The Influence of Hydrology on Lacustrine Sediment Contaminant Records

Michael R. Rosen

Abstract The way water flows to a lake, through streams, as runoff, or as groundwater, can control the distribution and mass of sediment and contaminants deposited. Whether a lake is large or small, deep or shallow, open or closed, the movement of water to a lake and the circulation patterns of water within a lake control how and where sediment and contaminants are deposited. Particle-associated contaminants may stay close to the input source of contamination or be transported by currents to bathymetric lows. A complex morphology of the lake bottom or shoreline can also affect how contaminants will be distributed. Dissolved contaminants may be widely dispersed in smaller lakes, but may be diluted in large lakes away from the source. Although dissolved contaminants may not be deposited in lake sediments, the impact of dissolved contaminants (such as nitrogen) may be reflected by the ecosystem. For instance, increased phosphorus and nitrogen may increase organic content or algal biomass, and contribute to eutrophication of the lake over time. Changes in oxidation-reduction potential at the sediment-water interface may either release some contaminants to the water column or conversely deposit other contaminants to the sediment depending on the compound's chemical characteristics. Changes in land use generally affect the hydrology of the watershed surrounding a lake, providing more runoff if soil binding vegetation is removed or if more impervious cover (roads and buildings) is increased. Groundwater inputs may change if pumping of the aquifer connected to the lake occurs. Even if groundwater is only a small portion of the volume of water entering a lake, if contaminant concentrations in the aquifer are high compared to surface water inputs, the mass of contaminants from groundwater may be as, or more, important than surface water contributions.

Keywords Hydrology · Lake sediments · Contaminants · Surface water · Groundwater · Organic pollutants · Inorganic pollutants

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Introduction

Contamination of sediments (both marine and lacustrine) can cause chronic health issues or can be toxic to aquatic organisms and animals that feed on aquatic organisms (Malins et al. 1984; Adams et al. 1992; Long et al. 1995). This can be caused by bioaccumulation through the foodweb, or by directly ingesting contaminated sediment while feeding. Where contaminated sediment comes to rest in a lake is largely the result of hydrological processes that contribute sediment to the lake. Yet, the role that natural or human-induced hydrologic changes in a lake basin plays in the development of the contaminant history of lake systems is often only indirectly assessed through analysis of mass accumulation rates, sediment focusing, or changes in land use. Seasonal or long-term changes in hydrology (changes in circulation patterns, sediment input from rivers, lake levels, or stratification), either natural or human caused, may impact contaminant concentrations in lakebed sediments and may change distribution patterns of contaminants spatially within a lake. Although sedimentary archives of lakes are often used to understand regional or local climate change, evidence of hydrological changes are often preserved that can be used to determine changes in inflow, lake level, precipitation, or groundwater input (i.e. Bradbury et al. 1989; Benson et al. 1991; Arnaud et al. 2005). These changes can be used to track locally derived pollution in the watershed (Spliethoff and Hemond 1996), diagenesis of metals deposited with the sediment (Callender 2000), and constrain interpretations of pollution from outside local areas (Blais 2005; Mahler et al. 2006; Chalmers et al. 2007).

Contaminant deposition in lake sediments can vary depending on the hydrology of the lake and the chemistry of the contaminants (Fig. 1). Local or regional input from rivers and groundwater, or regional to global input from atmospheric sources, may be able to be differentiated in lake deposits with careful observation. Some contaminants, such as nitrates or pharmaceuticals, are more water soluble (hydrophilic) than other contaminants that bind to organic matter or sediment (particle-associated, hydrophobic, or lipophilic). Both particle-associated and hydrophilic contaminants are able to move through groundwater depending on pore-throat sizes of the aquifer material and the redox condition of the water, and enter lakes as non-point source pollutants. Some metals that are soluble under anaerobic conditions in the groundwater may precipitate in nearshore areas of the lake as the groundwater interacts with the oxygenated lake water and the precipitates coat the sediment (Rosen et al. 2002). Hydrophilic compounds may not be directly present in the sedimentary record. However, contaminants such as nitrate may stimulate algal production in the lake, increasing organic nitrogen content of the sediments (Herczeg et al. 2003). Increased algal production due to nutrient inputs may lead to deposition of different species of diatoms (Fritz 1989; Hall and Smol 2010) or other algae, and ultimately lead to eutrophication of the lake. Increased nutrient concentrations may then be represented as increased biogenic silica concentrations in the lakebed sediment, or increased total organic carbon or total nitrogen concentrations in the sediment.

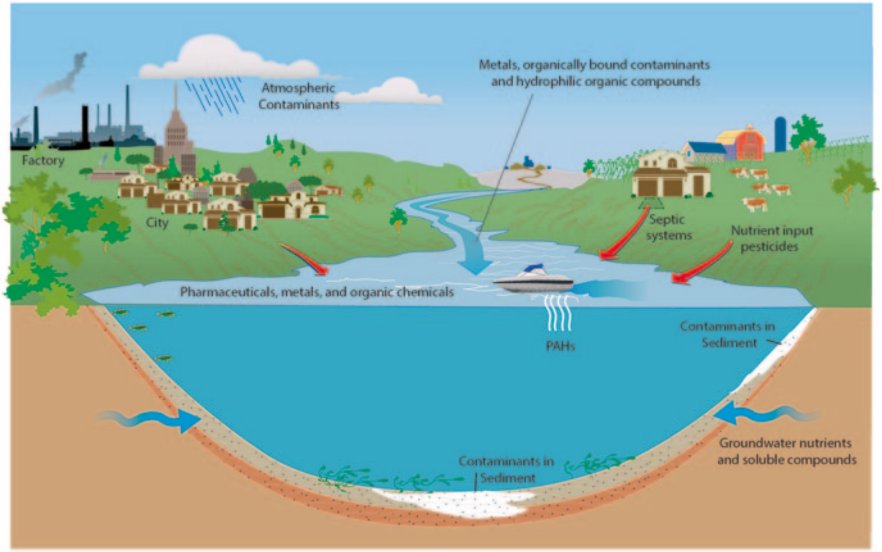

Fig. 1 Diagram of possible important hydrological sources of contaminants to a lake. Surface runoff from cities can be due to paved surfaces and buildings, improperly impounded construction sites, or overflowing sewers. Direct discharge may be from industrial sites or treated municipal wastewater. Groundwater contaminants may accumulate near the discharge point or may circulate throughout a lake if the contaminant is water soluble. Some contaminants may originate from activities on the lake, such as boat traffic

The role that different hydrologic processes play in lake systems, such as groundwater input, seasonal changes in watershed river flows, or human influences on the hydrologic system, is controlled by many factors including the size of the lake and watershed, the regional or local climate, the degree to which the basin is hydrologically open or closed, and the urbanization of a lake's watershed. All of these factors may change over time and be reflected vertically and spatially in the contaminant archive preserved in the lake sediments. Therefore, except perhaps for lakes that receive uniform inputs, spatially distributed sediment cores are likely to enhance the interpretation of contaminant archives in lakes by providing information on the hydrology of contaminant inputs. However, it has been demonstrated in man-made reservoirs of various sizes that sediment distributions appear to be mostly controlled by tributary inputs of sediments of different geologic origin (Abraham et al. 1999). The overall variability in geologic source material and human-derived contaminants implies that the uniform distribution of contaminants in a lake is unusual rather than typical in all lakes. This chapter examines many of these hydrological influences on contaminant records in lakes and illustrates how to utilize this information to enhance interpretations of contaminant histories in lake sediments.

Hydrologic Processes in Lake Systems

Hydrologic processes in lake systems vary in importance for the deposition of contaminants depending on climatic factors, relief of the basin, geology, and watershed and lake area (Fig. 2). In large, deep lakes[1], river inputs generally dominate volumetrically over groundwater inputs (e.g. Schouten 1983; Quinn and Guerra 1986). This pattern is observed because the flow needed to maintain large lake systems must arrive quickly or lake levels would decline. Groundwater, which usually moves slowly (feet per day to much longer time spans), generally is not capable of sustaining deep lakes except in fractured rock or karst terrains where groundwater flow is similar in volume to riverine input (Soler et al. 2007). During drought conditions, most large lakes will shrink even though the flux of groundwater input may actually increase during these times because the elevation between the groundwater table and the lake becomes steeper (Winter 1999).

Although the volume of water entering a lake may be vastly greater from rivers and overland flow in large lake systems, the mass of contaminants entering a lake from groundwater flow may be important because concentrations in the groundwater may be much higher than in the surface water. In Lake Taupo, New Zealand, for example, the volume of inflow from almost 30 rivers entering the lake and direct rainfall onto the lake accounts for approximately 95 % of the water input (Schouten 1983). Yet, the mass of sodium and chloride in the lake is significantly lower in the inflow than in the outflow from the lake, indicating an additional source of these elements that is not included in the dominant freshwater inflows. Lake Taupo was formed from volcanic eruptions and flows, and still has an active volcanic center. Evidence of continued geothermal input to the bottom of the lake has been documented by sampling the deepest parts of the lake bottom (de Ronde et al. 2002). Upward fluxes of Na, Cl and other constituents such as Hg, Sb, Au and S in current and paleo-vent structures into the bottom of the lake indicates that geothermal fluids contribute a large mass of constituents into the lake (Jones et al. 2007), even though the water volume of the geothermal fluids has been calculated to be less than 1 % of the total inflow to the lake (Schouten 1983). In addition, cold groundwater has been calculated to comprise less than 5 % of the water inflow to Lake Taupo (Schouten 1983), yet the mass of nitrogen entering the lake from groundwater has been estimated to be 20–30 % of the total (Rosen 2002). Changes in land use from forestry and natural habitats to sheep and dairy farming since 1960 have been linked to declines in water clarity in the Lake, and a potentially large portion of this decrease in clarity is likely to have come from groundwater nutrient inputs (Hadfield et al. 2001; Rosen 2002).

Conversely small and shallow (or deep) lakes may be dominated or highly influenced by groundwater input (Winter 1976, 1999). Australian shallow coastal lakes may be almost exclusively fed by groundwater (von der Borch 1975; Warren 1982;

[1] No attempt is made to quantify the terms large, deep, small or shallow lakes. This is because the degree to which a hydrologic process is important will vary depending on the combination of these measures of size.

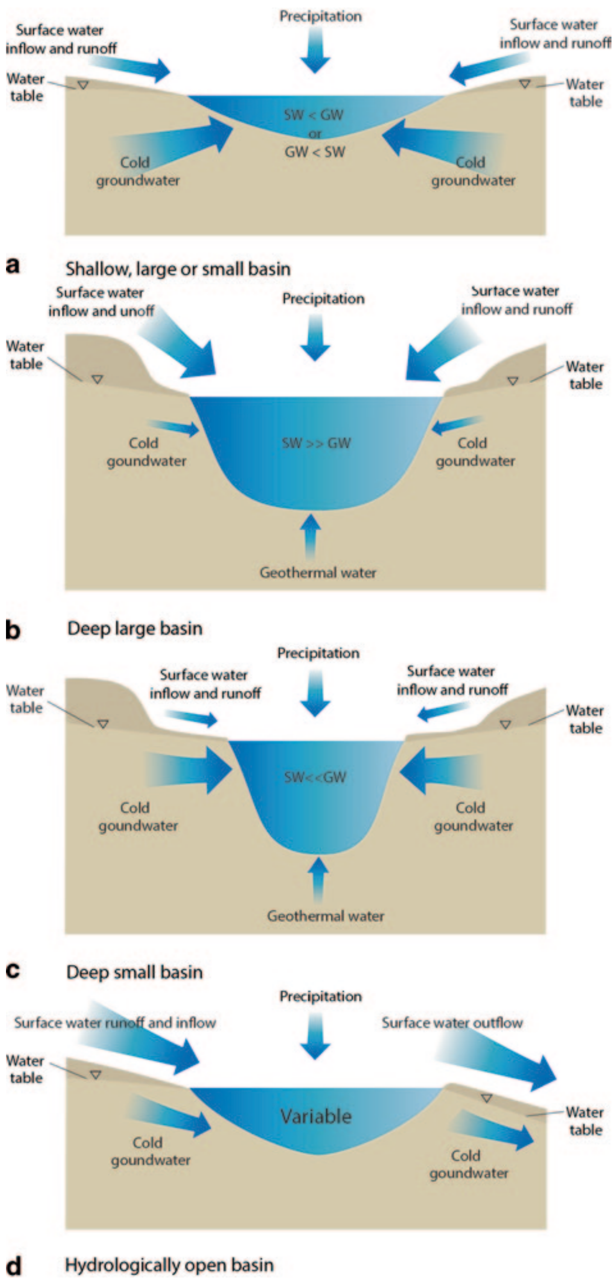

Fig. 2 Schematic of different hydrologic situations in lakes where groundwater or surface water may dominate (relative magnitude of input is indicated by *arrow* size). In shallow closed basins (**a**), large or small, groundwater may be the dominant water source, partly because if constant surface water is entering a lake, the lake will either get deeper or overflow. Groundwater can also dominate shallow hydrologically open basins. In deep large basins (**b**) either hydrologically -open or closed, surface water almost always dominates over groundwater. This is because groundwater flow is generally not fast enough to keep a large deep basin filled. Deep basins that are small (**c**) can be dominated by groundwater where porous rocks or fractures allow swift inflow of groundwater (see text). In hydrologically open basins (**d**) groundwater may be dominant but generally surface water controls water balances and is a greater part of the hydrologic budget than groundwater. Exceptions to all of these generalizations occur and this diagram is intended as a guide

Rosen et al. 1996), and some deep (>50 m) but spatially small maar lakes around the world are also supported mostly by groundwater (e.g. Russell 1885; Kebede 2013). In these types of lakes, water soluble contaminants (i.e. nutrients, major ions, some metals, and organic compounds with low octanol-water partition coefficients (K_{ow}) of less than 3, may be transported through the groundwater (Burne and Moore 1987; Rosen et al. 1996; Vroblesky et al. 1991) or may come from regional atmospheric inputs. Karst lakes may be dominated by regional or local groundwater flow that may travel long distances before reaching a lake (Katz et al. 1995).

Steep watershed topography (the topography of the area surrounding the lake that contributes runoff to the lake) or steep slopes of the lake basin (the slope of the lake bottom) are likely to result in accelerated sediment accumulation rates compared to more gently sloping areas. This may contribute particular architecture to the sediment cores, such as turbidite intervals caused by severe storms or seismic activity, or homogenites caused by rapid groundwater influx after heavy rainfall (Osleger et al. 2009; Twichell et al. 2005; Valero-Garcés et al. 2014). Steep topography can contribute sediment from less human-impacted areas of the watershed that may dilute contaminant concentrations as it enters the lake. Therefore, knowledge of the mass accumulation rates and potential for sediment focusing (see below) need to be taken into account when assessing contaminant contributions from different sources.

Differences in Sedimentation Between Natural Lakes and Reservoirs

Lakes and reservoirs have sometimes been considered to be synonymous because processes such as internal mixing, redox reactions, nutrient cycling, and primary production occur in both lakes and reservoirs (Thornton 1984). However, the variables driving these processes for lakes and reservoirs may not be identical so the response of these two systems may be different. For example, the stream channel up-gradient of the reservoir is likely to aggrade for some distance above the reservoir because of backwater effects on sediment transport. The formation and growth of deltas accelerate and extend the process even farther upstream. Thus channel gradients become flatter, channel cross sections become smaller, flooding occurs more frequently, and drainage of floodplain lands is slowed due to reservoir sedimentation (Glymph 1973). Other factors that distinguish sedimentation in reservoirs versus natural lakes is that reservoirs often have greater drainage area (DA) to surface area (SA) ratios, greater mean and maximum depths, greater shoreline development, and larger areal water loads than natural lakes. The greater DA/SA ratio in particular allows a potential for greater hydrologic and sediment (and contaminant) transport and loading to reservoirs (Thornton 1984). The greater areal water load leads to shorter hydraulic residence time of reservoirs, and allows faster input of contaminants and the potential for greater accumulation (Thornton 1984). For example, at Lake Mead, USA, sedimentation rates were high enough near Hoover Dam at Lake Mead that another reservoir was constructed upstream near Page, Arizona (Lake Powell) to control sedimentation rates in Lake Mead (Rosen and Van Metre 2010).

Many reservoirs, particularly large reservoirs, have been established in major incised river valleys. Another example from Lake Mead illustrates this point. Lake Mead was established by damming the well-entrenched Colorado River that had formed deep canyons in the bedrock (including the Grand Canyon upstream of Lake Mead). After the establishment of Hoover Dam, most of the sediment that had flowed down the entrenched river course began accumulating in the reservoir near the dam (Smith et al. 1960). However, sediment from the surrounding watershed that was washed into the lake became focused into the deep paleo-river channels (Twichell et al. 2005), with less than 1 m of sediment accumulating outside of the deep paleo-channels (Turner et al. 2012). Therefore, sediment deposition in the lake is non-uniform and leads to variable sedimentation patterns and thicknesses in the reservoir. This may be important to consider for contaminant deposition in any reservoir that dams a well-entrenched river.

In smaller reservoirs, sedimentation rates may be variable, with some annual depositional rates that are at the high end for natural lakes (Einsele 2000). Sedimentation rates can be high even in areas with low topographic gradients in the watershed. Measurement of sedimentation rates in eight small reservoirs with watershed areas less than 52 km^2 in Kansas, USA, exhibited sedimentation rates between 0.14 and 9.7 mm/year (Juracek 2004). The topographic gradients in these watersheds are relatively low and smaller watershed areas generally having the higher sedimentation rates. Some of these reservoirs were half-filled with sediment at the time of the measurements, so some of these reservoirs are filling relatively quickly. Not all small reservoirs have high sedimentations rates, but in many cases, reservoirs are located in climatic areas where natural lakes don't exist (Thornton 1984). This may lead to high sedimentation rates because rivers in high topographic gradient regions may carry high sediment loads, particularly during flood conditions. In low gradient areas, watersheds with highly erodible sediment that would naturally have been carried downstream would be trapped behind dams.

Land Use Change as a Surrogate for Hydrology

Changes in land use within a lake's watershed can dramatically affect the hydrology of a lake. Yet, there are relatively few papers that specifically set out to determine hydrologic changes in a watershed caused by human development over time, and without continuously recorded flow information, it is difficult to make this type of assessment. However, changes in land use over time can be used as a surrogate for changes in hydrology. Deforestation of a watershed (Fritz 1989) and changes in agricultural practices (Jacob et al. 2009) can lead to more erosion and runoff entering a lake and will also likely increase stream flow as well (Glade 2003). Irrigation for crops in arid areas may lead to a rise in the water table and increase groundwater flow to a lake (Hutchinson 1937; Rush 1972). The development or growth of cities in a watershed will likely increase runoff due to pavement and runoff from roof tops (Lindström 2001; Chalmers et al. 2007). The use of stream flow or groundwater for drinking water supplies or irrigation may reduce water flow to lakes (Paul and Meyer 2001; Liu and Chen 2006).

The implementation of more sustainable land management practices in agricultural or urban areas may subsequently decrease flows and contaminant inputs to lakes (Makarewicz et al. 2009). Lakes that may have become larger due to increased runoff or groundwater input, may shrink if land use practices change (Scott et al. 2011), potentially impacting wildlife that have grown accustomed to the additional open water. Therefore, it is important to consider the potential impact that various land use changes (for better or for worse) may have on the sediment input to a lake and how the hydrologic character of the lake may have changed independently of climate.

Sources of Contaminants

There may be multiple hydrologic sources of contaminants to a lake from rivers, groundwater, biologic transportation to a lake, or atmospheric inputs. In this chapter, only direct hydrologic inputs, surface water (including rivers, runoff and point sources) and groundwater will be considered. Point sources, such as factories, leaking pipelines, road runoff, wastewater discharge, and leaking landfills, as well as non-point sources, such as agricultural chemical use, closely spaced septic systems, regional smoke stack emissions, and land use change (Fig. 1) may deliver contaminants to lakes somewhat uniformly or in certain areas of lakes. Large lakes with complex basin structure and shorelines and multiple riverine inputs will likely have non-uniform contaminant inputs (e.g. Rosen and Van Metre 2010); whereas, atmospheric contaminants may show relatively uniform distributions in lake sediments (e.g. Heyvaert et al. 2000; Yang et al. 2002). In the following sections, examples of different hydrological inputs are shown as well as post-depositional hydrological changes that may change where sediments are located in lake basins. Contaminant inputs from dry or wet fallout from atmospheric sources are not considered in this chapter but are discussed in Korosi et al. (this volume), Kirk and Gleason (this volume), Catalan (this volume), Gabrielli and Vallelonga (this volume) and Kallenborn (this volume). This is because the majority of rainfall will enter lakes through watershed hydrologic process (runoff, rivers, and groundwater) except in those basins that have extremely limited catchments. Where catchment size is exceptionally small, groundwater may be the main source of water to the lake.

River Inputs and Localized Sources

The most obvious way for contaminants to enter lakes is through runoff to rivers or from direct point-source releases to rivers or lakes from industrial or municipal wastewater. These types of releases may result in more localized sediment contamination than atmospheric inputs because particulate-associated contaminants may not cover the entire lake floor in large lakes. Large lakes and lakes with complex shoreline morphology and multiple river inputs may also help to localize contaminant inputs because of uneven or variable lake bathymetry (e.g. Gewurtz et al. 2008; Rosen and Van Metre 2010). Particulate-bound contaminants entering Lake Mead, USA, from urban runoff, historical industrial complexes, and municipal wastewater are generally confined to one bay (Las Vegas Bay) in the lake (Fig. 3) due to the complex shape of the lake, circulation pattern, and relatively narrow passages between sub-basins. Although dissolved contaminants travel farther into the lake than particulate-bound contaminants (Rosen et al. 2010), the highest concentrations of particle-associated contaminants are found in Las Vegas Bay (Rosen and Van Metre 2010). For example, concentrations of DDE (dichlorodiphenyldichloroethylene), a metabolite of DDT (dichlorodiphenyltrichloroethane), were found in cores from Las Vegas Bay, but not in other areas of the lake (Rosen and Van Metre 2010). The peak in concentration is later than in most areas because DDT was not used in Las Vegas but was manufactured there close to Las Vegas Wash. Leaking holding ponds and careless disposal of waste after the plant closed led to DDT entering Las Vegas Wash. Concentrations declined after cleanup of the site in 1980 rather than after it was banned from use in the early 1970s (Fig. 4).

In Lake Ontario, PCB concentrations were uniformly distributed in deeper sub-basins of the lake, and lower concentrations were found in nearshore areas and areas outside of these deep basins (Oliver et al. 1989). Earlier studies done when many of the PCB generating industries were still operating showed the opposite trend, with higher concentrations in the nearshore areas close to where the contaminants were discharging from rivers. This change was attributed to the fact that these deeper basins accumulate the long-term sediment and contaminant loads and the nearshore loads may be ephemeral or perhaps buried. In addition, lake-wide circulation patterns in morphologically simple large lakes, such as Lake Erie, may redistribute local river inputs long distances from their source (Smirnov et al. 1998).

Industrial complexes may also release contaminants into localized areas of lakes that are reflected in sediment cores near the source. Given enough time and sufficient contaminant releases, these local inputs may affect the entire hydrology of the lake (Rowell 1996). These changes may include changing the timing and duration of stratification in a lake (Owens and Effler 1989) and changes in dissolved oxygen content at the sediment-water interface due to differences in oxygen consumption and release from the sediment over time (Matthews and Effler 2006). High ionic strength wastes from an alkali plant that was located on the shores of Lake Onondaga in New York State, USA, from 1880 to 1986, increased the salinity of the lake, prevented the lake from turning over in some years, and caused it to stay stratified longer each year than it would have naturally (Owens and Effler 1989). Sediment

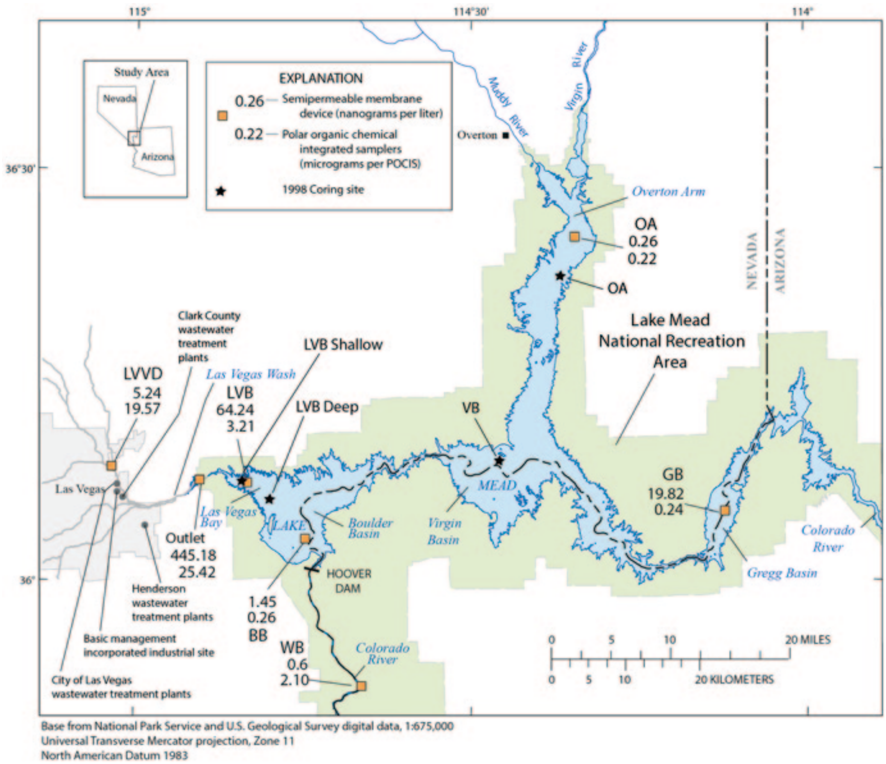

Fig. 3 Map of Lake Mead showing 1998 coring sites from Rosen and Van Metre (2010) and surface water total organic compound concentrations collected by semipermeable membrane devices (hydrophobic compounds) and polar organic chemical integrative samplers (hydrophilic) reported by Rosen et al. (2010). The greatest concentration of organic compounds is coming from Las Vegas Wash downstream of the industrial complexes and wastewater treatment plants. Note that both the hydrophilic and hydrophobic compounds extend more than 10 km out into the lake. The relatively high concentration of hydrophobic compounds in Gregg Basin (*GB*) is likely from boating traffic on the lake at that time

cores taken in the north and south of the lake show similar patterns of calcium carbonate increases caused by the influx of salts from the alkali plant located on the western shore of the lake. In addition, metal and Hg contaminant patterns from urbanization and steel manufacturing also show similar patterns throughout the lake even though the dominant urbanization and industrial activities occur in the south near the city of Syracuse (Rowell 1996). Although Onondaga Lake has two sub-basins divided near the middle of the lake by a 2–3 m high sill, the outlet of the lake is at the north end. This may explain why contaminant profiles in both sub-basins are similar.

Direct hydrological change can lead to rapid eutrophication of the lake. Mathewes and D'Auria (1982) used a combination of lake coring (metal and pollen analysis), sediment trap data, and historical records to outline the contaminant history

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