Potential use of composts and vermicomposts as low-cost adsorbents for dye removal: an overlooked application

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Acknowledgements

Dr R. Paradelo and Dr X. Vecino are grateful to the Spanish Ministry of Economy and Competitiveness (MINECO) for award of a *Ramón y Cajal* fellowship (RYC-2016-19286) and a *Juan de la Cierva* contract (ref. IJCI-2016-27445), respectively.

Summary

The use of composts and vermicomposts as adsorbents is an important topic of study in the field of environmental remediation. These materials are rich in organic matter and have functional groups that can interact with organic and inorganic compounds. They also contain microorganisms that can promote biodegradation of organic substances. Composts that cannot be used for agronomic purposes (owing to e.g. low nutrient levels or phytotoxicity) may be valuable for soil remediation or pollutant removal. In this review, we discuss papers on this topic, with the objective of drawing attention to the potential use of composts/vermicomposts and to recommend further investigation on this subject. Few published studies have investigated the use of composts/vermicomposts to remove dyes and other coloured compounds. However, preliminary results show that these materials are potentially good adsorbents, at least comparable to other low cost adsorbents, and that, in general, basic dyes are more efficiently removed than direct, reactive or acid dyes. The results of the works reviewed also show that dye removal takes place by adsorption mechanisms, in most studies following a Langmuir model, and that the kinetics of removal are fast and follows a pseudo-second order model. However, there remain several uncertainties regarding this application. For example, very few dyes have been studied so far, and little is known about the influence of the properties of composts/vermicomposts on the dye removal process. Moreover, the possible use of compost/vermicompost to enhance biodegradation processes has not been explored. All these questions should be addressed in future research.

Keywords: Organic waste; Waste management; Waste water; Coloured compounds; Adsorption.

1. Need to treat dye-contaminated effluents

The extensive use of dyes in many industries, such as the dyestuffs, textile, paper and plastic industries, has led to the production of huge amounts of coloured waste water (Crini 2006). In general, dyes are not readily degraded under the aerobic conditions of biological treatment plants, and the effluents discharged from these plants are usually coloured. The presence of dyes in waste water, even in small amounts, is a major problem for these industries, as colours and dyes are inherently highly visible, which greatly affects the perception of water quality by the public (Slokar and Majcen Le Marechal 1997; Crini 2006). In addition, dye compounds can interfere with the growth of aquatic organisms through absorption and reflection of sunlight (Slokar and Majcen Le Marechal 1997), and some azo dyes are also suspected of being mutagenic/carcinogenic and toxic to aquatic life (Gottlieb et al. 2003). The removal of dyes from waste water is a challenging task because of the inherent properties of these molecules, including colour fastness, stability and resistance to degradation (Sun and Yang 2003). Among the different types of dyes, reactive and acid dyes are the most problematical, as they usually pass unaffected through conventional treatment systems (Willmott et al. 1998; McKay et al. 2011).

Dye-containing effluents are currently treated in two ways: by chemical or physical methods of dye removal, or by biodegradation (Slokar and Majcen Le Marechal 1997). Among the physical and chemical techniques used for dye removal, adsorption is the most common procedure and has been shown to produce the best results for different types of dyes (Ho and McKay 2003; Jain et al. 2003). Activated carbon adsorption is one of the best available control technologies in this respect, according to the US Environmental Protection Agency (Derbyshire et al. 2001), although its use for the treatment of high volumes of waste water is restricted by its high cost. Alternative adsorbents must be found in order to reduce the cost of the treatments, and there is an abundance of literature on the use of inexpensive materials for the removal of dyes from waste water, with several review articles dealing specifically with this subject (Table 1). An adsorbent can be considered low-cost if it requires little processing, is abundant in nature or is a by-product or waste material from another industry (Bailey et al. 1999). In addition, it should ideally be efficient for the removal of a wide variety of dyes, have a high adsorption capacity and be tolerant to a wide range of physicochemical parameters of waste water (Crini 2006). Extensive reviews of low-cost adsorbents proposed for dye removal have been provided, for numerous materials, including agricultural waste, industrial waste products, biomolecules such as chitosan, peats, and more (e.g. Crini 2006) (Table 1). In contrast to the large amount of research on the use of other types of waste material as low-cost adsorbents for dye removal, existing research on the use of composts and vermicomposts for this application is relatively scarce, despite the potential advantages of using these materials.

2. Properties of composts and vermicomposts

Composts and vermicomposts are produced as a result of the transformation of different types of organic waste. Composting is the biological decomposition of the organic matter contained in wastes by mesophilic and thermophilic microorganisms, leading to stabilized organic amendments with fertilization potential (de Bertoldi et al. 1983). Vermicomposting is used to valorize organic waste, and the process relies on the action of epigeic earthworm species to accelerate biodegradation and stabilization processes (Gómez-Brandón and Domínguez 2014). Urban waste was initially the most common type of feedstock used for composting, including municipal solid waste and sewage sludge, in response to the needs of waste management in growing cities. Extension of composting and vermicomposting to the treatment of materials of agricultural and industrial origin has resulted in the worldwide use of these processes. Composting and vermicomposting are now routinely used in the treatment of municipal solid waste (Farrell and Jones 2009) and sewage sludge (U.S. EPA 2002), animal residues such as solid and liquid manures (Larney et al. 2006), agricultural waste (Mortier et al. 2016), food-processing waste such as winery or olive oil mill waste (Cegarra and Paredes 2008), and industrial waste, including textile, papermill and tannery waste (Bhat et al. 2018).

In addition to their use as waste management treatments, composting and vermicomposting also yield valuable products. Composts and vermicomposts are typically stable, safe products, with neutral or slightly alkaline pH. They are rich in organic matter (often more than 50% of weight) and, as a consequence, they are highly porous and have a low bulk density and a high water-holding capacity. They are also characterized by high concentrations of plant nutrients (N, P, K) in different forms and by large amounts and varieties of microbial communities. Thus, the composts and vermicomposts produced by these processes have many properties that make them valuable resources in several fields.

3. The use of composts and vermicomposts as adsorbents

Composts and vermicomposts have several applications, including agronomic uses as organic amendments in agricultural soils (Hargreaves et al. 2008; Diacono and Montemurro 2010), as components in soil-less horticultural substrates (Carmona and Abad 2008; Paradelo et al. 2012, 2019), and for the remediation of degraded or polluted soils (Semple et al. 2001; Paradelo et al. 2007, 2009a,b, 2011; Park et al. 2011; Huang et al. 2016). In addition, the use of composts and vermicomposts as adsorbents for the treating polluted waters is a promising field of study. Composts and vermicomposts can be produced inexpensively from by-products and waste materials, and their elaboration usually requires little processing. Compared with the cost of activated carbon (> 300 \$ m^{-3}), composts can typically be produced from urban waste at much lower cost, typically around 20-30 \$ m^{-3} (Eggerth et al. 2007; Askarany and Franklin-Smith 2014).

Composts and vermicomposts are rich in organic matter with functional groups (polar ionisable and non-ionisable groups, non-polar aromatic and aliphatic groups) that can interact with neutral, cationic and anionic compounds and provide a high adsorption capacity for a wide range of organic and inorganic substances. They are also rich in microorganisms that can promote the biodegradation of organic compounds. These characteristics make composts and vermicomposts of potential value for the treatment of waters polluted with a range of substances, including metallic elements and pesticides and organic compounds such as dyes. This represents an excellent alternative use for several types of composts that are not suitable for agronomic purposes because of e.g. phytotoxicity or low nutrient contents.

As a result of these characteristics, composted and vermicomposted materials have frequently been studied as potential biosorbents for the removal of pesticides and inorganic pollutants from water (Boni and Sbaffoni 2009; Kocasoy and Guvener 2009; Paradelo and Barral 2012; Carrillo-Zenteno et al. 2013; Barral et al. 2014; Singh and Kaur 2015; Cancelo-González et al. 2017; He et al. 2017), and many works have addressed the topic (Table 2). By contrast, few studies have considered the use of these products for dye removal, and research is still at a preliminary stage.

4. Studies on the use of compost and vermicomposts for dye removal

After running a Scopus search with the terms “dye” and “adsorption” or “removal” and “compost” or “vermicompost”, we found the 13 papers listed in Table 3. Most existing studies concern pure dye solutions (20 different molecules), and among these, methylene blue and malachite green (also known as Basic Green 4) are the most commonly studied dyes, with almost all studied only once. In addition, some studies have investigated coloured effluents from the wine industry (Paradelo et al. 2009; Pérez-Ameneiro et al. 2014, 2015), which are distinctly red and similar to the colour of commercial amaranth dye (Pérez-Ameneiro et al. 2014). Figure 1 shows the chemical structures of the dyes studied. They are all ionisable organic molecules with aromatic structures, so they can interact with the functional groups present in composts and vermicomposts either through electrostatic interactions or through non polar dispersion forces.

Regarding the chemical nature of the molecules in the research involving pure dye solutions, most studies have investigated basic dyes, which are cationic molecules at most pH values. Only three studies included more than one type of dye (Tsui et al. 2003; Jozwiak et al. 2013; Toptas et al. 2014), of which only one compared four types of dyes (Tsui et al. 2003). Anionic dyes, including acid dyes, direct dyes (molecules with high affinity for fibre) and reactive dyes (molecules that react with fibre) have been much less well studied than basic dyes. This precedence is probably due to the fact that composted materials have higher retention capacity for cations than for anions, which should result in a better performance for the removal of basic dyes relative to anionic dyes.

Approaches to the treatment of dye-contaminated waters also vary widely: some studies have included detailed adsorption studies (Kyziol-Komosińska et al. 2010; McKay et al. 2011; Jozwiak et al. 2013; Toptas et al. 2014; Bhagavathi et al. 2015; Anastopoulos et al. 2018), whereas others have only investigated dye removal without considering the adsorption process (Tsui et al. 2003; Bhagavathi et al. 2016a,b; Anastopoulos et al. 2018). In addition to studies focused on eliminating dyes from water by contact with an adsorbent, one of the studies addresses bioremediation during composting (Dey et al. 2017).

4.1 Dye removal capacity and nature of the removal process

Although simultaneous comparison of the removal of different types of dyes by composts and vermicomposts is not common, basic dyes are consistently more efficiently removed than direct, reactive or acid dyes. This has been observed by Tsui et al. (2003), Jozwiak et al. (2013) and Toptas et al. (2014) after comparison of the adsorption of several dyes on the same composts. Overall, the findings of all the research reviewed here sustains this observation (Table 4, Figure 1). As composts are mainly negatively charged, due to the presence of functional groups –COOH and –OH, stronger interaction will take place with positively charged compounds (cationic dyes), as a result of electrostatic attraction, than with anionic compounds, as a consequence of electrostatic repulsion.

Regarding the nature of the process, some studies have shown that removal takes place by adsorption mechanisms. In most studies, the adsorption process follows the Langmuir model (Kyziol-Komosińska et al. 2010; McKay et al. 2011; Jozwiak et al. 2013; Toptas et al. 2014; Bhagavathi et al. 2015; Anastopoulos et al. 2018), which assumes that adsorption takes place on a homogeneous surface by monolayer adsorption, with no significant interaction between the adsorbed molecules. This model implies that retention reaches a maximum due to the limited number of sites available for adsorption.

Regarding kinetics, there is also some agreement among these studies that adsorption of dyes follows a pseudo-second order model (McKay et al. 2011; Toptas et al. 2014; Bhagavathi et al. 2015). This assumes a chemisorption mechanism in which the rate-limiting step is the surface adsorption; it has commonly been observed that sorbent-mediated removal of pollutants follows this model (Ho and MacKay 1999). There is evidence that the removal process occurs rapidly: high removal percentages are generally reached within short contact times, in some cases of minutes. De Godoi Pereira et al. (2009) observed quantitative retention of crystal violet onto a vermicompost after 10 minutes and of methylene blue after only one minute. Anastopoulos et al (2018) also observed maximum adsorption of methylene blue after one minute of contact. For other molecules, equilibrium times of 3-5 hours were observed (Tsui et al. 2009; Jozwiak et al. 2013; Toptas et al. 2014). The faster adsorption kinetics of methylene blue may be explained by steric hindrance, as methylene blue molecules are smaller than in other dyes. This is important for the treatment of waste water in continuous flow systems, in which high reaction rates imply low contact times and higher flows, thereby increasing the capacity of water treatment systems.

Regarding biodegradation as an alternative to adsorption for dye removal, Dey et al. (2017) studied the removal of methylene blue from sugarcane bagasse during vermicomposting with *Eisenia foetida*. These researchers observed that the combined activities of earthworms and microbes led to the removal of 61% and 98% of methylene blue after 30 and 60 days, respectively.

4.2 Factors in dye removal

Among the conditions that affect the process of dye removal, the pH of the solution influences both the charge of the adsorbent and the ionic forms of the dye in solution. As expected, the effect of pH has been shown to depend on the chemical nature of the dye: the adsorption of anionic dyes increases at low pH values, with optimal values around 2-3 for acid and reactive dyes (McKay et al. 2011; Jozwiak et al. 2013; Toptas et al. 2014). In turn, adsorption of basic dyes increases with pH, with variable optimal values always over 5 (Jozwiak et al. 2013; Toptas et al. 2014; Bhagavathi et al. 2015). Basic dyes are generally best removed at neutral or slightly alkaline pH, whereas anionic dyes are optimally removed at acid pH (Jozwiak et al. 2013; Toptas et al. 2014). This occurs because anionic dyes generally have functional acid groups that are negatively charged at pH values above the pKa, and neutral at lower pH. As composts are also negatively charged, decreasing the pH will lead to a reduction in anion repulsion and therefore to potentially higher adsorption.

The properties and composition of the compost or vermicompost will obviously also have a strong influence on their capacity to adsorb dyes. Factors related to composition include pH, cation and anion exchange capacity, organic matter content and nature, and the specific surface area. In this respect, Kyziol-Komosińska et al. (2010) reported the compost had a lower adsorption capacity than several peats, probably due to the lower organic matter content and specific surface area and higher pH of the compost, as the combination of both factors will reduce the capacity to adsorb for anionic dyes. Bhagavathi et al. (2016a) compared the adsorption of crystal violet on four composts, but sound conclusions cannot be reached regarding the influence of these factors on adsorption, because of the narrow ranges of pH and OM content considered.

Regarding the role of the properties of compost or vermicomposts on their efficiency as adsorbents for dye removal, the properties of the final composts are not the only important factors. Physicochemical properties are modified during composting of organic wastes, and the process of composting/vermicomposting itself may affect the capacity of materials to remove dyes. Only one of the studies reviewed here compared dye removal with composted and non-composted waste. Thus, Anastopoulos et al. (2018) examined the effect of composting on the capacity of olive tree pruning waste to adsorb methylene blue. These researchers found that the maximum adsorption capacity was 250 mg g^{-1} for composted material and 130 mg g^{-1} for non-composted material, indicating that composting greatly improved the adsorptive properties of pruning waste. Similar findings may be obtained with other materials and dyes, as some researchers have observed that composting can increase the capacity of waste to adsorb other pollutants (Liu et al. 2018). The maturity of the compost may also affect its capacity for dye removal, as the nature and composition of organic matter evolves during composting. In this respect, Lashermes et al. (2010) observed that the maturity of compost modifies its capacity to adsorb organic pollutants, and the same may be true for organic dyes.

4.3. Application to coloured effluents

In addition to pure dye solutions, some studies have addressed the treatment of coloured effluents from the winery industry, which are rich in natural colorants. The compounds producing the colour of red vinasses include polyphenols, melanoidins produced by the reaction of sugars and proteins with furfurals (Pant and Adhleya 2007). Pérez-Ameneiro et al. (2014, 2015) used an adsorbent based on composted grape marc, immobilized in calcium alginate beads, to remove pigments from vinasses. The immobilized composted grape marc was able to eliminate between 95% and 100% of the pigments present in winery effluents. As with pure dye solutions, the adsorption process followed a pseudo-second order kinetic model (Pérez-Ameneiro et al. 2014); the adsorption equilibrium process was described by the Freundlich isotherm (Pérez-Ameneiro et al. 2015). Paradelo et al. (2009c) used several non-conventional low-cost natural adsorbents to remove the colour from red wine vinasses. Among the materials assayed, grape marc vermicompost produced the best results (80% colour removal), comparable to those achieved with activated carbon. The good results obtained by the vermicompost in this study may be partly related to its high organic matter concentration (91%). Nevertheless, this was not the only factor, as composted pine bark also has a high organic matter content (98%), but it did not yield a very high level of colour removal (below 15%). The nature of organic matter is therefore expected to play an important role in the performance of the adsorbent.

4.4 Pre-treatment and possible modifications of biosorbents

In most works, pre-treatment of the composts/vermicomposts before use as biosorbents is not necessary or is limited to simple conditioning operations such as air-drying or sieving. Washing some composts before use has been recommended (Toptas et al. 2014) in order to remove excessive amounts of soluble C, which can affect negatively the dye removal process. However, composts and vermicomposts may also represent a source of more efficient biosorbents via modifications that increase the adsorption capacity of the original material. For example, in a study with winery effluents, Paradelo et al. (2009c) observed that grinding or boiling the biosorbents before treatment improved colour removal, as a consequence of the increased surface area, in the first case, and temperature-mediated activation in the second.

On the other hand, there is also evidence that activated carbon or char produced from compost could also be used for dye removal (Qian et al. 2008; Yang et al. 2016). Qian et al. (2008) studied the adsorption of methylene blue on activated carbon prepared from composted cattle manure and found maximum adsorption capacity of around 500 mg g^{-1} (higher than those reported for other composts), due to the high surface area and large mesopore volume. Yang et al. (2016) observed that increasing the temperature of carbonization had contrasting effects on the adsorption capacity of a vermicompost, increasing the adsorption of Congo red (a direct dye) and decreasing that of methylene blue. Carbonisation of composts will probably increase the dye removal capacity, at least for some molecules; however, unfortunately, neither of these studies compared the performance of the original compost/vermicompost with that of the charred products.

Therefore, the available direct evidence is not sufficient, and further studies should compare the adsorption capacity of charred and non-charred materials for dyes, as done for other organic pollutants (Tsui and Juang 2010).

4.5 Limitations and future development

Overall, this review of existing studies show that there remain some uncertainties regarding the potential application of composts and vermicomposts for dye removal. Further research is therefore necessary in order to overcome these limitations. The small number of studies, in addition to the variable experimental conditions (common in adsorption studies), make it difficult to reach sound conclusions about this issue. Varying conditions of ionic strength, pH or solid/liquid ratios during adsorption experiments are problematical as regards the valid comparison of different studies. Moreover, the study results are not always reported in the same way: although in most cases complete adsorption parameters are provided, some researchers only report removal percentages. Another drawback is the scarcity of results regarding the removal of each dye, with the exception of methylene blue. The studies reviewed here involve a total of 20 dyes (Table 3), and although some researchers report the removal of more than one dye by the same adsorbent and under comparable conditions, additional studies are necessary before strong conclusions can be reached about each compound, in particular direct dyes.

In most of the studies reviewed here, details are not given about the feedstock used to produce the composts or the characteristics of the composting process, despite the potential influence of these factors on the performance of the adsorbents. Future studies should include more detailed information about the composting process, the feedstock used and the maturity of the materials used for dye removal. Additional studies should be conducted to compare composted/vermicomposted and untreated waste.

However, studies comparing the performance of several composts with different properties on the removal of one or more dyes remain scarce. In the absence of direct comparison of dye removal by several composts, indirect inference by comparison of the results from different studies may also be helpful. Unfortunately, these studies do not always provide details of the characteristics of the composts used, so comparison is difficult. There is therefore a need for further studies that compare the removal of one or more dyes by several composts under comparable experimental conditions.

Finally, most of the studies reviewed consider adsorption as the removal method, although this is not the only possible mechanism of removal during contact between dye solutions and compost. The main processes regulating dye removal are adsorption and biodegradation. Although the factors that affect adsorption are well known, because they have been clearly established for organic pesticides, including the nature of the molecule (ionizable or non-ionizable, pKa, solubility, etc.), biodegradation is also important and has been less well studied. Indeed, among the studies reviewed here, only one considered the biodegradation process. The contribution of biodegradative mechanisms to dye removal should also be explored further.

5. Conclusions

Composts and vermicomposts have properties that make them potentially valuable for use as adsorbents of a range of substances, including metallic ions and pesticides and organic compounds such as dyes. However, this last application has received little attention, especially relative to the numerous studies concerning other types of biosorbents, and it represent a promising line of research. The results of the studies reviewed here show that:

- Basic dyes are more efficiently removed than direct, reactive or acid dyes.

- Dye removal takes place mainly by adsorption mechanisms, in most studies following a Langmuir model.
- Kinetics of adsorption are fast and follows a pseudo-second order model.
- pH during dye removal and properties of the composts/vermicompost such as organic matter content and nature are important factors conditioning the efficiency of the process.

Moreover, these studies highlight both the unexplored potential of the process of dye removal by adsorption onto composts and vermicomposts and the need for further investigation of the processes leading to colour removal. In particular, review of the existing literature suggests significant research gaps:

- Very few dyes have been studied to date, and additional research on dyes of all classes is required to determine which compounds or groups of compounds are most suitable for this treatment.
- Further studies evaluating the removal of one or more dyes by several composts under comparable experimental conditions are also required in order to obtain further information about the role of the properties of composts on the process of dye removal.
- In this respect, a more detailed characterization of the composts and vermicomposts used in dye removal studies is also necessary, as well as more complete descriptions of the feedstocks and the characteristics of the process of composting or vermicomposting in each case.
- Finally, studies on the use of biodegradation mechanisms involved in removal are also necessary.

Future research on the use of compost and vermicomposts as low-cost sorbents for dye removal should focus on resolving these shortcomings.

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FIGURE CAPTIONS

Figure 1. Molecular structures of dyes used in the studies reviewed here.

Figure 2. Maximum adsorption capacities (Qm, from the Langmuir model) reported for basic, acid, reactive and direct dyes on composts and vermicomposts.

Table 1. Review papers published between 2005 and 2018 dealing with the use of low-cost adsorbents for dye or colored compounds removal.

Reference	Adsorbent	Dye classes
Ahmad and Danish (2018)	Banana waste	*Acid dyes*: orange 52 (Methyl orange), Egacid Orange II, Brilliant blue *Basic dyes*: blue 9 (Methylene blue), green 4 (Malachite green), violet 10 (Rhodamine B) *Direct dyes*: red *Other dyes*: Safranin, Novacron blue FN-R, Methyl violet, Methyl red
Momina et al. (2018)	Clays	*Acid dyes*: blue 193, orange 52 (Methyl orange) *Basic dyes*: red 2, green 4 (Malachite green), blue 9 (Methylene blue) *Reactive dyes*: black 5
Pandey (2017)	Clays	*Other dyes*: Amido black 10B
Sulyman et al. (2017)	Agricultural wastes	*Acid dyes*: blue 25, blue 92, orange 52 (Methyl orange) *Basic dyes*: red 29, blue 9 (Methylene blue), violet 3 (Crystal violet), green 4 (Malachite green), violet 10 (Rhodamine B) *Direct dyes*: red 28 (Congo red) *Reactive dyes*: Remazol brilliant yellow *Other dyes*: Methyl violet, Methyl red, Red MX 3B, Cibacron yellow
De Gisi et al. (2016)	Agricultural and household wastes, industrial waste, soil and ore materials, metal oxides and hydroxides	*Acid dyes*: yellow 36, blue 92, blue 25, blue 80, blue 264, red 14, red 114, orange 10, orange 52 (Methyl orange), violet *Basic dyes*: blue 9 (Methylene blue), blue 69, red 22, green 4 (Malachite green), violet 10 (Rhodamine B), red 9 *Direct dyes*: red 28 (Congo red), red 80, red 81, blue 71 *Reactive dyes*: Brilliant green *Disperse dyes*: orange 25
Adegoke and Bello (2015)	Agricultural wastes	*Acid dyes*: violet 17, red 119, blue, blue 15 *Basic dyes*: blue 9 (Methylene blue), yellow 21, violet 10 (Rhodamine B), green 4 (Malachite green) *Direct dyes*: F. Scarlet, red 23, red 28 (Congo red) *Reactive dyes*: red 23, blue 19 *Other dyes*: Red brown C4R
Kharat (2015)	Agricultural wastes	*Acid dyes*: green 20, orange 7, blue 29 *Basic dyes*: blue 3, blue 9 (Methylene blue), violet 1, violet 3 (Crystal violet), violet 10 (Rhodamine B), Rhodamine 6G, green 4 (Malachite green) *Direct dyes*: red 28 (Congo red), orange 26 *Reactive dyes*: red 3 BS, red 2, red 120, red 198, red 228, orange, orange 16, black 5, Turquoise blue QG, Remazol black B, Remazol brilliant blue R, Remazol brilliant red *Disperse dyes*: red 1 *Other dyes*: Erichrome black T, Dark green PLS, Coomassie brilliant, Methylene red, Ethylene blue, Acridine orange, Aniline blue, Safranine O, Alpacide yellow, Tartrazine
Geetha and Velmani (2015)	Agricultural wastes, industrial waste	*Basic dyes*: yellow 21 *Direct dyes*: red 28 (Congo red) *Reactive dyes*: orange 107, Remazol yellow
Koay et al. (2014)	Agricultural wastes	*Reactive dyes*: black 5
Ong et al. (2014)	Agricultural wastes, industrial waste, clays	*Acid dyes*: Egacid Orange II, blue 74 (Indigo carmine), orange 52 (Methyl orange) *Basic dyes*: blue 3, blue 9 (Methylene blue), red 46, yellow 21, violet 3 (Crystal violet), violet 10 (Rhodamine B), green 4 (Malachite green) *Direct dyes*: navy blue 106, blue 86, yellow 12, red 28 (Congo red) *Reactive dyes*: blue 19 *Other dyes*: Lanaset grey G, Astrazon yellow

Yagub et al. (2014)	Agricultural wastes, industrial waste, clays	*Acid dyes*: yellow 23, yellow 36, yellow 132, blue, blue 25, blue 29, blue 80, blue 193, blue 256, blue 264, red 18, red 27, red 73, violet, violet 17, orange 7, orange 10, orange 52 (Methyl orange), green 25 *Basic dyes*: blue 3G, blue 9 (Methylene blue), blue 69, blue 86, yellow 2, red 13, red 18, red 46, green 4 (Malachite green), violet 3 (Crystal violet), violet 10 (Rhodamine B) *Direct dyes*: brown 2, red 23, red 28 (Congo red) *Reactive dyes*: black 5, orange 16, blue 19, red 4, red 5, Brilliant green, Brilliant red HE-3B, Remazol black, Brilliant blue *Disperse dyes*: red 1 *Other dyes*: Eriochrome black T, Astrazone black, Naphthol green B, Indosol black, Methyl violet, Yellow X-GL, Vat red 10, Vat orange 11
Bello et al. (2013)	Various sand types	*Basic dyes*: blue 9 (Methylene blue), green 4 (Malachite green) *Other dyes*: Neutral red dye, Coomassie blue**,** Safranin orange
Doke and Khan (2013)	Agricultural wastes, industrial waste	*Acid dyes*: violet 54, orange 52 (Methyl orange), Egacid Orange II *Basic dyes*: orange, blue 3, blue 9 (Methylene blue), yellow 28, violet 3 (Crystal violet), green 4 (Malachite green) *Direct dyes*: red 28 (Congo red) *Reactive dyes*: black 5, Brilliant red HE-3B, Remazol brilliant orange 3R *Other dyes*: Methyl violet, Eriochrome black T, Neutral red, Tartrazina, Amaranth
Ahmad et al. (2012)	Agricultural wastes	*Acid dyes*: green 25, black 26, blue 7 *Basic dyes*: blue 9 (Methylene blue) *Reactive dyes*: Remazol
Sharma et al. (2011)	Agricultural wastes, industrial waste, clays	*Acid dyes*: yellow 23, yellow 36, yellow 99, yellow 117, blue, blue 9, blue 25, blue 29, blue 74 (Indigo carmine), blue 80, blue 113, blue 264, orange 7, orange 10, orange 51, blue 80, red 91, red 114, brown 283, violet, Brilliant blue, Egacid orange II, Egacid red G, Egacid yellow G *Basic dyes*: blue, blue 4, blue 9 (Methylene blue), blue 41, blue 69, violet 1, violet 3 (Crystal violet), violet 10 (Rhodamine B), green 4 (Malachite green), yellow, yellow 28, red, red 18, red 22, red 46, brown 1 *Direct dyes*: yellow 11, yellow 12, yellow 28, yellow 50, red 12b, red 28 (Congo red), red 89, black, black 19, brown *Reactive dyes*: blue 2, yellow 2, yellow 23, red, red 2, red 4, red 120, red 141, Ramazol yellow, Ramazol back, Ramazol red *Disperse dyes*: blue 79, red 1 *Other dyes*: α-picoline, Safranine, Midlon black VL, Brilliant green, Polar yellow, Polar blue RAWL, Methylene yellow, Methyl violet, Methyl red
Ahmad et al. (2011)	Agricultural wastes	*Basic dyes:* blue 9 (Methylene blue), green 4 (Malachite green) *Direct dyes*: blue 71, red 28 (Congo red) *Reactive dyes*: black 5, red E *Disperse dyes*: blue, red
Ahmaruzzaman (2010)	Industrial wastes	*Acid dyes*: red 91, blue 9, blue 29, Egacid orange II, Egacid red G, Egacid yellow G *Basic dyes*: blue 9 (Methylene blue), violet 3 (Crystal violet), violet 10 (Rhodamine B), green 4 (Malachite green) *Direct dyes*: red 28 (Congo red) *Other dyes*: Rosaniline hydrochloride, Midlon black VL, Orange-G
Rafatullah et al. (2010)	Agricultural wastes, industrial waste, clays	*Basic dyes*: blue 9 (Methylene blue)
Demirbas (2009)	Agricultural wastes	*Acid dyes*: blue 15, blue 74 (Indigo carmine), red 119, violet 17, violet 49, orange 52 (Methyl orange) *Basic dyes*: blue 9 (Methylene blue) *Direct dyes*: blue 86 *Other dyes*: Erythrosine, Quinoline yellow

Gupta and Suhas (2009)	Agricultural wastes, industrial waste, clays	*Acid dyes*: brilliant blue, blue, blue 9, blue 25, blue 29, blue 40, blue 74 (Indigo carmine), blue 80, red 88, blue 92, blue 113, blue 193, blue 256, blue 264, red 1, red 14, red 18, red 51, red 73, red 88, red 114, yellow, yellow 11, yellow 17, yellow 36, yellow 99, yellow 117, yellow 132, brown, brown 283, black 26, green 25, orange 7, orange 10, orange 12, orange 52 (Methyl orange), violet, Ethyl orange *Basic dyes*: blue 3, blue 6 (Meldolás blue), blue 9 (Methylene blue), blue 47, blue 54, blue 69 (Astrazone blue), green 4 (Malachite green), red 2, red 13, red 18, red 22, red 29, red 46, yellow 21, yellow 24, orange 2 (Chrysoidine G), violet 3 (Crystal violet), violet 10 (Rhodamine B), violet 14 (basic fuchsin) *Direct dyes*: black 168, brown, brown 1, red, red 12b, red 23, red 28 (Congo red), red 79, red 80, red 81, red 89, blue, blue 86, yellow 12, green 26 *Reactive dyes*: black B, black 5, blue 2, blue 19, blue 114, yellow 2, yellow 23, yellow 64, yellow 86, yellow 176, Levafix, green 12, orange 16, red X6BN Sandoz, red 2 red 120, red 124, red 141, red 189, red 222, red 239, Remazol golden yellow, Remazol red BB, Remazol black B, Remazol blue *Disperse dyes*: blue, red, red 1, orange 25 *Other dyes*: Metomega chrome orange, Sella fast brown H, Brilliant Red E-4BA, Vat blue 4
Wang and Wu (2006)	Industrial wastes	*Acid dyes*: blue 9, blue 29, blue 40, red 1, red 88, red 91, Egacid orange II, Egacid Red G, Egacid yellow G *Basic dyes*: blue 9 (Methylene blue), violet 3 (Crystal violet), violet 10 (Rhodamine B) *Direct dyes*: red 28 (Congo red) *Other dyes*: Rosaniline hydrochloride, Midlon Black VL
Crini (2006)	Agricultural wastes, industrial waste, clays, soil materials, peat	*Acid dyes*: yellow, yellow 17, yellow 36, yellow 99, yellow 117, yellow 132, blue, blue 9, blue 25, blue 29, blue 40, blue 80, blue 113, blue 193, blue 256, blue 264, red 4, red 18, red 73, red 88, red 114, violet, violet 17, orange 10, orange 12, orange 52 (Methyl orange), Ethyl orange, green 25 *Basic dyes*: yellow, yellow 21, yellow 24, red 2, red 13, red 18, red 22, red 29, red 46, blue 9 (Methylene blue), blue 47, blue 69, green 4 (Malachite green), violet 3 (Crystal violet), violet 10 (Rhodamine B), violet 14 (basic fuchsin) *Direct dyes*: red, red 28 (Congo red), red 81, brown 1, yellow 12 *Reactive dyes*: red, red 2, red 4, red 5, red 120, red 124, red 141, red 189, red 222, red 239, E-4BA, yellow 2, yellow 23, yellow 64, yellow 86, yellow 176, yellow 208, blue 2, blue 19, blue 114, black 5, orange 16, orange 107, Remazol yellow, Remazol BB, Remazol blue *Disperse dyes*: red 1 *Other dyes*: Alizarin sulfonic, Sella fast brown H, Methyl violet
Crini (2005)	Synthetic biopolymers	*Acid dyes*: blue 113, yellow 36, red 114 *Basic dyes*: violet 3 (Crystal violet) *Direct dyes*: red 28 (Congo red) *Reactive dyes*: red 2, red 189, red 141, red 189, blue 2

Table 2. Papers on compost and vermicompost use as adsorbents for pollutant removal.

Pretreatment	Composition of compost	Contaminant	Reference
Eisenia fetida earthworm (40 days)	-Sewage sludge from the second municipal wastewater treatment plant. -Different additive materials (soil, straw, fly ash, and sawdust) were mixed with sludge.	Heavy metals (Pb (II) and Cd (II)) and tetracycline (TC)	He et al. (2017)
Pyrolysis at 300ºC, 500ºC and 700ºC	Vermicompost biochars	17β-estradiol	Wu et al. (2016)
Slow pyrolysis	Vermicompost biochar	Rhodamine-B	Wang et al. (2015)
E. fetida earthworm	Cattle dung vermicompost	Cu, Mn, Fe, Zn	Singh and Kaur (2015)
Pyrolysis at 400-700ºC	Vermicompost biochar	Heavy metal ions, dyes and organic contaminants	Yang et al. (2015)
	Temple wastes: vegetable crop residues, grass residues, dry mango leaf litter, regular farmyard manure and cow dung vermicompost	Cd	Pradhan et al. (2014)
	Commercial cattle manure vermicompost	Cd	Carrillo Zenteno et al. (2013)
Dried in an oven at 60ºC, and sieved less than 150 µm	Commercial vermicompost as gardening humus	Methylparathion	Mendes et al. (2012)
Air-dried for 72 h and sieved a 2 mm	Cattle manure vermicompost	Cu (II), Cd (II)	Jordão et al. (2011)
E. fetida earthworm Sun-dried and sieved to 300 µm	Vermicompost	Cr, Pb, Ni	Parra et al. (2010)
Air-dried for 72 h	Cattle manure vermicompost	Al (III), Fe (II)	Jordão et al. (2010)
Dried at 70ºC for 4h	Cattle manure vermicompost	Zn (II)	Jordão et al. (2009)
	Local vermicompost	Pb, Ni, V, Cr	Urdaneta et al. (2008)
Dried and sieved between 75-150 µm	Local vermicompost	Cd (II), Pb (II)	De Godoi Pereira et al. (2004)
Dried and sieved size of ≤ 150, ≤ 355 or ≤ 600 mm	Local vermicompost	Cd (II), Cu (II), Pb (II), Zn (II)	Matos and Arruda (2003)
Dried at 60°C for 24 h	Vermicompost samples were obtained from different regions of Minas Gerais and São Paulo States (Brazil)	Cd (II)	Pereira and Arruda (2003)

Table 3. Papers on compost and vermicompost use as adsorbents for dye removal.

		Colorant(s) studied				
Reference	Adsorbent	Basic dyes	Acid dyes	Reactive dyes	Direct dyes	Other
Tsui et al. (2003)	Compost	Basic Blue 9, Basic Green 4	Acid Black 24, Acid Orange 74	Reactive Orange 16, Reactive Red 2	Direct Blue 71, Direct Orange 39	
de Godoi Pereira et al. (2009)	Vermicompost	Crystal violet, methylene blue				
Paradelo et al. (2009c)	Urban waste compost, grape marc vermicompost, pine bark compost					Winery wastewater
Kyziol-Komosińska et al. (2010)	Green waste compost		Acid Blue 193, Acid Black 194			
McKay et al. (2011)	Urban waste compost			Reactive Red 234		
Jozwiak et al. (2013)	Sewage sludge green waste compost	Basic Green 4, Basic Violet 10		Reactive Yellow 84, Reactive Black 5		
Toptas et al. (2014)	Spent mushroom compost	Basic Red 18	Acid Red 111	Reactive Brown 37		
Pérez-Ameneiro et al. (2014)	Grape marc compost					Winery wastewater
Bhagavathi et al. (2015)	Kitchen waste compost	Malachite green				
Bhagavathi et al. (2016a)	Kitchen waste compost, leaf waste compost, paper waste compost, water hyacinth compost	Crystal violet				
Bhagavathi et al. (2016b)	Water hyacinth compost	Methylene Blue, Malachite Green, Basic Blue 41				
Dey et al. (2017)	Sugarcane bagasse vermicompost	Methylene Blue				
Anastopoulos et al. (2018)	Pruning waste compost	Methylene Blue				

Table 4. Summary of adsorption parameters of dyes on composts/vermicomposts. Qm: maximum adsorption capacity at the Langmuir equation; t_{eq}: contact time for maximum adsorption.

	Adsorption	Kinetics		
Dye	Q_m (mg g^{-1})	t_{eq} (min)	Optimal pH	Reference
Methylene blue	5.47	500		de Godoi Pereira et al. (2009)
Methylene blue	296	1		Bhagavathi et al. (2016b)
Methylene blue	250	1	no optimal pH	Anastopoulos et al. (2018)
Malachite green	151	300	8	Bhagavathi et al. (2015)
Basic Green 4 (malachite green oxalate)	26.41	300	5	Jozwiak et al. (2013)
Malachite green	153	240		Bhagavathi et al. (2016b)
Crystal violet	0.78			de Godoi Pereira et al. (2009)
Basic blue 41	158	10		Bhagavathi et al. (2016b)
Basic red 18	400		6	Toptas et al. (2014)
Basic violet 10/Rhodamine B	27,2	180	5	Jozwiak et al. (2013)
Basic Blue 9	0.08	180		Tsui et al. (2003)
Acid Orange 74	0.005	180		Tsui et al. (2003)
Acid red 111	141		3	Toptas et al. (2014)
Acid Black 24	0.014	360		Tsui et al. (2003)
Acid Blue 193	9.3			Kyziol-Komosińska et al. (2010)
Acid black 194	15.9			Kyziol-Komosińska et al. (2010)
Reactive yellow 84	2.15		3	Jozwiak et al. (2013)
Reactive black 5	4.79	180	3	Jozwiak et al. (2013)
Reactive Red 234	0.718	24 h	2.3	McKay et al. (2011)
Reactive Red 2	0.003	180		Tsui et al. (2003)
Levafix Braun (reactive brown 37)	169.5		2	Toptas et al. (2014)
Direct Orange 39	0.002	180		Tsui et al. (2003)