Water Quality and Sediment Behaviour of the Future: Predictions for the 21st Century (Proceedings of Symposium HS2005 at IUGG2007, Perugia, July 2007). IAHS Publ. 314, 2007.

The distribution of heavy metals in a highly regulated river: the River Murray, Australia

MARTIN C. THOMS
Riverine Landscapes Research Laboratory, Resource and Environmental Sciences, University of Canberra, ACT 2601, Australia
martin.thoms@canberra.edu.au

Abstract Dams and weirs are efficient traps for sediments and associated pollutants. They interrupt the downstream movement of material, leading to changes in sediment composition. The lower reach of the River Murray, in southeast Australia, is regulated along its 830 km length by a series of ten weirs constructed between 1929 and 1935. Large amounts of sediment have accumulated in each weir pool, and in response to flow regulation the river has initiated a series of channel adjustments. Surface sediment samples taken along 154 km of the river between Locks 2 and 4 reveal the impact of these structures on the textural and geochemical composition of the sediment. Downstream of each weir, surficial sediments were found to be well-sorted medium sands, while poorly sorted fine sands, silts and clays were found downstream (above the next successive weir). Concentrations of sediment-associated chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) were up to five times background levels, presumably as a result of increased agricultural and urban development. Peak heavy metal loadings in the sediment were found in the depositing areas above each weir. It thus appears that maximum environmental disturbance occurred some distance from urbanization. Since heavy metal loads are amplified by changes in sediment texture, the spatial concentrations of these pollutants reflect sediment-transport factors associated with the presence of weirs. We also calculate, for this section of the River Murray, the long-term heavy metal concentrations arising from unabated pollutant runoff from urban areas, and the results provide cause for concern.

Key words serial impoundments; sediment quality; urbanization

INTRODUCTION

Dams and weirs are efficient traps for sediments and pollutants, and the way they interrupt the downstream movement of material causes progressive changes in the sediment composition of a river, both within its impoundments and along its course. Reservoirs retain a certain amount of the imported sediment, the fraction depending on the size of the impounding structure relative to the upstream catchment and the size of material supplied to the reservoir (Annandale, 1987). Large dams with relatively small catchments can trap nearly all the incoming bed-load material and appreciable amounts of the coarser suspended load. The result is that finer sediments are carried to downstream reaches and major changes to the composition of the river bed's sediment can occur. It is now accepted that a major shift in the composition of bed sediment is just one inevitable outcome among the cascade of morphological adjustments brought about by regulating a river (Petts & Thoms, 1986).

Fine river bed sediments are important in the management of riverine ecosystems. Studies have shown the effects of land-use changes and direct channel modifications on sediment loads and channel processes. Although an “excess” accumulation of in-channel sediment occurs following catchment disturbance, this is often of a short-term nature, and over time there is a tendency for reduced sediment loads and the gradual removal of this material (Thoms, 1987). In addition, changes in sediment geo-chemistry, especially those associated with urban development, become an even longer-term issue. Increases in pollutant loads from urban areas and their impact on riverine ecosystems are well documented (Hall, 1987).

Each riverine ecosystem is subject to different stressors, many of which are comprehensively set out in of the literature (Costa *et al.*, 1995; Gurnell & Petts, 1995). Increasingly, rivers are subject to multiple stressors and interactions between individual stressors, either through positive or negative feedback loops, which can enhance or diminish expected individual impacts. Despite the ubiquity of multiple stressors, there have only been a few studies on the combined impacts of multiple stressors on riverine ecosystems (Newson, 1995). The present study investigates the impact that urban development has had on the textural and geochemical character of river bed sediments in the highly regulated lower reaches of the River Murray in southeast Australia.

STUDY AREA

The Murray-Darling river system drains the inland slopes of the Great Dividing Range in southeast Australia. It has a catchment of 1.07 million km^2, with the Murray catchment having an area of 310 000 km^2 above its confluence with the Darling. Most of the basin west of the Great Dividing Range is arid or semi-arid, with mean annual evaporation (1200 mm) considerably exceeding mean annual rainfall (450 mm). Both seasonal and annual streamflows in the basin are highly variable. At Blanchetown, 274 km upstream from the mouth of the Murray, annual discharges for the period 1950–1998 ranged from 6 to 519 $m^3 s^{-1}$ (mean 318 $m^3 s^{-1}$). Flows in the Murray–Darling system are regulated by a series of 21 headwater dams and over 8000 low-head weirs. The Murray meets the Darling 830 km upstream from its mouth, and there are no significant tributaries to the Murray below the Darling junction; this long, slow-moving reach is called the lower River Murray (Fig. 1).

The lower River Murray is a “suspended load channel” (cf. Schumm, 1968) characterized by low bed slopes (mean 0.000055 km^{-1}), low sinuosities (1.2–2.1), low stream powers (0.44–5.25 W m^{-2}) and highly cohesive bank material (silt:clay ratios of 12–41%) (cf. Thoms & Walker, 1993). This section of the river cuts into predominantly sand-sized Tertiary alluvium that in South Australia is up to 60 m thick (Twidale *et al.*, 1978). There are two distinctive geomorphological sections of the lower Murray: one above and another below Overland Corner (near Weir 3, Fig. 1) (Thoms & Walker, 1992). In the Gorge Section, below Overland Corner, the river channel is confined to a limestone gorge 2–3 km wide and 304 m deep, and the flood plain soils are heavy, saline grey clays with poor infiltration rates and low hydraulic conductivities. Upstream, in the Valley Section, the river flows in a valley trough 5–10 km wide, and the channel is flanked by alluvial and lacustrine sediments up to 60-m deep

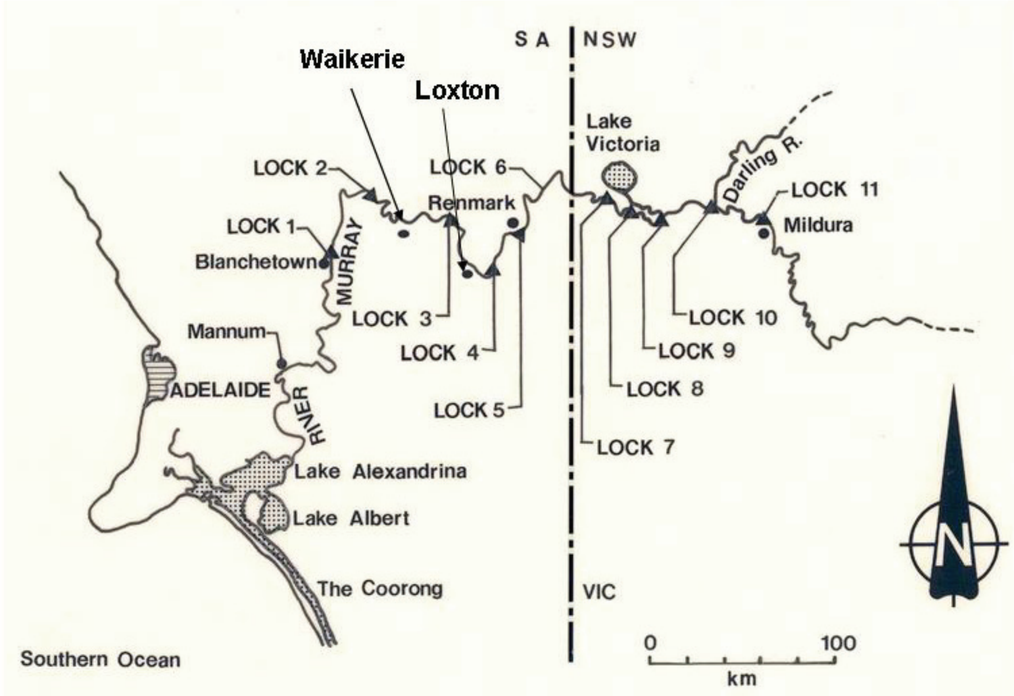

Fig. 1 The lower River Murray, SE Australia. The study reach is that between Locks 2 and 4.

(Twidale *et al.,* 1978). Here, the floodplain soils have a higher sand content, and thus higher infiltration rates and hydraulic conductivities (Cole, 1978). The riverbank soils of the Valley Section are also less cohesive and more easily eroded than those of the Gorge Section.

The weirs on the lower River Murray were originally designed to assist riverboat navigation, but are now important flow regulators. They are operated to maintain a constant upper-pool water level of 50 mm, except when flows exceed storage capacity. Each weir includes a collapsible, navigable pass consisting of 1-m^2 Boule panels suspended between needle beams and trestles, a sluice section comprised of bays of 400-mm deep concrete "stop logs", and an adjacent lock chamber. These structures are referred to as Locks and are numbered 1–10 in upstream sequence (in this paper, the pools associated with the locks are numbered in the same way). This study concerns the 154 km of river occupied between Pools 2 and 3 (Fig. 1) which are governed by Locks 2, 3 and 4 (each in turn located 350, 430 and 514 km upstream from the Murray mouth). The two Pools have similar dimensions and were constructed between 1929 and 1935. The trap efficiency of Lock 2 is 8% and that of Lock 3 is 13%, and they have, respectively, retained 80 723 and 267 470 tonnes of sediment since their construction (Thoms & Walker, 1993). Inevitably, flow regulation along the lower River Murray has changed the morphology of the river channel between Locks 2 and 4. A comparison by Thoms & Walker (1993) of bankfull cross-section data from 1906 and 1988 for this reach showed significant changes in channel morphology, including a reduction in average bed slope of up to 44% in Pool 3; in addition, bankfull cross-sections became wider and shallower below each lock and deeper and narrower in the weir pool immediately upstream of the locks.

The townships of Loxton and Waikerie are located on the mid reaches of each weir pool: Loxton on Pool 3 and Waikerie on Pool 2. These urban areas represent significant point and nonpoint pollutant sources. They are also regional centres for an intensive fruit industry that has traditionally relied upon the use of heavy-metal-based pesticides. In addition, both townships are serviced by combined sewer–surface runoff systems which overflow to the Murray during periods of heavy rainfall.

METHODS

Exactly 100 surface sediment samples were collected along the study reach at approximately equal river distances between Lock 2 and 4. At each site, 10 sub-samples were randomly collected across the channel with a stainless-steel pipe dredge. During retrieval, care was taken to avoid the loss of the finer sediment fractions. In order to reduce errors introduced by site variability and the sampling method, the 10 sub-samples were combined to form a representative composite (as recommended by Mosley & Tinsdale, 1985). The mass of the composite samples ranged from 8 to 12 kg.

Samples were air-dried at room temperature and sieved to obtain statistical measures of the sediment population at each site. Results of the textural analysis were expressed in phi units (ϕ); where $\phi = -\log_2$ (mm). Prior to sieving, a 500-g sub-sample was taken and ultrasonically dispersed in deionized water and then passed through a 4 ϕ (63 μm) stainless-steel sieve. To analyse for heavy metal concentrations, we used this fine sediment fraction since heavy metal concentrations increase with decreasing grain size (Hall, 1987). Total concentrations of chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) were determined by X-ray fluorescence spectrometry. These metals were chosen because they have been reported to reach high levels in urban runoff and sewage (Thoms, 1987). Calibrations were performed against a range of geochemical reference standards and count times provided a detection limit of 1 ppm and precision of ±5%. The overall geochemical character of the surface sediment was also assessed for 12 samples, six from each weir pool. These samples were sieved at half-ϕ intervals and heavy metal concentrations determined for these individual size fractions. The organic content of each composite surficial sample was also estimated by recording the loss on ignition at 550°C for 2.5 h.

The textural data were analysed using a multivariate statistical procedure involving entropy (Forrest & Clark, 1989) which identifies groups of samples with similar grain size distributions.

RESULTS

Surface sediments between Locks 2 and 3 in the lower River Murray were composed of particles ranging in size from fine gravels to silty clay. The contribution of the different size classes varied markedly along the study reach (Fig. 2(a)). In general, the sediments were dominated by medium and fine sands which contributed 23–60% (by weight) of the total sample. Marked changes in sediment composition were seen at three locations in the study area: the reaches located immediately upstream of Locks 2 and 3 (respectively 363–382 and 432–445 km upstream of the mouth) and the reach

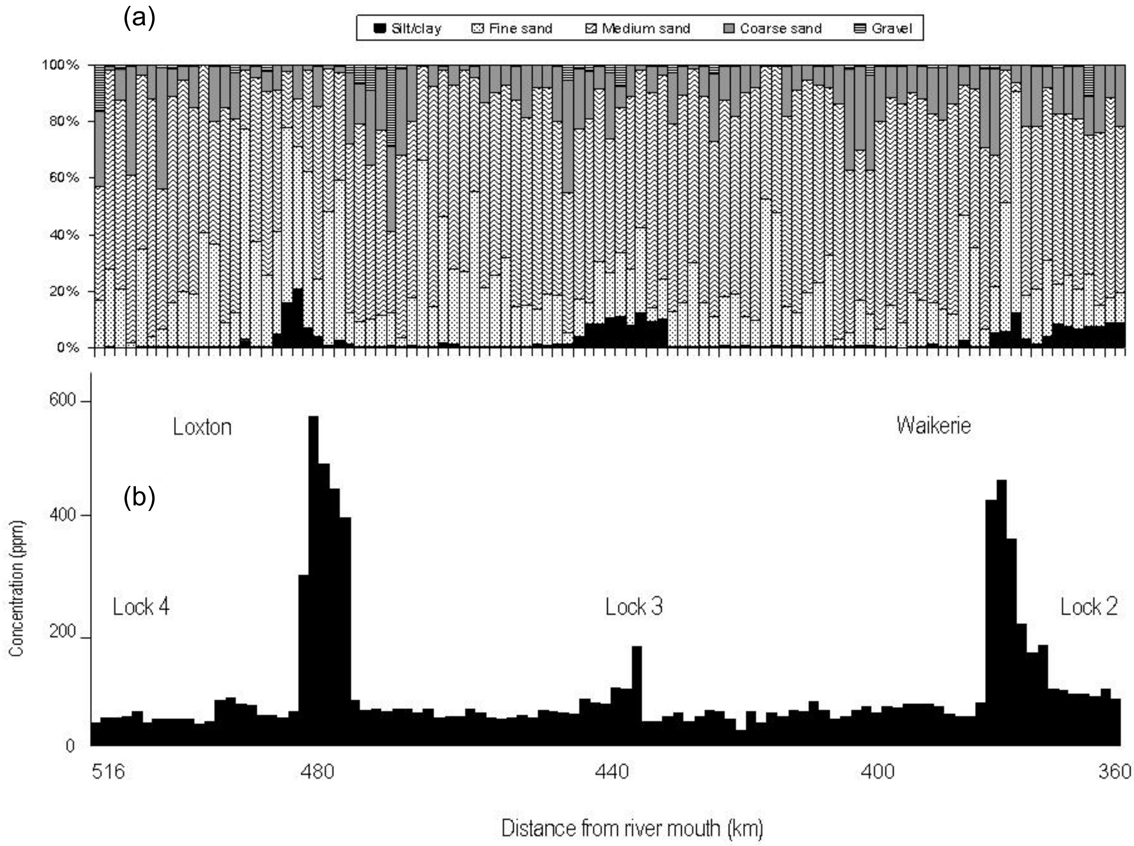

Fig. 2 Surface sediment composition along the lower River Murray between Locks 2 and 4: (a) textural composition; and (b) the concentrations of Lead (Pb).

479–489 km upstream; these were characterized by increases in the fine sand and silty clay fractions. In these reaches the average fraction of fine sand ranged between 16 and 47% (by weight) and silty clay from 7–9% (compared to an average for the whole lower River Murray of 1.6 and 21.2% respectively). Median grain sizes (D_{50}) for the study area ranged from 3.14 to –0.08 ϕ, with no statistical downstream trend being found for either the entire study reach or for individual weir pools.

Entropy analysis identified three distinct groups of grain-size distributions for the study area, accounting for 79% of the total variance between individual samples. Sediments in Group One were relatively coarser in size and better sorted in comparison to those in other zones. Group One sediments had a median grain size of 0.99 ϕ and a sorting coefficient (σ) of 0.87. The surface sediments of Group Three were finer in size, with intermediate sorting values ($D_{50} = 1.63\ \phi$; $\sigma = 1.23$) compared to the other groups. Group Two sediments had a D_{50} of 1.34 ϕ, a σ of 1.89 and were notable because of the variance in grain sizes within the group. However, an analysis of variance (ANOVA) revealed a significant difference between the entropy classes in terms of their median grain size ($f = 19.32$; d.f. = 3, 31; $\rho < 0.005$).

These three sediment groups had a distinct spatial arrangement within each Pool. The sub-reach extending downstream from each lock for approximately 12–14 km was dominated by Group One sediments, while the mid-reaches of each Pool were characterized by sediments from Group Two. The 15–20 km sub-reach upstream of the Locks was comprised of Group Three sediments.

Surface sediments in the study area displayed a similar geochemical feature of increasing heavy metal concentrations with decreasing particle size. Concentrations of most of the heavy metals in the sediment fraction finer than 4 ϕ were 75% higher than in the other sediment fractions. Mean concentrations of the five heavy metals in the study area (in ppm, Cr = 59, Cu = 41, Ni = 25, Pb = 61, Zn = 97) were generally comparable to natural background levels reported by Turekian & Wedepohl (1961) for similar rock types found in the catchment. However, heavy metal concentrations varied between the three sediment groups identified in the entropy analysis for each Pool (Table 1), with enrichment occurring in the mid and lower sub-reaches of each weir pool. For example, Pb had a mean background concentration of 12 ppm, compared to 17 ppm in the reach just downstream of Lock 3, which increased to 99 ppm in the mid reach and 35 ppm in the lower reach of Pool 3 (an enrichment of 41, 723 and 189% respectively). By way of contrast, in Pool 2 mean Pb concentration was 20 ppm in the upper reach, 39 ppm in the mid reaches and 122 ppm in lower reaches (an enrichment of 63, 226 and 913% respectively). Maximum heavy metal concentrations (in ppm, Cr = 135, Cu = 362, Ni = 37, Pb = 722, Zn = 592) were all recorded in the mid reach of Pool 2. Heavy metal concentrations displayed a similar pattern along each pool. For the most part, mean concentrations fluctuated about a value close to those of the geological source rocks, but there were three regions with elevated concentrations. These were located immediately downstream of Loxton and Waikerie, and in an area immediately upstream of Lock 3 (Fig. 2(b)).

Table 1 Heavy metal concentrations associated with surface sediments collected between Locks 2 and 4 along the Lower River Murray. Mean concentrations are given for the different sediment groups identified in the entropy analysis. All values are in parts per million (ppm).

Pool	Sub reach	Chromium	Copper	Nickel	Lead	Zinc
3	Upper	67.3	59.4	29.6	121.5	141.7
	Mid	54.0	26.4	22.6	39.2	104.2
	Lower	53.9	18.7	26.3	19.5	53.0
2	Upper	61.6	41.1	28.2	34.6	74.1
	Mid	64.8	62.3	25.4	98.7	124.5
	Lower	47.2	18.2	17.1	17.0	47.7

Large stores of heavy metals were found within this section of the regulated lower River Murray. When we combined the sediment-transport data of Thoms & Walker (1993) for Pools 2 and 3 with the heavy metal data from this study, our estimate is that 6.6 tonnes of heavy metals have accumulated in the study area since weir construction, with the majority (77%) stored in Pool 3. The quantity of heavy metals was found to vary along each pool, depending upon the textural composition of the surface sediment. Concentrations associated with the sediment finer than 4 ϕ were significantly higher (ANOVA: $f = 29.07$; d.f. = 6, 16; $\rho < 0.005$) in the two sub-reaches immediately upstream of Locks 2 and 3 (Fig. 3). For example, the average reach load for each of the five metals were calculated to be: 5.3 g m^{-3} for Cr, 4.6 g m^{-3} for Cu, 2.0 g m^{-3} for Ni, 9.0 g m^{-3} for Pb and 9.4 g m^{-3} for Zn (for comparison, the corresponding maximum figures were 61, 70, 11, 201 and 117 g m^{-3} recorded in either one of these two reaches).

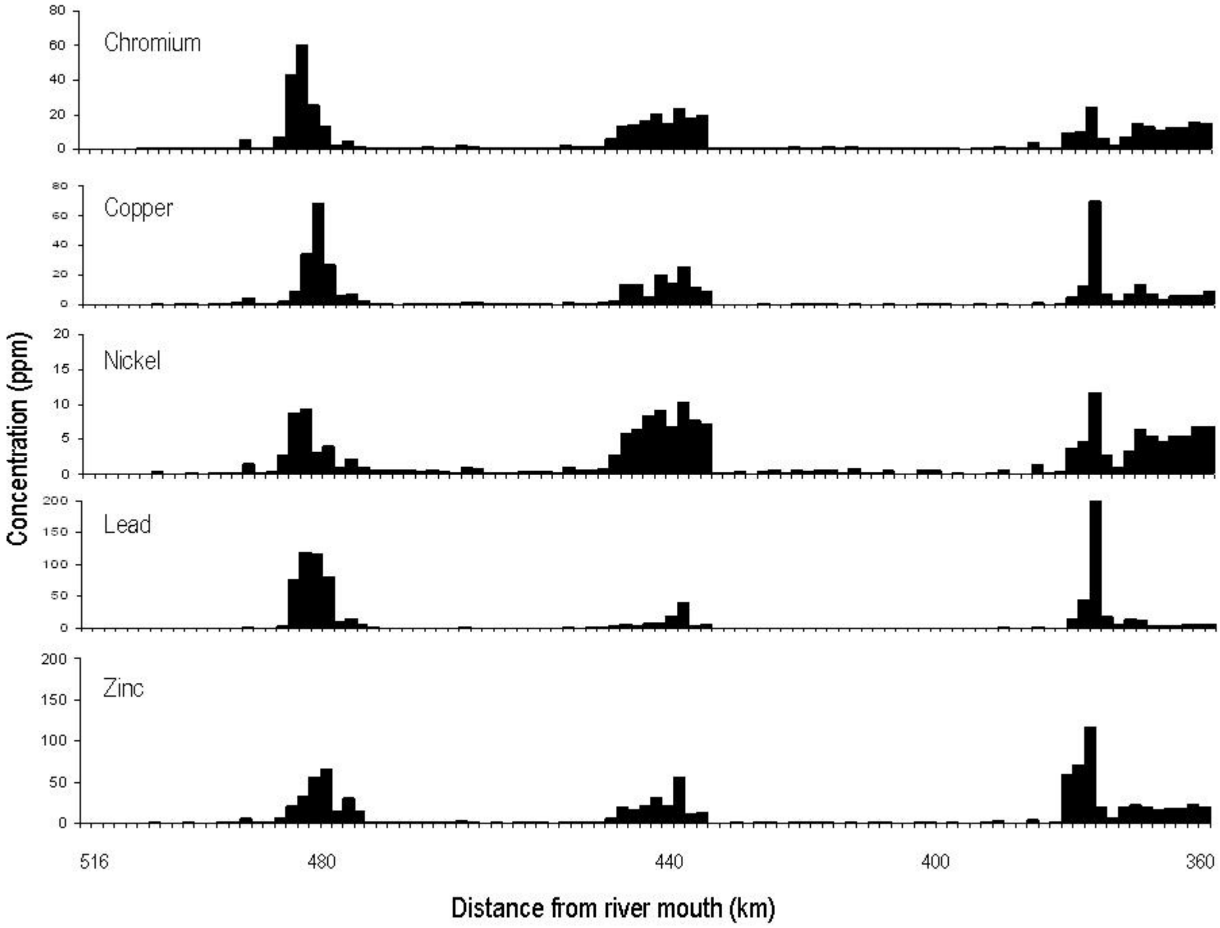

Fig. 3 Heavy metal loads along the lower River Murray between Locks 2 and 4.

Thus, in terms of heavy metal loads, the greatest disturbance along this section of the lower River Murray was not in the immediate vicinity of urban areas but at an appreciable distance downstream.

DISCUSSION

Urban development and the construction of dams and weirs rank among the most common and forceful impacts that humans have inflicted upon riverine ecosystems. Major changes in flow and sediment transport have been reported after catchments have been urbanized (Thoms, 1987) and after rivers have been impounded (cf. Petts, 1984, who illustrates upstream and downstream effects). Nonetheless, studies of the effect of urbanization on the quality of river sediments in regulated systems are few, especially those focusing on the textural and geochemical character of channel bed sediments. This lack has occurred despite the growing awareness of the impact of anthropogenic processes on riverine ecosystems and, in particular, the role of fine sediments in accumulating and transporting contaminants. This study has highlighted the influence of both urban development and flow regulation on the physical and geochemical character of river bed sediments in the lower River Murray. The role of the fine sediment component has been shown to be important and may have major repercussions for the long-term health of this riverine ecosystem.

Heavy metal enrichment in the surface sediments of the lower River Murray can be gauged using the "geoaccumulation index" (*Igeo*), as defined by Salomons & Forstner (1984). A standard background value is employed to calculate a quantitative measure of metal enrichment, i.e.:

$$Igeo = \log_2 (C_n / 1.5\, B_n)$$

where: C_n is the measured concentration of the element n in the given sediment fraction analysed; B_n is the background concentration for the catchment or that given by Turekian & Wedepohl (1961) for the catchment geology; and, 1.5 is a factor to compensate for variations in background data because of lithological effects.

There are seven basic *Igeo* classes: values less than one are essentially background, whereas values over six reflect an enrichment of over 100-fold above standard background concentrations and denote "extreme contamination". *Igeo* values therefore give a way of numerically gauging site contamination. Geoaccumulation values for the lower River Murray range from 0.2 for all metals (background values) to 4.6 for Zn, 7.3 for Cu and 12.7 for Pb (extreme contamination). All those samples collected from upper reaches of both Pools returned values less than one, but 40 and 45% of the samples from the mid and lower pool reaches, respectively, were strongly contaminated by heavy metals and had geoaccumulation values greater than 3.

The textural character of surficial river bed sediments reflects a complex set of hydrological and sedimentological variables. The decline in particle size of surface sediment with distance downstream – Sternberg's Law – has been extensively reported (Yatsu, 1955; Church & Kellerhalls, 1978). This diminution, commonly expressed as an exponential power function, is generally considered to be governed by two main variables. The first is the decline in stream power with distance downstream, a factor limiting transport competence and promoting the selective transport of finer particles (Scott, 1967); the second is the higher cumulative abrasion experienced by sediment as distance downstream increases (Bradley *et al.*, 1972). Although numerous studies have confirmed Sternberg's Law to be a reasonable approximation for the longitudinal sorting of alluvial river sediments, some anomalies have been reported (e.g. Shaw & Kellerhals, 1982). There is no doubt that flow regulation and urban development in the lower River Murray has had an impact on the downstream textural character of the surface sediment. The finer sediment immediately downstream of urban areas, and immediately upstream of each Lock, is enriched with fine sands and silty clay, whereas those downstream of each Lock are dominated by coarser sands. Accordingly, this change in the textural composition of sediment has affected the spatial distribution of heavy metals within the two Pools of this study.

Changes in the rate and composition of sediments accumulating in reservoirs over time are well documented in the literature (e.g. Bogardi, 1974; Garde & Ranga Raju, 1977). These changes have implications for the storage of sediment-associated pollutants in highly regulated riverine ecosystems. Using the reservoir sedimentation model of Annandale (1987), Thoms & Walker (1992) calculated the reach of the lower River Murray between Locks 3 and 4 to have "full" reservoir deposit, i.e. essentially no further accumulation of sediment could take place. However, the reach between Locks 2 and 3 has a river bed slope 50% less than its final predicted "equilibrium" slope. Thoms & Walker (1993) estimate that 80 000 tonnes of sediment needs to be deposited to provide such an equilibrium profile, and this equates to a further 5 tonnes of sediment-associated heavy metals. The Annandale model uses average stream power as an indicator of the sediment-carrying capacity of discharge through a reservoir (Annandale, 1987), and although it provides only a crude estimate of potential reservoir sedimentation, it suggests that changes in the composition of accumulating sediment are inevitable. Thus, the expected changes in river bed slope mean a reduction in sediment competence in the lower River Murray will occur, and hence, over time, finer sediments will accumulate in Pool 2. If so, then the above predictions for future heavy metal accumulations must be viewed as conservative.

CONCLUSIONS

The observed changes in the composition of the river bed sediment in our studied section of the lower River Murray demonstrate that impounding structures such as weirs have long-term impacts – for example, they lead to increases in heavy metals stored within the river substrate. Heavy metal enrichment in this particular section of the river should be of concern to water authorities, even though the full ecological significance has yet to be evaluated.

A broader approach to the assessment of the overall "health" of riverine ecosystems appears warranted. River sediments are key components in the cycling or spiralling (Thoms & Olley, 2004) of pollutants within fluvial systems. They also represent a large secondary contaminant source that may influence the health of a system over extended periods, especially if they are or become biologically available. At present, monitoring programmes only consider water quality or the condition of aquatic invertebrates. Given the known sediment–water interactions and the current emphasis on river health, it is perhaps surprising that no routine monitoring of the quality of fluvial sediments occurs in Australia. There are no guidelines as to the acceptable level of any sediment-associated pollutant. We believe that sediments and sediment storages should be of concern to both land and water managers.

Effective environmental management and the implementation of appropriate monitoring programmes both call for correctly identifying the core critical processes and the scale of their influence. A better geomorphological perspective of the functioning of fluvial ecosystems could lead to improved river health monitoring and management of fluvial systems. The geomorphological factors underlying riverine systems exert a considerable influence on river ecology, so understanding these complex dynamic systems is the first step in river management and restoration. This study has shown that large quantities of contaminated sediments have accumulated in the lower River Murray since the 1920s, and that their densities and dispersal varies along a weir pool. We conclude that the geomorphology of fluvial systems can either amplify or reduce the impact of human disturbances.

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