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A Sustainable Method to Reduce Vancomycin Concentrations in Water Using Timber Waste

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Abstract Emerging contaminants are becoming a significant challenge for modern society. Antibiotic resistance is notably among the most urgent public health concerns, and it is well recognised that the problem often originates from wastewater treatment facilities. In developed countries, where affordable and specified, activated carbon can be used as an efficient adsorbent of antibiotic residues present in wastewaters. However, this method is associated with high production and reactivation costs and has a significant carbon footprint. Research at Ulster University proposes a more sustainable and cost-effective solution to this problem. The technique involves the application of modified sawdust waste to treated effluent, reducing tertiary antibiotic contamination. The sawdust used in the current study was from two sources: ash or a mixture of tree species. These materials, in unmodified and modified forms, were applied as the adsorbents in model systems containing vancomycin (antibiotic) dissolved in distilled water at concentrations ranging between 30 and 150 mg/L. It was found that such adsorbents are highly efficient at removing vancomycin from contaminated waters. Antibiotic removal levels reached 63.0% ($\sigma = 2.3\%$) for the modified mixed sawdust and 56.6% ($\sigma = 5.1\%$) for the modified ash sawdust. Post-treatment, the sawdust can undergo a thermal conversion for energy production. The preliminary findings of this scoping work indicate the feasibility of converting sawdust waste into a novel means for wastewater treatment systems capable of dealing with antibiotic pollutants. The simplicity of the method offers many developing and developed economies, a sustainable means of managing this dangerous emerging problem.

Keywords Antibiotic resistance · Lignocellulosic materials · Sustainable wastewater treatment · Vancomycin · Environmental protection · Sawdust

1 Introduction

There are now increasing concerns regarding the impact of overuse or misuse of antibiotics on human health (WHO, 2020a). In addition, residual amounts of these medications are directly and indirectly discharged into the environment. Consequently, their effectiveness is diminished as bacteria metamorphose to become more resilient to antibiotic treatments (Gilbert, 2019). If antibiotic resistance continues to evolve at current rates, it is expected to kill more people than cancer by 2050 (UN, 2016). It has also been

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forecasted that 1 in 5 infections in the UK could be caused by antibiotic-resistant bacteria (Roxby, 2021). The aspiration for the UN Sustainability Development Goals (SDGs), including good health and well-being, clean water and sanitation, sustainable cities and communities, and finally responsible consumption and production, cannot be achieved unless the threat of antimicrobial resistance (AMR) is addressed (WHO, 2020b).

The prevalence of AMR has been attributed to many factors (Puvača et al., 2022), but the work of Jury et al. (2011) and Rodríguez-Molina et al. (2019) have conjectured that wastewater treatment plants (WWTPs) may pose a potential rich breeding ground for the pathogens. A review of EU CD91/271/EEC (1998) denotes no stated limits or treatment standards for the removal of antibiotics. As stated by Coimbra et al. (2021), WWTPs are not designed to treat this source of pollutants, and the non-existence of discharge standards for pharmaceuticals is the catalyst for their proliferation. Therefore, WWTPs are not currently equipped to effectively remove or accept-ingly lower the concentrations of antibiotics from the effluent (Pazda et al., 2019). Sewage is a fertile breeding and collecting ground for pathogens and antibiotics. Therein, their metabolites and bacterial interactions cause the more virulent bacteria to thrive, thereby diminishing the effectiveness of antibiotics against these microorganisms (Jury et al., 2011). Furthermore, the effect of climate change is expected to dramatically alter storm patterns, creating significant variations in discharge patterns of untreated wastewater through combined sewer overflows and storm overflow discharges (Department for Environment, Food & Rural Affairs, 2022). This will effectively exacerbate the uncontrolled release of emerging contaminants into the environment (Corada-Fernández et al., 2017). As stated already, there are no regulations in the UK or the EU to guide WWTPs to limit or appropriately control this dangerous process (Bengtsson-Palme and Larsson, 2016) .

A WWTP designer can potentially adopt different techniques to remove antibiotics and trace elements from tertiary wastewaters. The scope is a function of plant size needed and available funding. For example, chlorination at WWTPs has been shown as an effective means of removing antibiotic-resistant bacteria but has been proven to be less effective at removing the antibiotics species (Yuan et al., 2015). Zheng et al. (2017) found that UV light decreased the abundance of antibiotic-resistant genes but resulted in apoptosis, where bacterial DNAs are released into the environment. These processes alter the antibiotics or antibiotic-resistant bacteria in complex ways, whereas direct adsorption is a more efficient and safer technique (Metcalf et al., 2014). Activated carbon (AC) has been studied in depth over the past decade and has been proven to be very efficient in reducing the levels of contaminants in water, such as pharmaceutical pollutants (Mazille, 2010). However, Mulder et al. (2015) estimated the production costs of a cubic meter of activated carbon for removal of micropollutants to be in the region of 1200–1500 €. Furthermore, the operational cost of filtration through the AC is between 0.20 and 0.27 € per 1 m^3 of treated water, including the replacement/regeneration of the AC. These costs are prohibitive in many developing countries, where AMR is becoming a significant problem (Founou et al., 2017).

Recently, sawdust waste products have been considered as a medium for filtration and adsorption of polluted waters (Alidadi et al., 2018; Juela, 2022; Sayen et al., 2018; Tretsiakova-McNally et al., 2020). The sawdust obtained from wood processing and manufacturing activities represents 0.4 million tonnes per year in the UK alone (Community Wood Recycling, 2018). Compared to the AC, sawdust is a cheaper, more accessible and, potentially, a more sustainable alternative.

The research presented in this paper is focused on lowering the concentration of vancomycin dissolved in water, by means of adsorption on the particles of sawdust. Vancomycin is usually prescribed as a last-resort drug, and an increase in enterococcus vancomycin-resistant bacteria has been observed since the 1990s (McDonald et al., 2005). A systematic review of this antibiotic has highlighted outbreaks of vancomycin-resistant *Enterococcus faecium* in haematology and oncology departments in numerous countries (Ulrich et al., 2017). Unfortunately, these hospital departments typically have a significant immunocompromised population, and outbreaks have been linked to increased mortality (Ulrich et al., 2017).

The information on the concentration levels of vancomycin or the efficiency of its removal at WWTPs remains limited in the literature. Eco-toxicological bioassay evaluating the half-maximal effective concentration (EC_{50}) — i.e. the concentration of antibiotic leading to a 50% reduction in bacterial bioluminescence — found that the EC_{50} for vancomycin was in the region from 370

to 546 mg/L (Havelkova et al., 2016). Some studies have assessed the predicted no-effect concentration (PNEC) concerning the discharge of vancomycin into the aqueous environments to be in the range of 0.0006–0.0080 mg/L (Bengtsson-Palme and Larsson, 2016; Kümmerer and Henninger, 2003) . More recent publications have reported the presence of vancomycin in tested wastewaters to be at levels ranging from 0.0009 to 0.0437 mg/L in WWTP influent (Giebułtowicz et al., 2020; Tran et al., 2016) and up to 0.0085 mg/L in WWTP-treated effluent (Dinh et al., 2017). While ecotoxicity and PNEC levels differ, it remains vital to reduce the residual concentrations of antibiotics present in water discharges, particularly from hospital and pharmaceutical industry effluent (Verlicchi, 2021).

In the current paper, the authors report on a proof-of-concept study that sought to evaluate the efficiency of two types of sawdust in eliminating vancomycin from aqueous solutions with concentrations varying from 30 to 150 mg/L. With this in mind, the initial goal was to assess the removal capacities for relatively high concentrations, which may only be seen in pharmaceutical or healthcare settings (e.g. direct discharges to sewers). Furthermore, antibiotic concentrations observed in pharmaceutical plant effluent vary significantly, from 0.0005 to 31.00 mg/L (Larsson et al., 2007; Wang et al., 2021). In addition, hospital effluent is also known to be a source of antibiotic resistance, where concentrations have been seen to be ranging from 0.00002 to 0.54 mg/L (Aydin et al., 2018; Yao et al., 2021).

Sawdust has only been recently considered as an adsorbent for antibiotics (Alidadi et al., 2018; Juela, 2022; Sayen et al., 2018; Tretsiakova-McNally et al., 2020) and, hence, for a very narrow range of antibiotics. To the best of the authors' knowledge, the processes of vancomycin adsorption on sawdust particles have not been investigated previously. Therefore, this research was aimed to study vancomycin in a higher concentration interval, which, once proven to be effective at this range, will be applied to the aqueous systems with lower concentrations of the antibiotic in further works.

2 Materials and Methods

2.1 Materials and Reagents

All chemicals, reagents and solvents were purchased from Merck, formerly Sigma-Aldrich (UK), and used without any further purification. The vancomycin hydrochloride in a powdered form (pharmaceutical secondary standard) was purchased from Fisher Scientific (molecular formula: $C_{66}H_{76}Cl_3N_9O_{24}$, molecular weight: 1485.73 g/mol). The antibiotic was stored in the dark, at 4–8 °C. The chemical structure of vancomycin hydrochloride is given in Fig. 1.

The vancomycin stock solution, with a concentration of 10,000 mg/L, was prepared by dissolving a pre-weighed amount (1000 mg) of antibiotic powder in 100 mL of distilled water, in a volumetric flask. The glass volumetric flask was wrapped in aluminium foil to prevent any possible degradation of the antibiotic caused by indirect exposure to the UV light and stored in a fridge at 4–8 °C. All solutions were prepared by diluting the stock solution with distilled water.

AC, purchased from Sigma-Aldrich, was a black powder with an average particle size of 150 μm and was used as a control adsorbent. Two types of sawdust (SD) were used in this study: the sawdust from a single tree species, i.e. from ash, and the sawdust from different mixed tree species, with the main fraction originating from pine trees. The ash SD was obtained from a carpentry woodshop in Co. Monaghan (Ireland), and the mixed SD was collected from MacBlair, Coleraine (UK).

2.2 Preparation of Sawdust Adsorbents

Initially, the raw sawdust was ground to obtain smaller particles using an electrical grinder ('Sage Smart Grinder Pro'). It was then sieved with the aid of an 'Endecott 2MKII' sieve shaker, where sawdust within the 210–300 μm range was retained for further treatment. This particle size was chosen as it was found to be the most effective in similar studies involving sawdust (Acquah et al., 2016). This fraction of SD was then mixed and stirred intensively with distilled water, at room temperature, for 1 h, and then vacuum filtered. This process was repeated five times to obtain clean sawdust, without any water-soluble compounds. Finally, it was placed in a fan-assisted oven at 90 °C for 20 h to extract any excess water. The sawdust samples in this form were labelled as 'unmodified SD'.

The moisture content (W) for each sawdust material was assessed using Eq. (1) as follows:

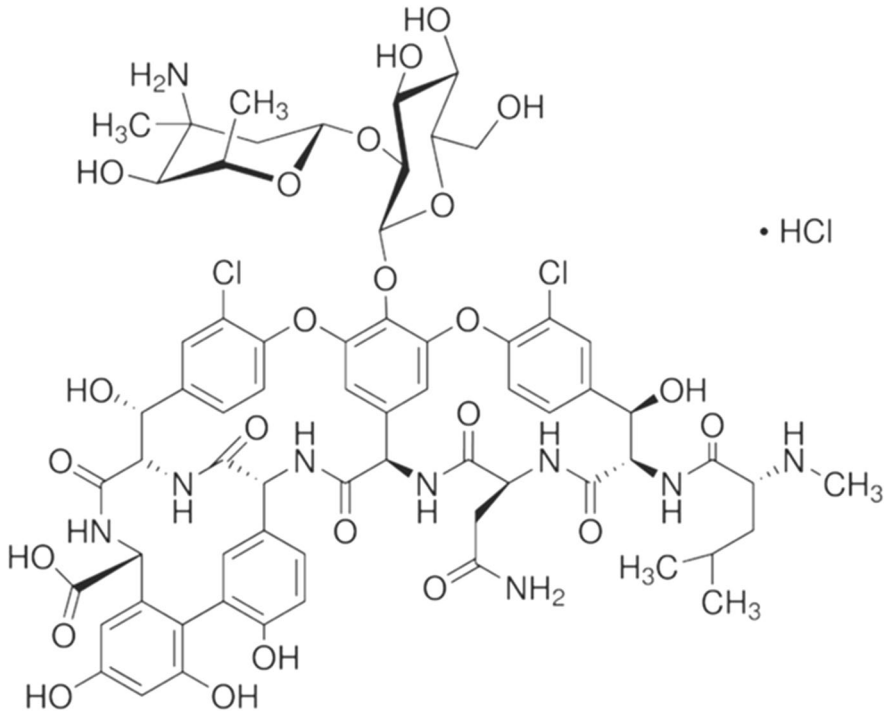

Fig. 1 Chemical structure of vancomycin hydrochloride

$$W(\%) = \frac{m_w - m_d}{m_w} \times 100 \quad (1)$$

where m_w is the mass of SD before drying in a fan oven (g) and m_d is the mass of sawdust after drying in a fan oven (g).

The SD treatment method used in this paper was adopted, with some minor modifications, from the works carried out by Akinsanmi et al. (2019), Alidadi et al. (2018) and Sayen et al. (2018). Both types of SD were modified by mixing the pre-weighed amounts with the acid or base solution, for 4 h at room temperature and for 2 h at 60 °C, in the following sequence: (1) 10 wt% aqueous solution of *o*-phosphoric acid (H_3PO_4), (2) 20 wt% aqueous solution of sulfuric acid (H_2SO_4), (3) 2.8 wt% solution of potassium hydroxide (KOH) in ethanol (Ph. Eur. Reagent, Sigma-Aldrich) and (4) distilled water. A 10 wt% solution of *o*-phosphoric acid was obtained by diluting concentrated 85 wt% H_3PO_4 (ACS reagent, Sigma-Aldrich) with the required amount of distilled water. A 20 wt% solution of sulfuric acid was prepared by diluting 97 wt% H_2SO_4 (ACS reagent, Sigma-Aldrich) with distilled water. The material was isolated by filtration and air-drying after treatment of SD with each solution. Finally, the sawdust was washed, under vacuum filtration, until the pH levels of the washings reached neutral values, followed by oven-drying at 60 °C for 24 h. The sample of sawdust used in the adsorption/filtration experiments was labelled as 'modified SD'.

2.3 Characterisation of SD

To enable a full understanding of the treatment effect on the sawdust, multiple experiements were carried out in relation with the SD characterisation. Firstly, the particle size distribution was carried out before the grinding of the raw SD using an 'Endecott 2MKII' sieve shaker and a range of sieves with mesh sizes ranging from 75 to 5000 µm. The samples (*ca.* 100,000 mg) were mechanically shaken for 2 h, according to the requirements of ISO 9276–5:2005. In addition, the moisture content of the sawdust was evaluated gravimetrically, according to CEN/TS 14,774–1:2004 by oven-drying all the materials at 110 ± 2 °C for 24 h.

Secondly, the Fourier transform infrared (FT-IR) spectra were recorded on a Thermo Nicolet FT-IR, Nexus model 470 spectrometer, for the samples of unmodified and modified SD. The spectra were obtained over a frequency range of 4000–600 cm^{-1} in the attenuated total reflectance (ATR) mode with

a resolution set at 4 cm^{-1} and 64 scans. Before spectral collection, each sample was air-dried in a fume hood and ground using a pestle and mortar to ensure a good level of homogeneity.

Thirdly, the Brunauer–Emmett–Teller (BET) method was used to determine the surface areas of sawdust with the aid of a Micromeritics Tristar 3020 instrument. The measurements were based on the adsorption of nitrogen at − 196 °C (77 K). Besides, the elemental compositions (ultimate analysis) of unmodified and modified SD were performed on a Perkin Elmer PE2400CHNS elemental analyser.

The proximate analysis evaluated the yield of the products obtained under controlled heating conditions. These parameters included volatile matter, ash and fixed carbon. Initially, each sample of SD was placed in a fan-assisted oven at 105 °C for 1 h. Once removed from the oven and cooled, the first portion (*ca.* 1000 mg) of the material was taken to determine the volatile matter, whilst the second portion (*ca.* 1000 mg) was used to evaluate the ash content. The samples of dried SD were pre-weighed in ceramic crucibles and placed in a muffle furnace (Carbolite, Hope, UK). Each of these experiments was conducted in duplicates.

The volatile matter (VM) content is the amount of sample matter that became volatile during controlled heating of the sample. Approximately 1000 mg of a sample, preliminary dried in a fan-assisted oven, was placed into a ceramic crucible and then put in a muffle furnace at 925 ± 10 °C for 7 min. After that, the crucible was taken out from the furnace and allowed to cool down to room temperature in a desiccator. The final mass was measured on a Mettler Toledo analytical balance. The VM content was calculated according to Eq. (2):

$$\mathrm{VM} = \frac{m_{\mathrm{dry}} - m_{\mathrm{furn}}}{m_{\mathrm{i}}} \times 100\% \quad (2)$$

where VM is volatile matter (%); m_{i} is the initial mass of SD, not oven-dried (g); m_{dry} is the mass of SD, oven-dried (g); and m_{furn} is the mass of SD after treatment in a muffle furnace (g).

The ash is the solid residue left after the burning of the material in a controlled manner. The oven-dried sample of SD (*ca.* 1000 mg) placed in a ceramic crucible was put in a muffle furnace at 575 ± 10 °C for 4 h. The residue obtained was taken from the furnace, cooled down to room temperature in a desiccator and weighed out using an analytical balance. The ash content was calculated from Eq. (3):

$$\text{Ash content} = \frac{m_{\mathrm{furn}}}{m_{\mathrm{dry}}} \times 100\% \quad (3)$$

where m_{dry} is the mass of SD, oven-dried (g), and m_{furn} is the mass of residue obtained after heating SD in a muffle furnace (g).

Fixed carbon content (in %) is the percentage of solid residue left after heating a sample of SD to achieve devolatilisation and is calculated from Eq. (4):

$$\text{Fixed carbon} = 100 - (W + \mathrm{VM} + \text{Ash content}) \quad (4)$$

The heating values were measured in a 'bomb' calorimeter IKA C200 (IKA, Oxford, UK) according to the standard requirements (BS EN ISO 1716: 2018). The SD (in modified or unmodified form) was compacted into a pellet, weighed out and placed in a clean and dry crucible, which was then positioned in a crucible holder of the bomb. The firing wire was connected to both electrodes and had to be in contact with the sample to ensure proper ignition. After that, 5 mL of distilled water was carefully added to the bottom of the bomb vessel. Then, it was tightly closed, filled with oxygen (up to 30 bar) and finally placed into the calorimeter. The measurements of heating values were done in duplicates on the samples of SD before and after the adsorption of the antibiotic (the SD samples post-adsorption were air-dried in a fume cupboard before pelleting).

2.4 Adsorption and Filtration Experiments

The adsorption and filtration experiments were carried out on the SD and AC particles (*ca.* 1000 mg) packed in a fixed filtration bed inside a glass column (internal diameter 22 mm), equipped with a PTFE stopcock. The filtrate flow was kept continuous using the stopcock to ensure the contact time between antibiotics and adsorbents remained identical for all experiments. A constant level of the antibiotic solution was kept above the top surface of the adsorbent. A funnel lined with a Whatman filter paper was set directly below the column to catch any larger solid particles that may accidentally have entered the filtrate. One hundred milliliters

of vancomycin solution was used per filtration run. The filtrate was collected in five 20-mL labelled plastic vials with screw caps and stored in the fridge, away from the UV light until spectroscopic analysis. Each experiment took 100 min from the start to the end of the filtration and was kept consistent between each type of adsorbent. The adsorption/filtration experiments were carried out at room temperature (20 ± 1 °C). The content of the antibiotic was analysed in the second, third and fourth portions of the filtrate, whilst the first and the last portions were discarded.

The concentration of vancomycin in solutions before the adsorption (c_i) and after the adsorption (c_e) was determined using UV–Vis spectrometry, on a Thermo Scientific instrument (Evolution 220 model). These measurements were carried out in triplicates at a wavelength of 281 nm, which corresponded to the maximum adsorption of the vancomycin dissolved in water. The concentration of vancomycin was assessed from a calibration line plotted in the concentration interval from 30 to 200 mg/L ($R^2 = 0.9959$).

The percentage of antibiotic removal from the water was calculated using Eq. (5), whilst the adsorption capacity of the adsorbent in question was evaluated from Eq. (6):

$$\text{Removal}(\%) = \frac{(c_i - c_e)}{c_i} \times 100\% \quad (5)$$

$$q_e = \frac{(c_i - c_e)V}{m} \quad (6)$$

where c_i is the initial concentration of antibiotic in water before the adsorption (mg/L), c_e is the concentration of antibiotic in a filtrate after the adsorption (mg/L), V is the antibiotic solution volume (L) and m is the mass of moisture-free adsorbent (mg).

The isotherms of adsorption for unmodified and modified sawdust samples were plotted using vancomycin solutions, in a concentration range between 30 and 150 mg/L, by adding 25 mL of each solution to *ca.* 500 mg of each type of sawdust. The mixtures of sawdust and vancomycin solutions were stirred for 2 h, at 22 ± 1 °C, using a magnetic stirrer at 100 rpm, and after 48 h, they were filtered using gravity filtration.

3 Results and Discussion

3.1 Modification of Sawdust

Previously, Tretsiakova-McNally et al. (2020) attempted to use a single agent, 2 M sulfuric acid, for 24 h, under modest heating for treatment of SD with the purpose to increase the adsorption capacity. The main components of wood (cellulose, hemicellulose and lignin) underwent a hydrolysis at elevated temperatures and under acidic conditions. Cellulosic 3D structures are stabilised by different types of bonding (hydrogen, van-der-Waals or hydrophobic interactions) between the so-called macrofibrils and microfibrils. The process of hydrolysis, catalysed by sulfuric acid, is likely to have led to the destruction of these intermolecular bonds within the cellulose fibres and resulted in the production of individual non-bonded fibres. However, it was found that aromatic structures originating from lignin may have interfered with the detection of some antibiotics in water (Tretsiakova-McNally et al., 2020).

In the current study, the treatment of SD was achieved through a consecutive use of several chemical agents: 10 wt% aqueous solution of H_3PO_4 followed by 20 wt% aqueous solution of H_2SO_4 and, finally, by 2.8 wt% solution of KOH in ethanol. The duration of treatment was also reduced compared to the one in our first report (Tretsiakova-McNally et al., 2020). The treatment of SD used in this study significantly increased the hydrophilicity of the adsorbent. It was observed that when passing the antibiotic solution through the column filled with unmodified sawdust, it took over an hour for the SD particles to saturate, whereas this was achieved within a minute for the modified sawdust. Also, the proposed treatment resulted in an almost threefold increase in the BET surface area of modified SD compared to unmodified material (Table 1).

3.2 Sawdust Characterisation

The particle size distribution analysis shown in Fig. 2 confirms that in an initial state, the mixed SD is a finer material than the ash sawdust. Before grinding, the mixed SD already contained 22% of the total material within the required particle size (210–300 μm), versus

Table 1 Characteristics of the unmodified and modified sawdust, including moisture content, BET surface area and proximate and elemental analyses

	Moisture content (*W*, wt%)	BET surface area (m^2/g)	Proximate analysis	Elemental analysis					
			VM (%)	Ash content (%)	Fixed carbon (%)	C (%)	H (%)	N (%)	S (%)
Unmodified Ash	5.23	0.7052	94.13	0.02	0.62	47.24	6.01	< 0.30	< 0.30
Modified Ash	1.49	2.0451	94.71	0.01	4.33	47.71	6.17	< 0.30	< 0.30
Unmodified mixed	5.55	0.8261	94.11	0.03	0.31	47.06	5.96	< 0.30	< 0.30
Unmodified mixed	1.19	2.1827	94.82	0.02	3.97	47.17	5.89	< 0.30	< 0.30

only 3% for the ash type (Fig. 2). Consequently, significantly more time was required to grind the initial ash SD compared to the mixed material.

The initial storage conditions impacted the moisture content (*W*) of both types of SD in their raw form, particularly for the mixed material (which was stored outdoors) with an average value of 18.0 wt% compared to 7.1 wt% for the ash sawdust. The moisture contents after the chemical treatments and oven-drying had dropped significantly for both sawdust-based adsorbents (Table 1).

The FT-IR (ATR) spectroscopy was carried out to analyse the spectral pattern of SD samples pre- and post-chemical treatment (see Figs. 3 and 4). The wavelength range between 900 and 1430 cm^{-1} corresponds to the signals of cellulose and hemicellulose, whilst the range between 1465 and 1740 cm^{-1} is associated with lignin (Ardhiansyah, 2019; Bodirlau et al., 2012). The broad bands at 3300 cm^{-1} correspond to the stretching of hydrogen-bonded hydroxyl (–OH) groups. The bands registered at 2900 cm^{-1} can be assigned to C–H stretching in methylene (CH_2) groups, and the bands at 1730 cm^{-1} and 1620 cm^{-1} are attributed to the carboxylic groups (–COOH). A set of signals in the region from 1500 to 1000 cm^{-1} is assigned to various lignin fragments such as guaiacyl and syringil units (Sayen et al., 2018). It can be demonstrated that the chemical treatment disrupted a complex system of hydrogen bonding in the cellulose, hemicellulose and lignin components of wood. Furthermore, the chemical treatment of the sawdust increased the band intensity registered at 1730 cm^{-1}, which corresponds to –C = O stretching (Ardhiansyah et al., 2019; Bajpai et al., 2012). It was evident that the adopted treatment of mixed SD fibres resulted in the smaller areas under the bands at 3346 cm^{-1} and 1029 cm^{-1}. Interestingly, this trend was not observed for the modified ash SD.

The heating values of SD before antibiotic adsorption, determined through 'bomb' calorimetry, were in the range of 15.5 to 18.5 MJ/kg (in both unmodified and modified forms). It was found that the adsorption

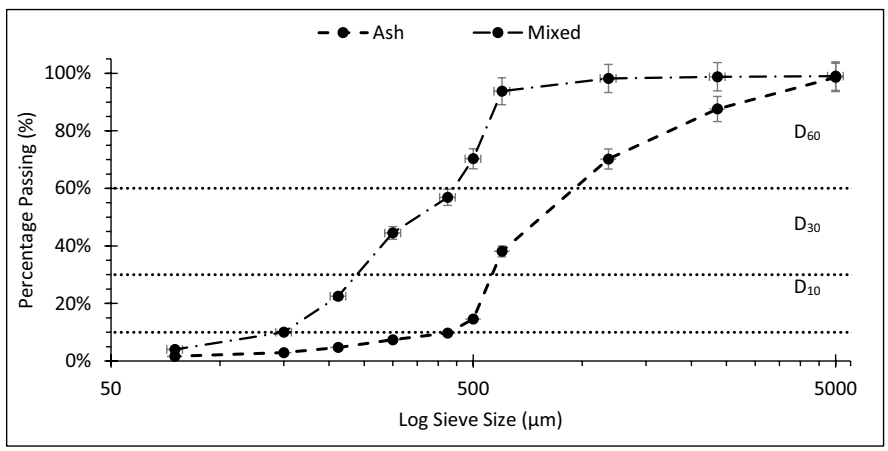

Fig. 2 Particle size distribution of the unmodified sawdust, before grinding and chemical treatments

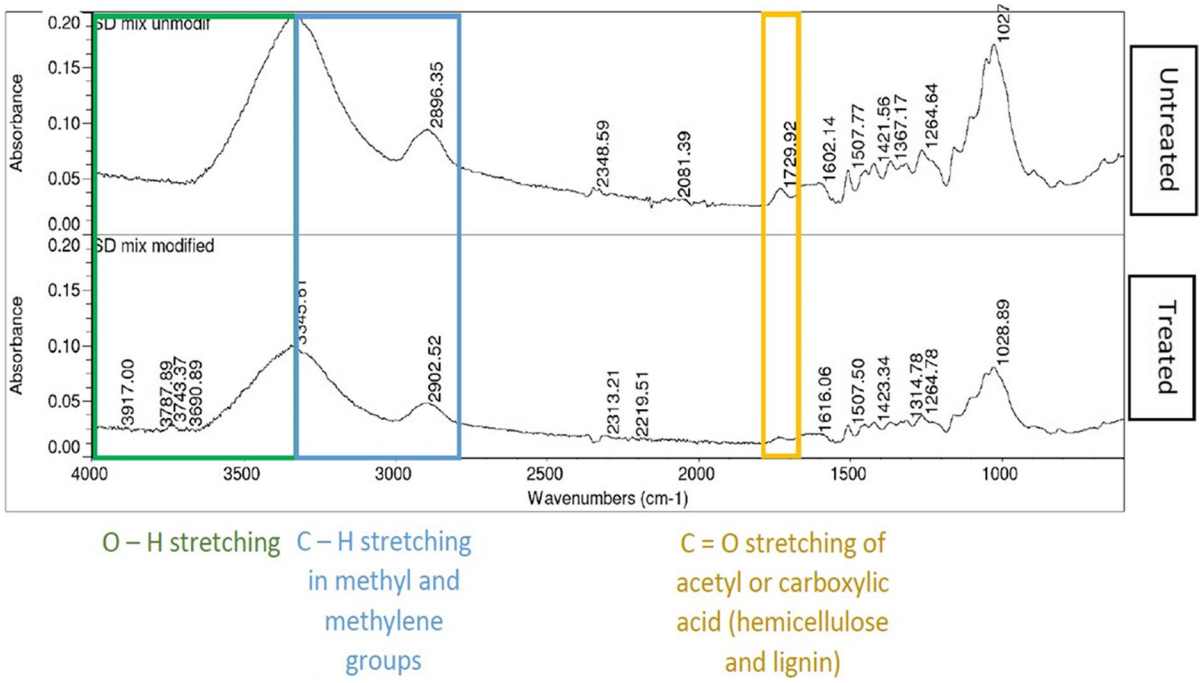

Fig. 3 FT-IR spectra recorded on untreated and treated mixed sawdust

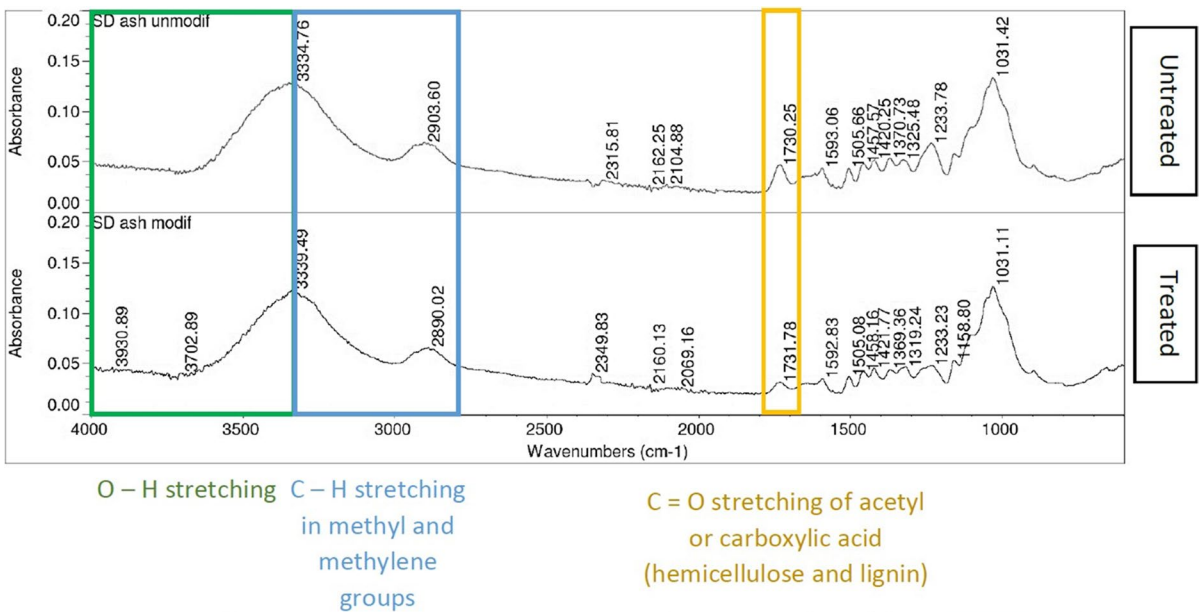

Fig. 4 FT-IR spectra recorded on unmodified and modified ash sawdust

of vancomycin virtually had no impact on these values. The heating values of SD constituted 65–90% of those characteristics for the AC (20.5–23 MJ/kg (Bouabid et al., 2013)), indicating its recycling potential.

The elemental analysis demonstrated a high carbon content (47–48%), highlighting the potential use of post-adsorption SD material as an energy source. Nominal quantities (less than 0.3%) of sulphur and

nitrogen were found in the studied samples of SD, which is a key to cleaner combustion (Table 1).

The proximate analysis indicated a high content of VM, ranging between 94.11 and 94.82% (Table 1). These values are higher than the 70–80% reported by Cheng (2018). The VM values found in the present study are closer to the characteristics of other biomass materials, for example for red pepper waste with VM percentages ranging from 93 to 96% (Maia & De Morais, 2016). The analysis in this paper also showed that the presence of the antibiotic sorbed onto SD particles did not affect the VM value (±0.5% between samples). Ash contents were found to be close to zero, 0.01–0.03 wt%, minimising the solid waste quantities left after processing.

3.3 Adsorption of Vancomycin from Water

Figure 5 shows the levels of vancomycin removal from aqueous solutions with the aid of modified forms on sawdust and AC. As expected, the control adsorbent, powdered AC, was characterised by the highest vancomycin removal level of 98.2% ($\sigma = 2.9$) which is comparable to other studies (Alacabey, 2022). The chemically modified forms of mixed SD offered a 63.0% ($\sigma = 2.3\%$) vancomycin removal from water, compared to 56.6% ($\sigma = 5.1\%$) for ash SD in the same state. The higher efficiency of powdered AC could be explained by the much smaller particles, with an average particle size of 150 µm versus 250 µm for sawdust, and the more efficient adsorption properties of the material. The unmodified sawdust provided a very low removal rate (less than 15%, $\sigma = 6.7\%$). In addition, due to the lower wettability of unmodified forms of SD, these adsorbents were not studied further.

These vancomycin removal rates were comparable to the observed values for the sorption of other antibiotics in previous studies, ranging between 70 and 98% (Akinsanmi et al., 2019; Alidadi et al., 2018; Sayen et al., 2018; Tretsiakova-McNally et al., 2020). Other adsorbent materials, such as wheat grains or rice straw biochar, were observed to have removal rates ranging between 84 and 97% (Boukhelkhal et al., 2015; Wang et al., 2017). The molecules of vancomycin are larger and heavier compared to the antibiotics used in the abovementioned studies, and therefore, its removal percentages can be considered competitive.

The evaluated adsorption capacity of the modified ash SD was 2.79, and the mixed SD was 3.11 mg/g, which is about a third of the capacity of the control adsorbent AC (Fig. 6). Furthermore, vancomycin levels in untreated wastewaters are reported to be in the range between 0.0009 and 0.0437 mg/L (Giebułtowicz et al., 2020; Tran et al., 2016) and even up to 0.0085 mg/L in treated wastewaters (Dinh et al., 2017). Therefore, the high adsorption capacity proves to be essential in maintaining a cost-effective operation.

To investigate the effect of adsorbent quantity, the same experiments were carried out but using two different sawdust quantities. It was found that using higher modified sawdust quantities increased the removal rate, with an increase from 36.2% ($\sigma = 10.2$) with 1.0 g of modified ash SD to 56.6% ($\sigma = 5.1$) with 2.0 g of the same material. The same effect was observed with the modified mixed SD, increasing

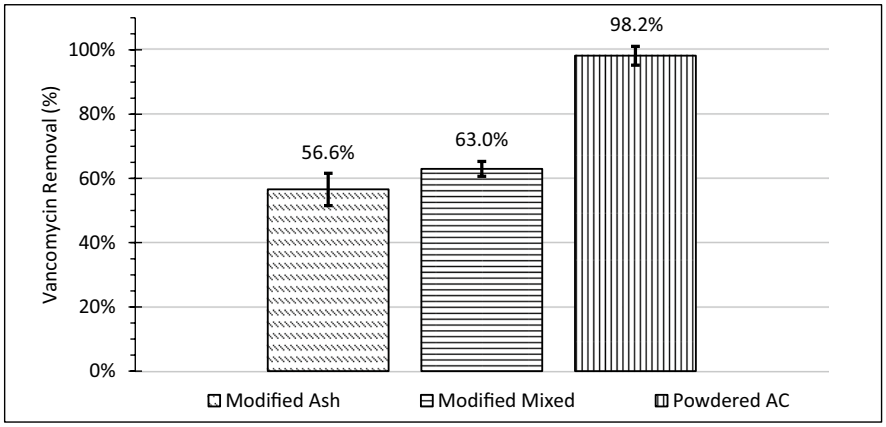

Fig. 5 Average vancomycin removal (%) of modified ash SD, modified mixed SD and powdered AC. All parameters (contact time = 100 min, temperature = 20 °C, adsorbent quantity = 2.0 g and antibiotic concentration = 100 mg/L) were identical for all the experiments

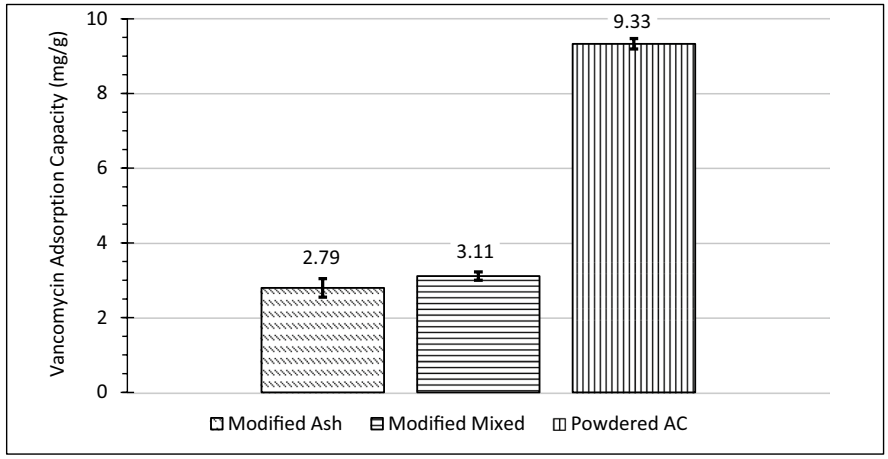

Fig. 6 Vancomycin adsorption capacity (mg/g) of modified ash SD, modified mixed SD and powdered AC. All parameters (contact time = 100 min, temperature = 20 °C, adsorbent quantity = 2.0 g and antibiotic concentration = 100 mg/L) were identical for all the experiments

from 48.2% (σ = 9.6) with 1.0 g to 63.0% (σ = 2.3) with 2.0 g (Table 2). It can also be observed that the increase in adsorbent quantity from 1 to 2 g significantly decreased the adsorption capacity of both sawdust forms. All tests were replicated for quality control.

The impact of the antibiotic concentration on the vancomycin removal from water is given in Fig. 7 in unmodified and modified forms (note: for the mixed SD, the results were variable, and clear trends could not be observed). It was noticed that the treated ash SD produced more uniform removal rates at all concentrations. The removal of the antibiotic by both forms of ash SD appears to stay in the region from 29.5 to 49.0% in the studied concentration interval. The highest removal level was 49.0% and 40.0% for the unmodified ash SD and modified ash SD, respectively. It appears that removal levels might have plateaued beyond the concentration point of 125 mg/L for the unmodified form, whilst for the modified ash SD, they remained almost constant in the studied range.

From Fig. 7, it can be observed that the removal rate stayed consistent when vancomycin concentration varies, with an adsorption capacity of 2.79 mg/g as seen in Fig. 6. If this was applied to a real-world scenario, considering an average wastewater effluent with a vancomycin concentration of 0.0085 mg/L, it is estimated that 1 g of modified ash could treat up to 330 L of wastewater.

As follows from Fig. 8, the absorption capacity for both types of ash SD gradually increases in the studied concentration range of vancomycin. At lower vancomycin concentrations, the absorption rate is between 0.85 and 1.65 mg/g, but increases up to around 4.00 mg/g at high vancomycin concentrations. The proportional increase in adsorption capacity, in relation to the increase in vancomycin concentration, confirms a consistent vancomycin removal throughout all studied concentrations.

Table 2 Vancomycin average removal rate and adsorption capacity of modified ash SD, modified mixed SD and powdered AC in 1.0 g and 2.0 g quantities

Sawdust type	Sawdust quantity (g)	Average removal rate (%)	Adsorption capacity (mg/g)
Modified ash	1.0	36.2 (σ = 10.2)	3.58
	2.0	56.6 (σ = 5.1)	2.79
Modified mixed	1.0	48.2 (σ = 9.6)	4.76
	2.0	63.0 (σ = 2.3)	3.11
Powdered AC	1.0	97.2 (σ = 2.9)	9.23
	2.0	98.2 (σ = 2.9)	9.33

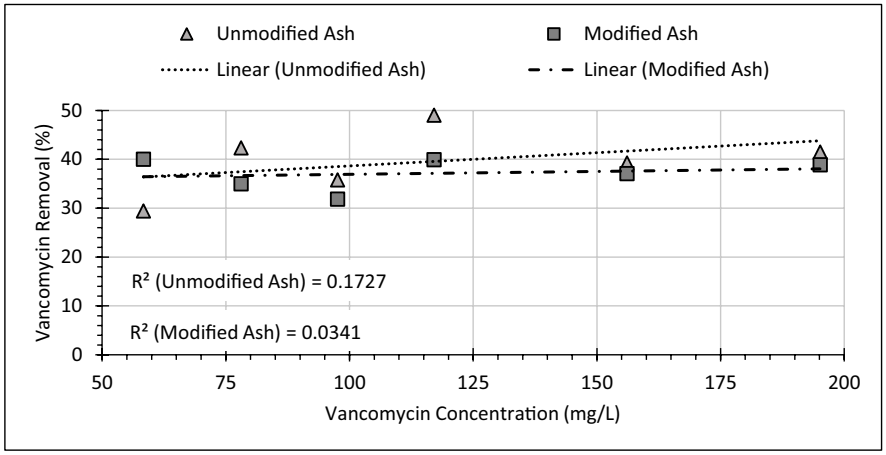

Fig. 7 Vancomycin removal (%) versus its initial concentration in aqueous solutions by the unmodified and modified forms of ash SD

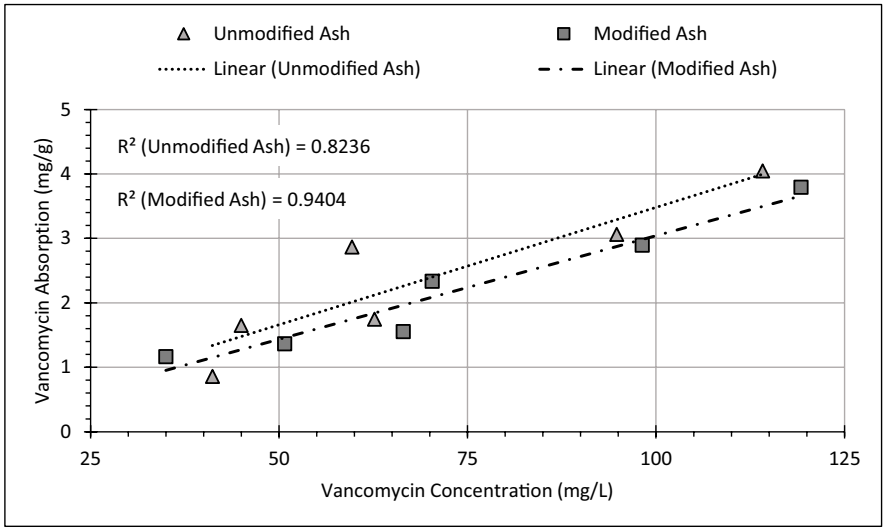

Fig. 8 Absorption capacity of unmodified and modified ash SD versus initial vancomycin concentration in aqueous solutions

3.4 Adsorption Isotherms

In the present study, the authors considered monocomponent adsorption isotherms according to Langmuir (Langmuir, 1918) and Freundlich (Freundlich, 1906) equations (Eqs. (7) and (8), respectively):

$$q_e = \frac{q_{max} \times K_L \times c_e}{1 + K_L \times c_0} \tag{7}$$

$$q_e = K_F \times c_e^{\frac{1}{n}} \tag{8}$$

where q_e is the adsorption capacity of the antibiotic at equilibrium (mg/g), q_{max} is the maximum adsorption capacity of the antibiotic (mg/g), c_e is the equilibrium concentration of the antibiotic (mg/L), c_0 is the initial concentration of the antibiotic (mg/L), K_L is the Langmuir isotherm constant (L/mg), K_F is the Freundlich isotherm constant (mg/g) and n is the non-linearity constant. The parameters found from the fitting of both models are summarised in Table 3.

This paper has used the vancomycin adsorption data for non-linear fitting to Langmuir and Freundlich isotherms, using the Excel Data Solver function, to evaluate which model would appear best suited to explain the adsorption mechanisms. The isotherms of vancomycin adsorption on the SD in the unmodified and modified forms are presented in Figs. 9 and 10.

Table 3 Parameters of Langmuir and Freundlich isotherms for vancomycin adsorption onto the particles of SD

Type of adsorption isotherm	Parameter	Unmodified ash SD	Modified ash SD
Langmuir	q_{max} (mg/g)	5.252	12,179.5
	K_L (L/mg)	0.008	2.47×10^{-6}
	R^2	0.779	0.943
	χ^2	0.379	0.165
Freundlich	K_F (mg/g)	0.138	0.028
	n	1.631	0.954
	R^2	0.761	0.839
	χ^2	0.419	0.645

The Langmuir isotherm assumes a formation of a saturated monolayer of an antibiotic on a surface having a limited number of adsorption sites. The separation factor (R_L) was calculated from Eq. (9):

$$R_L = \frac{1}{1 + K_L \times c_0} \quad (9)$$

This dimensionless value indicates the favourability of an organic compound's sorption to the Langmuir isotherm. The adsorption is favourable when $0 < R_L < 1$. It was found that the value of R_L ranges from 0.38 to 0.69 for the unmodified SD, which indicates that the Langmuir isotherm shape is favourable towards vancomycin adsorption. As for the modified SD adsorbent, the R_L remains almost constant and is very close to 1, confirming the linearity and the quality of the dataset. The maximum adsorption capacity (q_{max}), evaluated from the Langmuir isotherm model, is much higher for the modified form of SD, compared to the unmodified SD and reaches 12,179.5 mg/g, with a correlation coefficient of 0.943 (Table 3). It is also clear that the data for the adsorption of vancomycin by the modified SD fitted the Langmuir adsorption isotherm better than the data obtained on the unmodified SD.

The Freundlich isotherm assumes the adsorption characteristics on a heterogeneous surface with an unlimited number of sites for adsorption (Aksu & Tunç, 2005). The values of the constant n are linked to the adsorption intensity and could be indicative of the favourability of the adsorption. The sorption is considered to be linear when $n = 1$, whereas with $n > 1$, the shape of the isotherm is unfavourable (Worch, 2012). The n values for the adsorption of vancomycin sorption onto modified SD were below 1 (Table 3), confirming that the Freundlich adsorption isotherm shape is favourable. Conversely, the use of the unmodified SD adsorbent has resulted in n values being higher than 1 (1.631). The data for the adsorption of vancomycin onto the modified SD fitted better to the Freundlich isotherm than the antibiotic's adsorption onto its unmodified form with the correlation coefficients being 0.831 and 0.761, respectively

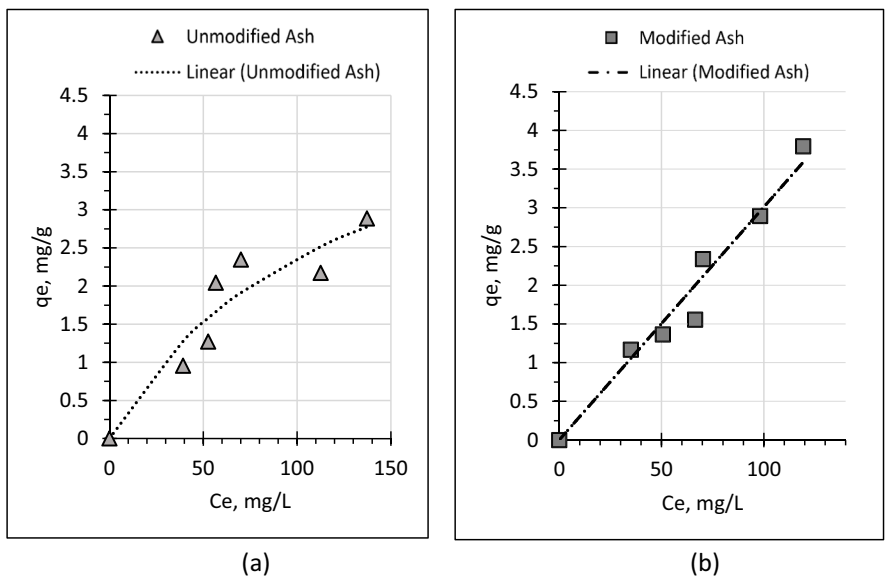

Fig. 9 Experimental data of vancomycin adsorption onto unmodified (**a**) and modified (**b**) SD fitted to the Langmuir isotherm model (dotted line)

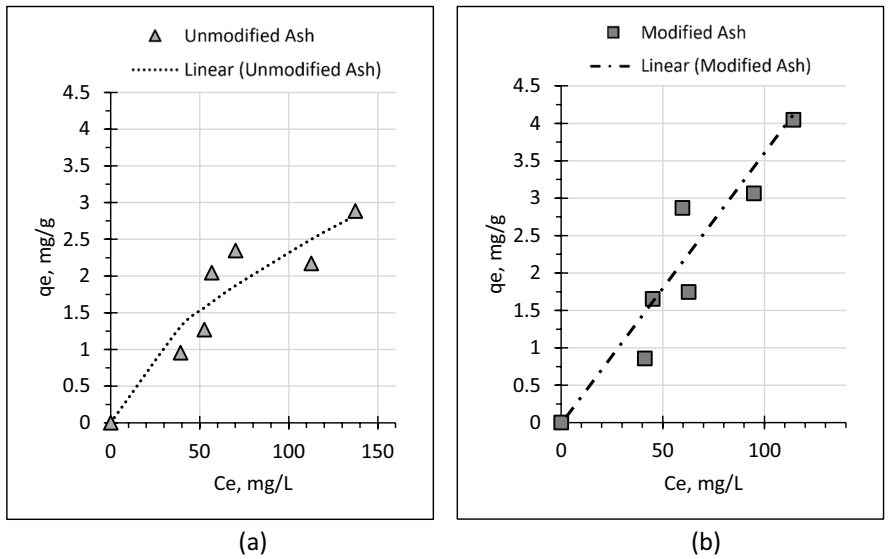

Fig. 10 Experimental data of vancomycin onto unmodified (**a**) and modified (**b**) SD fitted to the Freundlich isotherm model (dotted line)

(Table 3). Overall, the adsorption parameters fitted better the Langmuir model, yielding higher R^2 and χ^2 values for both unmodified and modified SD (Table 3). This result indicates a monolayer adsorption of vancomycin onto the SD characterised by a homogeneous adsorption surface with identical adsorption sites (Sayen et al., 2018).

3.5 Suggestions for Further Investigation

The proposed methodology is a simple and sustainable approach based on the utilisation of various biomass-derived materials or agricultural wastes. These materials can be potentially translated into working prototypes for tertiary wastewater treatment. Figure 11 shows how the prototype could be included in a sustainable process. Initially, the unwanted SD waste could be recycled into an efficient low-energy adsorbent through the chemical treatment proposed in the current paper. Then, the modified SD would be used to adsorb and reduce the concentration of antibiotics in the pre-treated wastewater, with the following liquid being safely discharged into the environment. Once the SD adsorbent reaches a saturation threshold, most of the moisture (considered leachate) would be collected after compacting the used sawdust through a press, similar to an activated sludge press. The small amount of leachate would subsequently be recirculated into the WWTP to ensure the removed antibiotics are fully captured and the dry SD pressed residue could be transferred to a pyrolysis reactor (Ramakrishnan et al., 2014). This process would convert the adsorbed antibiotic to a small amount of ash to be discharged into a sanitary landfill (Sayen et al., 2018). Dry sawdust could also be revalorized by co-composting the sawdust (Bello & Jimoh, 2021) or used as a growing media for mushrooms (Rambey et al., 2020).

Combining pyrolysis with an energy recovery system would maximise energy efficiency by redirecting some of this energy to a nearby facility requiring electricity. Alternatively, the power generated could be used to power the WWTP, thus reducing running costs whilst minimising the CO_2 footprint of the operation. As discussed earlier, the heating value potential of sawdust is close to the one of activated carbon. Besides, the nominal quantities of bound sulphur and nitrogen, high volatile matter and near-zero ash quantities make the sawdust a great candidate for a wider circular economy, by providing energy at the end of its life.

4 Conclusions

This paper has shown that sawdust (SD) can be an effective alternative to activated carbon (AC) for removing antibiotics from water. The proposed chemical treatment of SD fibres with solutions of *o*-phosphoric acid, sulfuric acid and potassium hydroxide was found to be efficient, by providing a five-fold

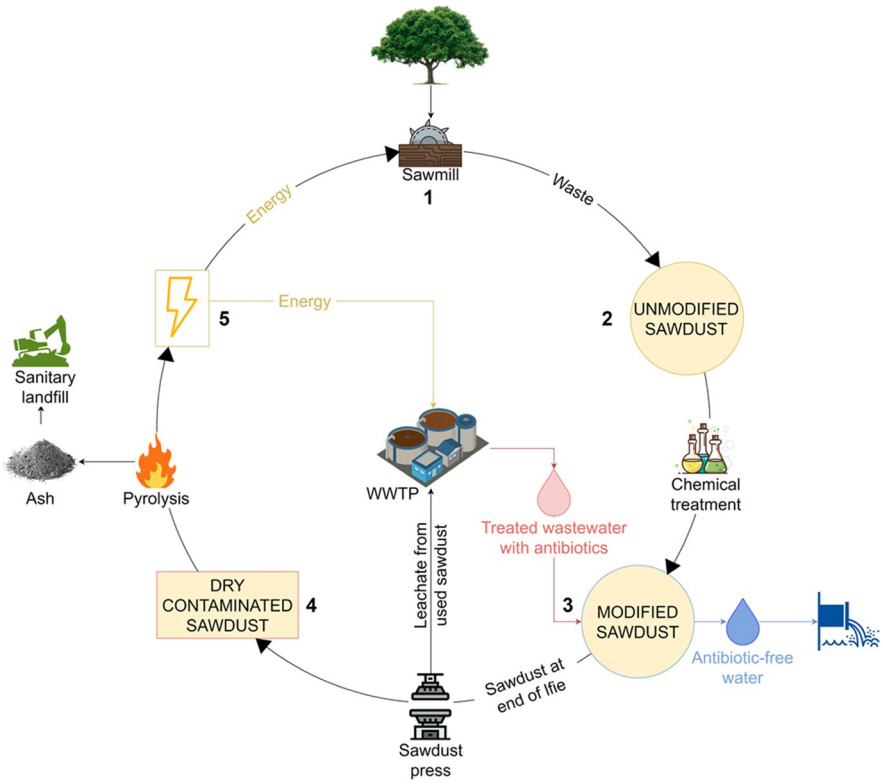

Fig. 11 Sawdust as part of a five-step circular sustainable process

increase in the adsorption of vancomycin compared to the unmodified materials. The removal levels of vancomycin reached 63.0% ($\sigma = 2.3\%$) for the modified mixed material and 56.6% ($\sigma = 5.1\%$) for the modified ash sawdust, which were lower than the levels obtained for AC (98.2%, $\sigma = 2.9\%$). However, given the recent austerity, WWTPs are likely to seek an adsorbent that is cheaper than AC. For instance, the adsorption of antibiotics on AC has been estimated to cost 0.20–0.27 € per cubic meter of treated water (Mulder et al., 2015). The proposed treatment of SD, using inexpensive common chemical reagents, could be considered as a viable route to obtain effective adsorbents, without a significant carbon footprint. Thus, the availability of SD, even with removal capacities lower than AC, makes it an affordable competitive adsorbent for both developing and developed countries. In addition, the heating values of SD were found to be in the region from 15.5 to 18.5 MJ/kg, indicating its potential use for energy production through thermo-chemical conversion methods. The method suggested above and the reuse of the spent material as a fuel will allow WWTPs to practice energy recovery, and this will help in achieving the UN Sustainability Development Goals of good health, clean water and sanitation and sustainable cities and communities. The study showed that increasing the SD quantity from 1 to 2 g can significantly improve vancomycin removal (from 30.7 to 56.4%) and decrease the adsorption capacity (from 22.1 to 34.7%). The adsorption parameters indicated a possible monolayer adsorption of vancomycin onto the SD characterised by a homogeneous adsorption surface with identical adsorption sites.

It should be noted that this initial scoping study was designed to test the viability of the process as a means of removing antibiotics from wastewater. As it has been denoted in the published literature, the non-existence of discharge limits of pharmaceuticals is at the core of the proliferation of this problem within the natural aquatic environment. This work is a stepping stone towards the development of a low-cost viable means of dealing with this issue in developed and developing countries alike. Further research using a lower range of the antibiotic concentration, and other types of antibiotics, will be necessary to confirm the initial findings, as well as to carry out kinetics and desorption studies.

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Author Contribution Benjamin Delmond: collected the data, performed the analysis and wrote the paper.

Svetlana Tretsiakova-McNally: conceived and designed the analysis, contributed to writing the paper, contributed data and reviewed the paper.

Brian Solan: conceived and designed the analysis, contributed to writing the paper and reviewed the paper.

Rodney McDermott: reviewed the paper.

Alexandre Audoin: contributed data on the analysis of sawdust.

Data Availability The datasets generated and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of Interest The authors declare no competing interests.

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