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Melamine degradation to bioregenerate granular activated carbon

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ABSTRACT

The industrial chemical melamine is often detected in surface water used for drinking water production, due to its wide application and insufficient removal in conventional wastewater treatment plants. Melamine can be removed from water by adsorption onto granular activated carbon (GAC), nevertheless, GAC needs periodic reactivation in costly and energy intense processes. As an alternative method, GAC can also be regenerated using biomass capable of degrading melamine in a process called bioregeneration. We assessed melamine biodegradation in batch experiments in fully oxic and anoxic, as well as in alternating oxic and anoxic conditions. Additionally, we studied the effect of an additional carbon source on the biodegradation. The most favourable conditions for melamine biodegradation were applied to bioregenerate GAC loaded with melamine. We demonstrate that melamine can be biodegraded in either oxic or anoxic conditions and that melamine degrading biomass can restore at least 28% of the original GAC adsorption capacity. Furthermore, our results indicate that bioregeneration occurs mainly in the largest pore fraction of GAC, impacting adsorption kinetics. Overall, we show that bioregeneration has a large potential for restoring GAC adsorption capacity in industrial wastewater.

1. Introduction

Melamine is an organic micropollutant frequently found in the environment (Ruff et al., 2015; Seitz and Winzenbacher, 2017; Zhu et al., 2019; Zhu and Kannan, 2020). This micropollutant is used as an industrial chemical compound for a wide range of applications, including the synthesis of melamine-formaldehyde resins to produce laminates, plastics and coatings (Smit, 2018). Melamine can be released to the environment both during the production process, as well as by using products containing melamine and its derivatives (Smit, 2018). This micropollutant is poorly removed from water in conventional wastewater treatment plants due to its low biodegradability by and low adsorbability to conventional activated sludge (An et al., 2017; Xu et al., 2013). As a consequence of insufficient removal in wastewater treatment plants (WWTPs) melamine is discharged into surface water. From 2015–2018, melamine was frequently detected in rivers used for the production of drinking water at concentrations above the signalling value of 1 μg/L (RIWA-Maas, 2017, 2016; RIWA-Rijn, 2018, 2016). Given its ubiquitous presence in surface water, a temporary derogation limit was proposed by Dutch authorities, allowing the intake of water to produce drinking water with a maximum concentration of 5 μg/L melamine. The possible contamination of drinking water sources with a micropollutant such as melamine highlights the need for improving current water treatment technologies in order to increase the removal of melamine from wastewater and prevent its presence in drinking water.

Micropollutants, such as melamine, can be removed from (waste) water using adsorption to activated carbon (AC) (Guillossou et al., 2019; Stackelberg et al., 2007). AC is a commonly used adsorbent with affinity for several micropollutants, including melamine (Piai et al., 2019). In adsorption processes with AC, the adsorbed compounds accumulate on the AC surface and the AC adsorption capacity decreases with its usage until the AC needs to be either replaced or reactivated (Worch, 2012). Reactivation of AC used in water treatment is most commonly achieved by applying high temperatures (700–900 °C) which makes it an energy intensive process (Worch, 2012). Biological regeneration is also possible as long as adsorbates can be desorbed and microorganisms capable of degrading them are present and active (Abromaitis et al., 2016; Aktaş and Çeçen, 2006). Biodegradation of micropollutants in the liquid phase can reduce their concentrations to such levels that desorption occurs (Abromaitis et al., 2016). This process continues, consequently regenerating the GAC adsorption capacity, as long as desorption is not hindered and enough biodegradation takes place.

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Microorganisms capable of degrading melamine were first reported by Cook and Huetter (1981). The authors isolated microorganisms from soil, and these microorganisms were capable of degrading melamine by using it as a sole nitrogen source and by using lactate or glucose as carbon source. After that, more authors reported melamine biodegradation using other carbon sources, such as glycerol, methanol and ethanol (Shelton et al., 1997; Takagi et al., 2012). Furthermore, Wang et al., 2014 observed melamine biodegradation without additional carbon sources. Given the low C:N ratio in melamine, an additional carbon source likely increases melamine biodegradation rate by promoting biomass growth.

The biodegradation pathway of melamine was first proposed by Jutzi et al. (1982) and later confirmed by other authors (El-Sayed et al., 2006; Shelton et al., 1997). The biodegradation occurs through consecutive hydrolytic deamination of the ammonia groups attached to the aromatic ring, producing ammeline, ammelide and cyanuric acid, followed by ring cleavage (Fig. 1). Melamine biodegradation results in release of ammonium, which can accumulate in the media (Takagi et al., 2012). However, in the presence of oxygen and nitrifying microorganisms, ammonium can be oxidized to nitrate. The produced nitrate can then be denitrified if denitrifying microorganisms are present and active. The resulting nitrogen removal through nitrification and denitrification creates nitrogen limiting conditions which can promote further melamine degradation, given that this micropollutant is consumed as a nitrogen source.

The combination of biological treatment with granular activated carbon (GAC) offers several advantages compared to biological treatment alone. Higher removal efficiencies of micropollutants with attached growth compared to suspended growth processes have been reported, as a consequence of washout of slow growing organisms in suspended systems (Falås et al., 2013). Furthermore, GAC is known to be an excellent carrier material for biomass since adsorption can remove toxic or inhibitory compounds that might be present in the water (Çeçen and Aktaş, 2012). Additionally, melamine adsorption to GAC can buffer variable influent concentrations, which is beneficial for biodegradation since it avoids peak loads and starvation conditions (Bourneuf et al., 2016; Li and Moe, 2005).

We propose that a combination of adsorption to GAC and biodegradation can be used to treat water containing melamine. Furthermore, melamine degrading biomass can be used to bioregenerate GAC loaded with melamine. To bioregenerate GAC, the optimal conditions for melamine biodegradation were studied. We assessed melamine biodegradation in fully oxic and anoxic conditions, as well as alternated oxic and anoxic conditions, with and without an additional carbon source. The most favourable conditions for melamine biodegradation were applied in a 3-step batch experiment to bioregenerate GAC loaded with melamine, and the bioregeneration extent was assessed. Our results provide additional insights in the optimal conditions for melamine biodegradation, GAC bioregeneration, and the limiting factors for GAC bioregeneration.

2. Materials and methods

2.1. Chemicals

Melamine 99% purity (CAS 108-78-1) was purchased from Sigma Aldrich. Ammeline 90% purity (CAS 108-78-1) was purchased from Alfa Aesar. Ammelide 99,4% purity (CAS 108-78-1) was purchased from Dr. Ehrenstofer GmbH. Cyanuric acid for synthesis (CAS 108-80-5) was purchased from Merck KGaA. Phosphate buffer for the mobile phase for liquid chromatography (LC) was prepared with Na_2HPO_4 (Merck) and $NaH_2PO_4{*}1H_2O$ (Sigma Aldrich).

2.2. Melamine degrading biomass

Melamine degrading biomass in activated sludge was obtained from a WWTP in Romania treating industrial wastewater containing melamine at an average concentration of 58 mg/L. The treatment includes nitrification and denitrification steps and uses methanol as carbon source. After being collected, the sludge was stored up to 2 months before being used. The sludge was collected in two different dates, May 2018 and January 2020. The volatile suspended solids (VSS) in the first and second batch of sludge were 2.45 and 9.24 mg/L respectively. To obtain the desired VSS concentration for each experiment, the sludge was centrifuged and resuspended in media or buffer before use (media and buffer composition are given in Table S1).

2.3. Melamine biodegradation in oxic or anoxic conditions

Melamine biodegradation was tested in oxic and anoxic conditions, with and without methanol, in duplicates. Anoxic conditions are defined in this work as redox conditions that allow for denitrification. Methanol was chosen since this is the carbon source in the WWTP from where the biomass was obtained. Anoxic conditions were achieved in the bottles by flushing the media with nitrogen gas for 10 min and then exchanging the gas phase with nitrogen gas in 5 cycles using a gas exchanger. Oxic conditions were achieved by ensuring sufficient head space and mixing in the batches. Oxic and anoxic conditions were verified by measuring the gas phase of the batches regularly (Table S4). Batches contained 0.5 mL melamine degrading sludge centrifuged and resuspended in nitrogen free mineral salts medium, 38 mg/L melamine (25 mg-N/L) and mineral salts medium (Table S1) to a total liquid volume of 150 mL in 250 mL serum bottles. Methanol, when present, was spiked at 8.5 mg/L. The VSS concentration in the batches was 51 mg/L. The bottles were cultivated at 20 °C with constant mixing at 120 rpm. Melamine and ammeline concentrations were analysed on days 0, 1, 3, 6, 8 and 10. Ammonium, nitrate and nitrite concentrations were analysed on day 6 and 10.

2.4. Effect of alternating redox conditions on melamine biodegradation

The effect of alternating redox conditions and supply of additional carbon source on melamine biodegradation and nitrogen removal was assessed in a batch experiment. Melamine degradation was studied in 5 different conditions, as depicted in Table 1. Anoxic periods were applied

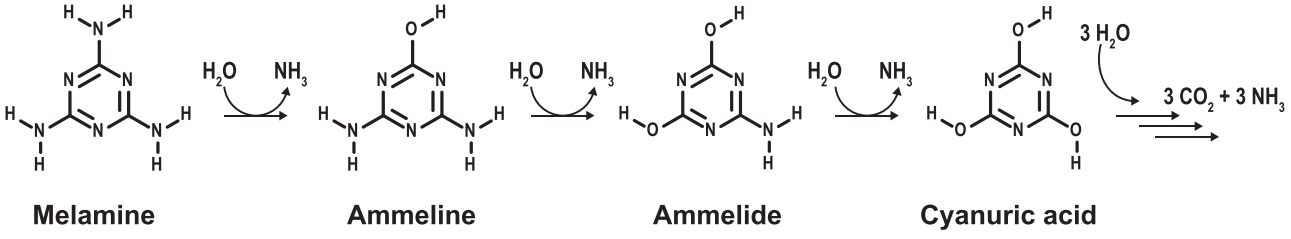

Fig. 1. Melamine biodegradation pathway (Jutzi et al., 1982).

Table 1
Melamine biodegradation test in alternating redox conditions.

		DAY				
		0-3	4-13	14-20	21-30	31-39
CONDITION	1	MeOH	MeOH	MeOH	MeOH	MeOH
	2		MeOH		MeOH	
	3					
	4	MeOH		MeOH		MeOH
	5					

Blue indicates oxic periods and red indicates anoxic periods; MeOH indicates addition of methanol. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

in 3 of the 5 conditions to allow denitrification. Anoxic conditions were achieved as described in 2.3. To restore oxic conditions, the same procedure was done using compressed air instead of nitrogen gas. The inoculum used in this experiment consisted of melamine degrading biomass pre-cultured for 7 days in oxic and for further 6 days in anoxic conditions. Details about the inoculum are described in Section 2.1 of Supplementary Information.

Batches contained 10 mL of inoculum, 36 mg/L melamine (24 mg-N/L) and nitrogen free mineral salts medium (Table S1) to a total liquid volume of 150 mL in 250 mL serum bottles. The VSS concentration in the batches was 54 mg/L. The bottles were cultivated at 20 °C with constant mixing at 120 rpm. Methanol was added to bottles of conditions 1, 2 and 4 at different days as indicated in Table S2. At the start of the oxic periods (days 0, 14 and 31), 50 μL 10% methanol was added to obtain a final methanol concentration of 26 mg/L. In the anoxic periods, different volumes of 1% methanol was added to a final concentration of 10.5–15.8 mg/L. The exact amount of methanol added at each day is given in Table S3. Samples for melamine, ammeline, nitrate and nitrite measurements were taken on days 0, 2, 4, 7, 14, 18, 21, 28, 31, 35, and 39. Ammonium samples were taken and directly analysed on days 0, 4, 14 and 21 in one replicate of each condition.

2.5. Melamine adsorption and desorption isotherm

Adsorption and desorption isotherms for melamine were obtained to: i. assess reversibility of melamine adsorption, a requirement to bioregenerate loaded GAC; ii. determine isotherm coefficients so that melamine load on GAC in different steps of the bioregeneration process could be calculated. Eight different melamine initial concentrations were used, ranging from 8 to 387 mg/L, in a total volume of 50 mL mineral salts medium (Table S1). Relatively high melamine concentrations were used so that the initial concentration of the bioregeneration step would be within the concentration range of the isotherm. A mass of 0.05 g GAC was added to bottles with different melamine initial concentrations and mixed at 120 rpm at 20 °C for 21 days. AcquaSorbTM K-CS (Jacobi®) was used in our experiments, a coconut shell-based activated carbon, that is thermally activated and which has 96% of internal surface area as micropores (Piai et al., 2019). Before being used, the GAC was sieved to obtain particle sizes between 0.5 and 1 mm diameter.

Samples for equilibrium concentration were taken at day 21. A long time interval was used to ensure equilibrium conditions. GAC was filtered from the solution using filter papers. GAC was dried for approximately 1 h and transferred to new bottles for desorption. These bottles contained 47 mL mineral salts medium and 3 mL inactive biomass to a final VSS concentration of 77 mg/L. The media had the same composition as in the adsorption step but without melamine. Inactive biomass consisted of melamine degrading sludge, centrifuged and resuspended in buffer (Table S1), inactivated by autoclaving it twice in consecutive days. Bottles were kept at 20 °C, mixed at 120 rpm and desorption equilibrium samples were taken after 21 days to measure melamine concentration. A control without GAC and with melamine was done to verify that the inoculum was inactivated.

The Freundlich isotherm (Eq. (1)) was used to fit the adsorption data using a non-linear optimization as suggested by Tran et al. (2017), starting from the linearized form of the model (Eq. (2))

$$q_e = K_{f,ads} c_e^{\,n} \quad (1)$$

$$\log q_e = n \log c_e + \log K_{f,ads} \quad (2)$$

where q_e is the melamine load on GAC (mg/g) at equilibrium, $K_{f,ads}$ is the adsorption Freundlich constant $(mg/g)(L/mg)^n$, c_e is the melamine concentration at equilibrium (mg/L), and n is the Freundlich intensity parameter (dimensionless).

Data from the adsorption isotherms were used to determine the coefficients $K_{f,ads}$ and n. The n value obtained for the adsorption isotherm was used as input, together with the desorption data, to determine the desorption Freundlich constant ($K_{f,des}$, in $(mg/g)(L/mg)^n$) similar to the procedure followed by Aschermann et al. (2019). The purpose of this approach is to compare adsorption and desorption affinities based on a single coefficient (K_f).

2.6. Bioregeneration of GAC loaded with melamine

Melamine degrading biomass was used to bioregenerate GAC loaded with melamine in a 3-step batch experiment as follows (Fig. 2):

1) GAC loading: fresh GAC was loaded with melamine;
2) GAC bioregeneration: melamine degrading biomass was added to melamine loaded GAC;
3) GAC reloading (assessment of bioregeneration extent): bioregenerated GAC was reloaded to assess its remaining adsorption capacity and quantify the extent of bioregeneration.

2.6.1. GAC loading

In the GAC loading step, 0.15 g GAC was used to adsorb melamine at an initial concentration of 246 mg/L in 150 mL of mineral salts medium (Table S1). This relatively high melamine concentration was used so that the melamine load on GAC would be close to saturation levels and the effect of bioregeneration would be detectable. Bottles containing GAC, melamine and mineral salts medium were mixed at 120 rpm at 20 °C for 7 days.

Liquid samples (3 mL) were taken approximately at the following time points: 0.5, 1.5, 2.5, 3.5 and 4.5 h, and on days 1, 2, 3, 6 and 7. A total of 4 replicate bottles were prepared and sampled either every hour until 4.5 h or daily until day 7 to avoid subtracting more than 10% of liquid volume by sampling during the adsorption periods.

2.6.2. GAC bioregeneration

After the loading step, GAC was separated from the solution using filter paper, dried for 1 h and transferred to a clean bottle containing 140 mL mineral salts medium (Table S1) spiked with 36 μL methanol and 10 mL melamine degrading biomass centrifuged and resuspended in buffer (Table S1). The VSS concentration in the bottles was 82 mg/L. Pure methanol was added in fractionated doses (6 μL each dose) between days 0 and 6, to avoid methanol toxicity. An abiotic control was taken along, with autoclaved biomass as inoculum. Methanol was added to the control only on the first day and demineralized water was added in the following days to keep the total liquid volume the same in test and control conditions.

After 7 days of incubation under oxic conditions, head space was replaced by 100% N_2 to obtain anoxic conditions and 300 μL 1%

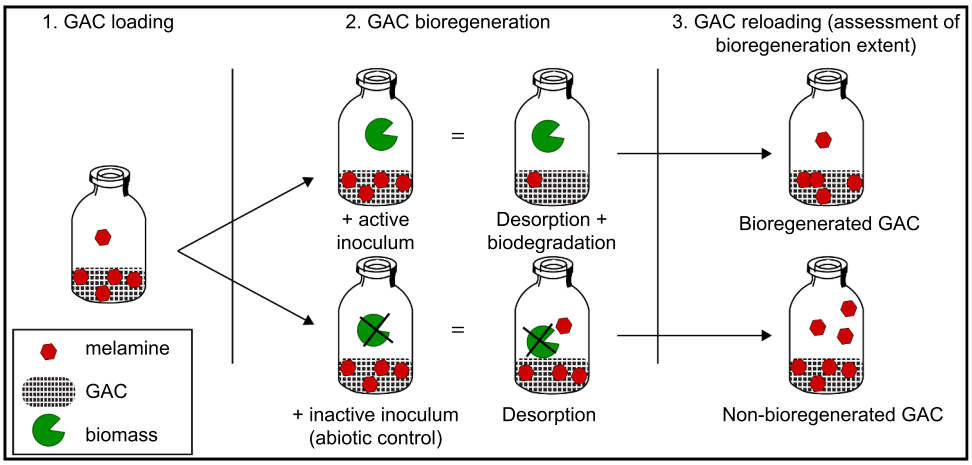

Fig. 2. Schematic representation of steps in GAC bioregeneration experiment.

methanol was added. The bottles remained anoxic for 7 days. After the bioregeneration period (14 days), liquid samples were taken from the biologically active bottle and abiotic control for measuring melamine concentration, followed by autoclaving the bottles and cooling. GAC was separated from the solution using filter paper and added to bottles with fresh medium with melamine to reload the GAC and assess the extent of bioregeneration in the third step of our experiment (2.6.3).

Residual melamine load on GAC (in mg/g) for the bioregenerated GAC ($q_{0,2bio}$) and abiotic control ($q_{0,2abio}$) at the end of this step were calculated based on the melamine concentration in the liquid phase in each treatment ($c_{0,2bio}$ and $c_{0,2abio}$ in mg/L) and by using the desorption isotherm coefficients (see 2.5), according to Eq. 3.

$$q_{0,2(a)bio} = K_{f,des} c_{0,2(a)bio}{}^{n} \quad (3)$$

2.6.3. GAC reloading (assessment of bioregeneration extent)

To assess the extent of bioregeneration, bioregenerated GAC and the abiotic control were reloaded at the same conditions as the GAC loading step, i.e., 150 mL mineral salts medium, 251 mg/L melamine and the full amount of GAC from step 2. Bottles were sampled at the same time points as in the first GAC loading step (2.6.1).

Melamine concentration on day 7 was used to calculate melamine load on GAC which we assumed to be at equilibrium, based on the melamine mass balance, according to Eq. (4).

$$q_{e,i} = \frac{(c_{0,i} - c_{e,i})v}{m} \quad (4)$$

where $q_{e,i}$ (mg/g) is the melamine load on GAC at equilibrium (day 7), $c_{0,i}$ (mg/L) is the initial melamine concentration, $c_{e,i}$ (mg/L) is the melamine concentration at equilibrium (day 7), v (L) is the solution volume and m (g) is the GAC mass. The index i refers either to the GAC loading (*1*) or reloading (*2bio* and *2abio*) steps.

Bioregeneration efficiency (%) was determined by subtracting the adsorption capacity of the abiotic control from the adsorption capacity of the bioregenerated GAC, normalized on the adsorption capacity of the abiotic control, according to Eq. (5):

$$Bioregenerationefficiency \quad (\%) = \frac{q_{e,2bio} - q_{e,2abio}}{q_{e,2abio}} x100 \quad (5)$$

where $q_{e,2bio}$ and $q_{e,2abio}$ are the adsorption capacity of the bioregenerated GAC and the abiotic control on day 7 of the reloading step, based on the mass balance as described in Eq. (4).

Total melamine load on GAC after reloading was calculated for the bioregenerated GAC and the abiotic control, according to Eq. (6).

$$q_{total} = q_{e,2(a)bio} + q_{0,2(a)bio} \quad (6)$$

where q_{total} (mg/g) is the total melamine load on GAC after the reloading step.

2.7. Adsorption kinetics

A pseudo-first-order kinetic equation, as described by Ho and McKay (1998) (Eq. (7)), was used to fit the adsorption data and compare the adsorption rates of fresh GAC, bioregenerated GAC and abiotic control. We used a non-linear optimization as suggested by Tran et al. (2017), starting from the linearized form of the equation (Eq. (8))

$$q_{t,i} = \quad q_{e,i}(1 - e^{-k_1 t}) \quad (7)$$

$$\ln(q_{e,i} - q_{t,i}) = -k_1 t + \ln(q_{e,i}) \quad (8)$$

where $q_{t,i}$ (mg/g) is the amount of melamine adsorbed at time t (h), $q_{e,i}$ (mg/g) is the amount of melamine adsorbed at equilibrium (day 7) and k_1 (1/h) is the adsorption rate constant. The index i refers either to the GAC loading (*1*) or reloading (*2bio* and *2abio*) steps.

2.8. Chemical analysis

All liquid samples were filtered with 0.2 µM polyethersulfone membrane filters and either analysed immediately (for ammonium), within 7 days (nitrate and nitrite) or stored at − 20 °C until analysis (for melamine and ammeline).

Melamine and ammeline concentrations were measured by LC coupled to UV-detection. In the same system, ammelide and cyanuric acid were detected but could not be separated and, therefore, could not be quantified. The LC consisted of a Ultimate 3000 coupled to a 4 channel UV detector (Dionex) and the compounds were detected at 220 nm. First, a method was used in which sample volumes of 50 µL were injected onto a Luna Omega column (150 mm × 4.6 mm, 3 µm) maintained at 40 °C. The compounds were separated using isocratic elution of a mixture of 90% sodium phosphate buffer 100 mM at pH 7.0 and 10% methanol with a flow of 1 mL/min. Retention times were: melamine, 2.3 min; ammeline: 2.0 min. The limit of quantification was 0.05 mg/L for both compounds. Thereafter, the method was changed and sample volumes of 5 µL were injected onto a Luna CN column (250 mm × 4.6 mm, 3 µm) maintained at 35 °C. Compounds were separated using isocratic elution of sodium phosphate buffer 5 mM at pH 6.7 and a flow of 1 mL/min. Retention times were: melamine, 5.6 min; ammeline: 3.8 min. The limit of quantification was 0.05 mg/L for ammeline and 0.09 mg/L for melamine.

Nitrate and nitrite were measured using ion chromatography, as described in (Saha et al., 2020). Ammonium was measured using Hach

Lange colorimetric kits (LCK 303 and LCK 305) and a spectrophotometer (Hach Lange DR 3900).

3. Results and discussion

3.1. Melamine biodegradation in oxic and anoxic conditions

Fig. 3a shows that melamine was biodegraded in both oxic and anoxic conditions and that the biodegradation rate was faster in the latter. Ammeline, the first degradation product of melamine, was also degraded faster in anoxic conditions (Fig. 3b). Ammelide and/or cyanuric acid were still present in oxic bottles on day 10 whereas they were not detected in anoxic bottles on that day (data not shown). Methanol did not increase the melamine biodegradation rate.

The addition of methanol mainly affected the nitrification rate, rather than the melamine biodegradation rate. In the oxic bottles with and without methanol, 36% or more of the nitrogen was still present as ammonium on day 6 (Fig. 4a). The accumulation of ammonium shows that the nitrification rate was slower than the melamine biodegradation rate. In the oxic bottles without methanol, 85% of the nitrogen from melamine was converted to nitrate in 10 days. When methanol was present, less than 45% of nitrogen was converted to nitrate (Fig. 4b), despite all other initial conditions being the same in the bottles with and without methanol. No nitrate was detected in the anoxic conditions, confirming the absence of nitrification.

The negative effect of methanol on nitrification rate is not fully understood. First, methanol did not result in a limiting oxygen concentration, as presented by the oxygen concentrations in our batches. Sufficient oxygen (> 14%) was present in the oxic bottles with and without methanol (Table S4). Furthermore, nitrite was not detected in any of the samples, also indicating that oxygen did not limit nitrification. As nitrite oxidation is affected by low dissolved oxygen levels to a larger extent than ammonium oxidation (Soliman and Eldyasti, 2018), nitrite would have accumulated in the media if oxygen had been limiting in our batches. Second, methanol can bind to the ammonium mono-oxygenase, one of the enzymes involved in nitrification, and hence inhibit it (McBride et al., 2019). Inhibition of nitrification by methanol has been reported in literature (Jönsson et al., 2001; Martin and Richard, 1982; Suzuki et al., 1976; Voysey and Wood, 1987), although most studies report inhibitory effects at levels at least 10 times higher than the initial concentration used in this experiment (8.5 mg/L). We did not expect toxic effects from methanol at this concentration, given that methanol is used as carbon source for denitrification in the WWTP where the sludge originated in doses up to 400 mg/L. Therefore, the observed inhibition of nitrification in the presence of methanol requires further investigation.

Our results show that melamine can be biodegraded in both oxic and anoxic conditions, which is in agreement with previous studies. Most melamine biodegradation studies have been performed in oxic conditions (El-Sayed et al., 2006; Shiomi and Ako, 2012; Takagi et al., 2012; Wang et al., 2014) and the study of Jutzi et al. (1982) is the sole report of melamine biodegradation in anoxic conditions.

3.2. Effect of alternating redox conditions on melamine biodegradation

In a follow-up experiment, we assessed melamine biodegradation in alternating oxic and anoxic conditions. Melamine was biodegraded to a larger extent when oxic and anoxic conditions were alternated, compared to maintaining oxic conditions throughout the incubation period (Fig. 5). Comparing alternating redox conditions, melamine was degraded faster when methanol was supplied in both oxic and anoxic periods (Fig. 5a). When oxic conditions were maintained throughout the experiment, the addition of methanol did not increase the melamine biodegradation rate (Fig. 5b).

Ammonium did not accumulate in the batches, neither in the oxic, nor in the anoxic periods (Table S5), differently from our results when only oxic or only anoxic conditions (see Section 3.1) were applied, and up to 43 mg NH_4-N/L accumulated. In those oxic conditions, melamine degradation rate was 4.9 mg-N/L.d in the first 3 days (3.1, Fig. 3), whereas melamine degradation rate was 1.3 mg-N/L.d in the first 4 days in the experiment with alternating conditions (Fig. 5). This shows a difference in melamine biodegradation rates in the first days in oxic conditions, despite their similar set-up. The lower degradation rate was likely a result of the applied pre-culturing period. As a result, nitrification was the rate limiting step for nitrate production in Section 3.1, resulting in accumulation of ammonium in the medium, whereas melamine degradation was the rate limiting step for nitrate production in this experiment.

Ammonium accumulation was expected in the anoxic periods, when ammonium is produced from the degraded melamine, and nitrification does not occur due to the lack of oxygen. Considering the stoichiometry of melamine degradation and ammeline production in the first anoxic period (days 4–14), 1.0–4.6 mg-N/L of ammonium should be produced. However, this stoichiometry was not reached, as ammonium concentration in all batches in this period remained below 0.05 mg-N/L (Table S5). No nitrite was detected in this period, confirming that partial

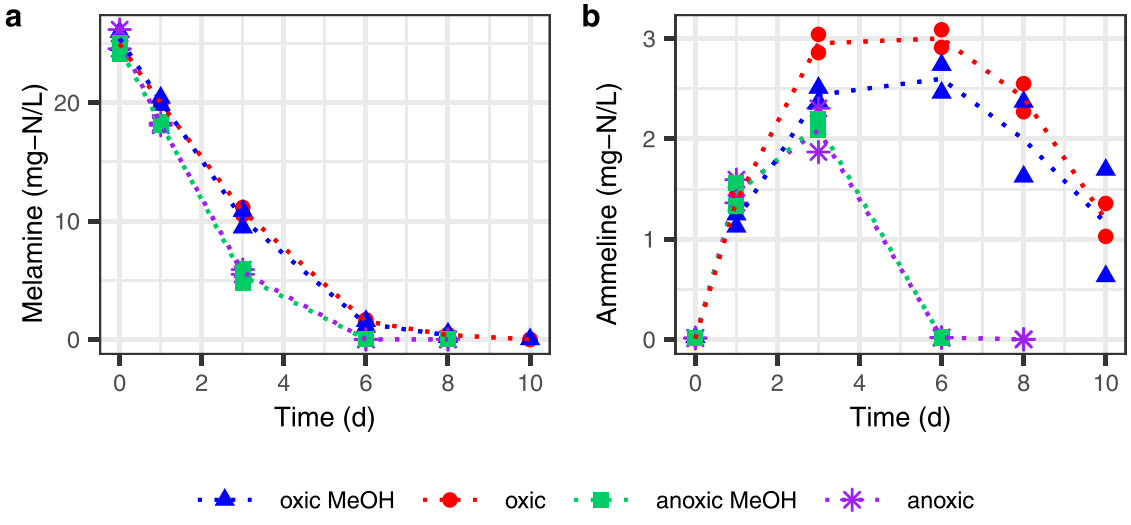

Fig. 3. Melamine (a) and ammeline (b) concentrations in oxic and anoxic conditions, with and without methanol. Duplicates are plotted individually and lines connect the mean value of the duplicates in each time point.

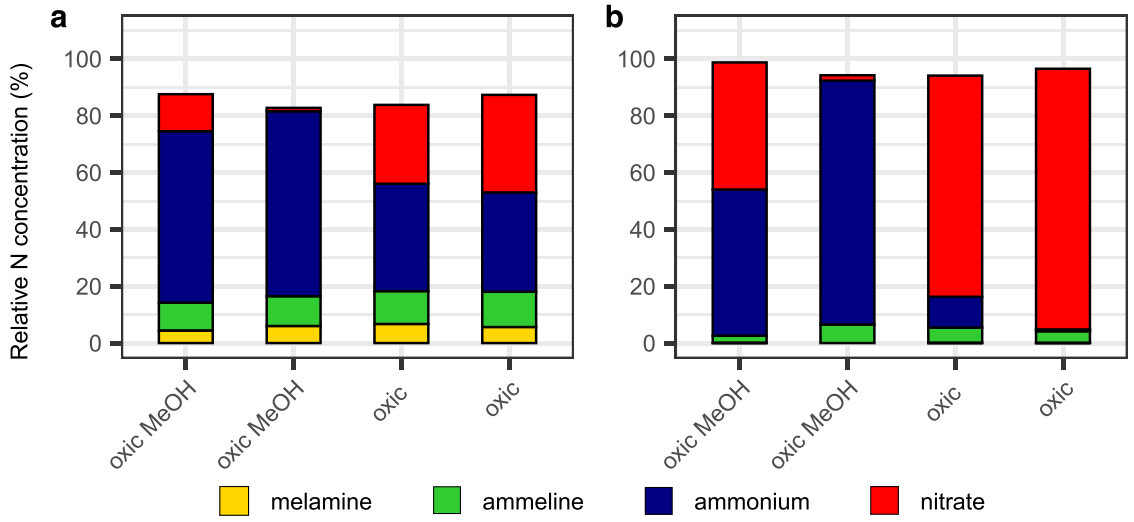

Fig. 4. Both duplicates are shown. a: day 6; b: day 10. Relative nitrogen concentration in melamine, ammeline, nitrate and ammonium in melamine biodegradation test, based on melamine as sole nitrogen source.

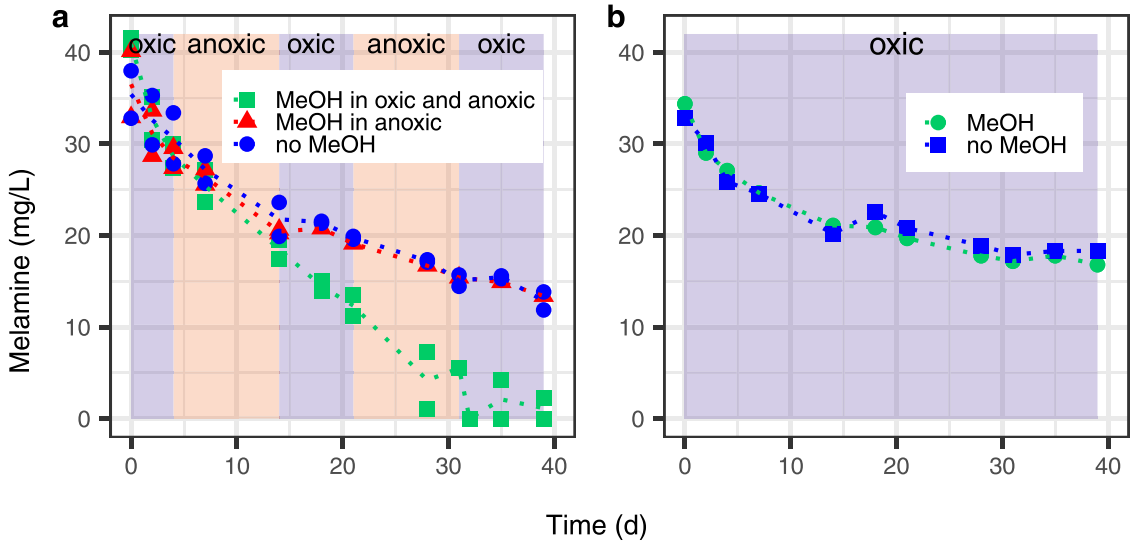

Fig. 5. Melamine concentration in biodegradation experiment. a: alternating oxic (blue background) and anoxic (red background) conditions. b: oxic conditions. For details of experimental setup, see Table 1. In plot a, duplicates are plotted individually and lines connect the mean value of the duplicates in each time point. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

nitrification did not occur. It is unlikely that the low ammonium concentrations measured are the result of artefacts during sampling and sample preparation, given that all samples were treated according to the same procedure in all experiments. A possible explanation for the ammonium fate in the anoxic periods would be the occurrence of anammox. However, this is a speculative hypothesis, given that the operational conditions in the WWTP where the sludge originated from are unfavorable for anammox bacteria (relatively short retention times and low temperatures).

When methanol was supplied in the anoxic period, the nitrate produced in the oxic period was removed by denitrification (Fig. 6a and Fig. S1a). Without methanol in the anoxic period, the nitrate concentration decreased slightly between days 21 and 30 (Fig. 6b). The decrease in nitrate concentration indicates that the carbon present in melamine becomes available when the aromatic ring is hydrolysed, and is used for denitrification. Biodegradable organic carbon in the inoculum was negligible, as confirmed by stable dissolved organic carbon concentrations in independent tests (data not shown). The denitrification rate in that case is much lower compared to denitrification with methanol. Furthermore, methanol had a negative impact on nitrification rates in this experiment, like in our experiments with either only oxic or anoxic conditions (3.1). This is shown by the average nitrate concentration on day 4, which was 20% lower in bottles with methanol, compared to bottles where no methanol was added. Overall, the results indicate that methanol increases melamine biodegradation rate when nitrogen limiting conditions are created, despite the negative influence of methanol on nitrification rates.

Considering our results on the effect of redox conditions and additional carbon source on melamine biodegradation, we conclude that the most favourable conditions for melamine biodegradation are created when nitrogen is removed from the media and methanol is supplied in both oxic and anoxic periods. Therefore, these conditions were applied to bioregenerate GAC loaded with melamine.

3.3. *Bioregeneration of GAC loaded with melamine*

3.3.1. *Reversibility of melamine adsorption*

Melamine adsorption and desorption isotherms were fit with the

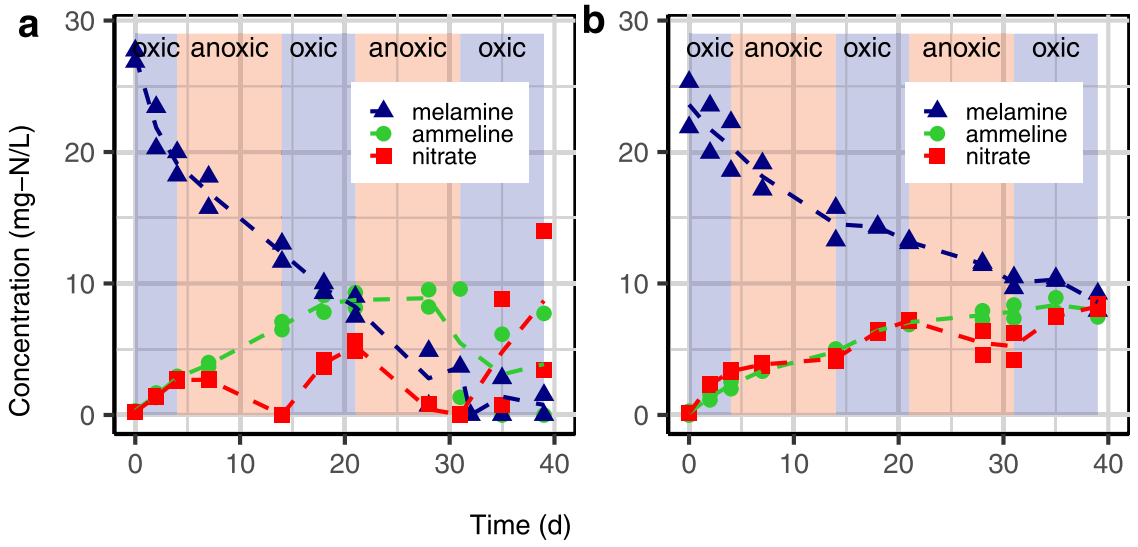

Fig. 6. Melamine, ammeline and nitrate concentrations during melamine biodegradation. a: alternating oxic and anoxic conditions with methanol in both periods. b: alternating oxic and anoxic conditions without methanol. Duplicates are plotted individually and lines connect the mean value of the duplicates in each time point.

Freundlich model to assess reversibility of melamine adsorption (Fig. S2). The calculated Freundlich constant for adsorption ($K_{f,ads}$) and desorption ($K_{f,des}$) were 30.4 and 30.8 (mg/g)(L/mg)^n respectively. The similar values of $K_{f,ads}$ and $K_{f,des}$ indicate that melamine adsorption is a fully reversible process on the conditions tested, i.e., in the presence of inactive biomass.

3.3.2. GAC bioregeneration

Bioregeneration was achieved by mixing loaded GAC with melamine degrading biomass (Fig. 2, step 2). The average melamine concentration at the end of the bioregeneration, i.e. after 14 days, was 0.6 mg/L in the biologically active bottles and 46 mg/L in the abiotic controls. These concentrations were used to calculate the melamine load on GAC at the end of the bioregeneration step ($q_{0.2bio}$ and $q_{0.2abio}$), by applying the desorption isotherm coefficients, according to Eq. 3 (Table 2).

Since melamine concentration was much lower in the biologically active bottles than in the controls, we decided to proceed to the next step, i.e., reloading the GAC, instead of applying a second oxic period.

After bioregeneration, biomass was inactivated and the remaining adsorption capacity of the GAC was assessed reloading the treated GAC. The extent of bioregeneration in this study was assessed by measuring the adsorption capacity of the bioregenerated GAC compared to the abiotic control which was exposed to inactive biomass (see also Fig. 2, step 3). The adsorption capacity of bioregenerated GAC at equilibrium was higher than of the abiotic control but lower than of the fresh GAC (Table 3).

A bioregeneration efficiency of 28% was calculated (Eq. (5)), which is lower than reported bioregeneration efficiencies for GAC loaded with phenolic compounds (Oh et al., 2011), but comparable to efficiencies obtained in batch systems for GAC loaded with surfactants (Klimenko et al., 2003). This relatively low bioregeneration efficiency in our study is partially due to the high adsorption capacity of the abiotic control when reloaded (97 mg/g). The high adsorption capacity of the abiotic control shows that significant regeneration was achieved as a consequence of the experimental design, i.e., simply by transferring the GAC to a solution without melamine (second experimental step).

Since GAC saturation was needed for assessing the extent of bioregeneration, we had to use high melamine concentrations in this study, which are representative of industrial wastewaters. In WWTP effluent and surface water, melamine occurs at much lower concentrations (ng/L to μg/L) (RIWA-Rijn, 2018; Seitz and Winzenbacher, 2017; Zhu and Kannan, 2020). The approach used here is therefore applicable to industrial wastewater when high concentrations of few pollutants are present. In order to apply bioregeneration to other streams, it is important to investigate the feasibility of this process at trace concentrations of micropollutants (ng/L to μg/L). At trace concentrations, micropollutants adsorb to high energy adsorption sites, which could lead to less desorption. However, this will not be the case in the presence of organic matter, where competition for adsorption sites results in higher surface coverage and, hence, to micropollutants adsorption also to low energy sites (Pikaar et al., 2006). Therefore, we do not expect that trace concentrations lead to more irreversible adsorption. Furthermore, at lower concentrations, biodegradation of micropollutants such as melamine can be challenged by mass transfer limitations and limited enzyme affinity (Blair et al., 2015; Joss et al., 2006). However, it is expected that micropollutants accumulation on the GAC surface facilitates their biodegradation at lower concentrations (Aktaş and Çeçen, 2007). Investigating the feasibility of bioregeneration in conditions

Table 2
Melamine concentration and calculated melamine load on GAC at the end of the GAC bioregeneration (step 2[a]) in the bioregeneration experiment (Fig. 2). Results of duplicates are shown individually.

	$c_{0,2}$ (mg/L)	$q_{0,2}$[b] (mg/g)
Biologically active	0.7	27.4
	0.8	28.4
Abiotic control	33.9	121.7
	34.5	122.4

[a] See Fig. 2 for the experimental steps.
[b] Based on Freundlich isotherm (Eq. (3)).

Table 3
Amount of melamine adsorbed by fresh GAC in the loading step and by bioregenerated GAC and abiotic control in the reloading step. Results of duplicates are shown individually.

GAC	Experimental step[a]	$c_{e,i}$ (mg/L)	$q_{e,i}$[b] (mg/g)
Fresh	1 (GAC loading)	78.9	167.1
		80.0	165.9
Bioregenerated	3 (GAC reloading)	124.8	126.3
		130.2	120.9
Abiotic control	3 (GAC reloading)	144.3	106.9
		164.6	86.6

[a] See Fig. 2 for the experimental steps.
[b] Based on mass balance (Eq. (4)).

representative of other water streams will expand the applicability of this process.

3.3.3. Limiting factors for bioregeneration

We compared the total melamine load on GAC after reloading (q_{total}, Eq. (6)), to the expected load based on the melamine concentration in the liquid phase at the end of the reloading step and the adsorption isotherm coefficients. We also compared the melamine load on fresh GAC ($q_{e,1}$) to the expected load based on the adsorption isotherm. Results show that for the fresh GAC and the abiotic control, the total melamine load on GAC matches the expected load based on the adsorption isotherm (Fig. 7). For bioregenerated GAC, the total melamine load on GAC is lower (27%) than the expected load based on the adsorption isotherm, indicating that not all adsorption capacity could be regenerated.

A possible explanation for the unexpected lower adsorption capacity of bioregenerated GAC is related to the adsorption of intermediate products of melamine biodegradation (ammeline, ammelide and cyanuric acid). These compounds were not detected in the end of the bioregeneration step and did not accumulate in any of the melamine biodegradation experiments. However, we may assume that melamine degradation products were present at concentrations below the detection limits of the analytical method, hence a fraction of them was adsorbed. Such a fraction would use adsorption sites in the GAC, thus contributing to the lower availability of adsorption sites for melamine.

Additionally, the reduced adsorption capacity of the bioregenerated GAC can also be related to the actual melamine GAC load at the end of the bioregeneration step ($q_{0.2}$, Table 2). We calculated $q_{0.2}$ based on the adsorption-desorption equilibrium relationship. However, in the presence of biomass, this only holds true when biodegradation is the rate limiting step. If desorption would be the rate limiting step, a higher GAC load is expected for a certain liquid concentration. Considering that the porosity of the GAC used in this study consists mainly of micropores (Piai et al., 2019), we cannot discard the possibility that desorption was the rate limiting step for bioregeneration. In this case, an underestimation of $q_{0.2}$ would explain why the total melamine load on bioregenerated GAC was lower than expected (Fig. 7). Finally, organic molecules produced by the active biomass could have hindered desorption by causing pore blockage (Smolin et al., 2020) and/or competed with melamine for adsorption sites in the GAC reloading step. Likely, all these factors combined contributed to limiting the extent of bioregeneration. Given the limiting factors of bioregeneration discussed above and its high compound-specificity, this method cannot fully replace thermal reactivation. Nevertheless, bioregeneration can contribute to reducing the frequency of thermal reactivation and, therefore, reduce the overall CO_2 footprint of a water treatment plant.

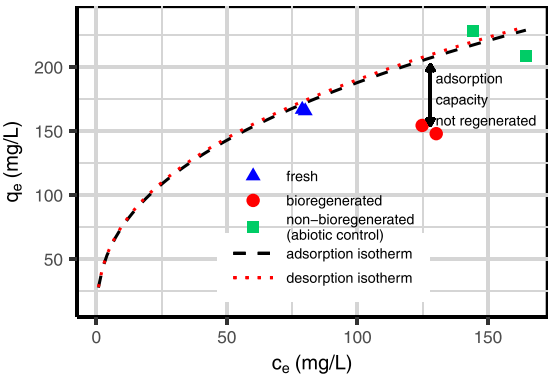

Fig. 7. Melamine load on fresh GAC, bioregenerated GAC and abiotic control compared to the expected load based on the adsorption and desorption isotherms. Duplicates are plotted individually.

3.3.4. Melamine adsorption rate

The adsorption rate of melamine to fresh GAC, to bioregenerated GAC and to the abiotic control was assessed to evaluate the effect of GAC bioregeneration on adsorption kinetics. Similar adsorption kinetics would indicate if adsorption sites in all pore sizes are subject to bioregeneration to the same extent. In the first 2.5 h, bioregenerated GAC adsorbed melamine to a similar extent as fresh GAC (Fig. 8a). After this period, the adsorption capacity of bioregenerated GAC started to diverge from the fresh GAC. With the non-bioregenerated GAC, melamine initially desorbed in the first minutes, and then started adsorbing, which resulted in negative values for q_t in the first 3.5 h. After 7 days the bioregenerated GAC showed an adsorption capacity that was intermediate between fresh GAC and non-bioregenerated GAC (Fig. 8b). The effect of bioregeneration on the adsorption rate of each GAC in the first hours and after 7 days can also be observed by comparing the first-order adsorption rate constants (k_1) of fresh, bioregenerated and non-bioregenerated GAC (Fig. 8). The values of k_1 are similar for bioregenerated and fresh GAC and more than 3 times higher than for non-bioregenerated GAC in the first 4.5 h of adsorption. However, the differences are smaller (less than double) when comparing the adsorption rates of all GACs in a period of 7 days. These results show that melamine biodegradation could restore part of the GAC adsorptive capacity, with a higher impact on adsorption rate than on the total adsorption capacity.

The faster adsorption observed for the bioregenerated GAC compared to non-bioregenerated GAC supports the hypothesis of

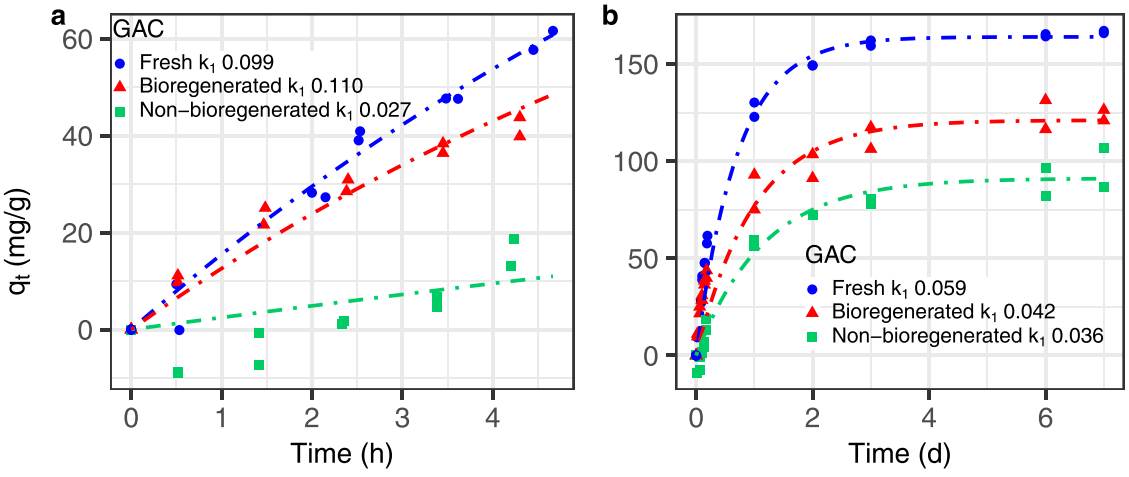

Fig. 8. Melamine adsorption by fresh and bioregenerated GAC abiotic control and respective first-order adsorption constant (k_1 in 1/h) in the period. a: First 4.5 h of adsorption. b: 7 days of adsorption. Data points represent experimental data. Lines represented fitted pseudo-first order kinetics equation (Eq. (7)).

melamine desorption hysteresis due to pore blockage. Pore blockage is likely to happen in the smallest pore fraction of the GAC (Aschermann et al., 2019). This means that melamine only desorbs from adsorption sites in the larger pore fraction and hence those are the pores subject to bioregeneration. Consequently, bioregenerated adsorption sites can be rapidly reached in the second adsorption test, reflecting in faster adsorption rates in bioregenerated GAC compared to non-bioregenerated GAC. These results have also been demonstrated in other modelling and experimental studies. Roy et al. (1999) modelled adsorption kinetics and calculated an intra-particle diffusion coefficient (D_s) for bioregenerated and fresh GAC, obtaining a higher D_s value for bioregenerated GAC. Furthermore, studies showed that the deepest layers of GAC loaded with surfactants or nitrophenol were not bioregenerated and most bioregeneration took only place in the mesopore fraction (Klimenko et al. 2003; Smolin et al. 2020). In our study, we provide additional evidence that bioregeneration happens mainly in the larger and more easily accessible pore fraction of the GAC.

4. Conclusions

Our results show that melamine is biodegraded in oxic and anoxic conditions. In addition, alternating oxic and anoxic conditions promote further melamine biodegradation when an additional carbon source is present, as nitrogen limiting conditions are created and melamine is used as a nitrogen source.

Supply of an additional carbon source and alternating redox conditions were successfully applied to bioregenerate GAC preloaded with melamine. A 3 step-batch experiment was used to allow dividing the processes (GAC loading, bioregeneration and reloading) in different steps and calculate the extent of bioregeneration. In practice, GAC is often used in fixed- or fluidized-bed filters (Worch, 2012), where bioregeneration can be achieved in-situ once biodegradation rates are higher than adsorption rates.

Bioregeneration partly restored the GAC's original adsorption capacity. The effect of bioregeneration is more pronounced in the first hours of adsorption. Bioregenerated GAC adsorbed almost as much melamine as fresh GAC in the first 4.5 h of adsorption, and after this period, the bioregenerated GAC showed less adsorption capacity than the fresh GAC. This indicates that the regenerated adsorption sites are mainly the ones present in the largest and more quickly accessed pore fraction of the GAC.

To conclude, we have shown that the use of biomass capable of degrading micropollutants has the potential to extend the lifetime of GAC, hence reducing the need for thermal GAC reactivation. Knowledge on optimal conditions for biodegradation of micropollutants allows designing bioregeneration processes and avoids GAC thermal reactivation.

CRediT authorship contribution statement

Laura Piai: Conceptualization; Methodology; Formal analysis; Investigation; Writing - Original Draft; Visualization; Project administration. **Albert van der Wa**l: Conceptualization; Resources; Writing - Review & Editing; Supervision; Funding acquisition. **Nadine Boelee**: Conceptualization; Methodology; Resources; Writing - Review & Editing. **Alette Langenhoff**: Conceptualization; Methodology; Resources; Writing - Review & Editing; Supervision; Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2021.125503.

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