	[86]
Calocybe indica	24.1							23.4		[80]

Table 2. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Cephalosporium aphidicola								36.9		[86]
Flammulina velutipes				7.2	7.9				6.3	[85]
Lentinus sajor-caju (free)			18.9							[87]
Lentinus sajor-caju (immobilized)			32.3							[87]
Mucor rouxii (alive)	8.5						11.1	35.7	7.8	[88]
Mucor rouxii (dead)	20.3						20.5	53.8	53.9	[88]
Pencillium purpurogenum	110.4				70.4			252.8		[89]
Pencillium simpliccium	52.5							76.9	65.6	[90]
Penicilium sp.		(97) *								[91]
Penicillium canescens	102.7				54.8			213.2		[92]
Penicillium chrysogenum							12.3			[93]
	56.0									[94]
Penicillium citrinum			(80)							[95]
Penicillium digitatum	3.5								9.7	[96]
Phanerochaete chrysosporium							55.9	53.6		[97]
	84.5									[98]
Pichia guilliermondii				20.0						[99]
Pleurotus eryngii				3.4	2.8				2.9	[85]
Pleurotus ostreatus			10.8	8.1			20.4		3.2	[100]
			(21)							[101]
				4.5	3.4				5.1	[85]
Pleurotus platypus	35.0							27.1		[80]
Pycnoporus sanguineus				2.8						[102]
Rhizopus arrhizus	(23)			10.8 (36)						[83]
	25.0									[50]
	30.0									[94]

Table 2. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Saccharomyces cerevisiae			32.6				46.3	270.3		[103]
	1.0									[104]
	31.8									[105]
							11.4			[106]
							9.0			[107]
			(87)							[108]
		11.3	3.3							[109]
Trametes versicolor							212.5			[110]
Mixed Media										
Bacteria. (*Lactobacillus delbruckii* var. *Bulgaricus* and *Streptococcus thermophilus*)									(92)	[111]
Fungi. (*Alternaria* sp., *Aspergillus* sp., *Pseudocamarosporium* sp., and *Penicillium* sp.)		(100)						(99)		[112]

Table 3. Heavy metal removal potential using terrestrial plant-derived materials in the developing world, in mg/g or (% removal) (Peruvian studies in black color). SNS stands for "species not specified".

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Fruits										
Quinoa (*Chenopodium quinoa*)			(55)							[113]
Taro (*Colocasia esculenta*)		6.1	1.4							[114]
Tuya oriental (*Thuja orientalis*)							12.4			[115]
Apple (*Malus* sp.)				10.8						[116]
Apricot (*Prunus armeniaca*)							101.0			[117]

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Leaves										
Amaltas (*Cassia fistula*)			4.5							[118]
Cactus (*Cumulopuntia unguispina*)	(97)					(96)				[119]
Cactus (SNS)		(55)	(20)							[120]
Mangrove (*Rhizophora mangle* L.)		6.5	5.7							[114]
Neem (*Azadirachta indica*)			16.0							[121]
				17.5						[122]
Pineapple (*Ananas comosus*)				9.3						[123]
Peels										
Avocado (*Persea americana*)						(88)				[124]
Banana (*Musa paradisiaca*)								(100)		[125]
Banana (*Musa* sp.)	34.1			52.4			54.4	25.9	21.9	[126]
				4.8			6.9	7.9	5.8	[127]
				7.4						[128]
		(95)								[129]
Banana, cortex (*Musa* sp.)	195.0	240.0							125.0	[19]
Cassava (*Manihot esculenta*)	119.6			127.3						[130]
							57.1			[131]
Chestnut (*Bertholletia excelsa*)								(59)		[132]
Sweet lemon (*Citrus limetta*)			250.0							[133]
Clementine, peel's pectin (*Citrus reticulata*)								(72)		[134]
Cucumber (*Cucumis sativus*)								(87)		[135]
Grapefruit (*Citrus paradisi*)	42.1						46.1			[136]
Kiwi, cortex (*Actinidia deliciosa*)	470.0	375.0							280.0	[19]
Lemon (*Citrus limon*)	54.6	(12) *		70.9			80.0	37.9	27.9	[126]
Lemon, peel's pectin (*Citrus x limon*)										[134]

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Orange (*Citrus senensis*)								(85)		[137]
				(60)						[138]
	41.8			63.3			81.3	27.1	24.1	[126]
				1.8			6.0	7.8	5.3	[127]
	293.0						162.0	476.0		[139]
								27.9		[140]
								46.6		[141]
				(92)						[142]
Orange, modified (*Citrus senensis*)				289.0						[143]
Orange, peel's pectin (*Citrus x sinensis*)		(18) *								[134]
Pomegranate (*Punica granatum*)							52.0			[144]
Potatoe (SNS)			3.3							[145]
Prickly pear (*Opuntia ficus*, var. *Indica*)								(99)		[146]
Tangerine, cortex (*Citrus reticulata*)	450.0	350.0							260.0	[19]
Walnut (*Bertholletia excelsa*)								(38)		[132]
Seeds										
Date (*Phoenix dactylifera*)	39.5			35.9						[147]
Papaya arequipeña (*Vasconcellea pubescens*)		(80)	(78)							[148]
Almond (*Prunus* sp.)		(60)	10.2 (24)							[120]
Coffee (*Canephora* sp.)	39.5			31.2			11.0	19.5	13.4	[149]
Stems										
Rose (*Rosa* sp.)								(97)		[150]
	(41)									[151]
Grape tree (*Vitis* sp.)				10.1						[152]
Shells										

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Cashew nut (*Anacardium occidentale*)							18.9			[153]
				20.0						[154]
Cocoa (*Theobroma cacao*)	0.2	0.2		0.5		0.1	0.2	5.2	0.0	[155]
Coconut (*Cocos* sp.)			18.7							[121]
				19.9						[122]
Coconut, green (*Cocos nucifera*)	(99)	(90)	(86)							[156]
Dye groundnut							7.5			[157]
Groundnut (*Arachis hypogaea*)				4.5			3.8		7.6	[157]
Hazelnut (*Corylus* sp.)				58.3						[158]
Nutshells (SNS)	19.4									[159]
Peanut (*Arachis hypogaea*)		27.9		25.4						[22]
			4.3							[118]
Walnut (*Juglans* sp.)								1.5		[160]
Wheat				10.8 (99)						[161]
Moss										
Moss (*Fontinalis antipyretica*)	28.0								15.0	[162]
Moss (*Hylocomium splendens*)	32.5	42.1								[163]
Moss (Irish peat)				17.6			14.5			[164]
Other Plant-Derived Materials										
Amicon regenerated cellulose									(99)	[165]
Apricot stone (*Prunus americana*)								(93)		[166]
Bagasse, sugar cane (SA-SC)	149.9									[167]
Bagasse, sugar cane (*Saccharum officinarum*)	(96)									[168]
	(96)									[169]
								87.0		[140]
							2.2			[170]

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Bagasse, sugar cane flyash (*Saccharum officinarum*)	6.2						6.5			[171]
Bark (*Abies alba*)				7.8 (83)						[172]
Bark (*Acer rubrum*)										[173]
Bark (*Azadirachta indica*)							30.4			[174]
Bark (*Hardwickia binata*)				(100)			(99)			[175]
Bark (*Lagerstroemia speciosa*)			24.4	11.3			12.2	6.7		[176]
Bark (*Moringa oleifera*)								(93)		[177]
Bark (*Pausinystalia johimbe*)								(100)		[178]
Bark (*Pinus nigra*)	5.7			13.5			6.3			[179]
Bark (*Pinus palustris*)									(73)	[180]
Bark (*Pinus palustris*)							(56)			[181]
Bark (*Pinus radiata*)				(98)						[180]
Bark (*Pinus* sp.)	8.2			(69)						[182]
Bark (*Sequoia sempenvirens*)	(97)				(97)					[183]
Bark (*Techtona grandis*)				(93)						[184]
Bark (*Techtona grandis*)					(99)					[181]
Bark (*Terminalia tomentosa*)			(94)				(68)			[185]
Bark (*Tsuga heterophylla*)								(99)		[186]
Bran, barley (*Hordeum vulgare*)								(59)		[187]
Bran, rice (Oryza sp.)			12.3							[121]
Bran, rice (Oryza sp.)				21.0						[122]
Cellulose bearing schiff				87.0				85.0		[13]
Coagulant, sentry plant (*Agave americana*)								(97)		[188]
Cork (*Pausinystalia johimbe*)				(85)			(91)			[175]
Cork, olive (*Olea europea*)		(67)								[189]

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Corncob (*Zea mays*)								(91)		[190]
	5.1									[191]
Corncob (*Zea mays*) (oxidized)	55.7									[191]
Endocarp, olive (*Olea europea*)		(60)						(89)		[192]
Fiber, ramie (*Boehmeria nivea*)	159.1							273.8		[193]
Hull, peanut (*Arachis hypogaea*)				21.3						[194]
				12.0						[195]
Husk, almond (SNS)							39.8			[196]
Husk, coconut coir (*Cocos nucifera*)	(75)							(80)		[197]
Husk, peanut (*Arachis hypogaea*)		7.7		10.2				29.1		[198]
Husk, rice (*Oryza* sp.)	16.6			10.9	36.1		5.5	58.0	8.1	[199]
			11.4							[121]
			(77)							[200]
				17.9						[122]
								(99)		[201]
	6.6				4.0				9.6	[20]
Leaf powder, neem (*Azadirachta indica*)								(93)		[202]
Lignin (SNS)	25.4			22.9			6.0	89.5	11.3	[203]
		18.0								[204]
Maize cobs (SNS)								(99)		[201]
Mat, reed (*Cannomois virgata*)		7.2	1.7							[114]
Oil cake, neem (*Azadirachta indica*)	11.8			9.4						[205]
Okra cellulosic (*Abelmoschus esculentus*)	121.5			72.7				274.0	57.1	[206]
Peach stone (*Prunus* sp.)								(97)		[166]
Peanut skins (SNS)					820.0					[207]
Pine needles (*Pinus* sp.)		(79)	21.5 (43)							[120]

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Pulp, sugar beet (SNS)				28.5						[208]
Roots, hyacinth (*Hyacinthus orientalis*)			15.3							[121]
				21.8						[122]
Sawdust (*Cryptomeria japonica*)	(75)									[209]
Sawdust (*Eucalyptus* sp.)		(93)						(99)		[201]
Sawdust (SNS)				1.0				1.0		[210]
										[120]
		(97)	15.8 (54)							[211]
			(95)							[118]
Sawdust (*Ziziphus mauritiana*)			3.7					(99)		[212]
Sawdust, beech (*Fagus* sp.)				4.5			4.0		2.0	[213]
Sawdust, fir-wood (*Abies* sp.)				(100)					(100)	[17]
Sawdust, maple (SNS)							0.3			[157]
Sawdust, meranti (*Shorea* sp.)		37.9		32.1			36.0	34.2		[214]
Sawdust, neem (*Azadirachta indica*)								(95)		[177]
Sawdust, papaya wood (*Carica papaya*)	(98)			(95)					(67)	[215]
Sawdust, poplar (*Populus* sp.)		5.5		6.6				21.1		[198]
Sawdust, teakwood (*Tectona grandis*)				4.9			8.1		11.0	[216]
Straw, barley (*Hordeum vulgare*)				4.6				23.2		[217]
							35.8			[218]
Straw, rice (*Oryza* sp.)			54.5							[121]
				18.4						[122]
Straw, rice (SNS)					280.0					[207]
Straw, wheat (SNS)		(80)								[12]
Straw, wheat (*Triticum aestivum*)	4.9			1.9						[219]

Table 3. *Cont.*

Absorbant	Cd (II)	Cr (III) or Cr Total *	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Tree fern (*Cyatheales* sp.)				10.6				39.8		[220]
									7.6	[221]
Water hyacinth (*Eichhornia crassipes*)		6.6	0.3							[114]
Water lily (*Nymphaea* sp.)		6.1	5.1							[114]
Wood, juniper (*Juniperus* sp.)	3.2									[221]
Mixed Media										
Peels. Four Peruvian potatoe species (*Solanum* sp.)			(98)							[222]
Corn husk (*Zea mays*) and rice husk (*Oryza sativa*)			(95)							[223]
Sawdust from oak (*Quercus* sp.) and fir-wood (*Fagus* sp.)				(98)						[17]

Table 4. Heavy metal removal potential using other agricultural and processing materials in the developing world, in mg/g or (% removal) (Peruvian studies in black color). SNS stands for "species not specified".

Absorbant	Cd (II)	Cr (III)	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Baby's breath (*Gypsophila elegans*)								(98)		[224]
Barely straw (Rosser)							35.8			[218]
Cabbage (*Brassica oleracea var. capitata*)								60.6		[225]
Cardboard				(95)						[17]
Carrot (*Daucus carota* L.)				12.4		5.2			6.5	[226]
Carrot (SNS)				32.7						[227]
Cauliflower (*Brassica oleracea* var. *botrytis*)								47.6		[225]
Coconut milk processing (*Cocos* sp.)					500.0					[228]
Coir pith (SNS)							9.5			[229]
Coir pith (SNS), modified							38.9			[229]

Table 4. *Cont.*

Absorbant	Cd (II)	Cr (III)	Cr (VI)	Cu (II)	Hg (II)	Mn (VI)	Ni (II)	Pb (II)	Zn (II)	Source
Dung, cow (*Bos taurus*)								(99)		[230]
Eggshells (SNS)			(89)							[231]
Feathers, Chicken (SNS)					24.4					[207]
Green bean (*Phaseolus vulgaris*)	(88)							(96)		[232]
Hemp (*Cannabis* sp.)							242.0			[233]
Larch tannin resin (*Larix gmelinii* Rupr.)			9.1							[234]
Organoalkoxysilane-grafted lignocellulose					(90)					[235]
Paper mill	14.8			13.9			13.7	14.1		[236]
Pistachio hull (*Pistacia vera*)			116.0							[237]
Rice bran (SNS)				27.8						[238]
Sour orange (SNS)				21.7						[239]
Soybean hulls (SNS)				154.9						[240]
Sporopollenin (*Lycopodium clavatum*)	1.6			1.2				8.5		[9]
Tea (*Camellia sinensis*)							90.9		166.7	[241]
	13.8								12.2	[242]
				43.2						[243]
							0.3			[244]
			54.7							[245]
							18.4			[246]
Tea (SNS)								1.6		[160]
Waste pomace of olive oil factory (WPOOF)							14.9			[247]
Wool, sheep (*Ovis aries*)				(71)						[248]
		(58)	41.2 (69)							[120]
Wool, sheep (SNS)					580.0					[207]
Mixed Media										
Kraft lignin from poplar and beech wood (SNS)			11.1							[249]

2.2. Most Promising Peruvian Materials

Of the 29 newly identified organic materials from southern Peru, 19 were able to remove more than 90% of the targeted metals from experimental waters. Waste products and other abundant and inexpensive organic materials such as those compiled here could be applied toward metal removal of river-derived waters prior to agricultural irrigation. This is important given their potential for bioaccumulation in commercial agricultural products and implications for human consumption and commercial export [250]. Materials could also be applied to the treatment of mining impacted waters associated with small-scale informal mining operations in the region. Applications using what would otherwise be waste materials could limit their direct introduction to landfills but also comes with a tradeoff of the need to subsequently dispose of metal-contaminated materials, treatment and subsequent regeneration as explored later in the manuscript.

While the tables below enable interpretation across elements, here we present Pb, Cd, and Cr as examples. Among the best Peruvian adsorbents (in terms of removal efficiency), green algae were capable of reducing metal concentrations of Cd below detection limits. Positive results were also obtained when combining different materials into a mixed adsorbent media, such as different species of fungi (for the removal of Cr and Pb), as well as fungi mixed with microalgae (Pb). Tested materials that showed almost complete metal removal (99%) were cow dung (Pb) and Peruvian prickly pear (Pb). Other efficient materials worth mentioning based on their high removal efficiency are baby's breath (Pb), sawdust from Eucalyptus (Cr), Peruvian cactus (Cd), Green bean (Pb), and rose stems (Pb), although, similar to the above, mixed materials also showed high efficiencies, highlighting four species of Peruvian potatoes (Cr). Peruvian materials that showed particularly low capacity for metal removal were walnut peel (Pb), chestnut peel (Pb), olive endocarp (Cr), quinoa (Cr), and rose stems (Cd), as shown in Tables 1–4. Table A1 provides removal capacities for comparatively less toxic aluminum and iron.

In addition to the above, some materials exhibited specific selectivity for different metals. For instance, data from a single study on avocado peels (*Persea americana*) indicated Al removal to below detection levels but only 88% removal of Mn (Tables 3 and A1). Similarly, certain algal species exhibited high selectivity such as *Scenedesmus obliquus*, which removed nearly 100% of Cd while others such as *Chondracanthus chamissoi* removed less than 50% of introduced Cu (Table 1). A mixture of these adsorbents, therefore, may allow for the removal of a broad range of heavy metals.

Along with removal efficiency, there are a number of additional considerations for selecting organic materials as a treatment technology, including (1) availability in large quantities; (2) the costs of large-scale production; (3) the potential to create other environmental problems; and (4) the social and cultural "fit" of the material and its associated technologies. Out of the 19 Peruvian materials (including those that were tested in combination with other materials) that showed more than 90% removal efficiency, these constraints limit real-world applicability of a number of organic materials. *Scenedesmus obliquus* (Table 1), for example, was able to remove 99% of Cd [51], but this green alga mostly occurs as a unique population in plankton [251], making it difficult to harvest in sufficiently large quantities for water treatment applications. This in turn limits applications such as combining these algae with the fungi *Wallemia sebi* that was shown to remove nearly all Pb from water [61]. Certain seashells, however, are locally abundant near the coast (e.g., [252]) and have shown excellent Hg and Cu removal efficiencies when combined with similarly abundant and inexpensive charcoal [62] and eggshells [63].

Ref [91] used *Penicillium* to remove Cr from water, reaching below detection levels (Table 2). This genus is ubiquitous and abundant in soils [253] but also produces antibiotics that while useful in biomedical applications, could challenge its application in this context [254] necessitating sterilization, strain selection or genetic engineering to limit antibiotic release. Similarly, the fungal genera *Alternaria* and *Aspergillus* are abundant in soils [255] and used to supply many industrial processes [256]. *Penicillium* and *Pseudocamarosporium* are abundant in wheat crops [257], highlighting the potential of a combination of these four fungal genera (*Alternaria*, *Aspergillus*, *Penicillium*, and *Pseudocamarosporium*) for removing Cr and Pb from waters [112]. Despite the potential applications of these fungi, production and extraction costs should be taken into consideration, as well as any potential effects on downstream aquatic ecosystems.

Even though [119] obtained excellent results using *Cumulopuntia unguispina* on the removal of Cd and Mn from water (Table 3), this cactus is a species that exists only in southern Peru [258], limiting its sustainable extrapolation for water treatment in other regions. However, the species is commonly used for local production in agroforestry systems (marmalades, nectars, alcoholic beverages, shampoo, slope stabilization, etc.), making it potentially feasible for regional water treatment projects particularly if residual wastes remain from those applications. Conversely, *Opuntia ficus* var. *Indica* (Peruvian prickly pear) showed great capacity to remove Pb [146]. It is abundant in Peru and elsewhere and used for industrial purposes around the globe [259], suggesting it holds higher promise for future large-scale water treatment projects. Similarly, rose stems (*Rosa* sp.) [150] and *Agave americana* [188] are also abundant on the continent and represent promising materials for Pb removal. Wood processing-derived materials such as sawdust from *Eucalyptus* sp. [209] may also be used in future water treatment projects to remove Cr, since they can be obtained in large quantities. Analogously, locally available mixed media made of peels from four Peruvian potato species (*Solanum* sp.) hold promise to remove Cr [222]. More globally, a combination of ubiquitous staple crop residuals such as corn husks (*Zea mays*) and rice husks (*Oryza sativa*) hold promise as abundant staples that also facilitate effective metal removal with complementary treatment attributes (Table 3).

Gypsophila elegans is an ornamental plant species that is found in many countries and has been found to remove Pb from water [260] though it has limited availability in southern Peru [224]. Cow dung (*Bos taurus*) (Table 4) showed excellent results when used to remove Pb from water [230], and it is an abundant material worldwide. However, special considerations must be taken, as it could create sanitary and ecological problems depending upon its downstream utilization and mandates for disinfection much as is implemented for municipal wastewater. In addition, public perceptions should be considered in conjunction with treatment and reuse to proactively address challenges that might be encountered in terms of impacts to crops and the potential proliferation of pathogens (e.g., [261]). As an example of food processing wastes, green bean residuals (*Phaseolus vulgaris*) hold promise for Pb removal from water in large-scale projects [232] depending on the local availability of this material.

2.3. International Analysis

These findings build on prior studies focused on materials available in southern Peru (e.g., [262,263]) and worldwide, as well as the continued identification of new materials that can be used to remove metals from water (e.g., [264]). Results from this analysis (a total of 203 studies; 36 Peruvian and 167 international) provide a synthesis and screening tool that can inform future studies and further inquiries into candidate materials to more effectively triage initial approaches. Heavy metal removal from water is affected by various water quality parameters such as organic matter, pH, ionic strength, contact time, total reactive sites and temperature (e.g., [8–10,25]). With these variables in mind, Peruvian results which are based on removal efficiency and not equilibrium loading or adsorption capacity can complement worldwide efforts. Collectively, this can provide a triage to inform future studies of promising materials as well as eliminate repetition of materials with limited potential. As shown in Tables 1–4, we propose that these different results can be binned for larger insights. Collectively, we have identified 12 new materials (detailed below) that could be used in large-scale water treatment projects to remove target heavy metals.

The present analysis of low cost organic materials used in southern Peru builds up on the results from earlier reviews (e.g., [14,25–27]), and other independent studies, expanding the state-of-the-art list from 185 known materials to 197, which is a 6% increase to the current knowledge on this topic. Promising synergies of high removal efficiency and large-scale applicability were identified including: two seashell-based mixed media (Table 1); one fungus and a mix of four more fungi (Table 2); three new plant species (Peruvian cactus, Peruvian prickly pear, and rose stems), peels from four Peruvian potatoes, lignocellulose-based sorbants such as sawdust, corn and rice husk (Table 3); cow dung and green bean waste (Table 4); and avocado peels (Table A1). Additionally, new metals were tested in Peru with already documented materials, as is the case of Salas and Sarcco (2017) who obtained positive results using orange peels (*Citrus senensis*) to remove Fe from waters. This material demonstrated past promise for Cd, Cr, Cu, Ni, Pb, and Zn (e.g., [126,127,139–142]) (Tables 3 and A1). While this tabulation enables the reader to triage and bin materials, it also highlights variability between studies and the need for more consistent protocols to enable future screening studies. For instance, the sorption of Cd onto *Escherichia coli* was reported as 51% by [73] while [72] obtained 79% in Portugal. Similarly, Ref. [138] removed 60% of Cu using orange peels, while [142] was able to remove 92% of the metal from regional waters in India.

2.4. Adsorption Isotherms for Promising Materials

Meta-analyses often suffer from inconsistencies in both testing methods and reporting standards. Of particular note, the curated studies expressed results in terms of both equilibrium solid phase concentration (mg/g) and removal efficiency (%). Unfortunately, these expression measures of adsorption efficiency can only be directly compared if all operating conditions are identical, which they were not. The initial adsorbent concentration, ionic strength, contact time, and adsorbent dose all can have an effect on heavy metals removal through adsorption (e.g., [11,265]). The pH of the water source similarly impacts adsorption capacity and solubility [266] and kinetics of mass transfer are impacted by temperature (e.g., [267]).

A conceptual framework for comparison is provided by an adsorption isotherm model such as Langmuir (non-linear form shown as Equation (1)) to yield coefficients that can be used to assess the characteristics and effectiveness of different adsorbents [268].

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (1)$$

C_e represents the concentration of adsorbate at equilibrium (mg/L); q_e is the adsorption capacity at equilibrium; q_m is maximum adsorption capacity (mg/g); and K_L is the Langmuir constant related to adsorption capacity (mg/g). With an underlying assumption of monolayer coverage, this can be correlated with variations of the surface characteristics of the adsorbent (e.g., surface area per mass adsorbent, porosity) where large surface areas and pore volumes will result in higher adsorption capacity. The maximum adsorption capacity can be experimentally derived using different contaminant and adsorbent concentrations and fitting with either non-linear or linear forms of the Langmuir model. Recent research has indicated that data fitting is best performed with the non-linear Langmuir model (Equation (1)), as linear model fits may result in model parameter error [269]. Ideally, maximum adsorption capacity should be reported together with the Langmuir constant so that solid phase sorbate concentration and removal efficiency can be estimated for a given treatment application.

While more empirical, Freundlich isotherms are better suited to heterogeneous materials and tends to fit a wider variety of adsorption isotherm data (e.g., [270]). The Freundlich isotherm model is shown as Equation (2) below, where K_F is the Freundlich adsorption capacity parameter (L/g) and 1/n is the Freundlich adsorption intensity parameter (unitless; [270]).

$$q_e = K_F C_e^{\frac{1}{n}} \quad (2)$$

For purposes of comparing different adsorbents for a given heavy metal, the authors recommend future research in the region to establish isotherms for a subset of the promising materials identified earlier such as sawdust, rice and corn husks. This is needed to understand the role of environmental conditions and regional water properties (pH, temperature, ionic strength, etc.), as well as active sites and capacities. Some of the identified Peruvian materials have been tested elsewhere in the developing world, such as seaweed (Table 1), fungi and bacteria (Table 2), peels from banana and orange, sawdust

(Table 3), and sheep wool (Table 4). Unfortunately, extrapolation is challenging due to variables in reporting convention, environmental conditions, and target metals.

2.5. Considerations of Material Feasibility and Applications in Water Treatment

In addition to material applications to water treatment, it is also important to consider the context in which this might be applied. The bacterium *E. coli* is readily studied in axenic cultures as a model laboratory organism and its capacity for metal sorption is important for fundamental study as well as applications (e.g., [72]). However, it is an enteric bacterium that includes pathogenic strains that have played prominent roles in food contamination and hence could be problematic in water treatment applications. Analogous concerns about the proliferation of pathogens can be raised for materials such as cow dung [230] and fungi [91,112]. Downstream disinfection could counter such adverse effects but would add complexity and cost that could limit adoption. Public perceptions of water treated with such materials could pose further challenges. On the other hand, non-potable uses for the treated water such as irrigation or mining processes could circumvent needs for disinfection. Increasing water availability for these applications will also decrease stress on potable resources which could increase adoption.

The regeneration and potential recovery of heavy metals from adsorbent materials is an important factor that influences material and disposal costs [271]. Although desorption and adsorbent regeneration has been well studied for commercial materials such as activated carbon and ion-exchange resin, the regeneration of the types of materials highlighted in this review have received much less attention [272]. A recent review [271] evaluated the literature on regenerating a wide range of adsorbent materials with a secondary focus on recovery of metals for recycling and fate of spent adsorbents. They concluded that more research is needed to evaluate the best regeneration methods for specific adsorbents, methods for recovering metals from regenerant solutions, and options for safely disposing of spent adsorbents. This is particularly applicable for the less studied, novel organic materials curated here highlighting the need to quantify and assess material regeneration, disposal, life cycle, and opportunities for economic metal recovery in conjunction with treatment efficacy.

Evaluation of materials prior to treatment applications should additionally include quantification of released constituents that could impact downstream use of the waters. The lifespan and breakdown of organic substances during treatment applications needs to be quantified to determine if adsorption capacity decays over time (e.g., [270]). Organic breakdown during treatment applications could result in the release of nitrogen, phosphorous and organic carbon that could contribute to eutrophication and increased oxygen consumption [273,274]. While this release could present a new set of environmental pressures, it could also provide opportunities if the water is used for applications such as agricultural irrigation, where the presence of introduced nutrients could enhance system productivity (e.g., [275]). In another vein, the decay and fermentation of organic substrates can sustain biogenic sulfide production in sulfate rich waters and in turn support passive treatment applications such as sulfate reducing bioreactors. In these systems, a combination of more labile and recalcitrant substrates such as alfalfa coupled to woodchips can sustain long-term immobilization of metal sulfides over a timeframe of years to decades [276,277]. Whether sorbed or precipitated, there are important and of yet unanswered questions associated with treatment lifespan as well as material disposal of heavy metal contaminated materials.

3. Conclusions and Recommendations

As water scarcity continues to increase in southern Peru (as well as other areas in Latin America) [278], it is important to investigate regionally abundant, low-cost materials that can be applied towards heavy metals removal. With limited water resources and projections of future scarcity, it is important to treat and reclaim supplies that have been impacted by natural and anthropogenic metal contamination. Regional studies have reported high metal content in locally grown food products in southern Peru (e.g., [250]), which highlight the need for cost-effective materials for metal removal in association with agricultural production. Water supply and quality have been the source of social conflicts in the region, often pitting mining and agriculture against each other (e.g., [279]). Low-cost remediation approaches could help promote coexistence between sectors. This benefit could extend to the artisanal and small-scale mining sector, which has not so far been a source of major conflict in southern Peru (e.g., [280]) by providing a potentially cost effective, small scale treatment application using waste or inexpensive local materials as a mechanism to reduce the likelihood of future conflicts.

For the new materials, such as plant and food wastes, that hold higher promise, there are associated variables that need further consideration such as their availability, mass demands for water treatment, acquisition/processing cost, ability to integrate into treatment schemes, environmental effects, and the social or cultural fit of the materials and associated technologies. Our curation of approximately 200 studies enables a preliminary screen and highlights several Peruvian materials that could be applied towards heavy metal attenuation. Those materials include two seashell-based mixed media, a mixture of fungi, three new plant species (Peruvian cactus, Peruvian prickly pear, and rose stems), lignin rich compounds such as sawdust from Eucalyptus, corn and rice husk, and food wastes derived from potato, avocado, and green beans. Further studies should explore the social and economic feasibility of these materials. In addition, the site-specific nature of remediation projects is enhanced by the involvement of local communities who are impacted by affected waters and live or work in these areas. In this context, project success and sustainability rely on stakeholder engagement and participation [281].

The synthesis and tabulation provided here can serve as an initial screening tool to triage materials and provide a hierarchy to inform future investigations. Comprehensive adsorption capacity values (mass adsorbate per mass adsorbent) were generally lacking, which limits extrapolation to treatment scenarios. Hence, systematic and standardized experiments (e.g., equilibrium isotherm experiments) should be conducted for promising materials to allow for comparison between materials and the extrapolation of the findings to real-world treatment scenarios.

Commonly used treatment technologies such as chemical precipitation are expensive and require specialized infrastructure and skilled operators for reliable treatment. This paper contributes to a growing body of work that demonstrates that metal-contaminated waters can be treated using inexpensive organic materials. Our focus on southern Peru highlights evidence from local studies and increases the available developing world's state-of-the-art list (based on earlier reviews and independent studies from the English literature) by 6%. Hence, this represents a valuable contribution to our current knowledge on this important branch of water quality remediation, which could be used in combination with naturally-inspired passive treatment technologies such as wetlands and sulfate reducing bioreactors [276,277] that require less maintenance and upkeep demands. Our synthesis can support further and more detailed studies on how heavy metals have been (and are being) removed from impaired waters using locally available and inexpensive materials and provides a framework for application to other countries in South America and beyond.

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Appendix A

Table A1. Removal of ubiquitous metals with limited toxicity in the developing world, in mg/g or (% removal) (Peruvian studies in black color).

Absorbant	Al (III)	Fe (II)	Source
Bacteria and Fungi			
Bacteria, *Bacillus licheniformis*		(52)	[69]
Bacteria, *Escherichia coli*		1 (100)	[72]
Plant-Derived			
Bagasse, sugar cane (*Saccharum officinarum*)		(94)	[167]
Husk, maize (*Zea mays* L.)		0.5	[282]
Peels, avocado (*Persea americana*)	(100)		[124]
Peels, orange (*Citrus senensis*)		(98)	[137]
Shells, cocoa (*Theobroma cacao*)		0.1	[155]
Mixed Media			
Bacteria. *Streptococcus thermophilus* and *Lactobacillus delbruckii*, var. *Bulgaricus*		(100)	[111]
Fungi. *Alternaria* sp., *Aspergillus* sp., *Pseudocamarosporium* sp., and *Penicillium* sp.		(96)	[112]

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