

Sewage history in the anoxic sediments of the fjord Nordåsvannet, western Norway: (II) The origin of the sedimented organic matter fraction

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Anoxic sediments retrieved from two locations in the deep parts of the fjord Nordåsvannet (located near the city of Bergen, western Norway) contain a record of changing sewage discharge from AD 1700 to the present. Four periods of sewage discharge history are documented: (1) Before 1700: Minor sewage discharge. (2) From 1700 to 1969: Increasing shallow water sewage discharge. (3) From 1969 to 1988: Major deep water sewage discharge. (4) After 1988: No sewage discharge. Loss-on-ignition (LOI, organic carbon) analysis, $\delta^{13}\text{C}$ and the C/N ratio of the total sedimentary organic matter fraction are used to distinguish between marine-produced, terrestrial and sewage-derived organic matter, and to relate these to the four periods. The sedimented organic matter composition shows three steps of development at both stations. (A) Below 15 cm: $\text{LOI} = 26 \pm 1\%$, $\delta^{13}\text{C} = -23 \pm 0.5\text{‰}$, $\text{C/N} = 11 \pm 0.5$ corresponding to the time of no sewage discharge before 1700. (B) From 15 to 5 cm: LOI increases from 25 to 30%, $\delta^{13}\text{C}$ varies between -23.7 and -25.8‰ , C/N between 10.6 and 13.7 corresponding to the period of shallow water sewage discharge from 1700 to 1969. (C) Above 5 cm: LOI decreases to 22 and 28%, $\delta^{13}\text{C}$ and C/N kept stable at $-25 \pm 0.2\text{‰}$ and 13 ± 0.6 , respectively, corresponding to the period of deep water sewage discharge from 1969 to 1988 and of no sewage discharge from 1988 to 1991. Further, step (C) is characterized by a marked increase in grain size from silty clay to sandy/clayey silt, increasing LOI mass accumulation rates (from 12 to 27 $\text{gC/m}^2/\text{y}$) and exponentially increasing trace metal content.

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Introduction

Tracing the origin of organic matter in marine sediments is important for the understanding of the geological record of a variety of processes. These include human-induced environmental changes (Nissenbaum & Kaplan 1972; Tan & Strain 1979; Arthur et al. 1983; Wiesner et al. 1990; Reimers et al. 1992), climatic variations (Müller & Suess 1979; Müller & Erlenkeuser 1983; Stein 1991), the formation of fossil fuels and gas (Emerson & Hedges 1988; Waples 1981) and the origin of black shales (Sackett 1964; Pedersen & Calvert 1990; Tyson & Pearson 1991).

Anoxic and semi-enclosed fjords with sedimentation rates in the order of millimetres to centimetres per year, contain well-preserved, high resolution signals of recent, i.e. younger than 150 y, environmental changes (Smith & Schafer 1987; Anderson et al. 1990; Haugen & Lichtenaler 1991; Parker & O'Reilly 1991; Calvert et al. 1992; Paetzel & Schrader 1992; Pedersen et al. 1992; Schimmelmann & Kastner 1993). The Nordåsvannet is such an environment, where the origin of the organic matter may be either marine, terrestrial or sewage. The carbon to nitrogen ratio (C/N ratio), the $^{13}\text{C}/^{12}\text{C}$ isotopic ratio ($\delta^{13}\text{C}$) of the total organic matter fraction, the bulk sediment organic carbon concentrations (presented as LOI) and mass accumulation rates (MAR, $\text{g/m}^2/\text{y}$) and diatom analysis are used to determine the origin of the organic material in the sediments.

Sedimentary organic matter content is high in the anoxic Nordåsvannet and the history of the sewage supply is well documented. In an earlier article the dating (^{134}Cs , ^{137}Cs , ^{210}Pb , lamina counting) of the sediments was dealt with, and the influence of the sewage discharge history on the sedimentary trace metal (Cu, Pb, Zn) accumulation (Paetzel et al. 1994) was discussed. The objective of this study is to develop models of variations in the organic carbon deposition through recent time (150 y) and to answer the following questions about the Nordåsvannet sedimentary record:

Can the origin of the organic matter fraction be distinguished in the sediment by the methods used?

How much organic matter from the different sources was deposited on the fjord bottom?

Does the composition of the sedimentary organic matter fraction indicate variations in the significance of the sources through time?

Throughout this study, the term '(sedimentary) organic matter' is used for the total organic material that was incorporated into the sediment. The term 'LOI' defines the carbon fraction of the organic matter, i.e. organic carbon, that was determined by the loss-on-ignition technique at 550°C . Age is given in AD (*anno domini*).

Environmental setting

Location and bathymetry

The Nordåsvannet is located about 5 km south of the city limits of Bergen (western Norway). It consists of two main basins which are separated by a 12 m deep sill. Maximum depth is 54 m in the outer and 88 m in the inner basin. The northeastern part of the inner basin at Fjøsanger is divided into three 40 m deep troughs which are separated from the deepest depression of the fjord by a 20 m deep sill. The western outlet of the fjord consists of two parallel channels at Straume, each 12 m wide, 30–40 m long and about 3 m deep. The Nordåsvannet is linked to the oxic Grimstadfjord and Raunefjord which are connected to the North Sea. Freshwater enters the fjord mainly through three river inlets in the north, east and south of the inner basin, and through a number of smaller inlets around the fjord (Fig. 1).

Most recent history

Settlement around the Nordåsvannet started about 1700. The population increased gradually until today, with a

major building activity from 1920 to 1950. We assume that only insignificant amounts of sewage reached the fjord waters before 1700. Sewage was surface discharged mainly to the inner basin through a number of sources until 1969. From 1969 to 1982, sewage was collected and discharged through two 40 m deep sewage outlets at Fjøsanger and Skjold (Fig. 1). The latter type of discharge continued during periods of heavy rainfall until 1988, but no sewage was discharged into the fjord after 1988. The sewage production in 1980 was about 30,000 person equivalents per year (Wassmann 1985) and has not changed significantly during the last 40 years (Hogne Hjelle 1992, *pers. comm.*).

Hydrography

Salinity (salinometer) and dissolved oxygen concentrations (Winkler titration) of the water column were determined through the following periods of time: 1912–14 (Gaarder 1916), 1941–42 (Wiborg 1944), 1969–70 (Johannessen 1972), 1979–84 (Johannessen 1988) and 1990–91 (Johannessen et al. 1990/91). Sampling frequency was one sample per depth every second month for most of the measured periods.

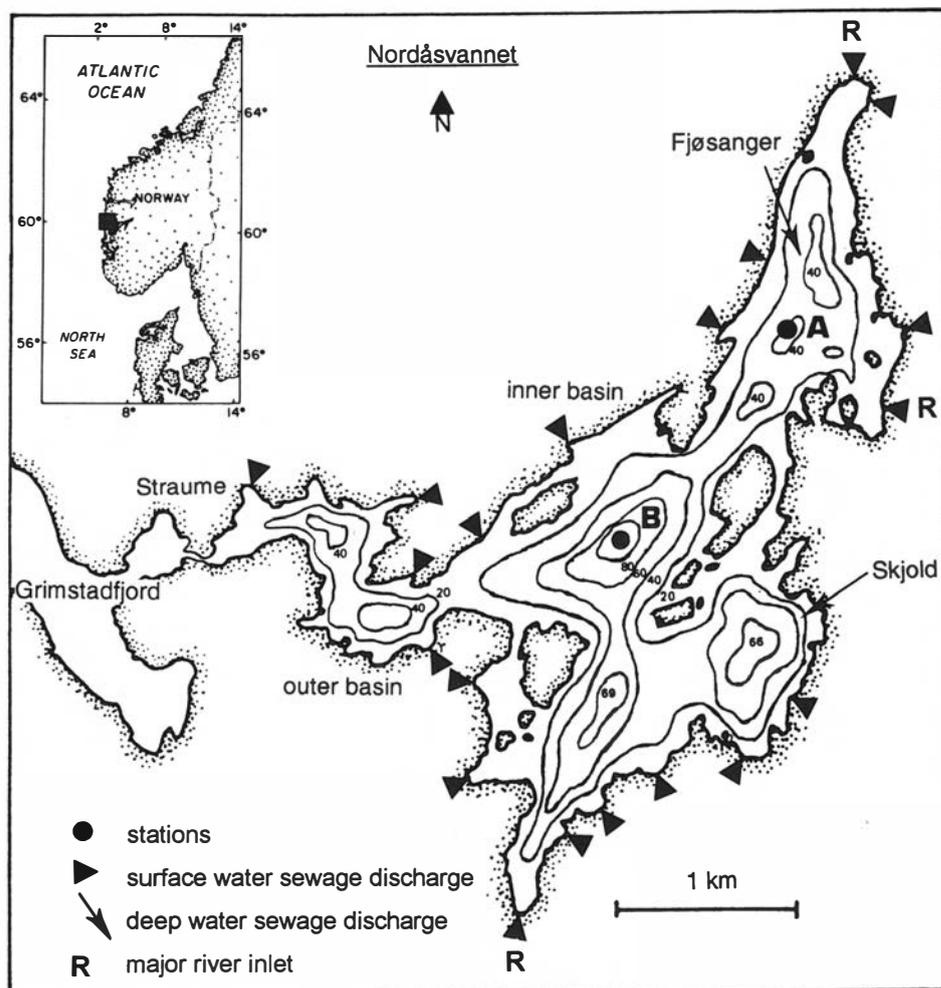


Fig. 1. Location map of Stations A and B (black dots), and bathymetry of the Nordåsvannet. Equidistance of depth lines is 20 m. Black triangles = surface water sewage discharge. Arrows = deep water sewage discharge. R = major river inlets.

Salinity did not vary significantly from the first investigation in 1941/42 until today. It stayed constant at 31‰ below 80 m and varied between 26 and 31‰ from 10 to 40 m. Surface water salinity was lowest in spring and autumn (3–10‰), and highest in summer and winter (21–27‰), depending on precipitation and meltwater volume.

The oxycline varied between 0 m in spring and autumn and 10 m during the summer. Dissolved oxygen content was 7–11 mlO₂/l at the surface and 2–8 mlO₂/l at 10 m. Oxygen usually was depleted below 20 m during autumn (0–0.2 mlO₂/l). Elevated oxygen levels were observed at 20 m from 1979 to 1984 when oxygen concentrations did not decline below 2 mlO₂/l. Oxygen renewal below 40 m was reported in the investigations carried out before 1943.

The total water volume of the Nordåsvannet is 1.4×10^8 m³. Precipitation and meltwater runoff discharge an annual freshwater volume of 1.3×10^8 m³ to the fjord. About 1.01×10^8 m³/year leaves the fjord with the tides through the channels at Straume. The annual surface sewage supply was 0.31×10^8 m³ before 1969 (Glennie & Simensen 1963). Approximately the same annual amount of sewage entered the fjord during the time of the deep water discharge from 1969 to 1988 (Hogne Hjelle & Alf Baadsvik 1992, *pers. comm.*).

Primary production and organic carbon sedimentation

Primary production culminates in three bloom maxima per year in the Nordåsvannet (Wiborg 1944; Braarud & Hope 1952; Wassmann et al. 1986). The spring bloom occurs as a light-induced maximum in March/April and another, meltwater induced, maximum in May/June. The summer period from June to August is characterized by a continuous production without any maxima. The autumn bloom is initiated by increased runoff in September. The production of particulate organic carbon is 190 gC/m²/y according to sediment trap data from a 20 m water depth (Wassmann et al. 1986).

Dissolved inorganic nutrients such as nitrogen and phosphorous do not limit primary production in the Nordåsvannet. The major limiting factor is light (Wassmann 1985). The sources of the nutrients in the photic zone are: The supply of phosphate and nitrogen compounds from the sewage. The accumulation of nitrate and phosphate during the winter when no consumption occurs, and when the fjord gains marine-supplied nutrients through the channel at Straume. Nutrients reach the photic zone by diffusion of nutrient-rich waters from below in periods of enhanced estuarine circulation.

El-Sayed (1982) documented an organic carbon content of 10–12% (LECO technique) in sediments taken at 88 m by a conventional gravity core. Grab samples of Johannessen et al. (1990/91) revealed a surface sediment LOI (organic carbon concentration determined by loss-on-ignition analysis) of about 29%.

Methods

Material

Undisturbed cores with anoxic sediments were retrieved from two stations in the inner basin of the Nordåsvannet in 1991 (Fig. 1). The samples were taken with a modified Niemistö corer which allows two samples sediments with intact sediment/water interfaces (Niemistö 1974). The sampling locations were: Station A at 37 m depth, 300 m southeast of the Fjøsanger deep sewage outlet (latitude 60°20.00'N, longitude 04°20.08'E), and Station B at 88 m depth, 2.5 km southeast of the Fjøsanger deep sewage outlet (latitude 60°19.34'N, longitude 04°18.98'E).

Plankton tows were made above Station A and B through the top 20 m of the water column using a 40 µm mesh-size plankton net with a 30 cm opening diameter. The plankton samples were stored in wide-mouth plastic bottles. Smear slides of the samples were taken immediately upon arrival at the laboratory. The plankton samples were stored at 4°C overnight, before further analysis. Bulk sewage samples were collected at the Knappen sewage treatment plant located at the Grimstadfjord.

Loss-on-ignition (LOI, organic carbon)

The sedimentary organic carbon concentrations, expressed as LOI, were measured on 3 cm thick sediment slices, using the loss-on-ignition technique at 550°C. The release of crystal water from clay minerals during the ignition process may result in an overestimation of the organic carbon percentages. According to Svensson (1989), this error is minor in organic-matter-rich sediments. However, the results of El-Sayed (1982) on LECO-derived organic carbon concentrations from Nordåsvannet sediments suggest that our LOI-derived organic carbon content is a maximum estimate. Mass accumulation rates (gC/m²/y) of the LOI concentrations were calculated using the formula of Dymond & Veeh (1975), and sedimentation rates and sediment physical parameters of Paetzel et al. (1994).

C/N ratio and δ¹³C isotopic ratio

The bulk plankton samples were filtered over a 0.45 µm NUCLEPORE filter using a water jet vacuum pump. After filtering they were frozen with liquid nitrogen, and freeze-dried. The bulk sediment and sewage samples were oven-dried at 50°C. These samples and the dry plankton samples were crushed to powder using an agate mortar and then filled into small glass beakers. CaCO₃ was then removed by twice adding 1M HCl and evaporating to dryness. About 5–10 mg of this sample was weighed into 5x9 mm aluminium capsules. The samples were analysed for total organic carbon, total organic nitrogen and ¹³C/¹²C isotope concentration using a CARLO ERBA Nitrogen Analyser and a FINNIGAN MAT Mass Spectrometer. The C/N ratio includes a relative error of ±5%

Table 1. Raw data from all analyses carried out on sediment subsamples taken at Station A and Station B in the Nordåsvannet. pl = bulk plankton sample. sw = bulk sewage sample. C/N ratio on weight basis.

| Depth cm | LOI % | Depth cm | C % | N % | C/N ratio | $\delta^{13}\text{C}$ ‰ | Depth cm | Sand % | Silt % | Clay % | Depth cm | Zn ppm |
|------------------|----------|-------------|--------|--------|--------------|----------------------------|-------------|-----------|-----------|-----------|-------------|-----------|
| <i>Station A</i> | | | | | | | | | | | | |
| 0.0–3.0 | 23 | 0.5 | 13.72 | 1.01 | 13.6 | –25.1 | 0.1 | 2 | 68 | 30 | 0.0–1.5 | 788 |
| 3.0–6.0 | 22 | 2.5 | 8.58 | 0.68 | 12.6 | –25.0 | 0.5 | 0 | 65 | 35 | 1.5–3.0 | 750 |
| 6.0–9.0 | 30 | 5.0 | 10.55 | 0.77 | 13.7 | –25.8 | 1.5 | 0 | 65 | 35 | 3.0–4.5 | 451 |
| 9.0–12.0 | 30 | 10.0 | 11.73 | 0.93 | 12.6 | –25.0 | 2.5 | 10 | 65 | 25 | 4.5–6.0 | 165 |
| 12.0–15.0 | 29 | 15.0 | 11.81 | 1.04 | 11.4 | –24.7 | 3.5 | 0 | 45 | 55 | 6.0–7.5 | 89 |
| 15.0–17.5 | 27 | 20.0 | 11.65 | 1.07 | 10.9 | –23.7 | 5.0 | 0 | 40 | 60 | 7.5–9.0 | 73 |
| 17.5–20.0 | 25 | 25.0 | 11.74 | 1.06 | 11.1 | –23.3 | 10.0 | 0 | 35 | 65 | 9.0–10.5 | 69 |
| 20.0–22.5 | 27 | 30.0 | 11.51 | 1.04 | 11.1 | –23.4 | 15.0 | 0 | 35 | 65 | 10.5–12.0 | 68 |
| 22.5–25.0 | 27 | 35.0 | 12.35 | 1.09 | 11.3 | –23.9 | 20.0 | 3 | 40 | 58 | 12.0–13.5 | 67 |
| 25.0–27.5 | 27 | 40.0 | 9.81 | 0.87 | 11.3 | –23.9 | 30.0 | 0 | 35 | 65 | 13.5–15.0 | 64 |
| 27.5–30.0 | 24 | 45.0 | 12.74 | 1.15 | 11.1 | –23.6 | | | | | 15.0–17.5 | 63 |
| 30.0–32.5 | 23 | | | | | | | | | | 17.5–20.0 | 58 |
| 32.5–35.0 | 27 | pl | 25.42 | 3.24 | 7.9 | –20.5 | | | | | 20.0–22.5 | 57 |
| 35.0–37.5 | 27 | sw | 20.85 | 1.69 | 12.3 | –26.4 | | | | | 22.5–25.0 | 56 |
| 37.5–40.0 | 26 | | | | | | | | | | 25.0–27.5 | 55 |
| | | | | | | | | | | | 27.5–30.0 | 53 |
| | | | | | | | | | | | 30.0–32.5 | 68 |
| | | | | | | | | | | | 32.5–35.0 | 66 |
| | | | | | | | | | | | 35.0–37.5 | 71 |
| | | | | | | | | | | | 37.5–40.0 | 64 |
| <i>Station B</i> | | | | | | | | | | | | |
| 0.0–3.0 | 28 | 0.5 | 8.72 | 0.71 | 12.3 | –24.7 | 0.1 | 0 | 40 | 60 | 0.0–1.5 | 807 |
| 3.0–6.0 | 36 | 2.5 | 10.44 | 0.76 | 13.7 | –25.0 | 0.5 | 0 | 40 | 60 | 1.5–3.0 | 506 |
| 6.0–9.0 | 30 | 4.0 | 6.37 | 0.60 | 10.6 | –23.7 | 1.5 | 5 | 50 | 45 | 3.0–4.5 | 110 |
| 9.0–12.0 | 28 | 9.0 | 9.81 | 0.79 | 12.4 | –24.6 | 2.5 | 5 | 47 | 48 | 4.5–6.0 | 93 |
| 12.0–15.0 | 26 | 14.0 | 10.43 | 0.87 | 12.0 | –24.6 | 3.5 | 0 | 30 | 70 | 6.0–7.5 | 112 |
| 15.0–17.5 | 24 | 19.0 | 11.81 | 1.11 | 10.6 | –23.8 | 5.0 | 0 | 37 | 63 | 7.5–9.0 | 96 |
| 17.5–20.0 | 24 | 24.0 | 14.73 | 1.41 | 10.4 | –22.9 | 10.0 | 0 | 40 | 60 | 9.0–10.5 | 88 |
| 22.0–22.5 | 26 | 29.0 | 11.70 | 1.15 | 10.2 | –22.5 | 14.0 | 0 | 40 | 60 | 10.5–12.0 | 81 |
| 22.5–25.0 | 26 | 34.0 | 11.44 | 1.09 | 10.5 | –22.7 | 19.0 | 0 | 30 | 70 | 12.0–13.5 | 73 |
| 25.0–27.5 | 24 | 39.0 | 10.45 | 1.01 | 10.3 | –22.9 | 34.0 | 0 | 25 | 75 | 13.5–15.0 | 75 |
| 27.5–30.0 | 25 | 44.0 | 10.08 | 1.01 | 10.0 | –22.4 | | | | | 15.0–17.5 | 71 |
| 30.0–32.5 | 27 | 49.0 | 10.06 | 1.00 | 10.1 | –22.4 | | | | | 17.5–20.0 | 72 |
| 32.5–35.0 | 26 | | | | | | | | | | 20.0–22.5 | 85 |
| | | pl | 20.83 | 2.64 | 7.9 | –19.6 | | | | | 22.5–25.0 | 81 |
| | | sw | 20.85 | 1.69 | 12.3 | –26.4 | | | | | 25.0–27.5 | 77 |
| | | | | | | | | | | | 27.5–30.0 | 74 |
| | | | | | | | | | | | 30.0–32.5 | 75 |
| | | | | | | | | | | | 32.5–35.0 | 76 |

of the measured values. The $\delta^{13}\text{C}$ isotopic ratio contains an absolute error of $\pm 0.05\text{‰}$.

Grain size and micro-organisms

MOUNTEX (MERCK, Article N°6184, refraction index ≈ 1.67) was used as a mounting medium for the smear slides. Grain size and siliceous microfossil analyses were done on a LEITZ SM-LUX POL polarizing microscope. The grain-size distribution was determined over the three fractions clay $< 4 \mu\text{m}$, silt 4 to $63 \mu\text{m}$ and sand $63 \mu\text{m}$ to 2 mm, according to the Udden-Wentworth size classification for sediment grains (Pettijohn et al. 1972). Pyrite was light microscopically determined according to the description by Rothwell (1989).

Smear slides for micro-organism analysis were taken continuously down-core across 2.5 cm thick sediment

slices. The sedimentary records of the planktonic diatom species *Tabellaria flocculosa* (freshwater) and *Chaetoceros sp.* (marine water) were used to indicate surface water salinity variations. A high abundance of *Chaetoceros sp.*, *Skeletonema costatum* and/or *Thalassiosira nordenskiöldii* was related to favourable nutrient conditions in the photic zone (Hustedt 1930–66). The occurrence of the high latitude silicoflagellate species *Distephanus speculum* was ascribed to predominantly marine surface water conditions with elevated nutrient concentrations (Lipps 1970).

Results

Raw data from all analyses carried out on sediment subsamples taken at Station A and Station B in the Nordåsvannet are presented in Table 1.

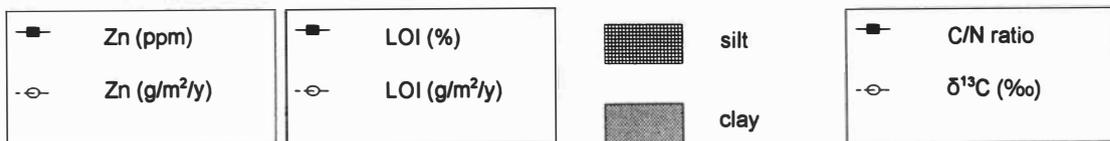
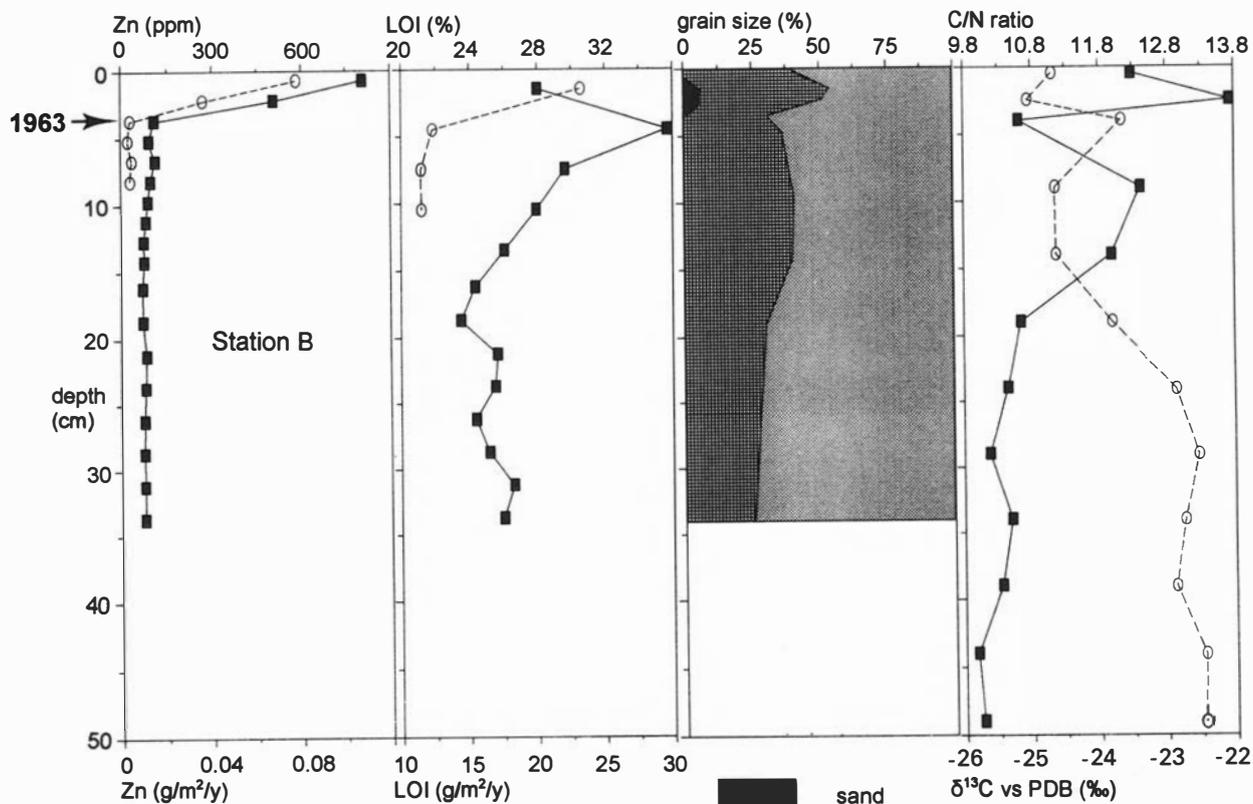
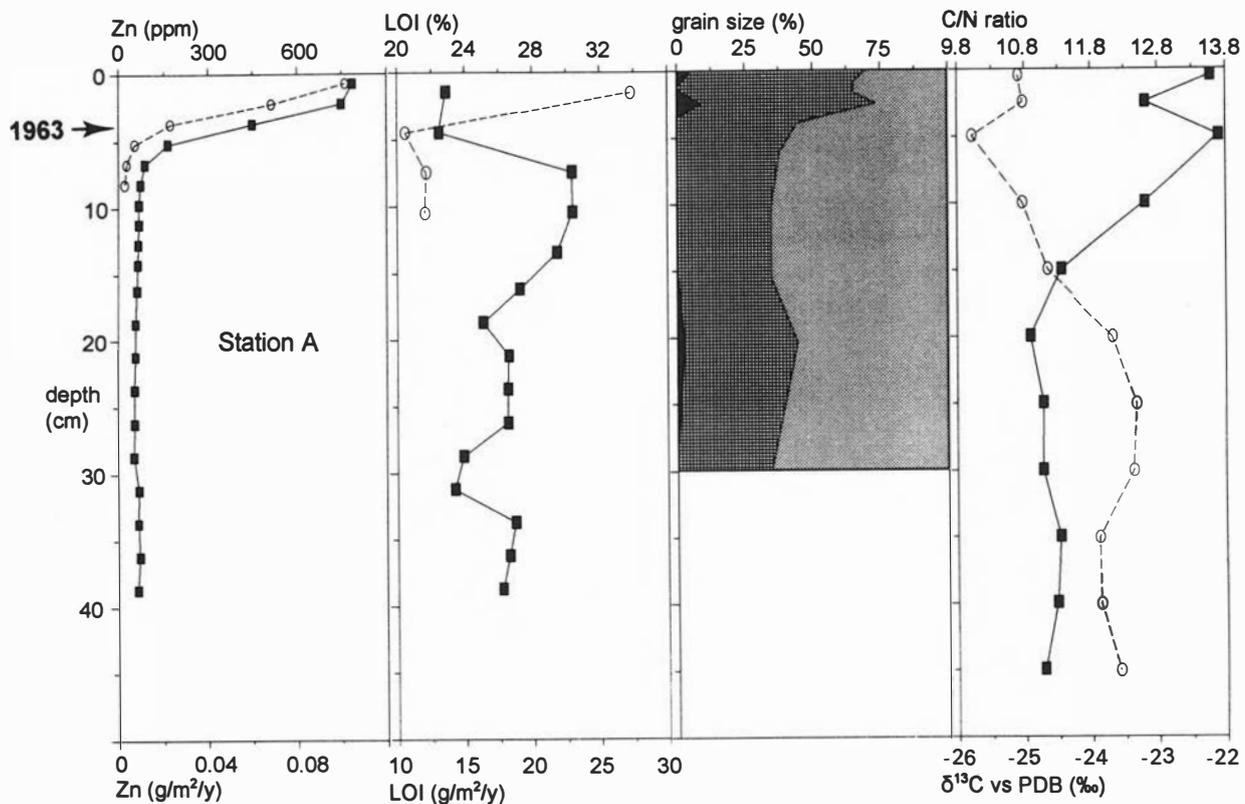


Table 2. Mean sediment parameter distribution below 15 cm ($n \geq 6$), from 15 to 5 cm ($n = 3$ or 4) and above 5 cm ($n \leq 2$) sediment depth. Calculation of accumulation rates ($\text{g/m}^2/\text{y}$) and Zn concentrations according to Paetzel et al. (1994). No standard deviation is indicated where $\sigma < 5\%$ of mean value.

| | LOI % | $\text{g/m}^2/\text{y}$ | C/N ratio | $\delta^{13}\text{C}$ ‰ | Grain size | | | Zn ppm | $\text{mg/m}^2/\text{y}$ |
|-----------------|------------|-------------------------|--------------|----------------------------|------------|------|------|---------------|--------------------------|
| | | | | | Sand | Silt | Clay | | |
| Station A | | | | | | | | | |
| Above 5 cm | 23 | 27 | 13 | -25 | 3 | 66 | 31 | 663 ± 150 | 96 ± 28 |
| From 15 to 5 cm | 28 ± 3 | 12 ± 1 | 13 ± 1 | -25 | 0 | 39 | 61 | 85 ± 33 | 7 ± 3 |
| Below 15 cm | 26 | — | 11 | -23 | 0 | 38 | 62 | 61 ± 6 | — |
| Station B | | | | | | | | | |
| Above 5 cm | 28 | 23 | 13 ± 1 | -25 | 3 | 44 | 53 | 657 ± 151 | 63 ± 17 |
| From 15 to 5 cm | 31 ± 3 | 12 ± 1 | 12 ± 1 | -24 | 0 | 37 | 63 | 91 ± 14 | 8 ± 2 |
| Below 15 cm | 25 | — | 10 | -23 | 0 | 28 | 73 | 76 ± 4 | — |

Structure and lithology

The sediments of both stations were finely laminated on a millimetre scale. The material was black, anoxic and rich in organic matter. Grain size of the minerogenic matter, excluding pyrite, increased at both stations from sand = 0, silt = 30–40 and clay = 60–70% below 5 cm, to sand = 3, silt = 40–60, and clay = 30–50% above 2.5 cm (Fig. 2, Table 2).

Organic matter from smear slides

Three different types of organic matter were distinguished from bulk sediment smear slides. These included marine organic matter, sewage organic matter and terrigenous organic matter. *Marine organic matter* (derived from primary production) consisted of light brownish, loosely packed, diffuse aggregates that contained minerogenic grains and opaline skeletons (diatoms and silicoflagellates). This fraction was dominant in all smear slides. *Sewage organic matter* was made up of light, colourless to yellowish, diffuse particles. This organic material was partly very similar to the diffuse marine organic matter aggregates. Therefore, additional smear slides from pure sewage samples were analysed for comparison. A significant part of the organic matter fraction ($> 5\%$) was sewage derived across the upper few centimetres of the surface sediments. The proportion of colourless diffuse particles decreased with depth. Dark brownish to blackish particles with well-confined shapes like terrestrial pollen and spores were classified as *terrigenous organic matter*. The content of terrigenous organic matter was less than 5% in all samples.

LOI, C/N and $\delta^{13}\text{C}$

Sediment LOI (organic carbon) concentrations, C/N ratios and $\delta^{13}\text{C}$ (versus PDB) isotopic ratios of Station A

and B are shown in Fig. 2 and Table 2. The general trends are similar at both stations: LOI concentrations (%) decrease and LOI mass accumulation rates ($\text{gC/m}^2/\text{y}$) increase across the upper 5 cm of the sediment. C/N ratios increase and $\delta^{13}\text{C}$ isotopic ratios decrease from 15 to 5 cm and no clear trend is observed across the 0–5 cm of the sediment surface.

The bulk plankton samples had an organic matter composition of $\text{C/N} = 7.9$ and $\delta^{13}\text{C} = -20.1 \pm 0.5\text{‰}$. They consisted mainly of diatoms, dominated by *Skeletonema costatum* and *Chaetoceros sp.*, some dinoflagellates and a few aggregates that contained minerogenic, clay-sized, grains. These samples represented the end-

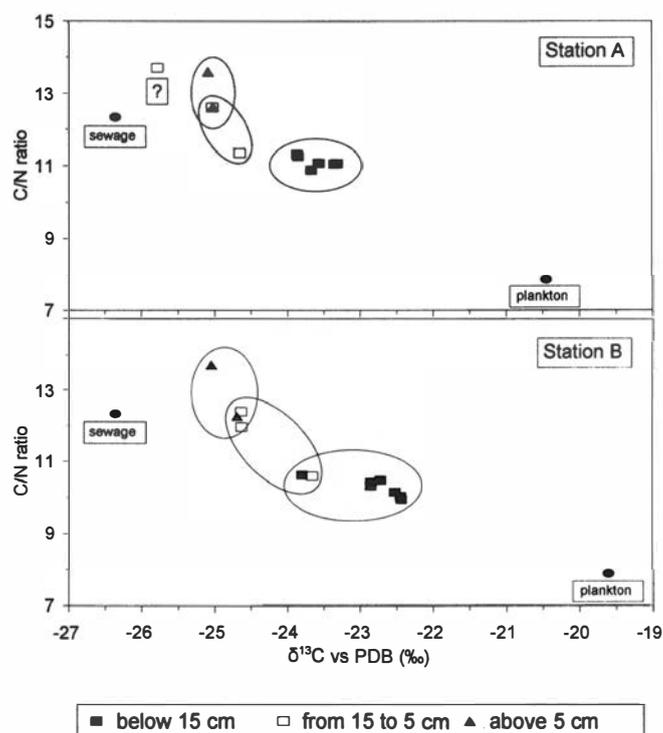


Fig. 3. C/N versus $\delta^{13}\text{C}$ of Station A (upper) and Station B (lower). Area of filled squares: Below 15 cm. Area of open squares: From 15 to 5 cm. Area of black triangles: Above 5 cm. Black dots mark bulk sewage and bulk plankton composition. Question-mark indicates erroneous high C/N and low $\delta^{13}\text{C}$ at 5 cm of Station A.

Fig. 2. Zn (ppm, $\text{g/m}^2/\text{y}$), LOI (%), $\text{gC/m}^2/\text{y}$, grain size (%), C/N ratio and $\delta^{13}\text{C}$ (versus PDB) isotopic ratio (‰) versus depth of Station A (upper) and Station B (lower). Arrow indicates the position of the maximum ^{137}Cs fallout peak of 1963.

member composition of marine organic matter from primary production of the photic zone. The bulk sewage composition was $C/N = 12.3$ and $\delta^{13}C = -26.4\text{‰}$, representing the sewage-derived end-member of the organic matter fraction. The C/N ratios and $\delta^{13}C$ isotopic ratios of both stations were plotted against each other and compared to the composition of the bulk plankton and the bulk sewage samples. Fig. 3 shows a continuous development from a more plankton-like organic matter composition below 15 cm to a more sewage-like organic matter composition from 15 to 5 and above 5 cm. No explanation has been found for the erroneously high C/N (13.7) and low $\delta^{13}C$ (-25.8‰) at 5 cm of Station A.

LOI mass accumulation rates were determined above 15 cm based on the dating of Paetzel et al. (1994). They were calculated to be $12 \pm 1 \text{ gC/m}^2/\text{y}$ in sediments deposited from 15 to 5 cm, they increased to $25 \pm 2 \text{ gC/m}^2/\text{y}$ above 5 cm. Similarly, the trace metal content, here represented by Zn, was enriched in the surface sediments. From 15 to 5 cm: $Zn = 88 \pm 24 \text{ ppm}$ and $8 \pm 3 \text{ mg/m}^2/\text{y}$. Above 5 cm: $Zn = 660 \pm 150 \text{ ppm}$ and $80 \pm 23 \text{ mg/m}^2/\text{y}$ (Fig. 2, Table 2). This exponential increase started around 1969.

Siliceous micro-organisms

All species, including *Chaetoceros sp.*, *Skeletonema costatum*, *Tabellaria flocculosa*, *Thalassiosira nordenskiöldii* and the silicoflagellate *Distephanus speculum* were well preserved across the whole sediment column. Preservation of the finely silicified diatom *Skeletonema costatum* indicated that the sedimentary signal of the micro-organisms was unaltered by silica dissolution. The siliceous organism composition was diverse and similar in all samples including both, marine and freshwater species. Above 5 cm, *Skeletonema costatum* was more abundant compared to sediment samples taken below 5 cm sediment depth. This higher abundance might be attributed to variations in nutrient composition or eutrophication within the photic zone.

Discussion

Decomposition of the organic matter fraction

Organic matter decomposes through microbial reduction of NO_3^- and NO_2^- into nitrogen and of SO_4^{2-} into sulphur in anoxic sediments (Price 1976; Berner 1980; Morse et al. 1987). This down-core decomposition of organic matter is usually reflected by a decrease in $\delta^{13}C$ (Emerson & Hedges 1988). Bacteria utilize preferentially ^{12}C during the fermentation of methanol into methane. This process enriches the organic carbon fraction in ^{13}C and $\delta^{13}C$ values will thus decrease with depth (Rosenfeld & Silverman 1959).

A downcore increase in C/N ratios, due to the production of nitrogen gas, is often used as an indication for the

anaerobic decomposition of organic matter in the sediment (Manheim 1976; Summerhayes 1983). C/N ratios decrease as a result of the release of CO_2 during sulphate reduction and methane fermentation in sediments. The influence of methane fermentation on the $\delta^{13}C$ and C/N composition is known from sediments deposited over several thousand years (Kvenvolden & Kastner 1990; Kvenvolden et al. 1990).

In what follows the question of whether organic matter decomposition could be the reason for the changes in LOI, $\delta^{13}C$ isotopic ratios and C/N ratios in the Nordåsvannet will be discussed.

Above 5 cm. – The composition of C/N and $\delta^{13}C$ was similar to that of the bulk sewage sample. The variation in C/N ratios and $\delta^{13}C$ may have been caused by variations in the composition of the sewage-derived organic matter. The depth resolution of this section was not high enough to indicate decompositional trends. LOI concentrations showed a clear minimum across this interval. This decrease coincides with larger grain-size supply due to increased sewage discharge. *From 5 to 15 cm:* LOI concentrations and C/N ratios decreased, $\delta^{13}C$ isotopic ratios increased down-core across this interval, changing from a sewage-like to a plankton-like organic matter composition. *Below 15 cm:* The organic carbon fraction revealed C/N ratios and $\delta^{13}C$ isotopic ratios that were more influenced by plankton-derived organic matter. The organic carbon composition and the LOI concentrations showed no or little variation. Either decomposition of the deeper sediments had reached a steady state, or the organic carbon fraction of the sediments originated mainly from primary production.

Sedimentary decomposition of organic matter should thus result in a down-core decrease of $\delta^{13}C$, and a down-core increase of C/N ratios. The opposite was found in the Nordåsvannet sediments. The $\delta^{13}C$ isotopic ratio increased and C/N ratios decreased down-core. Thus, changes in composition and sources of the organic matter mainly account for the sedimentary $\delta^{13}C$ and C/N ratio variations. The decrease in LOI (organic carbon) concentrations from 5 to 15 cm may be due to both anaerobic organic matter decomposition and increased organic matter supply. Larger minerogenic grain sizes dilute the LOI concentrations above 5 cm.

The origin of the organic matter fraction

Terrestrial or marine sources of organic matter can be distinguished by using C/N ratios, i.e. the different weight proportions of total organic carbon to total organic nitrogen in this material. Protein-rich marine phytoplankton has a C/N ratio of 6.6, with $C = 106$ and $N = 16$ (Redfield ratio, Redfield et al. 1963). The C/N ratio can vary between 3 and 9 in sedimentary marine organic matter (Bordovskiy 1965a, 1965b; Price 1976; Müller 1977; Cifuentes et al. 1988; Stein 1991; Hamilton & Lewis 1992). Terrigenous organic matter has a much

wider range of C/N ratios. Typical northern hemisphere sediment values of humid regions range from 12 to 35 (Cato 1990; Stein 1991; Tan et al. 1991).

The $\delta^{13}\text{C}$ method is often used to detect variations in the carbon isotope fractionation of land and marine-derived organic matter (Bordovskiy 1965a, 1965b; Price 1976; Stein 1991). Carbon isotopic compositions of < -22 to -32‰ (versus PDB) in sediments indicate a terrestrial origin of the organic material (Sackett 1964; Deines 1980; Waples 1981; Stein 1991). The sedimentary marine organic matter fraction has $\delta^{13}\text{C}$ isotopic ratios of > -22 to -10‰ , generally ranging between -21 and -19‰ (Brown et al. 1972; Fontugne & Duplessy 1981; Waples 1981; Cifuentes et al. 1988; Stein 1991).

C/N ratios, $\delta^{13}\text{C}$ isotopic ratios and smear slide analyses suggest two major sources of the organic matter fraction in the Nordåsvannet sediments:

Primary production. – Wassmann et al. (1986) described the Nordåsvannet as a natural eutrophic environment. They estimated primary production to be about $190 \text{ gC/m}^2/\text{y}$ with light as the limiting factor. The sedimentary record of the siliceous microfossils suggests that only minor variations in primary production have occurred during this century in the Nordåsvannet. We thus assume that the average primary production rate of $190 \text{ gC/m}^2/\text{y}$ (Wassmann et al. 1986) corresponds to the average primary production over this time span.

Sewage discharge. – The sewage discharge to the Nordåsvannet surface waters increased gradually from around 1700 until today. Deep water sewage discharge started in 1969 and lasted until 1988. We attribute the changes in the sedimentary organic matter composition during the last 250 years to the variations in the non-constant source, i.e. the sewage discharge, and relate them to the sewage discharge history.

Below 15 cm. – Consistent $\delta^{13}\text{C}$, C/N ratios and LOI concentrations correspond to the situation before 1700 when no significant amount of sewage was released into the Nordåsvannet. Smear slides indicate that less than 5% of terrigenous organic matter was incorporated into the sediment and that no changes occurred across the upper 20 cm of the cores. The C/N and $\delta^{13}\text{C}$ composition was plankton-like. The source of the organic matter was primary production during that time.

From 15 to 5 cm. – The gradual increase in sewage supply from 1700 to 1969 may account for the gradual increase in C/N and decrease in $\delta^{13}\text{C}$. Although C/N and $\delta^{13}\text{C}$ indicate a shift in the organic matter sources, accumulation of the sedimentary organic carbon (LOI in $\text{g/m}^2/\text{y}$) did not change significantly before 1969.

Above 5 cm. – The strong variation in C/N ratios, $\delta^{13}\text{C}$ and LOI concentrations coincide with the change from surface to deep water sewage release that started in 1969.

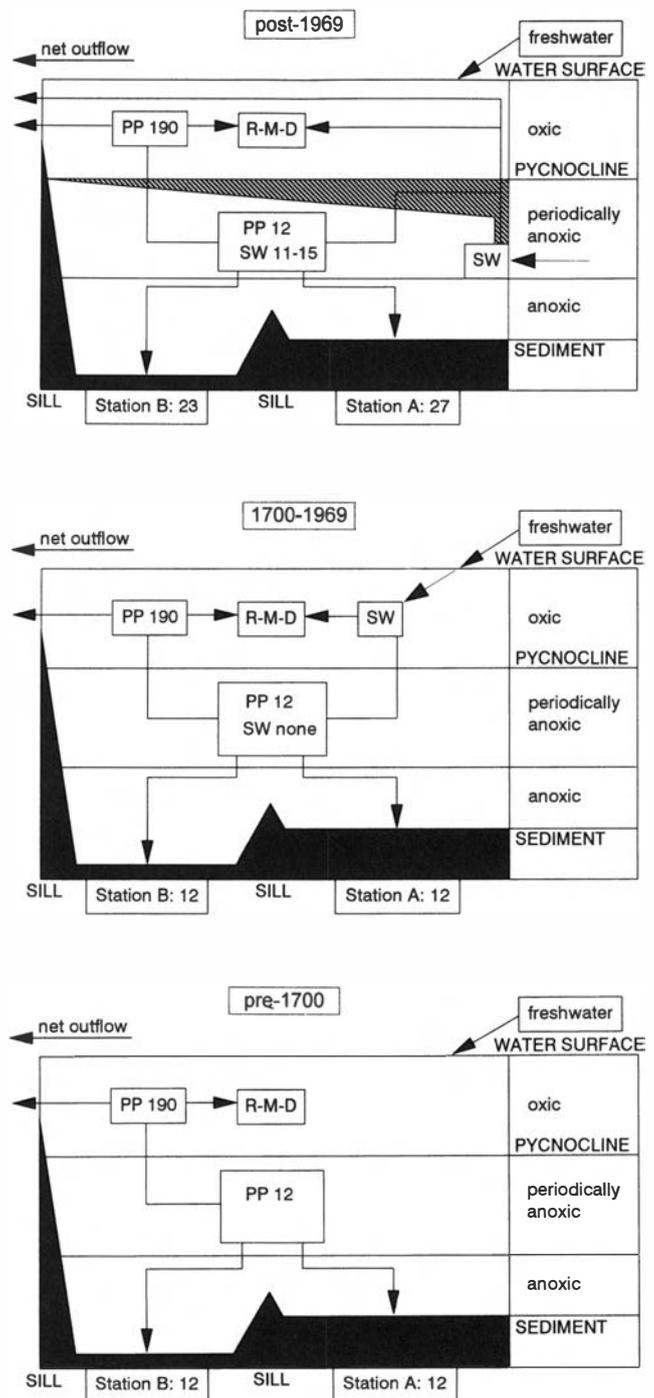


Fig. 4. Spatial and temporal (last 250 y) organic carbon deposition in the Nordåsvannet. PP = primary production. SW = sewage. R = respiration. M = mineralization. D = dissolution. Numbers without units are organic carbon in $\text{gC/m}^2/\text{y}$. Pre-1700: PP = 190, SW = no discharge, deposition = $12 \text{ gC/m}^2/\text{y}$. From 1700–1969: PP = 190, SW = 0 (surface water sewage discharge with only minor vertical transport), deposition = $12 \text{ gC/m}^2/\text{y}$. Post-1969: PP = 190, SW = 11–15 (deep water sewage discharge), deposition near SW source = 27, deposition distant to SW source = $23 \text{ gC/m}^2/\text{y}$.

The C/N and $\delta^{13}\text{C}$ composition is sewage-like. The increase in the organic carbon accumulation (LOI in $\text{g/m}^2/\text{y}$) is mainly attributed to this deep water sewage discharge (see model below).

In the Nordåsvannet, marine organic matter sources were distinguished from sewage derived organic matter

sources by using a combination of the C/N ratio method, the $\delta^{13}\text{C}$ method and smear slide analysis. The sedimentary organic carbon composition indicates a gradually increasing influence of sewage on the sediments from 1700 to 1969. The sudden increase in sewage-derived organic matter after 1969 coincides with the shift from surface water sewage discharge to deeper water sewage discharge. No changes in the terrestrial input of organic matter could be found.

The organic carbon (LOI) supply model

We propose the following model to describe the organic carbon supply (LOI in $\text{gC/m}^2/\text{y}$) to the Nordåsvannet bottom sediments (Fig. 4). According to the results of this investigation and those of Wassmann et al. (1986), primary production (PP) is assumed to have been constant ($190 \text{ gC/m}^2/\text{y}$) over the three scenarios presented in this model: (1) pre-1700; (2) from 1700 to 1969, (3) post-1969. An accumulation rate of $12 \text{ gC/m}^2/\text{y}$ responds to 6% deposition of organic carbon derived from primary production during the time before 1700 to 1969. After 1969, organic carbon accumulation increases to $23\text{--}27 \text{ gC/m}^2/\text{y}$. Subtraction of the sedimented primary produced organic carbon ($12 \text{ gC/m}^2/\text{y}$) from the total sedimentary organic carbon ($23\text{--}27 \text{ gC/m}^2/\text{y}$) indicates a sewage-derived organic carbon accumulation of $11\text{--}15 \text{ gC/m}^2/\text{y}$. Wassmann et al. (1986) postulated a sewage-derived organic carbon supply of $35 \text{ gC/m}^2/\text{y}$ to the Nordåsvannet. This would indicate that about 40% of the sewage-derived organic carbon became incorporated into the sediments during the period of deep water discharge.

Pre-1700. – Primary production was $190 \text{ gC/m}^2/\text{y}$, of which $> 90\%$ was respired (R), mineralized (M) or dissolved (D) above the pycnocline and transported out of the fjord with the strong tidal current. A salinity and/or temperature induced pycnocline exists across the upper 10 to 20 m of the fjord through most of the year (Glennie & Simensen 1963; Gade 1993, *pers. comm.*).

From 1700 to 1969. – Primary production and transport mechanisms of the organic carbon are assumed to have been similar to those suggested in the pre-1700 scenario. Sewage was additionally discharged to the surface waters. No changes occurred in the sedimentary organic carbon accumulation. Most of the surplus organic matter was dissolved, utilized by micro-organisms and/or transported out of the fjord together with the primary produced material. Only a minor fraction of the sewage was incorporated into the sediments.

Post-1969. – Primary production and transport mechanisms of the organic matter are still assumed to be unchanged. The sewage was discharged to the deeper water of the fjord and floated upward as a plume to the

depth of the pycnocline (Ravndal 1971). Part of the sewage-derived organic matter sank down to the fjord bottom ($11\text{--}15 \text{ gC/m}^2/\text{y}$) and part of this fraction was exported, dissolved and/or utilized. The organic carbon accumulation was higher near the Fjøsanger deep sewage outlet (Station A: $27 \text{ gC/m}^2/\text{y}$) than at greater distance (Station B: $23 \text{ gC/m}^2/\text{y}$), indicating a gradient from high organic carbon accumulation near the source and low organic carbon accumulation distant to the source.

Conclusions

1. The organic matter fraction in recently deposited (0–200 years) Nordåsvannet sediments originates from two major sources, marine, i.e. from primary production, and sewage, as determined by smear slide analysis, $\delta^{13}\text{C}$ and C/N ratios.
2. Accumulation of sedimentary organic matter increased by a factor of two after 1969 although no major changes in primary production or total sewage discharge have been recorded in the Nordåsvannet during the last 50–100 years.
3. The change in organic matter accumulation coincides with and can be attributed to a shift from surface water sewage discharge to deep water sewage discharge that occurred in 1969.
4. Our results point to the significance of stratified water masses on the sedimentation of sewage-derived organic matter and associated contaminants in the Nordåsvannet fjord system.

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