

Use of Tree-Ring Chemistry to Document Historical Ground-Water Contamination Events

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Abstract

The annual growth rings of tulip trees (*Liriodendron tulipifera* L.) appear to preserve a chemical record of ground-water contamination at a landfill in Maryland. Zones of elevated iron and chlorine concentrations in growth rings from trees immediately downgradient from the landfill are closely correlated temporally with activities in the landfill expected to generate iron and chloride contamination in the ground water. Successively later iron peaks in trees increasingly distant from the landfill along the general direction of ground-water flow imply movement of iron-contaminated ground water away from the landfill. The historical velocity of iron movement (2 to 9 m/yr) and chloride movement (at least 40 m/yr) in ground water at the site was estimated from element-concentration trends of trees at successive distances from the landfill. The tree-ring-derived chloride-transport velocity approximates the known ground-water velocity (30 to 80 m/yr). A minimum horizontal hydraulic conductivity (0.01 to 0.02 cm/s) calculated from chloride velocity agrees well with values derived from aquifer tests (about 0.07 cm/s) and from ground-water modeling results (0.009 to 0.04 cm/s).

Introduction

Ground-water investigations at hazardous-waste sites often confront a lack of historical data regarding the transport of subsurface contaminants. Such information may provide data necessary to assess the future movement of the contamination. Moreover, historical information on the transport velocity of conservative tracers is useful as an indicator of ground-water velocity, which can be used with a measured hydraulic gradient to calculate aquifer horizontal hydraulic conductivity. The necessary data may be difficult to obtain using existing methodology because tracer tests are time-consuming, aquifer pumping tests may discharge or affect the distribution of contaminated ground water, and slug tests are valid only for the screened interval of the well (Lohman, 1979). The purpose of this study is to determine whether such information can be derived from the chemical

content of growth rings of tulip trees (*Liriodendron tulipifera* L.) at a hazardous-waste landfill in Maryland (Figure 1). To test the hypothesis that the chemistry of tree rings may provide an areal and temporal record of geochemical changes in the ground water, the distribution of iron (Fe) and chlorine (Cl) in the growth rings of tulip trees near the hazardous-waste landfill was compared to the record of historical activity within the landfill. Fe and Cl are constituents of the ground-water contamination and have been shown not to be translocated across growth-ring boundaries in other studies (Hall, 1987; Wardell and Hart, 1973; Tout and others, 1977; McClenahan and others, 1989).

Tree-ring chemistry has been related to soil contamination (McClenahan and others, 1987, 1989; Meisch and others, 1986; Bowers and Melhuish, 1988; Bondietti and others, 1989) and surface-water contamination (Sheppard and Funk, 1975). A number of studies also have shown an apparent relation between air pollution and tree-ring chemical composition (Robitaille, 1981; Symeonides, 1979; Kardel and Larsson, 1978; Baes and McLaughlin, 1984; Bondietti and others, 1989) and between leaf and stem chemistry and ground-water chemistry (Zielinski and Schumann, 1987; Kalisz and others, 1988). Meredith and Hites (1987) noted that polychlorinated biphenyls (PCBs) were present in the bark of trees growing near a PCB-contaminated landfill, but

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they found none in the wood. However, the potential for using tree-ring chemistry to examine ground-water chemistry has not been examined. Because the availability of an element for uptake by trees may also be influenced by site microclimate and competition from other elements (Berry, 1986; Brooks, 1972), absolute concentrations probably are of secondary importance to elemental trends when relating tree-ring chemistry to ground-water chemistry.

Site Description and History

The study area is near a landfill (Figure 1) in Aberdeen Proving Ground, Maryland, that was periodically used for disposal of munitions, chemical-warfare agents, industrial solvents from mustard-gas manufacturing plants, and petroleum products. The types of chemical-warfare agents disposed potentially includes all that were developed prior to the late 1950s. The material was disposed on the ground or in unlined trenches to a depth near the surface of the water table.

The subsurface material in the vicinity of the landfill consists of unconsolidated sand and silt, with laterally discontinuous lenses of clay. The depth to the water table ranges from about 2.5 m near the edge of the fill to about 0.3

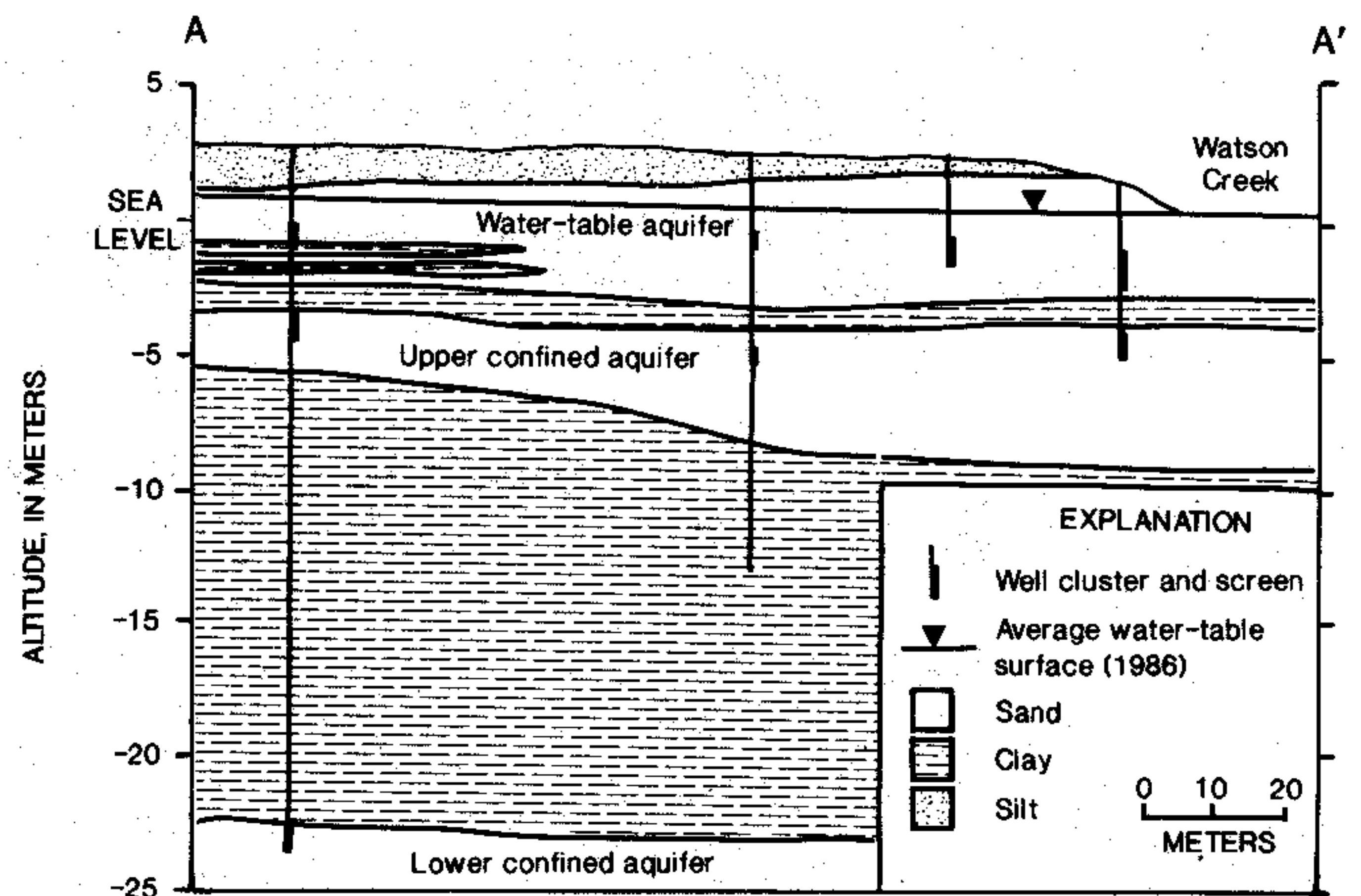


Fig. 2. Cross section A-A'.

m near the shoreline of Watson Creek. Borehole data and analysis of ground water indicate that much of the contamination is prevented from moving deeper than about 8 m below land surface by the presence of a laterally continuous layer of clay at that depth (Figure 2).

The shallow ground water is recharged by precipitation infiltration. Shallow ground-water movement beneath the disposal area is from southwest to northeast, toward Watson Creek, where it discharges (Figure 3). The ground-water velocity through the area is about 30-80 m/yr, based on ground-water flow modeling of the site and assuming a porosity of 0.3 percent (Vroblesky and others, 1989).

Ground-water samples collected quarterly during 1986-87, and weekly for selected wells, show that the water-table aquifer within the contaminated zone contains higher concentrations of iron (Fe) (19.8 to 87.3 mg/l), chloride (Cl) (52 to 2,150 mg/l), manganese (0.45 to 17 mg/l), zinc (0.88 to 8 mg/l), arsenic (0.11 to 2 mg/l), and aliphatic and aromatic organic solvents than are found in adjacent, uncontaminated ground water. Uncontaminated ground water contains approximately 0.1 mg/l Fe, 2-8 mg/l Cl, 0.05 mg/l manganese, 0.3 mg/l zinc, and no arsenic or organic solvents.

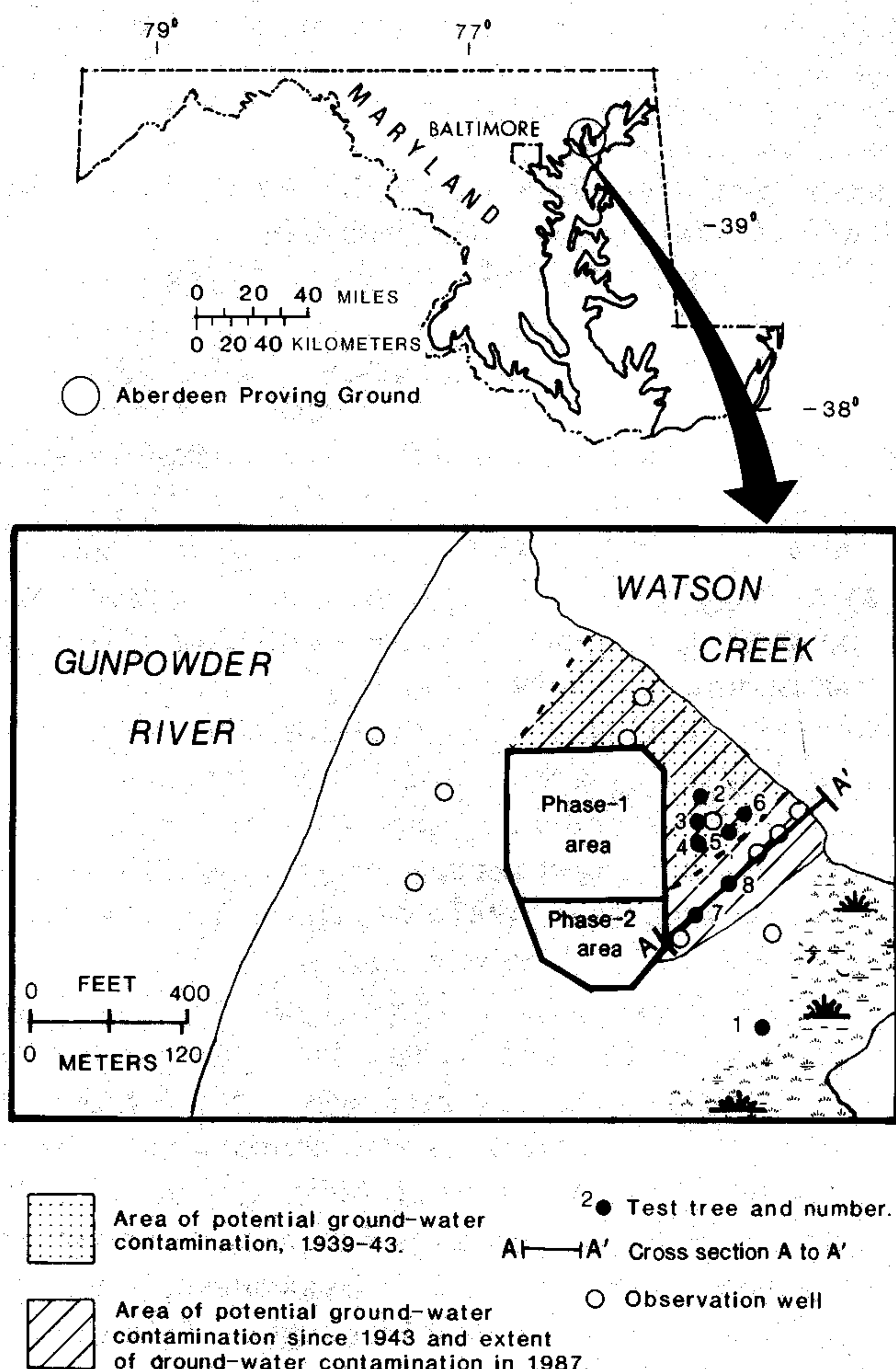


Fig. 1. Location of test trees, ground-water sampling points, boundaries of the landfill through time, the associated areas of potential or known ground-water contamination, and cross section A-A'.

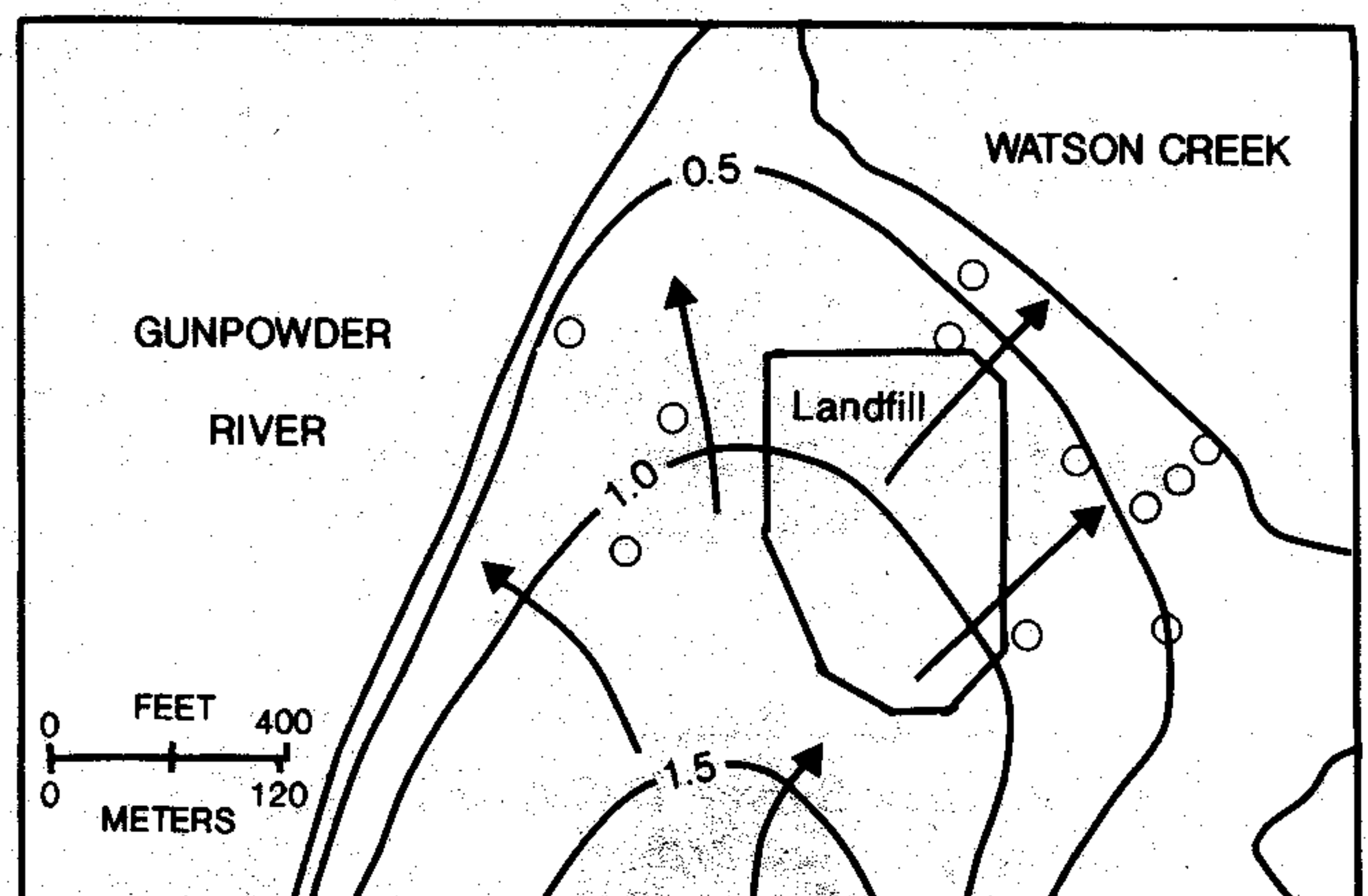


Fig. 3. Water-table map showing directions of shallow ground-water flow. The contour interval is 0.5 m, measured relative to sea level.

A major source of dissolved Fe appears related to the disposal of organic compounds. Contaminated ground water at the site is anaerobic, probably due to microbial degradation of disposed organic compounds (Vroblecky and others, 1989), whereas uncontaminated ground water in the area is aerobic. Reduction of ferric-oxide grain coatings in the presence of anaerobic ground-water contamination probably accounts for much of the Fe in solution. Because the disposal pits were unlined and the aquifer material is relatively permeable, ground-water contamination by the organic compounds probably occurred soon after disposal. Thus, it is likely that anaerobic ground-water conditions also existed during periods of maximum landfill use. Such periods would be expected to produce relatively high concentrations of dissolved Fe.

A variety of chemicals, termed decontaminants, were applied extensively to the site to render the chemical-warfare agents less toxic. The dominant decontaminants were DANC (an acronym for Decontaminating Agent NonCorrosive, a chloramide dissolved in tetrachloroethane), hypochlorite-based bleaching agents, and chlorinated lime. Decontaminants were used whenever the site activities, such as burial or munitions recovery, involved chemical-warfare agents. Munitions recovery and destruction were accomplished by removing, defusing, incinerating, or detonating the ordnance.

Much of the Cl contamination in the ground water appears to be due to the application of decontaminants (Vroblecky and others, 1989). Prior to 1951, the dominant decontaminants used at the landfill were DANC and lesser amounts of chlorinated lime (Gary Nemeth, 1988, U.S. Army Environmental Hygiene Agency, personal commun.). Limited amounts of Cl contamination may have occurred from the use of DANC. However, by 1953, after exhausting the stockpiles of DANC, hypochlorite-based bleaching agents became the dominant decontaminants (Gary Nemeth, 1988, U.S. Army Environmental Hygiene Agency, personal commun.) and would have been substantially greater sources of Cl.

Although information on historical ground-water chemistry of the site does not exist, much of the history of disposal and decontamination activities at the site is relatively well-known as a result of documentation by record reviews (Yon and others, 1978), written eyewitness accounts (Dickey, 1978), and interviews (Gary Nemeth, 1988, U.S. Army Environmental Hygiene Agency, personal commun.) with personnel involved with site activities. The landfill can be divided into two major areas, based on the timing of specific disposal operations. Disposal operations began in the phase-1 area (Figure 1) in the late 1930s. Maximum activity was in 1946 when tons of captured munitions and chemical-warfare agents were disposed on the ground or in unlined trenches. Some containers were reported to be leaking at the time of burial (Yon and others, 1978).

Disposal operations in the phase-1 area declined after 1946, and the last reported pit was excavated in 1948 (Yon and others, 1978). The primary decontaminant used during phase-1 operations was DANC, with lesser amounts of chlorinated lime (Gary Nemeth, 1988, U.S. Army Environ-

mental Hygiene Agency, written commun.). After a period of site cleanup in 1950, site operations were concentrated in the phase-2 area (Figure 1) of the landfill until 1965, when site-cleanup operations began (Dickey, 1978) and continued through the late 1960s and early 1970s (Nemeth and others, 1983).

Disposal operations in the phase-2 area (Figure 1) began in 1944. The landfill was officially closed to disposal in 1950 (Dickey, 1978); however, an additional disposal pit in the phase-2 area was excavated in 1953 and used until at least 1957 (Yon and others, 1978).

Little is known about site activities during the period 1957-65, although some decontamination apparently took place. Dickey (1978) reports that much site cleanup had been completed when he visited the landfill in 1965. The site was fenced, and access was to the phase-2 area; therefore, it is likely that most disposal or cleanup operations during 1957-65 would have occurred in the phase-2 area.

Activity at the landfill increased again in 1965 when the site was designated as an Explosive-Ordnance-Disposal Training area (Dickey, 1978). These operations involved munitions recovery and destruction. Peak activity during this period was in 1972, when tons of decontaminants were spread across the site. The training operations may have taken place in both the phase-1 and phase-2 areas of the landfill, but the descriptions of the types of ordnance removed imply that operations were concentrated primarily in the phase-1 area (Gary Nemeth, 1988, U.S. Army Environmental Hygiene Agency, personal commun.).

Methods

The tulip trees analyzed in this study were 50 to 100 years old. Seven trees grew in areas of known ground-water contamination and three in areas believed to be unaffected by the landfill (Figure 1). Two of the trees from uncontaminated locations are outside the area shown in Figure 1 but within a 300-m radius of the fill. Increment corings were extracted from trees at breast height, and borers were rinsed between corings with acetone.

Prior to element analysis, cores were oven-dried, mounted with cyanoacrylate glue into lucite holders, and shaved to a flat surface with a stainless-steel surgical blade. Nondestructive multielemental analysis of individual rings was performed by proton-induced X-ray emission (PIXE). In general, odd-numbered rings were analyzed beginning with the 1987 ring and ending with those from the 1930s. In a few cases, however, consecutive ring series were selectively analyzed as well, and, in some trees, fewer rings were analyzed. Fe and Cl in the wood of tulip trees were analyzed as elemental iron and chlorine. Comparisons of concentrations on either side of the heartwood/sapwood boundary were examined in background trees to assess the potential for lateral translocation in response to heartwood/sapwood differentiation.

Hydrogeologic data from the site were collected from 37 monitoring wells. Hydraulic gradients were determined using synoptic water-level measurements and data from digital water-level monitors that recorded water levels at 15-minute intervals over periods of up to 18 months.

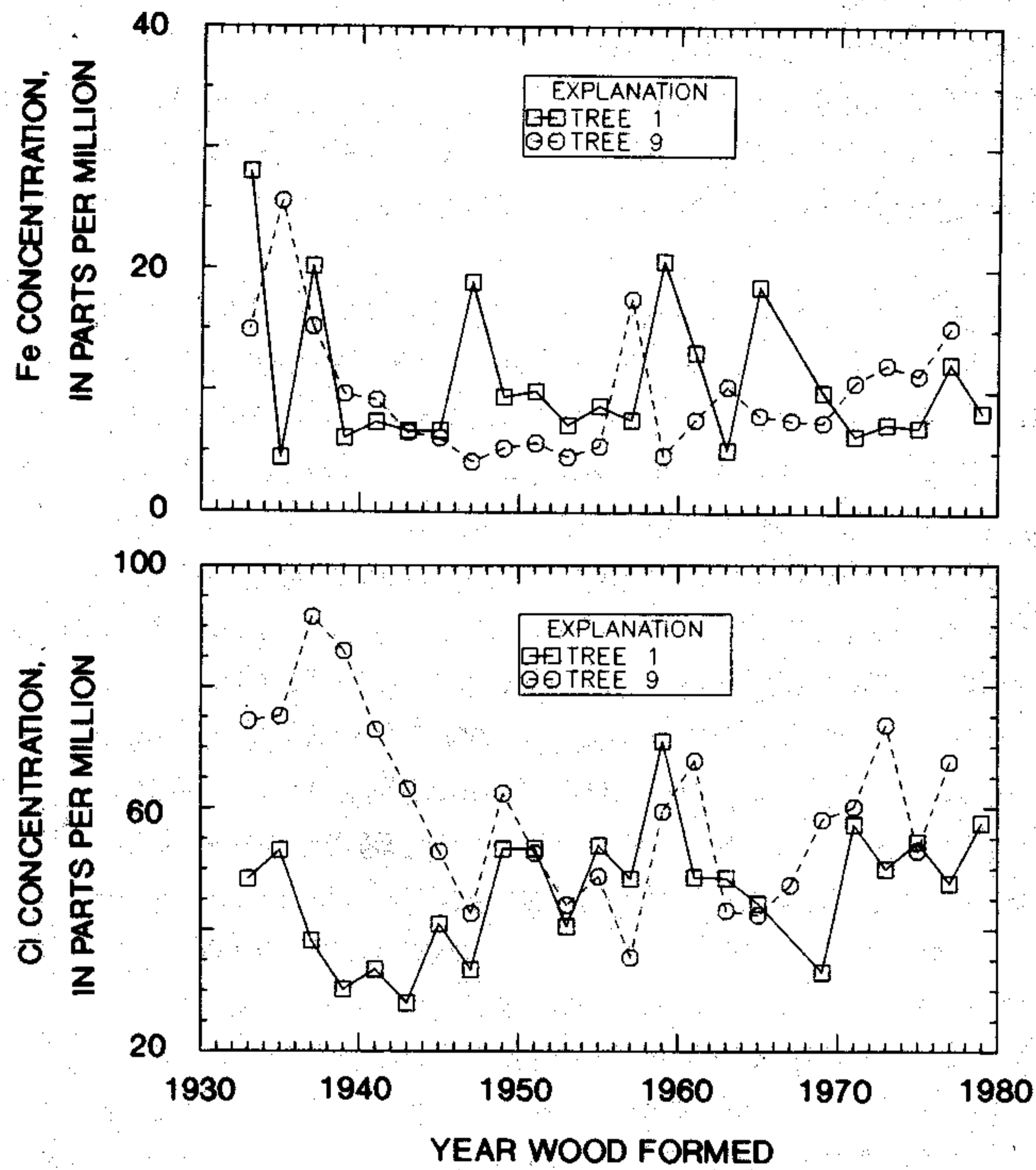


Fig. 4. Fe and Cl concentrations in background trees.

Hydraulic conductivity was calculated from slug-test data using the Hvorslev (1951) method.

Water samples were collected from selected onsite wells five times on a quarterly basis, beginning in December 1985 and ending in December 1986. Additional samples were collected more frequently from three wells within the area of existing ground-water contamination.

The ground-water velocity was calculated from the output of a three-dimensional digital ground-water flow model (McDonald and Harbaugh, 1984) that simulated steady-state ground-water flow in the upper two aquifers at the site (Vroblesky and others, 1989). Because the site is in a rural area where ground water is not pumped, recent hydrologic analyses were considered representative of historical ground-water conditions.

Results

Unlike the background trees (Figure 4), the Fe and Cl concentrations in most trees near the landfill typically became elevated across several growth rings during periods of maximum landfill activity (Figure 5). The Fe concentration in tree 4 increased beginning in about 1939 and reached a peak in 1947, followed thereafter by a decline (Figure 5). A

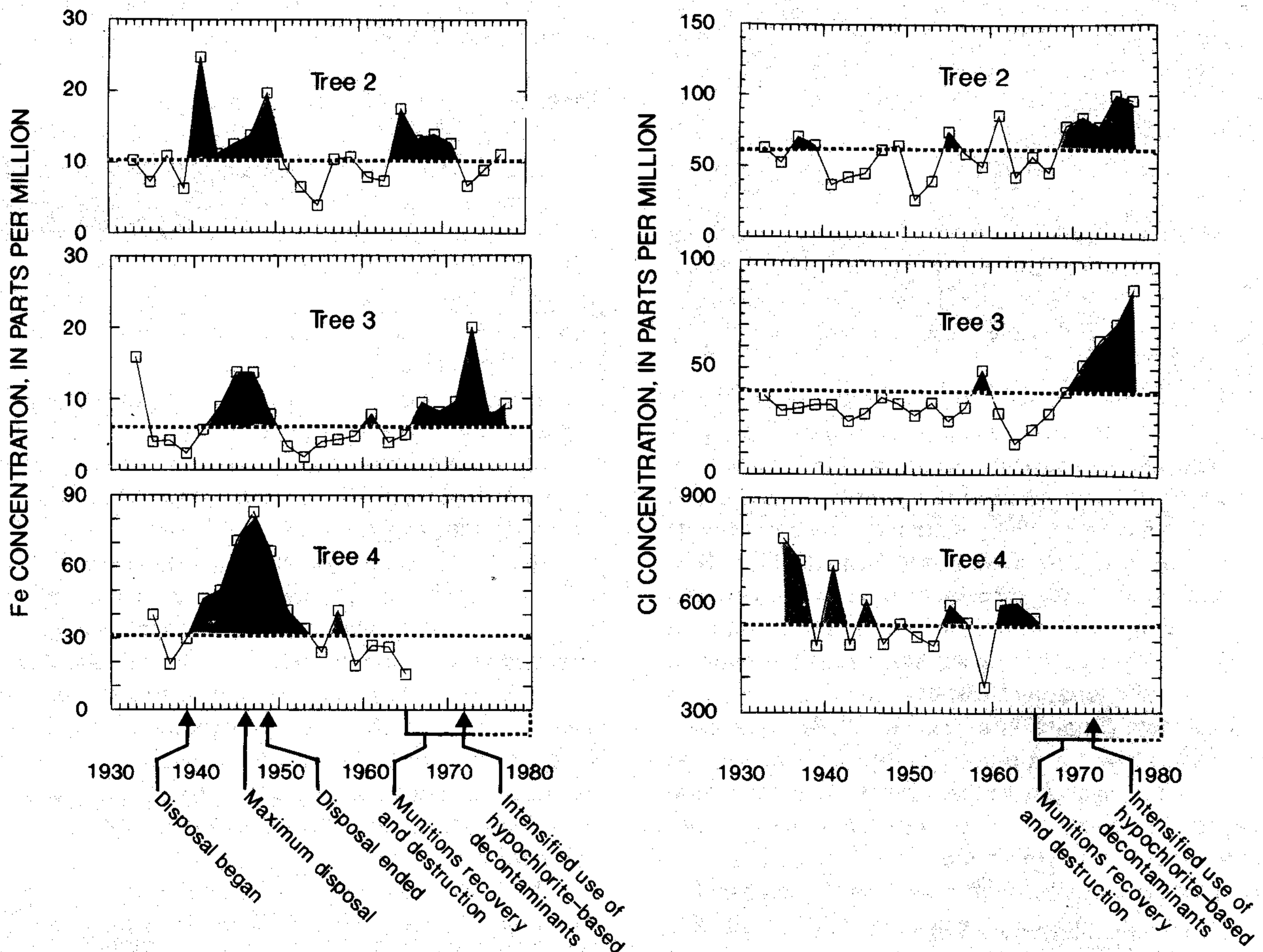


Fig. 5. Fe and Cl concentrations in trees located 30 m from the phase-1 area of the landfill in the direction of ground-water movement. Dashed lines within graph represent mean concentrations for the period of record. Periods of major site activity are noted along the x-axis.

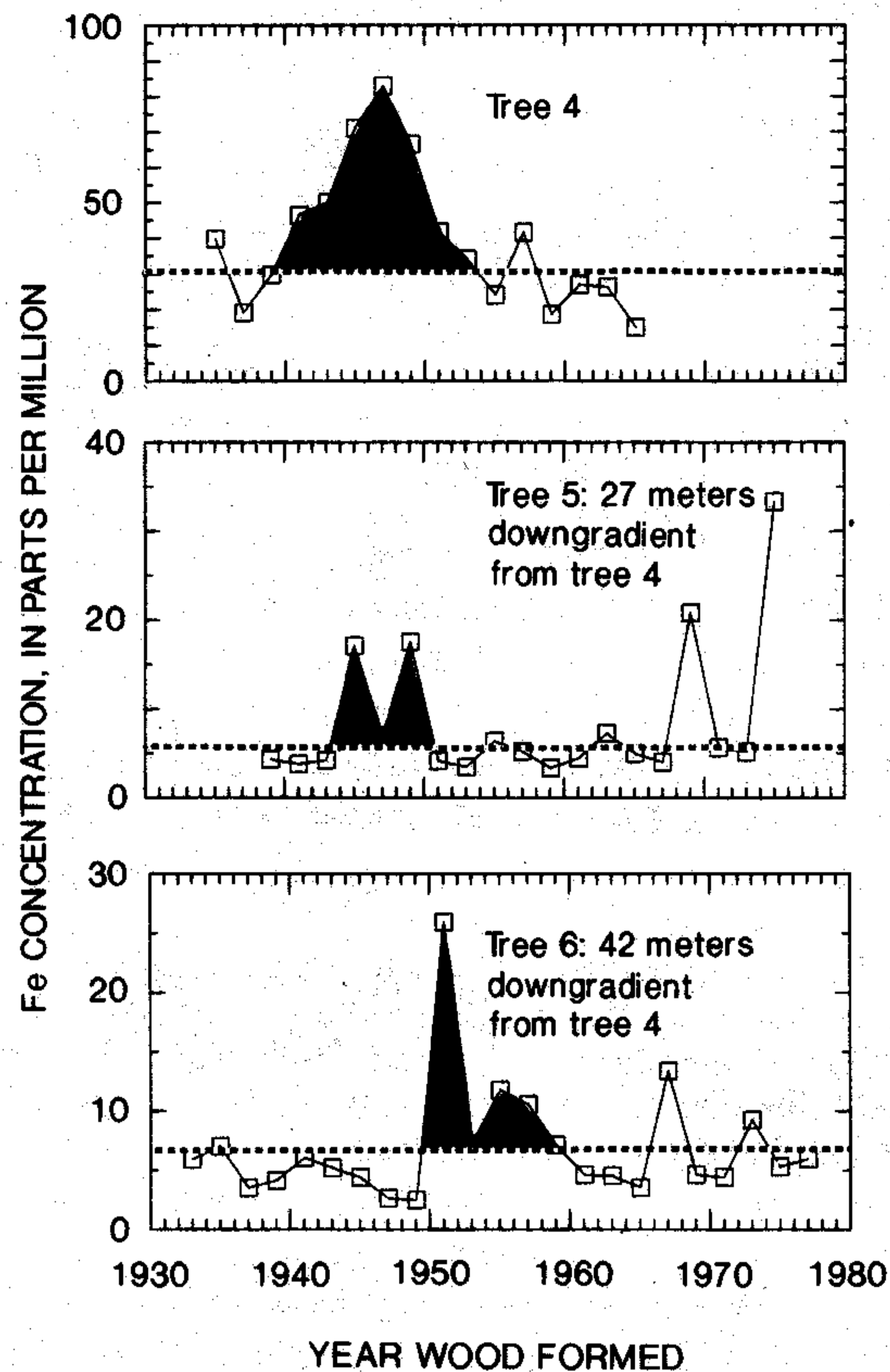


Fig. 6. Fe concentrations in trees successively distant from the phase-1 area of the landfill in the general direction of ground-water movement. Dashed lines represent mean concentrations for the period of record. Shaded peaks are used in text for velocity calculations.

similar trend was shown in tree 3, immediately to the north. Tree 2, north of trees 3 and 4, also had above average concentrations of Fe during this same period but did not have a similarly pronounced Fe peak. Increased Fe concentrations again occurred in trees 2 and 3 beginning in about the 1965 growth ring. Trees 2, 3, and 4 are located about 30 m

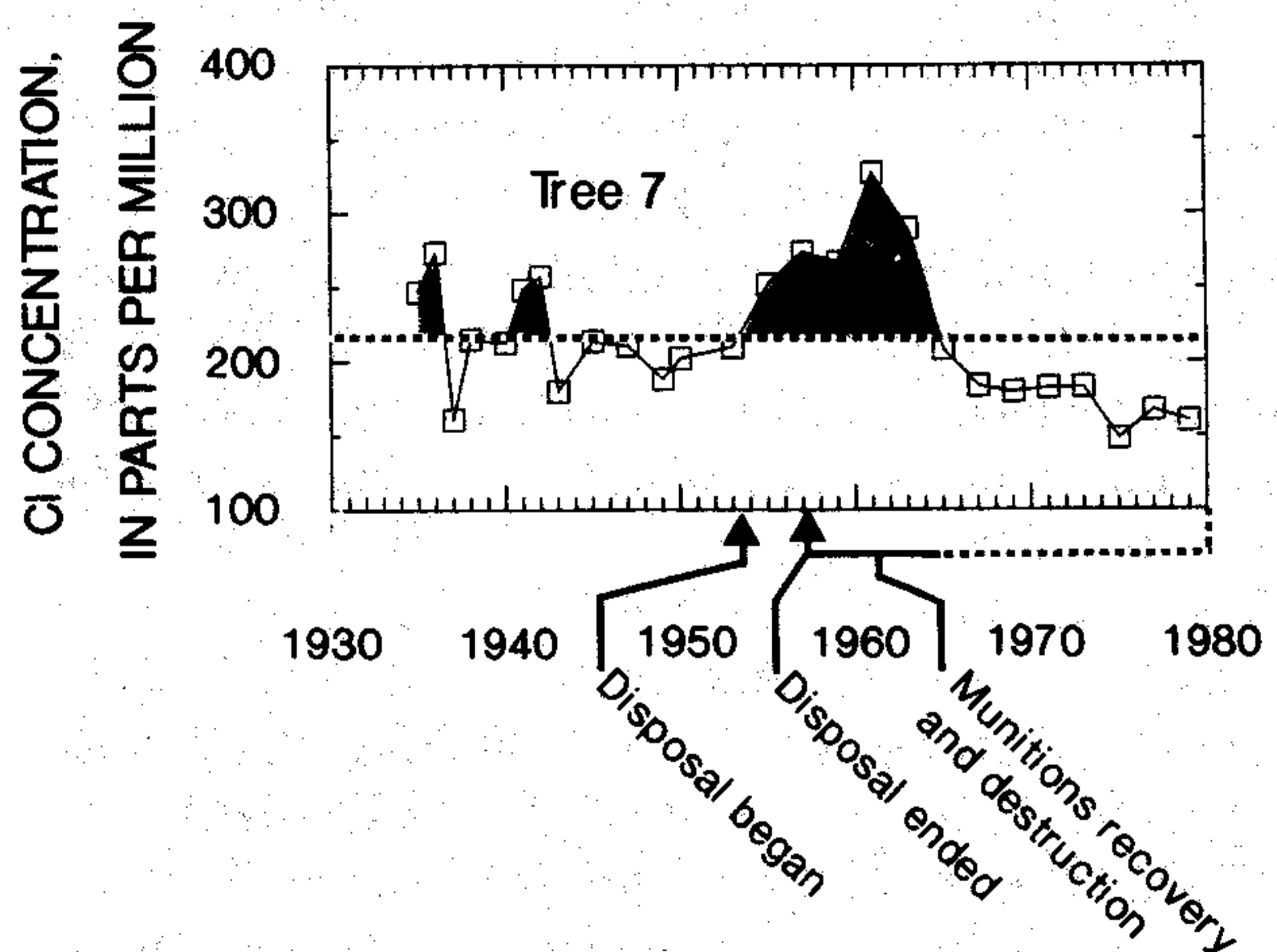
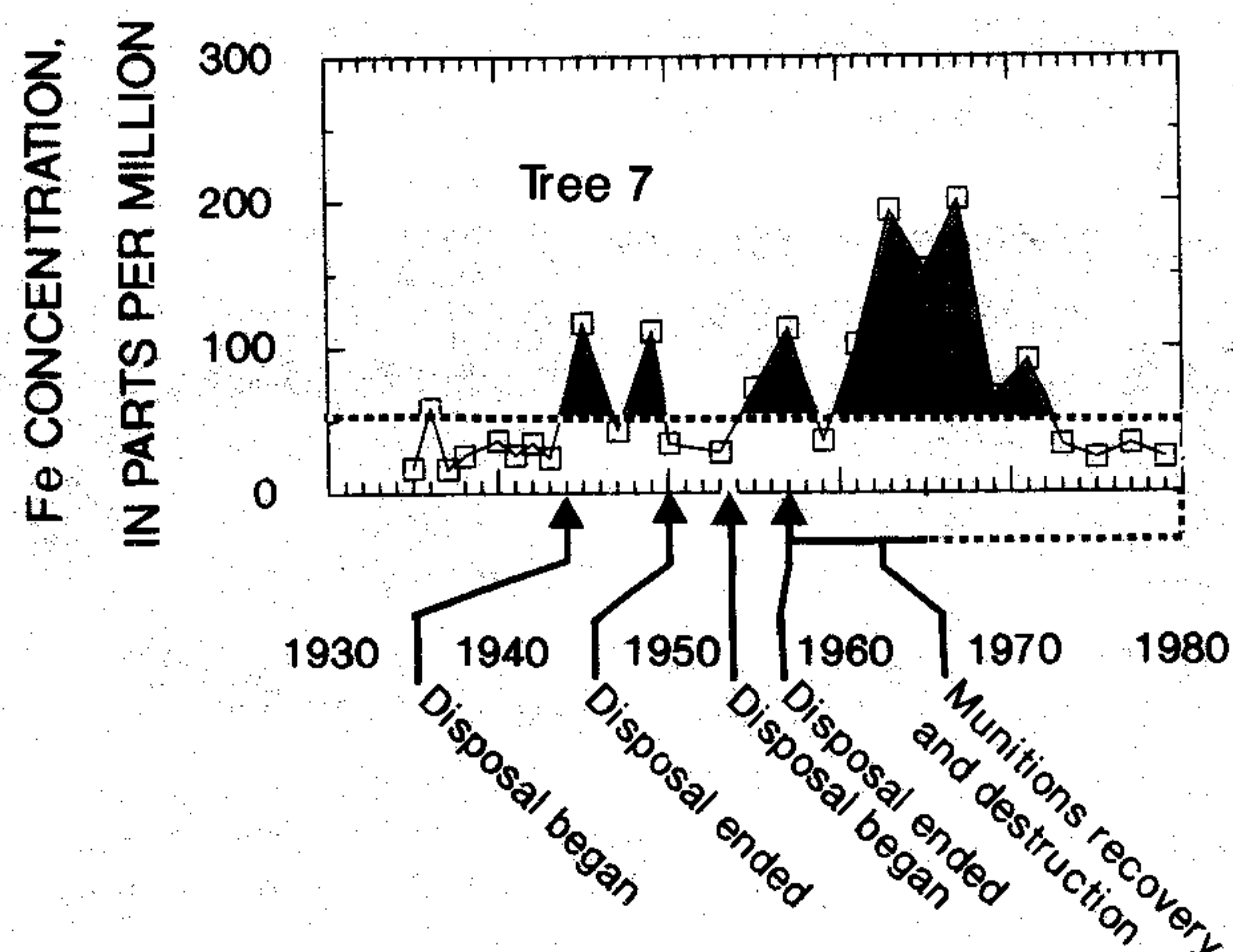


Fig. 7. Fe and Cl concentrations in tree 7, located 24 m from the phase-2 area of the landfill in the direction of ground-water movement. Dashed lines within graph represent mean concentrations for the period of record. Periods of major site activity are noted along the x-axis.

from the phase-1 area of the landfill along the direction of ground-water flow.

The Cl-concentration trends in the 1940 growth rings in trees 2, 3, and 4 showed no apparent similarity to the Fe-concentration trends. Although some of the Cl concentrations in tree 4 in the 1940s are above the mean concentration, the trend is irregular and the greatest concentrations are in the 1935 growth ring. Increasing trends of Cl concentration occurred in trees 2 and 3 in the mid to late 1960s.

Tree 5, about 27 m from tree 4 in the approximate direction of ground-water flow, showed increased concentrations of Fe beginning in about 1944-45 (Figure 6) and extending to about 1949. Tree 6, about 15 m from tree 5 and about 42 m from tree 4 in the approximate direction of ground-water flow, contain elevated concentrations of Fe beginning with the 1951 growth ring and extending to about 1959.

Tree 7 (Figure 7) is within the present-day (1988) zone of maximum ground-water contamination approximately 24 m from the southeastern end of the phase-2 area. Fe peaks in tree 7 did not occur until about 1945 (Figure 7). Concentrations were relatively low in growth rings from 1950-53, but increased in later growth rings. Additional peaks occurred from 1961-72. With the exception of two short-lived peaks in about 1936 and 1941, Cl concentrations in tree 7 showed no substantial increase until about 1954-55. Concentrations thereafter remained well above the mean until about 1965.

Tree 8, about 40 m from tree 7 along the approximate direction of ground-water flow, showed elevated Cl levels beginning at the same time as in tree 7, and extended to about the 1967 growth ring (Figure 8). Growth rings in tree 8 prior to 1949 were not analyzed.

Neither Fe nor Cl was preferentially enriched on either side of the heartwood-sapwood boundary. However, in cores from both contaminated and uncontaminated areas, there was a substantial increase in Fe concentrations in the most recently formed rings (about 1982-88) (Figure 9).

Discussion

The variability in concentrations of Fe and Cl detected in growth rings of background trees indicate that the trees are sensitive to environmental variations, such as yearly or seasonal effects. Such influences would also be expected to occur in trees near the landfill. However, in addition to such short-term variations, trees 2, 3, and 4, near the phase-1 area of the landfill in the direction of ground-water flow, showed a generally synchronous increase in Fe concentrations in growth rings corresponding to times when the adjacent part of the landfill was active (Figure 5). Increasing concentrations began soon after disposal began. Peak concentrations in tree 4 occurred in the 1947 growth ring, corresponding to the time of maximum disposal in 1946. Concentrations of Fe declined within a few years after the last disposal pit was excavated in 1948.

Increasing Fe concentrations in growth rings again occurred beginning in about 1965. The source of Fe in growth rings younger than 1965 is uncertain because of the limited amount of information on specific site activities during that time. However, 1965 was when the landfill was designated as an Explosive-Ordnance-Disposal Training area. Site training would have at least included disturbance of the soil and disposed material, destruction of munitions, and application of decontaminating agents.

The timing of Fe peaks successively later in trees along the general direction of ground-water flow appears to document the movement of Fe contamination away from the landfill (Figure 6). The transport velocity required to produce the offset peaks between trees 4 and 5 is 4.5 to 9 m/yr. The transport velocity required to produce the offset peaks between trees 5 and 6 is 2.1 to 3 m/yr and to produce the offset between trees 4 and 6 is 3.5 to 5 m/yr. The similarity of velocities provides evidence to support the hypothesis that the succession of Fe peaks in trees 4, 5, and 6 reflect the timing of Fe transport in the ground-water plume.

Disposal operations in the phase-1 area of landfill do not appear to have left a recognizable signature in the Cl trends of trees 2, 3, and 4 (Figure 5). The lack of response is expected because the dominant source of Cl, hypochlorite-based decontaminants, was used in substantial quantities only after the phase-1 area was closed. Cl concentrations in trees 2 and 3 increased in the late 1960s and continued to increase during the 1970s (Figure 5). A potential source of Cl during this period would have been the use of hypochlorite-based decontaminants. The application of such decontaminants was intensified in 1972 (Gary Nemeth, 1988, U.S. Army Environmental Hygiene Agency, personal commun.), a period that coincides with high Cl concentrations in the growth rings of trees 2 and 3.

Tree 7, located along the direction of ground-water flow from the phase-2 area of the landfill, also shows increased concentrations of Fe in growth rings that formed during periods of onsite activity (Figure 7). Fe peaks occur in 1945 and 1949, during the first period of activity (1944-50), when disposal of organic compounds was greatest. Additional Fe peaks occur in 1955-57 and 1961-77 and also correspond with Cl peaks during the same interval. Increased Cl concentrations also occur at the same time in tree 8

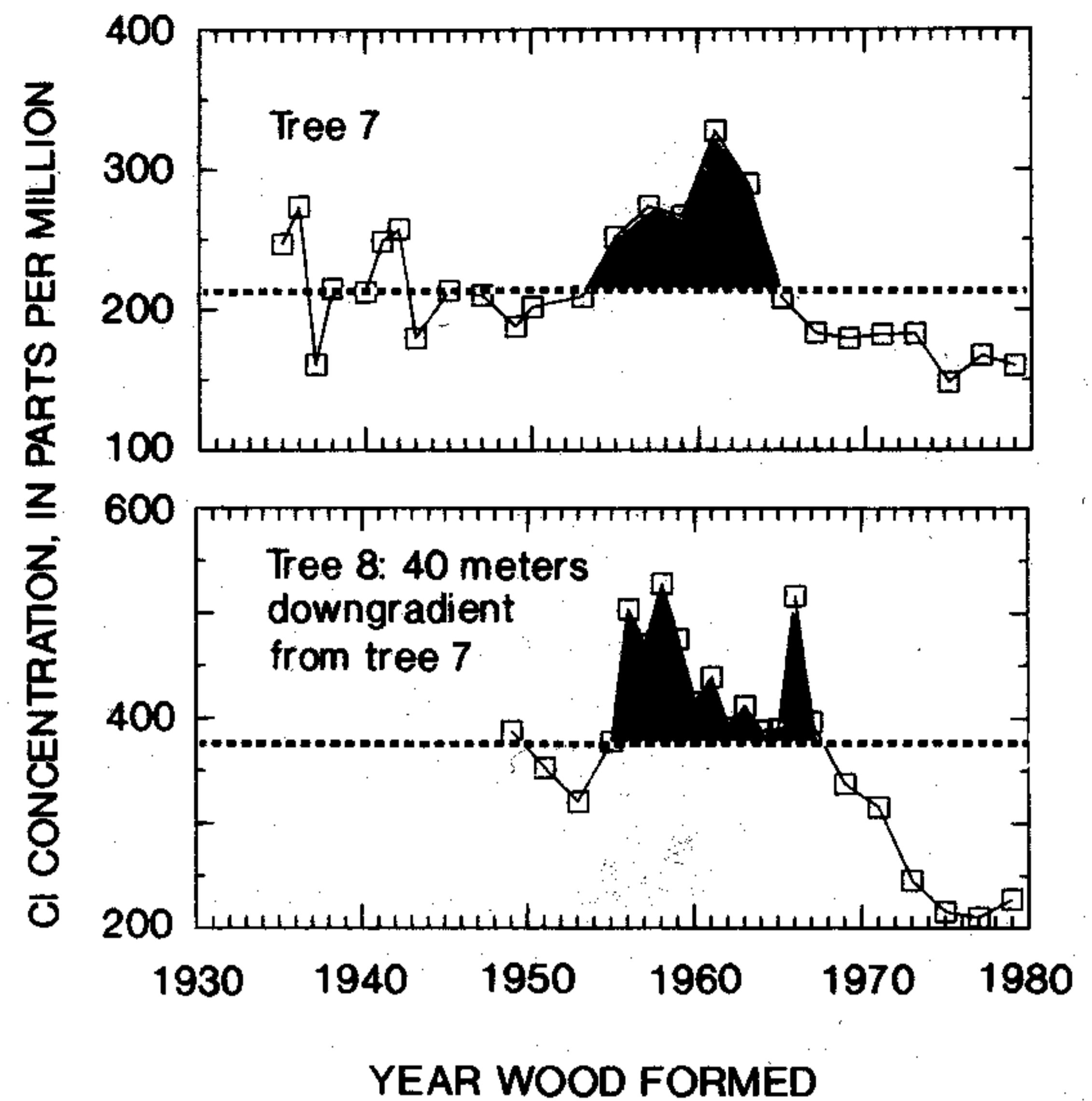


Fig. 8. Cl concentrations in trees successively distant from the phase-2 area of the landfill in the general direction of ground-water movement. Dashed lines represent mean concentrations for the period of record. Shaded peaks are used in text for velocity calculations.

(Figure 9), implying that the Cl and Fe trends represent a related event. The most likely event is the second period of disposal (1953-57) and subsequent site-cleanup operations (beginning about 1957).

Support for the hypothesis that the trees record movement of ground-water contamination can be seen by comparing transport velocity and horizontal hydraulic conductivity calculated from tree-ring data to values calculated by other methods. If increased Cl concentration in tree rings reflect ground-water-contaminant migration, it is then feasible to approximate an historical transport velocity of Cl through the water-table aquifer. Because the Cl peaks in trees 7 and 8 are synchronous, a maximum transport velocity cannot be calculated. However, if it is assumed that the peaks represent movement of ground-water contamination, then the transport between trees 7 and 8 appears to have occurred within one year. Thus, a minimum transport velocity of 40 m/yr can be estimated. The conservative nature of Cl dictates that its velocity should approximate that of

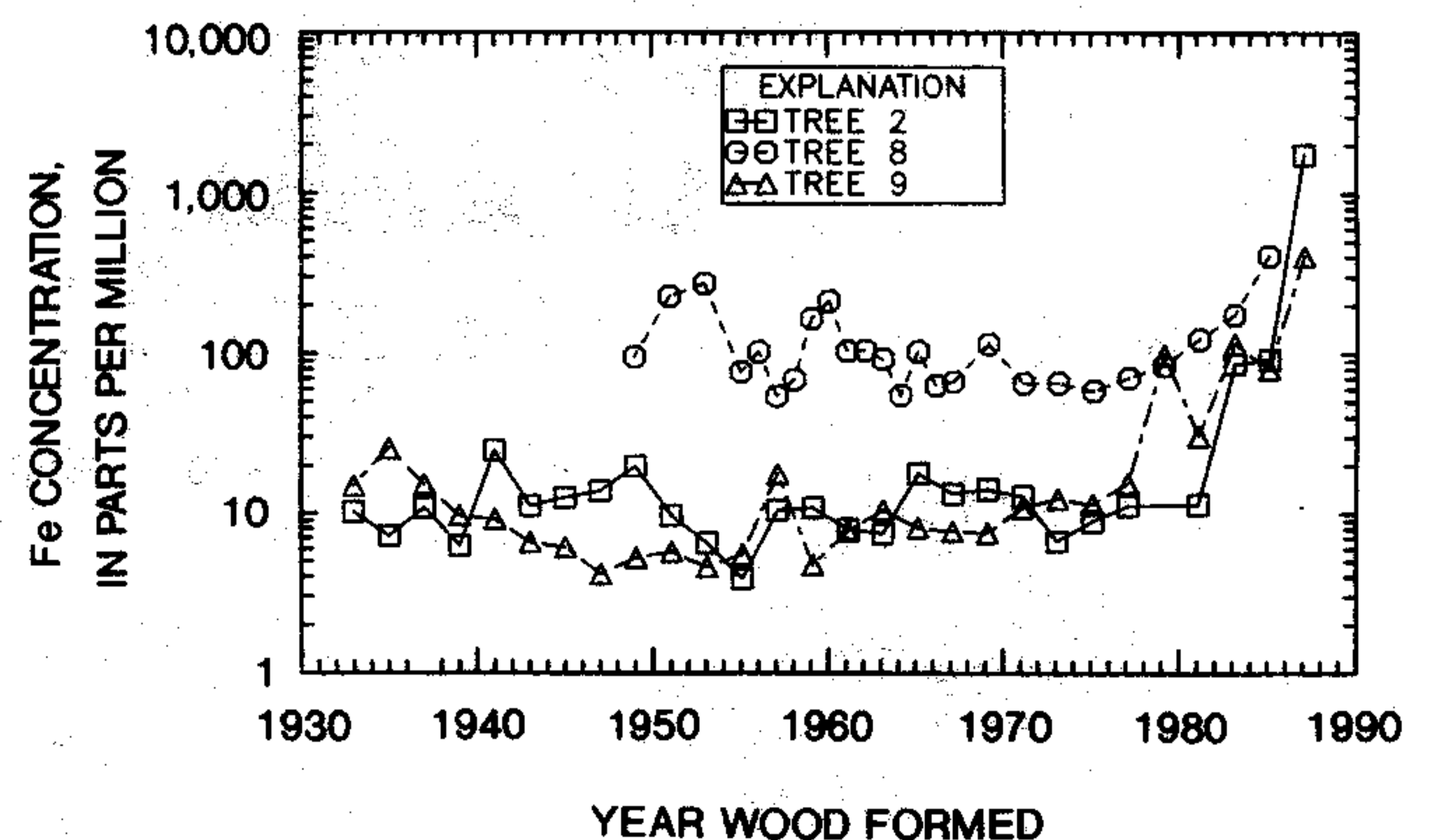


Fig. 9. Concentrations of Fe in selected trees showing increase in the outermost rings.

ground water. The tree-ring-derived velocity agrees well with the ground-water velocity (30-80 m/yr) calculated from ground-water-flow modeling (Vroblesky and others, 1989).

The minimum ground-water velocity estimated from the movement of Cl between tree 7 and tree 8 (40 m/yr) can be used in conjunction with the measured hydraulic gradient and an estimated porosity to estimate horizontal hydraulic conductivity from a modification of Darcy's Law,

$$K = vn/I,$$

where K is the horizontal hydraulic conductivity, v is the average linear velocity (40 m/yr), n is the porosity, and I is the hydraulic gradient. Using an average hydraulic gradient (based on field measurements) of 0.003 and a range of porosity typical for sand of 0.25 to 0.50 (Freeze and Cherry, 1979), a range of $K = 0.01$ cm/s to 0.02 cm/s is calculated. Although this K represents a minimum value, it agrees well with the K value calculated from three slug tests at the site (about 0.07 cm/s) and from a ground-water model (Vroblesky and others, 1989) of the site (0.009 cm/s to 0.04 cm/s). The close correlation of tree-ring derived K values to those calculated by independent methods strengthens the hypothesis that tree-ring chemistry documents temporal changes in ground-water chemistry.

Data from tree rings younger than 1980 were not compared to landfill activities because of the substantial increase in Fe concentrations in the outermost growth rings of the trees examined (Figure 9). The similar response of trees both inside and outside the contaminated areas implies that the increase is unrelated to ground-water contamination. Substantial increases of Fe in the outermost growth rings have also been reported in other studies (Baes and McLaughlin, 1984; Meisch and others, 1986), citing acid deposition from air pollution as a possible source.

Acid deposition by emissions from an industrial plant about 7 km west (typically upwind) of the site was considered as a possible source for the high Fe concentrations. Sulfur-dioxide emissions from the plant were at a maximum during the periods 1963-69 and since 1983. Although some trees show an increase in Fe corresponding approximately to 1963-69, most do not. Moreover, the increasing Fe concentrations in the outermost growth rings began before 1983 in some trees. Although it cannot be discounted, emissions from the plant do not appear to have produced an identifiable response in the trees.

Perhaps a more likely explanation is that the elevated levels of Fe in the outermost rings reflect the distribution of metabolic activity within the xylem. The outermost vessels of angiosperms contain the greatest amount of water in the sapwood because they are involved in the transport of large quantities of water and dissolved solutes from the roots to the crown of the tree (Stewart, 1966). Tree cores in this investigation were dehydrated in an oven. The evaporative process concentrates dissolved solutes in the residual water until they eventually precipitate out of solution. Thus, the increased Fe in the outermost growth rings may represent a combination of Fe incorporated into the wood structure and Fe that precipitated out of the residual water during evaporative drying of the cores. If so, the effects of residual

Fe precipitation would be limited to the outermost growth rings because the inner rings no longer actively conduct sap.

In either case, the increased Fe concentrations obscure the relation between tree-ring chemistry and ground-water chemistry in the outermost growth rings. Therefore, this study is only of the rings that formed during the period of 1930-1979, which includes the time of maximum landfill use and maximum probable ground-water contamination.

The lack of preferential enrichment of Fe or Cl on either side of the heartwood/sapwood boundary in background trees at the site implies that neither element is laterally translocated from ring to ring. These findings agree with those of other researchers who found that Fe (Hall, 1987; McClenahan and others, 1989) and Cl (Wardell and Hart, 1973; Tout and others, 1977) were not translocated across ring boundaries. The findings by McClenahan and others (1989) are particularly relevant because they deal with tulip trees. Thus, with the possible exception of the outermost rings, the yearly levels of Fe and Cl represent immobile constituents of rings that apparently reflect differences in the chemical environment during formation.

Conclusions

In the absence of historical data on the ground-water chemistry, it cannot be stated unequivocally that tree-ring chemistry documents past periods of ground-water contamination; however, a variety of factors suggests such a relation. Zones of elevated iron and chlorine concentrations in growth rings from trees immediately downgradient from the landfill are closely correlated temporally with activities in the landfill expected to generate iron and chloride contamination in the ground water. Successively later Fe peaks in trees increasingly distant from the landfill along the general direction of ground-water flow implies movement of Fe-contaminated ground water away from the landfill. Moreover, ground-water velocity and aquifer horizontal hydraulic conductivity calculated from the growth-ring chemistry agree well with those calculated by independent methods. Thus, tree-ring chemistry appears to be a potential tool for examining chemical histories of ground water. One important utility of such information is that it may be used to calculate horizontal hydraulic conductivity of the shallow ground-water flow system. Additional research needs include an improved understanding of the species-specific and metabolic controls on the uptake and incorporation of specific elements by trees. Use of this method at a variety of sites also would help determine the range of applications and constraints.

Acknowledgments

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