The effectiveness of conventional trickling filter treatment plants at

reducing concentrations of copper in wastewaters.

Dominik Ziolko^a, David Hala^a, John N. Lester^b and Mark D. Scrimshaw^{a,*}

^a Institute for the Environment, Brunel University, Uxbridge, Middlesex, UB83 PH, UK ^b School of Applied Science, Centre for Water Science, Cranfield University, Bedfordshire, MK43 0AL, UK

^{*} Corresponding author. Tel.: +44 (0)1895 267 299. E-mail: Mark.Scrimshaw@brunel.ac.uk.

Abstract

Eight different sewage treatment works were sampled in the North West of England. The effectiveness of the conventional treatment processes (primary sedimentation and biological trickling filters) as well as various tertiary treatment units in terms of both total and dissolved copper removal was evaluated. The removal of total copper across primary sedimentation averaged 53% and were relatively consistent at all sites, however, at three sites the removal of dissolved copper also occurred at this stage of treatment. Removal of total copper by the biological trickling filters averaged 49%, however, substantial dissolution of copper occurred at two sites, which highlighted the unpredictability of this treatment process in the removal of dissolved copper. Copper removal during tertiary treatment varied considerable even for the same treatment processes installed at different sites, primarily due to the variability of insoluble copper removal, with little effect on copper in the dissolved form being observed. The proportion of dissolved copper increased significantly during treatment, from an average of 22% in crude sewages to 55% in the final effluents. There may be the potential to optimise existing, conventional treatment processes (primary or biological treatment) to enhance dissolved copper removal, possibly reducing the requirement for installing any tertiary processes specifically for the removal of copper.

Keywords: copper; sewage; removal efficiency; mass balance; primary treatment; trickling filters

1. Introduction

Copper is one of the metals that are of particular concern in relation to water quality, because although an essential element, elevated concentrations in the aquatic environment may result in toxicological effects (de Oliveira-Filho et al., 2004; de Schamphelaere et al., 2004). Copper has been classified under the Water Framework Directive as a potential main pollutant (Annex VIII) for which an environmental quality standard (EQS), if required, should be established at the national level (European Commission, 2000). At present, in England and Wales for water bodies achieving good ecological status, the freshwater EQS for copper ranges from 1-28 μ g/l (dissolved copper annual average) depending on water hardness, falling to 5 μ g/l in coastal and estuarine areas (UKTAG, 2008). Discharge consents for sewage treatment works (STW) however, are set based on the total copper concentrations (particulate plus dissolved copper). Removal of both particulate and dissolved copper fractions during sewage treatment is therefore relevant to achieve compliance with increasingly stringent consents and EQS and achieving this with existing unit treatment processes may be challenging (Ziolko et al., in press).

In developed countries concentrations of copper in sewage have generally decreased in comparison to the late 1970s (Ziolko et al., in press). However, contemporary concentrations remain significant when compared with EQS and consents, especially for receiving waters that are soft in nature and offer limited natural dilution for final effluents. Thus STW, although not designed to do so, are expected to reduce the load of copper discharged to the aquatic environment. However, most contemporary data available on copper concentrations in sewage (Sorme and Lingerkvist, 2002; Karvelas et al., 2003; Buzier et al., 2006; Rule et al., 2007) focus on total metal, leaving a paucity of information on

the dissolved fraction, despite phase distribution being recognised as a significant factor in determining the mechanism and extent of copper removal in STW (Kempton et al., 1983; Rudd et al., 1983a; Lawson et al., 1984a; Kempton et al., 1987a, b).

There are effectively two treatment stages in conventional sewage treatment, primary sedimentation and biological treatment, where copper (Lester et al., 1979), and other elements (Stoveland et al., 1979a) may be removed. During primary treatment, removal is physical, and depends upon the presence of metal in an insoluble and settleable form (Kempton et al., 1987a, b). Two factors that have the biggest impact on metal removal at this stage are therefore copper partitioning between the particulate and dissolved phases in crude sewage and the efficiency of suspended solids removal (Lawson et al., 1984b, c, d; Kempton et al., 1987a). Typically no removal of dissolved to particulate copper in the settled sewage increases (Olivier and Cosgrove, 1974; Stoveland and Lester, 1980; Santarsiero et al., 1998; Karvelas et al., 2003).

Secondary biological treatment is predominantly based on one of two processes, a biological reactor consisting of either suspended growth (activated sludge) or an attached growth process (trickling filters) each followed by a phase separator (secondary sedimentation). In the activated sludge process the majority of the biomass removed in the secondary sedimentation tank is recycled to the aerator as returned activated sludge (RAS) a defining characteristic of the process (Metcalf and Eddy, 2004). Biological treatment processes are theoretically capable of removing both dissolved copper, mostly through sorption to the extracellular polymeric substances (EPS) produced by bacteria, as well as copper associated with solids, through settling with sludge flocs (Brown and Lester, 1979; Rudd et al., 1983b; Liu et al., 2001). However, due to the strong affinity of copper for dissolved organic ligands and/or possibility of release of EPS from the bacterial cell capsules, an increase in dissolved metal is also possible (Stoveland et al., 1979b; Rudd et al., 1988; Landner and Reuther, 2004; Ziolko et al., in press). The mechanism of copper removal in trickling filters is believed to be not dissimilar to that occurring in the activated sludge process (Lester, 1987) however, although trickling filters are extensively used in the UK, with over 800 STWs utilising them as a biological treatment stage in the South-East region alone (Koh et al., 2008), they have not previously been studied in terms of copper removal and therefore the exact mechanism of removal remains to be established.

The biological treatment stage is increasingly followed by a range of different types of tertiary treatment used to reduce biochemical oxygen demand, suspended solids and ammonia concentration in effluents. Such treatment may possibly assist in further reduction of copper concentrations in final effluents, however, these processes were not specifically designed to remove copper, and their effectiveness is not well documented (Kobler and Boller, 1997; Hamoda et al., 2004).

The aim of this study was to determine the concentrations of both total and dissolved copper in the sewage at the different stages of sewage treatment at a number of STWs in the North West of England in order to evaluate the effectiveness of various unit treatment processes in removing copper.

2. Methodology

2.1. Description of STWs and sampling strategy

Eight STWs were sampled between November 2006 and November 2007. Biological treatment at seven sites consisted of trickling filters (these locations are

identified as TF1 to TF7), and one site utilised an activated sludge plant, identified as ASP. The sampling dates, site characteristics and their operational parameters, such as proportion of returned liquors and recirculation over trickling filters, together with details of any tertiary treatment processes utilised at each STW are presented in Table 1. All catchments were predominantly domestic with less than 11% of the total copper load to the STWs from named traders, with the exception of TF3 where trade input was estimated to be 20% of total load in crude sewage (Pers. Comm. United Utilities).

Samples of the influent and effluent from each unit treatment process were taken every four hours from 10:00 on Monday through to 10:00 on Friday. Further sampling points were added where samples could beneficially be taken, such as combined flows of crude sewage with returned liquors or settled sewage with recirculated flows (Figs. S1-S8 in supplementary material). Samples of sludges and returned liquors, such as co-settled sludge from primary sedimentation, humus sludge (from secondary sedimentation following biological treatment), decant liquors from sludge treatment or backwashes from tertiary treatment processes were collected daily, or as dictated by operation at each site.

Table 1Site characteristics and operational parameters recorded during thesampling periods.

Site	Sampling dates	Design FTFT ^a (l/s)	Actual FTFT ^b (%)	Returns ^c (%)	Recirculation (%)	Population equivalent	Tertiary treatment
TF1	27/11 -01/12, 2006	1033	44	1	50	156,000	-
TF2	08-12/01, 2007	232	49	6	0	20,350	NTF ^d , COUF ^e
TF3	12-16/02, 2007	440	92	10	0	75,000	NTF ^d , RGF ^f
TF4	19-23/03, 2007	266	39	8	100	40,000	BAFF ^g
TF5	04-08/06, 2007	353	31	6	200	38,000	-
TF6	20-24/08, 2007	736	44	12	23	63,000	BAFF ^g
TF7	08-12/10, 2007	216	45	14	26	17,330	-
ASP	12-16/11, 2007	1655	56	7	98	332,932	-

^a Flow to Full Treatment. ^b Average FTFT as percentage of design during the week of sampling

^cReturns volume as percentage of FTFT. ^dNitrifying trickling filters (NTF) ^eContinuously operating up-flow filters (COUF) ^fRapid gravity filters (RGF) ^gBiological aerated flooded filters (BAFF)

2.2. Sample preparation

For the analysis of the concentrations of total copper in the influents and effluents, 40 ml samples were acidified with 2 ml of 70% Optima-grade nitric acid (Seastar Chemicals Inc., Pittsburgh, PA, USA) and then transferred to digestion vessels for microwave assisted acid digestion undertaken on-site according to EPA Method 3015, utilising a CEM Mars 5 digester with XP-1500 Plus vessels (CEM, Matthews, NC, USA). For sludge and returned liquors samples containing suspended solids concentrations greater than 1000 mg/l, prior to digestion (EPA Method 3051) samples were adjusted to 0.1 g/l of solids by dilution with Ultrapure water (Millipore UK, Watford, UK), and further acidified by addition of 1 ml of Optima-grade nitric acid.

Analysis for dissolved copper involved filtration of the sample through a 0.45 μ m cellulose nitrate membrane filter (VWR International, Darmstadt, Germany) combined with a glass micro-fibre GF/D pre-filter (Whatman, Maidstone, UK). Prior to filtration of the bulk sample, filters were washed with approximately 5 ml of sample, which was discarded. The filtrate (40 ml) was then acidified with 1 ml of Optima-grade nitric acid in order to solubilise copper prior to quantification. For sludge and returned liquors samples with high solids content, samples were centrifuged at 5000g for 30 minutes prior to filtration.

All glassware and digestion vessels used during the work were subject to a 2stage cleaning procedure using 3% solution of Decon90 (Decon Laboratories, Brighton, UK) and 3% solution of nitric acid (Merck, Darmstadt, Germany) and rinsed with Ultrapure water after each step.

2.3. Quantification of copper

Copper analyses were performed with a model 4110 ZL Zeeman-corrected graphite furnace atomic absorption spectrometer (Perkin-Elmer, Beaconsfield, UK) using a copper hollow cathode lamp (324.8 nm wavelength, 15 mA) and AS-72 autosampler with 2 ml polyethylene disposable sample cups. Calibration was undertaken using 1, 2, 5, 10, 20 and 50 μ g/l freshly prepared copper standards. Prior to analysis, where necessary, samples were diluted so as to allow for the detection of copper in the linear range of the calibration curve (Sterritt and Lester, 1980).

Five batches of unspiked samples together with samples spiked at low and high standard additions were analysed for both total and dissolved copper to characterise the method performance. Samples for both total (unfiltered and digested) and dissolved copper (filtered, but not digested) spiked at low and high additions exhibited no significant differences between batches with an overall recovery of 100% for total and 90% for dissolved copper. Recoveries for dissolved copper may be explained by matrix effects in undigested samples (Belarra et al., 2000). All reported results were corrected for these recoveries.

2.4. Data analysis

Statistical analyses were performed with JMP[®] 7 (SAS Institute, Cary, NC, USA). Variance in the measured concentration of dissolved copper in samples from the inlet and outlet of the unit treatment processes was analysed using the ANOVA test. The post-hoc Tukey's test was than used to determine where a statistically significant difference existed between mean concentrations before and after a treatment process.

2.5. Mass balance calculations

The mean daily flux of total copper through the unit treatment processes at each site was calculated using concentrations and flows from data collected over the sampling period. Changes in the flows resulting from the addition of returned liquors, recirculation over trickling filters or due to desludging and backwashing were taken into account. Data for fluxes through the works in many cases were calculated by summation of measured values where flows combined, or subtraction where flow streams split and these values are enclosed in boxes when presented in the mass balance diagrams. Due to the uncertainty regarding the exact volumes of sludges and returns and wide variability in their solid content, daily copper fluxes through the works were not calculated.

3. Results

3.1. Spatial and temporal variation in copper concentrations in the crude sewage

The average weekly concentrations of total and dissolved copper determined in the crude sewage at the eight sites are presented in Fig. 1. At the ASP it was not possible to obtain crude sewage prior to the addition of returned liquors and therefore the total copper concentration in the crude sewage was calculated using the mass balance approach described above, with no attempt made to estimate dissolved copper concentrations due to uncertainty regarding partitioning. There was some evidence of spatial variation in both total (14.8 μ g/l at TF1 to 107 μ g/l at TF4) and dissolved (3.0 μ g/l at TF2 to 7.7 μ g/l at TF4) copper concentrations. Copper in the dissolved phase accounted for between 7% (TF4) to 30% (TF3) of the total metal in the crude sewage, which demonstrates that not only the total concentrations of copper entering the STWs, but also the distribution between particulate and dissolved phases may vary considerably.



It was also apparent that there was temporal variation in the concentrations of both total and dissolved copper in the crude sewage entering each of the treatment works. A typical example of the dynamic nature of the crude sewage, which also demonstrates the link between diurnal flow patterns and copper concentrations in dry weather, is shown in Fig. 2. The distinct diurnal pattern can be distinguished with the lowest concentrations of copper consistently observed at the 06:00 sampling point. Even during wet weather, which occurred most frequently at TF3, with FTFT throughout the week averaging 92% of the design capacity (Table 1), a diurnal flow pattern remained evident. The increase in copper concentrations with the increasing flow rate during the day time implied the presence of a copper source with concentrations above those present in runoff. As estimated inputs from trade were mininal, the implication is that this source was of domestic origin.



Fig. 2. - Temporal variation in total (\blacksquare) and dissolved (\Box) copper concentrations (μ g/l) showing a diurnal pattern with flow (l/s) to full treatment (—) at TF5.

3.2. The flux of copper through the sewage treatment works

At all sites, before crude sewage entered the primary sedimentation tanks it was combined with the returned liquors streams. The concentrations of copper present in some of the returned liquors (Table 2) were up to two orders of magnitude above those observed in crude sewage (Fig. 1), indicating the potential impact that returns may have on the load to primary treatment. Concentrations of total copper varied considerably, not only between different return streams, but also at different sites for the same return, however, in all cases copper was predominantly associated with the solid phase.

Obtaining representative samples for flows of combined crude sewage plus returned liquors was, however, difficult as returned liquor flows were frequently unpredictable and access to suitable sampling points restricted. Fluxes to the primary sedimentation tanks were therefore calculated using appropriate data on concentrations and flows or volumes and compared to measured values (where available) for calculating and validating the mass balances for each site, which are available in the supplementary information (Figs. S1-S8). These figures present the flux of total copper in kg/day through the works, with values determined from measured concentrations in open text, and calculated values, from combing two or more flow streams, presented in text boxes. This approach was reliable for quantifying the flux of total metal, however, this method is unsuitable for quantifying the flux of copper present in the dissolved phase due to uncertainty related to partitioning when two or more flows combine.

Although the volume of returned liquors did not account for a large proportion of flow to the STWs, on average 8% (Table 1), their impact on the flux of copper to the primary sedimentation tanks was significant. Copper in the returns accounted for 27% (the ASP) to 66% (TF3) of the load of total metal to the primary sedimentation tanks with the major component typically being associated with biological solids, either humus returns or surplus activated sludge. At a number of sites, where representative samples of crude sewage plus returned liquors were obtained (e.g. TF3, TF4 and TF7), fluxes to primary sedimentation tanks which were based on measured concentrations at this same point, compared favourably with the calculated loads (Figs. S2, S4, S7). However, at TF1 and TF6 (Figs. S1, S6), it was apparent that sampling did not "catch" the total load present in the combined crude sewage and returned liquors, indicating the importance of calculating the load of the total copper by combining the flux in crude sewage and returned liquors.

3.3. The effectiveness of unit treatment processes at removing copper

The average removal of total copper based on fluxes through the STW (including the impact of relevant returns) and dissolved copper (based on measured

concentrations) during conventional primary and biological treatment are summarised in Table 3. The removal efficiencies for total copper have been calculated based on values obtained from fluxes through the STW and therefore they include copper in returned liquors and recirculation flows (where relevant), whereas values for the dissolved metal fraction are determined from measured concentrations which as explained, may not have captured internal return flows within each STW. However, it is known that concentrations of dissolved copper in the returns were relatively low (Table 2) and may not have any significant impact.

Site	Humus Return		Decant Liquor		Backwash	
bite	Total	Diss.	Total	Diss.	Total	Diss.
TF1	2144	0.5	-	-	-	-
TF2	6676	1.8	111	0.8	50	2.9
TF3	1854	1.5	1274	0.4	63	4.1
TF4	4689	3.9	809	1.2	663	3.7
TF5	375	4.0	249	1.1	-	-
TF6	380	2.8	1616	2.6	34.5	4.3
TF7	48	2.6	129	2.2	-	-
ASP	837 ^a	1.7 ^a	-	-	-	-

Table 2Mean weekly total and dissolved copper concentrations ($\mu g/l$) in returnflows.

Surplus activated sludge

The removal of total copper across the primary sedimentation tanks was generally consistent at all sites, ranging from 49% (TF4) to 60% (TF2), with the exception of TF6, where removal averaged just 38% over the sampling period. In addition to the removal of copper associated with solids, removal of dissolved copper was also observed at TF1, TF3, TF5 and TF7, with efficiencies reaching up to 49% (TF7). However, at the other sites, there was no difference between crude and settled sewage concentrations. The variability in the removal of dissolved copper was possibly influenced by both variations in the proportion of returned liquors added to the crude sewage prior to primary sedimentation (Table 1) as well as differences in the nature of the returned liquors, such as redox potential or suspended solids concentration, which may have influenced subsequent partitioning during primary sedimentation.

During biological treatment, at the majority of the sites utilising trickling filters removal of total copper was fairly consistent (Table 3), ranging from 44% (TF7) to 55% (TF5), although, the potential for variability was highlighted by two sites exhibiting more extreme removals of 22% (TF1) and 71% (TF3). The impact of the biological trickling filters on the concentrations of dissolved copper concentrations was not significant at most sites. However, at TF1 and TF5 significant differences between dissolved copper in settled sewage and the effluent from the biological treatment process indicate that substantial dissolution of copper occurred (76% and 94% respectively), highlighting the unpredictable nature of this treatment process in the removal of dissolved copper. It is possible that the rate of the recirculation over the trickling filters, which varied considerably (Table 1), may have changed the characteristics of the settled sewage by influencing redox potential (dissolved oxygen concentration) and decreasing suspended solids, which could in turn affect partitioning.

It was apparent that some of the tertiary treatment processes had the potential to further reduce the total copper load discharged to the environment in final effluents, although, their performance in this respect was variable. This was highlighted by the variable performance of the BAFF processes installed at TF4 and TF6 sites (Table 4). All tertiary processes designed to remove solids (RGF, COUF and BAFF) reduced the

load of total copper, probably as a result of decreasing suspended solids load, although only the BAFF at TF4 removed dissolved copper, and at TF2, the COUF was observed to increase concentrations of dissolved copper.

Table 3Average removal efficiencies (%) of total and dissolved copper acrossconventional unit treatment processes at each site. Negative values indicate anincrease in concentration.

Sito	Primary	r treatment	Biological treatment			
Site -	Total Dissolved		Total	Dissolved		
TF1	53	13	22	-76 ^a		
TF2	60	0	52	17		
TF3	59	41 ^a	71	16		
TF4	49	-1	50	-5		
TF5	54	28 ^a	55	-94 ^a		
TF6	38	-7	52	-7		
TF7	53	49	44	-17		
ASP	59	-	94	50 ^a		
^a Statistically significant (n. 0.05)						

Statistically significant (p=0.05)





Sita	NTF		BAFF		RGF/COUF		
Site	Total	Dissolved	Total	Dissolved	Total	Dissolved	
TF2	-15	20	-	-	45	-50 ^a	
TF3	-15	-3	-	-	32	-3	
TF4	-	-	61	52 ^a	-	-	
TF6	-	-	33	-2	-	-	
a Ctati	$\Omega(t_{1},t_{1}^{\prime},t_{2}^$						

Table 4Average removal efficiencies (%) of total and dissolved copper acrosstertiary treatment units. Negative values indicate an increase in concentration.

Statistically significant (p=0.05).

The overall performance of each complete STW, in terms of the removal efficiency calculated between crude sewage and final effluent, are displayed in Figure 3. Although an attempt has been made to present the sites grouped by the treatment processes present, there is no clear benefit displayed for the removal of copper when tertiary processes are present. It is apparent, however, that the ASP was most effective in removing total copper, but as crude sewage could not be sampled here, no value could be calculated for removal of dissolved copper.

3.4. Concentrations in final effluent

The average concentrations of total and dissolved copper in the final effluents from the STW ranged from 1.0 μ g/l (dissolved) at the ASP to 15.2 μ g/l (total) at TF4 (Fig. 4). As observed in crude sewage, there was a significant spatial variation in both dissolved and total copper concentrations. However, the phase distribution in the final effluents had markedly changed, in comparison to crude sewage, with a higher proportion of copper being present in the dissolved phase and accounting for 26% (TF4) up to 77% (TF5) of the total metal concentration. Additionally, the treatment smoothed the temporal variation initially present in the crude sewages and there was no diurnal pattern visible in the final effluent concentrations. In this set of data, the quality of the effluent from the ASP was noticeably superior, even when compared with the final effluents from trickling filter works incorporating tertiary treatment processes.

4. Discussion

At most of the sites investigated, the concentrations of copper present in the crude sewage were in the lower range of values recently reported (Karvelas et al., 2003; Rule et al., 2006; Olivieira et al., 2007). Industrial inputs were not significant at the sites studied, and diurnal variations indicated domestic users may be an important source of copper to the STWs. Leaching from copper pipes is considered to be the primary source of copper to STWs from domestic and commercial areas (Bouley and Edwards, 2000; Sorme and Lagerkvist, 2002; Landner and Reuther, 2004). There is, however, evidence that inputs from plumbing are decreasing, due to phosphate dosing of potable water, which is known to significantly reduce the leaching of copper from plumbing into the water supply (Edwards et al., 2002; Goh et al., 2008) and it is known that phosphate dosing of potable supplies in the area studied was instigated between 1997 and 1999 (United Utilities, 2001).



Fig. 4 - Total (\blacksquare) and dissolved (\Box) copper concentrations (μ g/l) in final effluents.

Regardless of the source, during this study copper in the influent was predominantly found in the particulate form (70-93%) i.e. associated with solid matter. This is in agreement with observation by other authors (Goldstone et al., 1990a; Karvelas et al., 2003; Buzier et al., 2006), and therefore, copper was expected to be efficiently removed during the primary sedimentation process. Moreover, the addition of retuned liquors that typically contained less than 1% of copper in the dissolved form further increased the ratio of total to dissolved metal. The removal efficiencies of total copper observed in primary treatment during this study averaged 53% and were generally in the higher range of reported values (Olivier and Cosgrove, 1974; Kempton et al., 1987a; Goldstone et al., 1990a; Karvelas et al., 2003). However, removal of dissolved copper during primary sedimentation was also observed at three STWs. This phenomenon appears to be a consequence of the addition of the returned liquors to the crude sewage. Retuned liquors typically contain a large amount of suspended solids that may enhance partitioning of dissolved copper onto the solid phase. Such behaviour has been reported previously (Goldstone et al., 1990a; Buzier et al., 2006), although there is also evidence that the addition of returned liquors may have the effect of increasing the concentration of dissolved copper in the settled sewage (Brown et al., 1973). An increase in suspended solids through the addition of returned liquors may also positively affect the removal of copper associated with particles by increasing flocculation (Stoveland and Lester, 1980; Kempton et al., 1987b). However, variability in removal efficiencies demonstrated that although the addition of returned liquors potentially enhanced copper removal, site specific factors such as the nature / type of returned liquors may have an impact on the fate of copper during primary treatment.

The removal efficiency of total copper during biological treatment varied from 22% to 71% on sites utilising trickling filters. Although during this study, the one site utilising the activated sludge process performed considerably better (94%) than the trickling filters, the removal values reported for this process also vary widely, from 25% (Roberts et al., 1977) through 43% (Karvelas et al., 2003) and 61% (Goldstone et al., 1990a) up to 79% (Lester et al., 1979). This implies that although biological treatment processes have the potential to significantly remove copper, the ultimate performance is highly site specific and may depend on both physico-chemical characteristics of the sewage (Sterritt et al., 1981) and operational parameters (Sterritt and Lester, 1981). If dissolution of copper occurs across the biological process, this is likely to reduce the effectiveness of any tertiary treatment process which reduces the suspended solids in reducing the final concentration of copper discharged.

5. Conclusions

There was a wide temporal variability in copper concentrations in crude sewage, however, they tend to follow the variation in diurnal flow which strongly suggest they are indicative of copper originating from the water supply systems.

There is a wide variation in the removal efficiencies of copper between sites, and this is especially evident for the dissolved metal fraction. Operational parameters such as returned liquors and recirculation over trickling filters may all have an impact on the removal of copper.

As the majority of the copper present following biological treatment was in the dissolved phase, optimisation of existing conventional treatment processes (primary and biological treatment) to maximise dissolved copper removal may have a substantial impact on the overall removal efficiency of the STW and be a more cost

effective option than installing any tertiary process to specifically reduce the discharge of copper.

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Table headers

Table 1Site characteristics and operational parameters recorded during thesampling periods.

Table 2 Total and dissolved copper concentrations (µg/l) in return flows.

Table 3Average removal efficiencies (%) of total and dissolved copper acrossconventional unit treatment processes at each site. Negative values indicate anincrease in concentration.

Table 4Average removal efficiencies (%) of total and dissolved copper acrosstertiary treatment units. Negative values indicate an increase in concentration.

Figure legends

Fig. 1. - Total (\blacksquare) and dissolved (\Box) copper concentrations ($\mu g/l$) in crude sewage. Data for the ASP is calculated from mass balances.

Fig. 2. - Temporal variation in total (\blacksquare) and dissolved (\Box) copper concentrations (μ g/l) showing a diurnal pattern with flow (l/s) to full treatment (—) at TF5.

Fig. 3 – The overall removal efficiencies from crude sewage to final affluent of total
(■) and dissolved (□) copper at the eight STWs. Negative removals indicate an increase in concentration was observed.

Fig. 4 - Total (\blacksquare) and dissolved (\Box) copper concentrations (μ g/l) in final effluents.