The relative roles of point and nonpoint phosphorus sources in the eutrophication of Lake Champlain as recorded in sediment cores

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ABSTRACT

Thirteen sediment cores from Lake Champlain (Vermont-New York- Quebec) were analyzed for sediment accumulation rate and eleven indicators of trophic status to reconstruct eutrophication history and relate it to phosphorus inputs from soil erosion, urban point sources, and agricultural runoff including P extracted from phosphate ore and brought to the catchment in fertilizer and livestock feed. Although the catchment was severely deforested and grazed by sheep and cattle in the 19th century, ten of thirteen sites accumulated excess sediment only in the 20th century, a consequence of slow sediment transport through the catchment. Of the three sites that received extra sediment, one remained oligotrophic, while two exhibited mild eutrophication. All sites underwent eutrophication in the 20th century. Three reached their current trophic status before agriculture's intensification, while point-source P input was maximal (1950–1980), four stayed in the reference state until agriculture intensified in the 1970s, and six responded incrementally first to point and then to agricultural P input. Pigment accumulation rates diminished at the surface of seven cores, evidence that the 1990s expansion of P removal from point sources reduced phytoplankton biomass. Additional water quality improvement depends on P management at the catchment border to balance inputs and outputs, and thus avoid P accumulation in soil that saturates phosphate sorption capacity, increasing the proportion of runoff P that is highly-bioavailable orthophosphate. Erosion control is of lower priority because the P native to the catchment's soil is of relatively low bioavailability, and retention structures do not retain dissolved P.

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Introduction

Eutrophication brought on by human activity continues to expand globally in freshwater despite increasingly aggressive management of phosphorus point and nonpoint sources (ILEC, 1994; US EPA, 2010). Where point sources contribute most excess P, eutrophication has been slowed, even reversed, through sewage diversion, phosphate precipitation from wastewater, and restrictions on the P content of detergents (Guilizzoni et al., 2012; Schindler and Vallentyne, 2008; Søndergaard et al., 2007), but today's P loading increasingly arises from nonpoint sources (ILEC, 1994; Søndergaard et al., 2007; US EPA, 2010), and few lakes affected by non-point loading have been restored through their management alone (Jarvie et al., 2013; Jeppesen et al., 2005; US EPA, 2010). Low success may be temporary if there are lag times between source

manipulations and reduced P supply to phytoplankton associated with the nutrient's transport through the catchment (Cassell et al., 2001; Meals et al., 2010; Sharpley et al., 2003) or to the processing in the lake of biologically-unavailable dissolved organic and particulate P (DOP and PP) into orthophosphate (PO_4^{3-}, HPO_4^{2-} and $H_2PO_4^{-}$; abbreviated PO_4), the P form that is transported across the phytoplankton cell membrane (Reynolds, 2006). In these circumstances, the lake will respond to the new catchment source output once legacy P has been removed from the catchment-lake system via outflow or burial in bottom sediment (Cassell et al., 1998; Meals et al., 2010; Sharpley et al., 2003). However, if the nonpoint sources targeted include substantial biologically-unreactive P, their management will not be as effective as the control of equal amounts of point-source P which is predominantly PO_4 (Dueñas et al., 2003), and is discharged directly into the phytoplankton habitat.

Historically, nonpoint source P has been largely in particulate (Meybeck, 1982; Reynolds and Davies, 2001) form from eroded soil and detritus although where wetlands are abundant, P may reside

primarily in colloidal humus-metal complexes (DeHaan and DeBoer, 1986; Meili, 1992). Orthophosphate generally is scarce, except where runoff passes over exposed phosphate rock, because it sorbs readily to particles and is taken up by microorganisms and plants (Reddy et al., 1999; Reynolds and Davies, 2001; Sharpley et al., 2003). The average PO_4 content of stream flow may have increased, however, (Meybeck, 1982; Seitzinger et al., 2005) as a consequence of the liberal application of commercial fertilizer to urban-suburban landscapes and cropland, and wastewater discharge to streams following secondary, but not tertiary, treatment. Phosphate fertilizer and many industrial P products are manufactured from mined phosphate rock, which is crushed and acidified to release PO_4 (Gowariker et al., 2009). Phosphate mining is relatively new, developed primarily after 1960 as part of agriculture's Green Revolution, which doubled food production globally over four decades, in part by increasing P supplementation of cropland to thirteen times the traditional rate (Tilman, 1999). With 11% of Earth's landmass cropland, the overall P demand has been high and most easily met through the manufacture of mineral phosphate (Gowariker et al., 2009). By 2000, enough P was extracted from inert rock to increase the mass of the global P cycle by three- to five-fold (Falkowski et al., 2000; Smil, 2000). The fate of this extra P may be pertinent to the eutrophication crisis that has recently emerged worldwide, and its effect likely most intense in agricultural areas (US EPA, 2010).

Only a portion of the P amendment to cropland is absorbed by the harvested crop; a larger portion is usually retained in the soil through sorption, microbial uptake and other mechanisms (Carpenter, 2005). Initially P immobilization may be nearly complete, but with repeated fertilizer addition, the soil retention sites saturate, so that newly introduced PO_4 can flush into overland flow or infiltrate to groundwater (Bennett et al., 2001; Carpenter, 2005; Daniels et al., 1998). Global soil surveys carried out around 2000, found P saturation in about 70% of cropland soil (MacDonald et al., 2011). The time needed for agricultural P to deplete soil sorptive capacity may explain why the eutrophication crisis in rural areas emerged in the 1980s or later (Gakstatter et al., 1978; Ryding et al., 1990; US EPA, 1990), decades after Hasler (1947) warned of the effects of domestic sewage influx on lake trophic status.

Much of the P captured in crops reaches agricultural soil as well, although generally in a different catchment, through the disposal of livestock manure and urban sewage sludge (biosolids) on farmland (Naylor et al., 2005; Pimentel and Pimentel, 2008). Underground septic systems, which handle roughly half of human waste in the United States (US Census Bureau, 2013), also rely on soil P retention capacity to protect groundwater from PO_4 input, and when operated for decades, eventually P-saturate the soil in leach fields (Gold and Sims, 2000). Moore et al. (2003) have documented the eutrophication of suburban Seattle lakes through septic P contamination of the groundwater, for example. Because animal wastes are alkaline, and hydroxyl ion competes with PO_4 for shared sorption sites on soil, their addition to soil reduces the overall P retention capacity as well as adding new P (Iyamuremye and Dick, 1996; Lijklema, 1980; Olila and Reddy, 1995).

The hypothesis that mined P is primarily responsible for surging lake eutrophication is currently popular (Bennett et al., 2001; Carpenter, 2005; Daniels et al., 1998), but challenged by the more traditional view that the P output from lake catchments is from internal sources. The global P cycle is, in fact, parsed into catchment units by the strong linkage of P transport to water flow, and mass balance studies indicate that soil erosion rates have been historically high in recent decades (Pimentel, 2006). The principal issue with this hypothesis is the uncertain bioavailability of soil sediment PP. Orthophosphate is the only P molecule small enough to be transported across cell membranes; therefore, PP must be transformed into soluble PO_4 before it can contribute to lake eutrophication. There are many processes in lakes that can accomplish this transformation, e.g. desorption, mineral dissolution, the decomposition of organic matter, and waste elimination from detritivores (Jensen and Andersen, 1992; Pettersson, 1998; Søndergaard et al., 2003). Each of these processes must occur as PP sinks through the water column, however, and may not

be completed before the particulate matter is buried in bottom sediment. Furthermore, all are sensitive to environmental conditions that may or may not be favorable. For example, the dissolution of iron oxy-hydroxides with sorbed PO_4 occurs when the sediment-water interface is anoxic (Nürnberg, 1984), not if dissolved oxygen is abundant, and PO_4 desorption occurs when pH is rising, because OH^- and PO_4 compete for sorption sites, but is unlikely at neutrality (Lijklema, 1980; Olila and Reddy, 1995). In short, considerable information about lake ecology is needed to predict the impact of soil sediment loading to a lake on its level of primary production. Nonetheless, catchment P management based on the total P (TP) content of the P sources present is common practice.

The dynamics of sediment transport in overland flow and streams (Gordon et al., 2013; Leopold et al., 1995) also complicates catchment P management. Orthophosphate and dissolved P are fully integrated into water flow, and thus spend only a few days in runoff before spreading out into the surface waters of a downstream lake. By contrast, particles require the turbulent lift generated by high current velocity to travel in suspension or bedload traction. For many streams, this critical velocity is attained only during spring snowmelt and storm events and subsides quickly. Consequently, the average sized particle often spends much more time deposited on the stream bottom than in transit. For example, Beach (1994) examined the fate of soil eroded from three Minnesota catchments during a forest clear cut 137 years earlier, and found 63 to 87% of the eroded material downhill, but still in the parent catchment. The late start of the eutrophication crisis in lakes that do not have a point-source P influx might be explained by the addition of particle travel time to soil erosion that began acceleration in the middle of the 20th century.

Although eutrophication by P species other than PO_4 should be explored through experimentation, much can be learned ahead of time through the monitoring of lakes impacted by excess erosion or agricultural importation of P. Regular water sampling to monitor trophic status change has been available only since the 1960s, too short a time for some of the processes involving PP mobilization to play out. Consequently, a blend of paleolimnology and neolimnology is necessary, at least in the early stages of research when hypotheses about processes evolve. Sediments collect remnants of past environments that can be quantified in the dated layers of sediment cores (Batterbee and Bennion, 2012; Smol, 2008), and related to recorded land use events. In this work, we analyze thirteen sediment cores from Lake Champlain (Vermont, New York, Quebec) for indicators of trophic status, including total phosphorus (TP), total nitrogen (TN), organic carbon (OC), fossil pigments, biogenic silica (BSi), and diatoms, over up to three centuries. Initially settled by Europeans in the 18th century, the lake catchment was severely deforested and eroded in the 19th century before sewage discharge began around 1900. Reforestation occurred during the 20th century, and reductions in the P content of point sources began around 1980. The intensification of agriculture to include fertilizer and CAFOs (concentrated animal feeding operations) began in the 1970s, but has been especially intense since 1990. We have previously presented data for four cores analyzed from the lake's highly eutrophic and hydrologically isolated northeastern arm (Levine et al., 2012) and focus here on nine cores collected elsewhere. In the final discussion, we evaluate the trophic history of all thirteen study locations to reach conclusions about the roles the major nonpoint and point sources of P have had in driving an initially oligotrophic water body towards eutrophy. The land use history for the catchment, and physical, chemical and biological characteristics of the lake pertinent to the observations made here are provided in Electronic Supplementary Material (ESM) Appendix S1.

Methods

Core acquisition and sampling

Short cores were retrieved at the locations shown in Fig. 1 using a gravity-corer fitted with a 2.5″ OD PVC tube, wrapped in black plastic