Monitoring of Hazardous Substances at Alcantarilha's WTP, Portugal

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Abstract A monitoring programme of hazardous substances was implemented in Alcantarilha's water treatment plant (Algarve, Portugal) since 2002, in addition to the legally established monitoring of standard physical, chemical and microbiological parameters. The objective of this programme was to ensure the drinking water quality regarding the waterborne disease organisms *Cryptosporidium*, *Giardia*, *Salmonella*, *Pseudomonas aeruginosa*, enterovirus and cyanobacteria, and the potentially harmful chemicals aluminium, cyanotoxins, and disinfection by-products (THM) and their precursors (TOC, DOC, UV_{254nm}, SUVA). Most of these parameters are new and still not regulated by the Portuguese and the European legislation. Data presented in this study refer to the period of August 2002 to October 2003. Results show that, despite the seasonal variations of the raw water quality, concentrations of the hazardous substances in the supplied drinking water were far below the legal standards and the WHO's and EPA guideline values, demonstrating the high removal efficiencies of this treatment plant.

Keywords Cyanotoxins, hazardous substances, microorganisms, monitoring programme, THM.

Introduction

Recently, particular attention has been given to the drinking water treatment due to the increasing concern with the protection of public health. New strict regulations for drinking water quality and drinking water sources have been imposed in different countries. These regulations should ensure the safety of drinking water through the elimination, or reduction to a minimum concentration, of the hazardous constituents in water. Thus, a new strict European legislation for drinking water quality was created, Directive n.º 98/83/CE, and subsequently transposed to the Portuguese legislation, DL 243/2001 which entered into force in 25 December 2003, replacing DL 236/1998.

Alcantarilha Water Treatment Plant (WTP), run by Águas do Algarve, SA, a holding of Águas de Portugal, SGPE, SA, is responsible for providing a reliable supply of safe drinking water to ca. half million people in southern Portugal (Algarve), since 2000. This WTP was designed to treat up to 3 m^3/s of surface water from Funcho Dam reservoir (200 km^2 and 43.4 hm^3), which has a history of cyanobacteria occurrence. The WTP conventional treatment sequence is preozonation, coagulation/flocculation/sedimentation (C/F/S), using aluminium polyhydroxichlorosulphate of high basicity and, when necessary, a flocculant, rapid sand filtration and chlorination (Figure 2.1). Alcantarilha WTP has to face a strong seasonal variation in raw water quality together with a seasonal water demand (in 2002, it supplied ca. 180,000 of people during winter and 650,000 people in summer). Continued monitoring showed that seasonal variations correspond to two major types of raw water quality: clear waters (1 – 6 NTU) and turbid waters (25 – 40 NTU) (Ribau Teixeira *et al.*, 2002). Increases in turbidity usually occur after intense rainfall periods and give rise to higher organic carbon contents (Ribau Teixeira *et al.*, 2002 and Ribau Teixeira and Rosa, 2003).

The initial objective of the drinking water quality management was to ensure that Alcantarilha WTP met the current Portuguese and European standards. Since 2002, monitoring of the drinking water supplied by this WTP include parameters of health and environmental concern. Some are new and still not regulated by the national or the European legislation, such as waterborne disease organisms, e.g. *Cryptosporidium*, *Giardia*, *Salmonella*, enterovirus, bacteriophages, *Legionella*, cyanobacteria, and cyanotoxins (microcystins, MCYST). Disinfection by-products, (trihalomethanes, THM) which became regulated in December 2003, are also included in the monitoring programme (WHO, 1993; 1996; 1998; 2002). The monitoring programme for hazardous substances includes those new parameters together with others legally established, namely, aluminium, turbidity, natural organic matter (NOM) and *Pseudomonas aeruginosa* (*P. aeruginosa*). Turbidity was included in the present work as an indicator of water quality, because it is used for water treatment optimisation. In addition high turbidity removal efficiencies correspond also to pathogens partial removal, especially of those pathogens that aggregate with particles (Cohn *et al.*, 2000). NOM was also included since it is a precursor of hazardous disinfection by-products (THM and other organochlorinated compounds).

This paper presents the monitoring programme of 2002/2003 implemented to assess the levels of different contaminants and to establish trends; to identify and track the occurrence of new hazardous chemicals; to assess and optimise the WTP treatment performance and also to provide data to help future developments in drinking water quality standards.

Methods

The monitoring programme refers to the period between August 2002 and October 2003. Samples were collected at different WTP treatment stages (Figure 2.1) and 16 parameters were analysed with different sampling frequencies (Table 2.1).

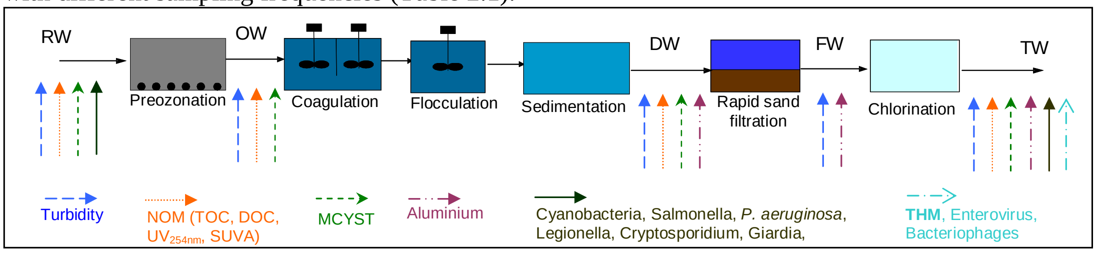


Figure 2.1 Hazardous substances analysed and respective sampling points at Alcantarilha WTP (RW, raw water ; OW, ozonated water; DW, decanted water; FW, filtered water; TW, treated water).

Table 2.1 Sampling frequencies of hazardous substances at Alcantarilha WTP

Parameters (units)	**Sampling Frequency**	**Methods**
Turbidity (NTU)	Twice a day	SMEWW 2130 B (Nephelometric Method)[1]
TOC and DOC (mg C.L^{-1})	Weekly	SMEWW 5310 B (High-Temperature Combustion Method)[1]
UV $_{254\ nm}$ (cm^{-1})	Weekly	SMEWW 5910 B (Ultraviolet Absorption Method)[1]
SUVA (L.(mg-m)$^{-1}$)	Weekly	SUVA is defined as the UV absorbance expressed as meter of absorbance per unit concentration of DOC in mg.L^{-1}
THM (µg.L^{-1})	Monthly	GC- MS (SPME Fiber PDMS 100 µm)
Aluminium (µg.L^{-1})	Twice a day	SMEWW 3500-Al B; SMEWW 3113[1]
MCYST extra[2] (µg. MC_LR eq.L^{-1})	Twice a month	Meriluoto and Spoof, 2003 (High performance liquid
MCYST intra[2] (µg. MC_LR eq.L^{-1})	Twice a month	Chromatography with photodiode array detection)
Cyanobacteria (cell.mL^{-1})	Twice a month	SMEWW 10200 F[1]; Uthermöhl (1958)
Enterovirus (No.100 L^{-1})	Annual	XP T90-451
Bacteriophages (No.50 mL^{-1})	Annual	ISO 10705-2:2000; ISO 10705-1:1995 ISO 10705-4:2000
Legionella (cfu.2 L^{-1})	Annual	ISO 11731
Cryptosporidium (P/A)	Annual	EPA 1623
Giardia (P/A)	Annual	EPA 1623
Salmonella (P/A. 2L^{-1})	Monthly	ISO 6340:1995 (E)
P. aeruginosa (cfu.100 mL^{-1})	Weekly	ISO 8360-2: 1988; NF T 90-421:1989

[1] SMEWW – Standard Methods (Clesceri *et al.*, 1998); [2] Analysed after February 2003; P/A- Presence/Absence.

Results and Discussion

Data obtained from the monitoring programme for hazardous substances at Alcantarilha's WTP, during the sampling period of August 2002 to October 2003, in raw (RW), ozonated (OW), decanted (DW), filtered (FW) and treated waters (TW), are presented in Figures 3.1 to 3.10 and Tables 3.1 and 3.2.

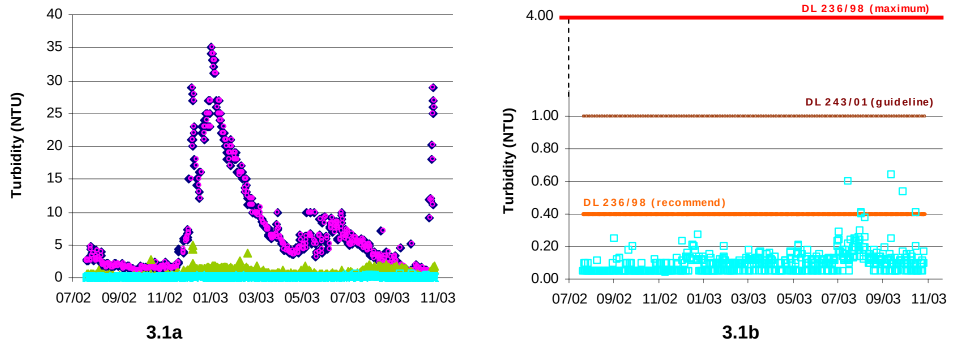


Figure 3.1 Turbidity values between Aug. 02 - Oct. 03: **a)** in RW (♦), OW (○), DW (▲) and TW (□); **b)** in TW and national standards for drinking water.

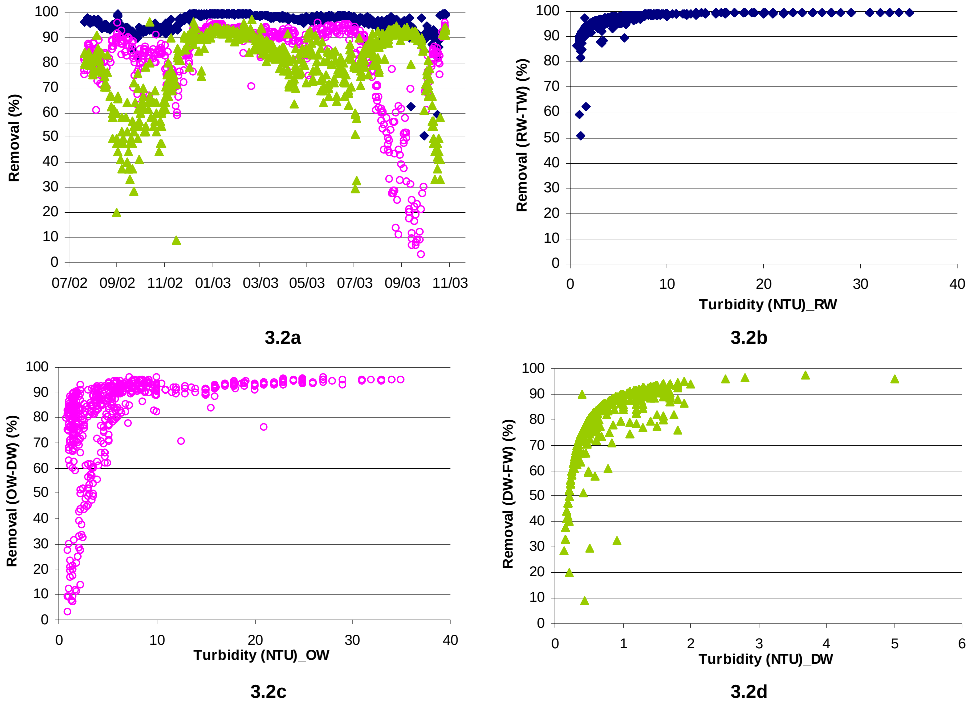
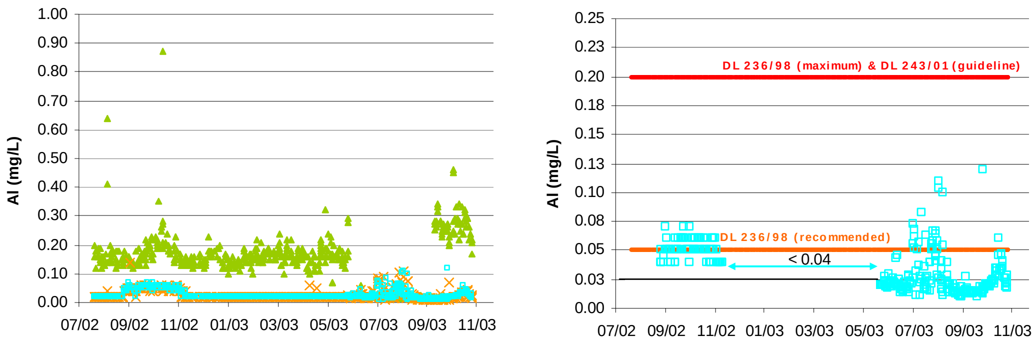


Figure 3.2 Turbidity removal: **a)** values between Aug.02 – Oct. 03, RW-TW (♦), OW-DW (○), DW-FW (▲); relationship with influent turbidity for: **b)** RW-TW; **c)** OW-DW; **d)** DW-FW

Turbidity values show a marked seasonal variation in both raw (RW) and ozonated water (OW) (Figure 3.1a). It is possible to distinguish two different periods related with wet and dry weather conditions. From August to November 2002 (dry months) turbidity was low, with an average of 2.10 ± 0.24 NTU. In December 2002, rainfall became more frequent and intensified the runoff to Funcho Dam reservoir, increasing turbidity significantly (up to 35 NTU registered in January 2003). After the intense rainfall period, RW turbidity decreased to a minimum of 3.10 NTU, in May 2003, as the result of particles deposition in the dam reservoir. A second period of turbidity increase (smaller than the winter time peak), was observed in June-July 2003. This rise was probably related to particles ressuspension in the WTP affluent main, due to the increase in flow rates to fulfil the water demand during the high season. In the period of August 2003 to the end of September 2003, clear raw waters, with an average of 3.71 ± 0.86 NTU, were registered. In contrast, another high peak (up to 30 NTU) was observed in October 2003, related to intense rainfall.

Turbidity after preozonation, the first WTP stage, show similar values to RW since no particle removal takes place in this stage.

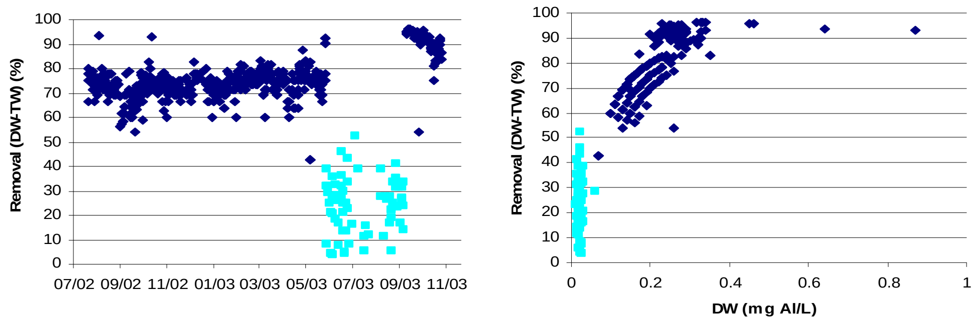
Despite the large turbidity fluctuations and the high influent values in wet months , turbidity of the TW presented very low and fairly constant values (0.12 ± 0.05 NTU, Table 3.1) demonstrating the high performance of Alcantarilha's WTP for turbidity removal. These values were far below the national standards for drinking water (4 NTU during the sampling period -DL 236/1998-, reduced to 1 NTU after 25 December 2003 -DL 243/2001). In fact, 99 % of the samples showed values below 0.4 NTU, the strictest standard (turbidity recommended value in DL 236/1998) (Figure 3.1b). As expected, C/F/S and filtration were mainly responsible for turbidity removal (Figures 3.1 and 3.2a). Figures 3.2.b, 3.2c and 3.2d show a turbidity removal increase with rising influent turbidity. The 99% plateau was reached for influent turbidity values above 9 NTU. Average turbidity removals for two classes of RW turbidity (< 5 NTU and 5-35 NTU range) are presented in Table 3.1. Considering that these waters present a low to moderate alkalinity (50-75 mg/L as $CaCO_3$), turbidity increases improve the colloidal matter removal by adsorption and charge neutralization mechanisms using low coagulant doses. In the case of low turbidity, high removals are achieved with the utilisation of a pre-polymerised aluminium coagulant of high basicity aided by a flocculant.



3.3a

3.3b

Figure 3.3 Aluminium values between Aug. 02 - Oct. 03: **a)** in DW (▲), FW (x) and TW (□); **b)** aluminium in TW and national standards for drinking water.

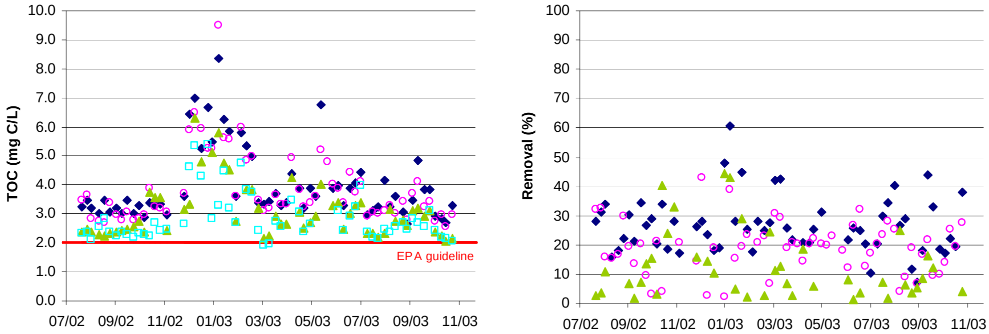


3.4a

3.4b

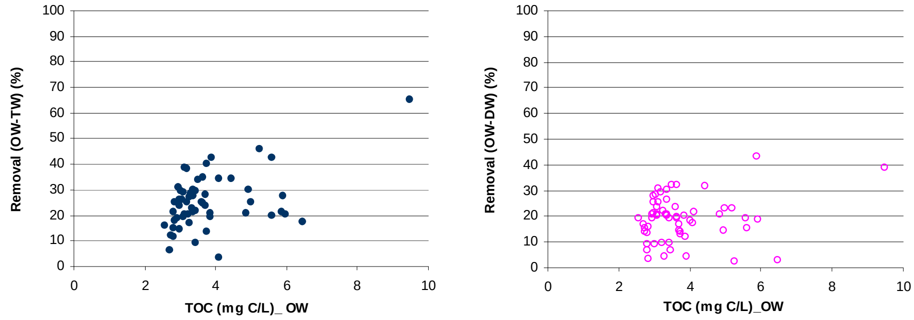
Figure 3.4 Aluminium removal: **a)** variation between Aug.02 – Oct. 03, DW-TW; **b)** relationship with Al concentration in DW (total Al (♦), dissolved Al (■)).

The main aluminium fraction present in water samples is residual aluminium from the coagulant addition. Taking into account that aluminium is mainly removed by sedimentation, the monitoring programme included sampling points for residual aluminium in DW, FW and TW. Figure 3.3a illustrates the important aluminium removal by filtration, allowing the compliance with the national standard of 0.2 mg Al/L (DL 236/1998 & DL 243/2001) and with the recommended value of 0.05 mg Al/L (DL 236/1998) in 92% of the samples (Figure 3.3b). Similarly to turbidity, aluminium removal (total Al, Figure 3.4b) increased asymptotically from 52% to 98% with increasing influent concentration, achieving a constant removal for influent concentrations above 0.4 mg Al/L. This guarantees the low DW aluminium residual even when higher doses of coagulant are used, as in the case of turbid RW.



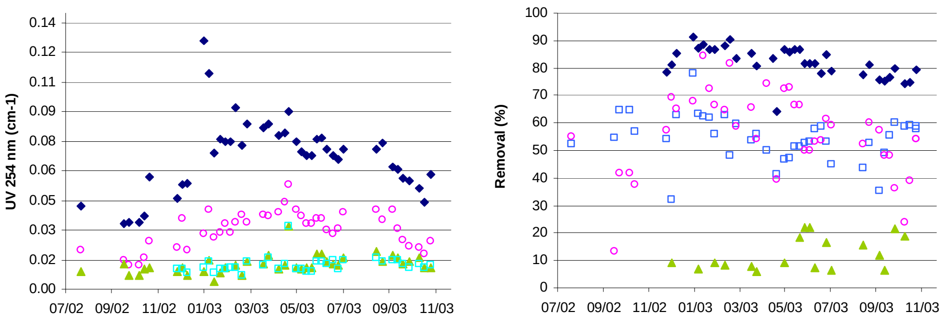
3.5a 3.5b

Figure 3.5 TOC values between Aug.02 – Oct. 03: **a)** in RW (♦), OW (○), DW (▲) and TW (□) and EPA (1999) guideline for THMFP control purposes; **b)** removals in RW-TW (♦), OW-DW (○), DW-TW (▲).



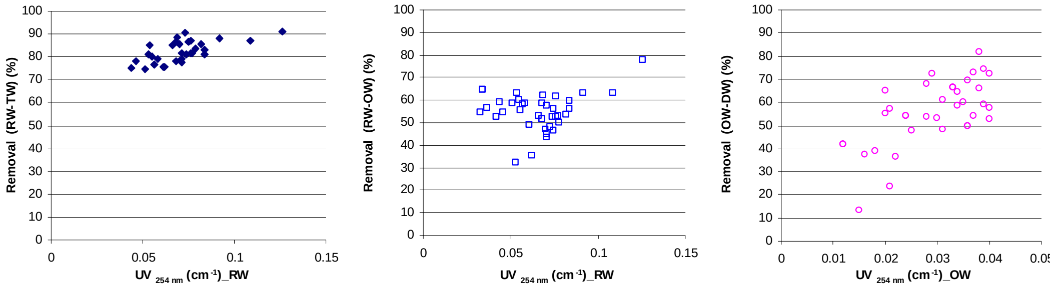
3.6a 3.6b

Figure 3.6 TOC removal *vs*. influent concentration: **a)** OW-TW (●); **b)** OW-DW (○).



3.7a 3.7b

Figure 3.7 $UV_{254\ nm}$ values between Aug.02 – Oct. 03: **a)** in RW (♦), OW (○), DW (▲) and TW (□); **b)** $UV_{254\ nm}$ removals in RW-TW (♦), RW-OW (□), OW-DW (○), DW-TW (▲).

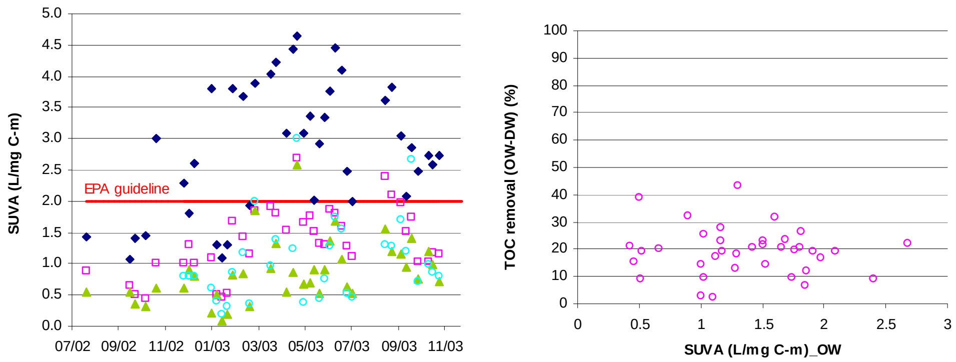


3.8a 3.8b 3.8c

Figure 3.8 UV $_{254\text{ nm}}$ removal vs. influent UV $_{254\text{ nm}}$: **a)** RW-TW (♦); **b)** RW-OW (□), **c)** OW-DW (○).

Figure 3.5a shows a seasonal variation for TOC content in RW, similar to the turbidity pattern (Figure 3.1a), which proves turbidity is a good and easy-to-assess indicator of water quality. RW TOC concentration increased in wet months, when rainfall became more frequent and runoff rose (yielding lower DOC/TOC ratios, Table 3.1). TOC removal efficiencies did not significantly increase with the influent concentration, and thus lower TW quality was observed during peak season (Figure 3.6). Table 3.1 presents the average TOC removals for two RW TOC classes, 2.0-4.0 mg C/L and 4.0-8.0 mg C/L, for which EPA indicates the need for removal of TOC by enhanced coagulation in plants using conventional treatment. In Alcantarilha's WTP, besides the hydrophilicity and the low molecular weight of NOM (SUVA below 4 L/(mg C-m)) registered in OW (influent to C/F/S stages), the high basicity of the pre-polimerised Al coagulant used did not allow the pH decrease required for enhanced coagulation to occur.

The relation between TOC removal and the influent concentration is established for C/F/S and filtration, where most of TOC removal is expected to occur (Figure 3.6). In Alcantarilha's WTP, the supernatant from sludge circular decanter and water from filters washing, return to ozonation effluent, often yielding higher TOC concentrations in OW than in RW. Results indicate that the increase in OW TOC does not increase the global neither the C/F/S removal efficiencies (Figure 3.6), once NOM removal depends greatly on the nature of dissolved organic matter. In this case, low SUVA values make the TOC removal more difficult (Figure 3.9b). In fact, ozonation oxidizes UV_{254nm} absorbing substances, further decreasing the already low SUVA values present in natural raw water (Table 3.1). OW SUVA was lower than 3 L/(mg C-m) (Figure 3.9a), which, according to Edzwald & Van Benschoten (1990), corresponds to hydrophilic NOM of low molecular weight, responsible for colloidal suspensions hard to destabilize. UV $_{254nm}$ absorbing substances were easier to remove by C/F/S than TOC and their removal by this process increased with the influent concentration. This relationship was not observed for the ozonation process (RW-OW) (Figures 3.5 and 3.6 vs. Figures 3.7 and 3.8).



3.9a 3.9b

Figure 3.9 SUVA: **a)** values between Aug.02 – Oct. 03 in RW (♦), OW (○), DW (▲) and TW (□), and EPA (1999) guideline for THMFP control purposes; **b)** relationship with DOC removal by C/F/S.

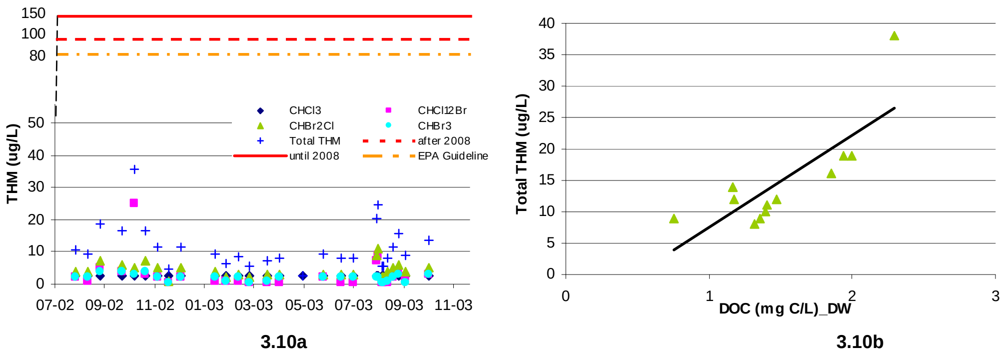


Figure 3.10 Total THM: **a)** variation between Aug.02 – Oct. 03 in TW, national standards and EPA (1999) guideline; **b)** relationship with DOC concentration in DW.

According to EPA guidelines for disinfection by-products formation (EPA, 1999), the low SUVA values obtained in DW (below 2 L/(mg C-m), Figure 3.9a) indicate a low THM formation potential (THMFP), since hydrophobic DOC has higher potential to form THM than hydrophilic DOC (Galapate *et al.*, 2000). In fact, total THM values in TW (14.2 ± 2.5 µg/L) were always far below the national and European standard (150 µg/L) as well as the stringent EPA (2003) guideline value (Figure 3.10a). Despite the low potential formation it was possible to establish a relationship between DOC concentration in DW and the total THM in TW, as shown in Figure 3.10b.

Alcantarilha's WTP treatment performance is summarised in Table 3.1.

Table 3.1 Relationship of global removal with concentration ranges in RW, and TW averages achieved by Alcantarilha's WTP (Aug. 02 – Oct.03) (Confidence interval was calculated with α = 0.05).

Parameters	RW Range	RW Average	TW Average	Removal Efficiency (%)	Remarks
Turbidity (NTU)	< 5	2.31 ± 0.13	0.12 ± 0.05	93.7 ± 0.5	Range: 51 – 98%
	5 - 35	--		98.6 ± 0.1	Range: 95 – 99.7%
Aluminium (mg/L)	--	--	0.038 ± 0.001	--	--
TOC (mg C/L)	2.0 – 4.0	3.34 ± 0.09	2.83 ± 0.21	25.8 ± 2.3	(1) 0 – 60 mg CaCO3/L: 35% 60 – 120 mg CaCO3/L: 25%
	4.0 – 8.0	--	--	29.9 ± 6.7	(1) 0 – 60 mg CaCO3/L: 45% 60 – 120 mg CaCO3/L: 35%
DOC/TOC*	2.0 – 4.0	0.61 ± 0.05	0.44 ± 0.07	--	--
	4.0 – 8.0	0.57 ± 0.11			--
$UV_{254\,nm}$ (cm^{-1})	--	0.07 ± 0.01	0.012 ± 0.006	82.0 ± 1.8	--
SUVA (L/mg C-m))	≤ 3	2.0 ± 0.3	1.0 ± 0.2	--	--
	> 3	3.8 ± 0.2			--
Total THM (µg/L)	--	--	14.2 ± 2.5	--	--

(1) EPA (1999); * calculated for TOC ranges

Table 3.2 shows microcystins and cyanobacteria data obtained during this study, including all the cyanobacterial genera identified and not only the potential producers of toxins. Cyanobacteria are present in the source water throughout most of the year (Rosa *et al.*, 2004), therefore reaching Alcantarilha's WTP (Table 3.2). Nevertheless, dissolved MCYST was never detected in RW (not even during the superficial bloom occurrence in Funcho dam reservoir in March 2003), and intra-cellular MCYST was quantified only once and in very low concentration (October 2003). Cyanobacteria were always absent in TW and MCYST were systematically below the quantification limit of 0.014 µg/L, much lower than the 1 µg MC-LR/L guideline value suggested by WHO (1998) for drinking water.

In March 2003, during a superficial bloom occurrence in source water, cyanobacteria were not detected in RW, indicating an adequate water depth abstraction at Funcho Dam reservoir.

Ocasionally, the monitoring programme implemented at Funcho Dam reservoir identified the presence of other cyanobacterial genera, which are potential producers of different toxins. Genera included *Aphanizomenon* and *Phlanktotrix* that may produce a potent neurotoxin, anatoxin-a, included in the monitoring programmes implemented for Funcho Dam reservoir and Alcantarilha's WTP since April 2004.

Table 3.2 Average values in raw and treated water for microcystins, cyanobacteria and other microorganisms.

Parameters		2002					2003										Standards
		Aug.	Sep.	Oct.	Nov.	Dec.	Jan.	Feb .	Mar.	Apr.	May	Jun.	July	Aug.	Sep.	Oct.	
MCYST_extra (μgMC-LR eq.L^{-1})	RW	-	-	-	-	-	-	-	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	1 [1]
	TW	-	-	-	-	-	-	-	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	
MCYST_intra (μgMC-LR eq.L^{-1})	RW	-	-	-	-	-	-	-	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	0.11	1 [1]
	TW	-	-	-	-	-	-	-	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	<0.014	
Cyanobacteria (cell·mL^{-1})	RW	-	243	3495	1902	202	9	1	0	1	4	4	0	18	151	414	-
	TW	-	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
Salmonella (P/A·2L^{-1})	RW	A/P	A	-	A	P	A	A	P	P	P	P	P	P	A	P	-
	TW	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	
P. aeruginosa (cfu·100 mL^{-1})	RW	0	0	0	0	3	15	1	1	1	0	0	0	0	0	20	0 [2]
	TW	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	

[1] WHO's guideline value; [2] DL 243/2001; P/A - Presence/Absence

As shown in Table 3.2, the pathogens *Salmonella* and *P. aeruginosa* were always removed in the treatment plant.

In the beginning of the high season (June 2003), when temperature increased together with the drinking water demand, *Cryptosporidium*, *Giardia*, and *Legionella* were analysed in both raw and treated water, and enterovirus and bacteriophages in the treated water. None of them were detected in both waters (RW and TW). Therefore, the absence of microcystins and the referred microorganisms in RW did not allow the checking of the WTP ability to remove these hazardous substances. In addition to the referred assessment through this monitoring programme, WTP management includes a 3 level strategy in terms of their removal to ensure a safe water supply:

1. Optimisation of WTP unit operations for removal of toxins and/or microorganisms – identification of the limiting steps (e.g. the recirculation of sludge treatment streams) and operating conditions (particularly, the type and dosage of oxidants, coagulant, flocculant and adsorbent, in this case, powdered activated carbon (PAC)). These procedures are being implemented in the WTP operation manual to guarantee their application whenever an episode occurs.
2. Development and optimisation of new technologies for WTP upgrade in case the monitoring programme indicates limited results from the strategy above. In fact, one can expect limited performance of the conventional treatment with preozonation and PAC adsorption if the occurrence of these hazardous substances becomes frequent. In this case, technologies, such as dissolved air flotation (for cyanobacteria removal), ultrafiltration (for particles removal, including bacteria and bacterial cysts, and virus (Ribau Teixeira *et al.*, 2004)), PAC/ ultrafiltration, nanofiltration and activated carbon filters with and without biological activity (BAC and GAC, respectively) (for further removal of organics of low molecular weight including toxins and THM precursors), would become very attractive.
3. Development of a contingency plan for management procedure whenever the monitoring programme indicates the treated water is not safe for human supply. This plan includes instructions to interrupt Alcantarilha's WTP production and to manage an alternative supply water system using water produced in other water treatment plants, which use different surface water and/or groundwater sources.

Conclusions

Results of this study emphasise the importance of hazardous substances monitoring in WTP, particularly when a conventional treatment is used. The monitoring programme implemented in Alcantarilha's WTP showed (i) a very good treatment performance of particulate matter and a reasonable performance for dissolved organics, despite the seasonal variations of the raw water quality, and (ii) the high quality standards of the supplied water (concentrations of the hazardous substances in the treated water are much lower than legal standards and the WHO's and EPA guideline values). Turbidity is a key parameter for raw water quality assessment and conventional treatment optimisation.

Futhermore, the monitoring programme was used to assess the levels and the normal variation of the different contaminants and to establish trends; to identify and track the occurrence of new hazardous chemicals; to assess and optimise the WTP treatment performance and to provide data that will help future developments in drinking water quality standards.

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