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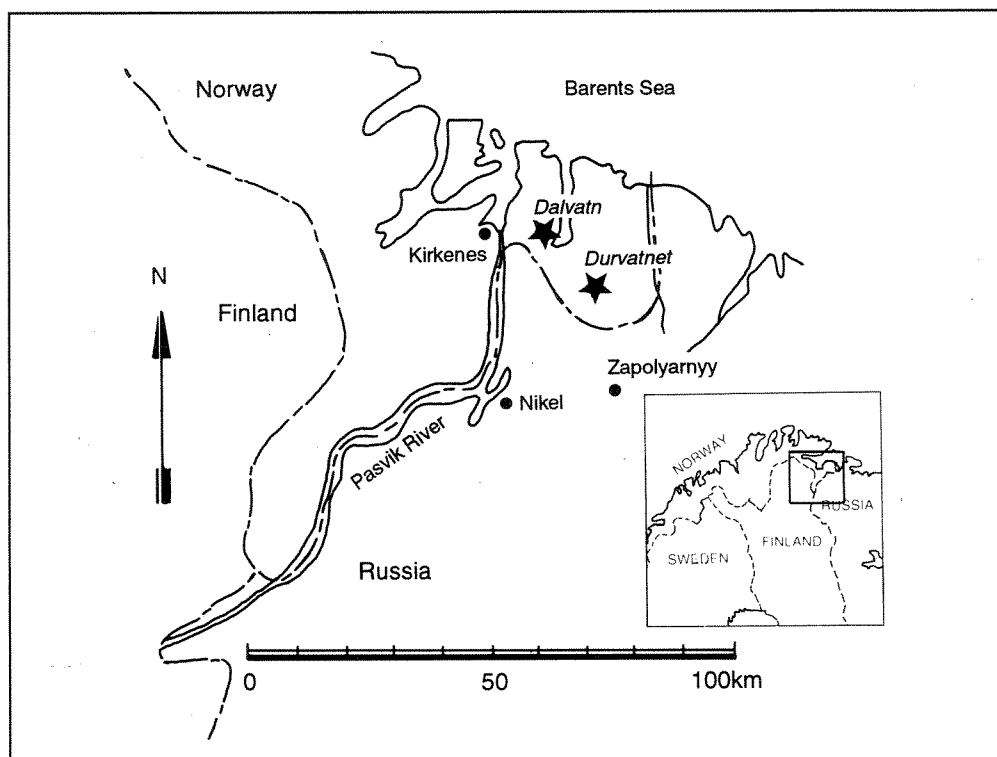
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# Trace Metal Pollution in Eastern Finnmark, Norway, as Evidenced by Studies of Lake Sediments.



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<b>Hovedkontor</b> Postboks 69, Korsvoll 0808 Oslo 8 Telefon (47 2) 23 52 80 Telefax (47 2) 95 21 89	<b>Sørlandsavdelingen</b> Televeien 1 4890 Grimstad Telefon (47 41) 43 033 Telefax (47 41) 44 513	<b>Østlandsavdelingen</b> Rute 866 2312 Ottestad Telefon (47 65) 76 752 Telefax (47 65) 78 402	<b>Vestlandsavdelingen</b> Breiviken 5 5035 Bergen - Sandviken Telefon (47 5) 95 17 00 Telefax (47 5) 25 78 90	<b>Akvaplan-NIVA A/S</b> Søndre Tollbugate 3 9000 Tromsø Telefon (47 83) 85 280 Telefax (47 83) 80 509
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Ekstrakt:

Sedimentkjerner fra Dalvatn og Durvatnet i Sør-Varanger er datert ved gammastråle-spektroskopi, og analysert for tungmetallene Cd, Co, Cu, Ni, Pb og Zn, samt radionuklider. De største sedimentasjonshastighetene for Co, Cu og Ni sammenfaller i tid med aktivitetene ved smeltverkene på Kola. Sedimentasjonshastigheten for Pb øker de siste 100 årene i et mønster som trolig reflekterer en generell forurensning av den nordlige halvkule. Radionuklider fra atmosfæriske prøvesprengninger av kjernefysiske bomber er registrert i sedimentkjerner fra begge innsjøene. I Dalvatn kunne man også spore nedfall fra Chernobyl-ulykken.

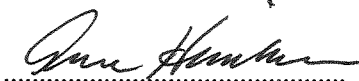
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2. Datering av sedimenter
3. Tungmetaller
4. Radionuklider

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## **Trace Metal Pollution in Eastern Finnmark, Norway, as Evidenced by Studies of Lake Sediments**

by

Stephen A. Norton

Geological Sciences<sup>1</sup>, University of Maine, Orono, Maine 04469, USA

Arne Henriksen

Norwegian Institute for Water Research<sup>2</sup>, 0808 Oslo, Norway

Peter G. Appleby

Department of Applied Mathematics, University of Liverpool, U.K.

Laura L. Ludwig<sup>1</sup>

Donald V. Vereault<sup>1</sup>

Tor S. Traaen<sup>2</sup>

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## **FORORD**

*Hensikten med denne undersøkelsen har vært å klarlegge nivået og tidsutviklingen av forurensningen av innsjøsedimenter i Sør-Varanger med tungmetaller og radionuklider. Undersøkelsen inngår i Statlig program for forurensningsovervåking som er administrert og finansiert av Statens forurensningstilsyn, SFT. Arbeidet er et samarbeidprosjekt mellom University of Maine, University of Liverpool og NIVA. Prosjektet inngår i kartleggingen av forurensningene i grenseområdene mellom Norge og Russland som samordnes av Arbeidsgruppen for vann og miljøproblemer under den norsk-sovjetiske/russiske miljøvernkommissjonen. Denne rapporten omhandler resultater fra norsk side av grensen. Det er planlagt en tilsvarende undersøkelse på russisk side.*

Tor S. Traaen

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## ABSTRACT

Sediment cores from Dalvatn and Durvatnet in eastern Finnmark, Norway have been dated by gamma ray spectroscopy and analyzed for content of water, oxidizable hydrocarbons, and the trace metals Cd, Co, Cu, Ni, Pb, and Zn. Radionuclides from atmospheric nuclear bomb testing are present in both cores; fallout from the Chernobyl accident was detected only in sediment from Dalvatn. Concentrations of Cd in the sediment are close to the detection limit, ca. 1 ppm. Concentrations of Zn are variable and different for the two lakes but show no systematic changes with time. Cobalt, Cu, Ni, and Pb have elevated concentrations in sediment dating from the last hundred years, for both lakes. Similarly, the accumulation rates of Co, Cu, Ni, and Pb increase relative to the gross sedimentation rate. The onset of increased accumulation rates predates the smelting activity at Nikel and Zapolyarnyy (in Russia, southeast from the coring sites) by several decades, reflecting long range transportation of metals. The maxima in accumulation rates for Co, Cu, and Ni are consistent with the history of regional Ni smelting activities. The accumulation rate for Pb increases over the last 100 years in a pattern consistent with that observed in southern Norway and eastern North America, probably reflecting northern hemispheric pollution.

## SAMMENDRAG

Sedimentkjerner fra Dalvatn og Durvatnet i Sør-Varanger, Finnmark ble datert ved gammastråle-spektroskopi og analysert for vanninnhold, glødetap, og tungmetallene Cd, Co, Ni, Pb og Zn. Radionuklider fra atmosfærisk testing av kjernefysiske bomber var tilstede i begge sediment-kjernene. Nedfall fra Chernobyl-ulykken ble registrert bare i sedimentene fra Dalvatn. Konsentrasjonene av Cd i sedimentene var nær deteksjonsgrensen, ca 1 ppm. Konsentrasjonene av Zn var variable og forskjellig i de to innsjøene, men viste ingen systematisk endring med tiden. Co, Cu, Ni og Pb har forhøyede verdier i sedimenter fra de siste hundre år i begge innsjøene. Sedimentasjonshastigheten for Co, Cu, Ni, og Pb øker tilsvarende, relativt til den totale sedimentasjonshastigheten. En økning i sedimentasjonshastighetene av metaller begynner flere tiår før oppstartingen av smelteverkene i Nikel og Zapolyarnyy, noe som indikerer fjerntransporterte forurensninger med metaller. De største sedimentasjonshastighetene av metaller sammenfaller i tid med aktivitetene ved Ni-smelteverkene i området. Sedimentasjonshastigheten for Pb øker i løpet av de siste 100 år i et mønster som også er observert i Sør-Norge og østre deler av Nord-Amerika. Dette skyldes trolig en generell forurensning av den nordlige halvkule.

## 1. INTRODUCTION

Eastern Finnmark, Norway is currently subjected to deposition of strong acids (primarily  $H_2SO_4$ ) and metals associated with emissions from the Ni smelters in the towns of Nikel and Zapolyarnyy in northwestern Russia. These installations have been active since 1932 and 1955, respectively (Wikan, 1990). As a result of the atmospheric emissions from these facilities, numerous studies have been conducted in eastern Finnmark to evaluate the impacts on vegetation (Ruhling et al., 1987), sediment chemistry (Rognerud and Fjeld, 1990; Rognerud, 1990; Traaen et al., 1990) and water (Traaen et al., 1990; Traaen, 1985; Traaen, 1987; SFT, 1985-1990). Based on these studies, it is clear that deposition of strong acids and metals have clearly impacted water quality and the chemistry of soils, vegetation, and sediments in the region.

This study was designed and conducted to evaluate the history of regional air pollution in eastern Finnmark, Norway using standard paleolimnological techniques. Such studies may provide a continuous record of conditions in the lake extending back in time through the period of interest. This approach has been utilized in many studies in the northern hemisphere to understand the history of emission of various air pollutants (Norton et al., 1990), as well as the impacts of these pollutants on ecosystems, especially lakes (Charles and Norton, 1985).

## 2. METHODS

### 2.1 Lake Selection

Lakes for paleolimnological studies are selected on the basis of having minimal disturbance in the watershed. Therefore, certain changes in the chemical record in the sediments may be attributed to atmospheric deposition. Further, the lakes should be sufficiently deep such that sediment deposition is continuous; this typically requires water depths in excess of 5 to 10 m, depending on lake area.

Dalvatn is located approximately 12 km east of Kirkenes and slightly north of Jarfjord at latitude  $69^{\circ}41'$ , longitude  $30^{\circ}22'$  (map on front page). Dalvatn is a drainage lake, receiving water from several headwater systems. Land use is restricted to the pasturing of sheep. The total watershed area is 220 ha; the lake area is 24 ha. The lake is approximately 1 km long and up to 0.5 km wide. The long dimension is oriented northwest-southeast. The maximum depth is 21 m. The water chemistry is given in Traaen et al. (1990). Approximately 1 km below the outlet, the discharge and chemistry have been measured "continuously" as part of the National Program for Monitoring Long-Range transported Air Pollutants administered by the State Pollution Control Authorities (SFT, 1990). Dalvatn, with typically over  $100 \mu eq SO_4 l^{-1}$ , is clearly under the influence of elevated deposition of non-marine sulfate from the Russian smelters.

Durvatnet is located approximately 25 km southeast of Kirkenes and 25 km northeast of Nikel, at latitude  $69^{\circ}37'$  and longitude  $30^{\circ}41'$  (map on front page). The lake, about 2 km long, is oriented in a northeast-southwest direction with three basins. The most southwesterly and deepest basin, from which the core was retrieved, is approximately 16 m deep. Approximately 12 cabins are located within the drainage basin; several are adjacent to the shore. No roads are present. Land use is restricted to open grazing. The water chemistry sampled on August 8, 1990 was as follows: pH, 6.78; Ca, 1.58 mg  $l^{-1}$ ; Mg, 0.77; Na, 2.67; K,

0.32; Cl, 3.6; NO<sub>3</sub>, <1; SO<sub>4</sub>, 4.7; TOC, 1.83. Durvatnet is also receiving considerable amounts of excess SO<sub>4</sub>.

## 2.2 Field Techniques

Cores were retrieved from the deeper parts of Dalvatn and Durvatnet on August 7 and 9, 1990, respectively. We used a stationary piston corer (Davis and Doyle, 1969) with a 2.5 inch diameter core tube.

The primary core from Dalvatn (Core 1) was taken in 17 m of water and was 55 cm in length. The core was vertically extruded and sectioned at the lake in 0.5 cm sections from 0 to 10 cm, and in 1.0 cm sections thereafter. Sediment was placed in Whirl-pack™ bags and returned to the University of Maine (USA) for further processing.

A second core was also retrieved from Dalvatn. It has been processed for water content and oxidizable hydrocarbon content.

A 1 cm<sup>3</sup> sample of sediment was removed from each of the samples from the primary core from Dalvatn for preliminary diatom analyses.

One core was retrieved from Durvatnet from a water depth of 15.5 m. The core was 50 cm long. The core was processed in the same manner as the Dalvatn core.

## 2.3 Sediment processing

Sediment was homogenized in the Wirl-Pack™ bags, transferred to porcelain crucibles, and air dried at 95°C. Weight loss was calculated as water content. Alternate dried intervals were sent to P. G. Appleby at Liverpool, UK, for gamma analysis for radiometric dating. Gamma ray spectroscopy for <sup>210</sup>Pb, <sup>226</sup>Ra, <sup>137</sup>Cs, <sup>134</sup>Cs, <sup>235</sup>U, <sup>238</sup>U, <sup>40</sup>K, and <sup>241</sup>Am was performed using a well-type, coaxial, low background, intrinsic germanium detector fitted with a NaI(Tl) escape suppression shield (Appleby et al., 1986).

The remaining sediment samples were heated in porcelain crucibles at 550°C ("ignited") for 3 hours. Weight loss was calculated as oxidizable hydrocarbons, or organic content.

Approximately 0.1000 g of "ignited" sediment was placed in solution using the methods of Buckley and Cranston (1971). This process places the entire sample into solution. Thus, "bulk" chemistry is determined.

Chemical analyses for various trace elements were performed by Atomic Absorption Spectrophotometry (Perkin-Elmer) with a graphite furnace (2200) or flame, whichever was appropriate for the element and concentration range. Analyses for Cd, Co, Cu, Ni, Pb, and Zn were performed on alternate intervals of sediment not analyzed for <sup>210</sup>Pb. Detection limits were: Cd, 0.06 ppm on an ignited weight basis; Co, 1.0 ppm; Cu, 0.7 ppm; Ni, 0.7 ppm; Pb, 1 ppm; Zn, 11 ppm.



### 3. RESULTS

#### 3.1 Radiometric Dating

For Dalvatn, the  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{241}\text{Am}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{40}\text{K}$ , and  $^{241}\text{Am}$  results are shown in Table 1. The  $^{134}\text{Cs}$  derives from Chernobyl fallout, and has been corrected for decay since May 5, 1986. The  $^{134}\text{Cs}$  data can be used to partition the  $^{137}\text{Cs}$  into its two components, derived respectively from weapons testing and Chernobyl fallout. Figure 1 displays the weapons fallout  $^{137}\text{Cs}$  and  $^{241}\text{Am}$ .

An unusual feature of the Dalvatn core is the layer of sediment between 3.5 and 5.0 cm which is characterized by enhanced  $^{226}\text{Ra}$  activities an order of magnitude higher than in the adjacent sediment intervals. These sediments have greatly reduced  $^{210}\text{Pb}$  activities. In particular, the layer 4.5 to 5.0 cm is devoid of unsupported  $^{210}\text{Pb}$ .  $^{222}\text{Rn}$  diffusion from the  $^{226}\text{Ra}$ -rich layer will, over time, result in supported  $^{210}\text{Pb}$  activities that are not in equilibrium with the  $^{226}\text{Ra}$ . Corrections have been made for this process in determining the unsupported  $^{210}\text{Pb}$  activities. Even with these corrections, the measured  $^{210}\text{Pb}$  activity for 4.5 to 5.0 cm was below the estimated supported activity. In determining the  $^{210}\text{Pb}$  chronology, this layer was assumed to have zero unsupported  $^{210}\text{Pb}$ . The estimated  $^{210}\text{Pb}$  flux to the coring site is about  $0.26 \text{ pCi cm}^{-2} \text{ yr}^{-1}$  and the integrated unsupported  $^{210}\text{Pb}$  is  $8.29 \text{ pCi cm}^{-2}$ . The 5.5 to 6.0 cm interval also has unusual activities of several nuclides.

$^{210}\text{Pb}$  chronologies have been calculated using both the CRS and CIC dating models (Appleby and Oldfield, 1978), and the results are shown on Figure 2 and in Table 2. Except for sediments within the anomalous intervals, the two models are in good agreement. Both indicate a fairly constant sedimentation rate of ca.  $0.0019 \text{ g cm}^{-2} \text{ yr}^{-1}$  for the past 60 to 70 years and a slightly lower rate of ca.  $0.0013 \text{ g cm}^{-2} \text{ yr}^{-1}$  in the latter half of the 19th century. The anomalous intervals are interpreted to represent an "instantaneous" in-wash event dating to approximately 1916-1917. A summary of these dates is given in Table 2.

The surficial sediments contain significant amounts of Chernobyl fallout. Allowing for this, the weapons fallout component of the  $^{137}\text{Cs}$  activity (Figure 1) still has its highest value in the topmost measured interval (0.5 to 1.0 cm). The highest values for  $^{241}\text{Am}$  also occur in this interval. The  $^{210}\text{Pb}$  chronology places 1963 (the weapons fallout maximum) at 1.0 to 1.5 cm. Because this sample was not analyzed, a precise fallout date cannot be determined. However, the next sample analyzed (1.5 to 2.0) is almost certainly-pre 1963, and the fallout nuclides appear to confirm the very low value of present day accumulation rates.

Table 3 displays radionuclide data for the core from Durvatnet. Dates were calculated using the CRS and CIC  $^{210}\text{Pb}$  dating models (Figure 3). There is no significant difference between the two sets of dates. Both models indicate a constant accumulation rate over the last 100 years of  $0.0024 \text{ g cm}^{-2} \text{ yr}^{-1}$ . Dates calculated with this value are given in Table 4.

$^{137}\text{Cs}$  and  $^{241}\text{Am}$  are also shown on Figure 4. The short-lived nuclide  $^{134}\text{Cs}$ , indicative of Chernobyl fallout, was not detected. Unlike the Dalvatn core, the  $^{137}\text{Cs}$  in Durvatnet cannot be partitioned into weapons testing and Chernobyl fallout. The  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  are of limited chronological value in this core. The  $^{137}\text{Cs}$  lacks a subsurface maximum; the maximum activity occurs in 0.0 to 0.5 cm interval. Further, the activity is nearly constant in the top 2.5

cm, presumably because of continuous in-wash of  $^{137}\text{Cs}$  from the watershed. It is possible that the surface sediments contain traces of Chernobyl fallout. At the time of measurement, only 11% of the short-lived  $^{134}\text{Cs}$  burden (if present) would remain and that may be undetectable. Although the  $^{241}\text{Am}$  profile does have a subsurface peak (Figure 4), the large standard errors in the  $^{241}\text{Am}$  values combined with the very slow accumulation rate make it difficult to assign a precise level to the 1963 fallout maximum. This feature can, however, be placed with confidence between 2.0 and 3.5 cm, date by  $^{210}\text{Pb}$  to the period 1957-1974. The  $^{210}\text{Pb}$  places 1963 at a depth of 3.0 cm. Thus the  $^{241}\text{Am}$  does provide some corroboration of the  $^{210}\text{Pb}$  dates.

### 3.2 Chemistry

Water content and organic content for the sediment cores from Dalvatn and Durvatnet are given in Table 4.

Water content in Dalvatn decreases from the surface downward to a minimum of 90% within the "in-wash" interval identified by radionuclide measurements. It then increases to pre-in-wash values and then varies unsystematically between 91 and 93% to the bottom of the core. Organic content decreases downward from 44% at the surface to a minimum of 27% at the in-wash interval. Then, organic content increases to values near 34% at the bottom of the core.

Water content in Durvatn sediment decreases monotonically from 99% at the surface to 89% at the bottom of the core with no suggestion of any change in the nature of the sediment. Organic content declines monotonically down core from 37% to 18%. These data suggest a very slow but consistent change in the nature of sedimentation, spanning hundreds of years.

Trace element concentrations (Cd, Co, Cu, Ni, Pb, and Zn) for the two cores are given in Figures 5 to 14.

For Dalvatn, Cd (not shown on any figures) was detectable only in sediment below 15 cm, ranging between 0 and 1 ppm. The anomalously high concentration at 32-33 cm is likely a laboratory error. Concentrations of Co range between 30 and 85 ppm up to 8.5 cm. Above 8.5 cm, they are elevated significantly with the exception of the 6.0 to 6.5 cm interval. The pronounced spike between 4.0 and 5.5 is clearly present. However, it was necessary to dilute these samples because of the high concentrations. Consequently, precision is poorer for these analyses. The concentration of Cu ranges between 130 and 215 ppm below 2.5 cm, with the 5.0 to 5.5 cm being outside this range and considerably lower than the long term average. This minimum corresponds with the minimum in organic content. From at least 2.5 cm to the surface, Cu increases to more than twice background. Nickel ranges from 33 to 83 ppm up to the 4.0 to 4.5 cm interval. A possible peak in concentration occurs in the in-wash layer; the upper three measured intervals have substantially increasing upward concentrations. Lead ranges between 8 and 18 ppm up to 5.0 cm, and thereafter increases to values approximately 17 times background. Zinc is highly variable in the bottom 20 cm of the core; it is less variable in the upper 20 cm, averaging about 200 ppm, except for the in-wash layer which is about 100 ppm.

For Durvatnet, the sample from the uppermost measured interval (0.5 to 1.0 cm) had too little mass for accurate analyses. Cadmium ranges from below the detection limit to slightly over 1 ppm, with no apparent trends. Cobalt is highly variable in concentration, with a broad peak centering on 20 cm. The upper 6 cm average about twice the Co concentration of older

sediment but with no trend in concentration. Copper increases upward from 36 ppm at the base of the core to 66 ppm at 6.5 cm. Upward, it increases dramatically to concentrations at least 10 times those at the base of the core. Nickel ranges from 0 to 50 ppm from the base of the core to 7.0 cm. It increases upward from there to nearly 400 ppm in the 1.5 to 2.0 interval. Lead values are quite low (by crustal abundance standards), less than 5 ppm at the base of the core, and increase irregularly to about 10 ppm at the 6.5 cm level. Upward from there the increase steepens with a maximum concentration of 46 ppm reached by 1.5 cm. The concentration of Zn ranges from 109 ppm to 250 ppm with no significant trends. The 5.5-6.0 cm interval has very high concentration, perhaps caused by an analytical error.

In summary, Cd and Zn display no systematic trends in concentration through either core. Cobalt shows enrichment in the upper part of both cores and also has a maximum deeper in Durvatnet's record. Copper and Pb have near surface enrichment which is nearly coincident in each core. Enrichment in Pb in both cores pre-dates (is deeper) than any of the other metals.

### 3.3 Trace element fluxes

Figures 15 to 24 depict the net flux of the trace metals to the coring site in the two lakes. The dates shown are the ages of the mid-points of sediment intervals. The accumulation rates are generated with knowledge of the concentration of each element in each interval (ppm), the sediment accumulation in each interval ( $\text{g cm}^{-2}$ ), the percentage of inorganic material in each interval (%), and the number of years represented by each interval (yr).

$$\text{Accumulation rate} = (\mu\text{g g}^{-1})(\text{g cm}^{-2})(\text{g g}^{-1})(\text{yr}^{-1})$$

Accumulation rates are for total metal content. For all metals in the sediment, there are two sources: the watershed and the atmosphere. Thus, the flux of Ni may be proportioned into a geologic component (derived from the bedrock) and an atmospheric component (derived from emissions). The accumulation rates for the sediment in Dalvatn is constant at  $1900 \mu\text{g cm}^{-2} \text{yr}^{-1}$  since the in-wash event, and  $1300 \mu\text{g cm}^{-2} \text{yr}^{-1}$  prior to that. Durvatnet has a constant rate of sedimentation ( $2400 \mu\text{g cm}^{-2} \text{yr}^{-1}$ ) throughout the period represented by the core. Thus, for both lakes, changes in the accumulation rates of trace metals in the last 50-60 years may be attributed largely to changes in the atmospheric flux term.

The  $^{210}\text{Pb}$  chronology may be used with confidence only back to about 1850 A.D. Ages of sediment, depicted on the figures, have been extrapolated farther back in time based on the accumulation rate in the 1850-1900 period and thus are qualitative.

For Dalvatn, the accumulation rate for Cd is less than  $0.0001 \mu\text{g cm}^{-2} \text{yr}^{-1}$  throughout the core, except for the apparently erroneous interval dated ca. 1386. Background accumulation rates for Co are about  $0.12$  to  $0.18 \mu\text{g cm}^{-2} \text{yr}^{-1}$  prior to 1800 but are approximately double this value since 1800 except during the in-wash event. Then (1916), the cumulative increase is approximately 100X background. The pattern for Cu is similar to that of Co with relatively constant values prior to 1800 at  $0.1$  to  $0.2 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . The increase in the 1916 event corresponds to about a 25X increase; Co is enriched relative to Cu by at least 4X. Cu increases from  $0.2$  to nearly  $0.5 \mu\text{g cm}^{-2} \text{yr}^{-1}$  from the end of the in-wash event to the 1986 sediment interval. The accumulation rate for Ni follows that of Cu, with a 50X increase of Ni during the in-wash event, followed by an increase from  $0.1$  to  $0.3 \mu\text{g cm}^{-2} \text{yr}^{-1}$  since then. Modern Ni values are approximately 8X background. The accumulation rate for Pb increases approximately 100X in the in-wash intervals and then increases steadily from the end of that

event until the present. Modern Pb accumulation rates are about 15X background. Zinc shows no long term trend in accumulation rates throughout the core except for the transitory in-wash event.

The in-wash event is characterized by an increase in sediment accumulation rate (dry) of about 25X over modern values and 40X over background values. This increase yields an increase in the accumulation rates for all elements for the sediment intervals between 4.0 and 7.5 cm. Cobalt and Ni concentrations increase in this interval; those of Cu, Pb, and Zn generally decline. Thus accumulation rates for Co and Ni increase more than the other metals. More recent increases in accumulation rates occur for Cu, Co (possibly), Ni, and Pb. The precise timing of the increases can not be established because of the overprint of the "instantaneous" in-wash event.

In Durvatnet, the accumulation rates for Cd are typically less than  $0.001 \mu\text{g cm}^{-2} \text{yr}^{-1}$ , suggesting that little or no atmospheric input is present (Norton et al, 1990). Since approximately 1900, the accumulation rate of Co has been elevated over values typical of the previous 200 years. However, values in the 17th century were also relatively high and thus the cause of the modern elevated values for Co can not be assigned with confidence to atmospheric emissions, although the timing is coincident. Copper shows a clear and strong increase in accumulation rate starting in sediment dated as early as 1910 and increasing at least to 1978. The increasing accumulation rate for Pb starts in the late 19th century, well before industrial activity in this region. A nearly ten-fold increase occurs in 100 years. The accumulation rate for Ni, the principal ore metal emitted from smelters in the region, increases by the end of the 19th century to a maximum at least as recent as 1978. The accumulation rate for Zn declines throughout the period represented by the entire core, with the exception of one interval. The latter anomaly is probably an analytical artifact.

#### 4. DISCUSSION

Of the six trace metals investigated, Zn and Cd show no pattern of increasing concentration or accumulation rates in young sediment that would suggest recent increased atmospheric deposition of these metals. This is consistent, for Zn, with the observation by Rognerud (1990) that Zn was not enriched in modern riverine sediments downwind of Nickel, compared with upwind. It is inconsistent with the slight enrichment he noted for Cd. Ruhling et al. (1987) found little enrichment of both these metals in successive collections and analyses of living mosses, compared with regional deposition in Fenno-Scandinavia.

Cobalt, Cu, Ni, and Pb are all enriched in concentration and have elevated accumulation rates in sediments dating from the last 100 years, in both lakes. Lead is not associated with the emissions from the Ni smelters. Further, the timing of the increases for Pb are too early to be associated with any of the near-by industrial activities. In North America, accumulation rates in lake sediments increase consistently from about 1850-1875 up to about 1975, and then decrease to the present (Norton et al, 1990). In Europe the early pattern is similar but with initial increases starting as much as 50-75 years earlier. Lead from anthropogenic sources has many origins including smelting, glass works, and the use of tetra-ethyl Pb as a gasoline additive. It is the cessation of the latter use that has caused the downturn in deposition in North America. Almost certainly, the increases in Pb may be attributed to very long range transport of Pb from North American sources and more southerly sources in Europe. The absolute values for the accumulation rates of Pb are about an order of magnitude less than those observed in southern Norway (Norton and Hess, 1980) or in eastern North America

(Norton et al., 1990). The more subtle increases in eastern Finnmark are detectable because of the relatively low sediment accumulation rate in both lakes and the very low background concentrations of Pb in the sediment. Similar increases have been observed in the Abisko, Sweden area (Norton, unpublished).

Co, Cu, and Ni are all emitted by the smelting activities at Nikel and elsewhere. This is clear from surveys of sediments (Rognerud, 1990), vegetation (Ruhling et al., 1987), and lake water chemistry (Traaen et al., 1990). In all cases, the increases in concentrations and accumulation rates pre-date the initiation of the smelting at Nikel and Zapolyarnyy by decades. Increases typically are detectable for sediment dating to the 1920's or 1930's. Two interpretations are possible. First, regional pollution (Co, Cu, and Ni) may have existed as a result of earlier activities known to be in existence further east and south in Russia, or even to the southwest in Sweden. More widespread paleolimnological studies could address this possibility. The strength of the recent signal in the sediments plus the regional surveys of vegetation and lake water chemistry leave no doubt that the Russian sources are now dominant. Second, trace metal pollutants in the lake water column may be deposited and redistributed into sediments which pre-date the atmospheric deposition of the metals themselves. Such diagenetic processes have been demonstrated for Zn (Carignan and Tessier, 1985) and other trace metals. Our data do not enable us to evaluate this possible artifact.

The apparent synchronicity of the pollution signal in sediments of two widely separated lakes suggests strongly that atmospheric deposition, rather than watershed-specific processes are responsible. This is not true for the "in-wash" event recorded in Dalvatn sediments. The sediment involved in the event is clearly old, probably greater than 1850; it contains no unsupported  $^{210}\text{Pb}$ . The strong enrichment in  $^{226}\text{Ra}$ , along with several other radiogenic and chemical anomalies, is most peculiar. This event may be anthropogenically linked.

## ACKNOWLEDGMENTS

We are very grateful to Einar Fiskebeck for courtesies extended to us while on site. The Sawyer Environmental Research Laboratory at the University of Maine provided chemical analyses of sediment at no charge. Richard F. Wright, Leif Lien, and Anne Norton assisted with the field work.

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## APPENDIX

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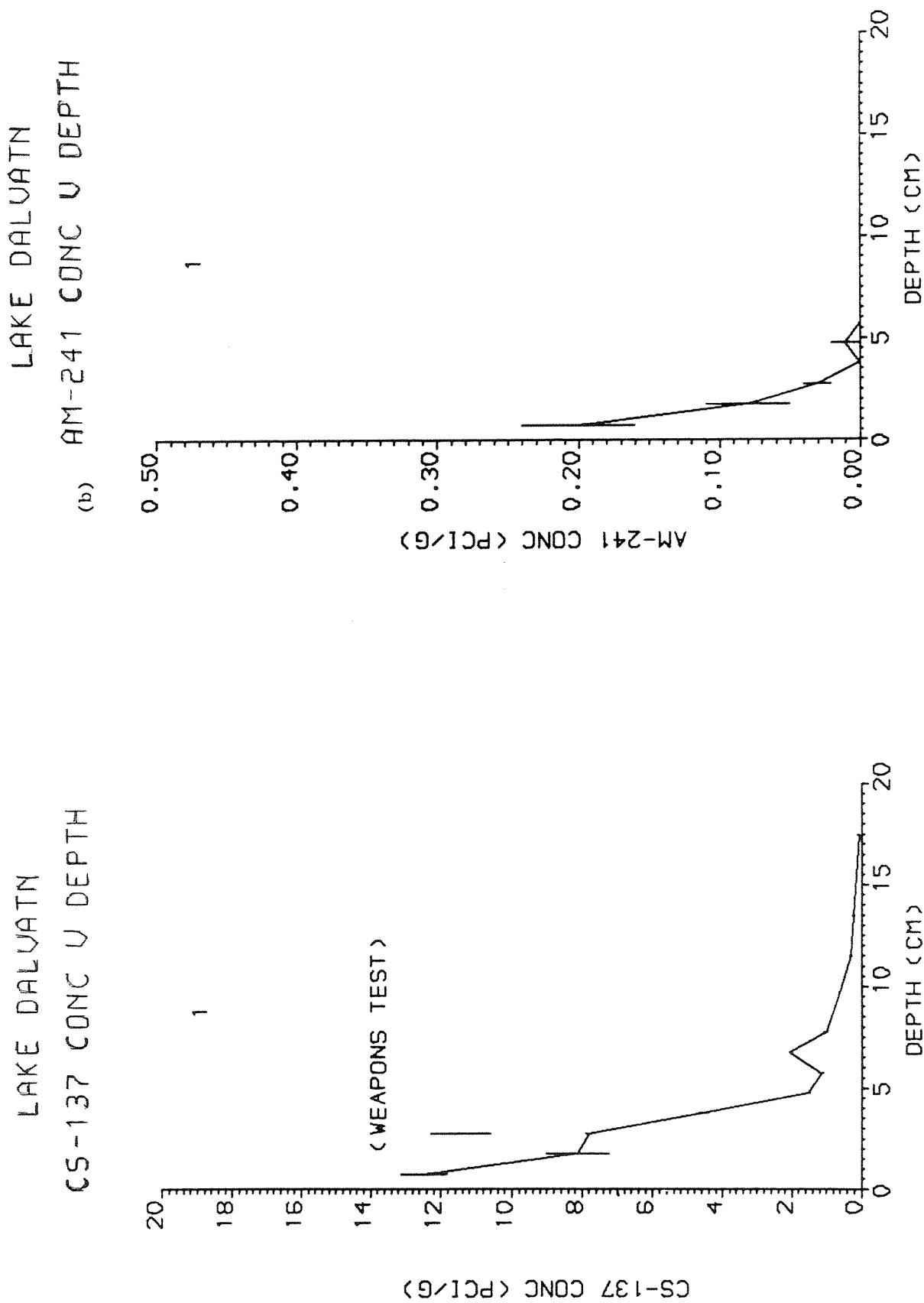


Figure 1. Weapons testing fallout ( $^{137}\text{Cs}$  and  $^{241}\text{Am}$ ) in a sediment core from Dalvatn, eastern Finnmark, Norway.

LAKE DALVATN  
DEPTH V AGE

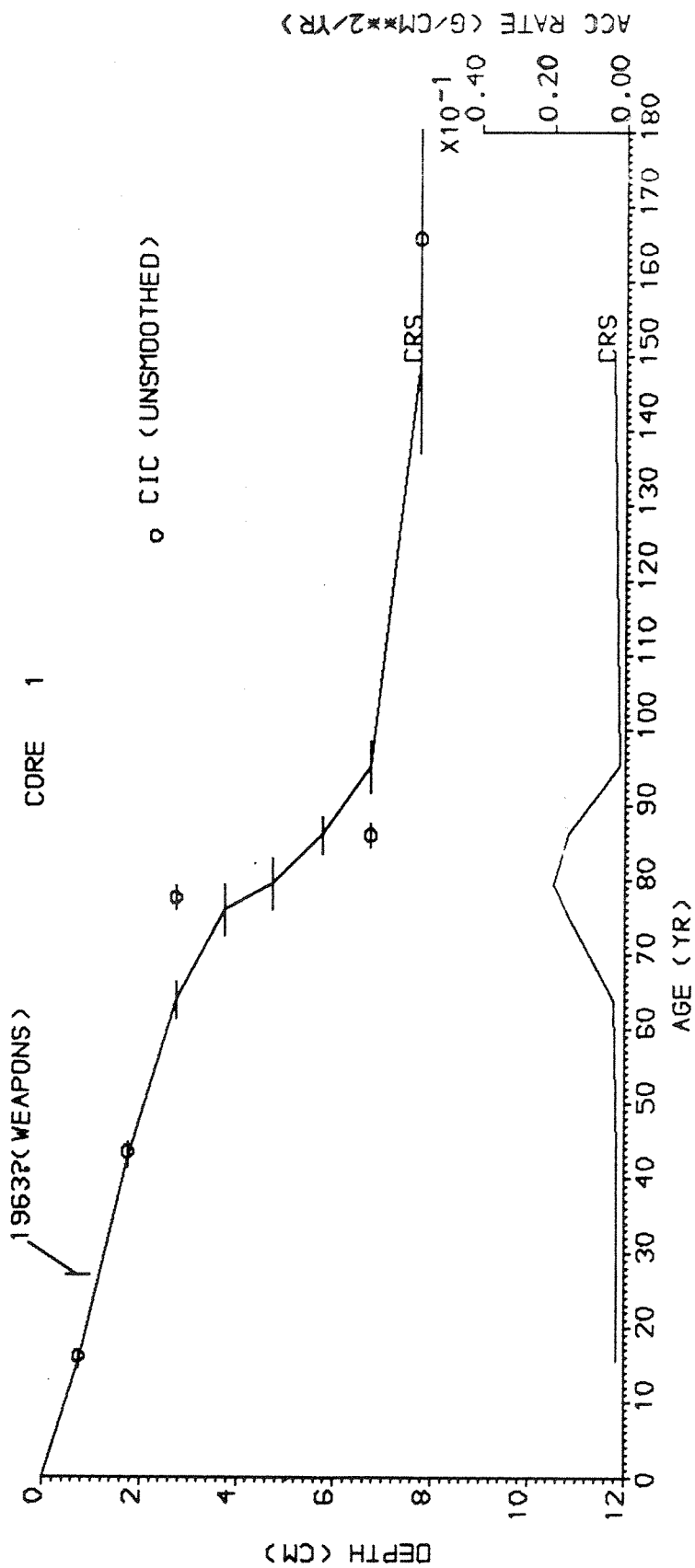


Figure 2. <sup>210</sup>Pb chronologies (CRS and CIC; Appleby and Oldfield, 1978) for a sediment core from Dalvatn, eastern Finnmark, Norway.

LAKE J  
DEPTH v AGE

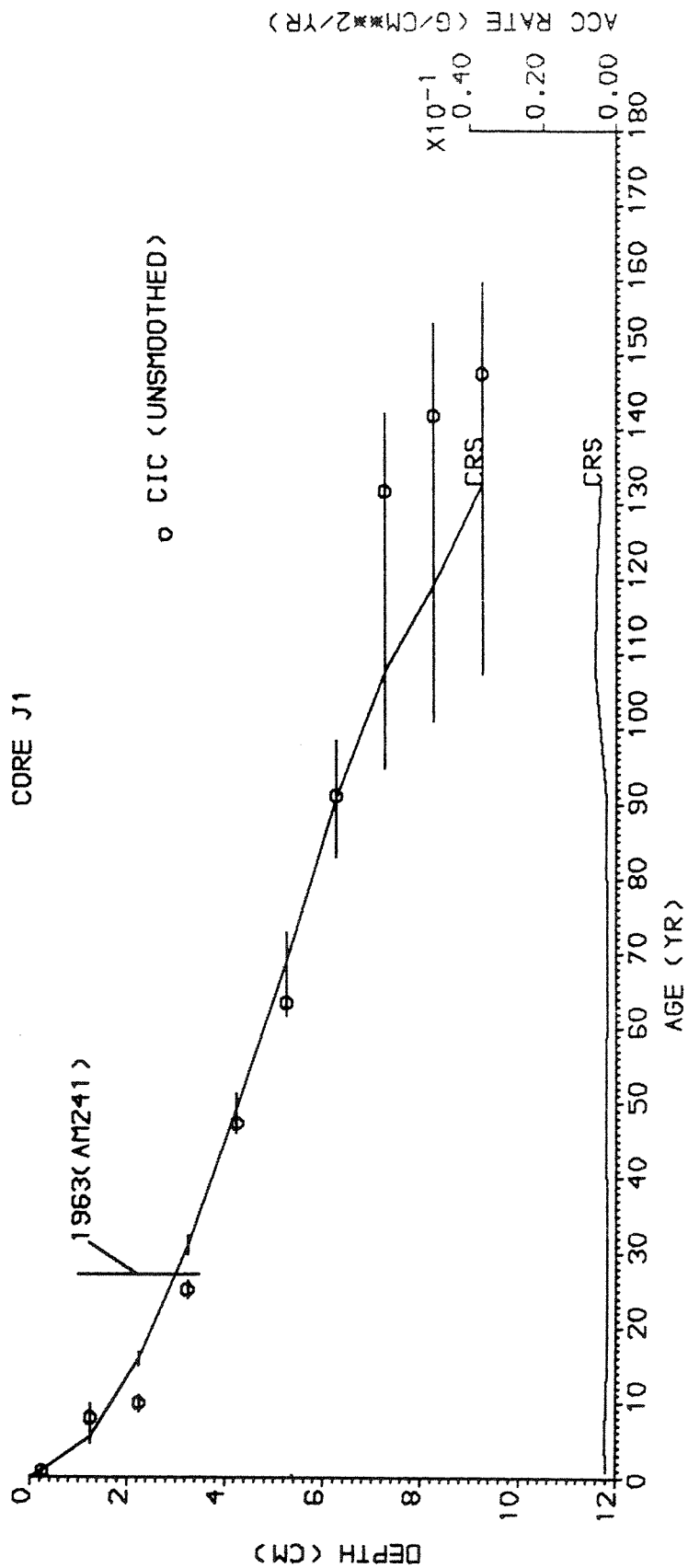


Figure 3. <sup>210</sup>Pb chronologies (CRS and CIC; Appeby and Oldfield, 1978) for a sediment core from Durvatnet, eastern Finnmark, Norway.

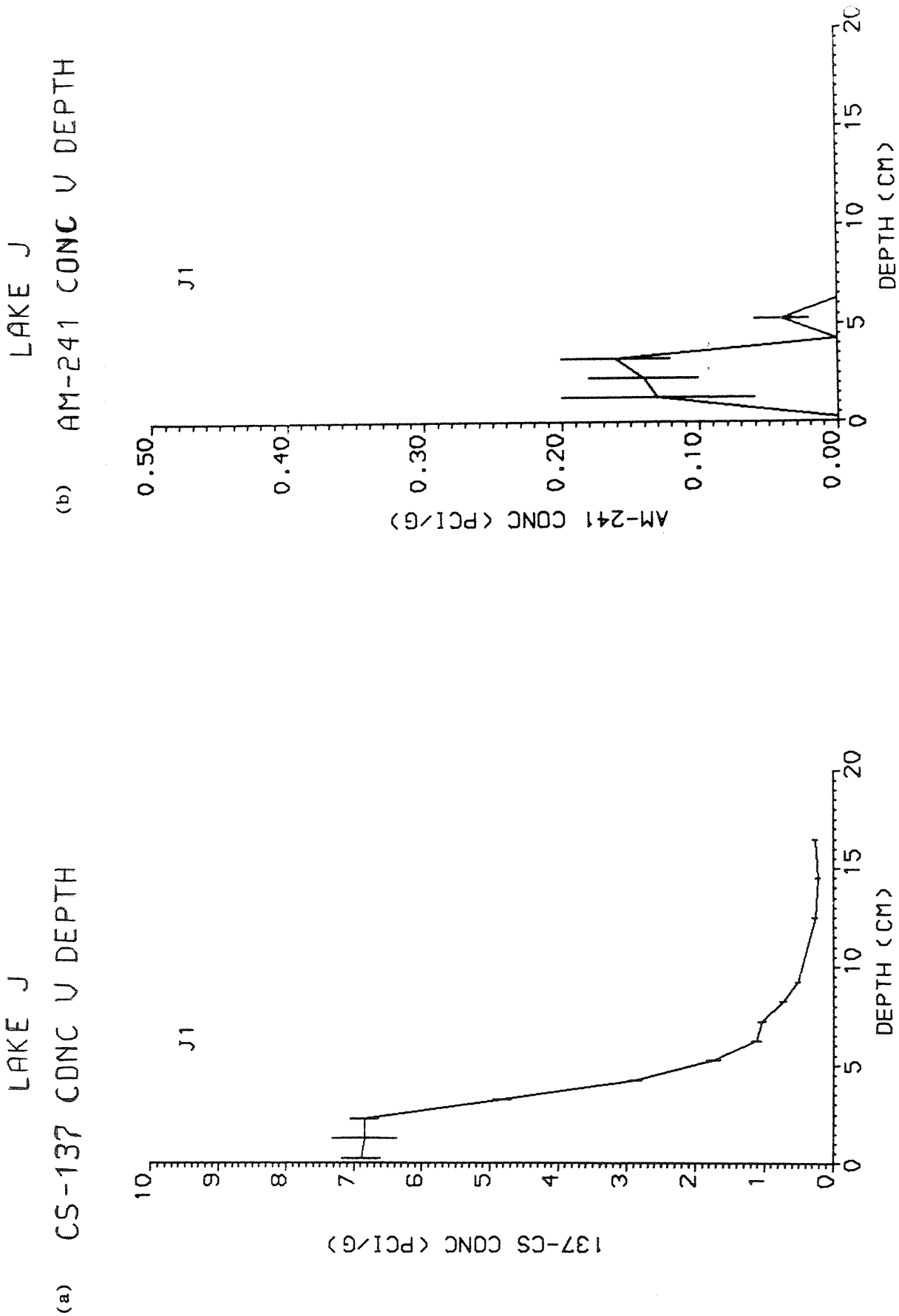


Figure 4. <sup>137</sup>Cs and <sup>241</sup>Am in a sediment core from Durvatnet, eastern Finnmark, Norway.

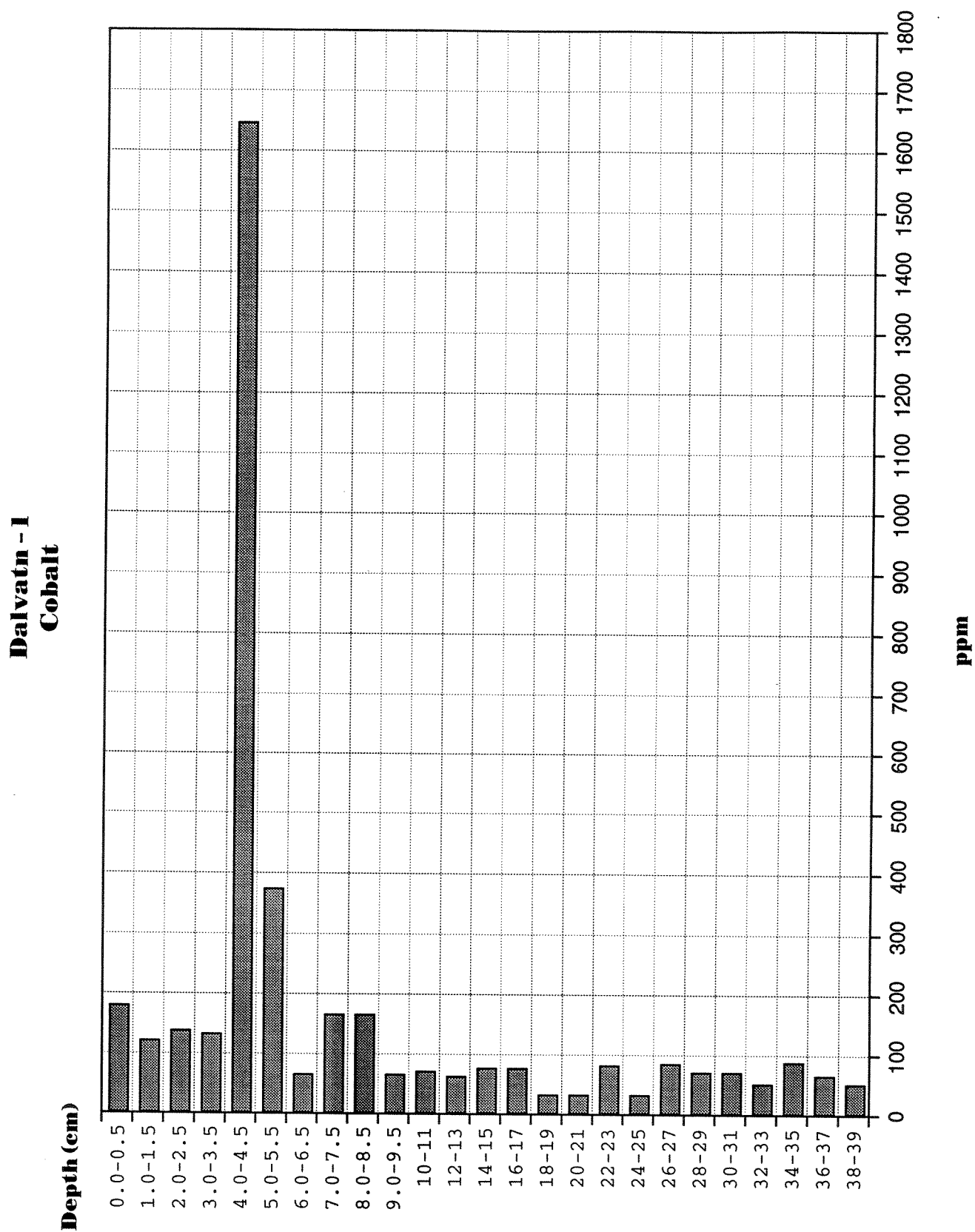


Figure 5. Concentrations (parts per million of ashed sediment, ppm) of Co in a core from Dalvatn, eastern Finnmark, Norway.

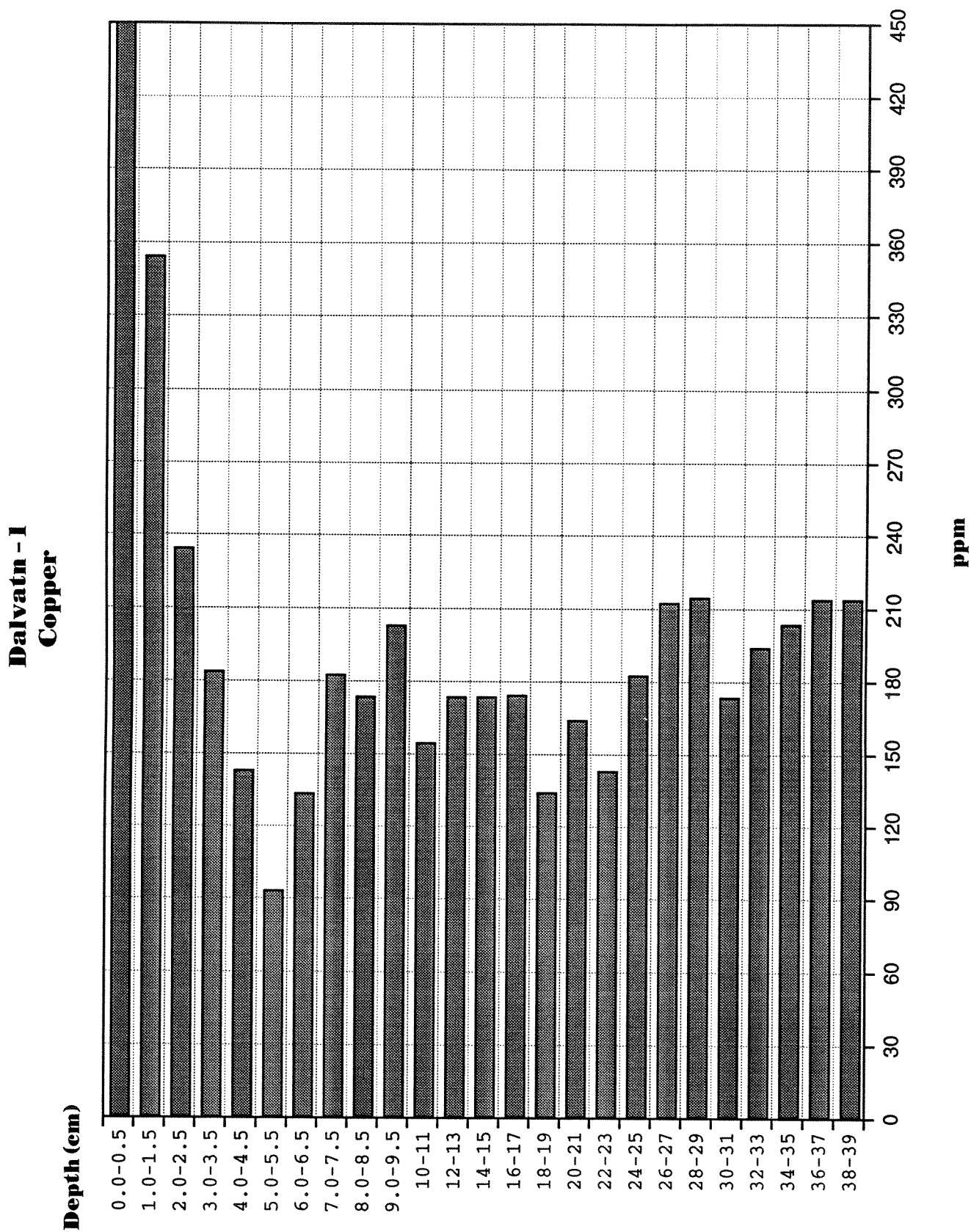


Figure 6. Concentrations (parts per million of ashed sediment, ppm) of Cu in a core from Dalvatn, eastern Finnmark, Norway.

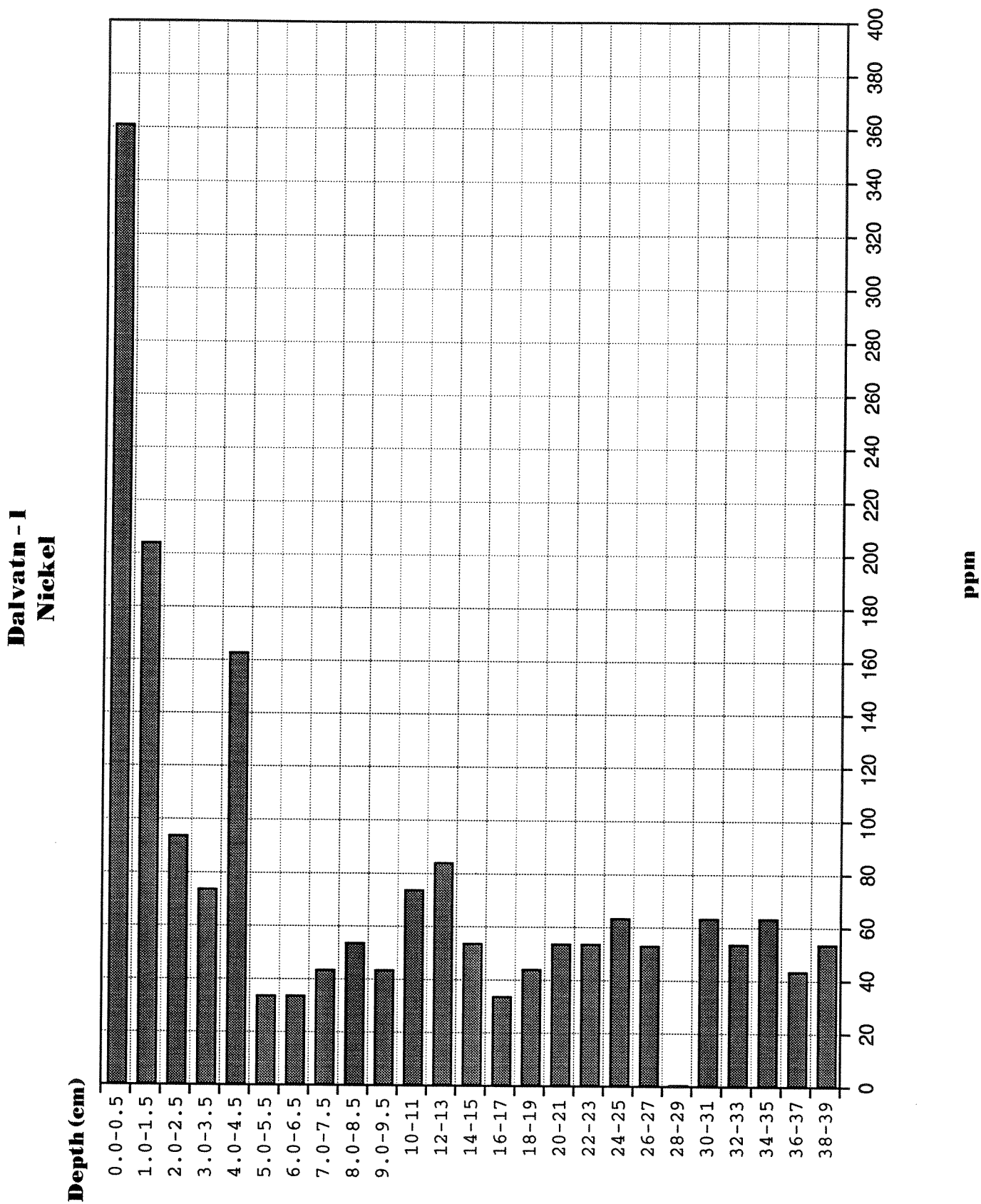


Figure 7. Concentrations (parts per million of ashed sediment, ppm) of Ni in a core from Dalvatn, eastern Finnmark, Norway.

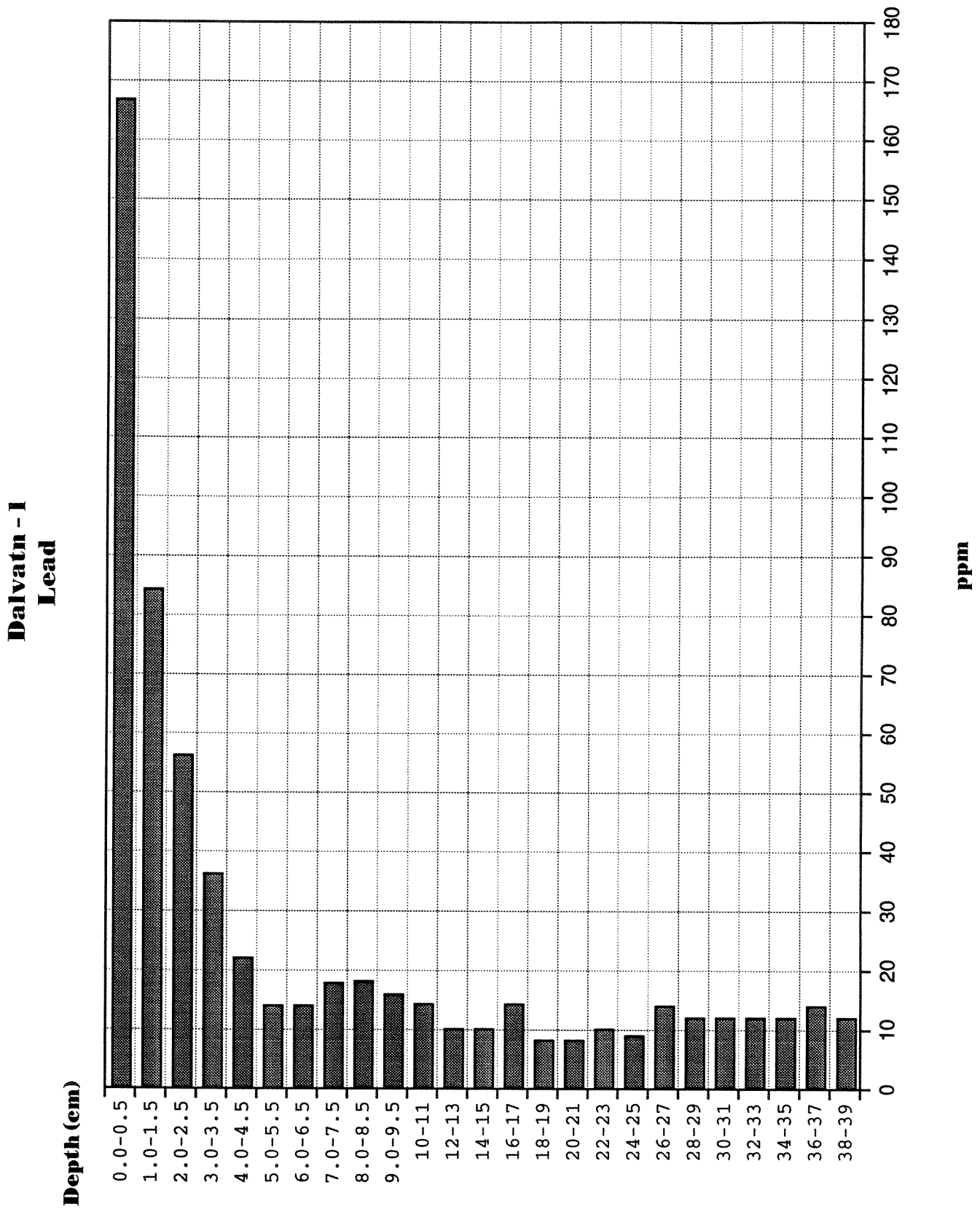


Figure 8. Concentrations (parts per million of ashed sediment, ppm) of Pb in a core from Dalvatn, eastern Finnmark, Norway.



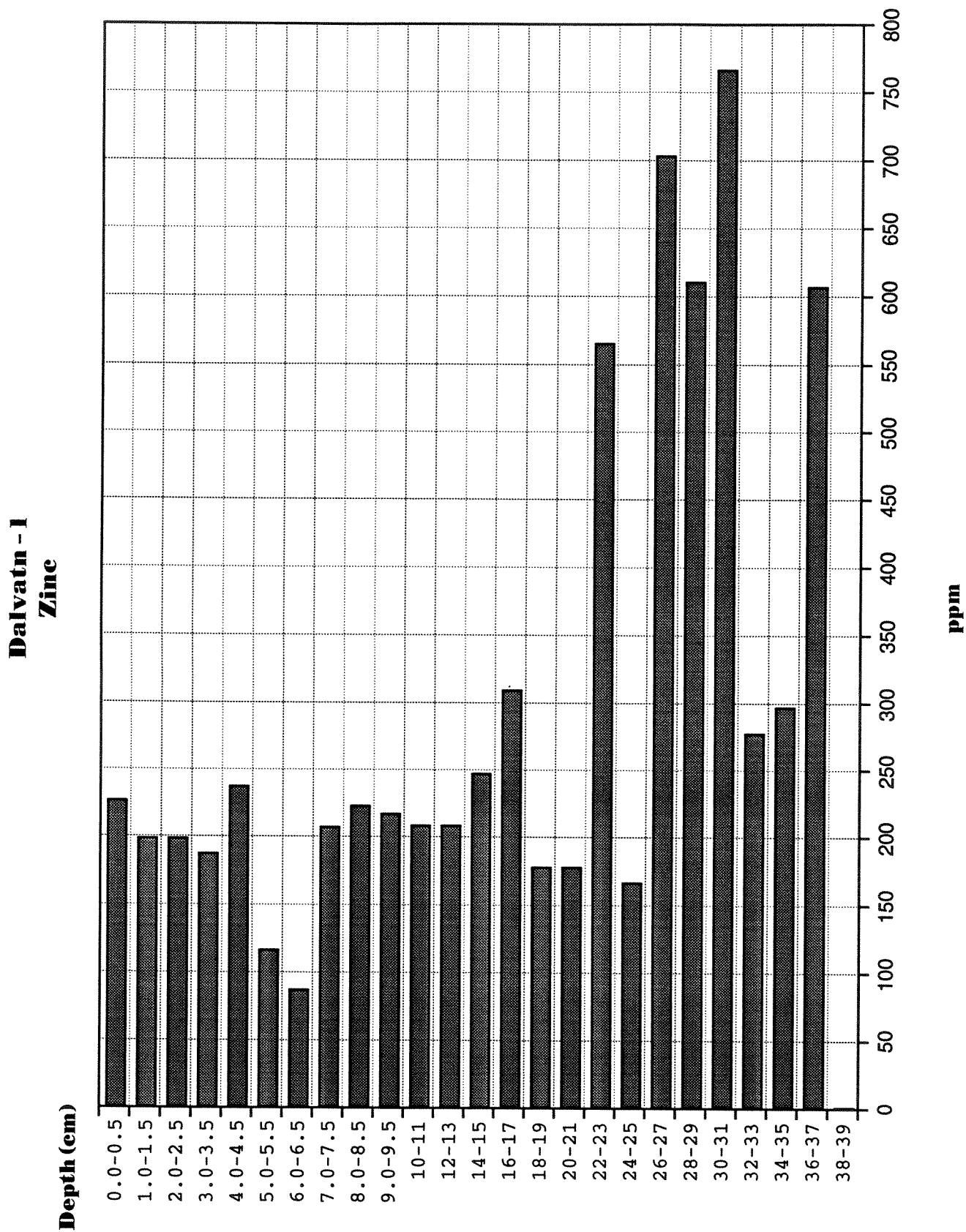
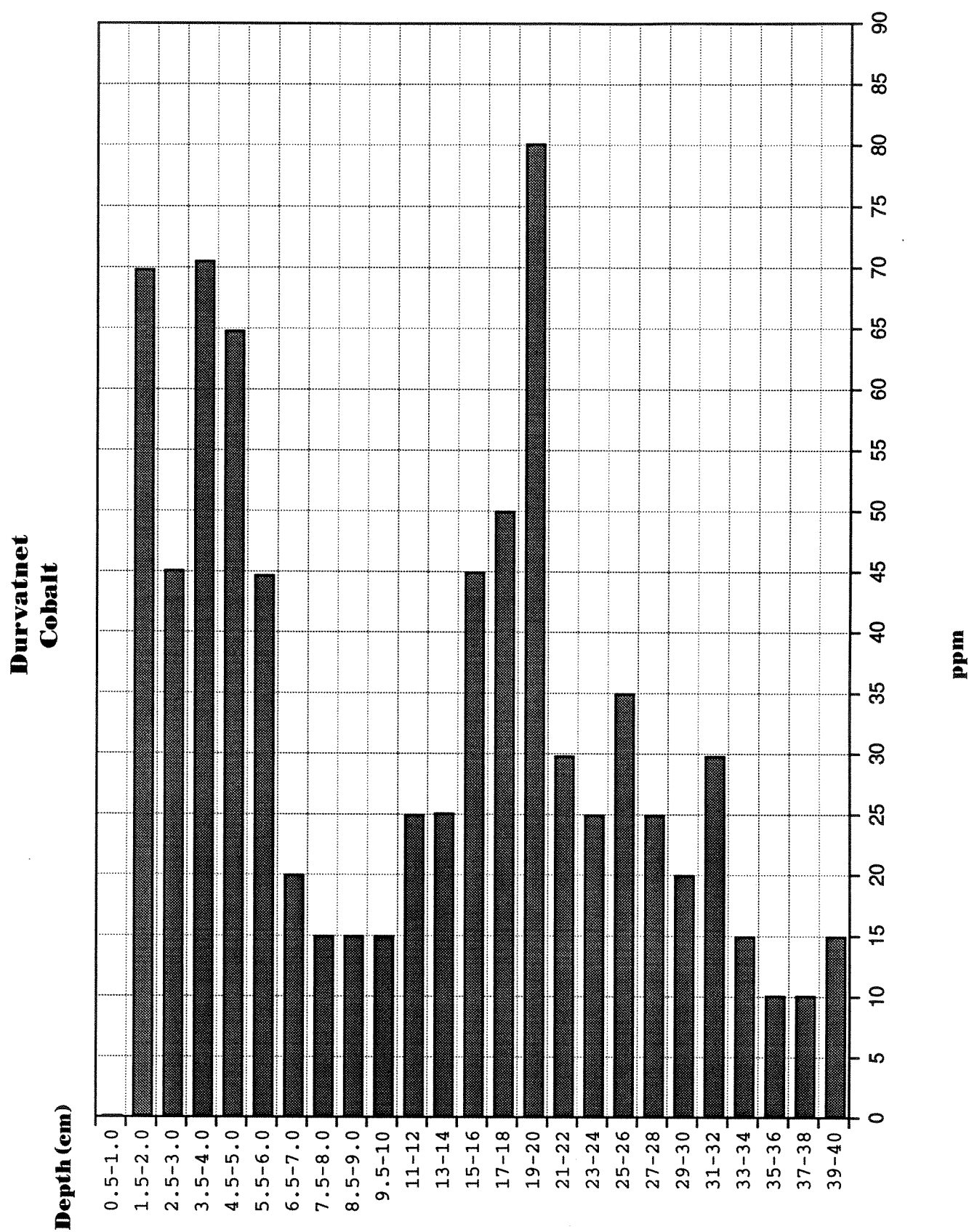


Figure 9. Concentrations (parts per million of ashed sediment, ppm) of Zn in a core from Dalvatn, eastern Finnmark, Norway.



**Figure 10. Concentrations (parts per million of ashed sediment, ppm) of Co in a core from Durvatnet, eastern Finnmark, Norway.**

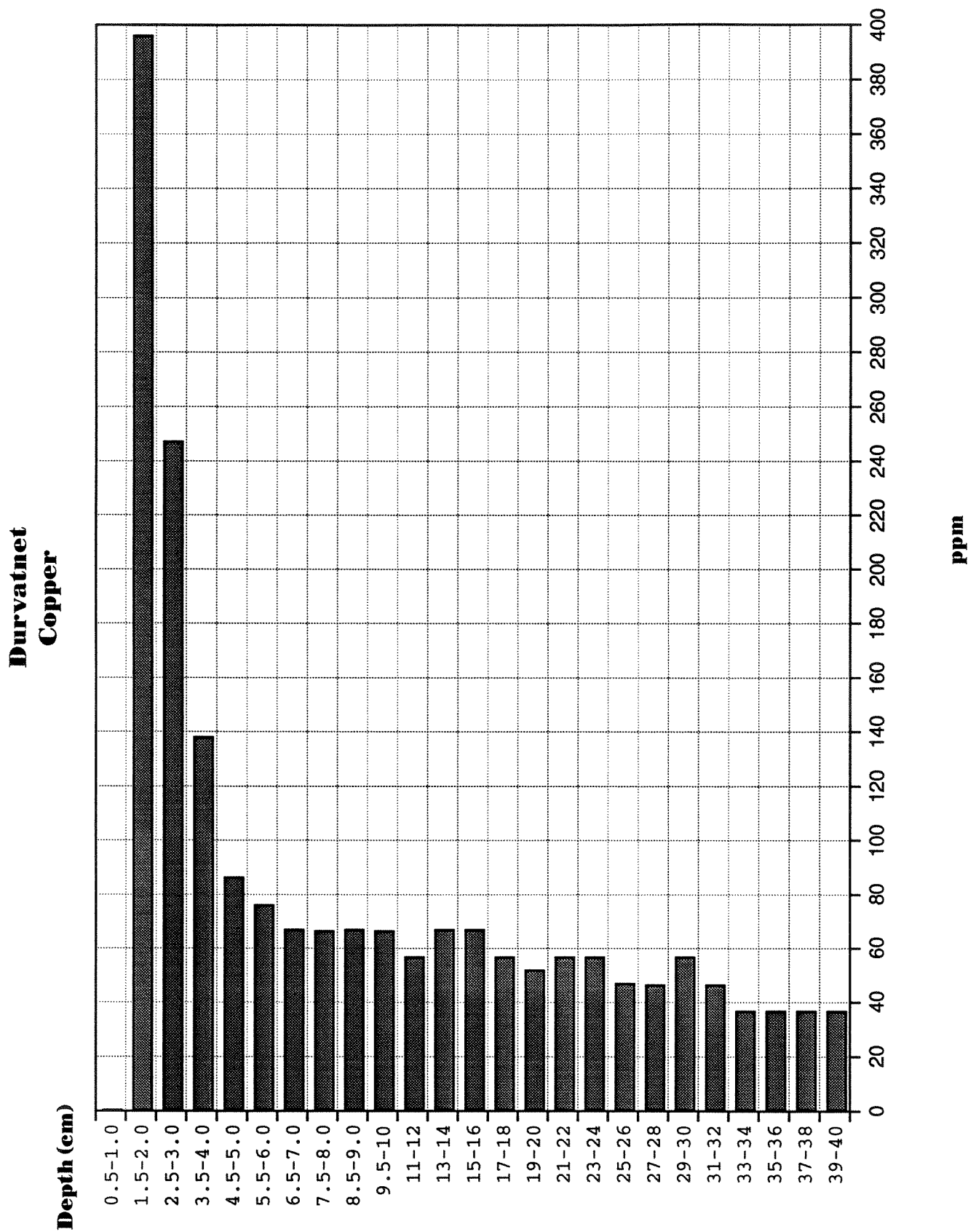
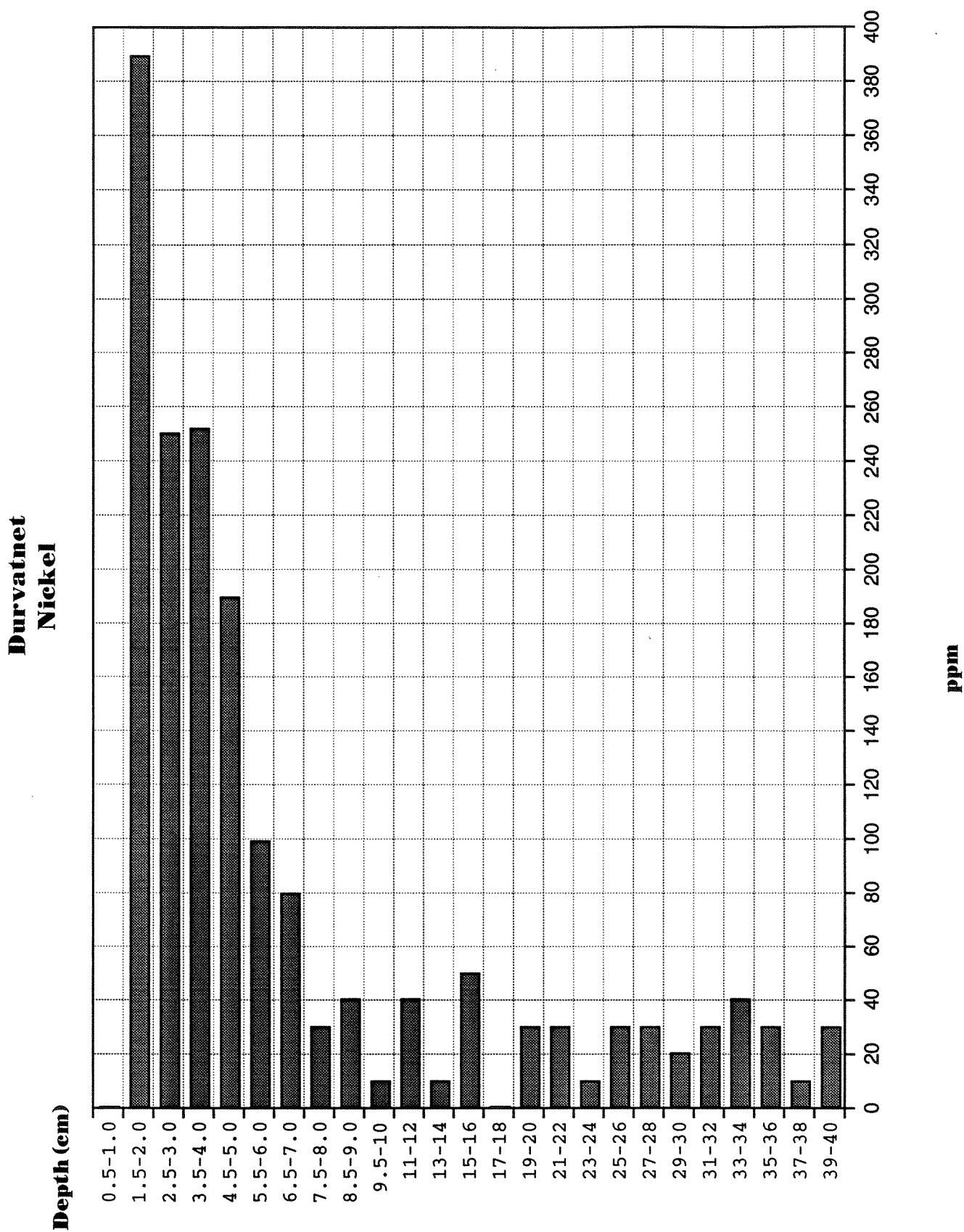


Figure 11. Concentrations (parts per million of ashed sediment, ppm) of Cu in a core from Durvatnet, eastern Finnmark, Norway.



**Figure 12. Concentrations (parts per million of ashed sediment, ppm) of Ni in a core from Durvatnet, eastern Finnmark, Norway.**

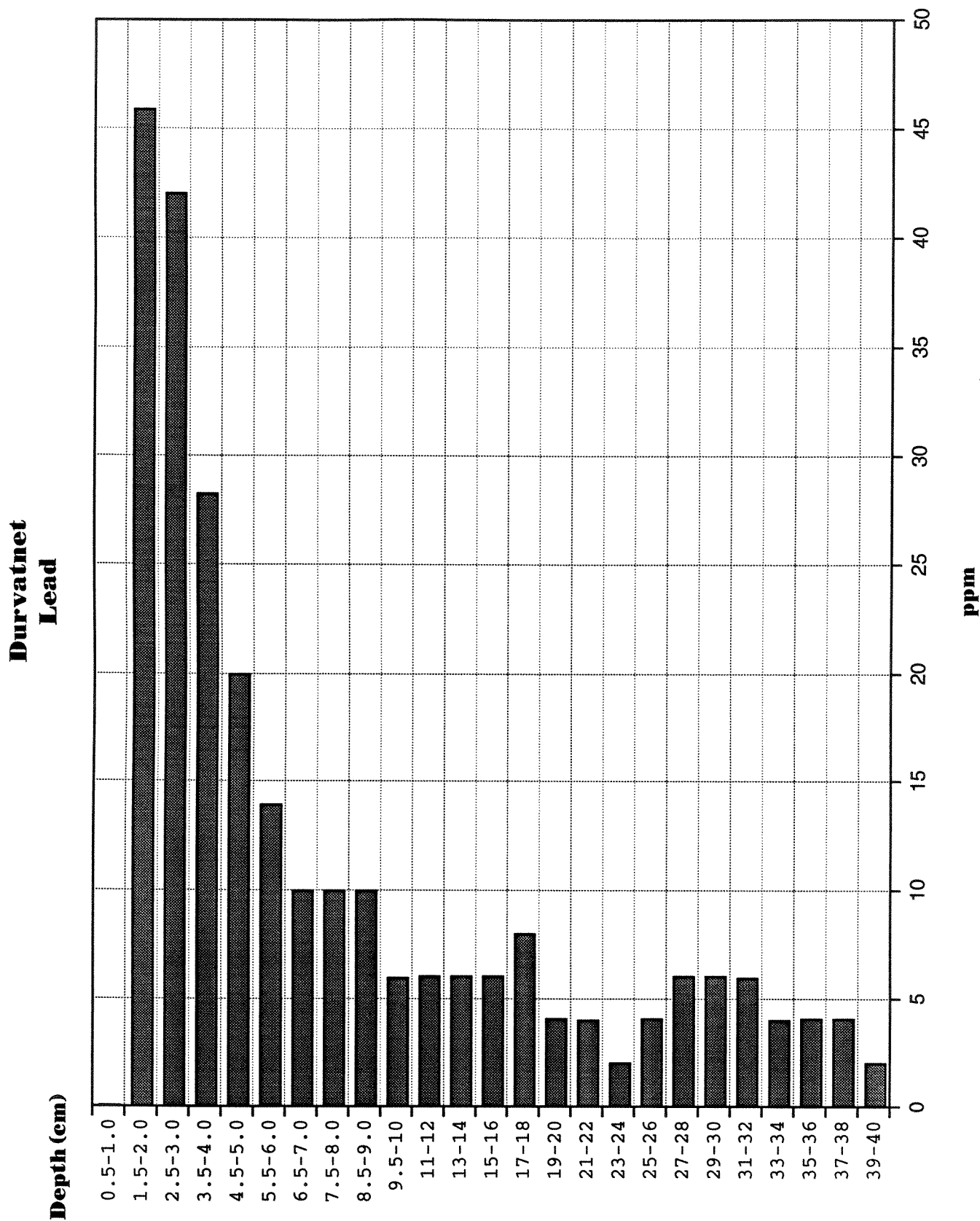
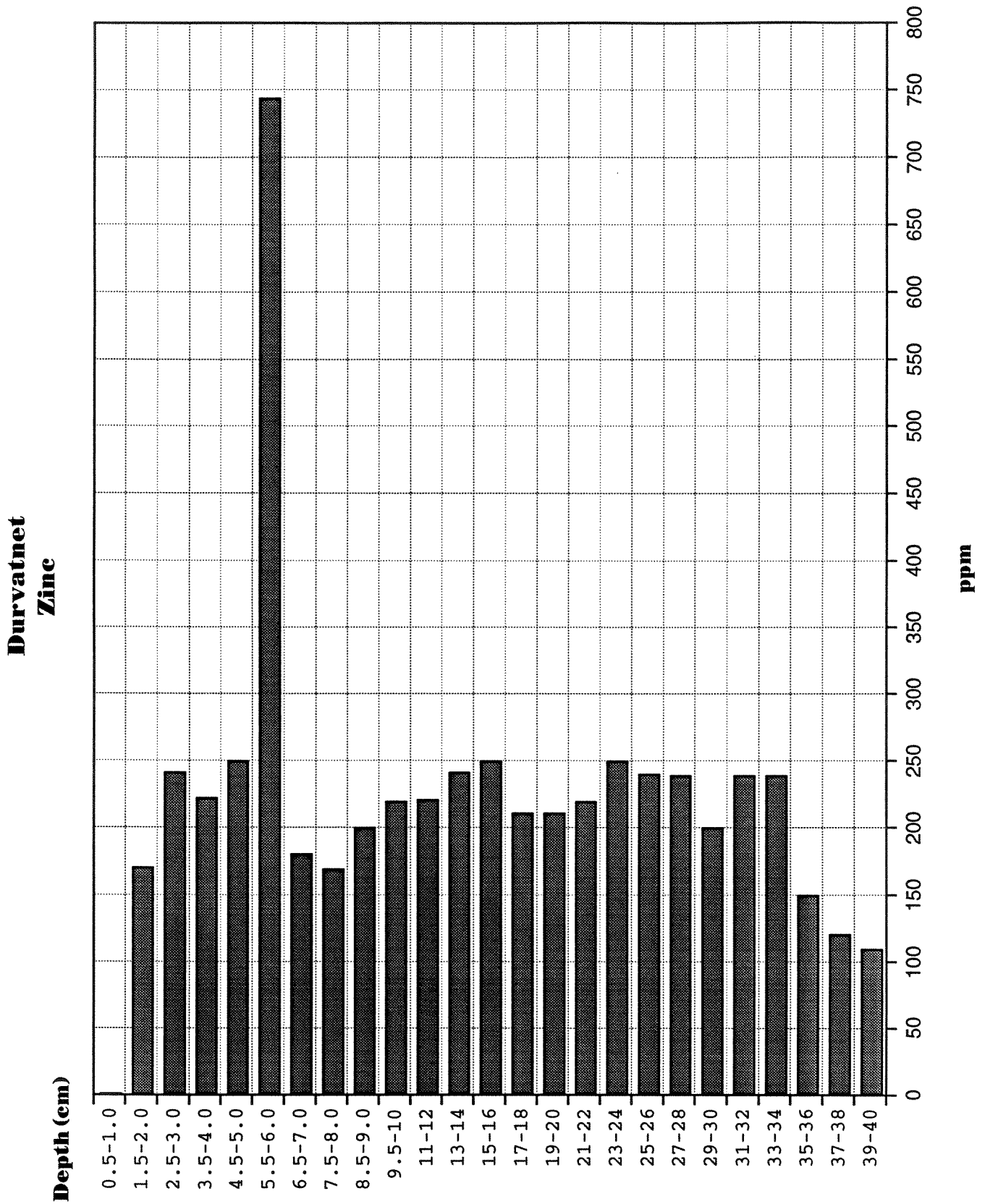


Figure 13. Concentrations (parts per million of ashed sediment, ppm) of Pb in a core from Durvatnet, eastern Finnmark, Norway.



**Figure 14. Concentrations (parts per million of ashed sediment, ppm) of Zn in a core from Durvatnet, eastern Finnmark, Norway.**

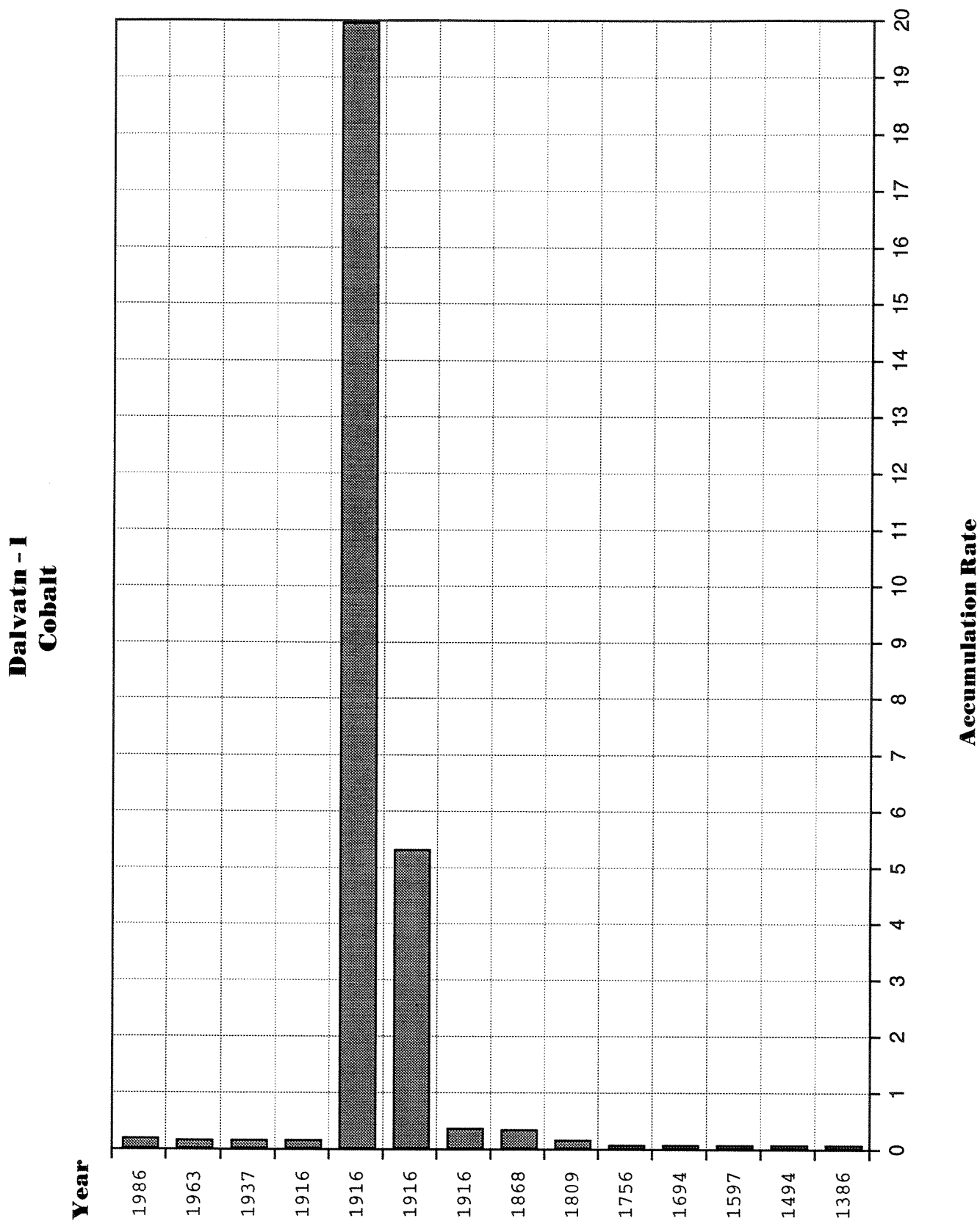


Figure15. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Co in a core from Dalvatn, eastern Finnmark, Norway.

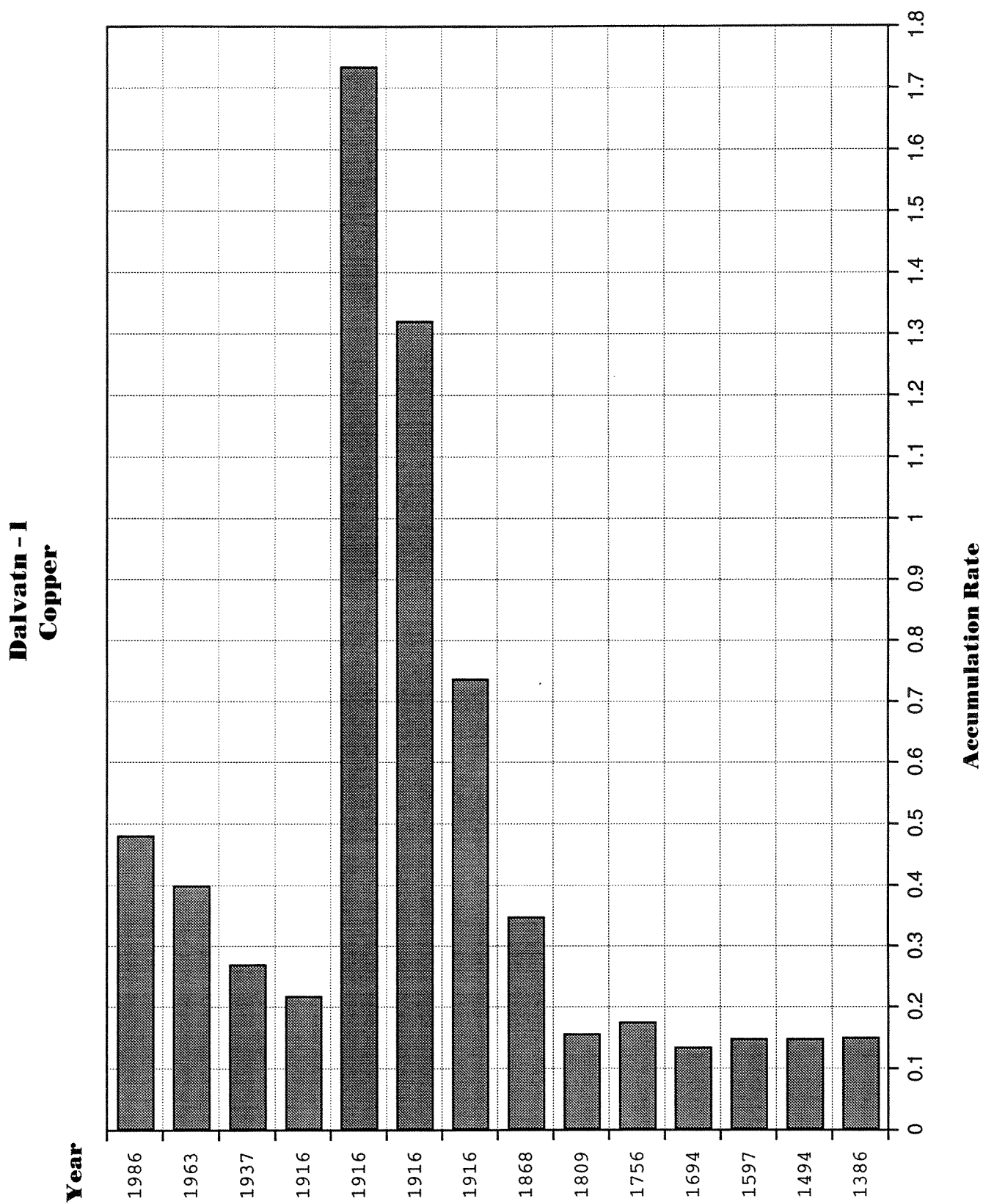


Figure 16. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Cu in a core from Dalvatn, eastern Finnmark, Norway.



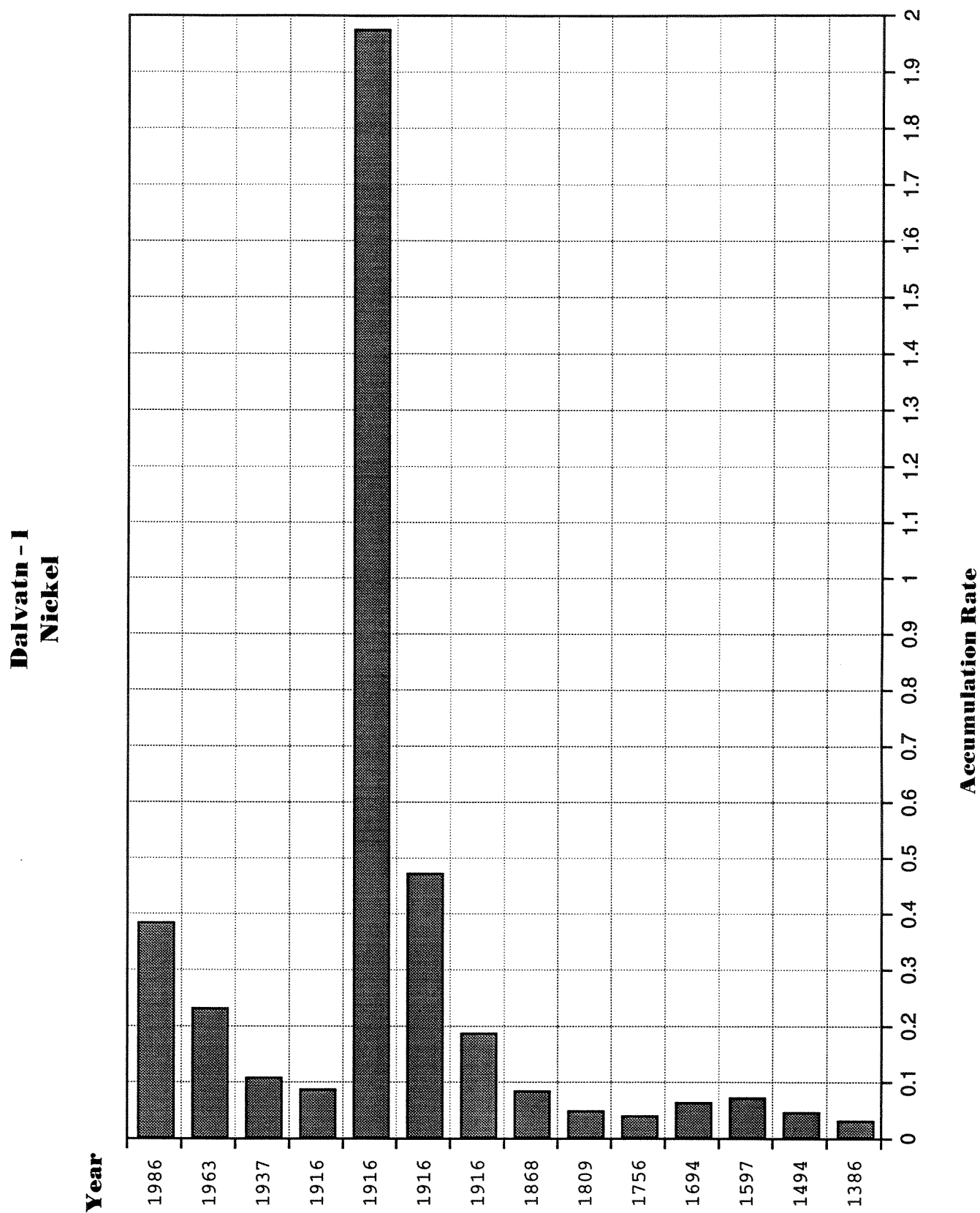


Figure 17. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Ni in a core from Dalvatn, eastern Finnmark, Norway.

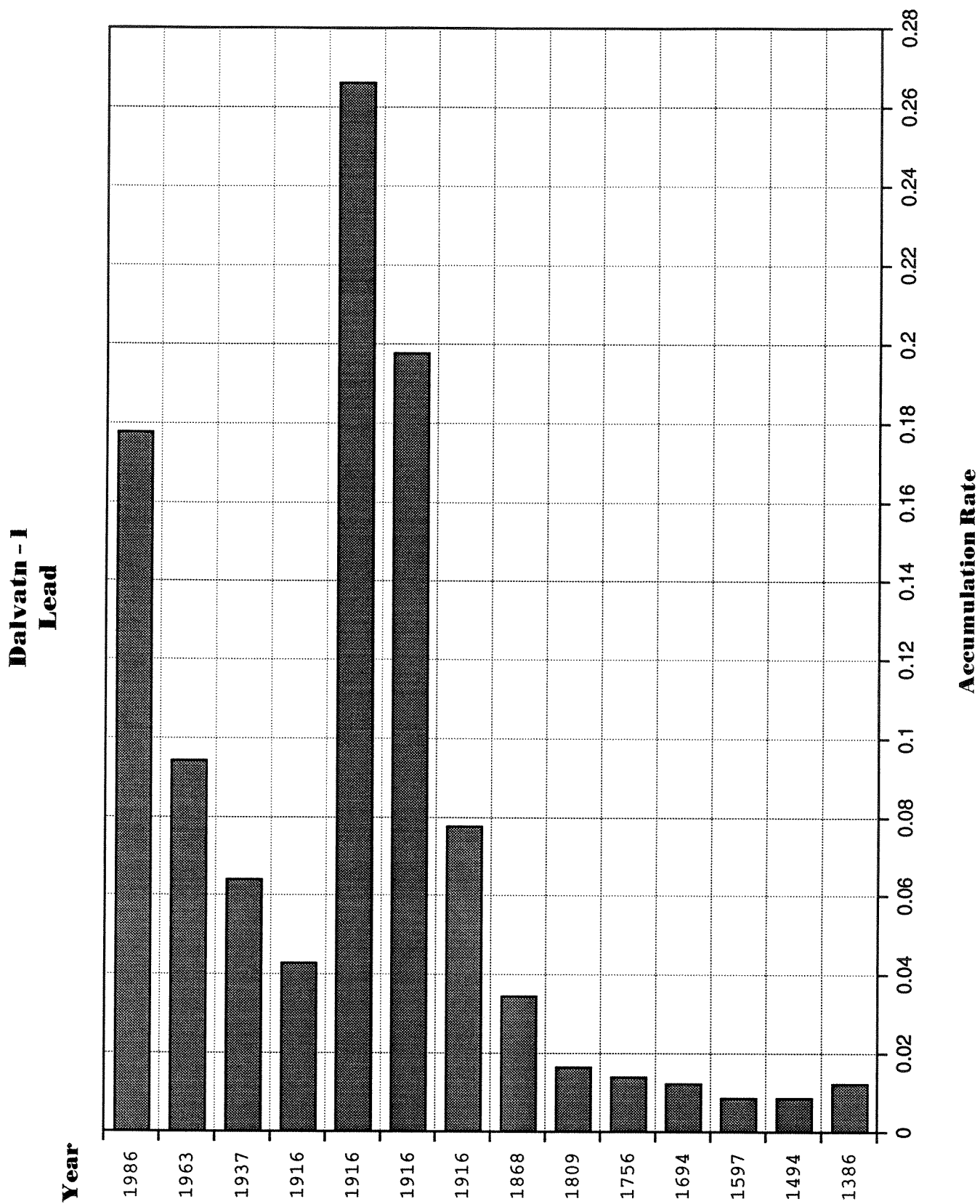


Figure 18. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Pb in a core from Dalvatn, eastern Finnmark, Norway.

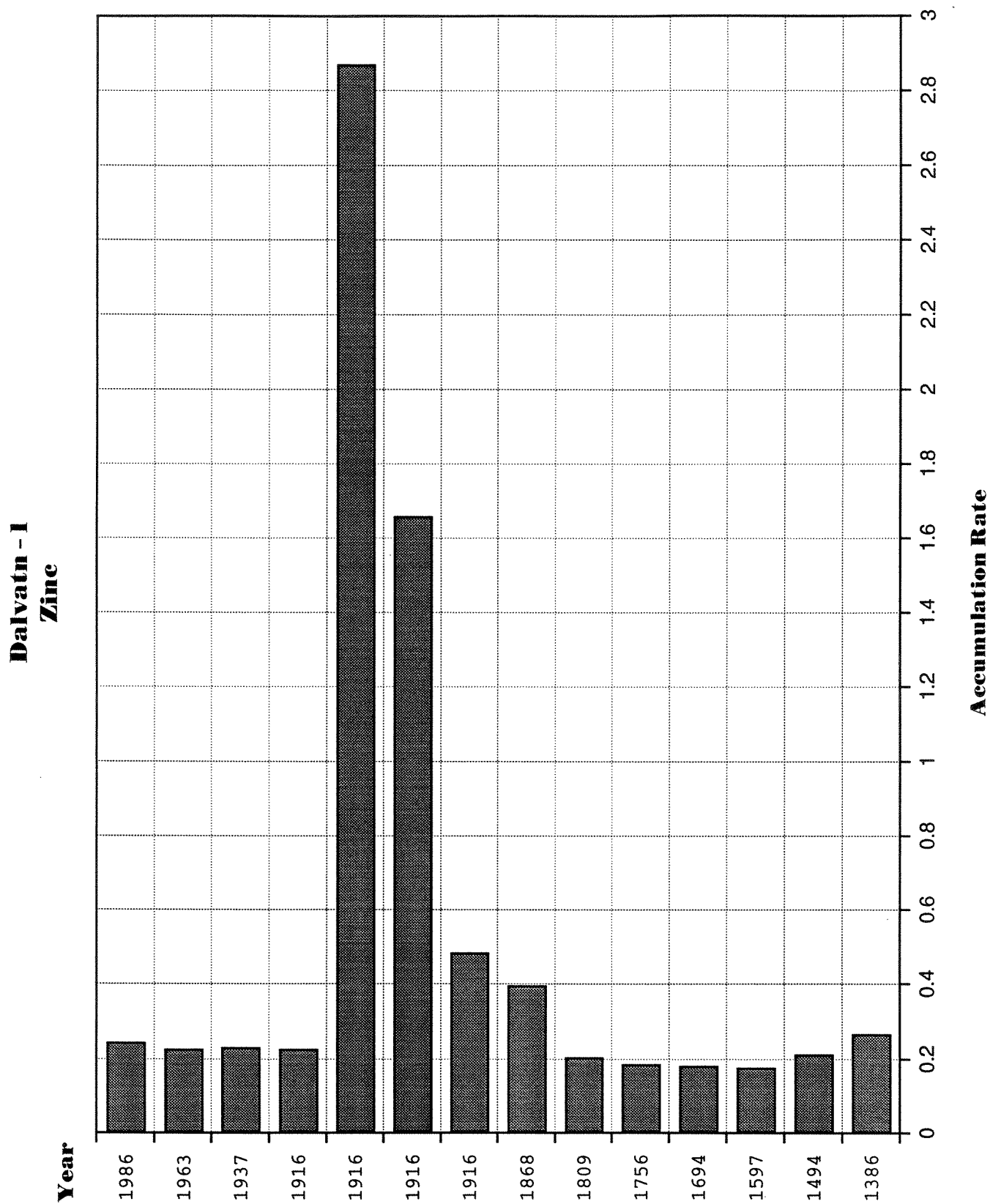


Figure 19. Accumulation rates ( $\mu\text{g cm}^{-2} \text{ yr}^{-1}$ ) of Zn in a core from Dalvatn, eastern Finnmark, Norway.

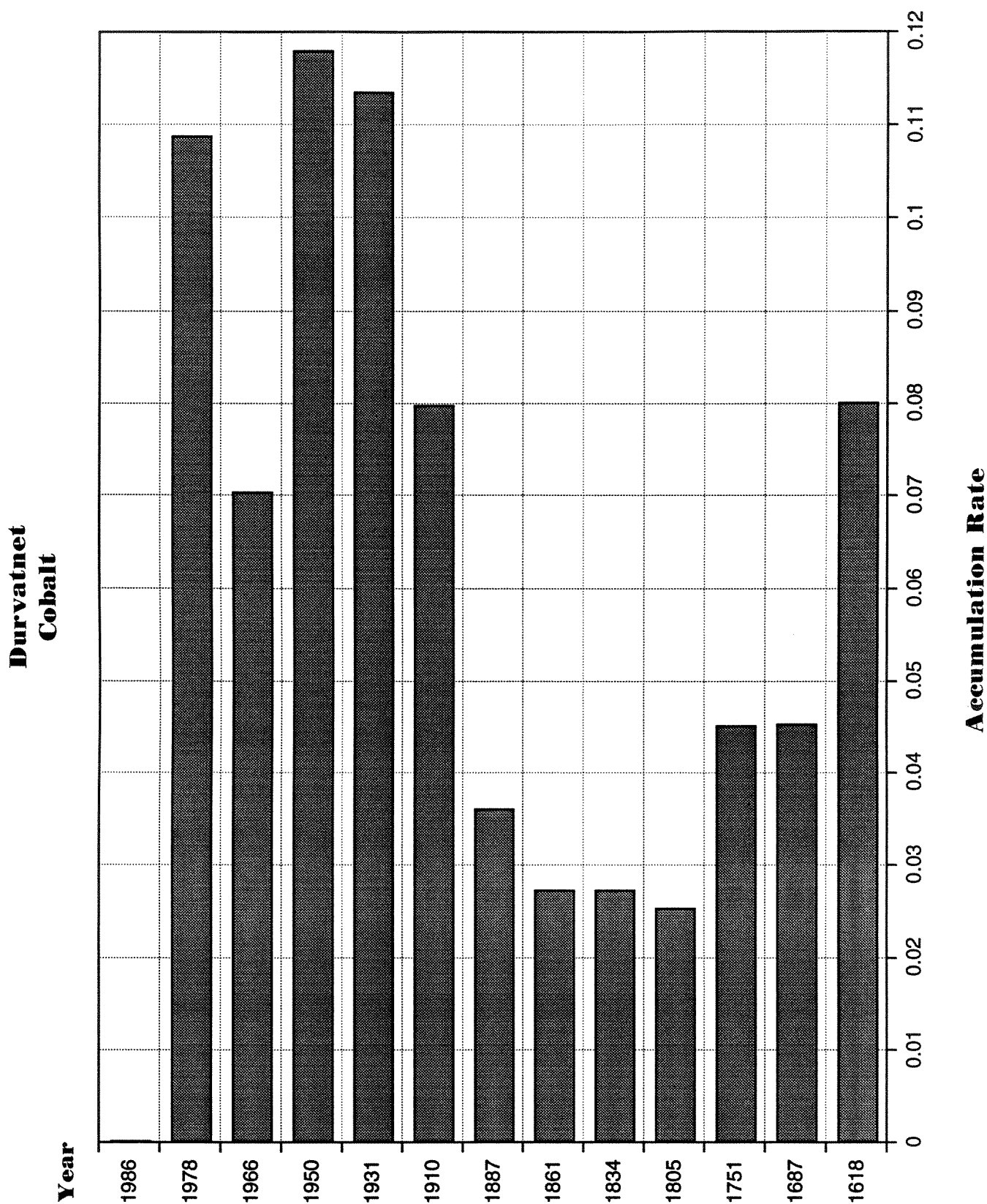


Figure 20. Accumulation rates ( $\mu\text{g cm}^{-2} \text{ yr}^{-1}$ ) of Co in a core from Durvatnet, eastern Finnmark, Norway.

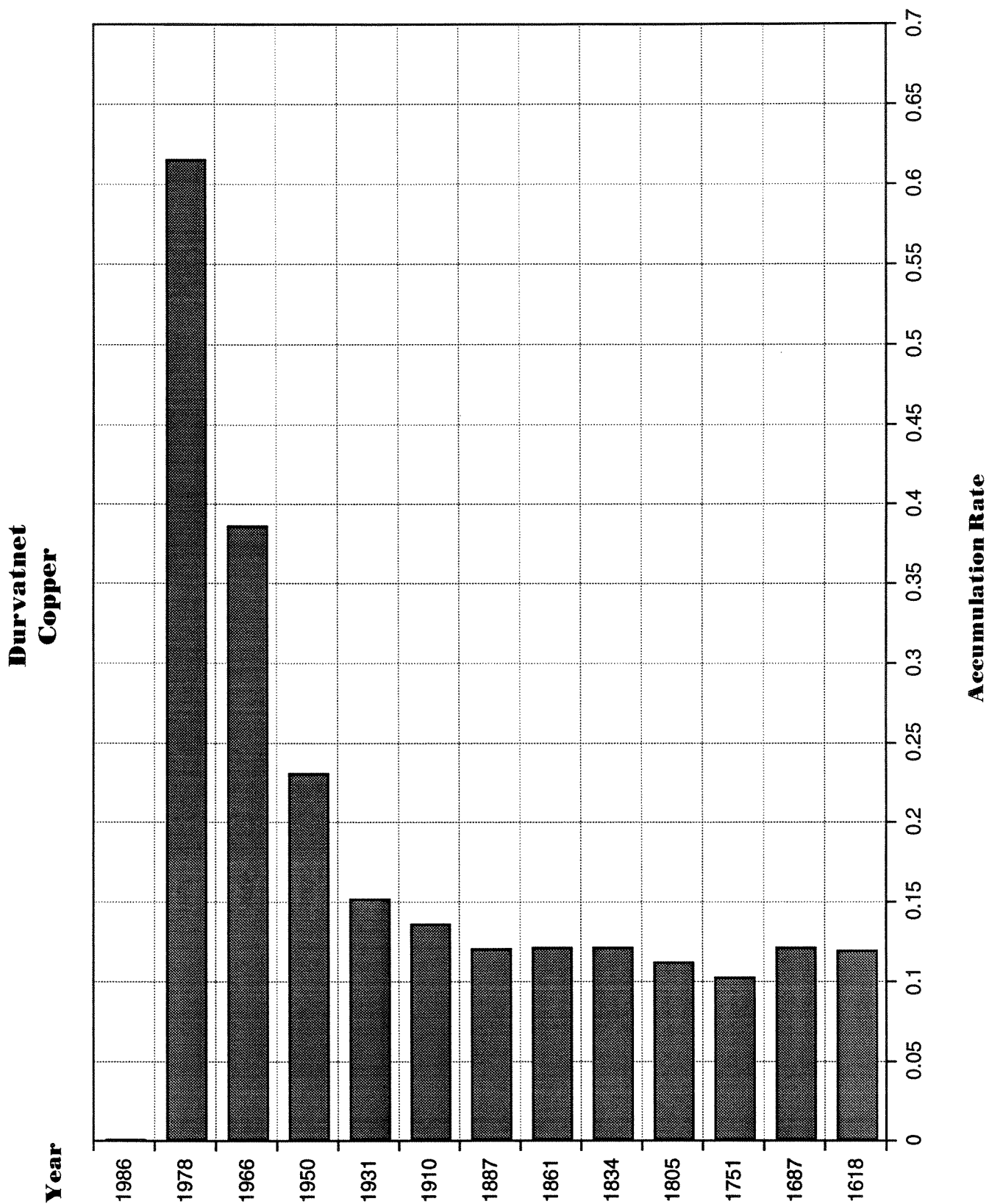


Figure 21. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Cu in a core from Durvatnet, eastern Finnmark, Norway.

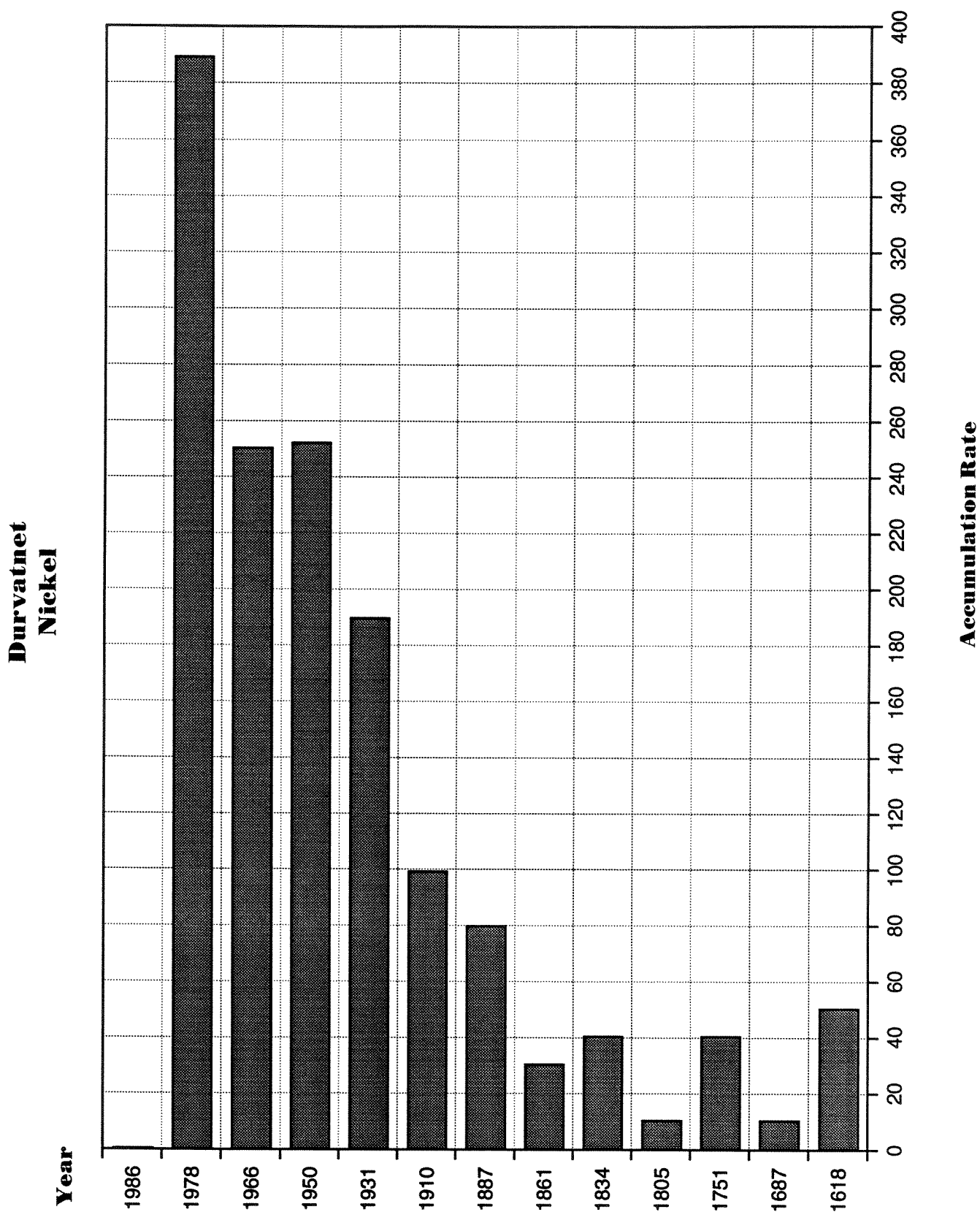


Figure 22. Accumulation rates ( $\mu\text{g cm}^{-2} \text{ yr}^{-1}$ ) of Ni in a core from Durvatnet, eastern Finnmark, Norway.

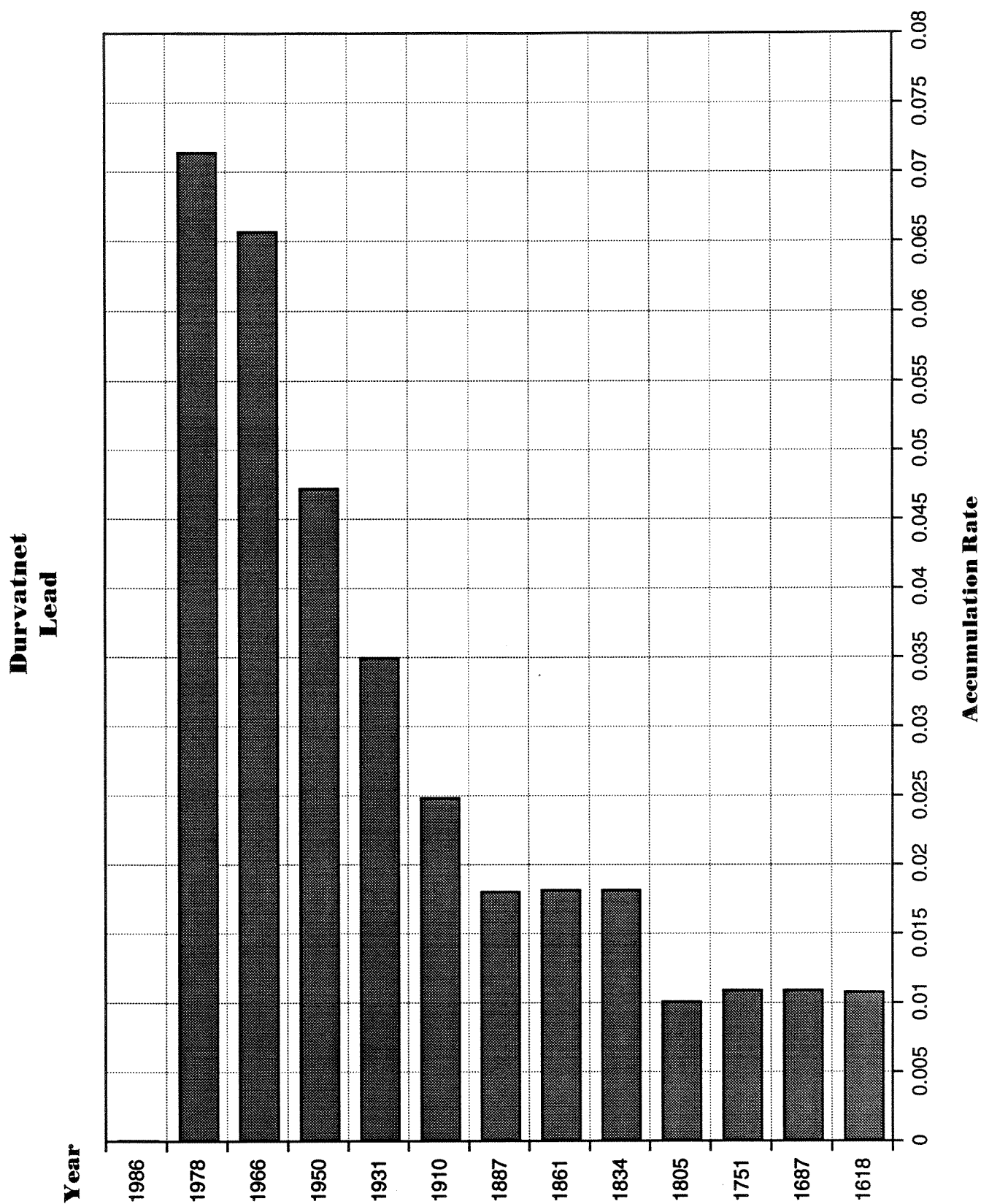


Figure 23. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Pb in a core from Durvatnet, eastern Finnmark, Norway.

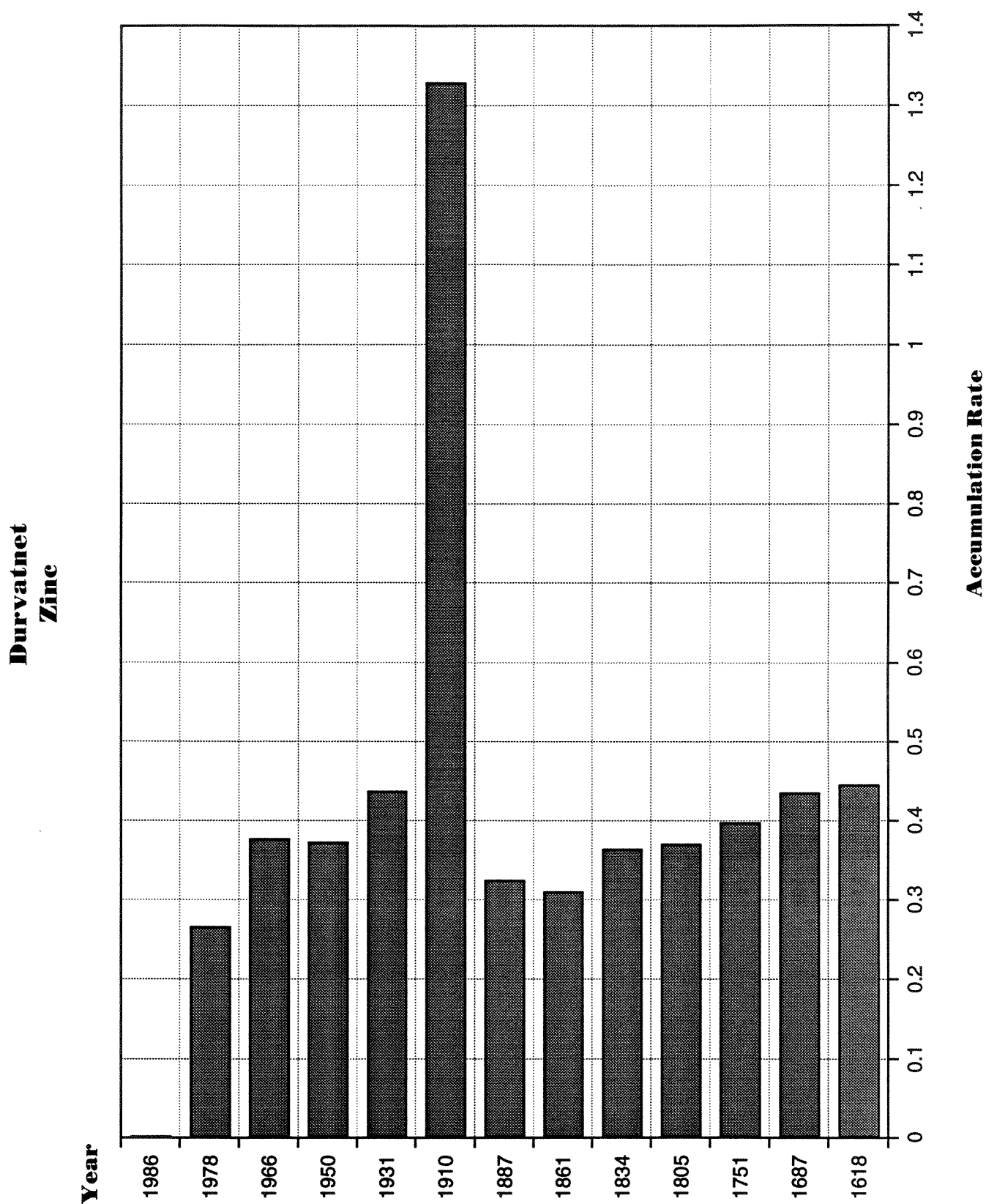


Figure 24. Accumulation rates ( $\mu\text{g cm}^{-2} \text{yr}^{-1}$ ) of Zn in a core from Durvatnet, eastern Finnmark, Norway.



Table 1. Activities of radionuclides in a sediment core from Dalvatn, eastern Finnmark, Norway.

Depth <sup>1</sup> cm	Dry Mass gcm <sup>-2</sup>	210Pb Conc. <sup>2</sup>		226Ra Conc. <sup>2</sup>		137Cs Conc. <sup>2</sup>		134Cs Conc. <sup>2</sup>		241Am Conc. <sup>2</sup>	
		Total pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	+ pCi g <sup>-1</sup>	
0.75	0.0242	53.65	0.83	0.74	0.17	16.03	0.19	3.30	0.38	0.20	0.04
1.75	0.0773	23.65	0.74	0.81	0.24	10.83	0.22	2.42	0.53	0.08	0.03
2.75	0.1255	9.25	0.26	0.81	0.12	6.98	0.10	0.00	0.00	0.03	0.01
3.75	0.1797	6.09	0.36	0.96	0.43	4.05	0.13	0.00	0.00	0.00	0.00
4.75	0.2481	2.83	0.23	-3.33	0.29	1.36	0.05	0.00	0.00	0.01	0.00
5.75	0.3512	1.91	0.24	0.30	0.37	1.01	0.08	0.00	0.00	0.00	0.00
6.75	0.4351	6.97	0.21	5.99	0.26	1.83	0.04	0.00	0.00	0.00	0.00
7.75	0.5196	1.79	0.19	0.50	0.23	0.91	0.06	0.00	0.00	0.00	0.00
9.75	0.6567	1.45	0.21	0.23	0.27	0.55	0.04	0.00	0.00	0.00	0.00
11.50	0.7644	1.27	0.18	-0.01	0.19	0.26	0.03	0.00	0.00	0.00	0.00
13.50	0.8935	1.19	0.21	-0.14	0.23	0.19	0.04	0.00	0.00	0.00	0.00
17.50	1.1742	1.29	0.18	-0.09	0.21	0.06	0.03	0.00	0.00	0.00	0.00

Inventories: 2.58±0.06 pCi cm<sup>-2</sup> 0.29±0.03 pCi cm<sup>-2</sup> 0.02±0.01 pCi cm<sup>-2</sup>

<sup>1</sup>mid-point of sediment interval  
<sup>2</sup>dry weight basis

TABLE 1 - Lake Dalvatn

**Table 2. Sediment age as a function of depth for sediment cores from Dalvatn and Durvatn, eastern Finnmark, Norway.**

Depth	Lake Dalvatn			Lake Durvatnet		
	AD	age	±	AD	age	±
0.00	1990	0	0	1900	0	0
0.25	1986	4	1	1989	1	2
0.50	1982	8	1	1988	2	2
0.75	1977	13	2	1986	4	2
1.00	1970	20	2	1985	5	2
1.25	1963	27	2	1984	6	2
1.50	1956	34	2	1981	9	2
1.75	1949	41	2	1978	12	2
2.00	1943	47	2	1975	15	2
2.25	1937	53	3	1973	17	2
2.50	1930	60	3	1969	21	2
2.75	1924	66	3	1966	24	2
3.00	1917	63	3	1962	28	2
3.25	1916	74	4	1959	31	2
3.50	1916	74	4	1954	36	2
3.75	1916	74	4	1950	40	2
4.00	1916	74	4	1945	45	3
4.25	1916	74	4	1941	49	3
4.50	1916	74	4	1936	54	3
4.75	1916	74	4	1931	59	4
5.00	1916	74	4	1926	64	4
5.25	1916	74	4	1921	69	5
5.50	1916	74	4	1916	74	5
5.75	1916	74	4	1910	80	6
6.00	1916	74	4	1905	85	7
6.25	1916	74	4	1899	91	8
6.50	1916	74	4	1893	97	9
6.75	1900	90	4	1887	103	9
7.00	1884	106	6	1881	109	10
7.25	1868	122	8	1875	115	11
7.50	1851	139	11	1968	122	12
7.75	1835	155	13	1961	129	12
8.00				1855	135	12
8.25				1848	142	13

Table 3. Activities of radionuclides in a sediment core from Durvatnet, eastern Finnmark, Norway.

Depth <sup>1</sup> cm	Dry Mass gcm <sup>-2</sup>	210Pb Conc. <sup>2</sup>		Unsupp. pCig <sup>-1</sup> ±		226Ra Conc. <sup>2</sup>		137Cs Conc. <sup>2</sup>		241Am Conc. <sup>2</sup>	
		Total pCig <sup>-1</sup>	±	pCig <sup>-1</sup>	±	pCig <sup>-1</sup>	±	pCig <sup>-1</sup>	±	pCig <sup>-1</sup>	±
0.25	0.0020	63.32	1.52	60.97	1.65	2.35	0.63	6.89	0.29	0.00	0.00
1.25	0.0151	49.50	2.58	48.87	2.75	0.63	0.94	6.84	0.48	0.13	0.07
2.25	0.0418	46.61	1.11	45.86	1.16	0.75	0.33	6.84	0.21	0.14	0.04
3.25	0.0758	29.38	0.72	28.63	0.75	0.75	0.21	4.81	0.14	0.16	0.04
4.25	0.1186	15.59	0.46	14.32	0.48	1.27	0.15	2.86	0.09	0.00	0.00
5.25	0.1662	10.20	0.39	8.66	0.44	1.54	0.21	1.73	0.10	0.04	0.02
6.25	0.2192	4.96	0.36	3.66	0.39	1.30	0.15	1.11	0.07	0.00	0.00
7.25	0.2785	2.63	0.31	1.03	0.34	1.60	0.15	1.03	0.06	0.00	0.00
8.25	0.3424	2.32	0.26	0.75	0.29	1.57	0.12	0.72	0.05	0.00	0.00
9.25	0.4088	2.17	0.22	0.63	0.24	1.54	0.09	0.51	0.04	0.00	0.00
12.50	0.6476	1.62	0.16	0.11	0.20	1.51	0.12	0.26	0.05	0.00	0.00
14.50	0.8109	1.08	0.21	-0.25	0.24	1.33	0.12	0.21	0.04	0.00	0.00
16.50	0.9781	1.49	0.20	0.22	0.23	1.27	0.12	0.25	0.04	0.00	0.00

Inventories:

		1.16±0.03 pCicm <sup>-2</sup>		0.01±0.004 pCicm <sup>-2</sup>	
		0.00		0.00	

1 mid-point of sediment interval  
2 dry weight basis

TABLE 3 - Lake Durvatnet

**Table 4. Water and organic content for sediment cores from Dalvatn and Durvatnet, eastern Finnmark, Norway.**

Depth Interval	Dalvatn		Durvatnet	
	% H <sub>2</sub> O	% Organic	%H <sub>2</sub> O	% Organic
0-0.15	97.2	44	99.2	
0.5-1.0	94.9		98.7	37
1.0-1.5	94.7	41	98.2	
1.5-2.0	94.5		97.2	35
2.0-2.5	94.5	40	96.9	
2.5-3.0	94.3		96.8	35
3.0-3.5	94.3	38	96.2	
3.5-4.0	95.0		95.8	30
4.0-4.5	92.9	29	95.5	
4.5-5.0	90.2		95.4	27
5.0-5.5	89.9	27	95.2	
5.5-6.0	90.1		94.9	26
6.0-6.5	90.8	29	94.5	
6.5-7.0	90.8		94.3	25
7.0-7.5	91.9	32	94.0	
7.5-8.0	91.9		93.8	24
8.0-8.5	92.3	31	93.8	
8.5-9.0	93.6		93.7	24
9.0-9.5	93.4	34	93.3	
9.5-10.0	93.7		93.2	30
10-11	93.4	34	93.0	
11-12	93.7		93.0	25
12-13	93.6	36	92.4	
13-14	92.8		92.2	25
14-15	93.1	35	92.1	
15-16	93.1		92.1	26
16-17	92.9	34	91.9	
17-18	91.9		91.7	26
18-19	92.8	34	91.5	
19-20	92.8		90.9	24
20-21	92.6	32	91.1	
21-22	91.1		90.6	23
22-23	91.9	33	89.9	
23-24	91.9		90.1	23
24-25	92.5	35	90.0	
25-26	92.4		89.4	21
26-27	92.1	34	88.8	
27-28	93.1		88.6	20
28-29	92.9	36	88.5	
29-30	92.5		88.4	19

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