Short-range atmospheric transport: A significant source of phosphorus to an oligotrophic lake

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Abstract

During summer stratification, particles transported by wind from surrounding forest were a major source of P into Mirror Lake, a small, oligotrophic lake in New Hampshire. These particles consisted largely of terrestrial insects, insect parts, and plant fragments of various sizes. This atmospheric fallout of P was greatest near the shoreline and decreased exponentially with increasing distance from shore. Integrated for the entire 15-ha lake, it represented an input of ~11-13 μmol P m⁻² d⁻¹ of lake surface in midsummer. The input of P from atmospheric fallout was highly significant to the summer P economy of the lake and was 50-fold to 70-fold greater than the input of P in either streamflow or rainfall in the same period.

Because P is one of the major nutrients limiting primary production in freshwaters (see Hecky and Kilham 1988), there has been much interest in quantifying its sources and sinks in lakes (e.g. Schindler and Fee 1974; Schindler et al. 1978; Bloesch et al. 1977; Carpenter and Adams 1977; Likens and Loucks 1978). In this vein, we have attempted to construct a complete and detailed P budget for the epilimnion of Mirror Lake, an oligotrophic lake in New Hampshire (Caraco et al. 1988). Our initial attempts suggested that during summer stratification the measured losses of P from the epilimnion far exceeded the inputs of P that we could measure; an analysis of these data and their associated uncertainties indicated strongly that we were underestimating some input of P to the lake or, more likely, had overlooked some unmeasured component altogether (Caraco et al. 1988). Thus, to achieve a balanced budget, we needed to examine nontraditional or rarely measured sources of P. In this work we directly measured P transported through the atmosphere and deposited on the surface of Mirror Lake.

Site description

Mirror Lake is a small (15 ha, $z_{max} = 11$ m), well-studied, oligotrophic lake located in the White Mountains of New Hampshire about 0.8 km southeast of the Hubbard Brook Experimental Forest (Likens 1985). The lake’s 85-ha watershed consists of mixed hardwoods and conifers, typical of the northern hardwood forest (Bormann and Likens 1979); there are several homes within the watershed boundaries. The lake is generally dimictic, although vernal mixis is often incomplete; the thermocline is generally about 7 m deep in midsummer (Likens et al. 1985). Water enters the lake by seepage and by three inlet streams (Likens et al. 1985; Winter 1984). It exits by a single outlet during higher flow periods and by seepage at all times.

Phytoplankton primary production accounts for about 80% of the organic C loaded to the lake annually. Much of this production, 42%, occurs during the peak of summer stratification in July and August alone (Jordan and Likens 1975). It is limited by the supply of both P and N (Gerhart and Likens 1975).

Methods

To measure directly the input of P by atmospheric fallout, we deployed a series of floating atmospheric deposition collectors on six transects covering the surface of the lake. These collectors were made of polypropylene, square in shape, with a collecting surface area of 0.057 m². The collectors were
7 cm tall; when 1 liter of water was added (below), the height of the wall above the water was 4 cm. During deployment, 1 liter of a dilute solution of deionized water and NaCl, with a final conductivity of 100 µmhos (about threefold greater than that of the lake water), was added to the collectors. The purpose of the water was to create a wet surface which, like the lake itself, would tend to entrain falling and blowing particles. Collectors without water may severely underestimate the input of particles (Lewis 1983).

The NaCl addition made it possible to determine if lake water had entered the trap; when this dilution occurred, the deployment was considered unsuccessful and excluded from analysis.

Each trap was deployed for 3–4 d, and the contents were size-fractionated and analyzed for P. The entire contents of each trap were passed through a 1-mm Nitex screen to harvest large particulate P (>1 mm); the harvested particles were rinsed off the Nitex screen and caught on a pre-rinsed Whatman GFF filter. Subsamples (100–500 ml) of the first filtrate from the 1-mm Nitex screen (e.g. the <1-mm size class) were filtered through pre-rinsed Whatman GF/F filters to harvest fine particulate P (from ~0.7 µm to 1 mm). The filtrate from the GF/F filters (e.g. the <0.7-µm size class) was analyzed for total dissolved P (TDP). In all cases samples were combusted and the resulting PO₄ was analyzed spectrophotometrically as molybdate-reactive P (Menzel and Corwin 1965). Particulate P samples were analyzed by a modification of the method of Solórzano and Sharp (1980). Particulate samples were combusted thermally, after drying, in borosilicate tubes at 500°C for 6 h. Resultant ash was then extracted in HCl by autoclaving for 1 h at 121°C, filtered (Whatman GF/F), and analyzed for PO₄. TDP samples were combusted in acid persulfate (2 h, 121°C) according to Menzel and Corwin (1965). Blanks for every step of this procedure were obtained by deploying deionized water (plus the NaCl) in containers with lids in the lake and then processing these samples exactly as described above.

In 1986, we put four traps on each of three transects and changed traps every 3–4 d from 22 July through 5 September, for a total of 64 individual, successful deployments, excluding blanks. During 1987, we used five transects with five traps in each transect (Fig. 1); traps were changed every 3–4 d from 28 June through 20 August, for a total of 155 successful deployments, excluding blanks. Of the 155 successful deployments, several samples were lost during processing. Thus, we have 150 samples for small particles, but only 144 samples for total P. Additionally, in 1987, we set an extra transect of five traps, also changed every 3–4 d, to identify the nature of the particles we caught (Fig. 1).

Results

Atmospheric deposition collectors and controls—We found variable but appreciable amounts of P in every collector that we deployed on the lake and relatively minor amounts of P in our covered "blanks." For all of the deployments in 1987, the average flux of total P from the atmosphere into the lake was 23.8 ± 3.9 µmol P m⁻² d⁻¹ (mean ±95% C.L., N = 138; expressed per unit of area of collector surface); the blanks averaged 1.5 ± 1.8 µmol P m⁻² d⁻¹ (mean ±95% C.L., N = 20). The presence of measurable P in the blanks is the combined result of leaching from the collectors themselves, P present in the NaCl and deionized water, and contamination during sample processing. The collectors were 0.057 m² in size.
and contained 1 liter of liquid, so the average blank represents a P concentration of only 0.25 μM above the P concentration in our deionized water—a reasonable value for contamination, considering the amount of handling the samples required (above).

The wet surface in the collectors strongly influenced the amount of material caught. As a test of the collectors, we deployed three of them set up in the usual way, i.e. containing 1 liter of dilute NaCl solution, and three identical collectors, which contained no liquid, in a large clearing about 0.5 m from the lake. The "wet" collectors caught 16.2 ± 10.91 μmol P m⁻² d⁻¹ during a 3-d deployment, while the "dry" collectors caught only 1.7 ± 0.6 μmol m⁻² d⁻¹ in the same period. The dry collectors did accumulate some moisture during deployment, so the actual difference in collection efficiency between a wet and dry surface is probably even greater.

**Particulate and dissolved P input**—Of the P we recovered in the collectors, much was particulate. Again using our 1987 data set, small plus large particulate P combined accounted for 66 ± 20% (mean ± 1 SD, N = 138) of the total atmospheric P flux into the collectors and ranged from 13 to 97% of the total. Small and large particles accounted for roughly equal, and equally variable, shares of this particulate input; 28 ± 21% for large particles and 38 ± 20% for small ones. The input of small and large particles was highly correlated (P < 0.001; r² = 0.41), suggesting either a common origin or a common input mechanism.

The P we measured as dissolved in the collectors may have entered the collectors in dissolved form or may have leached from the particles. The average concentration of dissolved P in particle-free precipitation collected at Mirror Lake is ~0.1 μM (Likens et al. 1985); the amount of dissolved P in our collectors was much higher than this and averaged 1.1 μM. We suspect that a large fraction of the dissolved P leached from particles, although direct sources of dissolved (Lewis et al. 1985) or even gaseous P (e.g. phosphine) are possible.

**Spatial and temporal variation in P deposition**—The amount of P caught in the traps varied considerably over the course of the summer. If we averaged by date and across all collectors, the flux of total P ranged from a high of 37.2 ± 19.7 μmol P m⁻² d⁻¹ (mean ± 95% C.L.) for the 12-16 July 1987 collection, to a low of 8.2 ± 2.5 for the 10-13 August collection (Fig. 2).

There was enormous spatial variability in the amount of P intercepted by collectors at different points on the surface of the lake (Fig. 3). Although much of this variance is explicable in terms of distance from the shoreline (below), a component of the variation may be controlled by wind direction. Averaging across all dates and distances from shoreline, the transect at the northwest end of the lake received significantly higher P deposition than any of the others (Fig. 3). The wind rose for Hubbard Brook is dominated by a flow from the northwest in summer (Likens et al. 1985). Thus the northwesternmost transect would be directly downwind from the surrounding forest more often than the other transects.

The amount of P caught in the collectors was highly dependent on the distance of the collector from the shoreline. Using the three components, date of deployment, distance from shore, and transect position, we performed a series of ANOVAs to determine which component accounted for most of the variation. Together these three factors ac-
counted for 57% of the total variance. Of the variance explained by these three variables, date of deployment was the least significant ($P = 0.027$). The compass location of the transect, although significant, ($P = 0.0025$) accounted for only 24% of the explained variation. Distance from shore was the most significant ($P < 0.001$) and accounted for 63% of the variation explained by the model.

If we average all of our data across all transects and all dates, the deposition of P showed an exponential decrease with distance from the shoreline. We were able to model these data as

$$\text{deposition} = k + A \times (\exp[-bX]) \quad (1)$$

where deposition is the amount of P intercepted per unit of time per unit of area of collector surface ($\mu$mol P m$^{-2}$ d$^{-1}$), $X$ is the distance (m) from the collector to the nearest point of land, and $k$, $b$, and $A$ are constants (below). This exponential model adequately represented the data for the total flux and the flux of large and small particles ($r^2 > 0.91$ in all cases), but had a poorer fit for the dissolved component (Fig. 4; Table 1). The smaller value for $b$ and $k$ in 1986 may reflect either real differences between years or may be a result of the poorer sampling of the shoreline region in 1986 (Table 2).

The constants in Eq 1. can be interpreted functionally; $k$ is the asymptotic value of flux at infinite distance and would, theoretically, represent the input of P at distances unaffected by the shoreline. Mirror Lake is small and surrounded by forest, however, and the maximal distance from the center of the lake to a shore is ~200 m. Thus $k$ should be thought of as a local, lakewide, asymptote rather than a regional one. Analogous to the constant in an exponential decay model, $b$ (the transport constant) represents the fraction of material that falls out per unit of distance from shore. A large value of $b$ means that a large fraction of the material is dropping out per unit of horizontal distance traveled. $A + k$ is the value of deposition when $X = 0$; this sum can be thought of as the magnitude of deposition at the source (shoreline). For a given value of $b$, deposition will depend on the magnitude of this source term.

Composition of the particles—The particles appeared to be of terrestrial origin, largely insect parts, whole insects, and leaf and bud fragments. Pollen was not an important component of this flux, probably because by late July relatively few trees are still in flower in central New Hampshire. Small plant fragments were numerically dominant, accounting for from 20 to 90% of the total number of identifiable particles we caught. Whole insects (and spiders), almost entirely of terrestrial origin, accounted
Table 1. Coefficients for Eq. 1 relating the deposition of P from the atmosphere to distance from shore. Coefficients are given with their standard errors (SE) and with the $R^2$ and $F$ ratio for the model with all three coefficients. Fits were obtained with a Marquardt procedure.

<table>
<thead>
<tr>
<th>Component</th>
<th>Observations (n)</th>
<th>A (μmol m$^{-2}$ d$^{-1}$)</th>
<th>B (μmol m$^{-2}$ d$^{-1}$)</th>
<th>R$^2$</th>
<th>F ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total P 1987</td>
<td>144</td>
<td>11.0 (1.7)</td>
<td>36.5 (4.6)</td>
<td>-0.24 (0.07)</td>
<td>0.95</td>
</tr>
<tr>
<td>Large particles 1987</td>
<td>144</td>
<td>3.7 (1.1)</td>
<td>18.2 (3.1)</td>
<td>-0.26 (0.11)</td>
<td>0.91</td>
</tr>
<tr>
<td>Small particles 1987</td>
<td>150</td>
<td>2.8 (0.40)</td>
<td>14.1 (1.1)</td>
<td>-0.25 (0.04)</td>
<td>0.98</td>
</tr>
<tr>
<td>Dissolved P 1987</td>
<td>144</td>
<td>4.4 (0.49)</td>
<td>3.8 (1.2)</td>
<td>-0.17 (0.13)</td>
<td>0.76</td>
</tr>
<tr>
<td>Total P 1986</td>
<td>64</td>
<td>2.3 (1.9)</td>
<td>19.9 (2.7)</td>
<td>-0.06 (0.02)</td>
<td>0.97</td>
</tr>
</tbody>
</table>

for up to 30% of the total number of particles caught at some locations; ants were the numerically dominant insect caught. Interestingly, although the total number of particles and amount of P deposition decreased with distance from shore, the composition of particles did not show any obvious trends with distance from shore (Fig. 5).

Calculation of P deposition for the entire lake—From a detailed map of the lake, we calculated its area at each 1-m interval from the shoreline. We then applied Eq. 1 and the constants in Table 1 to compute the deposition of P in each size class to each of the distance classes and summed to obtain a whole-lake value (Table 2). During summer stratification, on a lakewide basis, and using the coefficients we obtained for 1987, areally averaged total P flux was about 13 μmol P m$^{-2}$ d$^{-1}$, and evenly split between particulate and dissolved components (Table 2). During 1986 we had fewer collectors and less extensive coverage in the near-shoreline region; thus we may have underestimated actual deposition in 1986.

Discussion

Evaluation of deposition collectors—A collection device may intercept particles differently from the actual lake surface. We have tried to minimize such artifacts by designing a collector with a very low height-to-width ratio and by adding water to the collector to mimic the surface of the lake (see Mason and MacDonald 1982). With 1 liter of water inside the collector, the height-to-width ratio from the inside of the collector is < 1:6, which should minimize excessive overtrapping due to increased wind turbulence. Further, the test results comparing wet and dry collectors clearly show that the addition of a wet surface greatly increases trapping efficiency. Thus, al-

Table 2. Atmospheric fallout of P into Mirror Lake in summer 1986 and 1987. In all cases units are μmol P m$^{-2}$ d$^{-1}$ expressed per unit of lake surface area. Values were computed by integrating Eq. 1 with the appropriate coefficients for each class (Table 1) across 1-m distance classes for the entire lake (see text). In 1987 samples were separated into dissolved and two particle size classes; in 1986 samples were separated into only particulate and dissolved categories.

<table>
<thead>
<tr>
<th>Component</th>
<th>Summer 1986</th>
<th>Summer 1987</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dissolved</td>
<td>5.2</td>
<td>4.8</td>
</tr>
<tr>
<td>Small particles</td>
<td>NA</td>
<td>3.6</td>
</tr>
<tr>
<td>Large particles</td>
<td>NA</td>
<td>4.7</td>
</tr>
<tr>
<td>Small + large particles</td>
<td>5.7</td>
<td>8.3</td>
</tr>
<tr>
<td>Total</td>
<td>10.9</td>
<td>13.1</td>
</tr>
</tbody>
</table>

Fig. 5. Composition of particles falling into Mirror Lake in summer 1987. Each bar shows percent composition (by numbers of particles) averaged across date and for a single transect on the N side of the lake (see Fig. 1).
Table 3. Summary of studies for which there is an estimate of the atmospheric fallout of P in summer.

<table>
<thead>
<tr>
<th>System</th>
<th>Summer atmospheric P deposition (μmol P m⁻² d⁻¹)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lake Warniak, Poland</td>
<td>20</td>
<td>Kowalczewski and Rybak 1981</td>
</tr>
<tr>
<td>Como Creek, Colorado</td>
<td>9–23</td>
<td>Lewis et al. 1985</td>
</tr>
<tr>
<td>Lake Dillon, Colorado</td>
<td>70</td>
<td>Lewis et al. 1985</td>
</tr>
<tr>
<td>Piburger See, Austria</td>
<td>6</td>
<td>Psenner 1984</td>
</tr>
<tr>
<td>Woodland Pond, Pennsylvania</td>
<td>&gt;3*</td>
<td>Bowman 1988</td>
</tr>
<tr>
<td>Castle Lake, California</td>
<td>&gt;18†</td>
<td>Carlton and Goldman 1984</td>
</tr>
<tr>
<td>Mirror Lake, New Hampshire</td>
<td>11–13</td>
<td>This study</td>
</tr>
</tbody>
</table>

* Includes only deposition of terrestrial insects; assumes P is 0.24% of dry wt.
† Includes only deposition of ants during two swarms in August; assumes N:P ratio of 16:1 by atoms.

though the height-to-width ratio from inside the collector is about 1:3 for the dry collector (compared to 1:6 with the water added), the dry collector is much less efficient than the wet collector. Apparently the wet surface in the collector is a more important consideration than the possible artifacts caused by the collector itself. Nevertheless, although the patterns of deposition are relatively clear, some caution should be used in interpreting the absolute magnitude of the depositional flux, which may be somewhat larger or smaller than we have estimated it to be.

**Ecological significance of atmospheric P fallout** — Other researchers, using a variety of traps and collectors, have presented values for the fallout of P during summer into lakes (Table 3); the values range from ~3 to 70 μmol P m⁻² d⁻¹. Our best estimate for this flux into Mirror Lake is ~12 μmol P m⁻² d⁻¹, which is consistent with the few other studies that have measured atmospheric fallout in summer (Table 3). Even though our estimate is not large, atmospheric P fallout may represent an ecologically significant source of P to the lake during summer stratification. Our estimate of atmospheric fallout of P is 50–60 times larger than the fluvial input of P in the same period (Caraco et al. 1988; Likens et al. 1985). The input of P in direct, particle-free rainfall, based on long-term data from Mirror Lake, would represent an input of only 0.2 μmol P m⁻² d⁻¹ (see Caraco et al. 1988; Likens et al. 1985); our particle collectors include both precipitation and particle fallout, but collect about 65 times as much P as rainfall alone. Thus, the input of P in atmospheric fallout, a component ignored in most nutrient budgets, is much larger than some inputs that are more likely to be measured.

The input of P in atmospheric fallout may be important in the maintenance of P concentration in the epilimnion of the lake. The sedimentation of P from the epilimnion has been estimated at ~50 μmol P m⁻² d⁻¹ and is the major loss of P from the epilimnion during summer stratification (Caraco et al. 1988; Moeller and Likens 1978). Atmospheric P fallout, according to our estimate, could support as much as 26% of this output of P. Although we do not know if all of the P in the fallout material were directly available to phytoplankton, we do know that a large fraction of the organic matter initially produced by phytoplankton primary production ultimately leaves the water column by sedimentation and that the total P content of the water column does not decrease appreciably in summer. There must be a source of P to the system that balances output by sedimentation. These observations suggest that the input of P from particle fallout could account for as much as a fourth of this balance and could be significant to the maintenance of primary production in summer in Mirror Lake.

Although an important linkage between a lake and its watershed through airborne particles may seem unexpected, it may be of general ecological significance to small lakes. A number of researchers have pointed out the more obvious importance of autumnal litterfall to the C budgets of such lakes (e.g. Gasith and Hasler 1976; Rau 1976). In summer, when other allochthonous inputs are generally at their seasonal lows in many regions, atmospheric fallout of terrestrial particles can be a major input of N (Carlton...
and Goldman 1984) or P. Further, the input of terrestrial insects, and ants in particular, appears to be an important subsidy to the diet of fishes in some lakes (Carlton and Goldman 1984) and in streams (Mason and MacDonald 1982). In Mirror Lake, allochthonous food sources, including ants and terrestrial caterpillars, constitute at times as much as 50% of the diet of smallmouth bass (*Micropterus dolomieui*; Helman 1985); caterpillars are also significant to the diet of the spotted newt (*Notophthalmus viridescens*; Burton 1985).

There has been keen interest recently in the role that higher organisms might play in regulating phytoplankton (e.g. Carpenter et al. 1985). This control is usually envisaged as a cascading sequence of predator-prey interactions. If the nutrients in allochthonous particles and terrestrial insects were to become available to phytoplankton largely after digestion by fish or invertebrates, the process would represent an intriguing twist on the concept of "top-down" regulation of food webs.

**References**


---, ---, AND T. RUSZYNSKI. 1978. Phos-
Atmospheric transport of $P$


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