

SPATIAL AND TEMPORAL DISTRIBUTION OF  
MERCURY IN CABALLO AND ELEPHANT BUTTE RESERVOIRS  
SIERRA COUNTY, NEW MEXICO

By

Colleen A. Caldwell  
Principal Investigator  
New Mexico Cooperative Fish and Wildlife Research Unit

and

Christopher M. Canavan  
Department of Fishery and Wildlife Sciences  
New Mexico State University

TECHNICAL COMPLETION REPORT

Account Number 01345672  
May 1998

New Mexico Water Resources Research Institute

in cooperation with the

Department of Fishery and Wildlife Sciences  
New Mexico State University

The research on which this report is based was financed in part by the U.S. Department of the Interior, Geological Survey, through the New Mexico Water Resources Research Institute.

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## ACKNOWLEDGMENTS

The authors wish to thank Robert Deitner, Dr. William R. Gould, Timothy Thorn, Danny Borunda, Catherine Sikes, and Ed Weber for their assistance in the collection of the data. Forrest Mauldin and Chris Degase of Caballo State Park assisted locating collection sites, and devising and deploying site bouys. Raymond Lucero, Allen Tow, and Rick Williams of the Elephant Butte Field Division (U.S. Bureau of Reclamation) graciously provided the hydrographic information for Elephant Butte and Caballo reservoirs. Ernesto Reyes of the U.S. Forest Service (Gila National Forest) provided information on the Bonner Fire. Drs. Ron Rada and Charles Jagoe and Mr. Nicolas Bloom reviewed several versions of the report. A sincere thanks goes to the team at Frontier Geosciences, especially Nicolas Bloom and Sharon Goldblatt for their technical and moral support. Frontier Geosciences, a privately owned research laboratory, provided the training, instrumentation, facilities and supplies in support of Mr. Canavan's graduate research program. The New Mexico Cooperative Fish and Wildlife Research Unit and the Department of Fishery and Wildlife Sciences at New Mexico State University provided the boat, vehicle, collection equipment, and a portion of Mr. Canavan's salary.

## ABSTRACT

Caballo and Elephant Butte reservoirs are located in south-central New Mexico on the Rio Grande. The reservoirs are managed together for flood control and irrigation. As a result, Caballo Reservoir undergoes seasonal water volume fluctuations creating large littoral or shallow areas. Water and sediment samples were collected monthly for one year (July 1995 to June 1996) in Caballo Reservoir to examine spatial and temporal variability of total mercury (THg) and monomethylmercury (MMHg). Concentrations of THg and MMHg were greatest in water and sediments at the site located in the seasonally flooded area (Palomas) compared to five sites in deep water. In July 1995, the concentration of dissolved MMHg (filtered water sample representing "free" MMHg) from the Palomas site was 0.230 ng/L compared to 0.061-0.087 ng/L in five sites within deep water. Except for July and August 1995, MMHg concentrations in sediments (6.7-11.1 ng/g, dry weight) were greatest at the Palomas site. In contrast, concentrations of MMHg from the five sites were at or below 1.0 ng/g. The percentages of THg in the MMHg form (an index of methylation activity) was greatest in sediment collected from the Palomas site from September 1995 to June 1996 (5.4-33.8%) compared to sediment from the five sites in deep water (0.0-3.6%). By October 1995, a site above Caballo Reservoir in the Rio Grande had greater concentrations of dissolved MMHg (0.508 ng/L) than the Palomas site (0.411 ng/L). The presence of a potential source of contamination upriver, in addition to a series of unrelated events (fire and late summer rains), precipitated a second study from July 1996 to June 1997. Thus, the second year was initiated to determine the sources of THg and MMHg entering Caballo Reservoir. Collection of water and sediment samples was continued monthly from two sites in Caballo Reservoir (rather than six) and one site on the Rio Grande. Two sites were added upstream and downstream of the dam at Elephant Butte Reservoir. From July 1996 to October 1996, Elephant Butte Reservoir thermally and chemically stratified. Concentrations of dissolved MMHg downstream of Elephant Butte Dam increased 14.4 fold from 0.070 ng/L in July 1996 to 1.008 ng/L in October 1996. In contrast, concentrations of dissolved MMHg in water collected from Elephant Butte Reservoir were less than the detection limits (0.018 ng/L) from July 1996 to September 1996. By November 1996, when the Reservoir had undergone mixing, concentrations of dissolved MMHg had peaked at 0.074 ng/L while concentrations of total MMHg (unfiltered water representing both the bound and free fractions of MMHg) peaked at 0.262 ng/L. Coincidentally, stratification and turnover of Elephant Butte Reservoir were accompanied by the shutdown of hydroelectric power generation at the dam. The relationship between concentrations of MMHg, fall turnover, and hydroelectric generation are discussed.

**Keywords:** mercury, water, sediment, arid-lands reservoirs

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## INTRODUCTION

### JUSTIFICATION

Mercury (Hg) research has expanded from acute toxicity effects revealed during the Minimata tragedy in the 1950s to complex mass balance studies modeling atmospheric transport and deposition of Hg to lakes and oceans throughout the world. The source, fate, and effect of Hg in biological systems can be described by the following:

- a) Mercury undergoes a complex biogeochemical cycle within the aquatic environment that involves both biotic and abiotic processes resulting in a variety of chemical species.
- b) The toxicological effects are dependent on the chemical species ingested with monomethylmercury being the most toxic.
- c) Monomethylmercury bioconcentrates, bioaccumulates, and biomagnifies in the aquatic environment.
- d) The biotic methylation of Hg occurs as a result of microbial processes.

### The Mercury Cycle

After entering a body of water, Hg interacts in a complex cycle of physical, chemical, and biological processes resulting in a variety of chemical species, for example, elemental mercury ( $\text{Hg}^0$ ), two methylated forms (monomethylmercury [ $\text{CH}_3\text{Hg}^+$ ] and dimethylmercury [ $(\text{CH}_3)_2\text{Hg}$ ]), and complexes of inorganic mercury (Hg (II)) (Sommers and Floyd 1974; Eisler 1987; Amdur et al. 1993; Zillioux et al. 1993). The physical cycle of Hg can be described relative to inflow, outflow, bioaccumulation, and sedimentation. The inflow of Hg occurs via both wet and dry deposition, runoff, and direct point source inputs (Zillioux et al. 1993; Gustin et al. 1994; Rudd 1995). Atmospheric deposition is a significant source of mercury to freshwater ecosystems (Rada et al. 1989; Mierle and Ingram 1991; Fitzgerald et al. 1994). The largest Hg fraction in wet deposition is in the form of Hg (II) following oxidation of  $\text{Hg}^0$  (Bloom and Watras 1989; Munthe 1994). Significant amounts of MMHg, however, have also been reported (Bloom and Watras 1989; Lee and Iverfeldt 1991; St. Louis et al. 1994). Watershed runoff can also be a significant source of Hg to many freshwater ecosystems and have been observed in runoff from watersheds that contain wetlands or organic rich soils (Lee and Iverfeldt 1991; St. Louis et al. 1994; Verta 1994). Mercury flows out of lakes, estuaries, and reservoirs via outlets or volatilization to the atmosphere. Volatilization of  $\text{Hg}^0$  to the atmosphere is an important component of the biogeochemical cycle and represents a significant efflux of Hg from aquatic systems (Fitzgerald et al. 1994). Gustin et al. (1995)



observed air temperature and air currents were the predominant variables that increased efflux of Hg from contaminated substrates. Sediment acts as a sink to trap Hg both short- and long-term. The passive diffusion of dissolved MMHg from sediments reintroduces the Hg into the water column and to the biogeochemical cycle (Watras et al. 1995). Bio-uptake occurs when Hg is removed from the water column by aquatic plants and animals (*see* Bioconcentration, Bioaccumulation, and Biomagnification). Depending on the fate of the organism, Hg may be removed or recycled into the system.

The biogeochemical cycle is characterized by various chemical states of Hg. Elemental Hg is volatile and may exist in a vapor state. Mercury transformations can occur under either biotic or abiotic processes (Lee et al. 1985; Eisler 1987; Zillioux et al. 1993). Sunlight-induced formation of Hg<sup>0</sup> was described by Amyot et al. (1994). Research suggests that abiotic methylation of Hg can be an important part of the biogeochemical cycle in wetlands (Lee et al. 1985), and photochemical degradation of MMHg is an important component of the abiotic demethylation process (Sellers et al. 1996). The biotic processes probably produce the largest fraction of methylmercury in aquatic ecosystems (Zillioux et al. 1993; Weiner and Spry 1996).

### **Toxicity**

Mercury is a toxic metal with no known essential function in living organisms. Monomethylmercury is the most toxic of all the mercurial compounds and the most difficult for an organism to depurate (Eisler 1987; Amdur et al. 1993; Zillioux et al. 1993; Clarkson 1994). At relatively low concentrations, MMHg is known to have adverse effects on reproduction, growth, behavior, motor ability, appetite, metabolism, blood chemistry, osmoregulation, and oxygen exchange in both marine and freshwater organisms (Eisler 1987; Wiener and Spry 1996). Monomethylmercury perturbs chromosomal structure indicating DNA synthesis is a target for cytotoxicity (Massaro 1994).

Inorganic salts of Hg have low gastrointestinal absorptive rates (less than 7% in humans), facilitating rapid excretion and elimination compared to the organic forms of Hg (Amdur et al. 1993). Upon absorption, MMHg forms soluble complexes with amino acids and proteins containing sulfhydryl groups and will cross the blood-brain barrier and the placental barrier in humans with the thiol-containing amino acid cysteine (Clarkson 1994). The final compound mimics the amino acid methionine resulting in an accumulation of MMHg in the brain and central nervous system (USDHHS 1993). Because MMHg binds to the sulfur group of cysteine, it may be reabsorbed by the biliary tree and the gastrointestinal tract (Amdur et al. 1993; Clarkson 1994). Unlike Hg(II), which is readily excreted via the feces, a small portion of MMHg is eliminated via demethylation by intestinal bacteria in the gastrointestinal tract (USDHHS

1993) and the kidney (Clarkson 1994). Consequently, the half-life (the time in which half the Hg is eliminated) for inorganic Hg salts in humans is much less (40 days) compared to MMHg (70 days). Thus the higher rate of absorption for MMHg combined with its longer residence time (when compared to inorganic Hg species) results in greater toxicity.

### **Bioconcentration, Bioaccumulation, and Biomagnification**

Monomethylmercury is one of the few metals that is known to bioconcentrate, bioaccumulate, and biomagnify in aquatic food webs. The term “bioconcentration” refers to the path of uptake by the contaminant directly from the organism’s surrounding medium (e.g., uptake from water through the gills and epithelial tissue). The term “bioaccumulation” refers to the net uptake of a contaminant from all possible pathways (e.g., uptake via gut from ingestion of food, water, and sediment as well as via gills or epithelial tissue). The term “biomagnification” refers to the increase in contaminant concentration in organisms at successively higher trophic levels as a result of consuming organisms from lower trophic levels.

The primary route of MMHg accumulation is through ingestion, although minor uptake through fish gills has been documented (Ponce and Bloom 1991; Wiener and Spry 1996). As a result of its lipophilic properties and its high affinity for the sulfhydryl groups of proteins, its presence in the diet enables it to accumulate over the lifetime of the fish. Approximately 95 to 99% of the Hg in fish is sequestered in muscle as MMHg (Bloom 1992). As a result, food chain transfer of Hg in aquatic systems is considered to be the most important exposure pathway as MMHg moves from the lowest trophic levels to the higher consumer levels of piscivorous fish, birds, mammals, and humans.

### **Microbial Processes**

It has long been known that certain bacteria in the bottom sediments can methylate Hg (Jensen and Jernelov 1969; Ramlal et al. 1986; Zillioux et al. 1993; Weiner and Spry 1996). Ramlal et al. (1986) observed that methylation was several orders of magnitude greater under anoxic conditions. Sulfate-reducing bacteria, *Desulfovibrio desulfuricans*, living in the anoxic zone of the aquatic environment may be involved in the methylation of Hg(II) (Compeau and Bartha 1984; Gilmour et al. 1992). Watras et al. (1995) observed MMHg was produced in microbial populations inhabiting the anoxic boundary layer found in deep stratified lakes as well as in shallow littoral areas underlying oxic water.

The cycling of organic carbon plays an important role in the transport and bioavailability (biogeochemical cycle) of Hg in lake and reservoir ecosystems (McMurty et al. 1989; Jackson 1991b;

Miskimmin et al. 1992; Driscoll et al. 1994). Driscoll et al. (1994) and Wren et al. (1991) demonstrated that organic carbon correlated with a significant amount of the variation of Hg in fish in Ontario lakes, where MMHg concentrations were linked to the organic carbon released from associated wetlands within the basin. Others have demonstrated that organic enrichment of sediments resulted in greater methylation activity than less eutrophic sediments (Callister and Winfrey 1986). In contrast, Miskimmin et al. (1992) observed an inverse relationship between organic carbon and microbial methylation. Jackson (1991b) explained this inverse relationship to be the result of an elevated supply of organic substrate beyond a critical threshold. Thus the decaying organic matter that stimulates MMHg production may also inhibit the methylation process through the reduction of sulfates to the formation of sulfides that strongly bind to Hg, thereby lowering its availability for methylation.

Researchers have observed marked increases of Hg in fish of newly formed reservoirs. Increased microbial activity occurred as a result of organic matter introduced into the water column following inundation of plants and soil (Bodaly et al. 1984; Jackson 1988; Hecky et al. 1991). Wetlands associated with lakes and reservoirs can also contribute a significant portion of MMHg, presumably as a result of high microbial activity (St. Louis et al. 1994; Rudd 1995). Others have observed Hg discharged from reservoirs resulted in elevated Hg concentrations in fish downstream (Verdon et al. 1991). In a modeling exercise, Rudd (1995) demonstrated the most important source of MMHg in a reservoir was through internal production of MMHg if the percentage of the flooded area exceeded 10% of the reservoir area. Wiener and Spry (1996) reviewed causal mechanisms that resulted in an accumulation of high concentrations of MMHg in fish and noted that trophic structure, feeding habits of the predatory fish, lake morphometry, temperature, prevailing water and sediment chemistry, THg concentrations, and flooding of new impoundments all contributed to MMHg accumulation. Piscivorous fish in newly flooded reservoirs and impounded lakes accumulated high levels of Hg despite the absence of any known point source of Hg contamination (Bodaly et al. 1984; Jackson 1991a; Verdon et al. 1991). Verdon et al. (1991) demonstrated that Hg levels in northern pike (*Esox lucius*) increased from 0.6 µg/g in 1978 prior to impoundment to 2.99 µg/g ten years post-impoundment. Bodaly et al. (1984) observed Hg concentrations remained elevated for decades after impoundment in both walleye and northern pike. They hypothesized the continued bioaccumulation after flooding was due to enhanced microbial methylation of Hg prompted by the decomposition of inundated terrestrial material. Plourde et al. (1997) observed amplification rates of MMHg from water to zooplankton 4 to 9 times in newly formed reservoirs compared to natural lakes. Morrison and Therien (1995) observed elevated concentrations of Hg in the planktivorous whitefish (*Coregonus chupeaformis*) and the piscivorous northern pike in a newly flooded reservoir in Canada.

Mercury in whitefish, however, decreased with successive age classes whereas concentrations in northern pike remained the same with successive ages classes. Mercury concentrations increased immediately after flooding resulting in initially elevated bioconcentration and bioaccumulation rates of Hg in both species. As methylation activity in the reservoir decreased through time, concentrations of Hg diminished in the planktivorous whitefish while Hg concentrations remained elevated in pike as a result of their piscivorous diet.

In addition to organic matter, physical and chemical factors such as dissolved oxygen, temperature, water hardness, and acidity have a profound effect on the biogeochemical cycle of Hg in aquatic ecosystems (Jensen and Jernelov 1969; Bodaly et al. 1984; Clarkson et al. 1984; Eisler 1987; Miskimmin et al. 1992; Watras et al. 1995). Increased oxygenation of sediments decreased the methylation activity of microbes in the Wisconsin River (Callister and Winfrey 1986). Olson and Cooper (1976) observed the net amount of MMHg produced was greater in anaerobic sediments than in aerobic sediments. Lower temperatures inhibited methylation rates (Callister and Winfrey 1986; Jackson 1988). Callister and Winfrey (1986) observed a three-fold increase in methylation rate from 20°C to 35°C under laboratory conditions. Jackson (1988) demonstrated a direct effect of temperature on the accumulation of Hg by invertebrates. An inverse relationship between waterborne calcium and Hg concentrations in fish was observed by Rodgers and Beamish (1983) and McMurty et al. (1989). Rodgers and Beamish (1983) showed the relative efficiency of MMHg uptake by rainbow trout (*Oncorhynchus mykiss*) was much less in hard water than in soft water. A number of researchers have shown the relationship between a decrease in pH and increased Hg accumulation in fish. Wiener (1983) observed increased Hg concentrations in walleye from acidic lakes (<6.0) compared to circumneutral lakes in Wisconsin. Xun et al. (1987) demonstrated that lowering pH (4.5) resulted in a net increase in MMHg production in both the water column and surface sediments. Their study also showed that methylation rates in highly organic lakes would be low because of concurrent high pH (8.5) resulting from high primary productivity. McMurty et al. (1989) observed pH and alkalinity were negatively correlated with increased Hg concentrations of smallmouth bass (*Micropterus dolomieu*). Total Hg in the bass population increased as pH and alkalinity decreased.

## Regional Scope

Environmental legislation has reduced the toxicological impact of Hg contamination in the aquatic environment. Mercury contamination was assumed negligible when point-sources such as industrial discharge and mercurial fungicides were eliminated. Reports of Hg-contaminated fish stocks in the early 1980s as well as the discovery of Hg-contaminated fish in areas remote from human activities, however, have heightened an awareness and expanded research concerning its source, fate, and effect worldwide (Watras and Huckabee 1994). Consumption of Hg-contaminated fish represents the primary route of exposure in humans (Porcella 1994). In response to Hg contamination, 26 states have currently issued health advisories for fish consumers (Knuth 1994). Fish monitoring programs in New Mexico waterbodies have detected total Hg concentrations in edible fish tissues that have resulted in fish consumption advisories as early as 1971 (NMDH 1971). Presently, fish consumption advisories in New Mexico have identified elevated total Hg concentrations in fish throughout the state (NMDH 1991; 1993). A recent health advisory identified total Hg concentrations in fish ranging from 0.11 to 1.2  $\mu\text{g/g}$  wet weight. One walleye (*Stizostedion vitreum*; 720 mm, 1.2  $\mu\text{g/g}$ ) exceeded the U.S. Food and Drug Administration and the New Mexico Environment Department standards for human consumption of 1.0  $\mu\text{g/g}$  wet weight.

Total Hg concentrations (all species of Hg present) were recently measured in gizzard shad (*Dorosoma cepedianum*) (0.041-0.098  $\mu\text{g/g}$ , wet weight) and white bass (*Morone chrysops*) (0.219-0.355  $\mu\text{g/g}$ ) from Caballo Reservoir (Borunda 1997). Although concentrations were less than the recommended advisory levels for human consumption, the values indicate bioaccumulation and biomagnification are prevalent in a New Mexico waterbody. The New Mexico Cooperative Fish and Wildlife Research Unit (NMCFWRU) has recently examined the foraging strategies of a pair of resident bald eagles (*Haliaeetus leucocephalus*) at Caballo Reservoir and observed gizzard shad to be an important food source for these birds. In addition, NMCFWRU has also quantified Hg concentrations in eggs and feathers from nestlings of resident double-crested cormorants (*Phalacrocorax auritus*) at Caballo Reservoir. Although concentrations of total Hg were less than those reported to cause acute toxicological effects in piscivorous birds (Eisler 1987), the consequences of ingesting Hg-contaminated fish throughout the life-time of a bird may have long-term consequences as well as ecological risk implications.

The conditions favoring production of MMHg and its bioavailability in aquatic biota are of primary concern. The majority of Hg research in freshwater ecosystems has been conducted in Europe, Canada, and the northern and eastern United States. Thus, the goal of the research was to characterize the effect of physical and chemical environmental variables on the presence and availability of Hg in arid-lands reservoirs of southcentral New Mexico.

## Objectives

A two-year study (1995-1997) was conducted to characterize the combined effect of selected physicochemical characteristics of water (pH, alkalinity, hardness, dissolved oxygen, conductivity, and temperature) and reservoir volume on the presence and availability of total Hg (THg) and MMHg in water and sediment in Caballo and Elephant Butte reservoirs. Physical and chemical environmental variables were analyzed and discussed in relation to the Hg concentration in water and sediments.

From July 1995 to June 1996, the objectives were to:

- 1) determine general temporal and spatial trends for THg and MMHg concentrations in water and sediment at Caballo Reservoir, and
- 2) identify the physical and chemical factors influencing THg and MMHg concentrations in water and sediments.

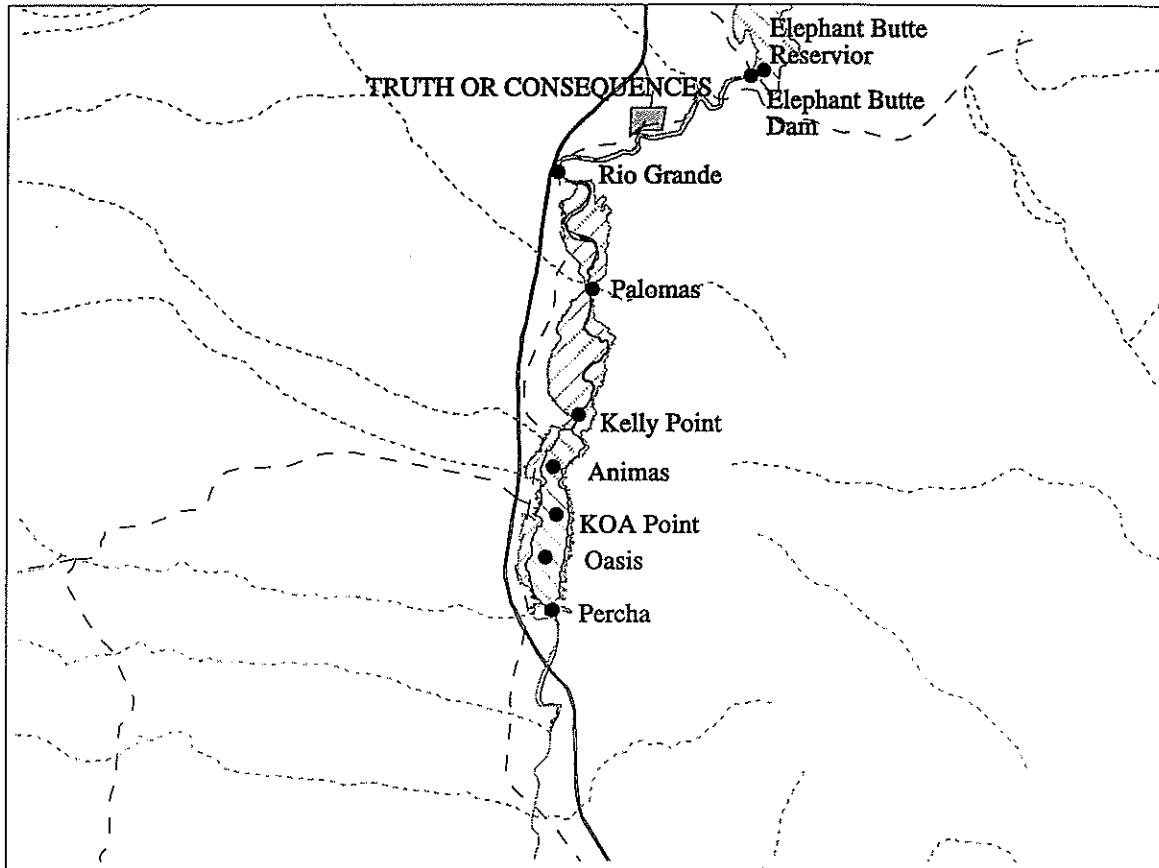
From July 1996 to June 1997, the objectives were to:

- 1) determine the source of THg and MMHg entering Caballo Reservoir, and
- 2) identify the physical and chemical factors influencing THg and MMHg concentrations in Caballo and Elephant Butte reservoirs.

## MATERIALS AND METHODS

### Study Area

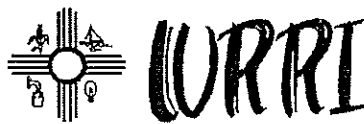
Elephant Butte and Caballo reservoirs are located on the Rio Grande in the Chihuahuan desert of south-central New Mexico (Figure 1). The two reservoirs are connected by a 35-km section of the Rio Grande. Elephant Butte, the upper reservoir, is considerably larger and deeper than Caballo Reservoir and is used for long-term storage. The volume of the reservoir is maintained at a maximum of  $2.5 \times 10^9 \text{ m}^3$  throughout the year (2,015,000 acre-ft). The U.S. Bureau of Reclamation operates a hydroelectric generating station at the dam. From October to November, water released from Elephant Butte Dam is reduced while the turbines are shut down for maintenance. By January or February, the turbines are brought back on-line and controlled releases of water from Elephant Butte Reservoir are resumed. Caballo Reservoir is much smaller and shallower than Elephant Butte Reservoir and has a maximum storage capacity of  $4.1 \times 10^8 \text{ m}^3$  (331,500 acre-ft). It is a shallow polymytic reservoir with a maximum depth of 18 m. The primary purpose of Caballo Reservoir is to store water for irrigation throughout winter and spring and to store excess water from Elephant Butte Reservoir. Once Caballo Reservoir reaches the prescribed fill volume, recharge rates are balanced with discharge rates until mid-summer when the Reservoir is drawn down. As the water level recedes, the previously flooded areas undergo re-vegetation with native annual and perennial plants that are further promoted with late summer rains.



Source: Caballo Reservoir Boundary, U.S. Geological Survey Caballo and Williamsburg 7.5' quadrangle maps. Roads, streams, and towns, RGIS, Albuquerque, NM.

Disclaimer - The information on this map was prepared from publicly available information. Any other use or recompilation of the information, while not prohibited, is the responsibility of the user. The information is for general location and should not be used for anything other than this intended purpose. It should not be used to establish legal title, boundary lines, or locations of improvements. The NMWRRRI expressly disclaims all liability regarding accuracy or completeness of the information contained on this map or the digital computer files.

- Sample Points
- Intermittent Streams
- Perennial Streams
- Secondary Roads
- Primary Roads
- Truth or Consequences
- Caballo Low Water Level (Approximately 75,000 acre-ft)
- Caballo High Water Level (Approximately 275,000 acre-ft)



New Mexico Water Resources Research Institute  
New Mexico State University - Box 310001, Dept. 31187, Las Cruces, NM 88003-0001  
Phone (505) 646-4157 FAX (505) 646-6418 World Wide Web: HTTP://WRRRI.NMSU.EDU

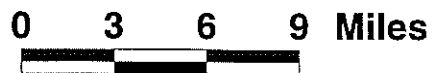
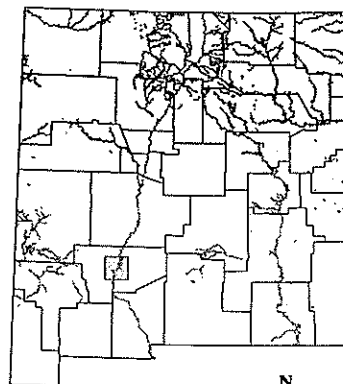


Figure 1. Study Area

The vegetation dies when the areas are inundated, releasing a large amount of organic matter into the water column.

Mean annual rainfall in the watershed surrounding the two reservoirs is 22.1 cm. Edges of the river and the reservoirs contain cottonwood (*Populus wislizenii*), salt cedar (*Tamarix chinensis*), shrub willow (*Salix* spp.), and tree willow (*Salix gooddingii*). Areas adjacent to the lake are composed primarily of creosote (*Larrea divericata*) and mesquite (*Prosopis juliflora*). Over 200 species of plants have been identified in the area, including grasses, forbs, and cacti. Both reservoirs support a number of game fish including white crappie (*Pomoxis annularis*), largemouth bass (*Micropterus salmoides*), white bass (*Morone chrysops*), channel catfish (*Ictalurus punctatus*), and walleye. Double-crested cormorants, western grebes (*Aeccmophorus occidentalis*), blue herons (*Ardea herodias*), and one pair of bald eagles live on and around the reservoirs. The reservoirs also provide winter habitat for golden eagles (*Aquila chrysaetos*), American white pelican (*Pelecanus erythrorhynchos*), and several species of ducks.

There are no known point sources for Hg in either Elephant Butte or Caballo reservoirs. Speculations have arisen that past mining activities in the watershed to the west of Caballo Reservoir may be a source of Hg contamination (Kidd et al. 1974). The impact of minor gold mining activities reported in the area, however, are unknown. Other theories include the weathering of rocks and increased erosion from livestock grazing. There are no known cinnabar (HgS) deposits in the area. Recent literature suggests that both wet and dry deposition play an important role in the addition of Hg to many "pristine" lake systems (St. Louis et al. 1994). Three major creeks (Percha, Animas, Palomas) empty into Caballo Reservoir during intense rain events. These creeks comprise the only drainage routes for a highly erosive watershed which is 19 km wide and 64 km long and extends from the western shore of the reservoir to the top of the Black Range. Small amounts of Hg deposition in this large drainage area could presumably result in a significant influx of Hg into the reservoir. The source of Hg, however, is unknown at this time.

### **Selection of Sampling Sites (July 1995 - June 1996)**

Water and sediment samples were collected monthly from six sites within Caballo Reservoir and one site upstream of the reservoir in the Rio Grande. The sites were chosen according to the following criteria: (1) spatial distribution within the reservoir, (2) sediment textural characteristics, (3) ease in locating and returning to the site, and (4) permanency of water. The six sites within the reservoir were distributed north to south. The most northern site was characterized by narrow margins having shallow littoral zones and intermittent flooding (Palomas) (Figure 1). The remaining sites were characterized by deep water (6-16 m) with limited littoral area (Kelly Point, Animas, KOA Point, Oasis, and Percha) (Figure 1). Beginning in August 1995, an additional site (Rio Grande) was established upstream of the reservoir approximately 16 km north of Palomas to determine



background levels of Hg entering the reservoir (Figure 1). To assist in site relocation, sites within the reservoirs were marked with buoys provided by the New Mexico State Parks and Recreation Department. In addition, a Magellan™ Global Positioning System was used to locate the sites in the event of buoy drift, removal, or destruction.

### **Selection of Sampling Sites (July 1996 - June 1997)**

Water and sediment samples were collected from three sites (Palomas, KOA Point, Percha) in Caballo Reservoir according to the criteria listed above for the previous year. The site on the Rio Grande was retained with the addition of one site in Elephant Butte Reservoir and one site downstream of the Reservoir near the dam (Elephant Butte Dam) (Figure 1). The addition of the two sites above and below Elephant Butte Dam was to determine the sources of Hg concentrations entering the Rio Grande. By September 1996, the volume of Caballo Reservoir had decreased and eliminated sediments at the Palomas site throughout the remainder of the study. Water continued to flow adjacent to the site (5 m), however, and was sampled for Hg and water quality determinations (see Appendix).

### **Sample Handling and Collection**

At each sampling site from June 1995 to July 1996, vertical profiles for pH, temperature (°C), dissolved oxygen (mg/L), and conductivity (µmohs/cm) were determined at one-meter intervals with a Hydrolab™ (Surveyor II, Hydrolab Corp), which was calibrated at the beginning and end of each collection date. Light penetration was measured with a secchi disk (m). Water samples for analysis of total suspended solids (TSS), alkalinity (mg CaCO<sub>3</sub>/L) and hardness (mg CaCO<sub>3</sub>/L) were collected at mid-depth with a Van Dorn bottle at each sampling site and on each collection date (APHA 1985). The water samples were placed in the dark on ice until returning to the lab where they were analyzed within 24 h. At each sampling site from July 1996 to June 1997, vertical profiles for dissolved oxygen and temperature were determined with a dissolved oxygen meter (Model 57) (Yellow Springs Instruments, Inc., OH).

### **Sample Collection and Preparation for Mercury Analysis**

Collection techniques ("clean hands-dirty hands technique") were consistent with specific collection protocols developed to prevent contamination (Bloom 1995). Water samples for Hg analysis were collected from the reservoir surface into rigorously cleaned teflon bottles (Bloom and Fitzgerald 1988; EPA 1995). Prior to the sample collection, the bottle was rinsed once with water from the reservoir or river. The rinses and final collection were taken by submersing the sample bottle, removing the cap to allow the water to flow in, and recapping while submerged. The water sample was placed on ice and held in the dark. Upon returning from the

field, the samples were shipped overnight to Frontier Geosciences, Inc., (Seattle, WA) for sample preparation, storage, and analyses. Upon arrival, an aliquot of each sample was filtered through an acid-cleaned 0.2  $\mu\text{m}$  Nalgene™ disposable filter to obtain the fraction of all species of Hg (THg) and MMHg dissolved in water (dissolved MMHg). The remaining sample was analyzed for total MMHg, which included the dissolved fraction and the fraction of MMHg bound to particulate matter. Monomethylmercury concentrations in filtered water are representative of MMHg that is either “free” or bound to fulvic or humic matter that can pass through a 0.2- $\mu\text{m}$  filter and is hereafter referred to as dissolved MMHg. In contrast, concentrations of MMHg in unfiltered water are representative of both the bound and “free” fractions and is hereafter referred to as total MMHg. Both fractions for each sample were preserved in 0.4 % HCl and stored in the dark until analyses for THg and MMHg.

Surficial sediments for THg and MMHg analyses were collected with a petite ponar dredge at each sampling site on each sampling date. A subsample (approximately 30 g) was removed from the top center of the dredged sample and stored in certified Hg-free glass jars (I-Chem™). Immediately upon collection, sediment samples were held in the dark on ice for transport to the laboratory where they were stored at  $-20^{\circ}\text{C}$  until analysis for THg and MMHg.

### Mercury Analysis

Both filtered and unfiltered water samples were prepared for THg analysis by digestion with the addition of 1% 0.2N BrCl. The samples were vigorously shaken to thoroughly mix and allowed to sit overnight. Digestion with BrCl breaks down covalently bound organo-mercurials to Hg(II) (Bloom and Crecelius 1983). Prior to analysis, 0.2 ml of 20%  $\text{NH}_2\text{OH}:\text{HCl}$  was added to the sample and swirled to reduce the excess BrCl. An aliquot of the digested solution was reduced with 0.3 ml of 10%  $\text{SnCl}_2$ , releasing  $\text{Hg}^{\circ}$ . The  $\text{Hg}^{\circ}$  was purged onto a gold trap with  $\text{N}_2$  stripping and sequentially desorbed with heat for final analysis by cold vapor atomic fluorescence spectrometry (CVAFS) using EPA method 1631 (Bloom and Crecelius 1983; Bloom and Fitzgerald 1988; EPA 1995). Analysis for THg in sediments used the same process described above with the exception of digestion with concentrated acid (7:3 solution of concentrated  $\text{HNO}_3/\text{H}_2\text{SO}_4$ , v/v). The digestion vial was swirled and placed on a hot plate at  $120^{\circ}\text{C}$  and allowed to digest for two hours. After cooling, 30 ml of 5% 0.2N BrCl was added to each vial. The vial was shaken to mix and allowed to sit 12 h. Subsequently, an aliquot was diluted to 100 ml and reduced with 0.3 ml of 10%  $\text{SnCl}_2$ . The  $\text{Hg}^{\circ}$  was purged onto a gold trap following the same procedure used for water.

Both filtered and unfiltered water samples were prepared for MMHg analysis by distillation followed by aqueous-phase ethylation (Horvat et al. 1993). The distillation was defined as complete when approximately 90% of the sample had been distilled over to the receiving vessel (3-4 h). Further distillation carries the HCl

over to the receiving vessel and causes interference during analysis. The sample was transferred to the reaction vessel followed by the sequential addition of 0.3 ml of sodium acetate buffer and 0.035 ml of sodium tetraethyl borate. The vessel was immediately sealed and allowed to react for 17 min. When the reaction was complete, the sample was purged with Hg-free N<sub>2</sub> onto a Carbotrap™ column for 17 min. The trap was removed and connected directly to the purge gas for 7 min to remove any moisture. The trap was thermally desorbed onto an isothermal gas chromatograph column for peak separation and analyzed by CVAFS (Bloom and Fitzgerald 1988; Bloom 1989).

Sediment samples were prepared for MMHg analysis by placing approximately 1.0 g of sample into a distillation vial. This was followed by the stepwise addition of 0.2 ml of 20% KCl, 0.5 ml of 9M H<sub>2</sub>SO<sub>4</sub>, diluted to a total volume of 24 ml with ultra-pure water (Milli-Q™) and 0.2 ml of ammonium pyrrolidine dithiocarbamate was added to the sample. Distillation was defined as complete when approximately 80% of the sample volume had been distilled over to the receiving vessel. Further distillation would carry the HCl over to the receiving vessel and cause interference during analysis. An aliquot of the distillate was added to the reaction vessel for aqueous-phase ethylation in the same manner as for MMHg described above.

After this work was completed, we have become aware of a positive artifact for MMHg which occurs during the distillation of sediments (Bloom et al. 1997). This artifact generally results in false positives on the order of 0.005-0.05% of THg present in the sample, and is positively correlated with organic carbon content. This artifact can be important in sediments where ambient MMHg is less than 1% of the THg. The fraction of MMHg was generally much higher in our sediments, thus, we do not believe the artifact to be of importance in our study.

### Quality Assurance

Analytical instrumentation for the CVAFS was calibrated with a five-point calibration curve prior to analysis each day. The calibration curve was prepared by serial dilution of a certified reference from the National Institute of Standards and Testing (NIST-3133). A verification check was run on the standard curve with a certified standard reference material of lyophilized dogfish muscle (DORM-2) from the National Research Council of Canada (NRCC). For every ten samples analyzed and at the end of the day, instrument calibration checks were performed with a mid-range concentration standard from the initial calibration curve.

Both the analysis for THg, total and dissolved MMHg call for the use of two types of “blanks.” A “bubbler blank” was used to determine the background Hg signal attributable to the analytical instrumentation including the purge bubblers. The “method blank” was used to determine the background Hg signal attributable to the entire method used to prepare the sample. A method blank was treated as a sample containing the same reagents and undergoing the same procedure.

Every 20 samples prepared for THg analysis in water included one duplicate sample set and one duplicate sample set with one matrix spike (Table 1). Every 25 samples prepared for THg analysis in sediments included two duplicate sample sets, one duplicate sample set with one matrix spike, and three method blanks (Table 2). A set of two subsamples of standard reference sediment (PACS-1) from NRCC were also included (Table 2).

Every 15 samples prepared for total and dissolved MMHg analysis in water included a duplicate sample set, one duplicate sample set with one matrix spike, and three method blanks (Table 3). Every 30 samples prepared for MMHg analysis in sediment included two duplicate sample sets, two duplicate sample sets with one matrix spike for each, and four method blanks (Table 4). In addition, a set of three standard reference sediment samples (IAEA-356, International Atomic Energy Agency ) were also included (Table 4).

The minimum detection limit (MDL) was computed as three times the standard deviation from the mean of the method blanks. The MDLs were 0.12 ng/L for THg in water, 0.57 ng/g for THg in sediment, 0.018 ng/L for total and dissolved MMHg in water, and 0.012 ng/g for MMHg in sediment. Precision was calculated from the relative percent-difference of the duplicates of an analytical set and did not exceed 25%. Bias was calculated from the percent recovery of the duplicate matrix spike samples and did not to fall outside  $\pm 25\%$  of the mean. The following summaries demonstrate the level of accuracy for THg and MMHg analyses in sediment and water and are deemed acceptable.

Table 1. Summary quality assurance statistics for THg in water.

Parameter	Units	Mean	Standard deviation	n
Method Blanks	ng/L	0.15	0.04	31
SRM (DORM-2) <sup>1</sup>	ng/L	4497	283	21
Lab Duplicates <sup>2</sup>	$\Delta\%$	12.3	11.8	20
Spike Recoveries	% recovery	92.7	8.5	16

<sup>1</sup> Standard reference material (SRM) DORM-2 certified range 4510-4830 ng/g (4670 ng/g  $\pm$  160).

<sup>2</sup> Duplicates were judged acceptable if less than 25% of the Relative Percent Difference ( $\Delta\%$ ).

Table 2. Summary quality assurance statistics for THg in sediment.

Parameter	Units	Mean	Standard deviation	n
Method Blanks	ng/g	0.24	0.19	12
SRM (PACS-1) <sup>1</sup>	ng/g	4984	409	12
SRM (DORM-2) <sup>2</sup>	ng/g	4515	187	11
Lab Duplicates <sup>3</sup>	Δ%	8.6	5.9	14
Spike Recoveries	% recovery	98.7	11.9	11

<sup>1</sup> Standard reference material (SRM) PACS-1 certified range 4380-4700 ng/g (4540 ng/g ± 160).

<sup>2</sup> DORM-2 certified range 4510-4830 ng/g (4670 ng/g ± 160).

<sup>3</sup> Duplicates were judged acceptable if less than 25% of the Relative Percent Difference (Δ%).

Table 3. Summary quality assurance statistics for total and dissolved MMHg in water.

Parameter	Units	Mean	Standard deviation	n
Method Blanks	ng/L	0.006	0.006	61
SRM (DORM-2) <sup>1</sup>	ng/L	4170	501	21
Lab Duplicates <sup>2</sup>	Δ%	7.4	8.8	30
Spike Recoveries	% recovery	95.9	11.7	25

<sup>1</sup> Standard reference material (SRM) DORM-2 certified range 4840-4100 ng/g (4470 ng/g ± 370).

<sup>2</sup> Duplicates were judged acceptable if less than 25% of the Relative Percent Difference (Δ%).

Table 4. Summary quality assurance statistics for MMHg in sediment.

Parameter	Units	Mean	Standard deviation	n
Method Blanks	ng/g	0.003	0.004	21
SRM (IAEA-356) <sup>1</sup>	ng/g	5.45	1.11	15
SRM (DORM-2) <sup>2</sup>	ng/g	4823	607	12
Lab Duplicates <sup>3</sup>	Δ%	10.2	12.3	10
Spike Recoveries	% recovery	97.6	7.9	14

<sup>1</sup> Standard reference material (SRM) IAEA-356 certified range 4.53-6.11 ng/g (5.32 ng/g ± 0.79).

<sup>2</sup> DORM-2 certified range 4840-4100 ng/g (4470 ng/g ± 370).

<sup>3</sup> Duplicates were judged acceptable if less than 25% of the Relative Percent Difference (Δ%).

## RESULTS AND DISCUSSION

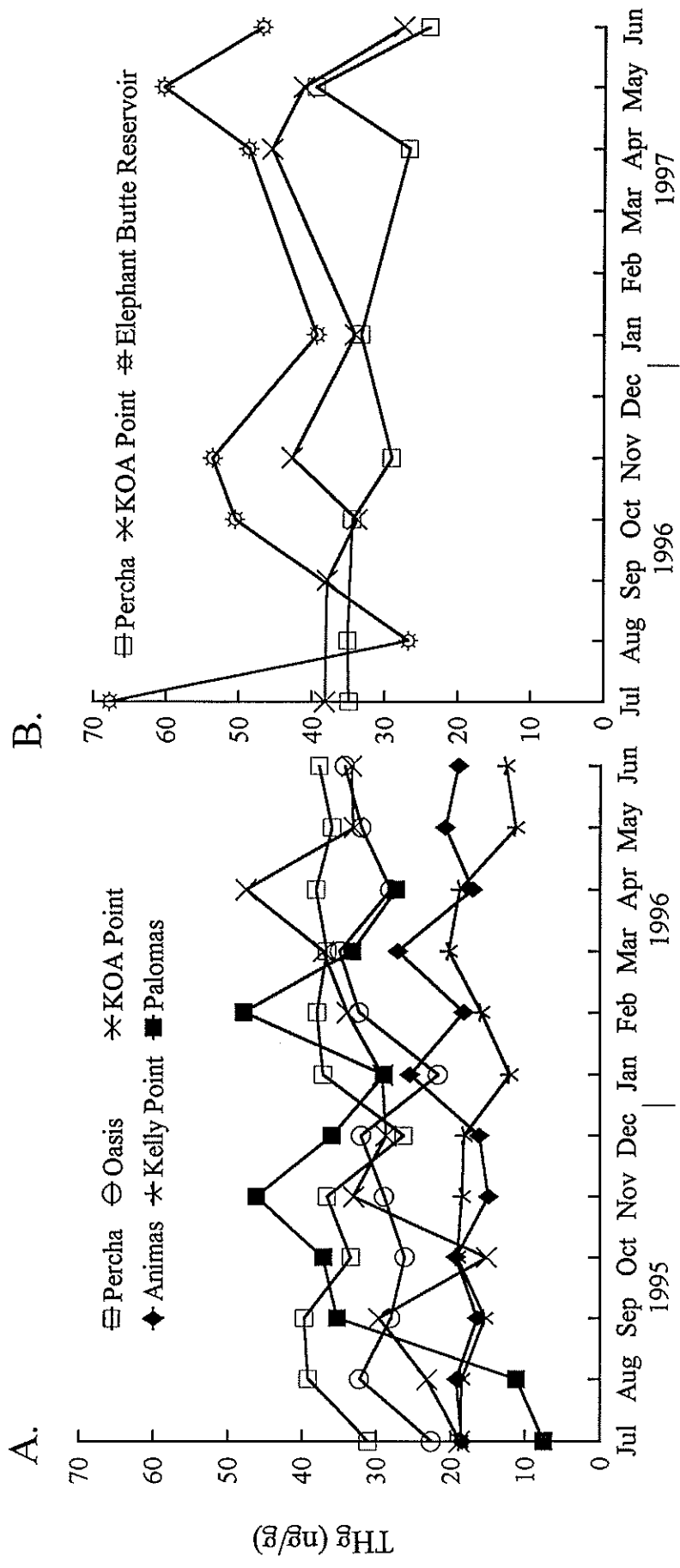
### Temporal and Spatial Trends for THg and MMHg Concentrations In Sediments

Concentrations of THg in sediments varied at all sites throughout the study with concentrations ranging from 7.5 ng/g at Palomas in July 1995 (Figure 2A) to 67.7 ng/g at Elephant Butte Reservoir in July 1996 (Figure 2B). Concentrations of MMHg in sediments were 1.0 ng/g, or less, July and August 1995 (Figure 3A). By September 1995, MMHg increased 10 fold in sediment from the Palomas site and remained elevated until April 1996 (Figure 3A). By May 1996, the reduced water levels resulted in the loss of the Palomas site through the remainder of the study. Concentrations of MMHg in sediment from the remaining sites were less than 2.5 ng/g throughout the entire study.

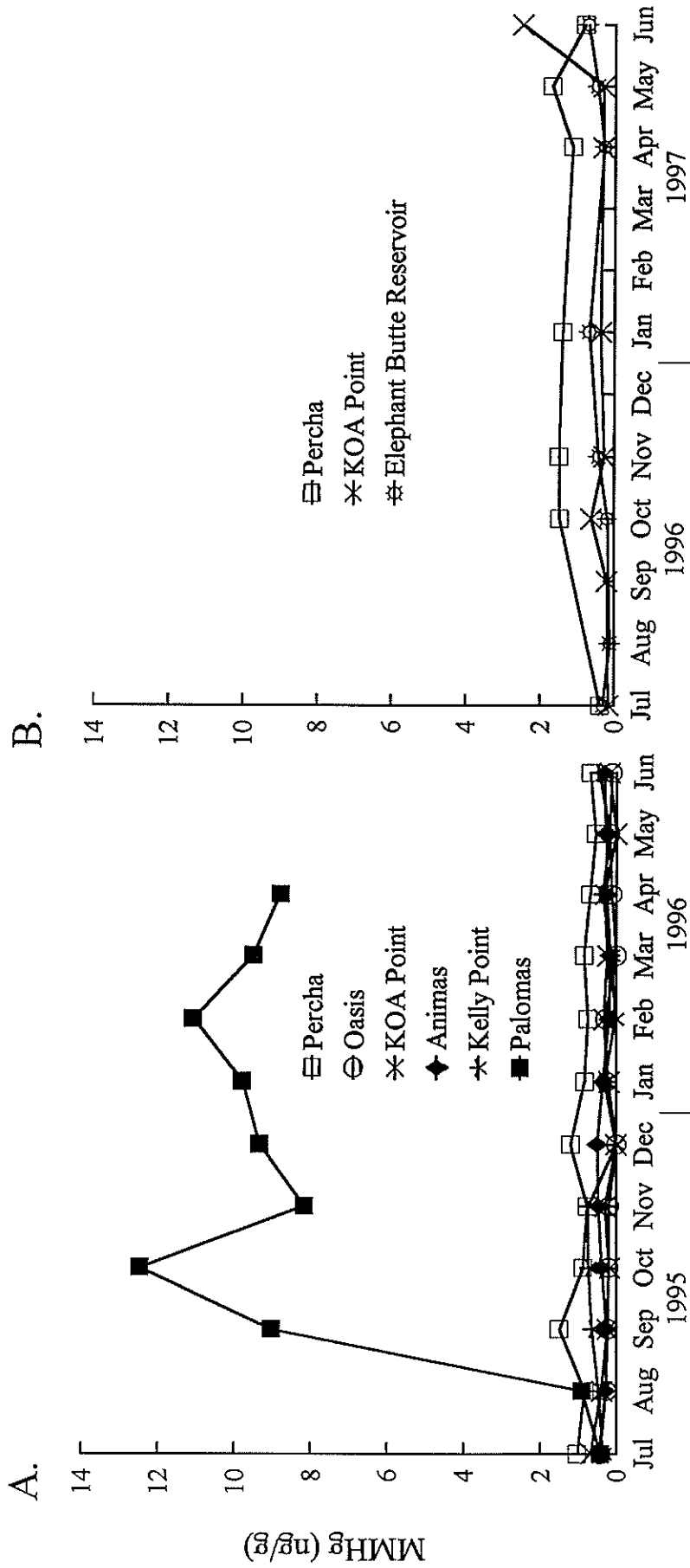
The ratios of MMHg:THg, an index of methylation activity, were greatest in sediment collected from the Palomas site from July 1995 to June 1996 (5.4-33.8%) compared to ratios in sediments from the remaining sites (0.0-3.6%) (Figure 4A). Prior to the Palomas site drying up in September 1996, the ratio of MMHg:THg had increased at the Palomas site to 26% (Figure 4B). We believe these elevated Hg concentrations in Palomas sediment were the result of a fire that occurred in the Black Range on 4 July 1995 (Personal communication: E. Reyes, U.S. Forest Service). The fire burned approximately 8,806 ha in the headwaters of the South Fork of Palomas Creek, which flows into Caballo Reservoir. Shortly thereafter, runoff from a series of rains carried an extensive amount of charred and uncharred vegetative material down the South Fork of Palomas Creek and dispersed the material throughout the northern end of the reservoir. The northern end of Caballo Reservoir (approximately 10 km<sup>2</sup>) was inundated coincidentally as a result of unseasonably large reservoir volume (4.34 x 10<sup>8</sup> m<sup>3</sup> or 250,000 acre-ft). When the volume of Caballo Reservoir decreased by nearly 1.74 x 10<sup>8</sup> m<sup>3</sup> (100,000 acre-ft) in September 1995, the northern reach of the Reservoir receded into a narrow river channel thereby transforming the recently inundated area into pastures containing ash and decomposing vegetation.

### Temporal and Spatial Trends for THg and MMHg Concentrations in Water

Total Hg in water is not presented due to a large number of contaminated samples. The majority of the water samples filtered for THg were contaminated when the receiving vessels, used to collect and store the filtrate, had adsorbed Hg during previous use. The source of contamination was identified and the process for cleaning containers and equipment was improved to prevent further contamination.

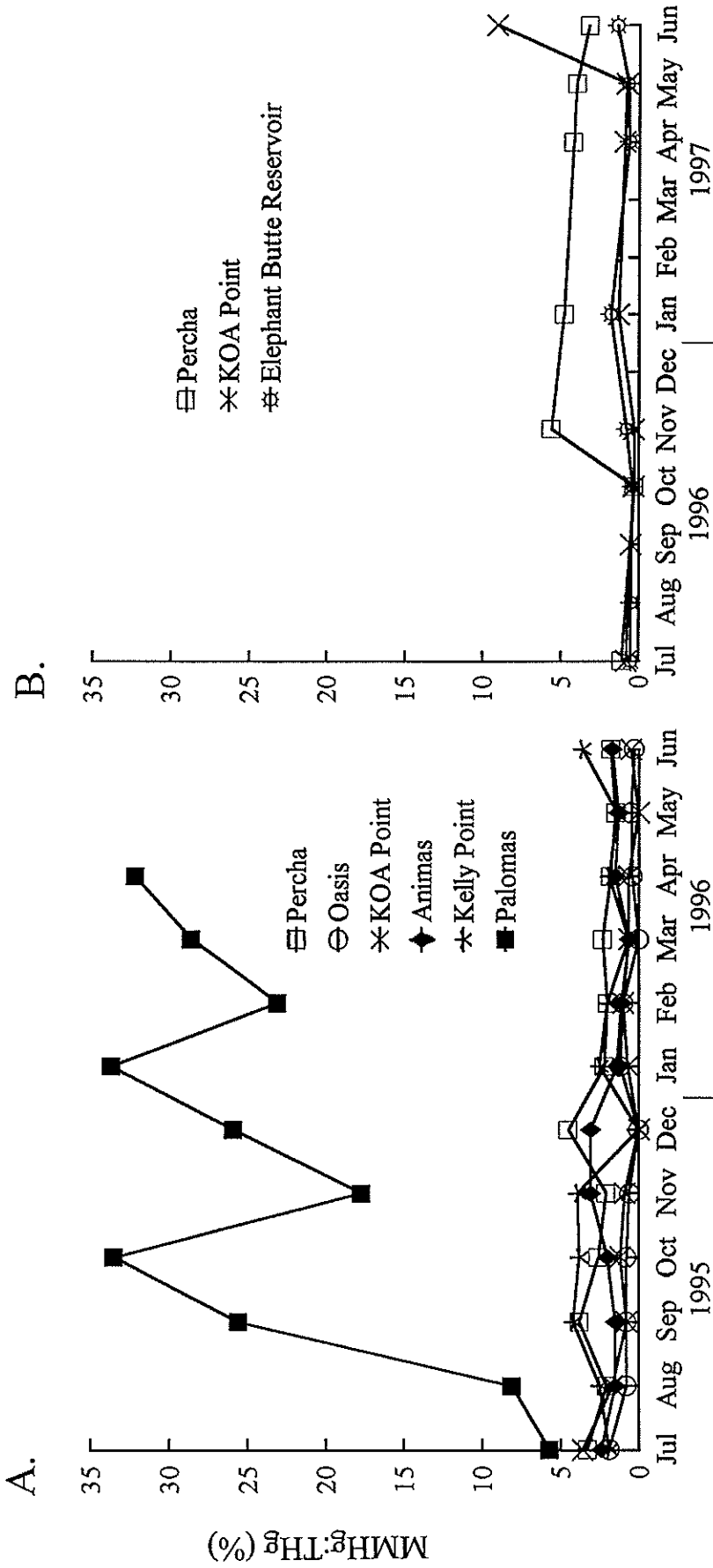


Figures 2 A and B. A. Temporal trends (1995-1996) for concentrations (ng/g) of total mercury in sediments (dry weight) from six sites at Caballo Reservoir. The Palomas site was dry in May 1996 and June 1996. B. Temporal trends (1996-1997) for concentrations (ng/g) of total mercury in sediments (dry weight) from Elephant Butte Reservoir, and two sites at Caballo Reservoir. The Palomas site remained dry from July 1996 to June 1997.



Figures 3 A and B. A. Temporal trends (1995-1996) for concentrations (ng/g) of monomethylmercury in sediments (dry weight) from six sites at Caballo Reservoir. The Palomas site was dry in May 1996 and June 1996. B. Temporal trends (1996-1997) for concentrations (ng/g) of monomethylmercury in sediments (dry weight) from Elephant Butte Reservoir, and two sites at Caballo Reservoir. The Palomas site remained dry from July 1996 to June 1997.



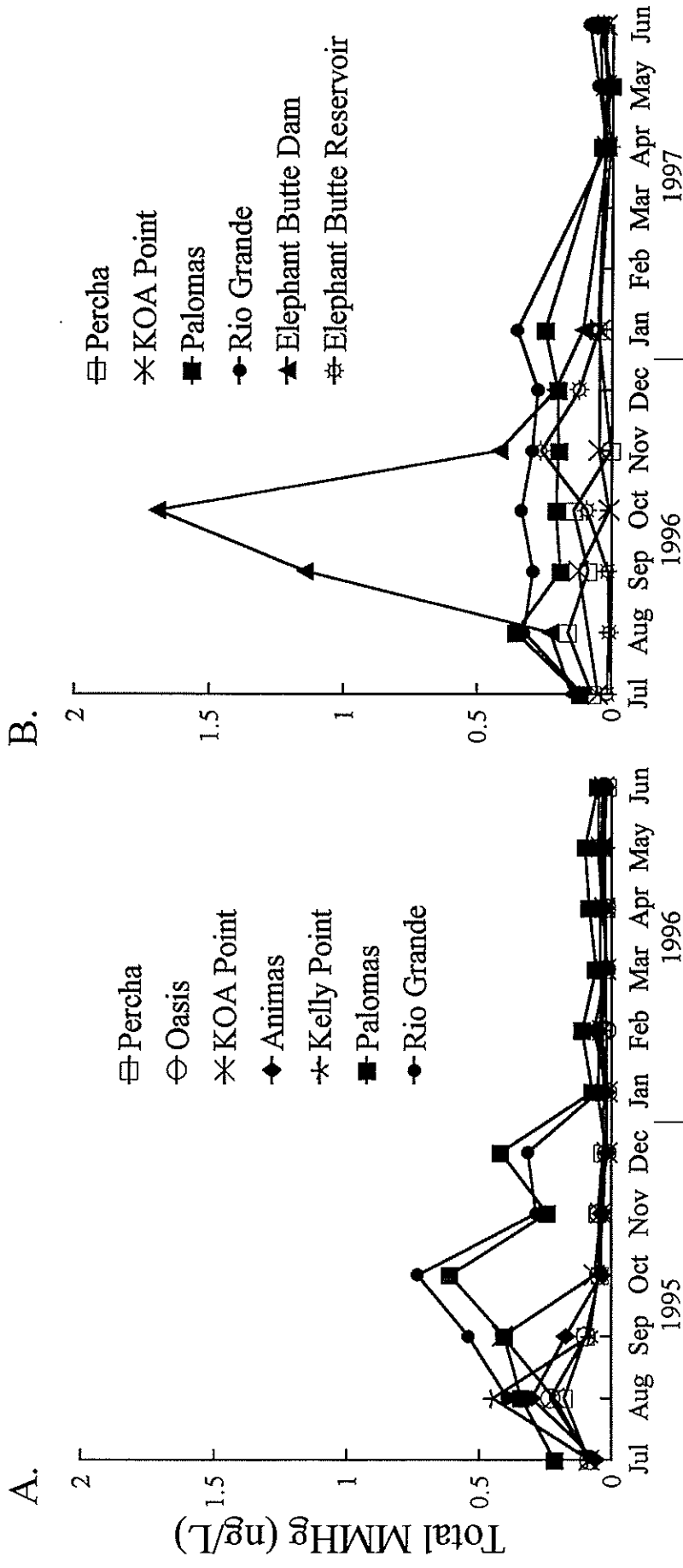


Figures 4 A and B. A. Temporal trends (1995-1996) of ratios for monomethylmercury to total mercury (MMHg:THg) (%) in sediments from six sites at Caballo Reservoir. The Palomas site was dry in May 1996 and June 1996. B. Temporal trends (1996-1997) of ratios for monomethylmercury to total mercury (MMHg:THg) (%) in sediments from Elephant Butte Reservoir, and two sites at Caballo Reservoir. The Palomas site remained dry from July 1996 to June 1997.

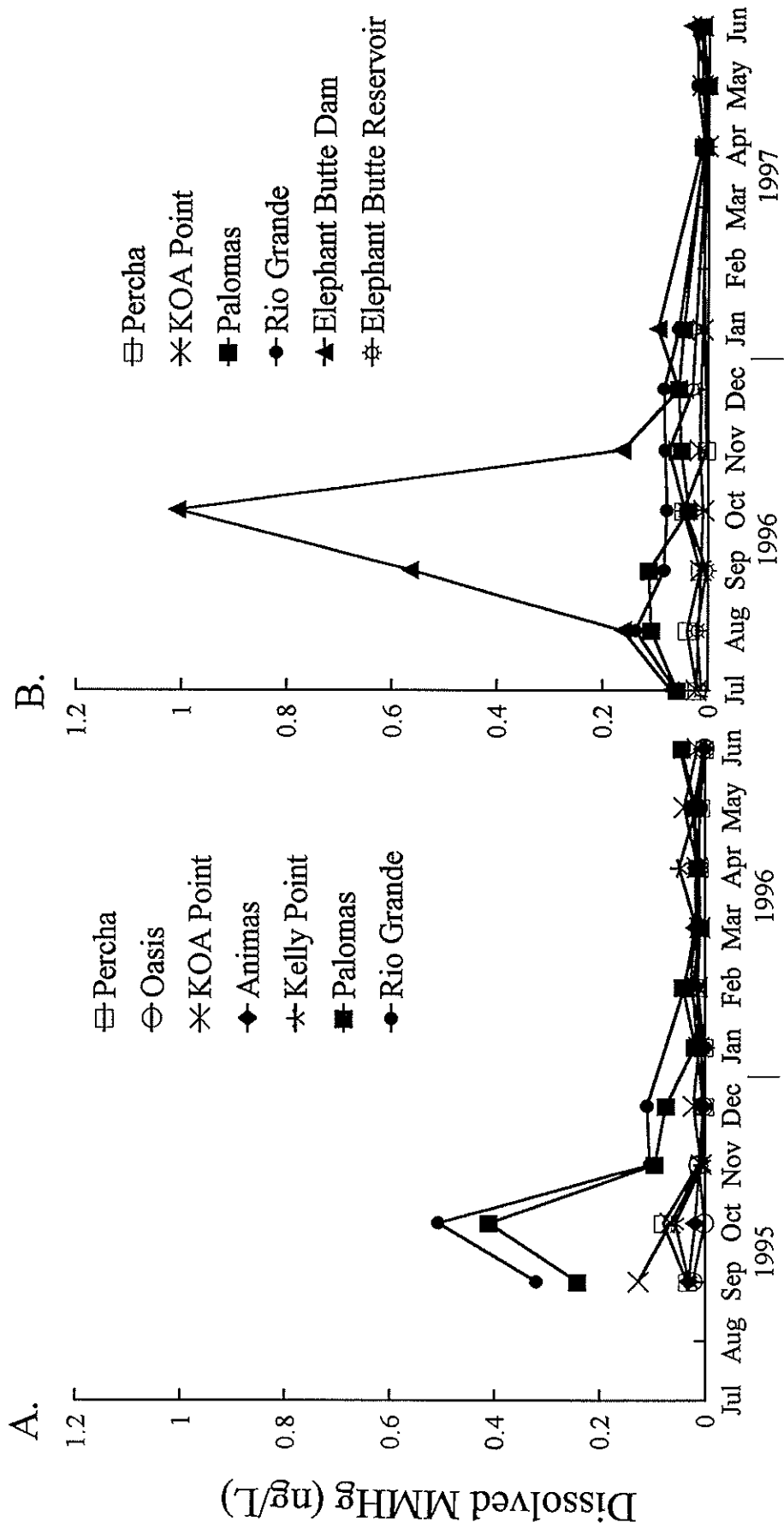
Concentration of total MMHg in water for July 1995 was 0.230 ng/L at Palomas (a site with an extensive shallow littoral area) (Figure 5A). In contrast, the remaining sites located in deep open water ranged from 0.061 ng/L (Animas) to 0.087 ng/L (Percha) (Figure 5A). By August 1995, total MMHg concentrations in water had increased at all the sites (range from 0.449 ng/L at Kelly Point to 0.181 ng/L at Percha). The Rio Grande site had the second highest total MMHg concentrations in water (0.392 ng/L). By October 1995, total MMHg concentrations in water were greatest at the Rio Grande and Palomas sites (0.732 and 0.611 ng/L, respectively) and subsequently decreased to 0.052 ng/L at the Rio Grande site and to 0.074 ng/L at the Palomas site by January 1996. Concentrations of total MMHg at the five remaining sites were at or below the MDL (0.018 ng/L) until June 1996.

Concentrations of total MMHg in water from the Rio Grande increased from 0.110 ng/L in July 1996 to 0.325 ng/L in August 1996 (Figure 5B). Similarly, total MMHg in water from Palomas increased from 0.117 ng/L in July 1996 to 0.355 ng/L in August 1996 (Figure 5B). Both sites remained elevated until April 1997 when total MMHg concentrations decreased an order of magnitude (near or below MDL) until the study concluded in June 1997. In contrast, concentrations of total MMHg in the deep open water of Caballo Reservoir varied only slightly throughout the year, with mean total MMHg concentrations of 0.044 ng/L ( $\pm$  0.035 standard deviation) at KOA Point and a mean total MMHg of 0.070 ng/L ( $\pm$  0.052) at Percha (Figure 5B). Concentrations of total MMHg near Elephant Butte Dam increased 11.4 fold from 0.149 ng/L in July 1996 to 1.695 ng/L by October 1996 (Figure 5B). Despite elevated total MMHg concentrations near Elephant Butte Dam, concentrations of total MMHg in Elephant Butte Reservoir were below the MDL from July 1996 to September 1996. Concentrations of total MMHg from Elephant Butte Reservoir increased to 0.264 ng/L by November 1996, subsequently decreased to 0.05 ng/L by January 1997, and then remained below the MDL until the end of the study.

Water samples for all sites were not filtered July and August 1995, thus dissolved MMHg concentrations are not available for those months (Figure 6A). Concentrations of dissolved MMHg in water from the Palomas site increased from 0.244 ng/L in September 1995 to 0.404 ng/L in October 1995 (Figure 6A). Concentrations of dissolved MMHg in the Rio Grande similarly increased from 0.321 ng/L in September 1995 to 0.508 ng/L in October 1995 (Figure 6A). By January 1996, dissolved MMHg concentrations in water from the Palomas and the Rio Grande sites had decreased to levels comparable to the five sites located in deep water of Caballo Reservoir (from Kelly Point to Percha) and remained low until June 1996.



Figures 5 A and B. A. Temporal trends (1995-1996) for concentrations (ng/L) of total monomethylmercury in water from the Rio Grande and six sites at Caballo Reservoir. B. Temporal trends (1996-1997) for concentrations (ng/L) of total monomethylmercury in water from Elephant Butte Reservoir, Elephant Butte Dam, the Rio Grande and three sites in Caballo Reservoir.



Figures 6 A and B. A. Temporal trends (1995-1996) for concentrations (ng/L) of dissolved monomethylmercury in water from the Rio Grande and six sites at Caballo Reservoir. Water samples were not filtered July and August 1995. B. Temporal trends (1996-1997) for concentrations (ng/L) of dissolved monomethylmercury in water from Elephant Butte Reservoir, Elephant Butte Dam, the Rio Grande, and three sites in Caballo Reservoir.

Although the Rio Grande site was originally selected to represent background Hg concentrations entering the reservoir, elevated concentrations of dissolved MMHg in September and October 1995 reflected a source of Hg contamination upstream of Caballo Reservoir. Water-release from Elephant Butte Dam was shut off from 1 November 1995 to 3 January 1996 and coincided with the decreases in total and dissolved MMHg in the November 1996 sample (Figures 5A and 6A). During this time, however, the Rio Grande entering Caballo Reservoir did not dry up. Water in the river was due to seepage through the dam and through the banks of the river (Personal communication: A. Tow, U.S. Bureau of Reclamation). Once water was again released from Elephant Butte Reservoir in January 1996, total and dissolved MMHg did not increase at the Rio Grande site.

Concentrations of dissolved MMHg in Palomas increased from 0.059 ng/L in July 1996 to 0.112 ng/L in September 1996 (Figure 6B). A similar increase in concentration of dissolved MMHg was observed in the Rio Grande (Figure 6B). By October 1996, dissolved MMHg concentrations had decreased to 0.037 ng/L in Palomas and 0.078 ng/L in the Rio Grande and remained at or below the MDL until the end of the study. Mean concentrations of dissolved MMHg for two sites within Caballo Reservoir from July 1996 to June 1997 was 0.013 ng/L ( $\pm 0.005$ ) for KOA Point and 0.025 ng/L ( $\pm 0.018$ ) for Percha (Figure 6B). Concentrations of dissolved MMHg in Elephant Butte Reservoir were at or below MDL from July 1996 to the end of the study (Figure 6B). In contrast, concentrations of dissolved MMHg near Elephant Butte Dam increased over 14 fold from 0.070 ng/L in July 1996 to 1.008 ng/L in October 1996 (Figure 6B). By December 1996, concentrations of MMHg near Elephant Butte Dam had decreased to 0.056 ng/L and remained at or below MDL until the end of the study.

The elevated MMHg concentrations near Elephant Butte Dam can be explained by considering the vertical profiles of dissolved oxygen concentrations at Elephant Butte Reservoir (Figure 7). As Elephant Butte Reservoir began to stratify July 1996, concentrations of total and dissolved MMHg near Elephant Butte Dam began to increase. In October, prior to mixing, concentrations of total and dissolved MMHg near Elephant Butte Dam were at their maximum. By November 1996, turnover began and mixing of the reservoir was completed by December 1996 (Figure 7). The onset of mixing presumably circulated MMHg (produced in the anoxic zone) into the watercolumn, resulting in an increase in total and dissolved MMHg concentrations in water. Concentrations of total and dissolved MMHg in Elephant Butte Reservoir were at or below MDL from July 1996 to September 1996 and did not reach maxima in Elephant Butte Reservoir until November 1996 after the onset of mixing. Overturn of the Reservoir presumably resulted in the movement of MMHg from the anoxic zone into the watercolumn and resulted in the increase in total and dissolved MMHg in water samples collected from the surface of the Reservoir. Concentrations of MMHg accumulated and were elevated in the anoxic hypolimnion compared to concentrations observed in the epilimnion of meromictic lakes (Cossa et al. 1994; Watras et al. 1995).

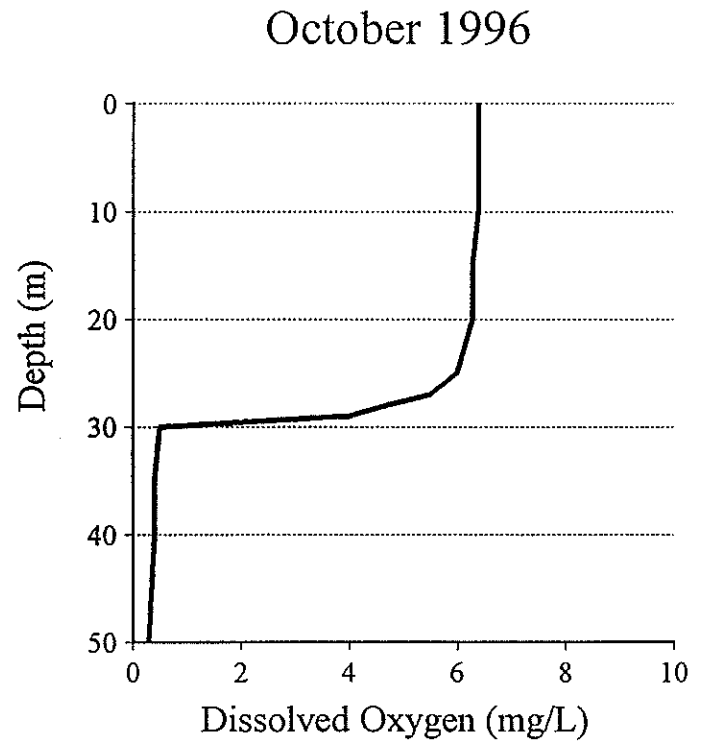
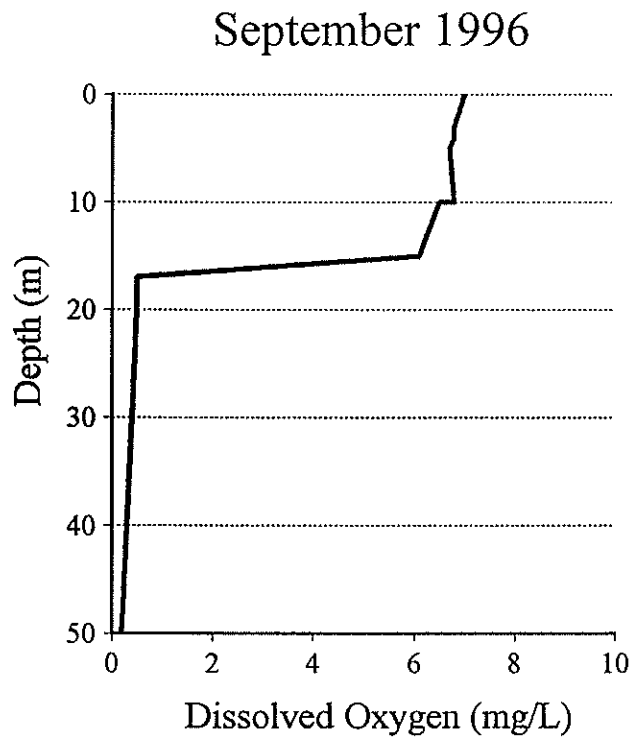
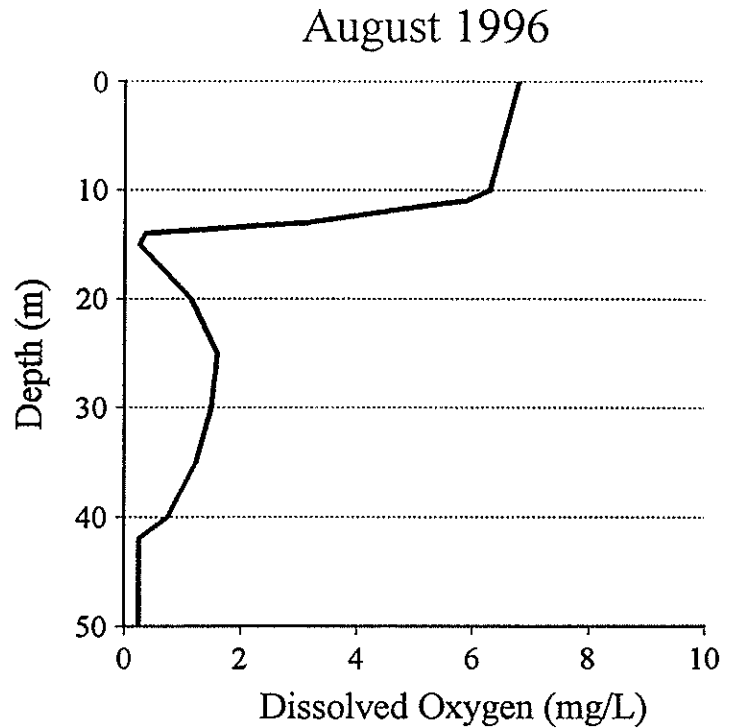
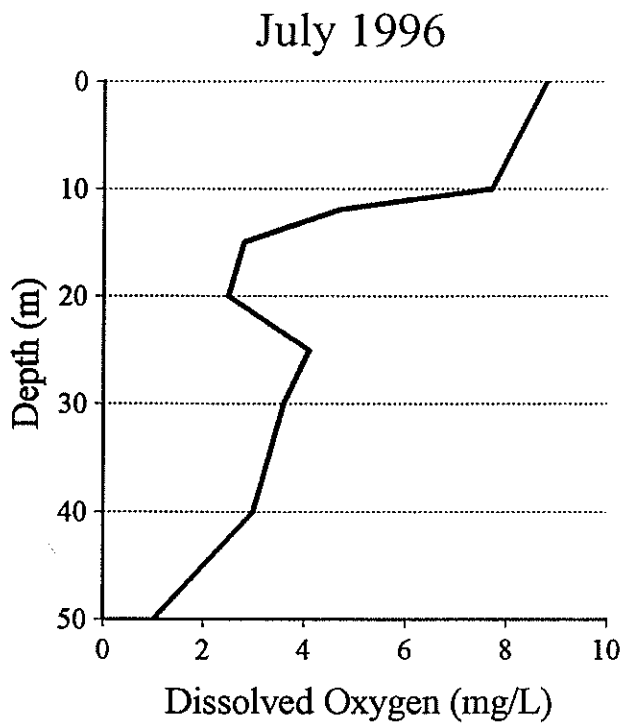


Figure 4. Dissolved oxygen profiles for Elephant Butte Reservoir from July 1996 to October 1996.

A second but related event may have contributed to the elevated concentrations of MMHg near Elephant Butte Dam from September 1996 to October 1996. Hydroelectric power generation at the dam was fully operational in July 1996 with all three turbines in use. By the end of August 1996, power generation was scaled back approximately 30% until 17 September 1996 when power was completely shut off. Power generation was eventually restored 27 January 1997. The Rio Grande did not dry up during those dates; water downstream of the dam was the result of seepage. Unlike water passing through the turbines, seepage water emanates from all levels of the dam including porewater from reservoir sediments. Thus, when Elephant Butte Reservoir stratified, a larger portion of the seepage water may have originated from the sediment layer in the reservoir carrying with it MMHg. Thus, stratification and the onset of mixing, when combined with the shutdown of power generation, could have resulted in elevated concentrations of MMHg downstream of Elephant Butte Dam in October 1996.

From September 1996 through November 1996, concentrations of total and dissolved MMHg were greater downstream of Elephant Butte Dam compared to the Rio Grande and Palomas sites, which are 16 km and 21 km downstream from the dam, respectively. Throughout the remainder of the year (January to June 1997), concentrations of total and dissolved MMHg were similar near Elephant Butte Dam, the Rio Grande, and Palomas sites. Although the residence time for water between Elephant Butte Dam and Palomas is less than one day, the reduced concentrations of MMHg may have been the result of a transformation of MMHg to other Hg species, MMHg becoming bound to particulates, removal by the biota, or a combination of these factors.

#### **Transport of THg and MMHg in Runoff Water From Two Drainages**

Concentrations of Hg were analyzed in water samples collected from Cuchillo Negro Creek that enters the Rio Grande between Elephant Butte Reservoir, and from Palomas Creek that enters near the Palomas site in Caballo Reservoir. The creeks were dry until the monsoon season from July 1996 to August 1996 when water drains off the Black Range into its tributaries. Mercury concentrations were initially elevated and subsequently decreased in a series of runoff events (Table 5). The timing between sample collection and when the creeks began flowing is not known. Although the data can not be used to quantitatively characterize export of Hg from the watershed, it does indicate that both THg and MMHg were mobilized from the watershed where it eventually entered the Rio Grande.

Table 5. Mercury concentrations (ng/L) in runoff from two tributaries.

	Date	THg (un-filtered) <sup>a</sup>	THg (filtered) <sup>b</sup>	Total MMHg	Dissolved MMHg
Cuchillo Negro Creek	7/12/96	49.5	6.13	0.287	0.128
	7/17/96	31.1	5.85	0.227	0.230
	7/22/96	4.01	3.23	0.354	0.136
Palomas Creek	7/15/96	80.0	6.13	0.510	0.097
	7/17/96	8.44	3.82	0.114	0.064

<sup>a</sup>THg (unfiltered) represents all species of mercury in unfiltered water containing Hg bound to particulate and dissolved in water.

<sup>b</sup>THg (filtered) represents all species of mercury that is either dissolved or bound to particulate that can pass through a 0.2- $\mu$ m filter.

### Temporal and Spatial Trends for Water Quality

Convictional circulation in combination with wind resulted in relatively little vertical variation of chemical and physical characteristics, which may be typical of warm polymictic lakes similar to Caballo Reservoir. Very few consistent trends were observed in the data for physical and chemical characteristics measured in vertical profiles and surficial samples for all sites. The average monthly vertical profile for conductivity reflected the greatest differences at the Palomas site compared to the remaining sites in Caballo Reservoir for both years (Appendix Table 2). The elevated conductivity at the Palomas site compared to the remaining sites may have been due to the shut-down of hydroelectric power generation at Elephant Butte Dam to allow dredging of the river channel upstream of the Rio Grande site. The high conductivity might also have been due to seepage of water containing minerals from the soil below the dam and into the river. The concentration of total suspended solids, pH, and temperature, similarly, reflected the greatest differences at the Palomas site compared to the remaining sites in Caballo Reservoir for both years (Appendix Tables 3, 5, 7, respectively). The Palomas site was shallow (0.25-5.0 m) and resulted in greater wind-induced suspension of particulate matter resulting in higher concentrations of TSS. Monthly vertical profiles for pH were lower in Palomas compared to the five sites for the entire study (Appendix Table 5). Monthly vertical profiles for temperatures were slightly lower in Palomas compared to the remaining sites and may be due to cooler hypolimnetic or seepage water (7-15 °C) which flowed directly into the Palomas site via the Rio Grande from Elephant Butte Reservoir (Appendix Table 7).

Dissolved oxygen concentrations in July, August, and September 1995 and June, July, and August 1996 reflected stratification of Caballo Reservoir at all sites except for the Palomas site (Appendix Table 9).



The stratification may have been related to differences in specific gravity and not density differences related to temperature (Personal communication: R.A. Cole, New Mexico State University). Dissolved oxygen concentrations were greatest and nearly constant (80-100% saturation) within 5 to 7 m from the surface at Caballo Reservoir and then decreased to <1.0 mg/L to the bottom. Despite the presence of this nearly anoxic layer, which promotes methylation activity, the ratio of MMHg:THg in sediment throughout the reservoir were much less than the Palomas site. Monthly hardness concentrations were greatest at the Palomas site compared to the remainder of the sites in Caballo Reservoir from July 1995 to December 1995 and were repeated from June 1996 to December 1996 (Appendix Table 11). Similarly, monthly total alkalinity concentrations were greatest for the Palomas site on the same dates as above for total hardness (Appendix Table 13).

## SUMMARY

Total and MMHg concentrations in sediments suggest a substantial amount of Hg was added at the northern end of Caballo Reservoir as a result of the Bonner fire and subsequent late-summer rains. The two events may have had a two-fold effect on Hg concentrations observed at the Palomas site. The fire followed by rain may not only have brought increased allochthonous organic matter into the reservoir but may have also mobilized and transported Hg from the watershed into the reservoir. August 1995 (one month after the fire) was the only month with elevated total and dissolved MMHg concentrations in water at all sites. It is widely accepted that both wet and dry deposition can be a significant source of Hg to aquatic environments (Bloom and Watras 1989; Porcella 1994), and a watershed as large as the Black Range could receive a substantial amount of Hg. In addition, the east side of the Black Range contains numerous abandoned gold and silver mines from which Hg might leach into the watershed during rain events. Runoff containing allochthonous material can influence Hg transport from the watershed as well (Lee and Iverfeldt 1991; Mierle and Ingram 1991; Johansson and Iverfeldt 1994).

From September 1995 to December 1995, total and dissolved MMHg concentrations were elevated at both the Rio Grande and Palomas sites, indicating potential transport from upriver. In June 1996, two sites were added upstream and downstream of Elephant Butte Dam. Elephant Butte Reservoir is characteristic of a modified dimictic lake that began stratification in August 1996 and began mixing November 1996. Prior to mixing, however, the greatest concentrations of total and dissolved MMHg were recorded near the Dam downstream of Elephant Butte Reservoir, while concentrations of total and dissolved MMHg in the Reservoir were at or the MDL. By November 1996, Elephant Butte Reservoir had undergone mixing, resulting in the dispersal of MMHg into the water column and elevated concentrations of total and dissolved MMHg in the reservoir. With the shutdown of hydroelectric power generation at the dam, water in the Rio Grande downstream of the dam was the result of seepage through and beneath the dam originating from the sediment layer in the reservoir and carrying with it MMHg. Thus, stratification and the onset of mixing of Elephant Butte Reservoir, when combined with the shutdown of hydroelectric power generation, may have resulted in elevated concentrations of MMHg observed below Elephant Butte Dam in October 1996.

Additional research is needed to describe Hg concentrations in runoff from the Black Range watershed entering the Rio Grande and Caballo Reservoir. During the summer rains of July 1996 and August 1996, THg and MMHg were mobilized out of the Cuchillo Negro and Palomas watersheds into the Rio Grande. Total Hg and MMHg should also be determined in wet and dry deposition in the vicinity of Caballo Reservoir. This would not only provide an estimate of the amount and species of Hg entering the reservoir, but it would also provide background data to correlate with watershed runoff. Additional research is also needed to determine the influence of Elephant Butte Reservoir on the Hg cycle in Caballo Reservoir. This should include continued

collections of total and dissolved MMHg in the Rio Grande upstream of Elephant Butte Reservoir, within the reservoir near the dam, and in the Rio Grande downstream of the dam. Sample collections within the reservoir should continue to include vertical profiles of pH, dissolved oxygen, temperature, and conductivity. In addition, alkalinity, hardness, total suspended solids, organic carbon (total and dissolved), and color should be determined in the reservoirs and in the Rio Grande to assist in describing the biogeochemical cycle of Hg in this arid landscape.

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**APPENDIX**

**Water Quality Summaries (July 1995 - July 1997)**

Table 1. Average monthly conductivity ( $\mu\text{mhos/cm}$ ) from vertical profiles in six sites within Caballo Reservoir from July 1995 to June 1996. Ranges are in parenthesis.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	525 (525)	636 (609-651)	648 (648)	650 (648-651)	1,050 (1,050)	986 (968)	574 (574)	560 (560)	592 (591-593)	602 (602)	586 (584-588)	665 (665)
Kelly Point	555 (525-575)	588 (566-626)	610 (594-628)	658 (655-660)	709 (696-743)	711 (709-713)	684 (679-696)	608 (602-614)	618 (616-619)	631 (631)	620 (619-622)	580 (579-585)
Animas	557 (525-575)	582 (572-603)	598 (583-615)	630 (628-642)	650 (646-657)	683 (681-685)	692 (688-697)	635 (632-647)	645 (635-653)	648 (646-649)	642 (640-643)	617 (609-628)
KOA Point	558 (525-575)	577 (564-584)	585 (580-591)	630 (628-640)	648 (645-653)	668 (668)	683 (682-685)	648 (646-650)	653 (650-662)	650 (649-651)	653 (649-668)	625 (620-627)
Oasis	575 (575)	576 (574-584)	§	625 (621-629)	645 (645)	663 (662-664)	686 (685-687)	651 (650-653)	653 (648-659)	650 (650)	661 (659-662)	629 (626-630)
Percha	575 (575)	578 (574-587)	§	625 (624-626)	642 (641-643)	661 (660-662)	687 (686-689)	656 (655-659)	657 (656-659)	650 (650)	665 (662-670)	630 (626-632)

§ Data collection was prohibited by bad weather.

Table 2. Monthly conductivity ( $\mu\text{mhos/cm}$ ) from surface water samples collected from three sites within Caballo Reservoir and from one site within Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	a	637	1390	1790	400	410	440	§	§	b	638	b
KOA Point	a	§	660	630	780	b	950	§	§	600	664	b
Percha	a	547	610	720	700	b	990	§	§	560	663	b
Elephant Butte Reservoir	a	554	b	660	b	b	b	§	§	500	628	b

§ Data collection was prohibited by bad weather.

a Instrument failure.

b Error in analysis.

Table 3. Concentrations of total suspended solids ( $\mu\text{g/L}$ ) in surface water samples collected monthly from six sites in Caballo Reservoir from July 1995 to June 1996.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	35	45	44	19	12	46	25	a	5	7	4	45
Kelly Point	1	6	6	15	10	20	29	16	11	16	<1	11
Animas	3	2	15	9	5	8	27	15	10	9	<1	10
KOA Point	6	8	6	9	12	12	27	14	7	9	<1	6
Oasis	4	6	5	8	9	12	25	11	12	9	<1	7
Percha	3	9	3	8	11	44	26	14	7	8	<1	7

a Error in analysis.

Table 4. Concentrations of total suspended solids ( $\mu\text{g/L}$ ) in surface water samples collected monthly from three sites in Caballo Reservoir and from one site in Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1996	August 1996	September 1996	October 1996	November 1996	December 1996	January 1997	February 1997	March 1997	April 1997	May 1997	June 1997
Percha	8	8	24	47	-	a	12	§	§	13	5	10
KOA Point	8	a	30	82	-	a	10	§	§	20	11	13
Palomas	3	9	3	8	-	12	26	§	§	20	12	12
Elephant Butte Reservoir	3	a	4	4	-	<1.0	<1.0	§	§	2	9	3

§ Data collection was prohibited by bad weather.

a Error in analysis.

- Data were not collected.

Table 5. Mean monthly pH for vertical profiles from six collection sites within Caballo Reservoir from July 1995 to June 1996. Ranges are in parenthesis.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	8.2 (*)	7.9 (7.7-8.4)	7.6 (7.6)	7.6 (7.5-7.8)	8.2 (8.1-8.3)	7.9 (7.9)	8.2 (8.2)	8.4 (8.4)	8.1 (8.1)	7.7 (7.7)	7.8 (7.8)	7.7 (7.7)
Kelly Point	8.4 (*)	8.2 (7.3-8.8)	8.4 (8.0-8.6)	8.3 (8.1-8.5)	8.2 (8.1-8.3)	8.3 (8.3)	8.4 (8.4)	8.2 (8.2)	8.5 (8.3-8.6)	8.2 (8.2)	8.6 (8.4-8.6)	8.6 (8.6)
Animas	8.4 (*)	8.1 (7.3-8.7)	8.2 (8.0-8.5)	8.3 (8.1-8.5)	8.2 (8.2)	8.3 (8.3)	8.4 (8.4)	8.3 (8.3)	8.5 (8.4-8.6)	8.1 (8.1)	8.3 (7.9-8.5)	8.2 (7.6-8.5)
KOA Point	7.8 (*)	8.2 (7.4-8.8)	8.3 (7.9-8.6)	8.2 (8.0-8.4)	8.2 (8.2)	8.2 (8.2)	8.5 (8.5)	8.3 (8.3)	8.5 (8.4-8.6)	8.1 (8.0-8.2)	8.3 (7.9-8.5)	8.2 (7.6-8.5)
Oasis	8.4 (*)	8.3 (7.6-8.7)	§	8.2 (8.1-8.3)	8.2 (8.1-8.3)	8.2 (8.2)	8.5 (8.5)	8.3 (8.3)	8.5 (8.4-8.6)	8.0 (8.0)	8.3 (8.2-8.4)	8.3 (8.6-7.7)
Percha	8.4 (*)	8.3 (7.7-8.7)	§	8.2 (8.0-8.3)	8.3 (8.3)	8.3 (8.3)	8.5 (8.5)	8.3 (8.3)	8.5 (8.5)	8.0 (8.0)	8.0 (7.7-8.2)	8.3 (8.5-8.0)

\* A vertical pH profile was not collected. The data point represents one sample collected mid-depth.

§ Data collection was prohibited by bad weather.

Table 6. pH in surface water samples collected monthly from three sites in Caballo Reservoir and from one site in Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1996	August 1996	September 1996	October 1996	November 1996	December 1996	January 1997	February 1997	March 1997	April 1997	May 1997	June 1997
Palomas	8.2	8.0	8.7	8.2	8.2	8.0	7.8	§	§	8.2	8.3	8.0
KOA Point	8.6	-	-	8.2	8.4	-	-	§	§	8.3	8.3	8.4
Percha	8.2	8.7	8.7	8.2	8.2	8.0	7.8	§	§	8.2	8.3	8.0
Elephant Butte Reservoir	8.4	8.6	-	8.3	8.1	8.1	8.9	§	§	8.1	8.5	8.2

§ Data collection was prohibited by bad weather.

- Data were not collected.

Table 7. Range of monthly temperature (°C) from vertical profiles for six sites within Caballo Reservoir from July 1995 to June 1996.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	21-22	21-27	17-18	17	10	9	8	10	10	10	12	14
Kelly Point	21-26	23-28	20-23	15-17	11-12	9-10	7-8	10	13	16	20-21	21
Animas	21-26	24-29	21-23	16-18	12	9-10	7	10-11	13	16	21-22	18-22
KOA Point	21-27	24-28	22-23	16-18	12-13	9-10	7	10-11	12-13	16	19-22	20-22
Oasis	20-27	24-27	§	16-17	12-13	10	7	10-12	12-13	16-17	20-22	20-22
Percha	20-28	24-27	§	16-17	12-13	10	7	10-12	12-13	15-17	18-21	21-22

§ Data collection was prohibited by bad weather.



Table 8. Range of monthly temperatures (°C) from vertical profiles for three sites in Caballo Reservoir and from one site in Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1996	August 1996	September 1996	October 1996	November 1996	December 1996	January 1997	February 1997	March 1997	April 1997	May 1997	June 1997
Palomas	-	17.5	21.0	13.0	12.0	7.0	12.0	§	§	12.0	14.0	13.5
KOA Point	20.0-25.0	-	21.0	13.0	10.0	-	5.25	§	§	11.5	15-16.5	18-20
Percha	22.0-26.0	23.0-24.0	17.5-19.0	13.5	9.0	-	5.5	§	§	11.5-12.0	14.0-16.5	17.0-20.0
Elephant Butte Reservoir	13.0-25.0	13.0-24.0	13.0-21.5	13.0-16.0	13.0	11.0	8.0	§	§	8.0-12.0	10.0-17.0	13.0-21.0

§ Data collection was prohibited by bad weather.

- Data were not collected.

Table 9. Range of dissolved oxygen concentrations (mg/L) from monthly vertical profiles for six sites in Caballo Reservoir from July 1995 to June 1996. Maximum depth (m) is in parenthesis.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	5.6-5.9 (5)	6.3-10.4 (1.5)	5.8 (1.5)	5.5-5.7 (1.0)	7.7-7.9 (1.0)	8.6-8.9 (0.5)	10.6-13.5 <sup>B</sup> (2.0)	8.4-9.8 (3.0)	8.0-8.8 (1.5)	7.5 (1.0)	10.8-11.2 <sup>B</sup> (1.5)	7.5 (0.25)
Kelly Point	<1.0-11.8 <sup>A,B</sup> (10.0)	<1.0-13.8 <sup>A,B</sup> (6.5)	3.6-10.2 <sup>A,B</sup> (6.0)	6.6-8.1 (6.0)	8.0-8.6 (5.0)	7.5-8.9 (5.0)	9.0-12.6 <sup>B</sup> (6.5)	9.2-9.3 (6.0)	7.9-10.2 <sup>B</sup> (6.0)	6.5-8.4 (5.0)	8.4-8.7 (4.0)	9.7-10.4 (3.0)
Animas	<1.0-10.4 <sup>A,B</sup> (12.0)	<1.0-11.0 <sup>A,B</sup> (7.5)	2.6-9.3 <sup>A</sup> (8.0)	4.8-8.4 (8.0)	8.0-8.8 (7.5)	7.6-8.8 (7.0)	8.8-10.8 <sup>B</sup> (9.0)	8.5-9.8 (8.0)	7.4-9.0 (8.0)	7.5-9.2 (7.0)	8.1-10.5 <sup>B</sup> (6.0)	<1.0-8.6 <sup>A</sup> (6.0)
KOA Point	<1.0-11.8 <sup>A,B</sup> (15.0)	<1.0-14.2 <sup>A,B</sup> (11.0)	<1.0-10.4 <sup>A,B</sup> (13.0)	3.0-8.5 (9.5)	7.9-8.6 (10.0)	7.7-9.6 (10.0)	8.4-10.6 <sup>B</sup> (9.0)	8.0-9.7 (13.0)	7.9-8.3 a	5.9-9.0 (11.5)	8.6-9.7 (8.0)	1.2-8.0 (7.5)
Oasis	<1.0-12.4 <sup>A,B</sup> (17.0)	<1.0-10.7 <sup>A,B</sup> (16.0)	§	5.9-7.8 (9.5)	8.0-8.5 (13.5)	7.5-9.8 (10.0)	8.3-10.3 <sup>B</sup> (10.0)	7.5-9.4 (16.0)	8.4-8.6 a	3.9-8.6 (16.0)	8.6-8.8 (13.0)	<1.0-8.0 <sup>A</sup> (11.5)
Percha	<1.0-13.2 <sup>A,B</sup> (16.0)	<1.0-10.0 <sup>A,B</sup> (14.0)	§	4.5-7.8 (12.0)	7.9-9.0 (12.0)	7.0-9.4 (10.0)	8.1-10.1 <sup>B</sup> (10.0)	10.0-12.0 <sup>B</sup> (16.0)	9.0-9.7 a	6.5-9.5 (12.5)	7.9-8.7 (10.0)	4.9-7.8 (10.0)

§ Data collection was prohibited by bad weather.

<sup>A</sup> Profiles reflected stratification of dissolved oxygen concentrations; concentrations were greatest and nearly constant (80-100% saturation) within 5 - 7 m from surface, then decreased to <1.0 mg/L to bottom.

<sup>B</sup> Dissolved oxygen concentrations representative of supersaturation within 1.0 m of surface.  
a Equipment failure prevented depth determination.

Table 10. Range of dissolved oxygen concentrations (mg/L) for monthly vertical profiles for three sites in Caballo Reservoir and from one site in Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1996	August 1996	September 1996	October 1996	November 1996	December 1996	January 1997	February 1997	March 1997	April 1997	May 1997	June 1997
Palomas	-	8.8	9.2	10.4	10.2	10.6	9.2	§	§	10.8	10.8	a
KOA Point	2.5-10.8 (-)	-	-	8.6	10.0 (3)	-	11.4 (3)	§	§	9.0 (5)	3.0-7.8 (6)	4.5-7.9 (6)
Percha	1.8-7.5 (-)	<1.0-6.4 <sup>A</sup> (-)	-	7.5	8.4-9.0 (5)	-	11.4 (5)	§	§	8.9-9.4 (7.5)	5.3-6.0 (8)	5.6-7.9 (8)
Elephant Butte Reservoir	3.0-8.8 (50)	<1.0-6.8 <sup>A</sup> (50)	<1.0-6.8 <sup>A</sup> (50)	<1.0-6.4 <sup>A</sup> (50)	7.4-6.2 (50)	3.5-8.5 (50)	8.9-9.8 (50)	§	§	6.0-9.5 (50)	<1.0-10.8 <sup>A</sup> (50)	5.4-7.4 (50)

§ Data collection was prohibited by bad weather.

<sup>A</sup> Profiles reflected stratification of dissolved oxygen concentrations, concentrations were greatest and nearly constant (80-100% saturation) within 5 - 7 m from surface, then decreased to <1.0 mg/L to bottom. a Equipment failure prevented dissolved oxygen determination.

- Data were not collected.

Table 11. Concentrations of hardness (mg CaCO<sub>3</sub>/L) in surface water samples collected monthly from six sites in Caballo Reservoir from July 1995 to June 1996.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	163	213	175	181	248	226	156	*	160	155	155	162
Kelly Point	144	164	150	178	209	203	187	169	167	168	147	147
Animas	142	159	140	163	179	194	178	176	166	166	159	148
KOA Point	163	179	150	168	181	196	130	183	171	167	162	158
Oasis	146	155	144	182	175	200	176	175	174	170	163	179
Percha	127	159	143	170	170	210	157	187	167	172	167	158

\*Missing data due to error in analysis.

Table 12. Concentrations of hardness (mg CaCO<sub>3</sub>/L) in surface water samples collected monthly from three sites in Caballo Reservoir and one site in Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1996	August 1996	September 1996	October 1996	November 1996	December 1996	January 1997	February 1997	March 1997	April 1997	May 1997	June 1997
Palomas	157	165	196	335	322	330	161	§	§	158	160	159
KOA Point	123	-	191	190	210	-	252	§	§	162	174	163
Percha	123	132	167	197	200	-	*	§	§	159	158	164
Elephant Butte Reservoir	142	145	154	145	152	157	197	§	§	155	170	154

§ Data collection was prohibited by bad weather.

\* Error in analysis.

- Data were not collected.

Table 13. Monthly total alkalinity (mg CaCO<sub>3</sub>/L) from six collection sites in Caballo Reservoir from July 1995 to June 1996.

Site	July 1995	August 1995	September 1995	October 1995	November 1995	December 1995	January 1996	February 1996	March 1996	April 1996	May 1996	June 1996
Palomas	152	154	158	129	121	136	116	*	125	122	128	129
Kelly Point	125	130	139	131	131	130	126	123	131	134	123	110
Animas	129	123	146	122	125	130	118	128	133	140	129	118
KOA Point	148	123	133	120	125	129	128	135	136	133	131	125
Oasis	128	123	136	*	123	124	128	128	139	142	133	124
Percha	129	124	132	125	124	126	129	136	140	137	134	124

\*Missing data due to error in analysis.

Table 14. Concentrations of alkalinity (mg CaCO<sub>3</sub>/L) in surface water samples collected monthly from three sites in Caballo Reservoir and one site in Elephant Butte Reservoir from July 1996 to June 1997.

Site	July 1996	August 1996	September 1996	October 1996	November 1996	December 1996	January 1997	February 1997	March 1997	April 1997	May 1997	June 1997
Palomas	131	134	170	159	165	166	144	§	§	117	128	133
KOA Point	98	-	154	179	146	-	106	§	§	126	143	140
Percha	99	106	129	143	147	-	124	§	§	130	131	138
Elephant Butte Reservoir	118	113	122	124	127	128	132	§	§	109	129	127

§ Data collection was prohibited by bad weather.

- Data were not collected.