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Review

The Incorporation of Adsorbents with Contrasting Properties into the Soil Substrate for the Removal of Multiple Pollutants in Stormwater Treatment for the Reuse of Water—A Review

Paripurnanda Loganathan [1], Jaya Kandasamy [1], Harsha Ratnaweera [2] and Saravanamuthu Vigneswaran [1,2,*]

[1] Faculty of Engineering, University of Technology Sydney (UTS), P.O. Box 123, Broadway, Sydney, NSW 2127, Australia; paripurnanda.loganathan@uts.edu.au (P.L.); jaya.kandasamy@uts.edu.au (J.K.)
[2] Faculty of Sciences & Technology (RealTek), Norwegian University of Life Sciences, P.O. Box 5003, NO-1432 Ås, Norway; harsha.ratnaweera@nmbu.no
* Correspondence: saravanamuth.vigneswaran@uts.edu.au

Abstract

Stormwater carries significant amounts of pollutants—including metals, microorganisms, organic micropollutants, and nutrients—from land surfaces into nearby water bodies, leading to water quality deterioration and threats to both human health and ecosystems. The removal of these contaminants is essential not only for environmental protection, but also to enable the reuse of treated water for various beneficial applications. Common treatment methods include bioretention systems, biofiltration, constructed wetlands, rain gardens, swales, and permeable pavements. To improve pollutant removal efficiency, adsorbent materials are often incorporated into the soil substrate of these treatment devices. However, most research on adsorbents has focused on their effectiveness against one or two specific pollutants and has been conducted under static, short-term laboratory conditions rather than dynamic, field-relevant scenarios. Column-based dynamic filtration type studies, which are more informative for field applications, are limited. In one study, a combination of two or more adsorbents with contrasting properties that matched the affinity preferences of the different pollutants to the substrate media removed 77–100% of several heavy metals that occur in real stormwater compared to 38–73% removal with only one adsorbent. In another study, polycyclic aromatic hydrocarbon removal with zeolite was only 30–50%, but increased to >99% with 0.3% granular activated carbon addition. Long-term dynamic column-based filtration experiments and field studies using real stormwater, which contains a wide range of pollutants, are recommended to better evaluate the performances of the combined adsorbent systems.

Keywords: pollutants; heavy metals; nitrate; organic micropollutants; phosphate; adsorbents; stormwater treatment

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1. Introduction

Increasing urbanization and industrial activities worldwide have led to the contamination of adjoining lands and natural water bodies with pollutants to such an extent that, in many areas, the water has become unsuitable for both potable and non-potable uses. Stormwater, which drains from land areas following precipitation events—such as heavy rain or meltwater from hail and snow—carries these pollutants in dissolved form or when they are adsorbed to the soil particles eroded from land surfaces, depositing them into water bodies. The quantity and quality of stormwater can vary significantly based on the landscape, soil erosion, and the intensity of precipitation [1–5]. In natural landscapes, much of the precipitation is absorbed into the soil, replenishing groundwater. Furthermore, the plants growing in the landscapes help to mitigate stormwater by enhancing soil infiltration, intercepting rainfall, and absorbing water through their roots. In contrast, in urban environments, stormwater primarily falls on impervious surfaces—such as roads, sidewalks, rooftops, and parking lots—preventing its absorption into the ground [6,7]. As a result, the water flows across these surfaces as runoff, ultimately draining into nearby water bodies such as streams, rivers, wetlands, lakes, or oceans. This runoff can carry large quantities of pollutants and nutrients, leading to a degradation in water quality in these receiving bodies [8–13]. Such pollution poses significant risks to both human and ecosystem health, underscoring the need to remove pollutants from stormwater to prevent environmental harm.

Stormwater is increasingly recognized as a valuable resource, particularly as the demand for water continues to rise due to population growth and industrial expansion, especially in arid and drought-prone regions. In large urban centers, stormwater is considered less as a problem, but more as a potential water supply resource [6,10,14]. Over recent years, stormwater harvesting and reuse have become central to sustainable water management practices. By collecting and reusing stormwater, cities can secure an alternative water source for non-potable uses [6,8,10]. Stormwater reuse systems can be implemented at various scales, from individual properties to entire catchments or sub-catchments [8]. Common systems include rainwater tanks for collecting roof runoff or wet basins and constructed wetlands for larger-scale collection. Once treated to remove pollutants, the collected stormwater can be used for purposes such as industrial processes, irrigation, garden watering, and toilet flushing [8,15]. Stormwater harvesting and reuse involves the collection, storage, treatment, and use of runoff water from urban areas. Stormwater treatment can be divided into primary, secondary, and tertiary levels [8,14]. Primary treatment involves the screening of gross pollutants and the sedimentation of coarse particles. Secondary treatment includes the sedimentation of finer particulates and filtration. Tertiary treatment involves enhanced sedimentation and filtration, biological uptake, and the adsorption of dissolved pollutants onto sediments or specific adsorbents. Primary and secondary treatments are typically applied at the beginning of a treatment train, while tertiary treatments are used at the end. Among these three levels, the removal of dissolved pollutants at the tertiary level is particularly challenging, as it requires complex biological and chemical processes, in contrast to the simpler physical methods used in the primary and secondary stages. Dissolved pollutants encompass a wide range of inorganic and organic ions and compounds of varying molecular sizes, originating from both natural and anthropogenic sources. These pollutants include nutrients (such as nitrogen and phosphorus, which promote algal blooms), heavy metals, biological species (e.g., fecal coliforms), and organic micropollutants (OMPs), such as hydrocarbons and derivatives, pesticides, pharmaceuticals, and consumer products [1,6,11,12,16]. Recent studies have highlighted the presence of newly recognized pollutants in stormwater, including antibiotic resistance genes, microplastics, tire-wear particles, and emerging organic micropollutants (OMPs) such as natural organic matter (NOM), per- and polyfluoroalkyl substances (PFAS), bisphenol-A (BPA), and polycyclic aromatic hydrocarbons (PAHs) [1,3,9,17].

To address these contaminants, tertiary treatment systems, such as bioretention cells, biofilters, constructed wetlands, rain gardens, swales, and permeable pavements, are commonly implemented [7,10,18]. These systems facilitate pollutant removal through a combination of processes, including plant uptake, increased infiltration that limits surface runoff, microbial degradation, root-zone biochemical activity, and adsorption. Adsorption, in particular, plays a central role in removing dissolved pollutants, as many contaminants bind to the soil or engineered substrates integrated into the system. Notably, these treatment technologies often share comparable components and operating principles [13,19].

Among the various mechanisms, adsorption is the most chemically intricate due to the wide range of physicochemical characteristics of stormwater pollutants. These substances can be either organic or inorganic and exhibit diverse traits, such as electric charge, molecular weight, functional groups, hydrophobic or hydrophilic behavior, aromatic structure, hydrogen-bonding potential, and propensity for π–π interactions. Standard soil media may not provide effective removal across such a broad spectrum of contaminants, as their intrinsic properties often do not match the pollutant profiles. Therefore, to enhance removal performance, advanced adsorbents are sometimes incorporated into the soil matrix. These engineered materials typically possess desirable attributes, such as high surface area, porosity, mechanical stability, and tailored functional groups, that facilitate more selective and efficient adsorption [6,12].

Previous reviews of stormwater treatment technologies have focused primarily on traditional pollutants and the use of soil as the adsorbent medium, covering systems like swales, constructed wetlands, biofiltration, rain gardens, and permeable pavements [4,7,11,13,18]. However, these reviews have not addressed all of the pollutants found in stormwater, particularly those recently identified [1,3,9,17]. The objectives of this paper are as follows: 1. to provide a critical review of the literature on the removal of pollutants using traditional technologies and show that they are insufficient; and 2. to suggest modifications that can be made to improve removal efficiency by incorporating combinations of adsorbents with diverse properties that align with the affinity preferences of various pollutants into soil media [20,21]. This latter aspect, which has been underrepresented in previous reviews, is thoroughly addressed here. The paper primarily discusses modifications aimed at improving pollutant removal efficiency with applications in large-scale stormwater treatment in the fields of pollution prevention and water harvesting for reuse. Flocculation, advanced oxidation, and membrane filtration processes are not addressed in detail, as they are typically applicable to high-rate, small-scale stormwater harvesting practices, and are generally not cost-effective for large-scale implementation. Pilot-scale studies involving these high-rate technologies are included in the Supplementary Materials.

2. Conventional Tertiary Treatments

The tertiary stormwater treatment technologies commonly practiced include swales, rain gardens, biofiltration/retention basins, and both natural and constructed wetlands. These systems typically consist of a trench, which may or may not be vegetated, and an underlying soil or sand media [10,19]. Vegetation in these systems can include grass, submerged species, or trees. The treatment technologies remove pollutants through a combination of physical filtration, chemical adsorption, biological degradation (mediated by microbes and plant roots), and plant uptake [10,11]. The main differences among these technologies lie in their size, shape (including slope, landscape, and depressions), purpose, materials used, and drainage systems (natural or constructed). Of the different pollutant removal mechanisms, adsorption is the main one. Examples of studies where pollutants were removed by incorporation of adsorbents are presented below for two commonly used treatment technologies.

2.1. Constructed Wetlands

In constructed wetlands, pollutants, such as heavy metals, organic micropollutants (OMPs), and phosphates, are primarily removed through adsorption onto the soil matrix. However, natural soils often lack sufficient adsorption capacity for effective treatment. To improve performance, amendments with superior adsorption properties are commonly integrated into the wetland substrate. For example, Chen et al. [22] demonstrated that the addition of biochar to a constructed wetland improved the removal of metals like cadmium (Cd), copper (Cu), and chromium (Cr). This improvement was linked to biochar's high cation exchange capacity and the presence of functional groups—including carboxyl, hydroxyl, and phenolic moieties—which facilitate the retention of metal cations via surface complexation, ion exchange, precipitation, and other binding interactions [6,23].

Deng et al. [23] reviewed multiple studies where biochar was employed as a wetland substrate and consistently found enhanced removal efficiencies for various pollutants in wastewater applications. Beyond biochar, a wide range of other materials—such as naturally occurring minerals, synthetic chemical compounds, biomass-derived substances, and industrial or municipal by-products—have also been investigated as alternative substrates in constructed wetlands [24,25]. These materials are being explored to boost adsorption capacity, improve treatment effectiveness, and promote cost-efficient, sustainable water management.

2.2. Permeable Pavement Systems

Permeable Pavement Systems (PPSs) offer another stormwater treatment option, allowing for water to infiltrate into the ground after pollutants are removed by various layers of materials [26]. These systems may or may not incorporate vegetation for plant uptake. They are commonly used along roads to manage excess stormwater runoff and pollutants. A typical PPS consists of a surface paving layer, a base layer, and a sub-base layer [27]. The surface layer is typically made of permeable concrete, porous asphalt, or permeable interlocking concrete blocks [28]. Below this, the base and sub-base layers consist of sand and gravel, with varying thicknesses and particle sizes. The primary mechanism of pollutant removal in PPS is adsorption.

Sun et al. [26] reported that the efficiency of heavy metal removal from synthetic stormwater in a PPS was found to follow this order: surface layer > base layer > sub-base layer. Among the four metals tested, Pb and copper Cu were effectively removed, while Cd and Zn were less effectively treated. Beecham et al. [29] compared a PPS with a conventional pavement in a field experiment, finding that the PPS removed significantly higher percentages of nutrients, some heavy metals, and total suspended solids. This removal was attributed to the adsorption of pollutants onto sediments trapped in the PPS. Despite the high removal rates, concentrations of some pollutants in the effluent still exceeded the ANZECC [8] trigger values.

The results from both constructed wetlands and PPS studies highlight a key limitation: the soil materials naturally present in these systems have relatively low adsorption capacities for certain stormwater pollutants. Moreover, adsorbed pollutants can sometimes re-dissolve into the water, leading to increased concentrations [6]. As a result, materials with higher adsorption capacities need to be incorporated into the natural soil media to effectively remove all pollutants, including those recently identified in stormwater [12]. However, to remove multiple pollutants simultaneously, a combination of two or more adsorbents with contrasting properties is often required, as each adsorbent is typically optimized to target specific pollutants based on its characteristics. This issue will be further explored in the next section.

3. Composite Engineered Substrate Media for Enhanced Stormwater Treatment

3.1. Properties of Stormwater Pollutants and Substrate Adsorbents

3.1.1. Pollutants

Stormwater contains numerous pollutants which can be broadly categorized into undissolved solids and dissolved substances [10,12,17,18,21,27,30]. Undissolved solids are readily removed through straining and filtration during primary and secondary treatment processes. Dissolved pollutants, however, include organic ions, inorganic ions, and molecules with diverse properties. Microorganisms, as well as some of these dissolved substances, are addressed during tertiary treatment. While certain dissolved pollutants are removed through microbial degradation and plant uptake, others are most effectively removed by adsorption. This approach enhances pollutant removal by incorporating specialized adsorbents—each with contrasting properties and specific affinities for target pollutants—into the substrate media of treatment systems. To develop such composite engineered substrate media, it is essential to first understand the properties of both the pollutants and the adsorbents.

The main properties of the commonly identified pollutants that influence their removal by adsorption onto the treatment's substrate media are summarized in Table 1. The sources of these pollutants and environmental impact/health hazards have been presented in many review articles [11,25,31]. The pollutants originate from activities of various sources, namely industries, agriculture, mining, municipality, and pharmaceutical operations. Agricultural activities, namely crop cultivation and animal farming, add nutrient elements, such as nitrate and phosphate, which can potentially produce eutrophication, leading to algal blooms, causing hypoxia and biodiversity loss in aquatic environment. Many organic pollutants, especially OMPs (e.g., PAH, PFAS), are linked to an array of carcinogenic, mutagenic, and reproductivity disorders in aquatic invertebrates. Many heavy metals, which are added in some agrochemicals and industrial effluents, can cause human and aquatic toxicity.

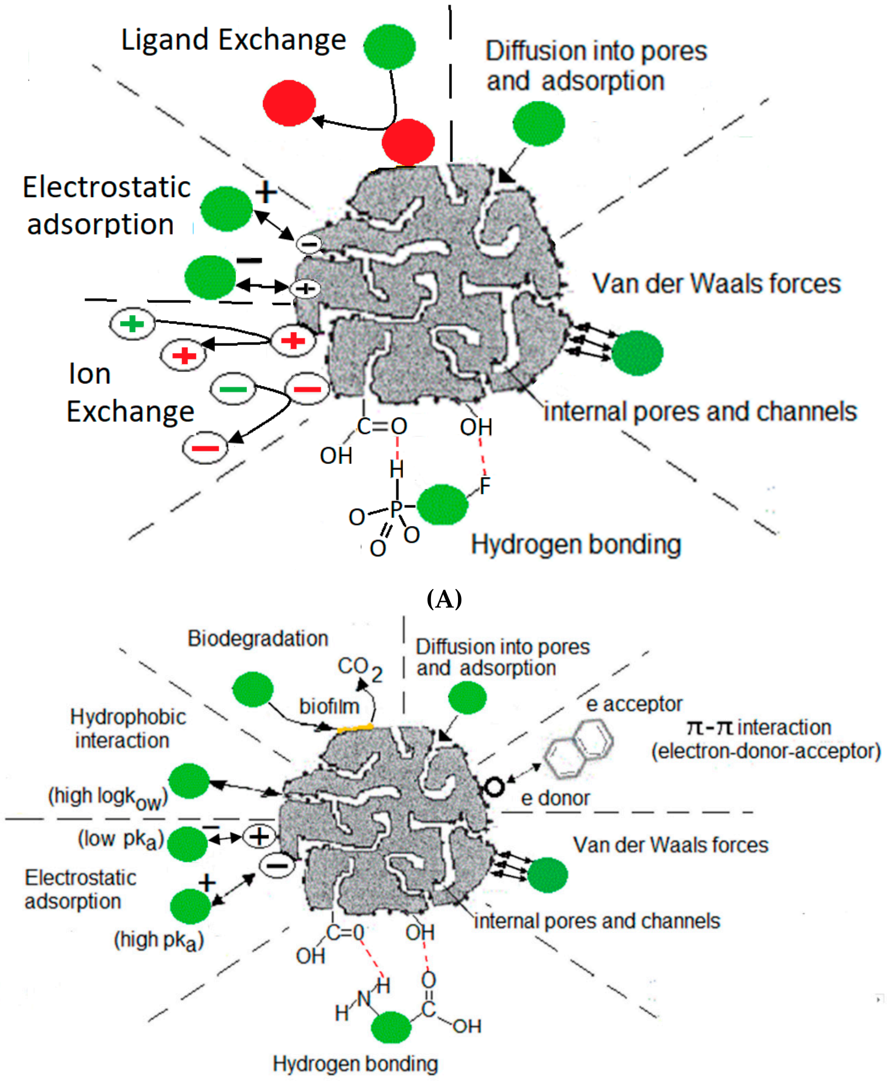
Table 1. Stormwater pollutants and their properties which control adsorption.

Properties	Inorganic Pollutants	Organic Pollutants	Microorganisms
	Metal ions, ammonium, nitrate, phosphate, arsenate, fluoride, total N and P	OMPs (NOM, PAH, PFAS, phenolic species, PPCP, pesticides), dyes, DOC, oil, grease	Bacteria, fungus, protozoa, microalgae, fecal coliforms
Size	Very small	Mostly very large	Very large
Charge at neutral pH (approximate pH of most stormwaters)	Negative for ions formed by non-metals (except ammonium) Mostly positive for ions formed by metals	Mostly negative	Mostly negative
Hydrophobic/hydrophilic.	Mostly hydrophilic	Mostly hydrophobic	Mostly hydrophobic
Hydrogen bonding	Few	Many	Many
π-π interaction	No	Those having aromatic rings	Those having aromatic rings
Van der Waals forces	Very weak	Very strong	Very strong
Microbial decomposition	No	Yes	Not applicable

3.1.2. Adsorbents

The adsorbents used in the substrates must have properties that are compatible with those of the pollutants to facilitate effective adsorption through one or more of the mechanisms presented in Figure 1. The adsorption mechanisms involve electrostatic attraction (adsorbent and pollutant having opposite charges), hydrophobicity/hydrophilicity, hydrogen bonding, π–π interactions, pore size diffusion, ligand exchange, or ion exchange [6,17,25,31].

The adsorbents used in treatment substrates can be categorized into several types: natural mineral materials, chemical products, biomass materials, industrial and municipal waste materials, metal oxides and hydroxides, modified functional materials, nanomaterials, and novel materials. Their properties and limitations have been described in many review articles [12,25,31,32]. They are summarized in the Supplementary Materials, Table S1. When selecting an adsorbent, it is important to consider not only its effectiveness in removing pollutants, but also factors such as cost, potential for reuse, and environmental sustainability. For example, some of the adsorbents, such as water treatment residuals, metal oxides, and metal-modified media, can, during long-term application, release toxic metals, such as Al and heavy metals, which can cause environmental damage, and, therefore, care should be taken in their use [6]. Some adsorbents can be efficient in removing pollutants, but may be costly when used in large-scale operations in the field by mixing it in large areas of land (e.g., constructed wetlands). However, in certain types of stormwater treatments, such as permeable pavements where the adsorbent is applied without mixing with soils, the spent adsorbent can be removed and regenerated and then reused, and this can cut down the cost of treatment [31].



(A)

(B)

Figure 1. Conceptual illustrations of the possible mechanisms for pollutant removal from stormwater by adsorbents used in the substrate media of treatment systems. (**A**). Inorganic pollutants [6,25,33,34] (**B**). Organic pollutants [6,17,25,33].

3.2. Pollutant Removal from Stormwater by Substrate Adsorbents

Only a few of the adsorbents have been tested in long-term experiments with real stormwater. Most experiments have used synthetic solutions in short-term static batch-type or dynamic column-type setups conducted in laboratory settings. Only a few studies have been conducted on a larger scale, such as in pilot plants or microcosms, to simulate real-world conditions over extended periods.

Column-type adsorption studies are particularly useful, as they simulate dynamic filtration processes more effectively than static batch-type adsorption studies, offering more realistic insights into stormwater treatment performance over time. The results from these column experiments can help predict the behavior of larger, practical treatment systems in the field, including the expected long-term effectiveness of pollutant removal [16,17,35,36].

Some long-term column-based adsorption studies on pollutant removal from stormwater are summarized in Tables 2–5, along with their key findings. Most of these studies have used a single adsorbent, which was often found to be ineffective at removing a mixture of pollutants with differing physicochemical characteristics. This is because the surface properties of the adsorbent were not compatible with all the pollutants it aimed to target. For example, Ray et al. [33] compared two polymer–clay complexes with biochar in a column experiment for the removal of OMPs and metals from stormwater. They found that the polymer–clay complexes were not effective at removing OMPs, but were better at removing metals than biochar. In contrast, biochar-based columns effectively removed OMPs, but performed poorly with metals. These results suggest that a combination of polymer–clay complexes and biochar could remove both metals and OMPs effectively. Examples of adsorbent combinations with contrasting adsorption properties for the simultaneous removal of multiple pollutants will be discussed in the next section.

Table 2. Substrate adsorbents primarily used in removing nutrients in dynamic filtration processes (column, pilot-plant, microcosm, or field studies).

Substrate Adsorbent	Method (Treatment, Water, Duration)	Pollutants	Results	Reference
AC and biochar amended sand or pumice.	A study to improve efficiency of biofiltration system of stormwater treatment. 50 mgL^{-1} nitrate pulse input (5 times each 500 mL) to columns containing sand or pumice amended with 5% AC (granular) or two biochar (lab-based study). Packing depth 4.56 cm.	NO_3^-	Biochar columns not as effective as AC columns due to negative surface charges on biochar particles causing repulsion of NO_3^- while AC had Fe and Al which provided positive charges to attract NO_3^-. No difference between sand and pumice for NO_3^- retention when amended with AC.	Kang et al. [37]
Sand (88%) + soil (12%), Biochar (4%) + sand +soil, Fe coated biochar (4%) + sand + soil.	Bioretention column (top to bottom: submerged layer 255 mm, mulch layer 100 mm, filter media layer 700 mm, gravel layer 150 mm). 18 h interval of 11 applications of synthetic stormwater (11 days). No plants.	NH_4^+, total P, NO_3^-	Fe coated biochar best removal of NH_4^+, total P (94–98%). Biochar total P removal (57%). Both these treat. good removal of NO_3^- (93, 97%). Adding biochar into submerged layer helped denitrification of NO_3^-.	Xiong et al. [37]
Sand/dolomite (10:1 ratio).	Constructed wetland, pilot plant study, outdoor planted with *P. australis*, synthetic wastewater. 3 months study. Gravel in top layer (3 cm depth), sand +dolomite layer (42 cm), gravel at bottom drainage layer (17 cm). 40 L of water 2 or 3 times a week applied 8 mgL^{-1} of P applied.	PO_4^{3-}	49% and 45% of added PO_4^{3-} was removed at the end of 3 months reaching 4.1 and 4.4 $mgPL^{-1}$ in the effluent for the 2 and 3 times per week applications, respectively. 6.5–18% increase in P accumulation in the substrate compared to unused media. Adsorption due on $CaCO_3$ and precipitation of Ca phosphate are the reasons. Dolomite to sand ratio can be increased because pH was not beyond 8 to affect plants.	Prochaske and Zouboulis [38]
Perlite + activated Al_2O_3 (PerlAlO), perlite alone, zeolite, GAC.	3 horizontal flow columns (18 cm long, volume 3.2 L) (simulates a Storm Filter cartridge in flow path). 0.5 mg L^{-1} influent P (pH 7), surface loading 43 Lm^{-2} min^{-1}. Empty bed (EB) contact time 2.6 min. Effluents sampled at 15 min interval for 1 h, then every 1–2 h. Conducted inside a laboratory.	Ortho-phosphate (HPO_4^{2-}, $H_2PO_4^-$).	Overall removal efficiency decreased as the number of treated EB volumes (EBVs) increased. To reach 50% overall removal, PerlAlO provided 838 EBVs, whereas GAC could only treat 12 EBVs. For the lifetime of media, PerlAlO outlasted GAC with 2297 EBVs, compared to 1000 EBVs, respectively.	Ma et al. [39]

Table 2. *Cont.*

Substrate Adsorbent	Method (Treatment, Water, Duration)	Pollutants	Results	Reference
Gravel, iron slag, marble stone, zeolite.	Pilot scale vertical-flow constructed wetland (VFCW) which contained 7.58 ± 2.80 mg/L TP and 5.30 ± 2.42 mg/L PO_4 -P at a hydraulic loading rate of 100 L/m^2 · d. P (1 m^2 wetlands, 30 cm depth) were operated for 3 months with and without plants. Anaerobically pretreated wastewater was used.	Total P and PO_4^{3-}	Iron slag was the most efficient material for the removal of total P and PO_4^{3-} both in the absence and presence of plants in VFCW compared to gravel, marble stone and zeolite. This was explained as due to high amounts of Al, Fe and Ca minerals within the slag adsorbing PO_4^{3-}.	Ayaz et al. [40]
Gravel, cocopeat, zeolite, limestone.	Constructed wetland column setup inside glasshouse. Substrate 0.7 m layer over 0.1 m deep gravel layer. Planted with *P. australis*. Synthetic very acidic wastewater leached for 13 weeks. Hydraulic loading 0.073 m^3 m^{-2} d^{-1}.	As, B, Fe	Limestone column removed 99% As, 98% Fe: zeolite 92% As, 86% Fe; Gravel 43%, no Fe removal; Cocopeat 9% As, 46% Fe. Only cocopeat removed B (by coprecipitation with Fe). Suggested that combination of media may be the best to remove all metals.	Allende et al. [41]

Table 3. Substrate adsorbents primarily used in removing heavy metals in dynamic filtration processes (column, pilot-plant, microcosm, or field studies).

Substrate Adsorbent	Method (Treatment, Water, Duration)	Pollutants	Results	Reference
Fe oxide coated sand, sand alone.	Permeable payment columns (laboratory based). Substrate packed length 612 mm. Synthetic stormwater (pH 6.5). Lateral flow rate 50 $mLmin^{-1}$. PH 6.5 is below the point of charge of Fe oxide of 7–8 providing positive surface charge.	Zn, Cd, Pb, Cu	Sand breakthrough rapid for all metals (<20 PV). Fe coated sand breakthrough delayed for all metals, especially for Pb (complete through >600 BV). When Fe sand partially substituted by porous payment aggregate, effluent pH 8.0 and no breakthrough for both Cu and Pb up to 1000 BV (due to pH effect).	Sansalone [42]

Table 3. *Cont.*

Substrate Adsorbent	Method (Treatment, Water, Duration)	Pollutants	Results	Reference
Manganese oxide coated sand.	Column experiment (laboratory based). Substrate packed to 36 mm height (10 g MnO_2). Upward flow of simulated stormwater in the presence and absence of natural organic matter (NOM). Flow rate 0.4 $mLmin^{-1}$ (9.5 cm h^{-1})	Zn, Cd, Cu, Pb	In the absence of NOM, Cu and Pb were removed for over 2000 pore volumes (PV), whereas Zn and Cd for several hundreds PV. In the presence of NOM, Cu and Pb removal diminished but that of Zn and Cd decreased only slightly.	Charbonnet et al. [43]
Al based drinking water treatment residual pyrolyzed and granulated with bentonite and corncob (DBG).	Synthetic stormwater containing Pb, Cu, Cd, PO_4^{3-}, with and without PAHs were added to columns every day for (15 storm events, 7 days drying period between each event) to columns having 4 layers (L) (bottom to top: support L, transition L, packing L, liquid distribution L). Packing layer 9.7 cm depth of sand + DBG 1:1). Surface velocity of influent solution 15 cm h^{-1} for 6 h a day.	Pb, Cu, Cd, PO_4^{3-}	The DBG column reduced influent PO_4-P from an average of 1.03 to 0.14 mg L^{-1} (due to Al in DRG) and achieved Cu, Pb, Cd removal efficiencies of all > 90% (due to CEC provided by bentonite/corn cob in DBG). Metals removal increased slightly when PAHs were present, due to PAHs mediated metal adsorption resulting from complexation of metals by adsorbed PAHs.	Wang et al. [44]
Wood mulch (WM) and Wood mulch coated with Fe based water treatment residual (WM-Fe-WTR).	Flow-through column studies with 5.1 and 10.2 cm filtration bed depths were performed using synthetic stormwater (100 µg/L Cu^{2+}, 100 µg/L Pb^{2+}, 600 µg/L Zn^{2+}, 3000 µg/L PO_4^{3-}-P). Fe-WTR to WM mass ratio 1:2. Column top layer 7.6 cm high glass beads, Filter bed 5.1 or 10.2 cm, Bottom 2.5 cm glass beads. 14 d treatment of 47 L water (239 and 120 bed volumes for the 2 bed depths.	Cu, Pb, Zn, PO_4^{3-}	WM-Fe-WTR performed better than WM for all pollutants and for both bed depths. In the 10.2 cm bed, 21% to 25% higher reductions were observed for metals and 8% higher reduction for P. Percent reduction ranking: Pb > Cu > PO_4^{3-} > Zn.	Sidhu et al. [45]
Zeolite (synthesized from illite).	Bioretention system 600 mm deep consisting of: top to bottom: a soil layer 90 mm, a filler 240 mm (gravel/soil), sand 60 mm, gravel 120 mm. Top layer: only soil, soil:zeolite 3:1 or 5:1. Synthetic stormwater added intermittently for 17 days.	Pb, Cu	Removal efficiencies of the system with zeolite for Pb and Cu were 44.4% and 44.9%, respectively, all of which were higher than those of the system without zeolite. Zeolite treatment effectively immobilized the trapped metals mostly in the top 10 cm and inhibited the trapped metals migrating to lower depths.	Shah et al. [46]

Table 4. Substrate adsorbents used in removing primarily OMPs in dynamic filtration processes (column, pilot-plant, microcosm or field studies).

Substrate Adsorbent	Method (Treatment, Water, Duration)	Pollutants	Results	Reference
Sand amended with biochar (BS) or granular activated carbon (GACS), Sand unamended (SU)	Laboratory column study: 14 cm depth geomedia with top and bottom 1 cm sand. Synthetic stormwater upward flow (1.84 mLmin^{-1}) 600 pore volumes (PV) of total flow.	Vehicle-related OMPs: 1H-benzotriazole, NN-iphenylguamidine, Hexamethooxymel Amine, (persistent mobile toxic (PMT).	C/C_0 breakthrough values for any PV lower for GACS than for BS for all three PMTs. The material life-time simulated by extrapolating the column data for BS was 30 yrs compared to 50 yrs for GACS. Higher GACS removal driven by hydrophobic, steric, coulombic effects.	Cruz et al. [17]
Sand mixed with activated carbon (AC) or biochars.	Synthetic stormwater containing OMPs (20 µg L^{-1}) and DOC (10 mg L^{-1}). 0.2–1% of 2 biochars or 0.4% AC to sand packed in column (12 cm depth). 8 days effluents (1–30 PV interval) collected for a total of 428 PVs after equilibrating stormwater overnight initially.	OMPs (2,4-D, TCPP, atrazine, orygalin, fibronil, benzotriazole, prometon, diuron).	Breakthrough for all OMPs in AC/sand 363–428 PV; no breakthrough within 428 PV for diuron. Biochar/sand breakthrough 100–200 BV. Case study simulations for atrazine breakthrough: 54 yrs for AC/sand, 5.8 yrs Biochar/sand. AC better substrate than biochar for OMPs but biochar cheaper and can have better removal of other pollutants (e.g., metals).	Ulrich et al. [35]
Polymer-clay complexes (Montmorillonite clay with PDADMAC or PVPeoS) and biochar.	Synthetic stormwater (10 µgL^{-1} each of OMP and trace metals). 12 cm length packing in columns of 3% adsorbent mixed with 97% sand. Barrier sand packed at top and bottom. Upflow 0,8 mLmin^{-1} (12 cmh^{-1}).	7 OMPs (2,4-D, ATR, PFOA, TEEP, DIU, FPR, PFOS) 6 trace metals (As, Cr, Cu, Zn, Ni, Cd).	Complete breakthrough (BT) within 5 Pore volumes (PV) for sand alone column for MOPs. Biochar column had no BT for most OMPs up to 150 PV tested. Polymer-clay columns complete BT near 50 PVs. They had the best removal of oxyanions As, Cr. All divalent metals well removed by all columns. NOM decreased all OMP removals but increased metals removal on poly clay column.	Ray et al. [33]
Wood biochar produced by pyrolysis and biomass gasification mixed with sand (1 to 10 weight % biochar).	Flow-through column (48 cm depth) experiments (up-flow mode at 1.2 mL min^{-1}), more than 8 months and 4000 pore volumes (PV). Synthetic stormwater (50 µg L^{-1} OMPs). Study simulates filters operated after the wetlands.	7 OMPs (atenolol, benzotriazole, dicamba, diuron, fipronil, mecoprop, terbutryn), 5 heavy metals (Cd, Cu, Pb, Ni, Zn).	Least retained OMP was dicamba (negatively charged) then fipronil and terbutryn. 20% breakthrough of dicamba was estimated at 1100 and 5300 PV Proposed that mixing biochar with other low-cost materials may enhance removal of organic and inorganic pollutants (metals, nutrients).	Spahr et al. [47]

Table 5. Substrate adsorbents used in removing mixed pollutants in dynamic filtration processes (column, pilot-plant, microcosm, or field studies).

Substrate Adsorbent	Method (Treatment, Water, Duration)	Pollutants	Results	Reference
Al-Mg-graphene oxide coated sand.	Actual surface runoff (SR) and synthetic stormwater spiked separately with individual pollutants and together. Column study (2–25 days). 26 cm substrate and 2 cm sand at top and bottom. Pore Vol 30 mL.	Caffeine, Zn, PO_4^{3-}	Mixed pollutants breakthrough: ~14,000, ~11,500, ~9500 pore volumes (PV) for PO_4^{3-}, Zn, caffeine-synthetic water; ~10,900, 6800, ~5400 PV- SR. Lower PV for SR due to competing chemical species. Life-time estimations to remove 90% PO_4^{3-}, Zn, caffeine were 81, 15, >100 yrs, respectively.	Vu and Wu [16]
AC amended sand (2%AC).	2 pilot-plant permeable pavements (PP) setup out- door open to atmosphere. Top layer 50 mm concrete blocks, then sand 100 mm, bottom gravel 100 mm. Natural stormwater (12 events, 1 yr) collected in the vicinity used.	Total N, NO_3^-, Cu, Dissolved organic carbon (DOC)	AC amended sand reduced total N and NO_3^- leaching compared to unamended sand PP by 40 ± 20% and 57 ± 15%, respectively. It also reduced Cu and DOC leaching.	Zan et al. [27]
Woodchips, straw (33%) (WCS): woodchips, biochar (33%) (WCB) + woodchips.	5 months laboratory pilot-scale study following 8 months of aging under field conditions, Synthetic stormwater spiked with 1 mg N L^{-1}, 50 μgL^{-1} each of 6 OMPs and 5 trace metals. Solutions pumped continuously into columns (50 cm depth) at 2 $mLmin^{-1}$.	OMPs (atrazine, fipronil, 2,4-D, diuron, TCEP, and 1H-benzotriazole) Ni, Cu, Cd, Pb, Zn, NO_3^-	NO_3^{-1} below detection limit for entire expt. WC acts as a C source for microbes to denitrify NO_3^{-1}. All treat. removed >80% Ni, Cu, Pb, Cd; But for Zn in WCB ~50%, in others ~ 20%. No significant breakthrough for MOPs in WCB. Others removed ~99% atrazine, 90% fipronil, but rapid breakthrough of TCEP, 2,4-D, 1 H-nenzotriazole, diuron. A diffusion-limited sorption model predicted breakthrough of OMPs to be 10,000–32,000 PV (decades of service time).	Ashoori et al. [36]

3.2.1. Nutrient Removal Using an Individual Adsorbent

Table 2 presents selected studies on the removal of nutrients from stormwater using adsorbents incorporated into treatment system substrates. Common nutrients contributing to water pollution include nitrate (NO_3^-), phosphate (PO_4^{3-}), ammonium (NH_4^+), and arsenic (As) ionic species. The primary mechanisms for removing these pollutants are ion exchange, ligand exchange, and hydrogen bonding.

For NO_3^-, which carries a negative charge, adsorbents must possess positive surface charges to facilitate adsorption. Organic wastes, biochar, and activated carbon typically have negative charges, making them ineffective for adsorbing NO_3^-. However, when activated carbon contains significant amounts of Fe and Al, it can acquire positive charges, enhancing its ability to adsorb NO_3^-, as demonstrated by Kang et al. [48]. Additionally, the carbon in organic materials like biochar can promote microbial growth, which facilitates nitrate reduction through denitrification under anaerobic conditions. Xiong et al. [37] demonstrated this method of NO_3^- removal when biochar was applied to the submerged layer of a column used for stormwater treatment (Table 2). Despite the fact that biochar contains Fe and Al, its net surface charge remains negative, as indicated by its measured zeta potential. This characteristic helps the adsorption of NH_4^+ through ion exchange.

In contrast, PO_4^{3-} is primarily removed through ligand exchange with hydroxide (OH^-), carbonate (CO_3^{2-}), and sulfate (SO_4^{2-}) ions, typically in inorganic compounds containing Al, Fe, Mn, and Ca. This adsorption mechanism is generally stronger than the non-specific ion exchange process used for NO_3^- removal (Table 2). Sidhu et al. [45] explained the higher adsorption of PO_4^{3-} by wood mulch coated with water-treatment residuals containing iron (WTR-Fe) compared to uncoated mulch. The improved removal was attributed to PO_4^{3-} binding to amorphous iron hydroxides in WTR-Fe through ligand exchange with OH^-. This results in an inner-sphere surface complexation, where PO_4^{3-} is tightly adsorbed [6]. Similarly, Nguyen et al. [49] explained the removal of arsenic anions by Fe-coated biochar via the same ligand exchange mechanism. In a constructed wetland setup, Allende et al. [41] found that zeolite substrates effectively removed As through a comparable ligand exchange mechanism, where arsenic anionic species displaced aluminol or silanol hydroxyl groups.

3.2.2. Heavy Metal Removal Using an Individual Adsorbent

Table 3 highlights selected studies that investigated the removal of heavy metals from stormwater using adsorbents integrated into the substrates of different treatment systems. Heavy metal cations are commonly removed via strong adsorption onto iron (Fe), aluminum (Al), and manganese (Mn) oxides and hydroxides. To enhance this process, low-cost materials—such as sand, organic waste products, and industrial by-products—are often coated with these metal oxides or hydroxides. Zeolite, a naturally occurring mineral, is another widely used substrate that is frequently modified with such coatings to improve its performance in stormwater metal removal.

The adsorption of heavy metals primarily involves strong interactions between metal hydroxide cationic species and the adsorbent surface, along with the surface precipitation of metal hydroxides [34]. The affinity of a metal for adsorption is largely influenced by its first hydrolysis constant (which governs the formation of MOH^+ species) and the solubility product of its hydroxide form. Metals with lower hydrolysis constants produce a greater fraction of MOH^+, which generally adsorb more effectively than their unhydrolyzed M^{2+} counterparts. Additionally, a higher-solubility product enhances the likelihood of metal precipitation, particularly on the surface of the adsorbent.

3.2.3. OMP Removal Using an Individual Adsorbent

Table 4 summarizes selected studies focused on the use of adsorbents for removing organic micropollutants (OMPs) from stormwater. Adsorption of OMPs is governed by multiple mechanisms, including hydrophobic interactions, electrostatic attraction or repulsion, hydrogen and covalent bonding, ligand exchange, Van der Waals forces, π–π interactions, and molecular diffusion into the porous structure of the adsorbent (Figure 1B). These same mechanisms also apply to natural organic matter (NOM), which is typically present in much higher concentrations than OMPs in stormwater. As a result, NOM can compete with OMPs for active adsorption sites, potentially reducing the efficiency of OMP removal [33].

Commonly used adsorbents for both NOM and OMPs include carbon-based materials such as biochar and activated carbon (AC), which tend to be hydrophobic and possess negatively charged surfaces. These materials are generally effective at removing the hydrophobic and positively charged components of NOM, as well as positively charged OMPs. However, their performance declines when targeting hydrophilic NOM and negatively charged OMPs. For instance, Spahr et al. [47] found that the herbicide dicamba was poorly removed by biochar, likely because its negative charge led to electrostatic repulsion from the negatively charged surface's functional groups on the biochar.

To broaden the range of pollutant types that can be effectively adsorbed, it is advisable to incorporate adsorbents with differing surface chemistries—specifically, combining materials that have hydrophilic properties and positive surface charges with those that are hydrophobic and negatively charged. This approach can enhance overall adsorption capacity and selectivity for a diverse set of NOM and OMP compounds.

3.2.4. Multiple Pollutant Removal Using a Combination of Adsorbents

This section reviews studies that demonstrate the benefits of incorporating adsorbents with complementary properties into filter media to improve the simultaneous removal of diverse pollutants from stormwater. Ekanayake et al. [50] evaluated the performance of a soil-based column filter (comprising a 50 mm soil layer sandwiched between coarse sand and gravel) in removing phosphate and three polycyclic aromatic hydrocarbons (PAHs) from synthetic stormwater. When 10% zeolite by weight was mixed with the soil, the removal efficiency of heavy metals and phosphate significantly improved. However, PAH removal remained low, ranging from 30 to 50% after 24 h. Introducing just 0.3% granular activated carbon (GAC) into the soil–zeolite mixture boosted PAH removal to over 99%. Zeolite enhanced phosphate removal through calcium phosphate ($Ca_3(PO_4)_2$) precipitation on its surface, supported by a bridging effect in which Ca^{2+} ions linked negatively charged zeolite and phosphate ($PO_4{}^{3-}$). The improved PAH removal was attributed to GAC's high hydrophobicity and capacity for π–π interactions. Although the impact of GAC alone on phosphate was not tested, other studies suggest that its performance for phosphate is limited relative to zeolite [51,52], indicating that both materials are necessary for effective multi-pollutant removal.

In a related study, Pritchard et al. [53] reported the improved removal of several OMPs—including six PAHs and various per- and polyfluoroalkyl substances (PFAS)—in a continuous-flow column filter packed with sand, zeolite, and activated carbon, operated over a three-month period. Comparable results were obtained when biochar, derived from wood pyrolyzed at high temperatures, replaced the activated carbon in the filter media, highlighting the versatility of carbonaceous materials when paired with mineral adsorbents.

Sounthararajah et al. [34] explored the removal of five heavy metals (Cd, Cu, Zn, Ni, Pb) in a 20 h column test (266 bed volumes) using a blend of sodium titanate nanofibers

(TNF) and GAC in a 25:1 ratio. This combination significantly outperformed GAC alone, especially for Ni, Zn, and Cd, for which GAC exhibited minimal adsorption. TNF, although used in small quantities due to its fine texture and resulting high pressure drop when used alone, provided most of the metal removal capacity. GAC contributed by offering mechanical support and facilitating the removal of organics like PAHs, as demonstrated in earlier studies. The synergy between TNF and GAC was essential for achieving high removal across all tested metals.

In a subsequent study, Sounthararajah et al. [54] applied TNF in a pilot-scale permeable pavement system (PPS) to treat synthetic stormwater over an 80 h period. TNF was introduced as a post-treatment in a horizontal filter filled with zeolite or basalt, mixed with GAC in a 25:1 weight ratio. Without TNF, metal removal ranged from 38 to 73%, with effluent concentrations failing to meet Australian and New Zealand guidelines [55]. When TNF was added, removal improved dramatically—77% for Ni and 99–100% for other metals—meeting marine water standards and nearly meeting freshwater criteria. These results confirm the efficacy of TNF as a supplementary adsorbent in systems aiming to meet stringent discharge standards. As adsorbents become saturated, they can be either regenerated or safely disposed of, as discussed by Baskar et al. [31].

Nagara et al. [56] developed a composite media combining aluminum-based drinking water-treatment residuals (WTR), sand, and a carbonaceous material (CM; granular non-activated carbon, ~2–3 mm particles). Following optimization in laboratory column experiments with synthetic stormwater, the media was tested under field conditions for four months across eight storm events in a parking lot. The composite media effectively removed dissolved Cu, Pb, and Zn, along with total petroleum hydrocarbons (TPH) and turbidity. WTR outperformed both sand and CM for metal removal, largely due to irreversible inner-sphere complexation, which led to a high sorption capacity and low desorption. Median metal removal efficiencies in the field were 27.4% for Cu, 36.3% for Pb, and 69.3% for Zn. CM contributed primarily to TPH removal (45.6% median efficiency) through hydrophobic, π–π, and hydrogen-bonding interactions. The dual-layer filter, consisting of 2.5 cm of WTR-amended sand over 7.5 cm of CM, provided effective turbidity removal (81.2% median) while maintaining suitable hydraulic conductivity. Although some WTR loss was observed due to the high flow rates, the combination of WTR and CM proved to be effective for treating multiple contaminants with diverse properties.

4. Challenges and Knowledge Gaps

Stormwater contains numerous dissolved and undissolved pollutants. Undissolved pollutants can be easily removed by screening and sedimentation. However, dissolved pollutants are chemically and biologically diverse. The biological pollutants, mainly microbes, are largely removed through biological processes involving soil media and plant rhizospheres in commonly practiced treatment systems.

4.1. Multiple-Adsorbent Combination Requiremen

Many chemical pollutants—both organic and inorganic—that vary in charge, molecular size, and other characteristics are removed by adsorption onto soil media. Removing all such pollutants through existing stormwater treatment methods remains a challenge. Since soil media cannot satisfactorily remove many pollutants, specific adsorbents are sometimes incorporated into the soil substrate to target additional contaminants. However, the incorporation of a single type of adsorbent only removes pollutants whose chemical characteristics are compatible with that adsorbent. Therefore, multiple adsorbents with contrasting properties need to be combined to remove a broader range of pollutants. Unfortunately, only a few studies have addressed this approach. Care should be taken that these adsorbents do cause environmental hazards, such as detriments to soil health, excessive heavy metals, OMPs, and other potentially toxic input into the land and aquatic environment.

4.2. Long-Term Column-Based Experiments on Real Stormwater Requirements

The existing studies were mostly short-term laboratory experiments conducted with synthetic stormwater. Long-term, dynamic, column-based filtration experiments using real stormwater—which contains a wide variety of pollutants—are recommended to better evaluate the performance of combined adsorbent systems. This represents a significant research gap in the field.

4.3. Cost

The cost of adsorbents used can have a wide range. Herazo et al. [57] presented the cost of 12 commonly used adsorbents in constructed wetlands and the cost of implementing them in the treatment process. The values reported varied very widely and, therefore, one should select the adsorbents that are cost-effective for practical application.

The few studies conducted on using multiple adsorbents have relied on expensive materials that are not economically viable for continuous, large-scale field applications over extended periods. Identifying cost-effective adsorbents that meet these requirements remains a major challenge. However, in certain treatment systems (e.g., PPS) where the adsorbents are not mixed with the soil, the adsorbents can be regenerated and reused to cut down the cost [31].

5. Conclusions

Stormwater harvesting and reuse schemes vary significantly between projects, but most typically consist of four main components: collection, storage, treatment, and the use of water. The level of treatment applied (primary, secondary, or tertiary) depends on the intended use of the harvested stormwater. Among these levels, tertiary treatment, which targets the removal of dissolved pollutants, presents the greatest challenge. Unlike the physical processes used in primary and secondary treatment, tertiary treatment primarily involves complex biological and chemical processes.

Dissolved pollutants in stormwater include a wide range of inorganic and organic ions and compounds of varying molecular sizes, originating from both natural sources and anthropogenic activities. These pollutants encompass nutrients (such as nitrogen and phosphorus, which contribute to algal blooms), heavy metals, biological species (such as fecal coliform bacteria), and OMPs including hydrocarbons, pesticides, pharmaceuticals, and consumer products.

Common technologies employed in tertiary-level treatment to remove dissolved pollutants include bioretention, biofiltration, constructed wetlands, rain gardens, swales, and permeable pavements. These systems often use special adsorbents in the substrate layers to enhance pollutant removal. The selection of adsorbents is a critical component of the treatment system. Numerous adsorbents have been tested for specific pollutants, but most of these studies have been conducted in short-term laboratory experiments using synthetic stormwater. In contrast, dynamic column-based filtration studies, which better simulate field conditions, have proven to be more useful for evaluating adsorbent performance.

Studies have shown that specific adsorbents can effectively remove individual pollutants, such as nutrients, heavy metals, or OMPs. However, to remove multiple pollutants simultaneously, a combination of two or more adsorbents with contrasting properties is often required. This is because each adsorbent is typically optimized to target certain pollutants based on characteristics such as charge, hydrophobicity/hydrophilicity, hydrogen bonding, π–π interactions, pore size diffusion, ligand exchange, or ion exchange.

Long-term dynamic column-based filtration experiments that use real stormwater containing a wide range of pollutants and that also use real-time performance monitoring are recommended to better evaluate the performance of these combined adsorbent systems. Such studies can provide valuable insights that can be scaled up to field operations for more effective stormwater treatment. The adsorbents selected should be cost-effective in large-scale field operations for extended periods of usage without causing any adverse environmental impact.

Supplementary Materials: The supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w17132007/s1. Table S1: Adsorbent types, properties, and types of pollutant removal. References [58–60] are cited in Supplementary Materials.

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