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Review

Potential Benefits and Disbenefits of the Application of Water Treatment Residuals from Drinking Water Treatment Processes to Land in Scotland: Development of a Decision Support Tool

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Abstract: Water Treatment Residuals (WTRs) are a by-product of the addition of chemical coagulants to water during the water treatment process and are a mixture of water and organic and inorganic matter that coagulates during the treatment process. WTRs often contain metals such as iron, aluminium, and manganese that have been oxidised as part of the process or are constituents of the coagulation chemicals used. The metals within WTRs are of interest with regard to applying these sludges to agricultural land. WTRs can also contain beneficial organic matter and nutrients (primarily nitrogen). The nature of the benefits delivered is largely dependent on the quality of the raw water and these beneficial components are generally found in much smaller quantities in WTRs than are found in sewage sludge produced from wastewater. However, WTRs can still be used to enhance the physical properties of soils. As urban populations increase in size, it is anticipated that the tonnage of WTRs will increase significantly in the future. At present, the majority of WTRs are disposed of in landfills; however, landfill charges are increasing significantly, making disposal of an increasing tonnage of WTRs financially unviable. In terms of a circular economy, the procedure of reusing WTRs for alternative applications satisfies the Scottish Government's goals in terms of waste prevention and reducing the amount of material being sent to landfill as set out in the Proposals for Legislation in 2019. Given the potential benefits in terms of cost savings and compliance with government legislation, and the complexities of understanding where and when WTRs can be used in land applications, we developed a Decision Support Tool (DST) that uses data obtained from an extensive review of approaches in other countries to assist in decision making. We also conducted a pre-application analysis and provided guidance on when and where WTRs can be used in land applications and when they are not suitable, presented in a simplified format that requires few inputs from the user in order to simplify the process and removes the requirement for a specialist operator during pre-application analyses.

Keywords: circular economy; decision making; decision support tool; waste; water treatment residuals

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1. Introduction

WTRs are generated as part of the drinking water treatment process. In Europe, several million tonnes of WTRs are produced every year, and there are considerable concerns about the costs associated with disposal of these materials [1]. Originally, WTRs were disposed of in the same water source where drinking water was taken from, until the introduction of stricter environmental regulations in Europe in the 1940s [1]. The management of WTRs as opposed to Waste Water Treatment Residuals (WWTRs), often referred to as 'sewage sludge' or 'biosolids', is a challenge because the lower nutritional and calorific value of WTRs make biological digestion or incineration more impractical [2], whilst the high metal content limits the reuse of WTRs in some applications and the large quantities of bound water make dewatering and transport difficult [1]. Despite the challenges associated with

WTR management, there has recently been much more interest in reuse. This is likely due to the increased costs of disposal and the move towards a circular economy model of waste management. Previous efforts to reuse waste materials to improve soil properties include the addition of crushed igneous rocks and crushed concrete to soils on managed brownfield sites and infrastructure projects which have the additional advantage of engineering soils for carbon capture [3,4].

The overall aim of this project is to identify information on best practice, outline the potential benefits and disbenefits of applying WTRs to land, and finally present this information in a user-friendly Decision Support Tool (DST).

2. Characteristics and Variability of WTRs

The standard processes for drinking (potable) water treatment typically include coagulation/flocculation, sedimentation, filtration, disinfection, and pH correction. Coagulation is used to remove sediment from the water before supplying it to customers. To achieve coagulation, aluminium (Al) salts are added to water, which results in formation of alum sludges [5–8] which are commonly referred to as WTRs. The Al salts typically used are aluminium sulphate and sodium aluminate as well as pre-hydrolysed polyaluminium chlorides such as Flokor 105B and PAX XL10 [9]. Using Al salts to achieve coagulation for water treatment is currently the main practice in Scotland (95% Al based); however, ferric salts are also used at a very small scale (5% are ferric based). After coagulation, WTRs can be dried to reduce the by-product's moisture and hence facilitate transportation. This method is also the most widely used treatment technique internationally [8].

It is estimated that water treatment results in 1–3% of WTRs by volume of the pre-treated water [10]. Internationally, a large portion of WTR disposal is via landfill, which results in disposal costs being added to overall water treatment costs [11]. When WTRs are deposited into landfills, the transport distance associated with disposal is of growing concern given the recently defined carbon emission targets internationally, nationally (i.e., Scotland's 2045 net-zero emission target [12]), and at a business level [13].

Recently in Scotland, the majority of WTRs have been used for land restoration (Table 1). Between April 2017 and March 2018, 92% was used for land restoration, similarly, between April 2018 and March 2019, 93% was used, and between April 2019 and November 2020, 95% of WTRs were used for land reclamation. With increasing numbers of completed reclamation projects (e.g., opencast coal mines), soon there will be a significant amount of WTRs in Scotland with no clear outlet for reuse. Assessing the suitability of WTRs for future land application in agriculture and forestry is important to explore future opportunities to reuse this potential resource. This report will focus on alum WTR, as treatment of water with Al salt is the technique generally used in Scotland.

Table 1. Summary of WTR production and reuse/disposal in Scotland (values in tonnes).

Area	April 2017–March 2018			April 2018–March 2019			April 2019–November 2020		
	Land Rest.	Landfill	Agri.	Land Rest.	Landfill	Agri.	Land Rest.	Landfill	Agri.
North	0	721	1895	0	714	1291	0	527	694
East	5671	0	0	5037	0	0	4118	0	0
West	12,228	0	0	11,089	0	0	10,145	0	0
South	12,199	0	0	11,042	0	0	8191	0	0
Total	30,098	721	1895	27,168	714	1291	22,454	527	694

The characteristics of WTRs depend on factors such as the initial characteristics of water and the method of treatment [11]. Water acquired from surface or underground water sources contains various types of suspended solids from clay to sand-sized particles. During water treatment, suspended solids are collected, which comprise a portion of WTR. Using Al salts for treatment of water is an effective and low-cost technique. Alternatively, ferric salts have been reported as a flocculant for water treatment [14]. However, alum

WTR is reported to be less toxic than ferric chloride sludge [8]. Porous organic polymers have been also reported as an effective coagulant for water purification [15].

WTRs are primarily composed of $Al(OH)_3$ or $Fe(OH)_3$ (depending on the coagulant used), organic matter, clay particles, nutrients, contaminant metals, and other impurities removed from treated water [16]. WTRs collected from the treatment plants contain a high water content (2–4% solids) and accordingly, dewatering is a common practice before transportation of WTRs [8]. The summary of WTRs physical and chemical properties are presented in Table 2.

Table 2. Summary of WTRs physical, chemical, and mechanical properties.

Property	Description
Particle size distribution	Uniform distribution of particle sizes [17]. Sand content 60.4–69.0%, silt content 17–23%, and clay content 14–16.6% [8].
Specific gravity	Normally lower specific gravity than topsoil which is attributed to the higher organic content in WTRs [17]. To produce fully dried WTR pallets, it is suggested room temperature drying for 3 days and then 24 h oven drying at a temperature of 110 °C [18]. The specific gravity of solids ranges between 1.8 and 2.2 [11].
Compaction	WTRs cannot be fully dried in situ as this results in destruction of soil structure and calcification of particles [17]. The bulk density and dry density values of partially dried but otherwise untreated (~10–40% *w*/*w* dry matter) range between 1.0 and 1.2 tonne m^{-3} and 0.12 and 0.36 tonne m^{-3}, respectively [11].
Shear strength	Values vary depending on solid content but in general increase with increasing solids content [17]. Effective cohesion value of zero and effective angle of shearing resistance ranges between 28 and 44° [11].
Atterberg limits	The Liquid Limit ranges between 100 and 550 (%) and the Plastic Limit ranges between 80 and 250 (%) [11]. WTR was partially dried but otherwise untreated (~10–40% *w*/*w* dry matter).
Nutrients	Contains four important nutrients: phosphorus, nitrogen, potassium, and sulphur [17]. For the range of values, see Table 3.
pH	Values vary but a significant difference in WTRs pH to the applied environment can have detrimental effects on the surrounding environment [17]. For the range of values, see Table 4.
Trace metals	Lower in WTR than WWTRs. Varies in WTRs depending on pre-treated water properties and treatment method but typically high in Al or Fe [17], depending on the coagulant used. For the range of values, see Table 4.

WTRs have a high porosity [7], resulting in a high water holding capacity. In addition, they have reactive surfaces due to presence of Al or Fe hydroxides, providing a high sorption capacity [19,20]. WTRs (air dried and crushed to less than 2.0 mm) typically have a high cation exchange capacity of around 13.6 to 56.5 cmol + kg^{-1} compared to around 3.5 to 35.6 cmol + kg^{-1} for typical soils [21]. It is suggested that WTRs can retain from 1740 to 37,000 mg phosphorus (P) kg^{-1} (it is not clear whether this is on a dry or wet weight basis), and for WTRs, the sorbed P is not readily desorbed [19]. An average nitrogen content of around 5 g kg^{-1} has been reported, with phosphorus, potassium, and magnesium contents of around 2.5 g kg^{-1} each [22]. Furthermore, 12.0 with a maximum recorded nutrient concentration of greater than 30 g kg^{-1} of the essential plant nutrient manganese was reported for WTR [22]. Table 3 presents the typical total major nutrient content of WTRs from the UK in general and Scotland in particular.

Table 3. Typical total nutrient content of WTRs from the UK and Scotland.

	Dry Matter Content	N	P_2O_5	K_2O	SO_3	MgO
	(%)	(kg Fresh Tonne^{-1})	(kg Fresh Tonne^{-1})	(kg Fresh Tonne^{-1})	(kg Fresh Tonne^{-1})	(kg Fresh Tonne^{-1})
UK mean values [23]	25	2.4	3.4	0.4	5.5	0.8
Scotland * [24]	18–25	1.7–4.4	0.3–1	0.04–0.1	2.8–4.4	0.16–0.2

* Treatment works include Amlaird, Auchneel, Bradan, Camps, Carron Valley, Glengap, Killiecrankie, Lintrathen, Loch Turret, Lochinver, Pateshill and Penwhim, Turriff, and Whitehillocks.

Table 4. Range of total element concentrations, organic matter, and pH for WTRs acquired from 18 peer-reviewed studies [11] and Scottish case studies [24].

Data Source	Al	Fe	P	Ca	Mn	Pb	Zn	Ni	Cu	Organic Matter	pH
Unit	g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	%	
Range of values from 18 peer-reviewed studies from around the world [11]	6.7–180	1.1–277	0.2–10	0.18–32	0.4–31.6	2.5–69	0.12–246	10.9–60	35–624	5.8–24.5	5.12–8
Mean values obtained from tests on three samples of WTRs from SW * Glenfarg treatment works in 2020 (this project).	120	4.3	1.7	3	0.3	4.1	40.5	11.1	34.6	52.1	6.5
Mean values obtained from SW tests on 19 samples of WTRs from SW Glenfarg treatment works from 2014 to 2018.	128	N/A	0.9	N/A	N/A	<10	60	11.4	33.9	N/A	6.3
Mean values obtained from SW tests on five samples of WTRs from SW Whitehillocks treatment works in 2020.	N/A	N/A	0.7	N/A	N/A	≤13.1	31.4	≤6.5	14.8	N/A	5.5
Values obtained from SW tests on four samples of WTRs from each of ten SW treatment works [a] in 2018 and 2019 (mean value from all samples tested)	75.6–222 (138)	8.4–15.1 (11.3)	0.2–18 [b] (1.3)	N/A	N/A	10–108 [c] (21)	15–722 (59)	6–54.4 (15)	6–153 (26)	N/A	4.5–7.1 (5.9)
Values obtained from independent tests on eight samples of WTRs from seven SW treatment works [d] from 2017 to 2019 (mean value from all samples tested)	N/A	N/A	0.3–1.3 (0.6)	1–3.7 (2.1)	N/A	11–47 (30)	30–71 (55)	4.4–15.1 (11)	11–70 (22)	58–67 (62)	4.4–6.7 (6)

* Scottish Water. [a] Treatment works include Auchneel, Bradan, Glengap, Killiecrankie, Lintrathen, Loch Turret, Lochinvar, Penwhim, Turriff, and Whitehillocks. [b] This value was highly unusual amongst the 40 batches tested and it may not be a true representation of this material. Of the remaining 39 samples tested, the next highest value for P was 2.5 g kg^{-1} and the mean value of the 39 other samples other than that with the abnormally high P concentration was 0.8 g kg^{-1}. [c] This value was highly unusual amongst the 40 batches tested and it may not be a true representation of this material. Of the remaining 39 samples tested, the next highest value for Pb was 20.6 g kg^{-1} and the mean value of the 39 samples other than that with the abnormally high Pb concentration was 42.2 g kg^{-1}. [d] Treatment works include Amlaird, Auchneel, Bradan, Camps, Carron Valley, Pateshill, and Penwhim.

A technical report [24] reported a summary of five case studies in Scotland, in addition to 18 peer-reviewed global studies on WTR total element concentrations, organic matter, and pH, originally presented by a research review paper [11] (shown in Table 4). The significant range of data indicates a wide variability in WTR characteristics due to varying pre-treated water characteristics and methods of treatment. Table 4 further proves that accurate identification of WTR characteristics from specific WTRs should be performed through a direct analysis, as using existing published data as an indicator could be associated with significant error.

3. Benefits and Disbenefits from Land Applications

3.1. Agricultural Land

Using alum-based WTRs as soil amendments in agriculture has been reported to enhance nutrient recycling and improve the soil's physical and chemical properties [8], as summarised in Tables 2–4. Table 5 summarises WTR application's impact on agricultural soils, in both sole application of WTRs and co-application with WWTRs.

3.1.1. Physical Properties

According to some studies [25,26], successful application of WTRs on agricultural land requires an evaluation of their effects on the soil's physical properties (such as cohesion, aggregation, strength, and texture, which affect the hydraulic properties of the soil), plant growth, and groundwater quality. Accordingly, consideration of all the above-mentioned factors before using WTRs for agricultural land applications is required. For most agricultural soils, particularly those under arable cropping, increasing the soil organic matter (SOM) content has a range of benefits such as increased crop yields and lowered additional nutrient input requirements. Increasing the organic matter content in soil also improves the soil structure, which results in better water infiltration and hence an improved drainage. Land application of alum-based WTRs could improve soil air and water holding capacity [27], which would result in a better root system performance which is required in agricultural soils.

WTRs can contain fine-grained particles such as clay [16], which if added to agricultural fields with fine particles could potentially reduce the soil porosity and lead to flooding risks, but could also be beneficial, increasing nutrients and water retention if added to a sandy soil. Accordingly, to avoid WTRs negatively impacting agricultural soils, the WTR and agricultural soil's particle size distribution should be analysed prior to the application to ensure that the WTRs are being spread in a suitable location.

3.1.2. Chemical Properties

The nutrients provided by WTRs are presented in Table 3. According to Table 4, WTRs pH ranges between 4.4 and 8. Maintaining the soil pH at optimal levels, generally accomplished in farming via the direct application of lime, has many well-documented benefits such as increasing microbial activity and maximising the availability of macronutrients N, P, and K [28]. Higher pH soils can be prone to deficiency in trace nutrient availability to crops, particularly if the soil pH is >7.5. In soils in Scotland, trace element deficiency, particularly manganese for cereals and cobalt for livestock, can be induced by pH values of >6.3, with the severity depending on the soil texture. Therefore, it is important to determine both WTR and soil pH before spreading a WTR on land. It has been demonstrated that the growth of ryegrass roots was reduced as the soluble Al concentration increased in four Scottish brown earth soils at pH 3.5–4.9 [29]. Accordingly, the impact of applying Al-based WTRs to agricultural land on the 'below ground' biomass should be considered. In SRUC Technical Note TN714 liming materials and recommendations [30], it is stated that at soil pH values below 5.6 in mineral soils in Scotland (independent of the application of WTRs), soluble aluminium inhibits root growth and reduces yields. Accordingly, WTR application could potentially exacerbate the Al toxicity under such conditions. Soil chemical properties play a key role in determining WTR application rates. The variability in the published WTR data shown in Table 4 indicates the importance of identifying the characteristics of specific WTRs, as referring to existing data may be associated with significant error. The aluminium content in WTRs and their provision of nutrients such as potassium and sulphur should be considered with respect to the receiving soil's chemical properties.

Several studies have reported benefits of adding WTR to other types of fertilisers for plant growing purposes such as mixing vermicompost [31] or poultry litter [32] with WTRs. An improvement in saline sodic soil physical properties and wheat yield as a result of combining vermicompost with WTR has also been demonstrated [31]. The authors of the study also reported that the mixing with vermicompost improves WTR efficiency in ameliorating the soil physical properties, especially in salt-affected soils (although not a common occurrence in Scotland), to improve the yield of wheat [31]. It has been demonstrated that applying WTRs at the rate of 50,000 kg ha^{-1} (50 t ha^{-1}) to box plots treated with 16,700 kg ha^{-1} (16.7 t ha^{-1}) poultry litter reduced runoff P by 14.0 to 84.9% [32].

According to one study [17], adding WTR to poultry litter results in a reduction in water-soluble P up to 87% depending on the dose and incubation period. Other studies [11] have reported benefits from using WTRs for agricultural production in areas that have contaminated soils requiring management and remediation.

Table 5. Impact and observations of using WTRs on agricultural lands in sole and co-application.

Impact of and Observation for Using WTRs on Agricultural Land	Reference
Long-term (7.5 years) study of the effect of WTRs on soil at a site in Michigan, USA, which had received >10 years application of poultry manure prior to the application of WTR. As a result of WTR application, Al-based WTRs immobilized P and remained stable 7.5 years following initial land application.	[33]
Mn release from WTRs from a treatment plant using KMnO4 was assessed as part of the treatment process and found increased extractable Mn concentrations in soils amended with Al WTRs enriched with Mn. The authors of the study suggested a WTR pre-screening procedure (testing elements such as Mn) to determine if land application of WTRs could release elements such as Mn that may cause plant growth problems.	[34]
The effects of different combinations of WTRs and biosolids (co-application) on two plant species in a laboratory were studied and showed that WTRs reduced plant-available P to both species. No visual P deficiencies were observed.	[35]
The long-term effects of WTRs–biosolid co-application on P cycling in semiarid rangelands at a site in Colorado, USA, were studied. Pathway analysis showed that even after 13 years following initial co-application, WTRs still acted as the major stable P sink. Additionally, differences in semiarid rangeland plant and soil microbial communities were noted 12 years after WTRs–biosolids co-application compared to soils affected by biosolid treatments alone. The effects were indicative of a successional shift from a community of low nutrient availability and tight nutrient cycling to one with more readily available resources and a decreased need for symbiotic arbuscular mycorrhizal fungi associations.	[36]
The long-term effects of a single co-application and the short-term effects of a repeated co-application of biosolids (10 t ha^{-1}) and Al WTRs (5, 10, 21 t ha^{-1}) on rangeland soils and plants were reported. No change in soil pH, EC, $NO^{3-}N$, $NH^{4-}N$, total C, or total N by WTR application was detected. However, extractable soil Mo decreased with the increasing Al WTRs rate, most likely due to WTR adsorption. The Mo content in the two dominant plant species decreased with repeated WTR application compared with a single WTR application. However, Mo deficiency was not observed.	[37]

3.2. Forestry

Forestry and land restoration are closely linked as WTRs, and other types of waste materials are used in the land remediation of former open cast lands to develop soil properties to a state fit for woodland creation. Table 6 summarises previous research into the WTR application impact on forests. Remediated land is frequently planted as woodland. Ecosystems provide living spaces for plants or animals; they also maintain a diversity of different breeds of plants and animals [38]. Various definitions of supporting ecosystem services exist; two of the most cited definitions are 'supporting services underpin almost all other services' and 'supporting services are those that are necessary to produce all other ecosystem services'. They differ from provisioning, regulating, and cultural services in that their impacts on people are often indirect or occur over a very long time, whereas changes in the other categories have relatively direct and short-term impacts on people [39]. Examples of supporting services required in forestry include soil formation, nutrient cycling, and water cycling.

Table 6. Impact of and recommendations for using WTRs for forest creation.

Impact of and Recommendation for Using WTRs for Forest Creation	Reference
Limited application of Al-based WTRs to forest soil in USA at the application rate of 1170 m^3 ha^{-1} showed the phosphate cycle and forest growth pattern were not affected.	[40]
Application of Al-based and polymer WTRs to forest soils at the application rate of 0.8 to 2.5 g kg^{-1} showed no effect on growth or nutrient content after at least 1 year.	[32,41,42]
Application of solid Al-based WTRs to forestry lands at the application rate of up to 2.5% by dry weight of the topsoil in USA showed no adverse effects. It was concluded that the WTR can be applied at high rates with no detrimental impact on the soil, groundwater, or tree growth up to 30 months after application.	[43]

In forestry applications, consideration of the existing soil properties (pH, soil nutrients, organic matter, etc.) is required. These properties play an important role in the requirement of WTRs and the application rate. In land restoration, due to the poor quality of soil, application of WTRs would provide benefits to the soil properties discussed above. These applications are discussed further in the next section.

3.3. Land Restoration

This section focuses on land restoration, and Table 7 summarises WTR application's impact on land restoration. Restoration sites predominately lack topsoil and suffer compaction, and provision of sufficient nutrients and organic matter is crucial. Accordingly, in restoration sites, inputs of organic matter which could be provided by WTRs will help to create a good soil structure, reducing the erosion of soil via wind and water.

Table 7. Impact of and recommendations for using WTRs for land restoration.

Impact of and Recommendation for Using WTRs for Forest Creation	Reference
WTR application's potential for land reclamation was evaluated in the USA. An analysis of WTR samples showed all of the samples were suitable as soil substitutes based on plant nutrients, with the exception of P. For crop growth of tomatoes, the vegetative yield and tissue P were poor. This was linked to phytotoxic nitrite-nitrogen ($NO^{2-}N$) (>10 mg kg^{-1}) generated during the bioassay or because of WTR P deficiency.	[21]
WTR was combined with vermicompost in Egypt to improve WTRs efficiency in ameliorating the soil's physical properties. It was concluded that WTRs can be used as an ameliorating material for the reclamation of salt-affected soils.	[31]

4. Field and Lab Trial Application Rates for Spreading in Agriculture and Forestry

In an experimental plot in Australia, Al-based WTRs were applied (in a dry state) to ryegrass plots at the rates of 0, 200, 400, 800, and 1600 t ha^{-1} with lime at the rates of 0, 1.8, 3.6, 7.2, and 14.4 t ha^{-1} [44]. The mixing of WTR and lime with soil was accomplished in a laboratory incubation experiment. At all application rates, the authors of the study realised positive impacts on the soil's physical properties, including a reduced dry bulk density and an increased porosity and infiltration rate. However, there were no observable visible impacts on plant growth or yield, even at the highest rate of application.

In the USA, researchers studied the application of Al-based WTRs to P-deficient bahiagrass (Paspalum notatum) plots at rates equivalent to 0, 10, and 25 g kg^{-1} (oven dry basis) with four P sources at two application rates [45]. These application rates were equivalent to approximately 0, 40, and 100 t ha^{-1}, assuming a bulk density of 1.33 g cm^{-3} and an application depth of 0.3 m. The authors of the study realised an increase in the P storage capacity with the application rate of WTRs, with the degree of the increase varying with sources of P acquired from conventional fertilisers such as manure and biosolids. The study reported an increase in soil soluble P concentrations as the soil P storage capacity was reduced, and a change point was identified at 0 mg kg^{-1} soil P storage capacity in the glasshouse and field studies [45]. The authors of the study identified a change point in the bahiagrass yield at a tissue P concentration of 2 g kg^{-1}, corresponding to a zero soil P storage capacity (considered as an agronomic threshold above which yield and P concentrations of plants declined and below which there is little or no yield response to increased plant P concentrations). Researchers also investigated the application of Al-based WTRs to bahiagrass plots at the rate of 0, 35, and 70 t ha^{-1} [46]. The authors of the study realised a reduction in grass yield in the first year, when the WTRs were applied at the highest rate, which is believed to be due to disturbance of the bahiagrass stand, by disking for the amendment incorporation, and not due to the effects of the Al WTR. Other researchers also evaluated the use of two Al-based WTRs with application rates of up to 45 t ha^{-1} combined with 6.7 t ha^{-1} of poultry litter to grass pasture [47]. The authors of the study realised a reduction in the mean dissolved P as a result of the WTR application compared to dissolved P pre-WTR application, resulting from P fixation, and reported no adverse effect on grass yields.

In a six-year study [33], two trial fields that received 0 or 114 t ha^{-1} dry Al-based WTR were investigated and realised a reduction in dissolved P and bioavailable P by >50% as compared to the control plots. The authors of the study reported no negative impact on crop yields.

In the USA, two studies [36,37] investigated the application of three different Al WTRs (at 5, 10, and 21 t ha^{-1}) co-applied with a single biosolid application (rate 10 t ha^{-1}) to a loamy soil pasture (shortgrass prairie with animals excluded). One study [36] realised the significant capacity of WTRs as a stable sink for P when mixed with biosolids. Others [48,49] reported the application of an Al WTR to a pasture at a cumulative rate of 78 metric t ha^{-1} over two years, and researchers evaluated the effects of dietary Al from the Al WTR on cattle over two years [48]. The results showed that Al WTR had no adverse effects on growth, development, or blood plasma mineral concentration of the cattle, likely due to low Al availability from the Al WTR. WTR application did not adversely affect forage mineral concentrations. The researchers concluded that Al WTRs are safe and could be applied to pastures at low to moderately high rates (=78 metric tons ha^{-1}) to help reduce P runoff and leaching as investigated in the study [48].

Compared with studies of WTR spreading on agricultural land, comparatively few studies of WTR spreading at different rates for forestry land can be found. In one study [43] in the USA, researchers applied Al-based WTRs to forestry plots (with an original soil pH = 4.8) at the rate of 2.5% by dry weight in the topsoil (or 1100 t ha^{-1}, assuming a bulk density of 1.33 g cm^{-3} and an application depth of 0.3 m). They concluded that there was no detrimental impact on the soil, groundwater, or tree growth up to 30 months after application.

Besides the studies that are discussed in this section, no further information was found on the impacts of long-term use of WTRs on land. It appears that routine monitoring of soil properties and crop/tree growth is either not widely carried out at WTR spreading sites or data from such monitoring have not been widely reported, leading to limited knowledge of the long-term benefits/impacts of WTR use on both agricultural and forestry land.

5. Potential P Immobilization Issues Caused by WTRs

Phosphorus exists in the soil within organic material, occluded in hydroxides, and as phosphate salts formed from rock weathering. Available quantities are usually low, and farmers may apply phosphorus fertilisers. Overapplication of P to soil can result in accumulation, with the associated risks of release under specific conditions and resultant transport to water bodies leading to eutrophication. A major process involved in this release is soil transport through erosion, although surface water runoff and leaching also play a part.

In Scotland, leaching of P is less likely than in other parts of the world, where the impacts of WTR spreading have been researched, as significant P fixation by Al occurs at pH values below 6. According to SRUC Technical Guidance Note TN714 [30], 64% of arable and grassland soils in Scotland have a soil pH of 5.5–6.25, with 20.3% above and 16.1% below this range. Erosion and runoff are therefore more likely mechanisms for P transport to streams and water bodies associated with WTR spreading to land in Scotland. Lateral flow transport of dissolved P through the soil is also a risk if there is heavy rainfall soon after WTR application.

There is concern that WTRs may immobilise P that is already within soils; if this does occur (through processes including fixation on hydro(oxides) that are present in high concentration in WTRs), then it would likely exacerbate the existing Al-based immobilisation at low pH values, resulting in further reduced available P.

There are no references in the literature containing all four key phrases 'phosphorus immobilisation', 'soil', 'sludge', and 'Scotland'. However, removing 'Scotland' from this search reveals 92 publications on Web of Science. Of these 92, many relate to sewage sludge and only seven were considered relevant with a focus on the topic of concern (WTRs) under comparable climatic conditions. There were also several publications that focused on the potential use of sludge-derived biochar and other products for immobilisation of contaminants, including heavy metals and elements such as Cu, Zn, P, and S, if present at levels high enough to become toxic (e.g., in soils containing mining residue). While not directly relevant to the topic of concern, they do highlight and reinforce the message that WTRs have the potential to immobilise soil nutrients important for plant growth.

Furthermore, most of the work on potential P immobilisation involves sewage sludge, rather than WTRs, and was focused on ways to immobilise P in the sewage sludge to control the release of available P. There was no information found on how the different characteristics of these two types of sludge would influence potential P immobilisation.

Studies have highlighted the potential of WTRs for the immobilisation of 'excess' nutrients in soil [11]. They also pointed out that application onto 'clean' land is well established in the UK and USA, and that a reduction in plant-available P has been observed following application (as well as reduced plant P concentrations). It is possible that this has occurred through immobilisation of P in soil depending on the organic matter and Al and Fe contents of the wastewater treatment residue, but it may also be caused by high nitrogen and comparatively low phosphate concentrations in wastewater treatment residues that are used as fertilisers. The wastewater treatment residue in these circumstances may provide enough nitrogen to support crop growth, but not enough P, which could lead to crops using up available P in soil and, over time, a decline in the soil available P concentration and ultimately a decline in P in plant tissues [50]. The Turner et al. review [11] is a comprehensive and extensive review relevant to this topic of P immobilisation and provides specific values on composition, application rates, and observed impacts (Table 2, Table 5 and Table 9 and Section 5.4 in that specific review paper [11]). It also describes the impacts of co-application of P fertilisers as an approach to reducing the P immobilisation effects of WTR application.

Zhao et al. [22] reviewed evidence of the impact of Al-based WTRs and determined that at pH values above 5, Al toxicity is unlikely. However, these higher pH levels are where Al-based immobilisation of P is less likely, highlighting that pH testing of soil prior to application may be necessary to allow for calculation of appropriate matching of application rates and residue chemical composition. WTR application can induce P deficiency in crops [19]. Lombi et al. [51] demonstrated reductions in plant growth (*Lactuca sativa*) caused by application-induced P deficiency to both acidic and neutral soils following WTR application.

Vasilyev et al. [52] found that application of WTRs to loamy brown earth soil decreased the nutritional content (K and starch) of potatoes grown in that soil due to immobilisation of potassium, while at the same time the soil nutrient (organic matter, total P, and total K) content increased. These effects appear to have been due to variations caused in soil chemical stoichiometry by sludge application. Tay et al. [53] found no impact on plant growth from P availability in an experimental application coupled with P fertiliser, while at the same time demonstrating reduced Cd and As concentrations.

Adding WTR sto green waste compost feedstock resulted in decreased CO_2 release, with this effect being reversed for biosolid compost [54]. These effects were attributed to reductions in available P and heavy metals, with the initial values of P in green waste compost being appropriate for microbial decomposition of organic matter but at toxically high heavy metal levels in biosolid compost. The P immobilisation effect of WTRs therefore needs to be considered in the context of the existing concentrations of P in the material to which it is being applied.

6. Technical Requirements and Management

6.1. Application Rates and Technical Requirements

Spreading WTRs on agricultural land is commonly achieved using conventional manure spreaders. However, for land restoration, WTRs are usually dug into the surface layer of de-compacted soil using an excavator [24]. The reported application rates from experimental studies, discussed in this review, are largely reported from small-scale experimental studies rather than large-scale agricultural and land restoration experiments. However, for agricultural purposes, WTR application rates of 50–150 t ha^{-1} have predominately been recommended in the published literature [24]. In England and Wales, application rates for agriculture are typically in the range of 20–60 t ha^{-1} [24]. The maximum application rate permitted is 250 t ha^{-1}, with a further limit on liquid applications of 50 m^3 ha^{-1} at any one time, and additionally no more than 250 kg N ha^{-1} may be applied to land according to waste management regulations in Scotland (Waste Management Licensing (Scotland) Regulations 2011).

6.1.1. Agriculture

The application rate is determined by the WTR characteristics, the nutrient requirement of the planned crop, and the frequency of application. In agriculture, application is anticipated to be on an annual basis. One of N, P, or Potentially Toxic Elements (PTEs, typically identified as Zn, Cu, Ni, Cd, Pb, Hg, Cr, Mo, Se, As, and F) usually limits the application rate; the maximum application rate will be determined by the requirement to avoid excessive supply of N or accumulation of P or PTEs in soil to excessive levels. Notably, the total N and P content may not necessarily provide sufficient N or P for crop growth due to nutrient losses [55]. In addition, the application of WTRs cannot satisfy the K need of the crops.

6.1.2. Land Restoration

In restoration, there is no N limit set in legislation in Scotland. However, a theoretical application rate of 588 t ha^{-1} of WTRs provides 1000 kg ha^{-1} N. The maximum addition rate of 1000 kg ha^{-1} N is listed in the 'Key principles for the remediation of former opencast coal sites for woodland establishment, v7.2′ [56]. Nitrogen can be added at rates up to this maximum, under the condition that PTE concentrations in soil will not exceed the maximum limits set out in the soil table in the 1989 Sludge (Use in Agriculture) Regulations. It should be noted that the WTRs (if used) are generally mixed with WWTRs, which provide the bulk of the nutrients.

6.2. Management

The Florida Department of Environmental Protection [57] provides guidance on management and handling of WTRs. They are considered along with biosolids, where these must not be stored, stockpiled, or staged for more than seven days, unless stored in accordance with an NMP (nutrient management plan) at an approved storage location that meets a 400 m setback from a building occupied by the public. Storage must be in accordance with the NMP and cannot cause or contribute to runoff, objectionable odours, or vector attraction.

Episodic rainfall events are a potential mechanism by which WTRs may be incorporated into runoff from farmland into drinking water supplies. This risk appears to be low under normal rainfall conditions [58], but climate-change-induced unseasonal extreme rainfall events may present a mechanism for this to become a problem in the future. However, any surface spread waste can become a problem for runoff into watercourses if spread in the wrong conditions (e.g., on top of snow or just before heavy rainfall). The risk of soluble compounds present in or mobilised from WTRs leaching into groundwater varies with climate conditions, increasing with temperature and precipitation [59].

In Scotland, under a paragraph 7 exemption, record keeping, reporting, and monitoring evaluation are key management activities that are required to demonstrate compliance with regulations. The maintenance of delivery records of WTRs is required, along with WTR application logs and records. In Florida, other records demonstrate compliance with the site nutrient management plan, including crop plantings and harvesting and applications of any other sources of nutrients. Appropriate site monitoring (soil testing and ground water monitoring) is required alongside soil fertility testing conducted at the frequency specified in the NMP. Annual soil pH testing of each application zone must be conducted to ensure the pH is at least 5 or greater [57].

7. Development of a Decision Support Tool

7.1. Purpose and Boundary of a Decision Support Tool

The development of a Decision Support Tool (DST) is required to support the relevant stakeholders in their decision-making process and allow them to explore the possibility of application of WTRs to land. The DST should also be designed to encourage the consideration of the additional benefits to the circular economy from spreading WTRs to land and further understanding the benefits of this for the soil's physical properties. The DST has been designed to include a screening process, as soil analysis and specific crop nutrient requirements need to be taken into consideration when determining the application rate for a specific location, and the specific location also needs to have the right properties that will allow for application. The boundary of the DST is shown in Figure 1.

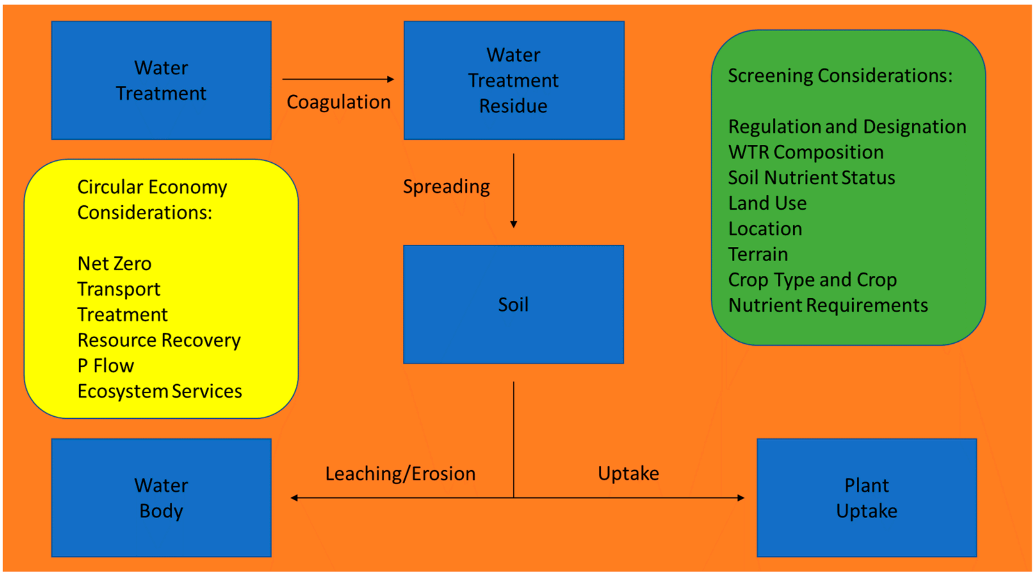

Figure 1. Decision Support Framework boundary.

7.2. Key Considerations in the Screening Process

The findings from the literature review suggest that there should be an initial screening process included in the DST to firstly inform the potential of reusing WTRs in land applications. The findings that form the basis of the key considerations to be included in the screening process are summarised in Table 8 below.

Table 8. Key considerations in the screening process taken from the literature review.

	Description	References
pH	In England and Wales, the application of Al WTRs is limited to soils above a pH of 6 due to the increased mobility of Al below a pH of 5 in soils, while Fe WTRs are limited to application to soils above a pH of 5. Application of WTRs to lands with pH < 5.2 should be avoided, given the potential for the Al in the WTR to become soluble and toxic to plants. Before the application, the properties of the WTR and receiving land (e.g., particle size distribution, pH, nutrition values, and organic content) should be analysed to evaluate the suitability of using WTRs for the specific application.	[11,24]
P fixing	WTRs may immobilise P that is already within soils; if this does occur, then it would likely exacerbate the existing Al-based immobilisation at low pH values, resulting in further reduced available P. Excessive WTR application can induce P deficiency in crops. WTR application can result in reductions in plant growth (Lactuca sativa) caused by application-induced P deficiency to both acidic and neutral soils.	[11,19,22,51]
Application rates	The application rate is linked to the requirements of the receiving soil. Application rates in England and Wales are typically in the range of 20–60 t ha^{-1}. In Scotland, application rates can vary between 50 and 150 t ha^{-1} based on N as the limiting factor.	[24]

In applying WTRs to land, it is important to consider the physical and chemical properties of both the WTR and the receiving soil. The benefits to the receiving soil's properties and the impacts of applying a WTR on the existing soil's properties are summarised in Table 9 below. These benefits are improvements in the soil properties and are summarised from the literature review.

Table 9. Impacts of applying WTRs on existing soil properties.

	Description	References
Particle size distribution	Some WTRs contain high percentages of fine particles, so if flooding is a concern, it may not be appropriate to apply fine-grained WTRs to land which will reduce hydraulic conductivity. If water retention is an issue, i.e., the receiving soil is sandy, then WTR application will result in an increase in water retention.	[16]
pH	The WTR pH ranges from 4.4 to 8. Maintaining the soil pH at optimal levels has important benefits, including increasing microbial activity in soils and maximising the availability of N, P, and K macronutrients. Higher pH soils can be prone to deficiencies in trace elements and therefore it is important to know the pH level of both the WTR and the receiving soil to ensure the pH of the receiving soil is maintained at optimum levels. In Scotland, with soil pH values below 5.6 in mineral soils, soluble aluminium inhibits root growth and reduces yields. Therefore, the application of a WTR could potentially exacerbate the Al toxicity.	[11,30]
Phosphorus	WTRs have effectively been used to reduce phosphorus in surface water runoff from agricultural lands. WTR application has reduced surface and groundwater phosphorus losses when added to Florida Spodosol with different P sources.	[32,33,47]
Organic matter	Increasing soil organic matter has a range of benefits such as increased agricultural productivity, good drainage, and low additional nutrient input requirements, as well as resulting in a better root system. Good soil structure is linked to a reduction in soil compaction and an increase in porosity. Introducing vermicompost with WTR application can increase the WTR efficiency in improving soil properties.	[8,27,32]

7.3. Structure of the DST

The DST has a three-stage structure, as shown in Figure 2.

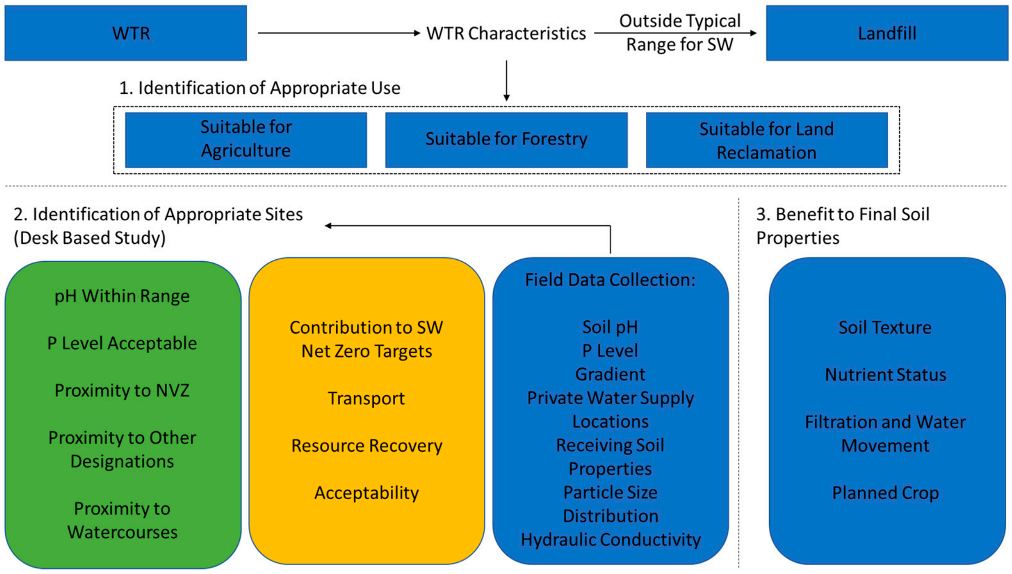

Figure 2. Decision Support Framework structure.

7.3.1. Stage 1: WTR Characteristics and Initial Screening

Stage 1.1—This stage allows for the WTR from a water treatment work (WTW) to be compared with an expected range arising from Scottish WTWs and previous results from the individual WTW across a range of characteristics. The tool automatically flags any parameters out of the expected range.

Stage 1.2—This allows for identification of appropriate sites. This initial, desk-based screening stage will use existing mapping data to help determine appropriate areas of Scotland's soils. Appropriateness will be based on key screening criteria and an analysis of additional circular economy benefits. The screening criteria were developed based on the results of the literature review and considerations of circular economy benefits will include elements such as distance from the WTW to the chosen site to allow for an assessment of travel-related carbon emissions. In Figure 2, the green-coloured text boxes represent physical criteria and the gold boxes show circular economy criteria.

The first step in the screening stage is the identification of farmer, landowner, or restoration site willingness to accept WTR spreading on land. It is anticipated that Stage 1.2 may only be needed when first establishing potential locations for WTR spreading arising from WTWs as it will exclude less preferable locations for spreading, saving the time and cost required for site-based data collection.

7.3.2. Stage 2: Field Data Collection

Stage 2.1—This stage identifies information required to be collected on site and describes the method of data collection. Following field data entry, the tool will identify, based on two key considerations of pH and P level, whether it is likely that WTRs can be applied at this location. Assuming the pH and P level are acceptable, then other site considerations are also stated which should then be considered.

7.3.3. Stage 3: Benefit to Final Soil Properties

Stage 3.1—This stage identifies the benefit of adding WTRs to different land uses. The benefits are similar for each land use type, but the application rate and timescale for benefits are different.

Stage 3.2—This stage calculates the maximum application rate from the WTR data entered in Stage 1. It references the need for nutrients for two crop types and land restoration to 20 cm. The DST developed is primarily a screening tool. In this respect, it can estimate what is likely to be a maximum suitable spread rate based on the WTR. This tool cannot provide the expertise of an agricultural adviser, given that there are more land uses and more recent soil analyses to be considered when determining the application rate for a certain field, but it can provide an initial assessment to assist in the decision-making process. Expert advice should still be sought to determine the required application rate.

8. Conclusions

This project evaluates the opportunities and implications of applying WTRs to land and the associated circular economy benefits based on an analysis of the international literature. This project also develops a user-friendly DST which can be used as a screening tool to help in the decision-making process and to allow stakeholders to identify potential sites for application and determine if the application of WTRs is possible at that specific site. This work has demonstrated that WTRs can provide a useful amount of major and secondary nutrients and can be used for land restoration, for both agricultural and forestry land, as either a sole treatment or co-applied with other biosolids.

The project has identified benefits in relation to the principles of the circular economy through increasing the use of WTRs on agricultural land. Benefits to the circular economy in Scotland are achieved by reducing the distance travelled from Scottish Water Waste Treatment Works to local application locations. Additional benefits are through the valorisation of the WTR where the beneficial nitrogen content of the WTR can displace the use of nitrogen-based fertilisers, leading to a reduction in CO_2 emissions, in keeping with the principles of a circular economy and contributing to climate change mitigation. More information is required to develop a full lifecycle analysis to explore the most beneficial use of WTRs within a circular economy. The lifecycle analysis process will enable a detailed comparison to be made between different outlets for WTRs alongside application to land, with a view to maximising the circular economy benefits.

Overall, the development of a DST has allowed stakeholders to use information developed from an in-depth review of the available literature and legislation to properly identify candidate sites for the reuse of WTRs, as opposed to disposing of such waste via landfill. Not only will this provide many societal benefits, such as a reduction in waste being sent to landfill, but it will provide business benefits, as costs are reduced as the volume of waste being sent to landfill is reduced. There is also the potential for cost recovery if such a waste material can be repurposed and sold to landowners looking to improve their soil properties.

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