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Anthropogenic tracers, endocrine disrupting chemicals, and endocrine disruption in Minnesota lakes

Jeffrey H. Writer [a], Larry B. Barber [a,*], Greg K. Brown [a], Howard E. Taylor [a], Richard L. Kiesling [b], Mark L. Ferrey [c], Nathan D. Jahns [d], Steve E. Bartell [d], Heiko L. Schoenfuss [d]

[a] U.S. Geological Survey, 3215 Marine Street, Boulder, CO 80303, United States
[b] U.S. Geological Survey, 2280 Woodale Drive, Mounds View, MN 55112, United States
[c] Minnesota Pollution Control Agency, 520 Lafayette Road, St. Paul, MN 55155, United States
[d] St. Cloud State University, WSB-273, 720 Fourth Avenue South, St. Cloud, MN 56301, United States

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ABSTRACT

Concentrations of endocrine disrupting chemicals and endocrine disruption in fish were determined in 11 lakes across Minnesota that represent a range of trophic conditions and land uses (urban, agricultural, residential, and forested) and in which wastewater treatment plant discharges were absent. Water, sediment, and passive polar organic integrative samplers (POCIS) were analyzed for steroidal hormones, alkylphenols, bisphenol A, and other organic and inorganic molecular tracers to evaluate potential non-point source inputs into the lakes. Resident fish from the lakes were collected, and caged male fathead minnows were deployed to evaluate endocrine disruption, as indicated by the biological endpoints of plasma vitellogenin and gonadal histology. Endocrine disrupting chemicals, including bisphenol A, 17β-estradiol, estrone, and 4-nonylphenol were detected in 90% of the lakes at part per trillion concentrations. Endocrine disruption was observed in caged fathead minnows and resident fish in 90% of the lakes. The widespread but variable occurrence of anthropogenic chemicals in the lakes and endocrine disruption in fish indicates that potential sources are diverse, not limited to wastewater treatment plant discharges, and not entirely predictable based on trophic status and land use.

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1. Introduction

Evidence of contamination of surface waters by trace concentrations of consumer product chemicals (Kolpin et al., 2002) has raised awareness about the connection between water supplies and anthropogenic activities. One of the potential consequences of exposure to these chemicals is endocrine disruption (Colborn and Thayer, 2000; McLachlan, 2001; Milnes et al., 2006), specifically reproductive impairment in fish from streams receiving wastewater treatment plant (WWTP) effluents (Jobling et al., 1998; Barber et al., 2007; Vajda et al., 2008). Most research on endocrine disrupting chemicals in the aquatic environment has focused on rivers susceptible to point-source influences such as WWTP discharges (Kolpin et al., 2002; Barber et al., 2006), surface waters influenced by non-point sources (Standley et al., 2000; Wilkinson et al., 2002), drinking water sources (Focazio et al., 2008), and groundwater (Swartz et al., 2006; Barnes et al., 2008; Barber et al., 2009). Endocrine disruption in lakes has received less attention (Rosen et al., 2006; Bogdal et al., 2009), but a lake dosing experiment in Canada showed that exposure to low ng/L concentrations of the synthetic estrogen 17α-ethynylestradiol adversely affected fish populations (Kidd et al., 2007; Palace et al., 2009). In addition to point sources such as WWTP discharges, contaminants can enter lakes from a variety of non-point sources, including land-applied biosolids, stormwater runoff, row-crop production, animal feeding operations, onsite wastewater disposal systems (which include septic systems), recreational activities, transportation, and atmospheric deposition.

The purpose of this study was to evaluate potential endocrine disruption in lakes across Minnesota representing a range of trophic conditions and land uses (urban, agricultural, residential, and forested), but that do not have WWTP discharges. Environmental concentrations of targeted endocrine disrupting chemicals and corresponding endocrine disruption biomarkers in fish were determined. The targeted chemicals were steroidal hormones, alkyphenols, and bisphenol A, which are generally the dominant estrogenic compounds in WWTP effluent (Desbrow et al., 1998; Barber et al., 2000; Vajda et al., 2008). Additional non-estrogenic organic and inorganic molecular tracers were used to characterize watershed influences on the lakes. The hypotheses tested were: (1) concentrations of endocrine disrupting chemicals should be higher in eutrophic than oligotrophic lakes as a result of greater anthropogenic influences, and (2) fish endocrine disruption (as measured by plasma

* Corresponding author. 3215 Marine Street, Boulder, CO 80303, United States. Tel.: +1 303 541 3039.
E-mail address: lbbarber@usgs.gov (L.B. Barber).

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vitellogenin and testicular histology) will be greater in eutrophic than oligotrophic lakes.

2. Methods and materials

2.1. Site selection and characterization

The Lake Survey Program (Minnesota Department of Natural Resources, 2010) was used to obtain information on the characteristics of lakes across Minnesota, 11 of which were selected for this study based on size (0.3–100 km^2), geographic distribution, trophic status, and land use (Fig. 1, Table 1). Land-use information was obtained at a spatial resolution of 30 m and processed using ArcGIS 9.1 (Environmental Research Systems Institute, Redlands, CA). Land use was classified using Landsat Thematic Mapper imagery as part of the US Geological Survey (USGS) Gap Analysis Program (USGS, 2010). Satellite imagery (Google Earth, Mountain View, CA) was used to estimate onsite wastewater disposal system density by counting residences located within ~0.1 km of the shoreline, and to estimate dock density.

This study focused on eutrophic, mesotrophic, and oligotrophic lakes (Carlson, 1977) that do not have direct WWTP effluent discharges (Table 1). Lake Owasso and Cedar Lake are eutrophic and surrounded by urban land use (residences are connected to a sewer system). Budd Lake is eutrophic and surrounded by mixed urban (residences connected to a sewer system) and agricultural land use. Sullivan Lake is eutrophic, surrounded by agricultural land use, and has high shoreline residential density (onsite wastewater disposal systems). Red Sand Lake is eutrophic, surrounded by forested land use, and has moderate shoreline residential density. Shingobee Lake is mesotrophic, surrounded by forested land use, and has low shoreline residential density. White Sand Lake is mesotrophic, surrounded by forested land use, and has high shoreline residential density (onsite wastewater disposal systems). Stewart Lake and Kabetogama Lake and are mesotrophic, lakes surrounded by forested land use, and have low shoreline residential density (onsite wastewater disposal systems). Northern Light Lake and Elk Lake are oligotrophic, surrounded by forested land use, and have no shoreline residential development. However, Elk Lake is located in an area that receives approximately 500,000 recreational visitors a year (Danae Fritz, Itasca State Park, Minnesota Department of Natural Resources, personal communication, June 11, 2010). All of the lakes are accessible by road and have recreational access points.

2.2. Sample collection

The 2008 field program concurrently collected water, sediment, and fish samples based on a one-time sampling event at each lake. Fish sampling was timed so that populations in each lake were at similar levels of biological development, (which is controlled in part by water temperature), and began in southern Minnesota in June and moved northward with the most northern site being sampled in October (Table 2). All fish sampling occurred in the littoral zone, and sites were selected to be consistent between lakes with respect to fish habitat. Grab water samples were collected from a dock or a boat ~10 m offshore using a stainless steel bucket, split into separate bottles for subsequent chemical analyses, and preserved following established protocols (USGS, 2008). Water samples for major and trace element analysis were filtered through 0.45 µm membranes, collected in acid rinsed polyethylene bottles, and preserved with nitric acid. Unfiltered water samples for organic analysis were collected in cleaned and burned amber glass bottles. Samples for carboxylic acid compound analysis were preserved with 1% (v/v) formalin. All water samples were stored at 4 °C until analysis.

Polar organic compound integrative samplers (POCIS; Alvarez et al., 2004) were obtained from Environmental Sampling Technologies (St. Joseph, MO) and deployed at a depth of ~1 m for 21 days at the same sites where water samples were collected. The deployment apparatus (one per lake) consisted of three sets of membranes containing a sequestration medium (OASIS HLB, Waters, Medford, MA). The above mentioned water samples were collected half-way through the POCIS deployment (Table 2). At lakes where a private dock was not available as an attachment site for the POCIS (Cedar, Red Sand, White Sand, Shingobee, Stewart, Elk, and Northern Light), a steel post was driven into the lake bed at the same location water samples were collected. To avoid potential tampering with the caged

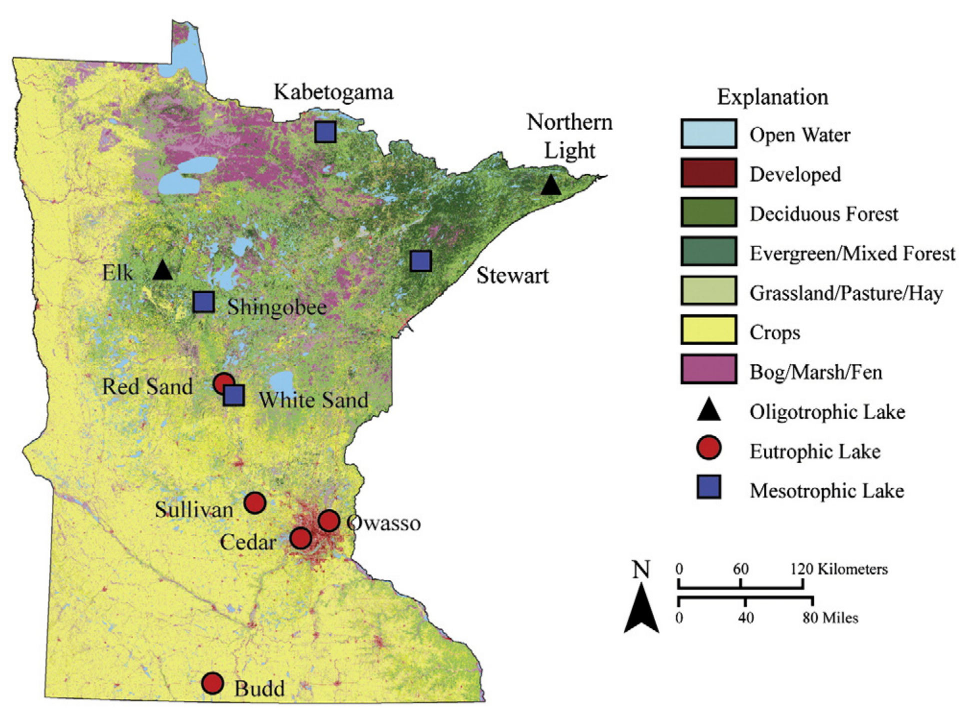

Fig. 1. Landcover and location of Minnesota lakes sampled during 2008.