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*Published in:*
Netherlands journal of geosciences-Geologie en mijnbouw

Netherlands Journal of Geosciences / Geologie en Mijnbouw 82 (2): 133-147 (2003)

# The origin of brackish and saline groundwater in the coastal area of the Netherlands

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Received: August 2002; accepted: March 2003

## Abstract

An explanation is presented for the origin of brackish to saline groundwater in the coastal area of the Netherlands based on geological, chemical (chlorinity), isotopic and geophysical data. A critical review of all possible salinization mechanisms shows that the origin of the brackish water is related to former transgressions. Both the vertical salinity distribution and the carbon-14 activity of the groundwater indicate that connate sea water from the Pliocene to Early Pleistocene is not the source of the brackish to saline waters in the overlying Pleistocene fluvial aquifers. Instead, it derives from Holocene transgressions. The salinization mechanism is discussed in relation to the paleogeographical development during the Holocene and the occurrence of low-permeability strata. Finally, freshening of the aquifers following retreat of the sea is briefly considered.

*Keywords:* carbon-14, coastal area, groundwater, Holocene, Netherlands, salinity

## Introduction

Coastal areas are attractive habitats for man for their favorable natural conditions (e.g. fertile soil, availability of food and water) and strategic position. They are among the most densely populated areas in the world. Often, these areas suffer from water management problems related to the occurrence of saline groundwater. This water derives from sources such as sea water intrusion due to overexploitation, former inundations, evaporation or dissolution of salt deposits. Knowledge of the origin of the saline water and its dynamics is a prerequisite for effective management of the available water resources.

A very complex salinity distribution is encountered in the coastal area of the Netherlands that is not in equilibrium with present-day hydrological boundary conditions (Oude Essink, 1996). As will be elaborated in this paper, the origin of the saline water is related to the geological history of the area that included multiple transgressions during the past 2 My. Understanding the genesis of the salinity distribution requires knowledge of the hydrological processes (e.g. diffusion, compaction-driven flow, free and forced convection) that control the salinity distribution on a geological time scale (> 1 kA). Identification of these processes is complicated because the present-day groundwater composition has been strongly affected by man-made changes during the recent past.

In the Dutch coastal area, a wealth of observations is available that are scattered across many different archives and reports. Various authors have addressed the origin of the brackish to saline ground water (e.g. Volker, 1961; De Vries, 1981; Beekman, 1991, Gieske, 1991; Meinardi, 1991; Stuyfzand, 1993), but few publications integrate data from these various sources. In this paper, a conceptual model of the genesis of the current salinity distribution will be presented based on a comprehensive collection of geological, chemical, isotopic and geophysical data. The data will be presented in the form of maps and charts and the possible sources of salt will be treated in detail. From the analysis, conclusions will be drawn on the origin of the saline water and the relevant hydrological processes will be identified.

## Physical setting

The coastal area of the western Netherlands forms a part of the south-eastern North Sea coastal plain that extends more or less continuously from northern Denmark to the north of France. The morphology generally consists of a polder landscape with an elevation near or below sea level, bordered on the western side by a coastal dune belt and on the eastern side by a fresh water lake (IJsselmeer), the more elevated terrain of ice-pushed ridges and the Brabant Massif and recent river deposits of Rhine and Meuse (Fig. 1).

### *Geological history*

The North Sea covered most of the Netherlands during the Early Pliocene (Fig. 2) but both the coastline and the depocenters were shifting toward the north-west at that time (Zagwijn, 1989). This shift continued into the Early Pleistocene and by 1.6 Ma BP. marine influences had disappeared from the present coastal area. The deltas of large rivers draining the Baltic region (the predecessors of the rivers Elbe, Weser and Ems, referred to as eastern rivers) and of the rivers Rhine and Meuse, merged to form one large delta complex. The progradation of the large delta continued until the early part of the Middle Pleistocene. The regression reached a maximum in the period between 900 and 450 kA BP., when the coastline was probably in the vicinity of the Dogger Bank (Zagwijn, 1989). It was not until the Holsteinian interglacial that the sea invaded the western part of the Netherlands again. Marine sediments from this period are found locally in deep valley systems that had formed during the Elsterian glacial stage (~ 300 kA BP.). During the following Saalian glacial (180 kA – 130 kA BP.), ice sheets covered approximately half of the Netherlands (Zagwijn, 1974), which had a profound impact on the morphology of the landscape. The ice formed subglacial basins of up to 100 meters deep and ice-pushed ridges with heights of more than 100 meters (De Gans et al., 1986). Sea level rose during the Eemian (130 kA – 110 kA BP.) and the sea penetrated the former glacial basins and deep river valleys (Zagwijn, 1983), resulting in widespread marine conditions in the western part of the Netherlands

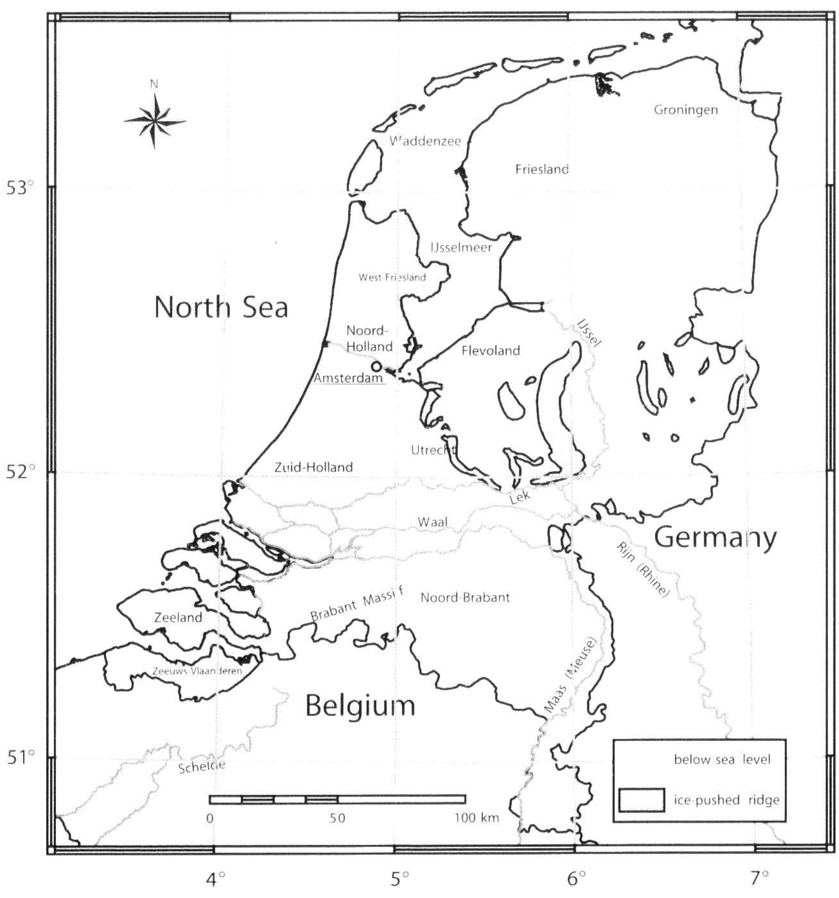


Fig. 1. Map of the Netherlands showing the area below mean sea level (shaded), ice-pushed ridges (hatched) and geographical names referred to in the text.


Fig. 2. Map showing the depth of the base of the Quaternary (Zagwijn & Doppert, 1978) and landward extent of the North Sea during the Pliocene and three Quaternary transgressions (Zagwijn, 1974; Zagwijn & Doppert, 1978; Zagwijn, 1986).

(Fig. 2). During the Weichselian glacial (110 kA – 10 kA BP.), periglacial conditions prevailed and the Saalian relief was further leveled through the deposition of so-called drift sands (fine-grained aeolian deposits) and strong erosion of elevated terrain (Zagwijn, 1974). The rivers Rhine and Meuse eroded two extensive east-west running valleys (in the present floodplain of the Rhine and in the IJsselmeer/West-Friesland area) that were separated by an area with little relief (De Gans & Van Gijssel, 1996).

These valleys were the first areas that were inundated when the North Sea reached the present coastline approximately 7.5 kA BP. Groundwater levels increased as a result of the sea level rise and due to the poor drainage of the relatively flat area, widespread peat formation occurred (Basal peat). Sea level continued to rise (Calais transgression: 8 – 3.8 kA BP.) and the sea flooded large parts of the peat areas (Fig. 2). Around 5 kA BP. barrier islands had formed at almost the same position as the present coastline, inland of which sand and clay were deposited in a lagoonal and tidal flat area. Several tidal inlets dissected the barrier islands (Beets et al., 1992) and Basal peat was eroded here. Marine influence gradually diminished inland of the barrier islands upon closure of the tidal inlets since 5.5 kA BP. (Beets et al., 1992) and once again peat growth occurred (Holland peat). Large peat bogs with an elevation of up to 5 meters above the present sea level developed (Pons, 1992). A fresh water lake existed in the IJsselmeer area between about 3 to 1 kA BP. (Zagwijn, 1986). Although the sea level rise had decreased to about 0.05 m/century around 2 kA BP. (Beets et al., 1992), large peat areas in the south-western and northern part of the coastal area were invaded and eroded by the sea (Duinkerke transgression: 3.5 kA BP. and later, Fig. 2). Dunes developed on top of the barrier islands from about 1 kA BP. An inland-sea (Zuiderzee) formed in the IJsselmeer area around 0.7 kA BP. following strong erosion of the northern peat area.

Since Roman times, man has dramatically changed the natural landscape by (amongst others) draining peat areas, reclaiming lakes and by building dikes. Drainage of peat areas started since Roman times and resulted in land subsidence owing to compaction and decomposition of peat. Vast areas of peat disappeared by erosion during floods and excavation for fuel, as a result of which many large lakes came into being. Several of these lakes have been reclaimed since the Middle Ages. Land reclamation also occurred on tidal flats in the south-western and northern part of the coastal area, where dikes were built to reclaim these areas from the sea. Dikes were also necessary to prevent rivers from flooding. Due to compaction, decomposition, erosion and sea level rise the elevation of the polders is currently below sea level. This way the characteristic polder landscape of the Dutch coastal area was created.

*Stratigraphy*

The study area is part of the subsiding North Sea basin that is filled with unconsolidated Cenozoic sediments, varying in thickness from 400 to over a 1000 meters, overlying Mesozoic strata (Zagwijn, 1989). The upper part of the Tertiary deposits consists of marine sand, sandy clay and clay (Zagwijn & Van Staalduinen, 1975). The depth of the base of the Quaternary sediments is shown in Fig. 2. The oldest Pleistocene layers consist of fine-grained marine deposits that reach a thickness of up to 200 meters (Zagwijn & Van Staalduinen, 1975). A thick sequence of continental deposits is overlying these marine sediments. In the northern part of the area, these consist of coarse-grained fluvial deposits of the eastern rivers and the river Rhine. The total thickness of these deposits locally exceeds 200 meters. In the southern part, the sequence has a maximum thickness of approximately 100 meters and consists of fluvial sands and clays of the rivers Rhine and Meuse. In both the north and the south, the upper part of the Pleistocene sequence consists of Eemian marine sediments, as well as aeolian and fluvial sediments from the Weichselian. Their total thickness generally does not exceed 20 to 30 meters. Peat, lagoonal- and marine clay together with fine-grained sand from the Holocene are found at the surface throughout most of the study area. Their average thickness amounts to 20 m although it increases from a few meters in the eastern part of the area to over 50 meters in former tidal inlets.

*Hydrogeology*

Significant lateral variations in lithology prohibit a simple subdivision of the subsurface into aquifers and aquicludes that is valid for the entire area (Dufour, 2000). In general, six aquifers are discerned which are shown schematically in a south-north cross section in Fig. 3. Locally, sub-aquifers are discerned where clay layers of significant thickness and extent occur (Van Rees Vellinga et al., 1981).

In the largest part of the area, the base of the aquifer system is formed by the clays of the Pliocene and locally, clays of the Early Pleistocene marine deposits (Pomper, 1983). In the province of Zeeland, the Tertiary marine sediments show a sandy facies and constitute an aquifer. Here, Oligocene clays form the low-permeability base.

Fine-grained layers that form more or less continuous aquitards in the south (for example, Lower Pleistocene clay layers), are absent in the northern part of the area. Conversely, confining units such as Saalian glacial deposits or Eemian marine clays are only present in the northern part of the area. It should be realized that all aquitards are very inhomogeneous both in their spatial distribution and hydraulic properties and many interconnections between the aquifers are present.

Groundwater levels are artificially controlled by a dense network of ditches and canals. Water levels in the polders are maintained by pumping excess rain and seepage water into canals and rivers that eventually drain to the North Sea. Drainage density is much smaller and groundwater levels are higher in the coastal dunes and the elevated Pleistocene terrain in the east. Groundwater flow patterns are mainly controlled by differences in surface water level between polders, lakes, canals and rivers, resulting in a complex system of local to regional groundwater flow systems (Fig. 4).

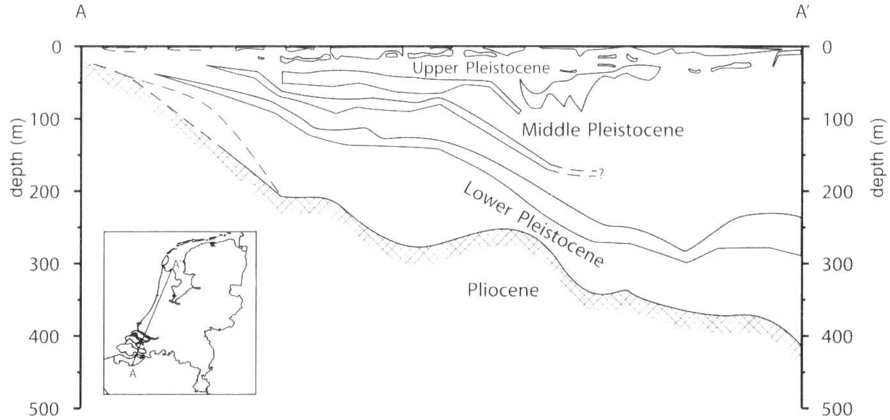


Fig. 3. Schematic diagram showing a generalized subdivision of the subsurface in aquifers and aquitards. The length of this section is roughly 200 km and depths (in meters) are approximate.

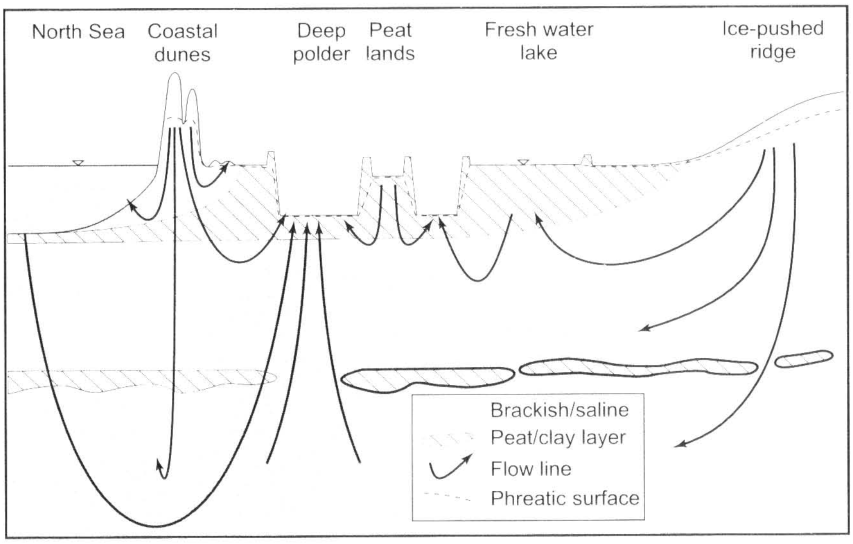


Fig. 4. Schematic representation of flow systems in the western part of the Netherlands (after Griffioen, 1994). The width of the cross-section measures approximately 60 km in reality.

The precipitation excess (precipitation – evapotranspiration) ranges from 200 to 350 mm/y (De Vries, 1974).

## Observations and methods

In the coastal area of the Netherlands, thousands of observation wells have been drilled and in many the water quality has been determined by chemical analyses. The abundance of the isotopes of the elements C, H and O have been measured in hundreds of water samples. Geo-electrical soundings have been carried out systematically to map the salinity distribution at a regional scale (Van Dam, 1976). Also, many resistivity logs from boreholes are available that provide information on the chloride concentration of the groundwater (Van Dongen & Boswinkel, 1982). In the present study, chloride concentrations in observation wells have been used to map the spatial salinity distribution, carbon-14 ($^{14}C$) was used to deduce the origin of water types and borehole resistivity logs were used to study salinity variations within clay layers.

Chloride analyses from water samples in observation wells were collected from the archives of TNO Netherlands Institute of Applied Geoscience (TNO-NITG) and the province of Noord-Holland. A three-dimensional version of Hardy's Multi Quadric Biharmonic interpolation method (Geenen, 1993; Van der Meij & Minnema, 1999) was employed to visualize the spatial distribution of chloride concentrations. Water samples analyzed prior to 1970 were excluded from the data set to avoid interference from temporal variations; if duplicate analyses were available, the most recent sample was included. 3868 chloride analyses were used to define the interpolation function that was used to calculate unknown chloride concentrations in a grid with a horizontal node spacing of 1 kilometer and a vertical node spacing of 10 meters.

Carbon-14 measurements of dissolved inorganic carbon (DIC) were retrieved from the archives of the Centre for Isotope Research in Groningen, The Netherlands and have been used in combination with chloride to assign a relative age to the water samples. Carbon-14 is subject to radioactive decay and can in theory be used to calculate the age of the groundwater according to:

$$T = -\frac{T_{1/2}}{\ln 2}\ln\frac{^{14}A_{sample}}{^{14}A_{nd}} = -8270\ln\frac{^{14}A_{sample}}{^{14}A_{nd}} \qquad (1)$$

where $T_{1/2}$ is the half life of $^{14}C$ (5730 y), $^{14}A_{sample}$ is the measured carbon-14 activity and $^{14}A_{nd}$ is the carbon-14 activity of the sample if no decay had occurred. The latter depends on both $^{14}A$ in the recharge area and the contribution of carbon sources and sinks in the aquifer to DIC (and thus $^{14}A$) of the sampled groundwater. Equation 1, however, could not be applied in this study as it is impossible to determine $^{14}A_{nd}$ because (1) $^{14}A$ of the recharge area cannot be determined, (2) carbon transfers are difficult to quantify, (3) $^{14}A$ of sources like carbonates, sediment organic matter and mixing waters are often unknown. Equation 1 has been used to assign a maximum (conventional) age to the water samples by setting $A_{nd}$ to 100 percent modern carbon (pmc). When groundwater has reacted with a carbon source that is younger than the water itself (for example Early-Pleistocene brackish water reacting with Holocene peat) equation 1 will underestimate the maximum age.

Information on the salinity of clay layers can solely be inferred from borehole resistivity logs as well screens have been installed in permeable layers only. For this purpose, a compilation of borehole measurements in the Netherlands by Van Dongen and Boswinkel (1982) was used. These authors used resistivity logs and water samples to reconstruct the chloride concentrations versus depth in 51 boreholes.

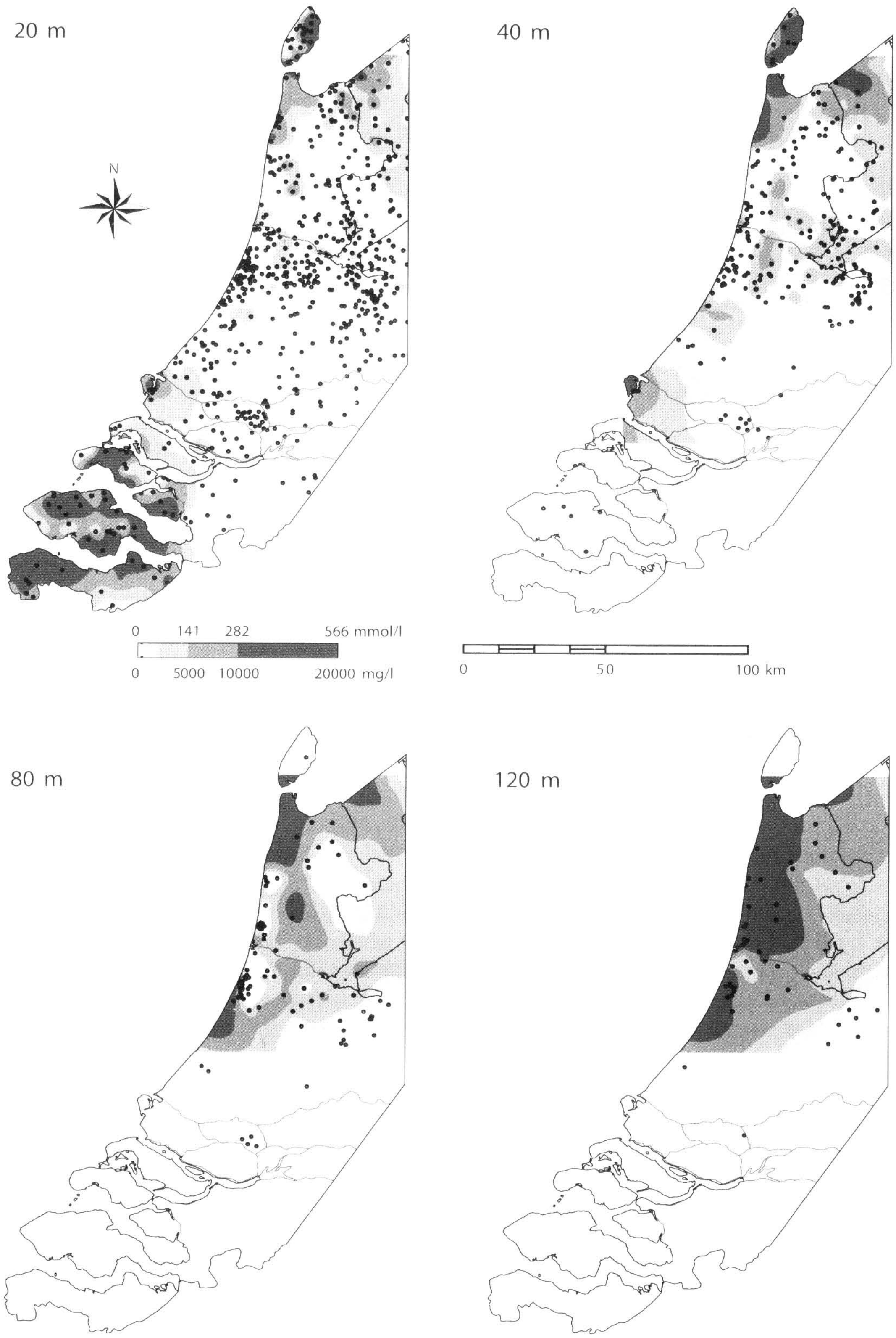


Fig. 5. Spatial distribution of chloride concentrations at 20, 40, 80 and 120 meters below sea level. Dots indicate the location of observation wells within 5 meters above or below the shown depth interval. No contours are shown in areas with too little data. Based on data from the province of Noord-Holland, Netherlands Institute of Applied Geoscience TNO - National Geological Survey and Ouwerkerk (1993).

In this paper, the term fresh is used to denote water with Cl < 8.4 mmol/l (300 mg/l), brackish refers to water with 4.2 < Cl < 282 mmol/l (150 – 10,000 mg/l) and saline signifies water with Cl > 282 mmol/l (10,000 mg/l).

## Results

The spatial distribution of chloride in groundwater is shown in Fig. 5. Since the result of the interpolation method is sensitive to variations in the density of data points, these maps serve only to indicate regional trends in chlorinity.

Actual measurements are plotted as an indication of the reliability. The maps show that major occurrences of brackish to saline water at shallow (< 20 m below the surface) depth are found in the province of Zeeland and the south-western part of the province of Zuid-Holland, the northern part of the province of Noord-Holland and the IJsselmeer area. Furthermore, upconing of brackish water is observed beneath deep polder areas in the provinces of Noord-Holland and Zuid-Holland. The most landward extension of brackish groundwater largely coincides with the landward extension of the Holocene transgressions (compare figures 2 and 5).

Fresh water up to depths of 100 meters below sea level or more is found below the coastal dunes, below the ice-pushed ridges and in the area of West-Friesland. Other occurrences of fresh water are in the province of Utrecht and the eastern part of the province of Zuid-Holland and the very southern part of the Zeeuws-Vlaanderen area (not shown in Fig. 5). Locally, such as south of Amsterdam, individual pockets of fresh water occur up to 80 meters below the surface.

At depths greater than 100 meters a seaward gradient is present, with chloride concentrations decreasing from sea water values near the coastline towards lower values more inland. Chloride and resistivity profiles measured in boreholes that penetrate the Pliocene- to Early Pleistocene marine deposits, generally show an inversion (i.e., a decrease of the salinity with depth) that coincides with the occurrence of low-permeability clay layers in these strata (Fig. 6). The occurrence of these inversions is ubiquitous in the area that has been affected by the Holocene transgressions (Fig. 7, cf. Fig. 2).

Inversions are also found within the upper part of the Pleistocene sequence, where they are mainly related to the occurrence of Lower Pleistocene fluvial clays, Saalian glacial deposits or Eemian and Holocene marine clays. The more saline water is found above or in (for marine clays) these low-permeability layers, the fresher water below (Pomper, 1981; Gieske, 1991).

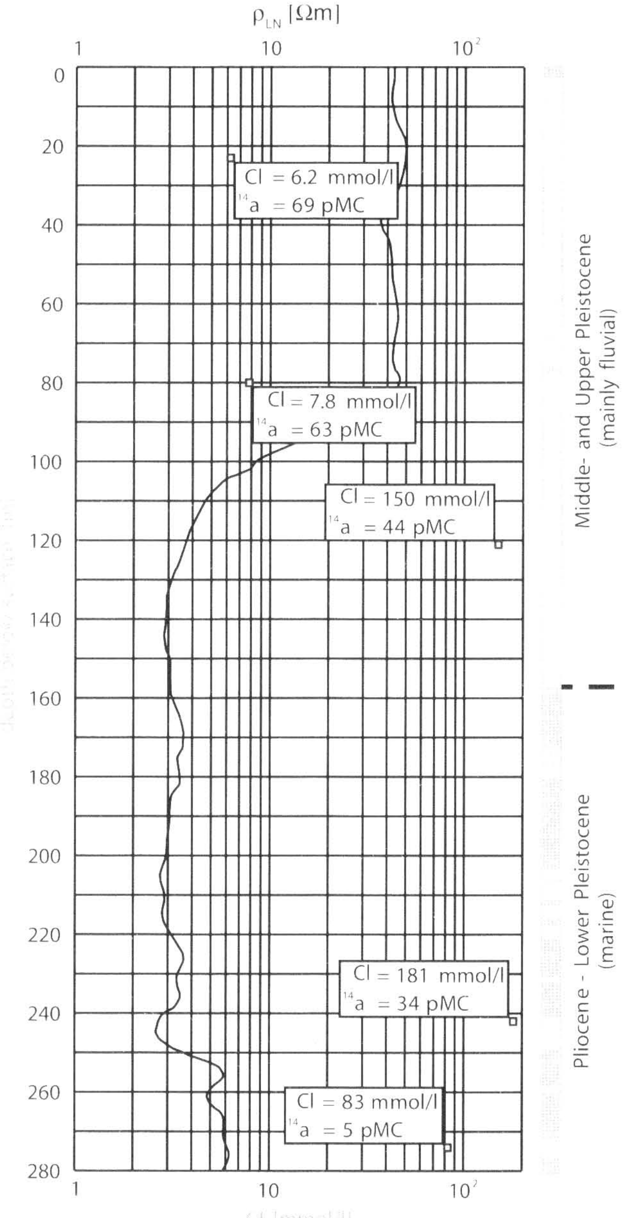


Fig. 6. Chloride concentration (mmol/l) and $\rho_{LN}$ vs. depth in well 31E-0176 (location shown in Fig. 7). Column on righthand side indicates sand (white) and clay layers (shaded). The top of the Early Pleistocene marine strata is found here at a depth of ~ 160 meters. The decrease of the Cl concentration below 245 m is very pronounced. Carbon-14 data by courtesy of R. Boekelman.

The cumulative frequency plot of $^{14}A$ (Fig. 8) of samples from the Pleistocene aquifers shows that of the samples with a chloride concentration > 10 mmol/l, approximately 80% has a conventional carbon-14 age < 10 kA. All but a few of these 48 samples are from the Pleistocene aquifers below the Holocene strata, so it seems unlikely that they represent old brackish water that has reacted with Holocene carbon sources. Therefore, the conventional age is considered

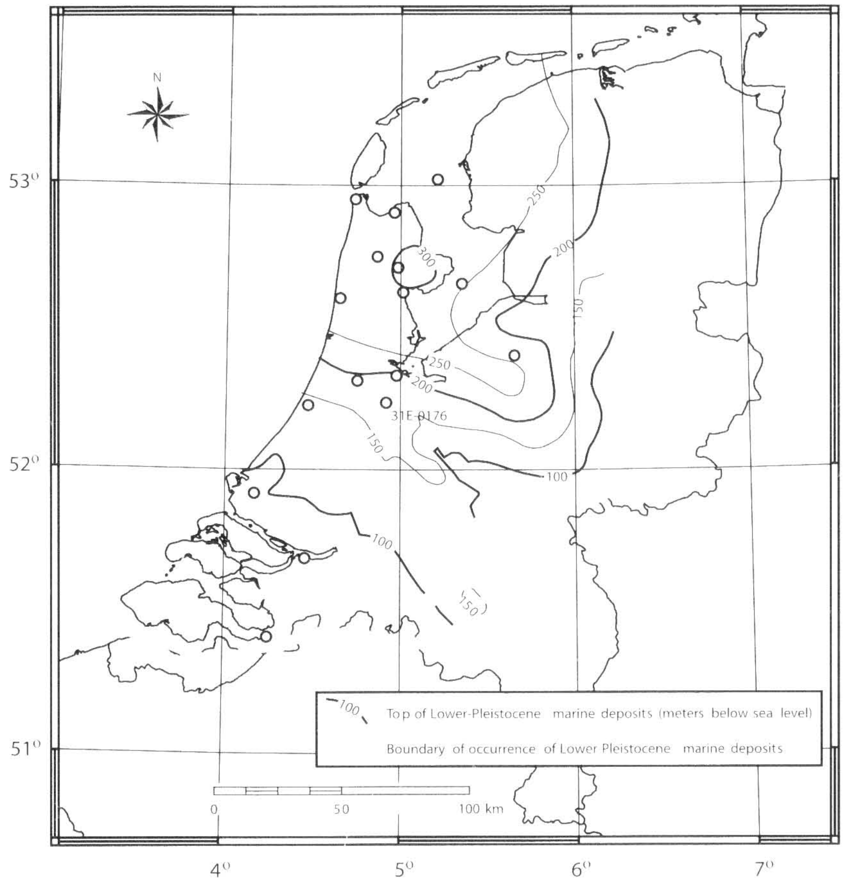


Fig. 7. Known occurrences of chloride inversions within Pliocene- to Early Pleistocene marine deposits, compiled from data by Meinardi (1974), Pomper (1981), Van Dongen & Boswinkel (1982), Van Wieringen & Willemsen (1982), Boswinkel & Ritsema (1984), Gieske (1991), Hobma (1993) and from the archives of the province of Noord-Holland. Contour lines represent the depth of the top of the Early-Pleistocene marine deposits below sea level (after Zagwijn & Van Staalduinen, 1975).

to represent the maximum age. The carbon-14 activities in borehole 31E-0176 (Fig. 6) show a sharp drop at the inversion in the Pliocene- to Early Pleistocene marine strata.

## Discussion

The spatial distribution of the chloride concentrations (Fig. 5) is not easily explained. The vicinity of the North Sea and former transgressions will have played an important role, but other factors that influence the chloride concentrations in groundwater cannot be ruled out a priori. Table 1 lists the sources and processes responsible for high-salinity waters in coastal areas in general (cf. Stuyfzand and Stuurman, 1994). Before discussing the role of transgressions and connate sea water, the relevance of the remaining entries from table 1 as a source of the saline waters in the Pleistocene aquifers will be addressed.

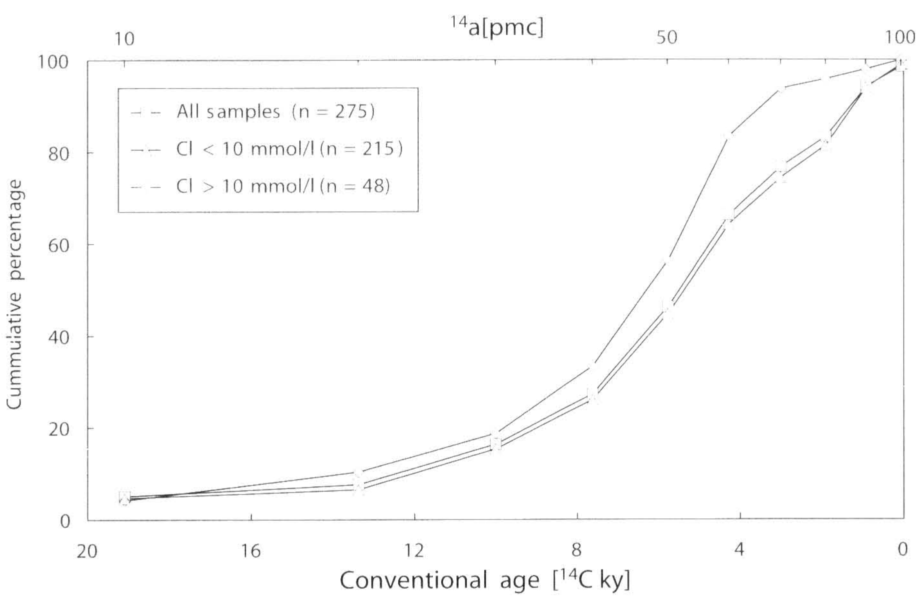


Fig. 8. Cumulative frequency distribution of the carbon-14 activities and corresponding conventional ages. The conventional $^{14}C$ age is an estimate of the maximum water age. It was calculated by setting $^{14}A_{initial}$ to 100 pmc, which represents atmospheric carbon-14 activity in 1950.

Table 1. Sources and processes responsible for high-salinity groundwaters in coastal areas and associated chlorinities.

| Source or process | Approximate chlorinity (mmol/l) | Remarks |
|---|---|---|
| modern sea water | 566 | Can be lower due to fluvial input |
| transgressions, floods | 566 | Can be lower due to fluvial input or mixing with fresh groundwater |
| connate sea water | 566 | Can be lower due to fluvial input or mixing with fresh groundwater |
| aerosols | 2 | Bulk precipitation along the coast (Stuyfzand, 1993) |
| salt formations | 2000 | Reported by Glasbergen (1981) |
| anthropogenic pollution (waste sites, road salt, salt disposal) | < 10 | |
| evapo(trans)piration | < 10 | evaporated rain water |
| hyperfiltration | < 100 | dependent on original composition |
| freezing | ? | dependent on original composition |

*Modern sea water*

Land subsidence and over-exploitation of groundwater resources results in subsurface encroachment of modern sea water in many coastal areas around the world, which is commonly referred to as sea water intrusion (De Breuck, 1991). For the western part of the Netherlands, basic hydrological calculations and hydrochemical data indicate that the inland presence of intruded contemporary North Sea water is limited to a region of about 2 to 6 km from the coastline (Stuyfzand, 1993). Therefore, modern sea water cannot be the source of the brackish/saline water that is found up to tens of kilometers inland.

*Aerosols*

The chloride concentration in bulk (dry + wet) precipitation near the coastline amounts to approximately 2 mmol/l and decreases exponentially inland to a background level of about 50 μmol/l (Stuyfzand, 1993). Of this value, 10 to 45% is due to dry deposition of chloride aerosols on the rain gauge (Ridder et al., 1984; Stuyfzand, 1993). The total amount of atmospheric deposition on land surfaces can be 10% higher than on bulk collectors if bare or scarcely vegetated, or 200 to 400% higher if covered by shrubs or trees (Stuyfzand, 1993). Chloride aerosols mainly derive from sea spray (Vermeulen, 1977). The low concentrations of chloride in precipitation that result from aerosols make them an unlikely source for concentrations > 10 mmol/l in groundwater.

*Evaporites*

Zechstein salt layers are found in the eastern and northeastern part of the coastal area and chloride concentrations of over 2 mol/l have been reported in Eocene aquifers at approximately 550 meters below the surface (Glasbergen, 1981). There is some evidence that shallow groundwater (within 100 meters below the surface) overlying salt domes (found at 200 meters below the surface) has also been affected by rock salt dissolution (Glasbergen & Mook, 1982; Post et al, in prep.). Rock salt does not occur in the western part of the Dutch coastal area and can be ruled out as a source of dissolved solids here.

*Anthropogenic pollution*

Little is know about the contribution of anthropogenic sources to the chloride concentration of groundwater in the western Netherlands. Locally, contributions can be expected from road salt (Werkgroep Midden West-Nederland, 1976), pollution by chlorinated solvents and waste disposal sites (Van Duijvenbooden & Kooper, 1981). An anthropogenic source that affects chloride concentrations on a regional scale is agriculture, where chloride is introduced to the environment through the application of fertilizers and manure. Studies in agricultural areas in the southern parts of the Netherlands, however, have shown that the highest chloride concentration of the groundwater beneath agricultural land is approximately 1.5 mmol/l (Broers & Griffioen, 1992). Agricultural activities in the coastal area are not more intensive than these investigated areas, so only a minor contribution to the total volume of salt groundwater is to be expected.

*Evapo-transpiration*

Strong evaporation sometimes leads to extreme salt concentrations in ground- and surface waters in arid areas. In the Netherlands, evapo-transpiration amounts on average to two-thirds of precipitation (Dufour, 2000), which will increase concentrations by a factor of 3. Locally, stronger evapo-transpiration occurs depending on the vegetation. For example, concentration factors as high as 5.8 have been found in the coastal dunes below pine trees (Stuyfzand, 1993). The highest chloride concentrations in recharge waters are in the order of 10 mmol/l in areas with high atmospheric deposition and strong evapotranspiration. Higher concentrations are to be attributed to other sources of chloride.

*Hyper-filtration*

Hyper-filtration (or reverse osmosis) occurs when groundwater flow due to a hydraulic head gradient occurs across a clay layer that acts as a semi-permeable membrane. Clay layers can act as natural membranes owing to their negative surface charge. Since they prevent the passage of charged solutes such as chloride ions, the concentration of groundwater on the inflow side of the clay layer will increase. Osmotic effects have been recognized in a harbour sludge depot directly overlying a brackish-water aquifer (Keijzer, 2000). To what extent this process plays a role in the coastal area of the Netherlands remains unclear. However, since the osmotic efficiency of natural membranes rapidly decreases with salinity, it is assumed that hyper-filtration is ineffective at sea water concentrations.

*Freezing*

It is known that freezing of groundwater causes migration of solutes and the role of this process has been recognized in the formation of saline groundwaters and brines (e.g. Bottomley et al., 1999; Bein & Arad, 1992; Herut et al., 1992). Permafrost conditions occurred in the coastal area during the glacial stages of the Pleistocene. Therefore, this mechanism must have influenced the salinity distribution at least to some extent, as was previously proposed by Pomper (1978). In the present study, there are no data that support the conclusion that freezing has contributed to the salinity of groundwater.

*Importance of transgressions*

From the foregoing discussion, it follows that salinization of the Pleistocene aquifers in the coastal area of the Netherlands must be due to either former transgressions or the presence of connate sea water in the subsoil. A distinction between the two is relevant here: salinization during a transgression refers to the introduction of solutes in a fresh water aquifer directly from the overlying or adjacent sea water, whereas salinization by connate sea water refers to the introduction of salts from marine sediments (deposited during a transgression) that still contain saline interstitial water.

The Netherlands has experienced several transgressions during the Cenozoic. The landward borders of the ones that are relevant here are depicted in Fig. 2. Marine sediments from the Pliocene to Early-Pleistocene, Eemian and Holocene transgressions still contain connate sea water (e.g. Van Dongen & Boswinkel, 1982; Van Rossum, 1998) and their potential contribution to the salinization of the Pleistocene aquifers will be discussed first. Then, salinization during the Holocene transgression will be elaborated in detail.

*Salinization from marine deposits*

The oldest marine deposits containing connate sea water that could act as a source of salts are the fine-grained sands and clays from the Pliocene and Early Pleistocene. Upward salt transport from these marine layers did undoubtedly cause salinization of the Pleistocene fluvial sands that originally contained fresh water. It is unlikely, however, that they constitute the source of the bulk of the brackish to saline water that is found in the aquifers today as suggested by Meinardi (1991), since measurements in boreholes and observation wells often show a chloride inversion near the boundary between Pliocene and Early Pleistocene marine strata and Pleistocene fluvial deposits. An example is given by well 31E-0176 (Fig. 6, see Fig. 7 for its location). If the connate sea water is the source of the saline water in the Pleistocene fluvial sediments an opposite gradient would be observed. The reverse is indicated: the marine strata have been freshened after deposition and they are currently salinized by salts from the overlying fluvial deposits. Similar findings were reported by Appelo & Geirnaert (1991), based on the geochemical characteristics of sediment samples. The large number of boreholes in which this inversion is observed and their distribution across the coastal area (Fig. 7) shows that this is a general phenomenon. This suggests that the occurrence of relic Pliocene and Early Pleistocene sea water in the Pleistocene fluvial aquifers is an exceptional condition.

Carbon-14 activities have been measured at 5 different depths in observation well 31E-0176 (data by courtesy of R. Boekelman). The deepest sample, which is from the Pliocene marine deposits, has a very low $^{14}A$ and represents connate, $^{14}$C-free sea water admixed with young (< 25 kA) water. It can not be excluded that this sample is actually $^{14}$C-free and that the measured activity is due to pollution by drilling fluid or shortcut flow in the well. The high $^{14}A$ of the samples at 121 and 242 meters (Fig. 6) points at a Holocene origin of the brackish water at these depths. The notion that the brackish water above the chloride inversion does not derive from the pore waters in the Early-Pleistocene to Pliocene marine deposits excludes the possibility that its carbon-14 signature is due to mixing of old sea water with Holocene fresh water. Since the chloride inversion at the base of the Pleistocene aquifer is a general phenomenon, high carbon-14 activities of brackish to saline samples from other parts of the coastal area are considered to be indicative for a Holocene origin. Samples with very little or no carbon-14 must date from either the Eemian transgression or the Pliocene to Early Pleistocene transgressions. Fig. 8 shows that 80 % of all water samples with a chloride concentration of at least 10 mmol/l have a carbon-14 activity higher than 30 pmc and thus date from the Holocene. Samples with an activity below 30 pmc possibly represent older waters but correction for geochemical reactions might well prove them younger than their conventional age.

Although its contribution to the present volume of saline water is small, large amounts of salt are likely to have entered the Pleistocene fluvial sediments over the past 1.6 My by compaction-driven flow (and also diffusion). Compaction-driven flow results from natural compression of the clastic sediments in response to sediment loading (Kooi & De Vries, 1998), leading to an upward flux of expelled pore water in the order of $10^{-4}$ m/y. The combined salt flux due to diffusion and compaction-driven flow is small compared to the average horizontal advective flux in the Pleistocene aquifers owing to topography-driven flow that is likely to have prevailed during the Pleistocene and hence most of this salt water has probably been removed.

There is little evidence of any large-scale contribution of connate water from Eemian marine deposits. The occurrence of Eemian brackish water has been documented in the northern part of the coastal area, probably related to stagnant flow conditions in the Pleistocene aquifer (Hoogendoorn, 1985). Although similar occurrences in the western part of the Netherlands cannot be ruled out, they are to be considered of local significance only as can be inferred from the fact that the majority of brackish water samples have a carbon-14 signature that points towards a Holocene origin. Following the Eemian transgression, a 100 ky period of fresh water circulation occurred during the Weichselian glacial stage. Despite uncertainties about the characteristics of the flow regime, such as reduced infiltration owing to permafrost (Bath et al., 1978), low precipitation rates (Zagwijn et al., 1992, cited in Van Weert et al., 1997) and increased groundwater velocities during glacial periods (Van Weert et al., 1997), significant flushing of the Eemian deposits is to be expected.

*Salinization during the Holocene transgression*

The North Sea gradually inundated the westward sloping Pleistocene surface as global sea level began to rise at the beginning of the Holocene. Brackish to saline water was carried on top of the fresh groundwater in the Pleistocene aquifers upon inundation. Owing to its higher density, saline water will sink into the fresh water, provided that the permeability of the underlying sediments is sufficiently high. In this process, referred to as free convection, a boundary layer develops as salts enter the aquifer by diffusion until it reaches a critical thickness at which it becomes unstable and breaks up into salt fingers that descend to the aquifer bottom. Fresh water is expelled by rising plumes. Whether or not free convection will occur is controlled by the non-dimensional aquifer Rayleigh number (Holzbecher, 1998):

$$Ra = \frac{\Delta\rho g k H}{\mu D_M} > 40 \qquad (2)$$

where $\Delta\rho$ is the density contrast ($ML^{-3}$), $g$ is gravitational acceleration ($LT^{-2}$), $k$ is intrinsic permeability ($L^2$), $H$ is the height of the aquifer, $\mu$ is dynamic viscosity ($ML^{-1}T^{-1}$) and $D_M$ is the molecular diffusion coefficient ($L^2T^{-1}$) including both tortuosity and porosity. Typical Rayleigh numbers for the sandy aquifers in the coastal area are in the order of $10^5$ – $10^6$. This indicates that salinization by free convection is likely to have occurred as these values greatly exceed the critical value of Ra ≈ 40. Associated vertical groundwater flow velocities are in the order of meters per year and the corresponding timespan for the salinization of an aquifer with a depth of about 200 meters is a few to tens of decades (Post & Kooi, 2003). These numbers hold for highly permeable aquifer sediments. Low-permeability strata will have greatly hindered free convection by (1) preventing the entry of saline water at the top of the aquifer system and (2) by obstructing the (vertical) passage of saline water as it moves through the aquifer. Both aspects will be discussed in more detail below.

Many studies have shown that clay layers can protect underlying fresh water aquifers from salinization for many thousands of years (e.g. Volker, 1961; Groen et al., 2000; Post et al., 2000) as salts, without water flow, need to pass primarily via diffusion. When the sea invaded the western part of the Netherlands some 7.5 kA BP., peat covered large parts of the sandy

Pleistocene surface, protecting the underlying aquifer. In tidal inlets, however, erosion removed the peat layers, so that the permeable aquifer sediments came in direct contact with the overlying sea water and became prone to rapid salinization by free convection. A complication arises from the fact that deposition of fine-grained material occurred on the "seafloor" during inundation, so that a hydraulic resistance was built up and the salinization process was greatly reduced. Furthermore, the salinity of the overlying water was lower during the initial stage of inundation than during later stages (Pons et al., 1963).

Gieske (1991) proposed that, in the southern part of the IJsselmeer area, salinization during the Calais transgression was followed by freshening during the formation of the Holland Peat and that clay and peat deposits from these periods protected the fresh groundwater from salinization during the subsequent Duinkerke transgression. This suggests that the main phase of salinization must have occurred during the early parts of the Calais transgression as during later stages free convection was hindered or even impeded due to the deposition or presence of clay layers and peat growth. In the northern part of the IJsselmeer area, however, marine conditions were absent until 1 kA BP. when the sea invaded the area and eroded the peat, resulting in favorable conditions for free convection. Strong erosion since the Middle Ages also removed large parts of the Holocene confining layers in the northwestern and south-western parts of the coastal area (Zagwijn, 1986). In the central part of the coastal area, widespread marine conditions have not occurred following the closure of tidal channels since 5.5 kA BP. (except for marine incursions along the inlets of the river Rhine and the Oer-IJ estuary), which means that in this area salinization mainly occurred during the Calais transgression.

Low-permeability layers have a retarding effect on salinization by free convection irrespective of their depth. Observations often show that chloride concentrations decrease significantly over clay layers (Pomper, 1981; Gieske, 1991) or that they contain relatively fresh water (Van Dongen & Boswinkel, 1982). At the same time vast amounts of Holocene brackish to saline waters are present below many other clay layers. This implies that (1) these layers are laterally discontinuous so that the salt water can flow around them and/or that (2) the permeability of these layers locally is large enough to permit the passage of salt water. Although the clay layers have retarded the salinization process, they apparently have not always been able to prevent the salt water from entering the underlying sediments. Numerical modeling of solute transport by diffusion and free convection should elucidate the precise effect these layers had on the salinization rate by free convection.

*Freshening of the aquifer system*

At present flushing of the saline groundwater takes place as fresh water infiltrates due to natural and man-made hydraulic gradients.

The first stage of freshening began with the closing of barriers and final sedimentation of back-barrier areas around 5.5 kA BP. Fresh water was introduced to the former marine areas by riverine input and precipitation (Zagwijn, 1986). Hydraulic gradients were small as only minor elevation differences existed in the coastal zone (e.g. former barrier islands and levees) and significant freshening of the subsurface is unlikely. Elevation differences of several meters were created upon formation of extensive peat bogs during the following 4 kA. Studies of modern undisturbed peat bogs have shown that vertical seepage losses to the underlying aquifer range between 10 and 30 mm/y (Van der Schaaf, 1998). Freshening of the aquifers up to tens of meters is therefore likely under favorable conditions. Significant freshening occurred below the coastal dunes that developed since the Middle Ages. Fresh groundwater is found here up to a depth of 120 meters.

The original peat bog topography has been almost completely transformed into a man-made topography (polder landscape). As elevation differences of several meters exist between individual polders and between polders and the nearby surface waters (lakes, rivers, drainage canals), groundwater flow systems developed within the coastal zone that extend up to tens of meters below the surface (e.g. Engelen & De Ruiter-Peltzer, 1986; Schot & Molenaar, 1992). Infiltration takes place from surface waters and elevated areas such as inverted tidal channels or peat remnants, thereby replacing the saline water that discharges in the deepest polder areas (Fig. 4). Lateral fresh water inflow is also from the coastal dunes (Stuyfzand, 1993) and the Pleistocene sand district (Schot & Molenaar, 1992). The presence of brackish water in Holocene marine deposits (e.g. Van Rossum, 1998) shows that recharge of the Pleistocene aquifer by precipitation mainly occurs via preferential flow paths in zones where the Holocene confining deposits are relatively permeable or thin or where they are dissected by tidal channels (De Louw et al., 2000).

## Conclusions

The salinity distribution of groundwater in the Pleistocene aquifers in the coastal area of the Netherlands shows a complicated pattern, especially in the upper 50 meters of the subsurface. The complexity of the observed pattern is mainly the result of Holocene marine influence as well as superimposed variations that are the result of local flow systems associated with man-made topography. At depths greater than 100 meters, groundwater with the salinity of sea water is found near the coastline and concentrations decrease landward. The origin of this water is linked to the Holocene transgression which can be concluded from: (1) indirect demonstration (the inability of other processes that increase groundwater salinity to explain the observed concentration patterns), (2) the occurrence of salinity inversions near the boundary between Pliocene and Early Pleistocene marine strata and Pleistocene fluvial deposits that exclude connate, Pliocene- to Early Pleistocene sea water as a source of saline water in the Pleistocene fluvial deposits and (3) carbon-14 analyses of groundwater that indicate that most water samples have a conventional age less than 10 kA.

Rayleigh numbers of the high-permeability Pleistocene sediments exceed their critical value by far. This suggests that salinization occurred mainly through free convection. Although especially Early Pleistocene and Saalian clay layers have protected the underlying aquifers from salinization, the lateral continuity and/or resistance to groundwater flow of most aquitards was insufficient to counteract the vertical intrusion of saline water.

Freshening of the aquifers occurred after retreat of the sea from the area. Except for the coastal dunes and the elevated Pleistocene outcrops in the eastern part of the area, natural hydraulic head gradients were relatively small compared to the gradients created by human activities. This means that, although freshening took place under natural conditions, the main phase of groundwater flow did not occur until man started to significantly alter the topography. Future developments are difficult to predict as infiltration of meteoric water through the Holocene confining deposits is along preferential flow paths, which complicates the quantification of fresh-water infiltration rates.

## Acknowledgements

We are greatly indebted to Lester Reiniers of the Province of Noord-Holland for generously providing the many chloride analyses, Harm-Jan Streurman and Bert Kers of the Centre for Isotope Research in Groningen for their help in digging up the carbon-14 data and to Reinder Boekelman of Delft University of Technology for providing the carbon-14 analyses of observation well 31E-0176. We would also like to thank the people whose critical comments have helped to improve the manuscript: Jasper Griffioen of Netherlands Institute of Applied Geoscience TNO – National Geological Survey and Victor Bense, Elmer van den Berg, Koos Groen, Henk Kooi and Co de Vries of the Vrije Universiteit Amsterdam.

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