

# THE HYDROGEOCHEMISTRY OF THE PECONIC RIVER WATERSHED: A QUANTITATIVE APPROACH TO ESTIMATE THE ANTHROPOGENIC LOADINGS IN THE WATERSHED

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## ABSTRACT

The Peconic River watershed, which forms the core of the Central Pine Barrens of Eastern Long Island is largely undeveloped. However, a comparison with the hydrogeochemistry of streams in the core of the New Jersey Pine Barrens indicates that the watershed is far from pristine. The magnitude of the anthropogenic loadings can be constrained using mass balances. Furthermore, by examining the chemical signatures of waters throughout the watershed the relative importance of various sources of anthropogenic loadings can be estimated.

## INTRODUCTION

One of the major challenges our community faces is the preservation of the Central Pine Barrens of Long Island (CPBLI). For socio-economic and political reasons, a return of this ecosystems to its pristine, pre-development condition is untenable. Instead, a compromise must be reached with the objective to protect the ecosystem from degradation while allowing some forms of land use to continue. A critical step in developing a best management plan for the CPBLI is to determine the impact the various forms of land use have on the ecosystem. For most other watersheds the problem is that the effects of local antropogenic inputs are difficult to separate from the two other major sources: atmospheric inputs and weathering. However, the CPBLI offers an unique opportunity to study the effect of limited development because the atmospheric sources are well constrained while contribution through weathering of soil minerals is probably small. Furthermore, the CPBLI may be compared with watersheds in the nearby New Jersey Pine Barrens (NJPB) which have varying levels of development.

## RESEARCH STRATEGY AND METHODS

Mass balances for all major constituents were calculated to constrain the magnitude of the anthropogenic loadings in the watershed. In essence, a mass balance calculation treats the watershed as a black box with inputs and outputs. Constraining the outputs is relatively easy. Nearly all the effective recharge leaves the watershed as riverine discharge (Stasko and Schoonen, 1993). By combining discharge data at the Riverhead gauge and water quality data, the discharge (i.e., export) of constituents can be calculated. Constraining the input is more difficult. Atmospheric loadings can be estimated based on precipitation data collected at Brookhaven National Laboratory in the headwaters of the Peconic River. Any mass imbalance for a constituent must be due to anthropogenic loadings and/or weathering.

While the mass balance approach provides a good frame work for estimating the non-atmospheric inputs or losses in the watershed, it provides little insight into where inputs may occur or what the chemical signature of these inputs is. Therefore, it is important to combine mass balance calculations with an examination of seasonal and spatial variations in the hydrogeochemistry throughout the watershed. In order to eliminate the effects of evapotranspiration, it is useful to compare water quality samples in terms of molar X/Cl ratios, where X is a major constituent. Chloride is used as a divisor because it is the most abundant anion in the waters and it is not taken up by vegetation or mineral surfaces.

To determine the atmospheric inputs for the Peconic River watershed, we made use of data available for precipitation collected between 1965 and 1990 at Brookhaven National Lab in the headwaters of the watershed (see also Proios and Schoonen, 1994). To determine the loss of major elements and nutrients from the watershed as well as the spatial and seasonal variation in chemical composition of the river water, a water quality monitoring program was maintained between 1990 and 1993. Water samples were collected at regular intervals at five sites along the river, two ponds that drain into the river, and two shallow wells (< 3 ft), see Fig. 1. The same field and laboratory procedures as described in Choynowski and Schoonen (1994) were used for the 200 samples collected during this study.

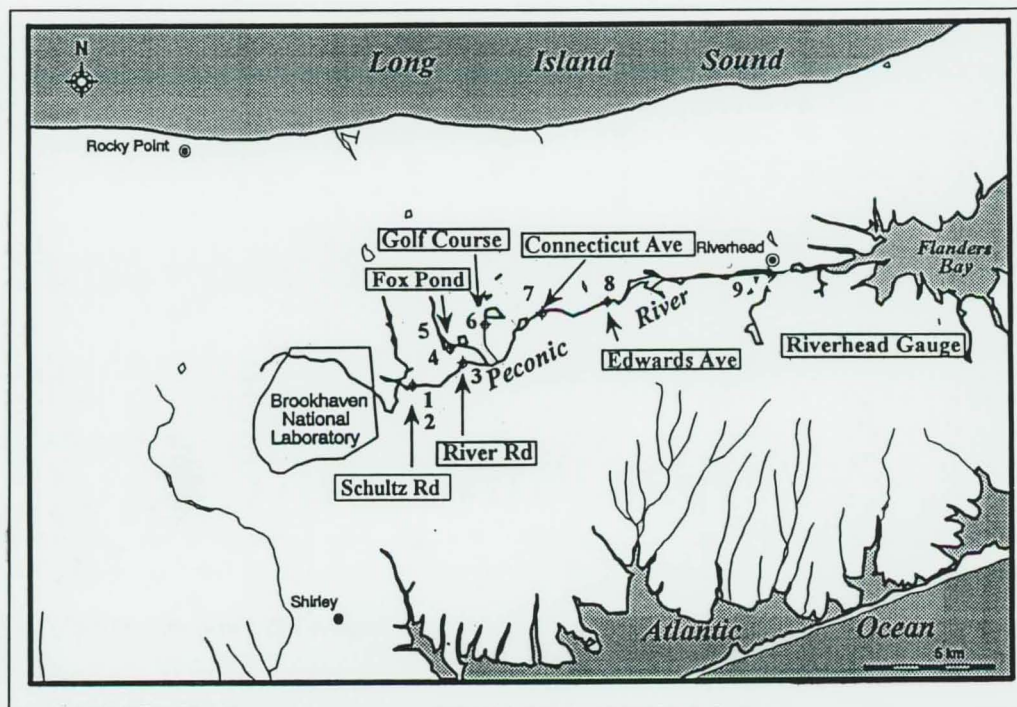


Figure 1. Location map of Peconic River watershed. Numbers are sampling locations. Stations 1,3,7,8, and 9 are along the river; stations 4 and 6 are two contributing ponds, and 2 and 5 are shallow wells. Modified after Xin (1993).

## RESULTS AND DISCUSSION

### *Precipitation Chemistry*

To constrain the atmospheric loading in the CPBLI, we made use of precipitation data collected at Brookhaven National Laboratory between 1965 and 1990. Between 1965 and 1978, BNL was used by the USGS to monitor the deposition of acid rain by analyzing one-month composite bulk precipitation samples. In the late 1970's, this program was superseded with the MAP3S program administered by the DOE and EPA. During the MAP3S program each precipitation event was sampled using a dry-wet collector. For this study, we obtained the raw data collected under the MAP3S program for 1982 to 1989 and made use of the summary of the USGS-data by Peters et al. (1982), which covers data collected between 1965 and 1978. Table 1 summarizes the data in the form of ion ratios, the mean volume-weighted major element concentrations, the atmospheric loading based on the precipitation data.

The composition of the precipitation collected at BNL is typical for precipitation along the Northeastern Atlantic Coast (Berner and Berner, 1987). The composition of the precipitation is strongly influenced by the proximity to the ocean and by acid deposition. The influence of the ocean can be seen from the Na/Cl and Mg/Cl ratios in the rain which are close to those in sea water. Acid deposition causes elevated  $\text{NO}_3$  and  $\text{SO}_4$  concentrations and loadings. Ammonia is derived from agricultural activity in the Mid-West (Berner and Berner, 1987). A more detailed discussion of the composition of the precipitation can be found in (Dana and Barchett, 19xx; Schoonen and Brown, 1994). In addition, the work by Proios and Schoonen (1994) examines the relation between weather patterns and chemical composition of the precipitation. Because the composition of the precipitation in New Jersey is very similar in chemical composition to that for Brookhaven, it is reasonable to assume that the chemical signature of the precipitation is the same for both the CPBLI and the NJPB (Schoonen and Brown, 1994).

The precipitation data collected at Brookhaven National Lab in the headwaters of the Peconic River watershed provide, however, only a constraint on the minimum atmospheric inputs. The Peconic River watershed receives a significant amount of its major constituents through atmospheric processes such as wet precipitation, dry precipitation and impaction. The difference in average loadings based on wet precipitation and those based on bulk precipitation (see Table 1) indicates that dry precipitation is an important process. However, loadings based on bulk precipitation do not account for the contribution of impaction of aerosols on trees. The contribution of impaction has been shown to be of great importance by Art et al. (1974) in a study on Fire Island. A second

problem in constraining the atmospheric input is that the bulk precipitation data date back to the 1970's. The loadings of marine-derived constituents (i.e., Na, Cl, Mg) are probably unchanged, but the loading of SO<sub>4</sub> and NO<sub>3</sub> may be lower now due to stricter limits on emissions (see Likens, 1992). Therefore, based on a study by Lord et al. (1988) which compared bulk versus wet precipitation, we multiplied the loadings based on wet precipitation for SO<sub>4</sub>, NO<sub>3</sub>, and NH<sub>4</sub> with a factor of 1.5. As seen below, this assumption is not critical but Table 3 (below) represents our best guess at the atmospheric loadings in the Peconic River watershed.

**Table 1. Summary of chemical composition of precipitation collected at BNL.**

|                     | CPBLI<br>Wet <sup>x</sup><br>X/Cl <sup>a</sup> | CPBLI<br>Bulk <sup>y</sup><br>X/Cl <sup>a</sup> |                               | CPBLI<br>Wet <sup>x</sup><br>[uM] <sup>b</sup> | CPBLI<br>Bulk <sup>y</sup><br>[uM] <sup>b</sup> |                                 | CPBLI<br>Wet <sup>x</sup><br>loading <sup>c</sup> | CPBLI<br>Bulk <sup>y</sup><br>loading <sup>c</sup> |
|---------------------|--|---|-------------------------------|--|---|---------------------------------|---|--|
| K/Cl                | 0.11   | 0.06  | pH                            | 4.38   | 4.29  | H <sup>+</sup>                  | 0.052   | 0.063  |
| Mg/Cl               | 0.10   | 0.15  | H <sup>+</sup>                | 41   | 51  | K <sup>+</sup>                  | 0.11  | 0.14   |
| Ca/Cl               | 0.06   | 0.22  | K <sup>+</sup>                | 3.5  | 3   | Mg <sup>2+</sup>                | 0.10  | 0.24   |
| Na/Cl               | 0.81   | 0.93  | Mg <sup>2+</sup>              | 3.3  | 8   | Ca <sup>2+</sup>                | 0.10  | 0.59   |
| SO <sub>4</sub> /Cl | 0.66   | 0.69  | Ca <sup>2+</sup>              | 2.0  | 12  | Na <sup>+</sup>                 | 0.72  | 1.41   |
| NO <sub>3</sub> /Cl | 0.14   | 0.5   | Na <sup>+</sup>               | 24.9   | 50  | Cl <sup>-</sup>                 | 1.36  | 2.36   |
| NH <sub>4</sub> /Cl | 0.31   | .20   | Cl <sup>-</sup>               | 30.2   | 54  | SO <sub>4</sub> <sup>2-</sup>   | 2.43  | 4.37   |
|                     |  |   | SO <sub>4</sub> <sup>2-</sup> | 20.0   | 37  | NO <sub>3</sub> <sup>-(N)</sup> | 0.34  | 0.46   |
|                     |  |   | NO <sub>3</sub> <sup>-</sup>  | 19.3   | 27  | NH <sub>4</sub> <sup>+(N)</sup> | 0.21  | 0.19   |
|                     |  |   | NH <sub>4</sub> <sup>+</sup>  | 12.0   | 11  |                                 |   |  |

<sup>x</sup> Wet precipitation collected between 1982 and 1989 (MAP3S Program). <sup>y</sup> Bulk precipitation collected between 1965 and 1978 (Peters et al., 1982). <sup>a</sup> Molar ratio. <sup>b</sup> Average volume-weighted concentration. <sup>c</sup> Average loadings in metric tons/yr/km<sup>2</sup>.

### *Hydrogeochemistry of Peconic River*

The water of the Peconic River and its tributaries is comparable to that in streams draining disturbed watersheds in the NJPB. Morgan and Good (1988) and Morgan et al. (1983) found that disturbed watersheds in the NJPB are primarily characterized by significantly higher pH, nitrate concentration and alkalinity. Furthermore Ca, Mg, K and SO<sub>4</sub> concentrations tend to be higher in disturbed watersheds. The chemical characteristics for disturbed and undisturbed NJPB watersheds are here summarized in Table 2. Table 2 shows average concentration data for sampling station 1 in the headwaters of the river (Schultz Road), a shallow well near Fox Pond (station 5), and sampling station 9 (USGS Gauge at Riverhead). From Table 2, it is clear that the water of the Peconic River has a chemical signature similar to that of the disturbed NJPB watersheds (see molar X/Cl ratios). However, the shallow well near Fox Pond, which is located in a pine-oak forest, has the chemical signature of an unpolluted water. The objective of this study is to further quantify these anthropogenic inputs and examine intra-watershed, or spatial variations that may be linked to these forms of land use.

### *A Chemical Budget for the Watershed*

In the preceding sections the concentrations and chemical signature of atmospheric precipitation as well as the stream water were presented. It is clear from this data that there are non-atmospheric sources in addition to atmospheric sources. By drawing up a constituent budget for the watershed it is possible to constrain the importance of non-atmospheric sources. To calculate the chemical budget for each major constituent, its atmospheric loading must be known as well as its discharge. In the Peconic River watershed, nearly all of the effective recharge leaves the watershed as riverine discharge (Stasko and Schoonen, 1993). Therefore, riverine discharge represents a good estimate of the total loss of major constituent from the watershed. The budget presented in Table 3 was calculated by combining USGS discharge data for the Riverhead gauge with our chemical composition data.

The budget presented in Table 3 indicates a large imbalance between atmospheric input and riverine output for nearly all major constituents. The watershed gains protons and nitrogen but loses all other major constituents. The only constituent that appears to be nearly in balance is sulfate. The nitrogen loss is due to uptake by vegetation (Brown and Schoonen, 1993; Woodwell, 1979), but the proton loss must be compensated with the release of another cation. Note also that the relative discrepancies for sodium and chloride are essentially the same. The riverine discharge of sodium and chloride is a little over twice as large as the minimum atmospheric

loading, while the riverine discharges for Ca, Mg, and K are nearly a factor 2.5 higher than the minimum atmospheric loading. It is clear from these budget calculations that (1) a significant Na and Cl source with a molar Na/Cl ratio close to unity is not accounted for and (2) that a significant Ca, Mg, and K source is present.

It is unlikely that impaction can account for the observed imbalance in Na and Cl. Art et al., (1974) found that on Fire Island impaction was about as important as bulk precipitation. However, the concentration of seasalt aerosols drops of rapidly away from the coast. Moreover, easterly and northeasterly winds are far less frequent than westerly and southwesterly winds (Miltenberger et al., 1990). Therefore, impaction is probably less important in the Peconic River watershed than on Fire Island.

**Table 2. Average Concentration and molar X/Cl ratios for waters in CPBLI and NJPB.**

| Constituent         | Station 1    | Station 5  | Station 9   | NJPB                     | NJPB                   |
|---------------------|--------------|------------|-------------|--------------------------|------------------------|
|                     | (Schultz Rd) | (Fox Well) | (Riverhead) | undisturbed <sup>a</sup> | disturbed <sup>a</sup> |
|                     | [ $\mu$ M]   | [ $\mu$ M] | [ $\mu$ M]  | [ $\mu$ M]               | [ $\mu$ M]             |
| Cl                  | 235          | 162        | 376         | 133                      | 145                    |
| K                   | 18           | 4          | 30          | 11                       | 21                     |
| Na                  | 249          | 144        | 363         | 114                      | 118                    |
| Mg                  | 51           | 18         | 89.8        | 20                       | 38                     |
| Ca                  | 73.2         | 14         | 151         | 14                       | 45                     |
| SO <sub>4</sub>     | 52.7         | 78         | 110         | 40                       | 65                     |
| NO <sub>3</sub>     | 5.11         | 0.71       | 9.47        | 1                        | 12                     |
| HCO <sub>3</sub>    | 185          | 55         | 297         | 0-50                     | >50                    |
| H <sup>+</sup>      | 1.61         | 28.8       | 0.25        | 30                       | 16                     |
| pH                  | 5.79         | 4.54       | 6.6         | 4.52                     | 4.79                   |
| K/Cl                | 0.08         | 0.05       | 0.08        | 0.08                     | 0.14                   |
| Na/Cl               | 1.07         | 1.28       | .97         | 0.86                     | 0.81                   |
| Mg/Cl               | 0.23         | .15        | 0.24        | 0.15                     | 0.26                   |
| Ca/Cl               | 0.34         | .13        | 0.40        | 0.11                     | 0.31                   |
| SO <sub>4</sub> /Cl | 0.23         | .69        | .29         | 0.30                     | 0.45                   |
| NO <sub>3</sub> /Cl | 0.02         | .004       | 0.03        | 0.01                     | 0.08                   |
| H <sup>+</sup> /Cl  | 0.007        | 0.38       | 0.001       | 0.22                     | 0.11                   |

<sup>a</sup> Data taken from Morgan and Good (1988). All other data taken from Schoonen and Brown (1994). Ratios are molar ratios.

**Table 3 Chemical budget for Peconic River at USGS gauging station near Riverhead.**

| Element                       | Minimum Atmospheric Input<br>(10 <sup>3</sup> kg/km <sup>2</sup> /yr) | Riverine Discharge<br>(10 <sup>3</sup> kg/km <sup>2</sup> /yr) | Uptake or Loss<br>(10 <sup>3</sup> kg/km <sup>2</sup> /yr) | %Uptake (+) or Export (-) |
|-------------------------------|---|--|--|---------------------------|
| H <sup>+</sup>                | 0.052   | 0.005  | 0.047  | +90                       |
| K <sup>+</sup>                | 0.14  | 0.49   | -0.35  | -250                      |
| Mg <sup>2+</sup>              | 0.24  | 0.83   | -0.59  | -246                      |
| Ca <sup>2+</sup>              | 0.59  | 2.20   | -1.61  | -272                      |
| Na <sup>+</sup>               | 1.41  | 3.38   | -1.97  | -139                      |
| Cl <sup>-</sup>               | 2.36  | 5.29   | -2.93  | -124                      |
| SO <sub>4</sub> <sup>2-</sup> | 3.64  | 4.05   | -0.41  | -11                       |
| N                             | 0.83  | 0.12   | +0.71  | +86                       |

### Spatial Variations

To evaluate the importance of non-atmospheric sources such as weathering, fertilizers, road salt, and septic tank effluent, it is useful to examine the chemical signature of the waters collected at the nine stations throughout the watershed. Figure 2 presents the chemical signature of the waters at each sampling station in the form of molar X/Cl ratios. Molar X/Cl ratios are a more useful basis for comparison than concentrations because fluctuations in evapotranspiration have no effect on X/Cl ratios. As seen in Fig. 2, the Na/Cl ratio is virtually constant throughout the watershed and slightly elevated with respect to the Na/Cl ratio in precipitation. Hence, the input of Na and Cl not accounted for by the estimated atmospheric input must have a 1:1 molar ratio. In contrast to Na/Cl, Ca/Cl, Mg/Cl, K/Cl, HCO<sub>3</sub>/Cl, and SO<sub>4</sub>/Cl show a significant spatial variation.

Based on Figure 2, it is clear that the elevated Ca/Cl, Mg/Cl, molar ratios found in Fox Pond, Swan Pond, and the well at Schultz Road are due to local disturbances. Upgradient from Fox Pond there are three houses, Swan Pond is adjacent to a golf course, and the well at Schultz Road may be influenced by several homes nearby and agricultural activity further to the south. Although the chemical signatures of these waters are significantly different from that in the river, the two tributaries examined here (Fox Pond and Swan Pond) appear to contribute only a minor volume to the river water because the chemical signature of the river remains nearly constant Figure throughout its course. The shallow water collected in the pine-oak forest at Fox Pond has a chemical signature close to that for precipitation and represents unspoiled pine barrens water (see Table 2). The chemical signature of the river water appears to be a mixture of pristine water and water affected by (local) disturbances.

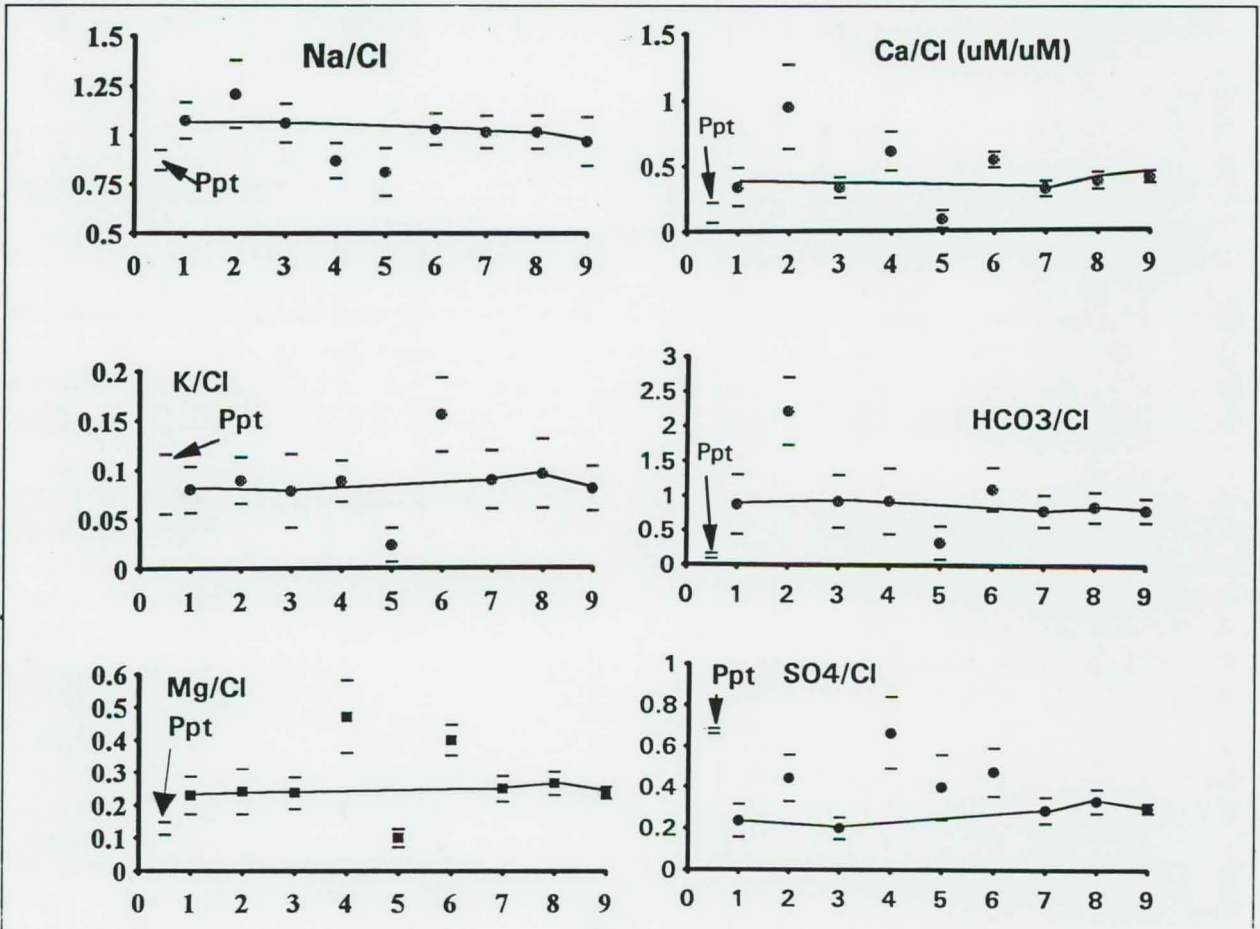


Figure 2. Spatial variation in watershed. Horizontal axis refers to sampling stations indicated on Figure 1. Stations along river are connected with line. For each station average value are given. Error bars represent  $\pm$  one standard deviation. Precipitation is presented as two average values (one for wet precipitation and one for bulk precipitation).

### *Anthropogenic Sources*

Possible anthropogenic sources in the Peconic River watershed are (1) road salt, (2) fertilizers and soil-pH adjusters, and (3) septic tank effluent. Per application 0.3 to 1 ton of NaCl is used per mile-lane per salting (Watson, Suffolk County Highway Department, pers. comm.). With about 80 lane-miles of primary and secondary roads in the watershed, one salting adds as much as 80 tons of NaCl. This is equivalent to 20 % of the estimated minimum atmospheric input. Because the river is fed by ground water, it may take several years for salt introduced near the groundwater divide to discharge into the river or its tributaries. Hence, it is not surprising that there is no significant difference in Na and Cl concentrations between the winter months and the rest of the year (Schoonen and Brown, 1994). Since neither Na or Cl are taken up by vegetation the 1:1 ratio in the river water may be caused by a significant input of road salt. However, road salt is only a source for Na and Cl. Therefore, Ca, Mg, HCO<sub>3</sub><sup>-</sup>, and K must be derived from another source which contributes little or no Na and Cl.

Fertilizers and soil adjusters are a necessity in the pine barrens if the soil is to be used for growing turf or for agriculture. The soil pH is around 4 which stunts crops and turf. To raise the soil pH lime (CaCO<sub>3</sub>) is added to the soil. Liming of soils throughout much of the watershed could be responsible for the uptake of protons and the discharge of calcium and much of the bicarbonate seen throughout the watershed. The neutralization of acid by lime is governed by the following reaction:  $\text{CaCO}_3 + \text{H}_2\text{CO}_3 \Rightarrow \text{Ca}^{2+} + 2\text{HCO}_3^-$ . The watershed takes up  $47 \cdot 10^3$  moles H<sup>+</sup>/km<sup>2</sup>/yr while it releases  $40 \cdot 10^3$  moles Ca<sup>2+</sup>/km<sup>2</sup>/yr and about  $80 \cdot 10^3$  moles of HCO<sub>3</sub><sup>-</sup>. Furthermore, liming would not change the Na/Cl ratio in the watershed. Fertilizers are rich in K, Ca, and Mg. All these constituents are elevated in Swan Pond which indicates that this water is significantly impacted by the golf course. N and P derived from fertilizers is probably largely taken up by the turf or increased primary production in Swan Pond.

Septic systems are a third source that needs to be evaluated. Septic tank effluent has typically about twice as much sodium than chloride by weight (Robertson et al., 1991). Hence, if the river received a significant amount of septic tank effluent we would expect a higher molar Na/Cl ratio than observed. Furthermore, the low population density in the watershed rules out that septic tank effluent is important in terms of volume. This is evident from a comparison of the volume of septic system effluent versus the discharge of the Peconic River. On average a person produces 166 L of waste water per day (Koppelman, 1978). In contrast, the average discharge at the USGS gauge is  $9 \cdot 10^7$  L/d. The population in the watershed is probably lower than 10000 (Miltenberger et al., 1990); however, using that number, less than 2% of the discharge could be derived from septic systems. Note, however, that while septic tanks are not an important in terms of volume, they may be important as a source for nitrogen, and phosphate. Septic tank effluent contains typically about 30 to 100 mg/L N, and 8 to 15 mg/L P (Robertson et al., 1991).

### *Weathering*

Some of the calculated losses in the watershed may be due to weathering of soil minerals and aquifer material. Studies by Woodwell (1979) at BNL indicate that the forest floor may yield K, Mg, and Ca. However, the Ca/Cl, Mg/Cl, and HCO<sub>3</sub>/Cl ratios found in water collected below a mature oak-pine forest are consistently lower than the ratios found in the river water. This is particularly evident if all observed ratios are multiplied by a factor 2 to correct for the fact that half of the Cl in the watershed is probably derived from road salt. Hence, it appears that while weathering is likely to contribute to the observed imbalance for Ca, Mg, and K, it is not the most important source.

### **CONCLUSIONS**

The water quality in Peconic River watershed is comparable to disturbed watersheds in the NJPB. Deicing of roads is probably responsible for about half of the Na and Cl exported from the watershed. Ca is primarily elevated due to liming which neutralizes much of the acidity in the watershed and also explains the formation of bicarbonate. Mg and K are major constituents in fertilizers. However, some Mg, K, and Ca may also be derived from weathering. Of course N and P are also added to the watershed with the use of fertilizers and the discharge of septic tank effluent. However, the ecosystem removes nearly all phosphorus and much of the nitrogen.

The consequences of the anthropogenic inputs on the ecology of the watershed are difficult to evaluate. However, several studies in the NJPB indicate that in disturbed watersheds the species characteristic for the pine barrens environment will decrease in numbers (see e.g., Ehrenfield, 1983; and Morgan and Phillip, 1986; Morgan, 1987).

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