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RESEARCH

Seasonal Variation of Iron Removal in Coal Mine Water Discharge Lagoons and Constructed Wetlands

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Abstract

Passive Mine Water Treatment Schemes (MWTS) have been developed to remove iron from coal mine drainage before discharge into the environment to prevent potential ecological environmental problems caused by coal mine drainage. The performance of these MWTS vary across distinct climatic conditions, based on variations in temperature and rainfall patterns. Therefore, this study analysed monthly iron concentrations of 10 settlement lagoons and 5 constructed wetlands (CWs) in 5 full-scale MWTS to assess the influence of varying seasons on the MWTS lagoons and CWs treatment performance using removal efficiency (R.E.). The comparative performance of monthly records of Ferric Iron (Fe^{3+}), Ferrous Iron (Fe^{2+}) and pH over a period of 12 years were conducted using comprehensive statistical analysis of Fe^{3+} R.E. Fe^{3+} R.E. obtained for the 5 MWTS ranged from 50.97 to 98.98% and is in the order Site A > Site C > Site B > Site E > Site D. pH increased from slightly acidic to neutral while Fe^{3+} and Fe^{2+} concentrations decreased. The MWTS achieved an average Fe^{2+} R.E. of 46.66–98.16%. Seasonal variation affected iron removal from the schemes, i.e., Fe^{3+} R.E. in the lagoons and wetlands was in the order summer > spring > autumn > winter. Due to increased rates of sedimentation, filtration and absorption in warmer seasons, treatment performance of the schemes was better compared to colder seasons. The experimental results obtained provide a scientific basis for MWTS operators, research scientists, policy makers to improve R.E. and ensure consistent treatment performance in different seasons throughout the year.

Highlights

- We investigated iron removal from coal mine drainage in different seasons.
- All MWTS lagoons and wetlands exhibited higher Fe^{3+} R.E. in summer and spring.
- Highest variation in Fe^{3+} R.E. across lagoons was 40% from cold to warm season.
- Greatest variation in Fe^{3+} R.E. across wetlands was 19% from cold to warm season.
- Four of the five full-scale passive MWTS achieved Fe^{3+} R.E. greater than 80%.

Keywords Iron removal · Seasonal variation · Water treatment · Full-scale lagoons · Full-scale wetlands · Mine discharge

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1 Introduction

Coal mining is one of the major industries that contributes to UK economy, but it also causes noticeable deterioration to the environment, such as acidification of the local watercourses, limitation of river ecology growth, surface and groundwaters pollution, and more (Rinder et al. 2020; Bowell et al. 2023). Discharging coal mine wastewater into waterways is frequently associated with unnatural alterations to water quality, identified as elevated salinity levels, modifications to pH and ionic balance, and higher amounts of heavy metals (Fleming et al. 2021). Coal mine drainage is characterized by elevated Iron (Fe) and Sulphate (SO_4^{2-}) concentrations, contributed by oxidation of pyrite (Kusin et al. 2021; Bowell et al. 2023). If not properly treated prior to discharge, mine drainage can cause serious impact on the environment, aquatic ecosystems, water quality and drinking water resources (Fleming et al. 2021; Walls et al. 2023).

In response to the deterioration and other various notable ecological problems associated with coal mine drainage, different schemes have been developed to remove contaminants from coal mine-impacted waters before being discharged into the environment. These include Passive Mine Water Treatment Schemes (MWTS), drainable limestone beds (DLB), passive vertical flow ponds (VFP) and wetlands, sulphate reducing passive biofilters (SRPB) or disperse alkaline substrate (DAS). These passive treatment technologies are being used all over the world for long-term remediation of mine water, due to their cost-effective operation, maintenance and monitoring (Hedin et al. 2019; Hedin 2020; Dube et al. 2021; Jacob et al. 2022).

Passive MWTS, comprising of settlement lagoons and aerobic wetlands, have been constructed with a main focus on Fe removal from coal mine drainage (Khachatryan et al. 2021; Walls et al. 2023; Welman-purchase and Hansen 2023). Fe is the primary metal of concern in mine drainage and its removal is prioritised in the design of MWTS as it potentially exists in association with other metals (such as Al and Mn), providing adsorption sites for the metals. Passive MWTS are therefore designed with the objective to encourage ferrous iron (Fe^{2+}) oxidation to ferric iron (Fe^{3+}), followed by Fe^{3+} hydrolysis to ferric oxyhydroxide (known as ochre) which then settles within settlement lagoon(s) (Strosnider et al. 2020; Khachatryan et al. 2021; Welman-purchase and Hansen 2023).

MWTS are strategically constructed adjacent to abandoned coal mines, known as collieries, which often contain significant amounts of Fe sulphide minerals; therefore, a fundamental indicator for evaluating MWTS performance is Fe removal (Chen et al. 2020; Opitz et al. 2022; Satterley and Moorhouse-parry 2023). Drainage from these areas contains high concentrations of unoxidized Fe, which can contribute to acidity and ochre formation, if released directly into the environment. Regulatory bodies in the UK, such as the Environment Agency, enforce strict limits on Fe concentration in treated mine water discharged into water bodies, as indicated in the Water Framework Directive (England and Wales) Regulations 2017. This is an essential measure for preserving aquatic ecosystems, wildlife biodiversity and human health from the adverse effects of Fe contamination. Effective Fe removal from mine water also aids the removal of other metals and contaminants by providing adsorption sites for other metals, therefore, contributing to an overall improved water quality.

A limited number of studies have been carried out on the removal of heavy metals in different seasons from lagoons and wetlands (Oujidi et al. 2020; Tuladhar and Iqbal 2020;

Singh and Chakraborty 2020; Tran et al. 2022). Compared to nutrient removal, where plant uptake is significant to the heavy metal treatment, plant biomass does not contribute to a significant increase of metal removal; instead, sediment and substrate are the major adsorbents leading to metal accumulation. Contaminant removal mostly occurs within the top few centimetres of soils and in a constructed wetland (CW) vegetated system (Tuladhar and Iqbal 2020). These studies have revealed that metal (Fe, Mn and Zn) retention in a studied wetland receiving agricultural runoff, was better during spring, compared to winter (Oujidi et al. 2020; Heiderscheidt et al. 2020). During summer, metal ions precipitates have been observed to be deposited onto root surfaces forming plaques which can adsorb a large capacity of other metals (Tuladhar and Iqbal 2020; Ma et al. 2020). Higher removal of metals in warmer temperatures has also been observed in previous studies (Ulén et al. 2019; Tuladhar and Iqbal 2020; Ma et al. 2020; Tran et al. 2022; Fan et al. 2023). However, during colder seasons, reduced uptake of Fe and Mn by plants and lower photosynthesis rates, lead to anoxic conditions possibly in the water column and in the sediments (Ma et al. 2020; Zhu et al. 2022; Tran et al. 2022).

The performance of CWs treating Fe removal has been observed to be significantly influenced by variations in seasons, such as vegetation growth, changes in air and water temperature, precipitation and flow rate variation. These variations influence the hydraulic and treatment performance of these systems. For instance, during growing season, plants tend to offer resistance to flow rate variation, affecting hydraulic performance parameters (Abbasi et al. 2019; Heiderscheidt et al. 2020; Tran et al. 2022). When these plants become mature, sedimentation also increases as a result of the matured vegetation high flow resistance which reduces the flow velocity of water, increasing the retention time and metals adsorption (Ioannidou and Pearson 2019; Kretz Id et al. 2021).

Overall, the above reported studies highlighting the relationship between treatment performance and seasonal variations, have revealed that treatment systems are more effective in warmer seasons than in colder seasons due to warmer seasons being favourable to plant growth, whose roots adsorb contaminants from the drainage. However, the above mentioned studies were conducted on other than mine discharge wastewater types, like municipal wastewater, agricultural runoff, sewage and dairy wastewater. In addition, most of those studies were carried out in pilot-scale systems, whereas studies within scientific literature on the effects of seasonal variation is lacking in full-scale systems receiving mine drainage.

To address this gap, this study examines for the first time the relationship between treatment performance and seasonal variations in full-scale and large MWTS which treat mine discharge, also employing long timeseries datasets. This paper presents findings on the influence of seasonal variation and plant growth on Fe removal from five full-scale MWTS and their respective lagoons and wetlands. Seasonal variation attributed to patterns, delineated by monthly segmentation, provides valuable insights into the seasonality of the MWTS ecosystems, and its influence on MWTS. It should be noted that the wetlands under investigation are CWs.

2 Methodology

2.1 Sites Overview

The study was conducted using data from five full-scale Passive MWTS located in the north of England, consisting of settlement lagoons and CWs. A total of 10 settlement lagoons and 5 Free Water Surface Flow (FWSF) CWs within the 5 schemes were assessed for their respective treatment performances. Table 1 summarises the number of lagoons and CWs. The 5 investigated MWTS vary in layout, and number of assets included.

Full details of the operational parameters and design aspects, i.e., shape, surface area, aspect ratio, position of inlet and outlets (configuration), number of inlets and outlets, of the studied MWTS lagoons and CWs have been published in Okeleji and Ioannidou (2024). Moreover, a summary of the design aspects, operational conditions and types of CWs studied is presented in Table 2. The lagoons and CWs short forms are listed in column 1 of Table 2, while the design and operational aspects of surface area, design depth, operational depth, aspect ratio, length, width, CW flow type and CW vegetation species are presented respectively in columns 2 to 9 of Table 2.

Schematic diagrams of the MWTS studied are provided in Figs. 1 and 2, illustrating the components involved, flow direction, layout, and shape of each scheme, while an overview of the 5 MWTS sites is presented as follows. Site A MWTS consists of the cascade which flows into 2 parallel lagoons- Lagoon 1 and Lagoon 2, as shown in Fig. 1a. the discharge from both lagoons is received by another cascade, which flows into a lagoon (Lagoon 3) that feeds into a set of wetlands connected in series (Fig. 1a), prior to the discharge of the treated mine water. Site B MWTS comprise of a cascade which flows into 3 parallel settlement lagoons (Lagoon 1, Lagoon 2 and Lagoon 3), which individually flow into 3 separate wetlands in parallel arrangement, as presented in Fig. 1b. As in Fig. 1b, these wetlands (Stage 1) are also followed by another set of 3 wetlands in parallel arrangement. However, it is important to note that the latter set of wetlands (Stage 2) is for maintenance.

Site C MWTS consists of a cascade, 2 settlement lagoons connected in parallel, and a wetland (see Fig. 2a). This scheme has a sludge bed which receives flow from the second lagoon before flowing into the wetland, after which the treated mine water is discharged. Site D MWTS receives the mine water directly into its lagoon, as there is no cascade present (see Fig. 2b). Flow from the lagoon is received by a long and narrow wetland for further treatment prior to discharge of the treated flow. Site E MWTS consists of an aeration cascade, a settlement lagoon and 3 wetlands connected in series as shown in Fig. 2c.

Table 1 Scheme name and associated features of studied passive MWTS

Scheme	No. of Lagoons	No. of Wetlands	Operation Type
Site A	3	3	Passive MWTS
Site B	3	3	Passive MWTS
Site C	2	1	Passive MWTS
Site D	1	1	Passive MWTS & Gravity Overflows
Site E	1	3	Passive MWTS

Table 2 Design aspects of the studied MWTS settlement lagoons and CWs

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)
	Surface Area (m^2)	Design Depth (m)	Water Level "Operational Depth" (m)	Aspect Ratio	Length (m)	Width (m)	CW Type	CW Vegetation
Lagoons								
A-Lag1	1314.60	3.00	2.00	2.0	51.28	25.64	-	-
A-Lag2	892.90	3.00	2.00	0.5	21.13	42.26	-	-
A-Lag3	1455.40	3.00	2.00	4.0	76.30	19.08	-	-
B-Lag1	2664.29	3.00	2.05	2.0	72.99	36.50	-	-
B-Lag2	3528.29	3.00	2.05	2.0	84.00	42.00	-	-
B-Lag3	2517.19	3.00	2.05	3.0	86.90	28.97	-	-
C-Lag1	1613.90	3.00	2.00	3.0	69.58	23.19	-	-
C-Lag2	1590.39	3.00	2.00	3.0	69.07	23.02	-	-
D-Lag	148.04	3.00	2.60	0.5	8.60	17.21	-	-
E-Lag	895.00	3.00	2.50	3.0	51.82	17.27	-	-
Wetlands								
B-Wet1	2613.98	1.65	1.65	1.5	62.61765	41.75	Free Water Surface Flow	Common reed
B-Wet2	2606.94	1.65	1.65	1.5	62.53327	41.69	Free Water Surface Flow	Common reed
B-Wet3	2593.20	1.65	1.65	2.5	80.51708	32.21	Free Water Surface Flow	Common reed
C-Wet	1597.11	1.00	1.00	1.5	48.94553	32.63	Free Water Surface Flow	Common reed
D-Wet	453.29	1.00	1.00	9.0	63.87182	7.10	Free Water Surface Flow	Common reed

2.2 Data Overview

This research is based on time-series data from full-scale MWTS. The data contains monthly records spanning over a period of 12 years, i.e., from January 2007 to September 2018. Site C, however, only includes data from January 2014 to September 2018. The dataset contains daily records of Fe^{3+}, Fe^{2+} and pH. These daily records were then averaged to create monthly records provided by the site operators for this study. ICP-OES method was employed for water quality analysis to determine the concentration of iron ions.

The removal efficiencies R.E. of the systems for the measured contaminants were calculated using Eq. (1) (Orden et al. 2021; Zhu et al. 2022; Okeleji and Ioannidou 2024):

$$R.E.\ (\%) = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \tag{1}$$

where C_{in} = influent concentration, and C_{out} = effluent concentration.

2.3 Statistical Methods

The influence of seasonal variation on the treatment performance of the schemes is explored by conducting statistical analyses. Descriptive analyses (minimum, mean, maximum and standard deviation) were used for data analysis and to compare the MWTS R.E. in different seasons. All statistical analyses were performed with Python 3.6.8 (Islam Khan et al. 2021; Taoufik et al. 2022).

Seasonal variation attributed to distinctive climatic conditions, such as variations in temperature and rainfall patterns, were investigated using the monthly data. Based on the months of data readings, 2 seasonal divisions were made to allow for comparison.

- Division of hydrological year into 4 seasons: Spring - March to May; Summer - June to August; Autumn– September to November; and Winter– December to February.
- Cold and Warm seasons based on MET Office temperatures history of the schemes over the period of study, with the Warm months being from May to October and Cold months from November to April. Warm months in the location of the MWTS experienced temperatures within the range of 11.56°C and 14.78°C, while in Cold months temperatures ranged from 2.37°C to 8.02°C over the study period.

The effect of seasonal variation on Fe R.E. in the studied MWTS was analysed using a two-way Analysis of Variance (ANOVA) to determine their statistical significance (Fernández-Pascual et al. 2019; Mayes et al. 2021).

3 Results and Discussion

The seasonal variation in this study is attributed to patterns segmented based on monthly delineations, as presented in Sect. 2.3, to analyse the influence of environmental dynamics, such as patterns of precipitation, varying temperature, and biological activity in the studied MWTS all through the year.

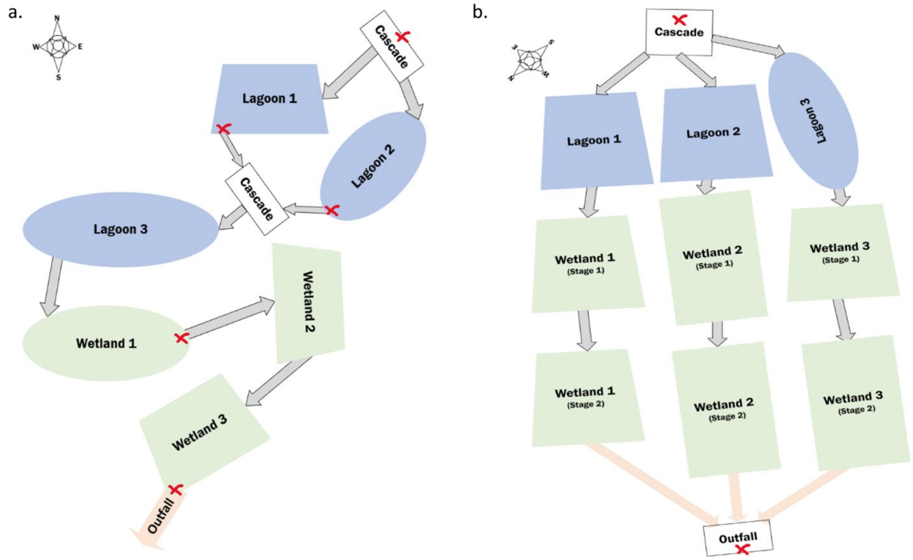


Fig. 1 Schematic Diagram of: (**a**) Site A MWTS; and (**b**) Site B MWTS (not drawn to scale). Sampling points for chemical analyses are indicated by X symbol

3.1 Contaminant Removal from the MWTS

The performance of the five full-scale MWTS investigated in this study is analysed using their respective Fe^{3+} removal efficiency. Other considered parameters are Fe^{2+} and pH.

3.1.1 Iron (Fe)

As discussed in Sect. 1, Fe removal is used as a key performance indicator for MWTS in the UK (Opitz et al. 2020; Li et al. 2020; Zhu et al. 2022). The highest Fe^{3+} R.E. of 98.98% among the 5 MWTS was observed at Site A. Overall, Fe^{3+} R.E. in the 5 schemes was achieved in the order Site A > Site C > Site B > Site E > Site D. While Site C recorded a Fe^{3+} R.E. of 96.58%, Site B, Site E and Site D achieved Fe^{3+} R.E. of 94.62%, 83.83%, and 50.97% respectively, as presented in Fig. 3.

Site A MWTS recorded the highest Fe^{3+} R.E. of 98.98% and lowest Fe^{3+} effluent concentration of 0.02 mg/L compared to the other schemes (Fig. 3). Compared to the other schemes, as presented in Fig. 1a, Site A has a greater number of assets for treatment which have contributed to the 98.98% Fe^{3+} R.E. Site B is similar to Site A as it also has 3 lagoons and 3 wetlands; however, it has only 1 cascade (Fig. 1b). Site B recorded a lower average Fe^{3+} R.E. of 94.62% and a higher average Fe^{3+} effluent concentration of 0.60 mg/L compared to Site A (Fig. 3). Site B had a cascade, but unlike Site A with serially connected wetlands, Site B wetlands were in a parallel arrangement receiving flow from individual lagoons and only converging at the outfall for exit. The design aspects of these scheme's asset, such as lagoons and wetlands, have been studied to assess the impact of factors like number of assets, position, inlet-outlet configuration on Fe R.E., with findings detailed in Okeleji and Ioannidou (2024).

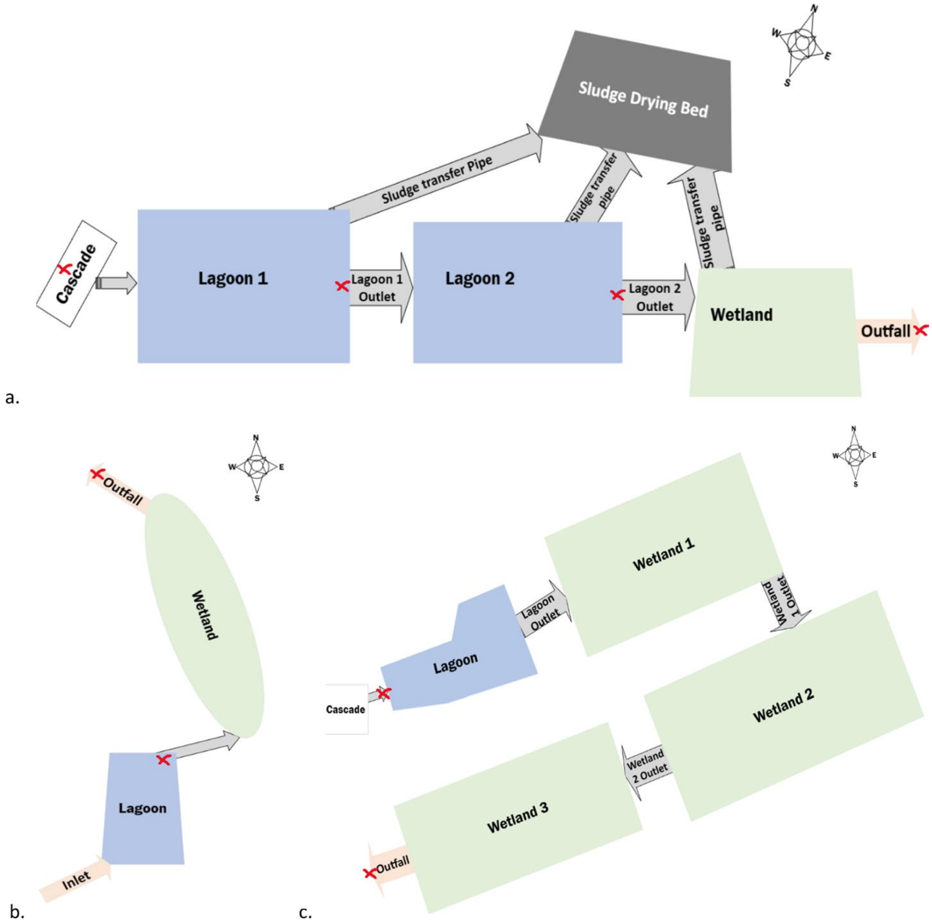
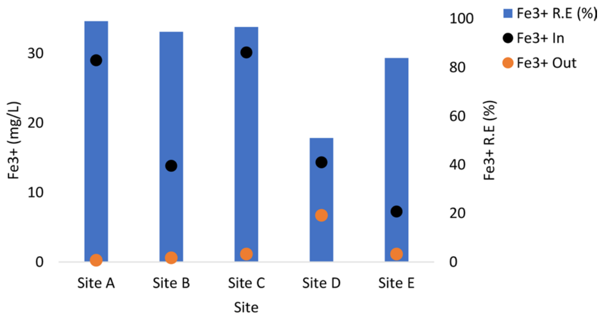


Fig. 2 Schematic Diagram of: (**a**) Site C MWTS; (**b**) Site D MWTS; and (**c**) Site E MWTS (not drawn to scale). Sampling points for chemical analyses are indicated by X symbol

Fig. 3 Average removal efficiencies, influent and effluent Fe^{3+} concentrations of the studied MWTS



Site C MWTS achieved a total of 96.59% Fe^{3+} R.E. The presence of a sludge bed for ochre removal prior to further treatment in the wetland might have contributed to the Fe^{3+} high R.E. in Site C MWTS. Sludge beds reduce the quantity of ochre, which accumulates at the bottom of the treatment system and limits contaminant adsorption on the system sediments (Hu et al. 2019). Site D, however, recorded the lowest Fe^{3+} R.E. of 50.97% compared to the higher R.E. of other schemes, and a Fe^{3+} discharge concentration of 6.69 mg/L (Fig. 3). This lower Fe R.E. could be due to its gravity overflow mode of operation, unlike the other studied schemes, or its design aspects (Okeleji and Ioannidou 2024). Overall, Site E MWTS achieved 83.83% Fe^{3+} R.E. As in Site A, Site E lagoon empties into 3 wetlands connected in series (see Fig. 2c) which retained more Fe^{3+} prior to discharge from the scheme.

Similar to previous studies, a full-scale MWTS achieved 85% Fe^{3+} R.E. over a 3-year period, with an average effluent Fe^{3+} concentration of 2 mg/L (Bowell et al. 2023). This is also comparable to an effluent Fe^{3+} concentration range of 0.10 to 5.0 mg/L from another full-scale MWTS monitored over a period of 4 years, which achieved approximately 90% Fe^{3+} R.E. (Moorhouse-Parry and Satterley 2023).

Fe^{2+} effluent concentrations were consistently lower than the corresponding influent, as observed in Fig. 4, which shows that Fe^{2+} is being oxidised to Fe^{3+} as the mine water flowed. Fe^{3+} then began to settle, resulting in the decrease of Fe^{3+} concentration in the flow. This also indicates that both the oxidation rate of Fe^{2+} and the settling rate of Fe^{3+} are important in the general treatability of the mine drainage (Kleinmann et al. 2021; Welman-Purchase and Hansen 2023).

A significant difference in both Fe^{2+} and Fe^{3+} concentrations at the influent and effluent can be observed in Fig. 4. In the schemes, most Fe is generally present in the form of dissolved Fe^{2+} at the start of the treatment. If a cascade is present, oxygenation will commence in the cascade initializing the conversion process of some of the Fe^{2+} present in the mine water to Fe^{3+} precipitates. More oxidations occur in the lagoon(s) converting Fe^{2+} to Fe^{3+}. As the retention time increases, the Fe^{3+} begins to settle, indicating that the rates of oxidation of Fe^{2+} and settling of Fe^{3+} are crucial to mine drainage treatment within the lagoon.

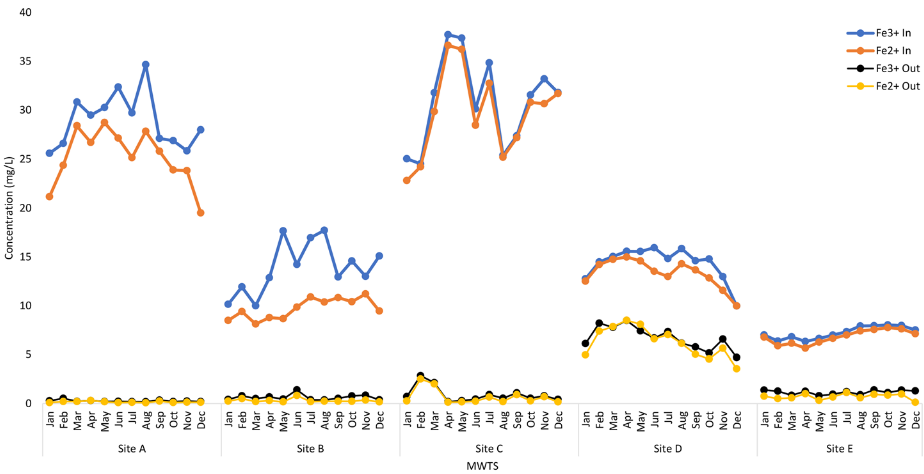


Fig. 4 Change in monthly average Fe^{2+} and Fe^{3+} concentration with time at the five MWTS over 12 years

For the five MWTS, mean Fe^{3+} effluent concentration ranged from 0.25 to 8.12 mg/L (Fig. 3). The studied schemes discharge into water bodies ranging from river tributaries, to dikes and estuaries. The discharge consent limit for Fe in treated mine water that is released into these water bodies varies and depends on the regulations of the country or region (Strosnider et al. 2020). In the UK, the discharge consent limit for Fe in treated mine water discharged into water bodies is set by the Environment Agency under the Water Framework Directive (England and Wales) Regulations 2017 (WFD).

According to WFD 2017/407, Fe discharge consent limit depends on the Fe loading rate, whether or not the discharge gives rise to a visible plume in the receiving water, and if a 'good' ecological status is achieved (Machado et al. 2019; Environment Agency 2022). Furthermore, although the Environment Agency set estuaries and coastal waters specific pollutants and operational environmental quality standards (EQS) to guide discharge concentrations, no maximum allowable concentration EQS is or has been set for Fe.

3.1.2 pH

It is vital to consider the relationship between Fe R.E. and pH, given that a corresponding increase in pH was observed as Fe^{3+} was removed (Fig. 5). The oxidation of Fe^{2+} by dissolved oxygen within lagoons results in higher pH, hence, an increase in pH can be observed in Fig. 5 (Moore et al. 2022; Bowell et al. 2023). This is similar to Bowell et al. (2023) full-scale MWTS study, where pH increased gradually over the monitoring period, with all values falling within the permitted discharge range. Furthermore, as pH values exceed 7, sorption of metals (e.g., Zinc, Nickel, Cadmium) eventually occurs as Fe oxyhydroxide (or ferrihydrite) forms, when Fe^{3+} precipitates begin to settle. The pH values obtained in the effluent of the studied sites were identified in the neutral range (6.93–7.97) and were in accordance with the maximum allowable concentration EQS of pH 6.5–8.5 for discharge into estuaries and coastal waters set by the Environment Agency (2022).

After mine water treatment, pH increase is a common characteristic of passive MWTS systems, which contain sufficient bicarbonate alkalinity (i.e., net-alkaline water). Alkalinity is not defined by the pH of the mine water but is rather a net effect of the presence of important constituents, such as carbonate (CO_3^{2-}), bicarbonate (HCO_3^-), and hydroxyl (OH) anions (Madzin et al. 2020; Walls et al. 2022). In a net-alkaline environment, as mine water flows through the MWTS, there would be a consumption of HCO_3^- by the acid released during the oxidation and hydrolysis of Fe^{2+}. The decrease in alkalinity (mg/L $CaCO_3$) in the schemes, as well as a value of pH > 6.3 is an indication that the treatment system is net alkaline (Hedin 2020; Moorhouse-Parry and Satterley 2023). The increase in pH may be

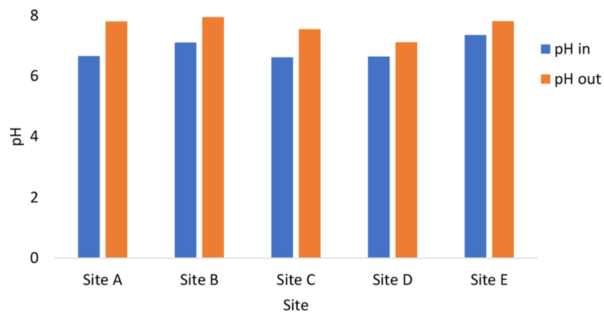


Fig. 5 Average pH values of the studied MWTS for influent and effluent

traceable to dissolved CO_2 degassing, which is often above CO_2 atmospheric partial pressure upon mine drainage emergence to the environment (Opitz et al. 2021; Moore et al. 2022). Degassing may decrease CO_2 via dissolution of HCO_3^-, releasing CO_2 (g), but also producing OH^- ions, and therefore increasing pH.

3.2 Seasonal Variation of Fe Removal Efficiency in the MWTS

The breakdown of the average seasonal efficiencies for the different MWTS is presented in Fig. 6. The statistical significance of the effect of seasonal variation on the Fe^{3+} R.E. was studied using two-way ANOVA with a significance probability associated with the F

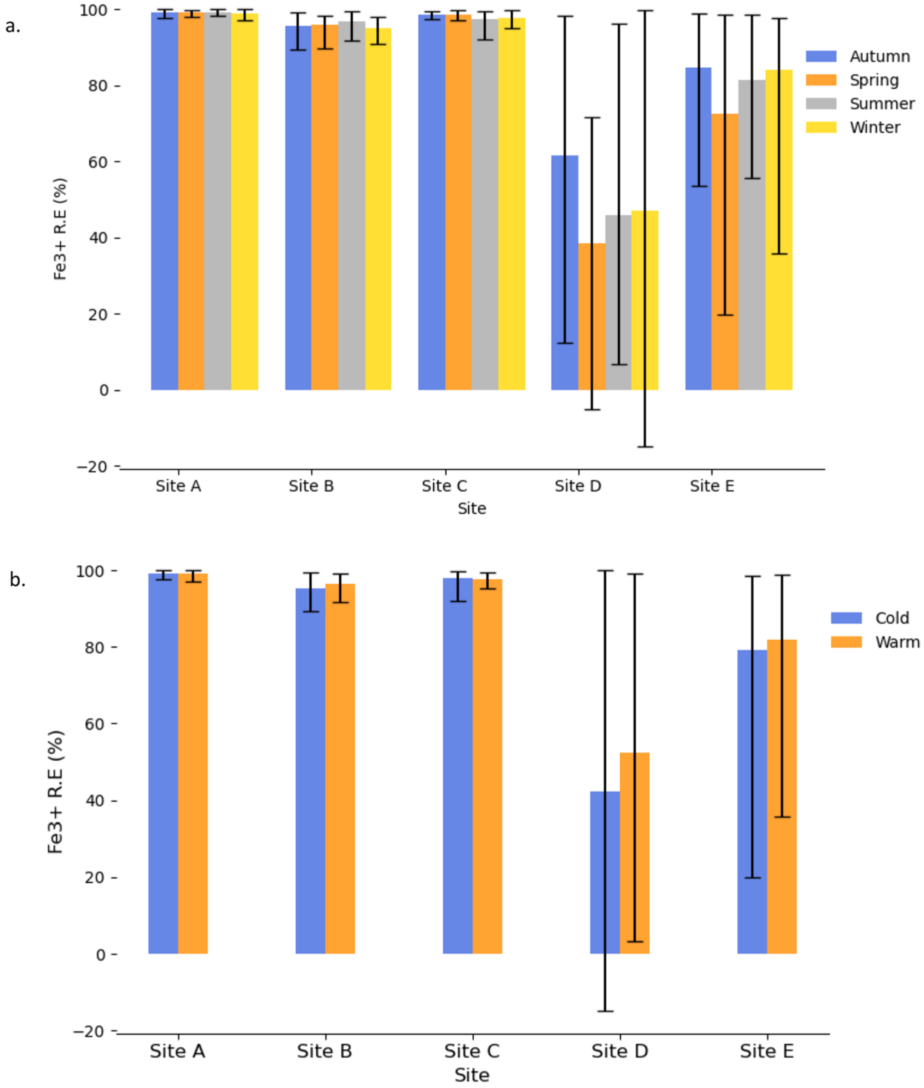


Fig. 6 Average Fe^{3+} R.E. of: (**a**) autumn, summer, spring, winter; and (**b**) cold, warm seasons ($p < 0.001$)

statistic: Pr(>F) < 0.001 (Fernández-Pascual et al. 2019; Mayes et al. 2021). This suggests that the interaction between the MWTS and season is statistically significant, and the effect of MWTS on Fe^{3+} R.E. depends on the season and vice versa, indicating thereby that some MWTS might perform better in specific seasons.

Overall, Fe R.E. was greater in summer for all schemes, followed by spring, autumn and winter. Warm season (defined as May to October in this study) also recorded higher Fe^{3+} R.E. for all the schemes, compared to cold season.

In summer and warm season, fewer rainfalls occur, resulting in higher pollutant concentrations, increased activity of algae and microorganisms breaking down contaminants, increase in rate of chemical reactions, higher sedimentation and filtration, and finally, higher removal efficiencies (Valkanas and Trun 2018; Fan et al. 2023). For instance, Valkanas and Trun (2018) observed that Fe concentration during summer was 50% greater than in winter in the full-scale passive abandoned coal mine drainage remediation system consisting of 5 lagoons and one wetland. In terms of Fe R.E., about 85% of Fe was removed in summer, while 51% in winter (Valkanas and Trun 2018).

In winter and cold season, the regions of the studied schemes experienced higher precipitation, coupled with drop in temperatures, which were associated to lower pollutant concentrations, reduced activities of microorganisms, reduced rates of chemical reactions, and lower sedimentation, filtration and adsorption (Banks et al. 2019). This has accounted for lower removal efficiencies occurring in winter and cold seasons in the treatment systems. Kuyucak et al. (2006) observed that due to freezing conditions in winter, there was reduced settling in a pilot-scale passive treatment system, which resulted in lower Fe^{3+} R.E., as high concentrations of Fe were present in the effluent, compared to Spring. Fe R.E. was 22.5% in winter, with spring achieving 95% (Kuyucak et al. 2006).

Contrary to other studied MWTS which experienced Fe^{3+} R.E. in the order of summer > spring > autumn > winter, Site D MWTS Fe R.E. followed the order summer > autumn > spring > winter. Site D Fe R.E. seasonal variation was similar to Chen et al. (2020) study which recorded higher Fe^{3+} R.E. in summer and autumn, compared to spring and winter. The plants in the pilot-scale passive treatment system treating mine drainage were also observed to be abundant in summer and autumn (Chen et al. 2020). Previous studies have demonstrated that the type of vegetation has a significant influence on wetlands treatment performance and should also be considered as a factor when evaluating seasonal variation of wetlands treatment performance (Wang et al. 2016; Li et al. 2018; Miranda et al. 2019).

3.3 Seasonal Variation of Fe R.E. From Lagoons and Wetlands

The seasonal variation of Fe^{3+} removal efficiencies in lagoons and wetlands for the 2 different season divisions are presented in Table 3 (lagoons) and Table 4 (CWs). The lagoon and CW short forms are listed in row 2 of their respective tables. This section further discusses the seasonal variation effect on the investigated lagoons and CWs.

3.3.1 Seasonal Changes in Fe Concentrations in Lagoons

The seasonal variation of Fe^{3+} R.E. in the studied lagoons for the different season divisions is presented in Fig. 7. As presented in Fig. 7a, in most of the lagoons the highest average Fe^{3+}

Table 3 Summary of treatment performance data for the settlement lagoons (±S.D.)

Lagoons	SITE A-Lagoon1	SITE A-Lagoon2	SITE A-Lagoon3	SITE B-Lagoon1	SITE B-Lagoon2	SITE B-Lagoon3	SITE C- Lagoon1	SITE C- Lagoon2	SITE D-Lagoon	SITE E-Lagoon
Lagoons short form	A-Lag1	A-Lag2	A-Lag3	B-Lag1	B-Lag2	B-Lag3	C-Lag1	C-Lag2	D-Lag	E-Lag
Influent Fe^{3+} (mg/L)	29.52±16.87	29.40±16.79	12.29±7.68	13.26±9.46	13.26±9.46	13.26±9.46	39.28±19.45	7.12±3.12	15.80±7.23	7.25±5.67
Effluent Fe^{3+} (mg/L)	13.25±3.58	11.62±5.79	1.33±2.26	2.58±2.82	1.79±2.47	2.87±2.24	6.82±3.74	3.23±3.28	13.63±5.98	5.02±3.19
Fe^{3+} R.E. (%)	54.94±19.36	60.34±18.03	89.21±12.91	80.29±12.10	86.48±14.94	78.10±12.49	82.65±6.61	54.71±15.99	13.72±14.28	30.48±15.81
Number of measurements	132	124	96	101	99	98	74	74	160	107
Seasonal Fe^{3+}R.E. (%)										
Summer	67.45±14.91	68.07±14.89	94.35±6.62	83.1±13.24	91.99±8.54	81.34±10.82	85.47±3.71	55.12±20.61	18.46±19.53	37.65±18.54
Spring	52.54±19.44	61.12±15.66	90.81±15.49	84.2±8.37	87.59±7.43	79.98±12.61	83.21±5.67	56.35±13.31	10.71±11.93	31.1±15.53
Autumn	53.83±16.12	61.18±17.17	88.87±12.80	83.15±9.01	87.29±12.71	78.84±9.99	83.37±4.37	53.66±17.35	14.44±10.74	27.12±14.80
Winter	46.00±20.49	50.79±20.44	83.13±12.64	71.06±12.63	79.79±22.86	72.27±14.94	79.50±9.00	53.36±14.49	10.46±10.65	25.87±11.60
Cold	48.21±19.33	54.73±18.95	86.18±14.58	76.19±11.39	82.97±17.68	73.92±12.93	81.47±7.88	56.54±13.34	11.30±11.71	29.37±15.52
Warm	61.68±17.03	65.78±15.39	92.37±10.13	84.3±11.51	89.93±10.75	82.11±10.71	84.21±4.05	52.30±18.86	15.85±15.99	31.53±16.16

R.E. was recorded in summer, followed by spring which achieved higher Fe^{3+} R.E. than in autumn and winter. The lagoons with this order of removal (summer > spring > autumn > winter) include A-Lag3, B-Lag2, B-Lag3, C-Lag1, C-Lag2 and E-Lag. However, in A-Lag1, A-Lag2 and D-Lag, the highest order of Fe^{3+} R.E. was summer > autumn > spring > winter, with a higher Fe^{3+} R.E. achieved in autumn than in winter. However, for all 10 lagoons, Fe^{3+} R.E. was higher in warm season than in cold season (see Fig. 7b).

From Fig. 7, it can be observed that the lagoons have recorded varying Fe^{3+} R.E., with most notable difference identified in D-Lag. The drop of Fe^{3+} R.E. in D-Lag is, however, not relevant to seasonal aspects (see Fig. 7), but is largely attributed to its significantly different design aspects, i.e., the lagoon significantly smaller dimensions and scale compared to the other lagoons (details of which are listed in Table 2). As reported in a study conducted by Okeleji and Ioannidou (2024), it is demonstrated that the design aspects of lagoons, such as water level and surface area, significantly influence their performance. These particular design aspects have been found to play a critical role in the overall treatment efficiency of Fe^{3+}, by affecting the development of dead zones and the percentage of effective volume of the system. Greater surface areas and lower water levels were observed to have resulted in a higher Fe^{3+} R.E. from the lagoons (Okeleji and Ioannidou 2024). It is therefore important not to overlook the influence of design aspects of lagoons when investigating their treatment performance throughout the year.

Unlike the other lagoons, B-Lag1 attained higher Fe removed in spring and autumn, compared to summer, following the order of spring > autumn > summer > winter. It can be inferred that within the scheme, the assets had different response to seasonal variation, as observed by their varying treatment performance. It is inferred that their hydraulic design aspects also influence their treatment performance in different seasons, given that design aspects have been recorded to impact retention time, mixing, effective volume involved in treatment, and other contaminant removal processes. For instance, if the effect of operational water depth is removed, surface sediments with and without vegetation will have similar metal retention capacity (Goulet et al. 2001; Hu et al. 2019; Li et al. 2019). Therefore, the impact of design aspects is an element that should also be understood and factored in the seasonal performance of lagoons to enable the effective development and implementation of strategies that can maximise R.E. and attain optimal and consistent treatment performance in different seasons throughout the year. For example, to optimize Fe R.E. from lagoons in varying seasons, the lagoon inlet could be modified in warmer seasons by adding baffles to improve mixing and prevent dead zone occurrence; in cold seasons, however, the flow rate of drainage into the lagoons could be reduced to increase the retention time of the drainage in the lagoon and allow for sedimentation and pollutants adsorption prior to discharge (Badhe et al. 2014; Lu et al. 2016; Ma et al. 2020).

Further investigation into the Fe^{3+} concentrations at the outlet of each asset is presented in Fig. 8, which suggest lower concentration in summer, and highest in winter for most of the lagoons. From Fig. 8, A-Lag1, A-Lag2, A-Lag3, B-Lag2, C-Lag1, C-Lag2 and D-Lag recorded the lowest Fe^{3+} concentrations in summer. B-Lag1, B-Lag3 and E-Lag, however, achieved lowest Fe^{3+} concentrations in spring. Overall, all the lagoons recorded highest Fe^{3+} concentrations in winter, which supports the lowest Fe^{3+} R.E. obtained as discussed above.

In winter and cold seasons, more precipitation and rainfall are expected to occur within this microclimate, which would result in higher amounts of drainage flowing into the lagoons and wetlands. This could lead to dilution of the mine water, reduced activity of

Table 4 Summary of treatment performance data for the wetlands (±S.D.)

Wetland	SITE B-Wetland 1	SITE B-Wetland 2	SITE B-Wetland 3	SITE C- Wetland	SITE D-Wetland
Wetland short form	B-Wet1	B-Wet2	B-Wet3	C-Wet	D-Wet
Influent Fe^{3+} (mg/L)	2.68±3.23	1.57±2.18	2.43±2.34	2.19±3.68	13.29±19.8
Effluent Fe^{3+} (mg/L)	0.89±2.16	0.52±2.33	0.40±1.34	0.64±2.19	7.75±4.38
Fe^{3+} R.E. (%)	66.74±17.16	66.80±21.52	83.33±15.96	70.85±19.24	41.72±20.77
Number of measurements	103	101	94	71	110
Seasonal Fe^{3+}R.E.(%)					
Summer	74.14±16.29	72.48±17.79	85.86±13.19	73.51±12.97	41.67±23.1
Spring	71.15±13.53	68.93±23.79	84.25±15.41	76.6±15.18	39.62±20.57
Autumn	64.31±18.04	65.08±20.47	82.7±18.61	62.88±25.90	46.62±16.31
Winter	58.49±16.85	61.65±22.91	80.55±16.32	67.71±21.44	39.05±22.72
Cold	61.01±17.04	65.56±20.70	82.81±14.43	70.33±18.70	39.96±20.87
Warm	72.82±15.21	68.06±22.47	83.84±17.44	71.47±20.17	43.49±20.71

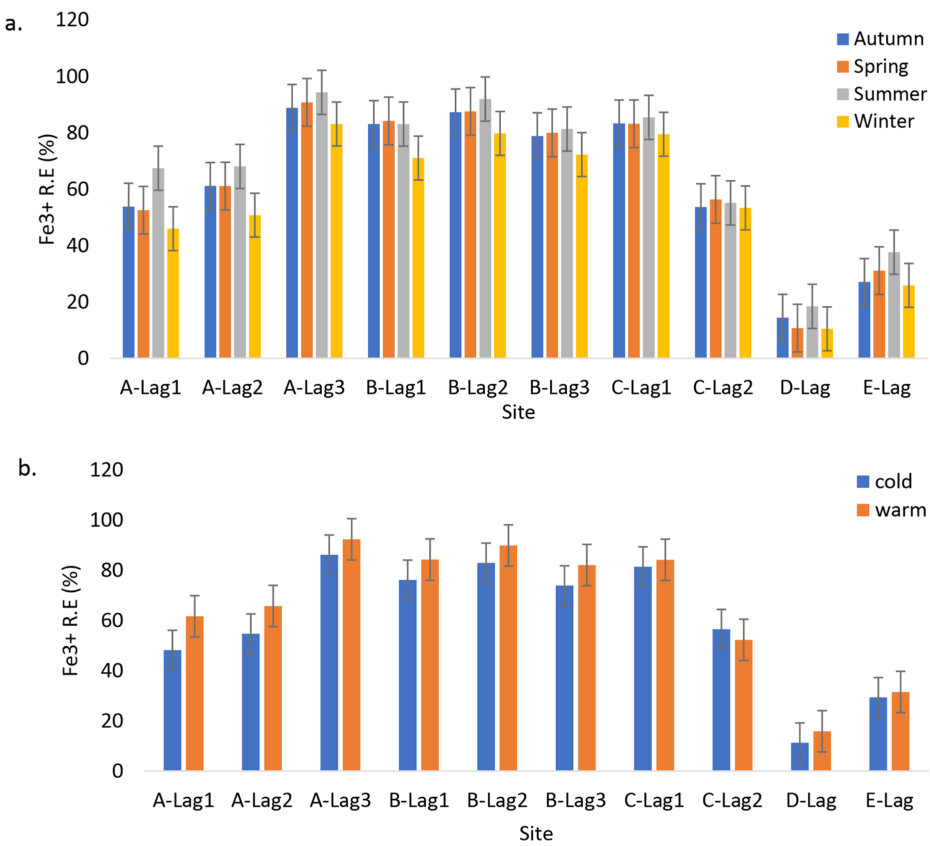


Fig. 7 Seasonal variation of Fe^{3+} removal efficiencies in the studied lagoons: (**a**) autumn, summer, spring, winter; and (**b**) cold, warm seasons ($p < 0.001$)

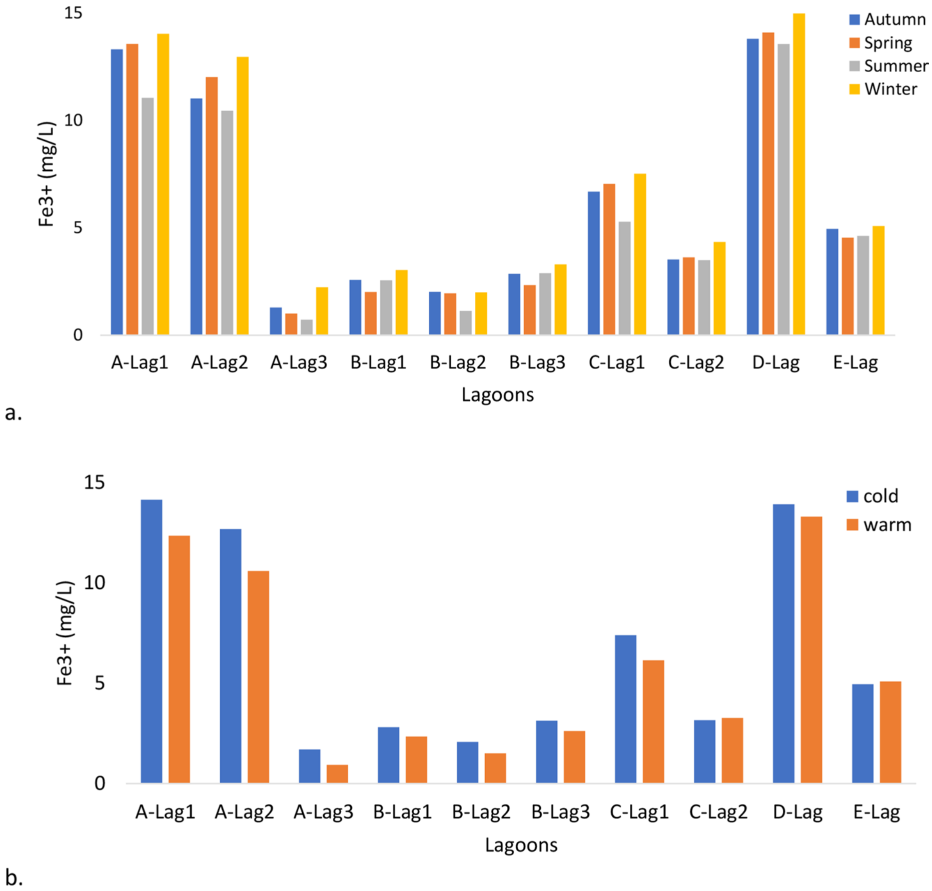


Fig. 8 Fe^{3+} discharge concentrations from the studied lagoons in different seasons: (**a**) autumn, summer, spring, winter; and (**b**) cold, warm seasons

algae and microorganisms breaking down contaminants, reduced rate of chemical reactions, reduced sedimentation and filtration, and ultimately lower removal efficiencies (Fan et al. 2023). However, in summer and warm seasons, fewer rainfalls occur, contributing to higher pollutant concentrations, increased activity of algae and microorganisms breaking down contaminants, increase in rate of chemical reactions, higher sedimentation and filtration, and finally, higher removal efficiencies (Fan et al. 2023; Valkanas and Trun 2018). For instance, Valkanas and Trun (2018) observed that Fe concentration during summer was 50% greater than in winter in the full-scale passive abandoned coal mine drainage remediation system in southwestern Pennsylvania, which consisted of five lagoons and one wetland. In summer, approximately 85% of Fe was removed, while 51% Fe R.E. was achieved in winter (Valkanas and Trun 2018).

3.3.2 Seasonal Changes in Fe Concentrations in Wetlands

With reference to CWs investigated in this study, as a general trend, warm season achieved higher Fe^{3+} R.E. compared to cold season. The wetlands had different orders for summer, winter, spring and summer divisions as presented in Fig. 9a. All three wetlands in Site B achieved Fe R.E. in the order of summer > spring > autumn > winter, while in C-Wet, spring had a higher average Fe^{3+} R.E. than summer (Fig. 9a), removing Fe in the order of spring > summer > winter > autumn.

However, the highest Fe^{3+} R.E. in Site D (D-Wet) was achieved in autumn. The Fe^{3+} R.E. in D-Wet was in the order of autumn > summer > spring > winter (see Fig. 9a). Although research has shown that plants (common reed) in CWs reach the end of their growth cycle at the end of autumn in the UK microclimate, and hence, adsorb fewer pollutants than in spring and summer (Abbasi et al. 2019; Ulén et al. 2019; Heiderscheidt et al. 2020), D-Wet recorded the highest Fe^{3+} R.E. in autumn. This finding is similar to Chen et al. (2020) study where the plants in the pilot-scale passive treatment system treating mine drainage were abundant in summer and autumn, recording the highest Fe^{3+} R.E. in both seasons.

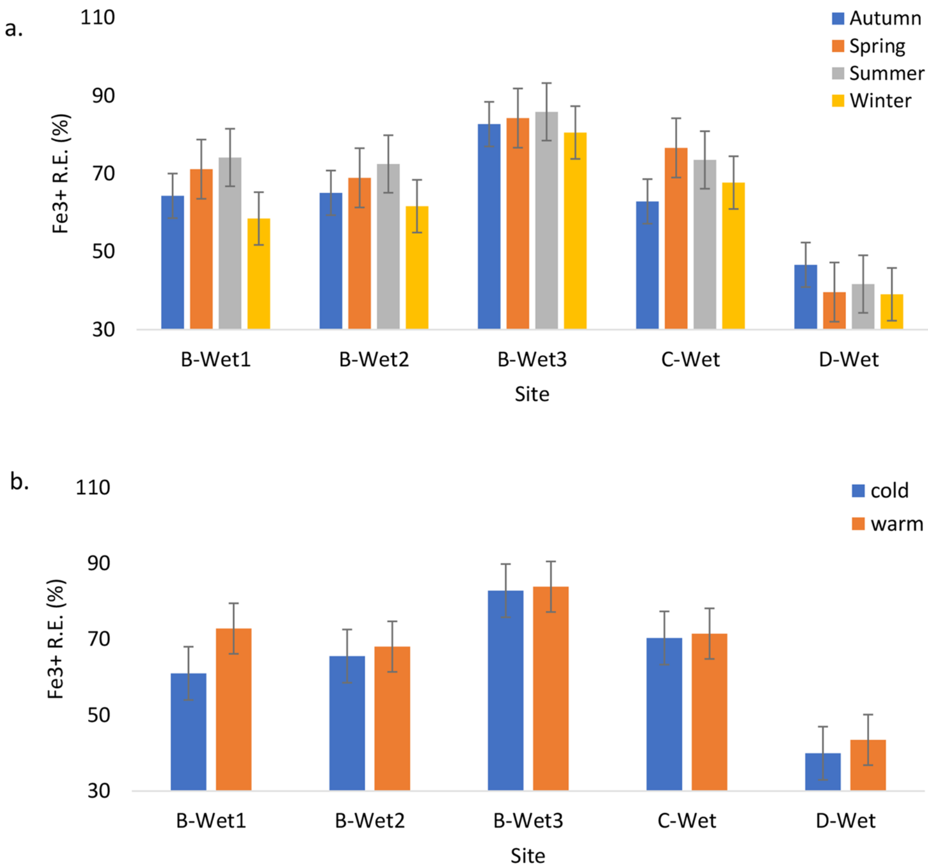


Fig. 9 Seasonal variation of Fe^{3+} removal efficiencies in the studied wetlands: (**a**) autumn, summer, spring, winter; and (**b**) cold, warm seasons

It is possible that the better treatment performance of D-Wet in autumn, and C-Wet in spring, compared to summer, could be attributed to the species of plants in their respective wetlands. Some plant species flourish better in a particular season which makes pollutant concentrations lower, and also the wetland achieve higher removal efficiencies in that particular season (Galal et al. 2017). It is, therefore, recommended for the plant species to also be considered when evaluating seasonal variation of CW treatment performance. In the UK, mine water treatment reedbeds are commonly planted with a mix of common reed (*Phragmites australis*) and bull rush (*Typha latifolia*), which have demonstrated enhancement of Fe^{3+} R.E., particularly across varying UK micro-climates and mine water concentration ranges (Ma et al. 2020; Bamforth and Satterley 2022; Satterley and Moorhouse-parry 2023).

In addition, in wetlands during high temperatures in summer, plant growth would cause the prevalence of anoxic conditions in the top layer of the wetland (Goulet and Roy 2000; Goulet and Pick 2001). Although this reduces the efficiency of the wetland for Fe precipitation, settling becomes prevalent, and since oxygen uptake occurs in plant roots, Fe is also reduced by plant roots uptake (Falagán et al. 2016). Plants also accumulate heavy metals from water and sediments in their shoots; more metals are accumulated in their roots compared to their shoots (Galal et al. 2017). Therefore, the presence of plants in a wetland can be said to encourage Fe reduction and mobilization, contributing to the decrease in Fe concentrations as drainage flows through. However, although wetland plants can maintain their aquatic ecosystem after accumulating heavy metals from water and sediments and temporary immobilizing them within the wetland, when these plants reach the dormant season, there is a tendency for these accumulated metals to return to the wetland (Meyer et al. 2015; Galal et al. 2017; Vymazal et al. 2021).

Furthermore, during autumn and winter, there are low photosynthesis rates taking place, which also reduce the enzymatic reactions of Fe^{3+} precipitates. This results in more dissolved Fe^{2+} oxides in the water column, due to decreased reactions meant to drive precipitation and settling; hence the wetland becomes then a metal source, and low removal efficiencies are recorded (Kadlec and Wallace 2009).

Finally, using Site D as an example, D-Wet was observed (see Fig. 10) to have the highest discharge concentrations among all the wetlands, while D-Lag also recorded the highest levels among all the lagoons (as presented in Fig. 8).

4 Conclusions

This study has investigated for the first time the link between Fe removal efficiency and seasonal variation (i.e., variations in temperature and rainfall patterns) in coal mine drainage employing time-series monthly datasets. The study has analysed the treatment performance of 5 full-scale passive mine water treatment schemes (MWTS) over a period of 12 years. Overall, Fe^{3+} removal efficiency (R.E.) ranged from 50.97 to 98.98% between the various schemes, achieving the order of Site A > Site C > Site B > Site E > Site D. The studied MWTS experienced overall Fe^{3+} R.E. in the order of summer > spring > autumn > winter. In addition, there was an overall increase in pH from slightly acidic to neutral in all the respective schemes studied, with Fe^{3+} and Fe^{2+} concentrations decreasing as pH increased. Results demonstrated that the treatment performance of the MWTS is affected by seasonal variation. Fe^{3+} R.E. for most of the studied lagoons and wetlands of the five MWTS was

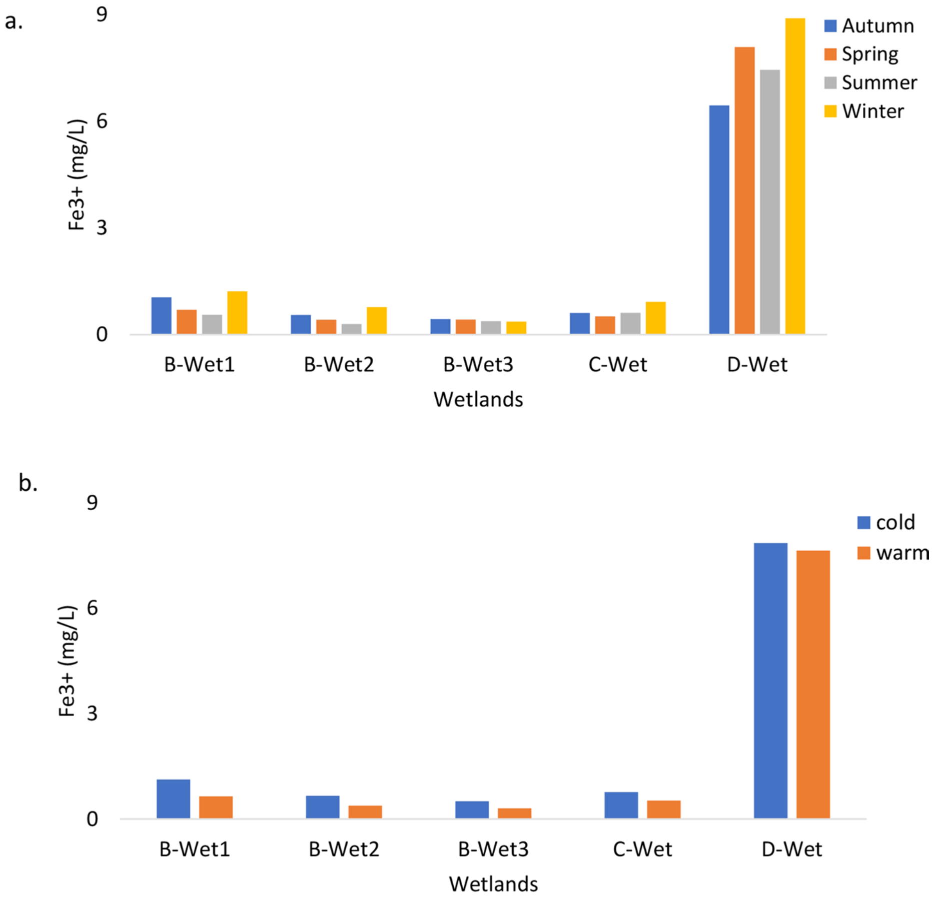


Fig. 10 Fe^{3+} discharge concentrations from the studied wetlands in different seasons: (**a**) autumn, summer, spring, winter; and (**b**) cold, warm seasons

observed to be higher in the warmer months. The higher R.E. identified in wetlands during summer and spring is attributed to the gradual increase in plant growth and volume during the growing season, compared to plant dormant season in winter in the UK microclimate. For the ten lagoons investigated, Fe^{3+} R.E. was higher in warm season with the highest average Fe^{3+} R.E. being recorded in summer, followed by spring, autumn and winter. Increased precipitation in winter, causes dilution of the mine drainage, reducing thereby the rates of sedimentation, adsorption and filtration, and thus, leading to decreased activity of algae and microorganisms breaking down contaminants. In order to maximise Fe^{3+} R.E. and attain optimal and consistent treatment performance of MWTS in different seasons throughout the year, it is recommended that the impact of hydraulic design aspects, e.g., retention time and operational water depth, are factored in the seasonal performance of lagoons and wetlands as part of the MWTS.

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Data Availability No datasets were generated or analysed during the current study.

Declarations

Competing Interests The authors declare no competing interests.

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