Ground Water Quality

Nitrate Distributions and Source Identification in the Abbotsford–Sumas Aquifer, Northwestern Washington State

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ABSTRACT

The Abbotsford–Sumas Aquifer is a shallow, predominantly unconfined aquifer that spans regions in southwestern British Columbia, Canada and northwestern Washington, USA. The aquifer is prone to nitrate contamination because of extensive regional agricultural practices. A 22-month ground water nitrate assessment was performed in a 10-km² study area adjacent to the international boundary in northwestern Washington to examine nitrate concentrations and nitrogen isotope ratios to characterize local source contributions from up-gradient sources in Canada. Nitrate concentrations in excess of 10 mg nitrate as nitrogen per liter (mg N L⁻¹) were observed in ground water from most of the 26 domestic wells sampled in the study area, and in a creek that dissects the study area. The nitrate distribution was characteristic of nonpoint agricultural sources and consistent with the historical documentation of agriculturally related nitrate contamination in many parts of the aquifer. Hydrogeologic information, nitrogen isotope values, and statistical analyses indicated a nitrate concentration stratification in the study area. The highest concentrations (>20 mg N L⁻¹) occurred in shallow regions of the aquifer and were linked to local agricultural practices in northwestern Washington. Nitrate concentrations in excess of 10 mg N L⁻¹ deeper in the aquifer (>10 m) were related to agricultural sources in Canada. The identification of two possible sources of ground water nitrate in northwestern Washington adds to the difficulty in assessing and implementing local nutrient management plans for protecting drinking water in the region.

The Abbotsford–Sumas Aquifer lies within the agriculturally productive Fraser–Whatcom lowlands that straddle the U.S.–Canada boundary (Fig. 1). Intensive agricultural practices in the lowlands, combined with high precipitation and well-drained soils, make the unconfined aquifer susceptible to nitrate (NO₃⁻) contamination (Tesoriero and Voss, 1997). In addition to land use in northwestern Washington, agricultural practices in British Columbia, Canada are affecting water quality in the northwestern Washington area because ground water in the aquifer flows south. Nitrate concentrations exceeding the USEPA maximum contaminant level (MCL) of 10 mg nitrate as nitrogen per liter (mg N L⁻¹) have persisted in ground water and surface water in northwestern Washington, despite the implementation of best management practices (BMPs) over the past two decades (Wills, 1998). This is problematic because the aquifer serves as a drinking water supply for rural residents and small communities in both the USA and Canada (Hii et al., 1999). In a step to improve water quality, in April 1998 the State of Washington signed into law the Dairy Nutrient Management Act. This law requires all dairy farms to develop and implement a dairy nutrient management plan by December 2003. Since agribusiness is an important economic base in this region, differentiating sources of NO₃⁻ to the aquifer is essential in the design and management of agricultural loading in northwestern Washington, both from a public health and economic standpoint.

Ground water NO₃⁻ contamination in shallow unconfined aquifers due to agricultural practices is a worldwide problem. Excess N in the form of NO₃⁻ accumulates in the soil when land application of animal manures and inorganic, commercial fertilizers exceed the N agronomically required by crops. The mobile NO₃⁻ ion is easily leached to the ground water during precipitation events and irrigation, especially in areas with well-drained soils and shallow water tables. Numerous studies have documented the extent of the problem and the relationship between agricultural activities and NO₃⁻ leaching in shallow aquifers (e.g., Power and Schepers, 1989; Spalding and Exner, 1993; Stuart et al., 1994; Puckett et al., 1999; Nolan and Stoner, 2000; Nolan, 2001). Nitrate contamination in the Abbotsford–Sumas Aquifer is unique because it spans extensive agricultural regions in both British Columbia and northwestern Washington, making it difficult to manage.

A section of the Abbotsford–Sumas Aquifer in southern British Columbia, referred to here as the British Columbia study area (Fig. 1), has been the subject of a number of studies in the last two decades because of its high density of agricultural land use. It is called the “raspberry capital of Canada” and also contains a large number of poultry farms and processing plants (Hii et al., 1999). Researchers have reported NO₃⁻ concentrations in the study area exceeding the 10 mg N L⁻¹ MCL (Liebscher et al., 1992; Wassenaar, 1995; Zebarth et al., 1998; Hii et al., 1999). Most recently, Hii et al. (1999) summarized NO₃⁻ results from piezometers sampled between 1991 and 1997 in the British Columbia study area and documented persistent high concentrations of NO₃⁻ in both shallow and deep samples in the aquifer. Wassenaar (1995) analyzed N and O isotope data for NO₃⁻ from 117 ground water samples predominantly

Abbreviations: DBWT, depth below the water table; MCL, maximum contaminant level.


from the British Columbia study area and concluded that the primary British Columbia source for NO$_3^-$ in the aquifer is poultry manure, and to a lesser extent inorganic, commercial fertilizers. Poultry manure was also indicated as the dominant N source to the British Columbia aquifer by Zebarth et al. (1998).

Like the British Columbia study area, agricultural land use in northwestern Washington is also contributing NO$_3^-$ to the aquifer. This region is the largest producer of raspberries (Rubus spp.) in the state of Washington and has many dairy farms; therefore, fertilization practices include a mix of inorganic, commercial fertilizers and animal manure (J. Gillies, personal communication, 1998). Nitrate concentrations above the 10 mg N L$^{-1}$ MCL have been reported at a number of locations in the aquifer in northwestern Washington (e.g., Erickson, 1998; Cox and Kahle, 1999). In addition, degraded surface water quality in the region also suggests contamination due to land use (Wills, 1998; Butkus et al., 2000). Because ground water flows south from British Columbia, accurate knowledge of NO$_3^-$ concentrations and source separation in the aquifer is important to manage agronomic loading adequately in northwestern Washington and to protect drinking water in the region. Nitrate concentrations in the northwestern Washington portion of the aquifer have not been comprehensively examined previously to assess this issue.

The objectives of this study were to (i) characterize the extent of ground water NO$_3^-$ contamination in a study area of the Abbotsford–Sumas Aquifer in northwestern Washington, and (ii) distinguish NO$_3^-$ sources originating up-gradient in British Columbia from sources localized in northwestern Washington. These objectives can be achieved by evaluating the horizontal and vertical variability of ground water NO$_3^-$ concentrations and N isotope values in a portion of the aquifer using domestic wells and linking them to land use information. We assume that deep wells in the study area are a proxy for ground water age and withdraw ground water derived up-gradient in British Columbia. Nitrate data from shallow wells and a surface water stream in the study area will be used as an indicator of localized sources of NO$_3^-$.

STUDY AREA

Hydrogeologic Setting

The study area encompasses approximately 10 km$^2$ of the Abbotsford–Sumas Aquifer in northwestern Washington, beginning at the international boundary (Fig. 2). Regionally, the Abbotsford–Sumas Aquifer is predominately unconfined and covers an area of about 200 km$^2$ (Liebscher et al., 1992). The aquifer is comprised largely of glacial outwash sand and gravel deposits with lenses of sand, silt, and clay deposited during the Sumas stage (11 000–10 000 yr ago) of the Fraser glaciation (Easterbrook and Hansen, 1974). The thickness of the unconfined aquifer is typically about 15 to 25 m, but can range from 5 to 70 m (Cox and Kahle, 1999). Because of the high percentage of sand and gravel, the aquifer has a large capacity to transmit water. Cox and Kahle (1999) used data from 170 completed wells in the aquifer to estimate a mean hydraulic conductivity of 82 m d$^{-1}$ that ranged from 2 to 2380 m d$^{-1}$. They also estimated a median lateral ground water velocity of 0.75 m d$^{-1}$ and a range from 0.06 to 8.8 m d$^{-1}$. These ranges of values are consistent with values determined for the British Columbia study area by Liebscher et al. (1992) and Hii et al. (1999), and by Stasney (2000) for our study area. Underlying the Abbotsford–Sumas Aquifer is a confining unit, primarily clay with an unsorted mixture of pebbly silt of glaciomarine origin (13 500–11 000 yr ago). The clay layer is believed to be continuous at depth and is interpreted as Bellingham glaciomarine drift (Easterbrook, 1976) in northwestern Washington and as the Fort Langley Formation (Halstead, 1986) of glaciomarine origin to the north in British Columbia. The dominant soil types in the study area include the Kickerville silt loam (coarse-loamy, isotic, mesic Typic Haplorthod), Jug very gravelly loam (sandy-skeletal, mixed, frigid Andic Haplohumod), and Skipopa silt loam (fine, isotic, mesic Aquandic Haploxer-
The soils are thin (0–70 cm) with permeabilities generally greater than typical precipitation rates (Cox and Kahle, 1999). Recharge to the aquifer is primarily by precipitation, of which 70% typically occurs between October and March. Because of the permeable soils and coarse-grained aquifer material, recharge is on the order of 60% of the annual precipitation (Cox and Kahle, 1999). The 30-yr rainfall average at the Clearbook, WA weather station near our study area is 1160 mm, annually. Yearly rainfall for 1997 and 1998 were 1.56 and 1.17 m, respectively (Stasney, 2000). Irrigation is also common in the summer months in many parts of the region and is typically on the order of 0.30 to 0.45 m (J. Gillies, personal communication, 2001). Discharge regions are small creeks, lakes, and streams. Water level measurements indicate that the water-table depth fluctuated from season to season in the study area and showed the largest variations between the spring and fall seasons of 1997–1999 (Stasney, 2000). The largest overall average variation in water table levels was approximately 2 m in the northern half of the study area and 1 m in the southern half.

Land Use

Land use in the study area is dominated by agriculture. In 1998, 57% of the study area was comprised of berry fields, dairies, corn (Zea mays L.) fields, and grasslands; raspberries occupied 50% of that area (Nanus, 2000). Most of the raspberry fields are fertilized by inorganic, commercial fertilizers and may require irrigation between bloom and post-harvest (May–August) due to the decreased precipitation during these months (Menzies, 1999). Fields are irrigated by drip tape, which is buried in the soil along the edge of each row of berries, or by overhead sprinkler irrigation systems. Because of the study area also has a high concentration of dairy farms, considerable amounts of animal waste are produced. Corn and grasses, grown by dairies for feed purposes, are typically fertilized by spraying liquid manure from dairy lagoons (J. Gillies, personal communication, 1998). Land use north of the study area, in the British Columbia study area (Fig. 1), is dominantly raspberry fields with numerous commercial poultry barns dispersed throughout the region (Zebarth et al.,
1998). The British Columbia region is fertilized primarily by poultry manure (Wassenaar, 1995).

Well Locations

We measured water levels and collected water samples from 26 domestic wells (Fig. 2). The selection of the well locations depended partly on the permission of landowners and wells were chosen to span a distribution along roads in the study area. Most wells were located down-gradient from raspberry fields and dairy operations. Surface locations of wells were established with a global positioning system (±3–5 m) in the field. Elevations of well casings were surveyed to ±0.003 m above mean sea level with a rod and level (Stasney, 2000). Driller-well logs were analyzed to determine the depths and screen interval lengths of the sampling wells.

MATERIALS AND METHODS

Water Level Measurements

The depth to water was measured monthly for 22 months at accessible wells, before any water was removed from the well, with a depth-to-water meter with an electrical sounding tape. The corrected static water level (CSWL) represents the median measured depth to the water table with respect to the ground surface. The CSWL also represents the median vadose zone thickness. The depth below the water table of the sampling location was estimated by subtracting the CSWL from the well depth and represents the approximate depth below the water table (DBWT) from which a sample was collected.

Data from well logs and information from a local drilling company (D. Hayes, personal communication, 2002) indicate that the well screen lengths are typically 1.5 m or less. Due to the inability to measure water levels in several wells, the DBWT was estimated for these wells using information from drilling well logs, topographic maps, reported well depths, and static water-level measurements from wells in the general area.

Water Sampling

Twenty domestic wells were sampled monthly between April 1997 and January 1999 to characterize spatial and temporal water quality parameters in the study area (Fig. 2). Six additional domestic wells (Wells 6, 16, 24, 25, and 26; Fig. 2) were sampled intermittently. Sampling of Wells 6 and 24 through 26 depended on permission from landowners, and Well 16 was replaced by deep Well 25 in August of 1998. Water samples were collected consistently from the same location, which was the closest tap to the well. After static water levels were measured, the wells were purged at the standpipe for approximately 10 min, or until the specific conductance stabilized to remove any stagnant water. A 1-L flow-through cell was used to minimize atmospheric interactions. Water samples were collected in 500-mL, 2 M hydrochloric acid-washed Nalgene bottles (Nalge Nunc International, Rochester, NY) for nitrate + nitrite and nitrite (NO\(_2\)) analyses. All bottles were rinsed three times with the sample before collection, then placed in an iced cooler for transport.

Surface water samples were collected in November 1997 and February and May 1998, at 12 locations along a 2.4-km reach of Pangborn Creek, which dissects the central portion of the study area (Fig. 2). Pangborn Creek is a discharge zone for the Abbotsford–Sumas Aquifer and also receives runoff from nearby croplands. Samples for laboratory analysis were collected at each site by placing a 250-mL acid-washed Nalgene bottle at elbow depth in the water. The bottles were rinsed three times with stream water before collection, then placed in an iced cooler for transport. To assess NO\(_3^-\) sources in the study area we analyzed N isotopes sampled from ground water at nine wells and two surface water sites. Wells 2, 5, 7, 9, 10, 13, 14, 17, and 18 were selected because they represent a spatial distribution in the study area and had high NO\(_3^-\) concentrations that would improve the likelihood of identifying the source of contamination. We sampled each of the nine wells and two surface water sites in February, June, and October 1998. Water samples were collected in 1-L Nalgene bottles, which contained acid before collection to fix the solution, and then placed in an iced cooler for transport.

Laboratory Analyses

The ground and surface water samples were analyzed for NO\(_3^-\) and NO\(_2^-\) + NO\(_3^-\) at the Institute for Watershed Studies laboratory at Western Washington University, a laboratory certified by the Washington State Department of Ecology. Field duplicates (5% of all the samples collected) and lab duplicates (10% of the samples) were used for quality assurance. Dissolved oxygen was measured in a flow-through cell in the field with a Model 85 water analyzer (YSI, Yellow Springs, OH) with a detection limit of 0.1 mg L\(^{-1}\). The water samples were filtered the day of the sampling with a 0.45-μm membrane filter. Laboratory NO\(_3^-\) tests were conducted within 48 h of sampling using a spectrophotometric method (USEPA 354.1) with a detection limit of 0.0009 mg N L\(^{-1}\). Nitrate + nitrite samples were frozen and analyzed in the lab within 60 d of sampling using automated cadmium reduction (USEPA 353.2) with a detection limit of 0.006 mg N L\(^{-1}\). Water samples were shipped to Coastal Science Laboratories (Austin, TX) for isotopic analysis. The method used by Coastal Science Laboratories for measuring the two stable isotopes of N (\(^{14}\)N and \(^{15}\)N) in NO\(_3^-\) is based on methodology discussed by Velinsky et al. (1989). Coastal Science Labs estimated standard deviations for ground water N measurements to be 0.8 to 1.0% for samples of commonly encountered concentrations (0.5–5 mg N L\(^{-1}\)).

Data Analyses

The ground water NO\(_3^-\) concentrations, reported here as mg N L\(^{-1}\), are categorized according to Tesoriero and Voss (1997). They used NO\(_3^-\) concentrations greater than 3 mg N L\(^{-1}\) as an indicator of contamination due to anthropogenic effects and to predict aquifer susceptibility and vulnerability in the Puget Sound region. The 10 mg N L\(^{-1}\) MCL was used as a second NO\(_3^-\) concentration category. The water quality data were not normally distributed and heteroskedastic; therefore, we examined the ground water NO\(_3^-\) concentrations using nonparametric graphical and descriptive statistics. These analyses include boxplots showing the minimum value, the 25th quartile, the median value, the 75th quartile, and the maximum value (Carlson and Thorne, 1997) and Kendall’s τ correlation analysis (Helsel and Hirsch, 1992). The DBWT was used to separate NO\(_3^-\) concentrations measured from deep and shallow regions of the aquifer. Wells with DBWT levels less than 10 m were considered to be shallow, whereas wells with DBWT levels greater than 10 m were considered to be deep.

Stable isotope ratios were used to differentiate between inorganic, commercial fertilizers and animal waste as NO\(_3^-\) sources (Flipse and Bonner, 1985; Exner and Spalding, 1994; Wassenaar, 1995; Igbal et al., 1997). Stable isotope ratios are usually expressed in a δ format, which for N indicates the relative value of the \(^{15}\)N to \(^{14}\)N isotopic ratio as compared with the atmospheric (standard), and is expressed as:

\[
\delta^{15}\text{N}(\text{‰}) = \left(\frac{^{15}\text{N}_{\text{sample}}}{^{14}\text{N}_{\text{sample}}} / \frac{^{15}\text{N}_{\text{standard}}}{^{14}\text{N}_{\text{standard}}} \right) - 1 \times 10^3
\]
\[(\frac{\delta^{15}N}{\delta^{15}N_{\text{standard}}}) \times 1000\] \[\text{[1]}\]

where \(\delta^{15}N\) has units of per mil (‰), \(\delta^{15}N/\delta^{15}N_{\text{standard}}\) is the stable isotopic ratio of the sample, and \(\delta^{15}N/\delta^{15}N_{\text{standard}}\) is the ratio of the atmospheric standard. Interpreting \(\delta^{15}N\) data, however, can be difficult when the values are derived from ground water samples rather than from soil pore water beneath a source, because it is possible that the \(\delta^{15}N\) values may have been enriched by denitrification or reflect a mixture of sources (Herbel and Spalding, 1993; Fogg et al., 1998). To maintain consistency, we compared our \(\delta^{15}N\) results (Nanus, 2000) to those of Wassenaar (1995), who used three classes to categorize NO\(_3^–\) sources in the British Columbia study area from ground water–derived \(\delta^{15}N\) values. Wassenaar’s accepted range of \(\delta^{15}N\) values for inorganic commercially derived NO\(_3^–\) in ground water was −1 to +2‰, whereas animal waste sources had \(\delta^{15}N\) values greater than +8 to +16‰. Although human waste–derived NO\(_3^–\) is indistinguishable from animal waste, Cox and Kahle (1999) qualitatively estimated that septic tank effluent accounted for less than 4% of the total N entering the ground water as NO\(_3^–\) in the region. Values between +2 and +8‰ were considered indicative of a mix of animal and inorganic, commercial fertilizer sources or derived from natural soil nitrate.

**RESULTS AND DISCUSSION**

**Nitrate Concentrations and Distributions**

A total of 504 ground water samples from 26 wells were analyzed over the 22-month period. Except for some outliers, NO\(_3^–\) concentrations were usually below the detection limit of 0.0009 mg N L\(^{-1}\). The minimum, maximum, and median NO\(_3^–\) concentration for each well is listed in Table 1. Most (63%) of all values and 17 well median values were greater than 3 mg N L\(^{-1}\) and indicate probable anthropogenic contamination (Tesoriero and Voss, 1997). In addition, 38% of all values and 12 of the median values were greater than the 10 mg N L\(^{-1}\) MCL. Of the 36 surface water samples analyzed, 34 had NO\(_3^–\) concentrations above 3 mg N L\(^{-1}\), and seven samples were greater than 10 mg N L\(^{-1}\).

To show the spatial distribution of NO\(_3^–\) concentrations, we plotted median values from each well location and two representative surface water sampling sites, along with the agricultural land use in the study area (Fig. 3). The distribution of elevated concentrations is characteristic of nonpoint agricultural sources and is consistent with the historical documentation of agricultural NO\(_3^–\) contamination in many parts of the Abbotsford–Sumas Aquifer area (Liebscher et al., 1992; Wassenaar, 1995; Erickson, 1998; Zebart et al., 1998; Cox and Kahle, 1999; Hii et al., 1999). The concentrations in some wells remained high throughout the duration of the study. In the last month of sampling (January 1999), NO\(_3^–\) concentrations in 14 wells were greater than 3 mg N L\(^{-1}\), and 9 wells yielded concentrations greater than the 10 mg N L\(^{-1}\) MCL.

Median NO\(_3^–\) concentrations for the wells are categorized by magnitude and by DBWT levels (Table 1; Fig. 4). Kendall’s \(\tau\) correlation analysis (Helsel and Hirsch, 1992) confirmed a statistically significant relationship between NO\(_3^–\) and DBWT, with higher NO\(_3^–\) concentrations at shallower depths and lower NO\(_3^–\) concentrations at deeper depths (Kendall’s \(\tau = -0.380; p < 0.001\)). Despite the significant correlation, there was

| Table 1. Well depth and values measured between April 1997 and January 1999 from wells in the northwestern Washington study area.† |
|---|---|---|---|---|---|---|
| Well number | Depth (m) | Median CSWL | Median DBWT | Minimum NO\(_3^–\) | Median NO\(_3^–\) | Maximum NO\(_3^–\) | DO (mg L\(^{-1}\)) | \(\delta^{15}N\) (‰) |
| Shallow |
| 24 | 9.1 | 6.4 | 2.7 | 0.2 | 25.3 | 28.7 | 8.3 | – |
| 14 | 8.5 | 7.2 | 1.3 | 18.2 | 22.9 | 32.4 | 4.4 | 5.6 |
| 18 | 13.1 | 7.8 | 5.3 | 13.4 | 19.9 | 32.3 | 5.6 | 5.5 |
| 2 | 10.4 | 7.1 | 3.3 | 12.7 | 14.5 | 16.4 | 6.2 | 3.4 |
| 1 | 9.1 | 1.5 | 7.6 | 9.8 | 12.1 | 14.2 | 5.3 | – |
| 10 | 15.2 | 9.2 | 6.1 | 7.9 | 11.2 | 12.6 | 5.6 | 1.5 |
| 17 | 11.3 | 8.8 | 2.5 | 9.0 | 10.6 | 11.6 | 2.6 | 6.9 |
| 13 | 12.2 | 8.8 | 3.4 | 8.4 | 10.4 | 25.5 | 4.8 | 5.0 |
| 16 | 18.3 | 10.7 | 7.6 | 5.3 | 8.7 | 12.3 | 6.4 | – |
| 22 | 10.4 | 7.5 | 2.9 | 3.6 | 5.3 | 10.3 | – | – |
| 15 | 10.7 | 7.8 | 2.9 | 2.0 | 3.6 | 9.9 | 4.5 | – |
| 13 | 13.7 | 4.2 | 9.5 | 1.5 | 2.3 | 2.5 | 6.1 | – |
| 19 | 13.7 | 3.9 | 9.8 | 1.5 | 2.3 | 3.3 | 6.4 | – |
| 11 | 21.6 | 12.9 | 8.7 | 0.006 | 2.1 | 5.4 | 3.9 | – |
| 4 | 9.1 | 7.1 | 2.1 | 0.04 | 1.8 | 6.6 | 3.3 | – |
| 8 | 18.3 | 9.3 | 9.0 | 0.4 | 1.2 | 13.0 | 2.2 | – |
| 12 | 22.9 | 13.1 | 9.7 | <0.006 | 0.0 | 0.03 | 0.8 | – |
| Deep |
| 25 | 30.5 | 11.4 | 19.1 | 11.7 | 13.5 | 14.2 | 0.5 | – |
| 6 | 29.0 | 15.5 | 13.5 | 11.3 | 12.3 | 14.1 | 7.9 | – |
| 9 | 21.6 | 9.3 | 12.3 | 4.2 | 12.1 | 26.7 | 1.4 | 9.5 |
| 5 | 30.5 | 19.1 | 11.4 | 8.0 | 11.2 | 16.1 | 3.5 | 8.3 |
| 7 | 23.5 | 11.4 | 12.1 | 5.5 | 8.9 | 9.9 | 4.3 | 0.3 |
| 26 | 24.4 | 8.0 | 16.4 | 5.5 | 7.6 | 11.5 | Na | – |
| 23 | 22.9 | 3.8 | 19.1 | 0.1 | 0.8 | 2.4 | 8.3 | – |
| 21 | 17.4 | 5.8 | 11.6 | 0.002 | 0.0 | 0.04 | 0.54 | – |
| 3 | 28.0 | 7.4 | 20.7 | <0.006 | 0.0 | 0.03 | 2.0 | – |

† Values include the median corrected static water level (CSWL); median well depth below the water table (DBWT); the minimum, median, and maximum nitrate concentration (NO\(_3^–\)); median dissolved oxygen value (DO); and median nitrogen isotope ratio (\(\delta^{15}N\)) value. The wells are categorized into shallow DBWT levels (<10 m) and deep DBWT levels (>10 m) and by NO\(_3^–\) magnitude. Values that are below the detection limit are identified by <0.006.
considerable variability among wells (Fig. 4), indicating that the concentration of ground water NO$_3^-$ was not just a simple bivariate correlation with DBWT. Gelinas (2000) used multivariate analyses (principal component analysis and metric and nonmetric clustering) to confirm the inverse relationship between NO$_3^-$ and DBWT and reveal three distinct groups of wells. Deep wells with moderately high NO$_3^-$ may represent an up-gradient NO$_3^-$ source, and were distinct from shallow wells with very high NO$_3^-$ concentrations (possibly from a local agricultural source), and both were distinct from low NO$_3^-$ “anomaly” wells, which were possibly influenced by denitrification.

Deep Aquifer Contamination

Deep Wells 5, 6, 7, and 9, located less than 1 km from the international boundary, have high NO$_3^-$ concentrations (Table 1; Fig. 2), as do the more southerly deep wells (Wells 25 and 26). It is unlikely that the elevated NO$_3^-$ concentrations at Wells 5, 6, 7, and 9 are related to northwestern Washington sources because it would
require large vertical gradients to deliver \( \text{NO}_3^- \) to these well depths, and the vertical hydraulic gradients are minimal in the region (Zebarth et al., 1998). The unconfined aquifer in this region is a highly conductive mixture of gravel and sand where ground water flow is dominantly horizontal to the southeast (e.g., Cox and Kahle, 1999; Stasney, 2000). The elevated \( \text{NO}_3^- \) concentrations in these wells are probably the result of sources further north (up-gradient) in British Columbia. It is also likely that the elevated \( \text{NO}_3^- \) concentrations of 13.5 and 7.6 mg N L\(^{-1}\) at the more southerly Wells 25 and 26, respectively, are related to British Columbia sources because of their depth. We estimated a regional background concentration in the study area to be 10.7 mg N L\(^{-1}\), which is the median concentration of the \( \text{NO}_3^- \) measurements from the contaminated deep wells in the study area (5, 6, 7, 9, 25, 26). This value is in the range predicted by Hii et al. (1999), who used hypothetical modeling to simulate a similar stratification and deep aquifer contamination observed in the British Columbia study area (Hii et al., 1999; Zebarth et al., 1998). Using flow net analyses and a 20 mg N L\(^{-1}\) load to the water table, Hii et al. (1999) predicted \( \text{NO}_3^- \) concentrations averaging 10 mg N L\(^{-1}\) crossing the international boundary into some locations in northwestern Washington.

Nitrogen isotope ratios analyzed from water samples from three deep wells (Wells 5, 7, and 9) were used to test a British Columbia source hypothesis. Two of these wells, Wells 5 and 9, had the highest median \( \delta^{15}N \) values of all the wells measured in the study area, 8.3 and 9.5‰, respectively (Table 1; Fig. 5). These values suggest an animal source, probably poultry manure (Wassenaar, 1995). Eighty percent of Wassenaar’s ground water \( \delta^{15}N \) values were between +7 and 16‰. The median \( \delta^{15}N \) value from the other deep well, Well 7, was 0.3‰, which is surprisingly small, but consistent with the types of inorganic, commercial fertilizers used in the British Columbia study area. Although limited, when coupled with
Fig. 5. Land use in the northwestern Washington study area and associated nitrogen isotope ratios ($\delta^{15}N$) at nine well locations and two surface water sites in the northwestern Washington study area. Nitrogen isotope ratios are proportional to the size of the well symbol. Individual well numbers are shown by the well symbols.

The proximity of the wells to the international border and depth, the $\delta^{15}N$ values suggest up-gradient sources. Temporal variations in climate and land use in British Columbia can result in NO$_3^-$ concentrations in excess of the 10 mg N L$^{-1}$ MCL and add to the difficulty in managing localized sources. Nitrate concentrations from several shallow and deep piezometers, grouped west and east of Judson Lake in the British Columbia study area, showed pronounced fluctuation of NO$_3^-$ concentrations beginning in 1995 and ending in 1997 (Hii et al., 1999). The increase was partly correlated to a rise in water table that was observed during the same period. The rise in the water table can liberate accumulated NO$_3^-$ from the vadose zone. Moreover, the higher water table corresponds to a larger recharge, which would increase the leaching of NO$_3^-$ from the vadose zone to the water table. Although our well measurements started in April 1997, Well 9 had been intermittently measured, using comparable laboratory procedures, by the Washington State Department of Ecology between June 1993 and March 1997 (D. Garland, personal communication, 1997). The NO$_3^-$ time series, combined with
Fig. 6. Time series of nitrate concentrations (mg N L$^{-1}$) measured at Well 9. “Historic” values were measured by the Washington State Department of Ecology and “current” values are from this study.

our data between April 1997 and January 1999, clearly depicts a significant plume migrating past Well 9 between mid-1995 and mid-1997 that is similar in shape to those observed in the British Columbia study area (Fig. 6). The drop in concentration is probably due to a combination of lower recharge and a change in N loading. Unfortunately, we do not have a consistent time series before April 1997 at the other deeper wells in our study area to document a similar pattern, but we believe this plume could have migrated through the entire study area.

**Shallow Aquifer Contamination**

Bivariate and multivariate statistics support a stratified distribution of NO$_3^-$ in the aquifer and suggest that land use in our study area contributed to ground water contamination. This was expected because the majority of the land use in the study area is agricultural (Fig. 3). If we assume a background NO$_3^-$ concentration around 10 mg N L$^{-1}$ in the aquifer due to up-gradient sources (using the contaminated deep-well median), then measured NO$_3^-$ concentrations equal to or greater than the MCL would suggest localized sources. Median values of eight shallow wells (1, 2, 10, 13, and 17) meet this criterion (Table 1), whereas Wells 14, 18, and 24 had median NO$_3^-$ concentrations of 22.9, 19.9, and 25.3 mg N L$^{-1}$, respectively (Table 1). Measured NO$_3^-$ concentrations in the wells also showed the most variation during the study (Fig. 3), suggesting that they are more responsive to land use changes because of their exceptionally shallow depth (<5 m).

Degraded surface water quality in Pangborn Creek also suggests contamination in the study area due to local contributions (Nanus, 2000). Median NO$_3^-$ values in Pangborn Creek increased from about 6.0 mg N L$^{-1}$ near Pangborn Lake to about 9.0 mg N L$^{-1}$ at the eastern-most site (Fig. 2). The highest NO$_3^-$ values measured in the creek were in May 1998 (median 8.8 mg N L$^{-1}$) and the lowest in November 1997 (median 5.9 mg N L$^{-1}$), and this range relates to seasonal rains and spring nutrient applications in the region. Median NO$_3^-$ values for two representative sites along Pangborn Creek are shown in Fig. 3, and these data are consistent with the results of Wills (1998) and Butkus et al. (2000). Pang-
Low Nitrate Anomalies

A number of shallow wells (Wells 4, 8, 11, 12, 19, and 20) and deep wells (Wells 3, 21, and 23) in the study area produced NO$_3^-$ concentrations below 3 mg N L$^{-1}$ and in some cases, values near zero (Table 1). We also show a weak correlation between NO$_3^-$ and dissolved oxygen, and this relationship suggests the effects of land use and/or fertilizer application. Our finding confirms a relationship found by Gelinas (2000) between low NO$_3^-$ and low dissolved oxygen (Table 1) at some of these locations (Wells 3, 4, 8, 11, 12, and 21). Wells 19 and 20 are shallow wells down-gradient of a large gravel mining facility in the southern portion of the study area, which suggests that land use may have influenced some of these low NO$_3^-$ concentrations (Fig. 3). The low NO$_3^-$ values may also be the result of denitrification facilitated by organic carbon or iron, or both. Denitrification has been shown to be reducing NO$_3^-$ concentrations near the water table in the aquifer, especially in areas where manure is applied (Paul and Zebarth, 1997a,b; Paul et al., 1997) and in riparian zones (Tesoriero et al., 2000). Although Wassenaar (1995) concluded that no significant denitrification was occurring deeper in the aquifer, it does occur in isolated zones having iron-containing minerals, where reduced forms of iron (e.g., pyrite) serve as electron donors in denitrification reactions (Tesoriero et al., 2000). We did not measure iron or byproducts associated with denitrification (i.e., N$_2$ gas).

CONCLUSIONS

For a period of 22 months, we measured NO$_3^-$ concentrations in shallow and deep domestic wells in a study area in northwestern Washington and confirmed elevated NO$_3^-$ concentrations, characteristic of nonpoint agricultural sources. The results suggest that NO$_3^-$ concentrations are stratified in the unconfined aquifer due to sources up-gradient in British Columbia and local sources in the study area. The highest NO$_3^-$ concentrations (>20 mg N L$^{-1}$) were found in the shallow region of the aquifer and were linked to northwestern Washington sources by using a combination of N isotope results, local agricultural land use relationships, and contaminated surface water in the study area. We also measured NO$_3^-$ concentrations in excess of 10 mg N L$^{-1}$ in deep wells in the study area and consider agricultural land use in British Columbia to be the NO$_3^-$ source deeper in the aquifer based on $\delta^{15}$N results and hydrogeology. Because of the low vertical gradients in the aquifer, long flow paths would be required to transport NO$_3^-$ to the deep wells near the international boundary. In addition,
N isotope ratios from two of the three deep wells sampled near the international boundary suggest animal manure as the source, which is consistent with poultry manure fertilization practices in British Columbia.

The identification of two possible regional sources and a NO$_3^-$ stratification in the aquifer adds to the difficulty in managing agricultural practices in northwestern Washington. Despite the implementation of best management practices for decades, elevated NO$_3^-$ concentrations have persisted in ground water and surface water in the region. This has driven the development of the State of Washington’s Dairy Nutrient Management Act (1998), which requires all dairy farms to develop and implement dairy nutrient management plans by 2003. To assess and refine these management plans, the magnitudes of the northwestern Washington contribution of NO$_3^-$ to the aquifer must be quantified. Our results show that it will be difficult to determine these contributions because of the spatial and temporal variability of NO$_3^-$ transported into the study area from up-gradient sources in British Columbia. In our analysis, we suggest a NO$_3^-$ stratification in the aquifer based on a background NO$_3^-$ concentration of about 10 mg N L$^{-1}$. However, the exact background level and spatial extent of background concentrations is not clear, nor is the degree of vertical and horizontal mixing of the two sources in the aquifer. Separating source contributions is complicated further by temporal changes in climate and up-gradient land use and aquifer heterogeneities that may cause ground water flow anomalies and localized denitrification. Our results suggest that using water measurements from domestic wells offers a cost-efficient approach in assessing NO$_3^-$ concentrations and sources as well as identifying NO$_3^-$ stratified regions in the aquifer. However, wide-spread implementation and monitoring of nested piezometers and soil lysimeters, coupled with nutrient loading information and extensive aquifer characterization and modeling, may be necessary to monitor NO$_3^-$ accurately and manage N loading in northwestern Washington. Because of these difficulties, conservationists and farmers are faced with a problematic task and economic challenges when having to consider multiple sources and up-gradient contamination to refine management plans.

REFERENCES


