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PAPER

History of the hydrogeochemical study of groundwater in the Netherlands and the research motives

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Abstract

An overview is presented of research on the hydrogeochemical aspects of groundwater resources in the Netherlands conducted since the early nineteenth century. The earliest studies investigated groundwater as a resource for drinking water. The first systematic, national study was in 1868 and was motivated by the cholera epidemics at that time. At the beginning of the twentieth century, research for drinking water production was institutionalised at national level. Since the 1960s, the range of organisations involved in hydrogeochemical research has broadened. Societal motives are also identified: shallow, biogenic methane as fossil fuel (already researched since the 1890s); groundwater contamination; freshening/salinisation of aquifers; ecohydrology and nature conservation; aquifer thermal energy storage; national and regional groundwater monitoring for policy evaluation; impact of climate change and weather variability; and occurrence of brackish groundwater and brines in the deeper subsurface. The last-mentioned has been driven by a series of motives ranging from water supply for recreational spas and mineral water production to subsurface disposal of radioactive waste. There have been two major scientific drivers: the introduction of techniques for using isotopes as tracers, and geochemical computer modelling. Another recent development has been the increasing capabilities in analytical chemistry in relation to the contamination of groundwater with emerging pollutants. Many of the motives for research emerged in the 1980s. Overall, the societal and associated technical motives turn out to be more important than the scientific motives for hydrogeochemical research on groundwater in the Netherlands. Once a research motive has emerged, it commonly tends to remain.

Keywords Drinking water · Groundwater management · Contamination · Nitrate · Netherlands

Introduction

Hydrogeochemistry is the scientific discipline that addresses the chemistry of groundwater and, according to some, also surface waters, particularly the relationship between the chemical characteristics and quality of waters and the regional geology. The primary aspects of hydrogeochemistry are the characterisation of the aqueous composition and interpretation of the controls in terms of recharge origin and geochemical processes during transport.

This paper is intended to give a brief overview of the hydrogeochemical research on groundwater in the Netherlands. Virtually all this research has been driven by management of groundwater resources and environmental management in a broader sense. Therefore, the paper presents, in a systematic way, a discussion of the emergence of research topics, the motivations for the research from a groundwater management perspective, the introduction of new international research methods, and insight into whether research on the topic did continue. With respect to the latter, a limited number of references is given as an example, without aiming to cite all available literature if valid.

The focus on hydrogeochemical research on groundwater implies that only studies that deal with *reactive* solutes will be considered. Thus, salinity as a more physical property, lies beyond the scope of this paper; it merits a separate study because so much research on groundwater salinity and its controls has been done in the Netherlands. Microbial research on groundwater quality is not covered here either, even though it may be interlinked with hydrogeochemistry.

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Methods and study area

Methods

This study comprises a review of the scientific literature, focusing on the scope of the research investigations as well as the methods employed. The core body of literature reviewed comprised articles and other manuscripts as collected by the author in the past 35 years. It has been augmented with peer-reviewed articles published since 1945 and found using scientific search engines and keywords such as groundwater, quality, Netherlands and nitrate. Older literature was collected from reference lists and report overviews in institutional anniversary manuscripts. Groundwater studies were classified as “hydrogeochemical” if they dealt with groundwater analyses (or the experimental equivalent) in some way.

Study area

The typical situation of the Netherlands with respect to groundwater is summarised as follows (Griffioen et al. 2013). The Netherlands has a temperate climate with an average 850 mm/year precipitation and 700 mm/year potential evaporation. The population density is ca. 500 inhabitants/km^2, which is high when compared internationally. Especially the western part is strongly urbanised and also industrialised. Two-thirds of the country has agricultural land use, which is commonly intensive.

Geologically, the Netherlands lies at the southeastern border of the North Sea sedimentary basin. The groundwater compartment is wedge-shaped, increasing in thickness from about 50 m in the east-southeast to about 400–500 m in the west–northwest (Dufour 2000). Reservoirs for oil, gas and geothermal energy production are present down to 4 km depth. The groundwater compartment contains multiple sandy layers that act as aquifers and the availability of groundwater is high compared to other countries. The Netherlands can be divided into a Holocene area and a Pleistocene area according to surface geology and related landscape (Fig. 1; De Mulder et al. 2003; De Gans 2007). The low-lying western and northern part of the country as well as the central, riverine part contain a Holocene layer at the top. This Holocene layer comprises of (1) surficial aeolian sediments along the North Sea, (2) marine sediments and peat in the northern part and also fluvial sediments in the western part, and (3) fluvial sediments and peat in the central, riverine part.

The aeolian, Holocene sands along the North Sea form dunes and barriers. The maximum elevation of these dunes is several tens of metres above mean sea level. Nowadays, they serve as a natural area of groundwater recharge for the adjacent polders or as an artificial area of recharge for the production of drinking water through the infiltration of prepurified River Rhine and Lake IJssel water. Large parts of the Holocene area consist of polders that are artificially discharged through a system of drains and ditches where surface water is pumped out of these polders. In the polders, the Holocene layer often acts as a semiconfining layer for the Pleistocene aquifers below, which are predominantly composed of fluvial sands; however, Pleistocene glacial sediments and Eemian marine sediments are also found in parts of the Netherlands. Many of these polders lie below sea level. Lakes are also frequently present in this area. Surface runoff and shallow subsurface runoff of rainwater are substantial in these polders, while the recharge of aquifers from relatively high-lying polders is limited to 30–100 mm/year (NHV 2004). Additional groundwater recharge occurs via infiltration from lakes, rivers and the North Sea or Wadden Sea and also from ditches when water from the large rivers is channelled into the polders during dry summer months. A considerable number of drinking-water production sites is present along the Rhine River and rely on riverbank infiltration. As indicated by tritium dating of groundwater, much shallow groundwater under the polders is “old”, i.e., has infiltrated before 1950 and may be brackish (Cl is 300–1,000 mg/L) or saline (Cl > 1,000 mg/L; Frapporti et al. 1993; Van den Brink 2007).

Fig. 1 The Netherlands, showing the occurrence of Holocene (light green) versus Pleistocene and older sediments (dark green) at the surface, the major rivers and the locations of the geographical names referred to in this paper

The Pleistocene area of the Netherlands comprises older fluvial deposits and glacial or periglacial deposits near the surface. Phreatic aquifers are found here and groundwater is recharged via rainwater infiltration. This recharge is around 350 mm/y but lower under pine forest. Groundwater is regionally discharged through a natural drainage system of rivers and brooks that is genetically controlled by the precipitation surplus, the hydraulic resistance of the subsurface and the topography (De Vries 1976). Tritium dating has revealed that most groundwater at depths between 10 and 30 m has infiltrated since 1950 (Frapporti et al. 1993; Van den Brink et al. 2007), indicating that most but not all shallow groundwater is "young". Many groundwater abstractions for drinking water purposes are present in this area. The southernmost part of the Netherlands has an entirely different shallow geology. Here, Pleistocene loess deposits lie at the surface, covering Cretaceous carbonate rocks or Tertiary clastic sediments. Several groundwater abstraction sites for drinking water purposes are present in this part of the country. It has been less well studied hydrogeochemically.

Three hydrogeological situations have been distinguished when it comes to hydrogeochemical studies of Paleogene and older deposits (Griffioen et al. 2016): (1) shallow, semi-confined aquifers, (2) deep, oil and gas reservoirs that also serve as reservoirs for geothermal energy and (3) deep, buried aquifers. The first deals with Palaeogene and older sediments lying close to or at the surface near the border with Belgium or Germany. The second deals with formation waters in Permian to Early Cretaceous oil and gas reservoirs at 950–4,000 m depth in western and northern Netherlands. The last deals with Upper Devonian to Palaeogene aquifers and buried deep at several hundreds to 1,100 m depth as especially sampled in southern Netherlands.

19th century: drinking water and fossil fuel

The oldest known hydrogeochemical study done in the Netherlands that presents groundwater analyses is that by Mulder (1827), who presented five groundwater analyses together with a series of surface water analyses. The groundwater samples were collected from wells in Amsterdam and the villages of Vianen and Rheden in the centre of the Netherlands and reported in terms of dissolved constituents of $NaCl$, $MgCl_2$, Na_2SO_4, $CaSO_4$, $CaCO_3$ and $MgCO_3$, as commonly done in the nineteenth century. His study addresses water quality for drinking water purposes, focusing on salinity and odour. The interpretation is strongly hypothetical. The PhD thesis by Harting (1852) on the geology underlying Amsterdam also includes a discussion on the quality of groundwater in Amsterdam for a depth range of 60 m below the surface (and one outlier down to 151 m). Using groundwater analyses from 1850–1851, Harting interpreted the hydrogeochemical data in terms of drinking water quality and hypothesised that infiltrating marine water during the Holocene marine transgressions was responsible for the high salinity of some of the groundwater. Here, one may realise that Amsterdam is situated in the coastal lowlands of western Netherlands where Holocene marine sediments lie at the surface. In his PhD study on rain, surface water and groundwater in the Netherlands, Gunning (1853) collected nine samples from groundwater wells producing the "best and tastiest drinking water". The wells were spread across the country. He paid considerable attention to the analytical aspects of water analyses and the origin of soluble (like carbonate and chloride) versus insoluble salts (like silicates) dissolved in water.

In the mid-nineteenth century, against a background of a growing population and outbreaks of cholera, physicians became increasingly aware of the inadequacy of the drinking water (PIE 1997). In response to this and because of the frequent occurrence of brackish and saline water in Amsterdam and other cities in western Netherlands, the first central drinking water supply was set up by the Dunewater Company at Leiduin in 1853 to supply Amsterdam with drinking water sourced from the dunes (Fig. 2). The city of Den Helder in northwest Netherlands soon followed with a central supply system. Unfortunately, no groundwater analyses associated with the early stages of dune water abstraction for central drinking water supply could be found. The oldest systematic investigation on regional groundwater quality was reported in 1868 (Report to the King 1868). The related commission had been appointed by the king in 1866 to investigate the relationship between the quality of

Fig. 2 The Oranjekom (Orange bowl; photographed in 1909), which was excavated by the Dunewater Company and named after King Willem van Oranje, who (as the 11-years-old crown prince) dug the first sod in the ground at the construction site of the first centralised drinking water production facility in the Netherlands (obtained from Noord-Hollands Archief, 1100 – Image collection of the municipality of Haarlem, inventory number 38723)

drinking water and the extent of the cholera epidemic. Of the 272 water samples collected throughout the Netherlands and analysed for their chemical constituents, 171 were groundwater. The suitability of surface water or groundwater for drinking water purposes was evaluated based on four criteria that addressed whether the sample had a pristine nature or wastewater characteristics. In the last three decades of the nineteenth century, 57 cities obtained a centralised drinking water supply thanks to the acceptance of the causal relationship between human health and safe drinking water and the realization that the need for safe drinking water was greatest in cities (PIE 1997).

Focusing especially on the west of the Netherlands, Lorié (1899) addressed groundwater quality with respect to salinity, dissolved Fe and high carbonate concentrations, based on a number of studies conducted in the second half of the nineteenth century. One unique set of 16 samples was collected by means of multilevel sampling of a geological boring to a depth of 335 m. Another boring went down to 200 m from which 11 samples were collected. The other borings resulting in single samples commonly reached down to 60 m below average sea level with three outliers of 178, 150 and 94 m below average sea level. The analyses were interpreted in terms of percentage of seawater, based on the Cl concentration, and deviations from this for Ca, Mg and SO_4 (indicated as SO_3) as other major ions were attributed to hydrogeochemical processes. Three issues remained enigmatic: the cause of the salinity inversion (i.e., fresh groundwater below saline groundwater that has a higher density) in the deep boring with multilevel sampling; the possible role of cation exchange in controlling Ca, Na, K and Mg concentrations; and the likelihood that SO_4 reduction and methane production are microbially mediated. These became frequently studied national and international hydrogeochemical research topics.

By the end of the nineteenth century, hydrogeochemical research was also investigating the presence of methane within the first tens of metres of groundwater in polder areas in the west of the Netherlands (Ribbius 1898a, b, c). This biogenic methane was first discovered in 1870 when drilling a groundwater well in Delft. In the next few decades, interest rose in the production of this methane as fuel, and hypotheses were put forward about the source of the methane and the flow of groundwater between the dunes and the polders. Methane in groundwater has remained a research issue until now, for various reasons: the need for water treatment, especially when groundwater is intended to be used for drinking water (cf. Kruithof and Koppers 1989; De Vet et al. 2013), technical complications arising from gas clogging when groundwater is reinjected in aquifer thermal energy storage installations (Fortuin and Willemsen 2005) and background concentrations in relation to potential leakage of thermogenic methane from the deeper subsurface, especially via leakage at gas wells (Schout 2020). A drinking water supply company has recently started to produce methane as fuel from extracted groundwater at a production station where groundwater contains on average 40 mg methane/L (P+ 2014).

Early 20th century: institutionalisation and health

At the beginning of the twentieth century, hydrogeochemical research relating to the drinking water supply was intensified, and it remains a major motivation for hydrogeochemical research today. A shortage of fresh groundwater reserves was noted in the dune area around 1900, at a time when interest in managed aquifer recharge (MAR) was already growing (Romijn 1973). Instead, deeper wells were installed in the dunes, resulting in overexploitation. Not until 1957 was artificial recharge with water from the River Rhine started, to ensure the supply of drinking water to Amsterdam (Roebert 1972), leading to hydrogeochemical research on MAR (e.g. Engelen and Roebert 1974; Stuyfzand 1993). More recently, hydrogeochemical research on MAR has extended beyond the drinking water supply to include water availability for greenhouse horticulture and agriculture (e.g. Zuurbier et al. 2016; Kruisdijk and Van Breukelen 2021).

An important early milestone is the PhD thesis of Van der Sleen (1912) on the groundwater chemistry of the dunes. Although it is not the first Dutch PhD thesis that deals with groundwater analyses, it is the first that applies hydrogeochemical principles. A year later, the Dutch Rijksbureau voor Drinkwatervoorziening (RBD; State Office for Drinking Water Supply) was established, together with the Centrale Commissie voor Drinkwatervoorziening (Central Committee for Drinking Water Supply). That office and its successor, the Rijksinstituut voor Drinkwatervoorziening (RID; State Institute for Drinking Water Supply), played a major role in hydrogeochemical research in the following decades. In 1984, the RID was merged into the RIVM together with the RIV (Rijksinstituut voor Volksgezondheid) and the IVA (Instituut voor Afvalstoffenonderzoek). Since the 1960s, hydrogeochemical research has been institutionally broadened: hydrogeological research began at the Vrije Universiteit Amsterdam in the late 1960s, TNO-DGV had been mapping groundwater resources since 1967, and KIWA (with the drinking water companies as shareholders) had been doing research on drinking water since 1972. Much research was published in Dutch as reports. Later, other organisations also became active in hydrogeochemical research, and from 1980 on, more and more of the findings were published in English-language peer-reviewed journals. Only two publications were published in peer-reviewed journals between 1945 and 1969, two in the 1970s, an average

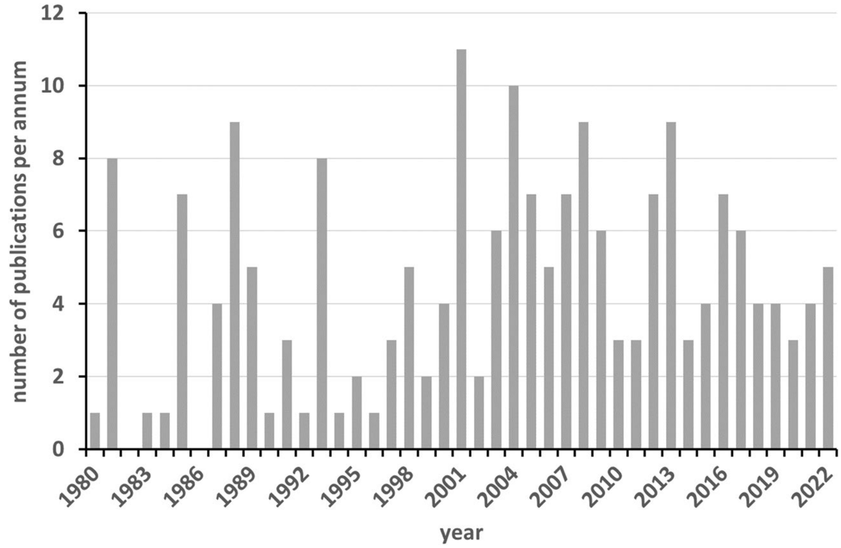

Fig. 3 Numbers of peer-reviewed articles per annum on hydrogeochemistry of groundwater in the Netherlands during the period 1980–2022

of 3.2/year between 1980 and 2000, and an average of 6.0/year since 2001 (Fig. 3).

Before World War II, RBD (i.e., State Office for Drinking Water Supply) had done research on iodide and fluoride in groundwater for reasons of human health: the realization that a lack of iodide in drinking water could induce goitre led to a national investigation of iodide in groundwater (Heymann 1925, 1927; Krul 1933). It is worth pointing out that these authors, who wrote in Dutch, noted that iodide may become incorporated in Ca carbonate as an impurity—this conclusion was presented internationally as a new finding nine decades later within the framework of subsurface disposal of radioactive waste (Claret et al. 2010). The practical environmental relevance of the finding is that ^{129}I is among the radioisotopes that obtain strongest attention in subsurface disposal of radioactive waste as it becomes produced in radioactive waste, has a long half-life of 16.1 million years and is potentially mobile in the groundwater environment.

Post World War II: intensification and internationalisation

The importance of hydrogeochemical processes during the freshening of saline aquifers was first recognised internationally by Rutten (1949) together with Foster (1950). The existence of a fresh Na–HCO_3 groundwater type in the Netherlands had been realised earlier, but Rutten was the first to explain it in terms of cation exchange during freshening. The presence of such a water type was initially enigmatic as it is commonly associated with the weathering of rocks rich in Na-feldspar like granite or dissolution of $NaHCO_3$ salts, which does not hold for the Netherlands. The topic of freshening and salinisation of coastal aquifers has remained a major research topic since then nationally and internationally, and it is also important within the framework of sea-level rise induced by climate change as well as urbanisation and associated overexploitation of groundwater resources. Some of this research has investigated groundwater salinity as a physical attribute, but the hydrogeochemical processes associated with freshening/salinisation have also been intensively studied (e.g. Geirnaert 1973; Beekman 1991).

In the late 1960s, isotope research at Groningen University led to tracers being adopted in groundwater research. Mook (1968) wrote his PhD thesis on the use of ^{14}C-carbonate, $\delta^{18}O$–H_2O and $\delta^{13}C$-carbonate analyses as tracers in surface water and groundwater. The first Dutch study using ^{3}H analysis of groundwater for groundwater aging was by Herweijer et al. (1985) and traced the history of manure leaching to groundwater. Later, Visser et al. (2007) used combined $^{3}H/^{3}He$ analysis for similar reasons, as ^{3}H analysis loses its sensitivity over time. Other isotopic and chemical tracers that have been used as tracers for contaminants and to elucidate the hydrogeochemical processes are $\delta^{7}Li$, $\delta^{11}B$, $\delta^{2}H$–CH_4 $\delta^{13}C$–CH_4, $\delta^{13}C$–DOC, $\delta^{15}N$–NO_3, $\delta^{18}O$–NO_3, $\delta^{34}S$–SO_4, $\delta^{18}O$–SO_4, $\delta^{37}Cl$, $^{87}Sr/^{86}Sr$, $\delta^{2}H$ and $\delta^{13}C$ of benzene and ethylbenzene, and the rare earth elements, particularly gadolinium (Mancini et al. 2002; Van Breukelen et al. 2003; Petelet-Giraud et al. 2009; Beekman et al. 2011; Zhang et al. 2012; Negrel et al. 2020).

Groundwater contamination emerged as a research topic at the end of the 1970s. Its importance was consolidated by the International Symposium on the Quality of Groundwater organised by the RID and held in 1981 in the Netherlands. Ten papers by Dutch researchers were among the proceedings papers published in a special issue of the journal *Science of the Total Environment* (Van Duijvenbooden et al. 1981). In particular, they addressed local contamination from landfills and diffuse contamination from agriculture. Soon thereafter, line contamination of groundwater by riverbank infiltration became a research topic, particularly in relation to the drinking water supply, as along the Rhine and Meuse there is a series of drinking-water-abstraction sites (Van der Kooij et al. 1985; Stuyfzand 1989).

Research on groundwater contamination has continued since then, and the types of contaminants studied have broadened from nutrients, heavy metals, acid rain and classical organic microcontaminants such as biocides (e.g. Peters and Den Blanken 1985; Krajenbrink et al. 1988; Van Bennekom et al. 1993; Gaus 2000; Griffioen 2001; Fest et al. 2007; Van der Grift and Griffioen 2008) and now include nanoparticles, gasoline additives, antibiotics, per- and poly-fluoroalkyl substances, etc. (e.g. Van Wezel et al. 2009; Eschauzier et al. 2013; Hamann et al. 2016; Bäuerlein et al. 2017; Kivits et al. 2018). A rarity is the study by Veen and Meijer (1989) on ^{137}Cs as fallout from the Chernobyl accident (in nowadays Ukraine) near the MAR site in the dunes at Castricum in western Netherlands. Contamination from agriculture has remained a primary concern, but other sources of pollution also need attention. The realisation that groundwater contamination poses a threat to drinking water production (as noted by the references mentioned in the preceding) was accompanied by the realisation that groundwater contamination also threatens surface-water quality. So, research on groundwater quality was combined with research on the interaction between surface water and groundwater, and the role of the riparian zone in the attenuation of contaminants such as nitrate also came to be investigated (e.g. Bleuten 1989; Hefting and De Klein 1998; Van Lanen and Dijksma 2004; Yu et al. 2019).

The issue of national-scale characterisation of groundwater quality returned at the end of the 1970s. From 1979 to 1984, the RID set up a national groundwater quality monitoring network, which was followed later by provincial groundwater quality monitoring networks. There were three official motives for the national network (Van Duijvenbooden et al. 1981): (1) to make an inventory of the current groundwater quality to complement the information already available; (2) to identify long-term changes in groundwater quality; (3) to provide the information required for adequate groundwater management. The introduction of the European Water Framework Directive in 2000 further stimulated interest in trends in groundwater quality. The data from these monitoring networks have been used in research to serve policy evaluations and in terms of data mining (e.g. Frapporti et al. 1996; Pebesma and De Kwaadsteniet 1997). Later, the data were also used in research to identify trends in groundwater quality, which is the purpose they were intended for (e.g. Broers and Van der Grift 2004; Visser et al. 2007). Alternative nation-wide investigations were conducted by Mendizabal et al. (2011, 2012), who took a hydrological system analysis perspective and used water quality analyses from drinking-water-production stations, and by Griffioen et al. (2013), who took a geochemical/palaeohydrological perspective and mined the groundwater quality database of TNO Geological Survey of the Netherlands.

Hydrogeochemical research in relation to drinking water production has remained important not only in relation to groundwater contamination, MAR and methane chemistry. Hydrogeochemical research on the technical problem of well clogging began in the 1980s, with papers by Van Beek and Kooper (1980) and Van Beek (1985). One of the aspects that subsequent studies have investigated is the feasibility of subsurface iron removal to avoid clogging (Appelo and De Vet 2003; Wolthoorn et al. 2004).

The composition of deep groundwater (frequently as brines, which are much more saline than seawater) also started to be investigated in the 1980s by Glasbergen (1981) and Coenegracht et al. (1984). Also worth mentioning in this context is an early study by Kimpe (1963), published in French, on the groundwater composition in the coal mining district in southeast Netherlands. In this area, brackish to highly saline groundwater or formation water is present in Paleogene and older geological units, and surficial recharge as a driving force for flow is absent or negligible in these buried layers (see Griffioen et al. 2016). There have been various reasons behind the research on deep groundwater, but especially important topics of investigation are the diagenesis of oil and gas reservoirs, water supply for recreational spas and mineral water production, subsurface disposal of radioactive waste, production of geothermal energy and the risk of leakage of brines to shallow groundwater.

Geochemical computer modelling arrived on the scene in the mid-1980s. Appelo and Willemsen (1987) coupled the geochemical model code EQ3/6 to a one-dimensional (1D) mixing cell transport model and used this to model freshening in the Groot Mijdrecht polder to augment theoretical modelling. Since then, geochemical batch models and 1D or multi-dimensional reactive transport models have been used in studies on groundwater in the Netherlands. Among the applications of this research are managed aquifer recharge and natural attenuation of landfill leachate plumes (e.g. Saaltink et al. 2003; Van Breukelen et al. 2004).

Meanwhile, Dutch ecohydrological research was becoming more internationally oriented. Tollenaar and Ryckborst (1975) published the results obtained from analysing data that had been collected as early as 1946–1962 from lysimeters in vegetated and bare sites in the coastal dunes. Additionally, the role of groundwater dynamics and the chemistry of groundwater began to be studied as abiotic aspects that influence the plant ecology in wetlands in terms of location factors (e.g. Grootjans et al. 1988; Wassen et al. 1989). Water-related research on nature-based solutions may also fall under ecohydrological research—an example is the research on groundwater composition in the Sand Motor, an experimental mega coastal sand nourishment that uses a nature-based approach, reported by Pit et al. (2017).

Another topic that emerged at the end of the 1980s was the hydrogeochemistry of aquifer thermal energy storage systems (ATES). Initially, research focused on technical complications, particularly on potential well clogging by carbonate precipitation ("scaling") in high-temperature ATES (Griffioen and Appelo 1993). In the early 1990s, interest in ATES as a research topic disappeared due to low oil and gas prices and associated lack of socio-economic interest, but since around 2000 it has revived in the context of the energy transition, first with a focus on low-temperature ATES and more recently focusing on high-temperature ATES too. The research scope has also been broadened to cover environmental effects in addition to technical complications (e.g. Bonte et al. 2013a, b, c) as well as physical attributes such as system interference.

The most recent major research motive that has emerged is the impact of climate change. Visser et al. (2012) addressed the impact of leaching of heavy metals near a historical Zn ore smelter. Earlier, the impact of weather variability had been investigated, especially in relation to leaching of nitrate from farmers' fields and its annual variability (e.g. Fraters et al. 1998; Boumans et al. 2001).

Discussion and conclusions

The preceding overview of the history of hydrogeochemical research on groundwater in the Netherlands shows that the motives for the research are diverse (see Fig. 4 for a summary). Most are societal in origin, with a reliable and safe supply of drinking water being the main driver of research for 200 years. There are technical motives associated with the research relating to drinking water supply. Energy supply and storage have also been important motives. Scientific curiosity and technical innovations per se have rarely been the primary motives, as the research objectives have usually been embedded in water resources management, although the research methods have certainly been determined by technical innovations, particularly in the case of analytical techniques (Johnson et al. 2013). In terms of the geographical extent of the research, groundwater contamination has been and continues to demand much research throughout the Netherlands because this is a rich industrialised country with a strong tradition in intensive agriculture and associated large emissions to the environment. The overview confirms the pivotal role that hydrogeochemistry has had in groundwater quality management (Edmunds 2009).

Some of the research motives were transitory, e.g., cholera outbreaks and concern about a lack of iodine in drinking water (the latter was subsequently dealt with by the introduction of iodised table salt). However, most of the motives remain relevant today, which illustrates how stubborn the problems associated with groundwater quality management often are. This persistence runs counter to what Meybeck and Helmer (1989) noted for rich, developed countries; they argued that water quality problems continue to rise until unacceptable conditions are reached and then decline after the problems have been recognised and effective measures have been implemented. Although this view may hold for many individual contaminants, it does not necessarily hold for groundwater resources as a whole. On the one hand, there is a continuously repeating water-quality-management process, with new contaminants demanding attention. On the other hand, recognition of contamination may not always lead to effective, preventive measures being implemented at the source. There may be no political will for such measures

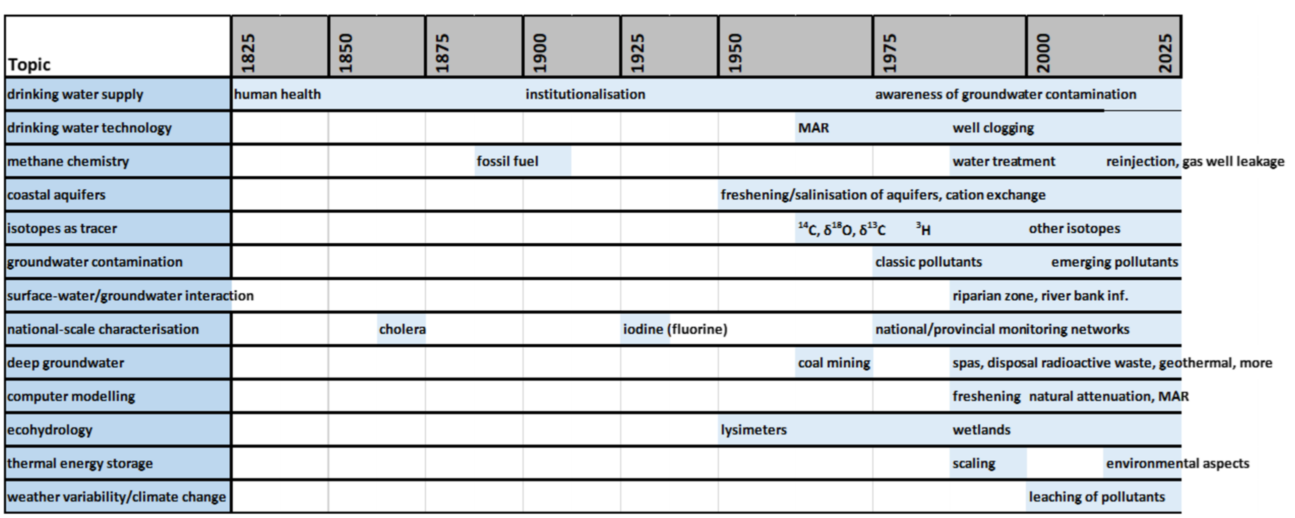

Fig. 4 Summary of the incentives that have driven hydrogeochemical research on groundwater in the Netherlands since its beginnings in the early nineteenth century

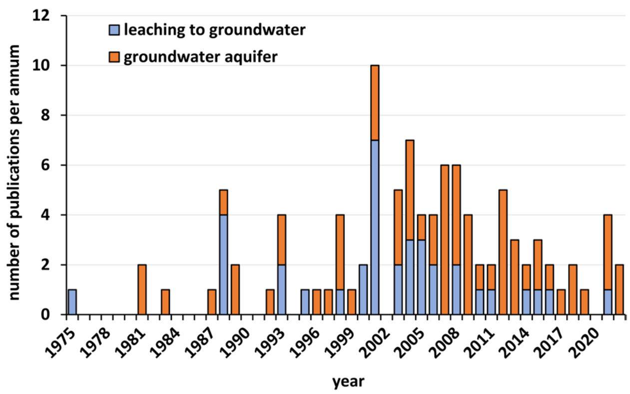

Fig. 5 Numbers of peer-reviewed articles per annum that address leaching of nitrate to the groundwater or the chemistry of nitrate in aquifers of the Netherlands for the period 1975–2022

(Dietz and Hoogervorst 1991). If that is the case, drinking water companies must resort to curative, end-of-pipe measures that are accompanied by hydrogeochemical and other kinds of research.

The situation described previously holds in the Netherlands for the implementation of the European Nitrates Directive and the Dutch Manure and Fertilisers Act. They came into force in 1991 and 1987, respectively, but their effectiveness has been limited, especially since about 2000 (Van Eerdt and Fong 1998; Van Grinsven et al. 2016; PBL 2017). The motive for research associated with nitrate pollution also remains valid because the problem did not get solved and new research questions arose such as the impact of climate change, which is illustrated in Fig. 5 and shows the number of groundwater-related publications per annum that address nitrate chemistry in groundwater in the Netherlands. Two types of research can be distinguished: (1) hydrogeochemical studies on the chemistry of nitrate in groundwater aquifers and (2) soil chemistry studies on the leaching of nitrate to groundwater that present chemical analyses of groundwater sampled at the groundwater table. The first addresses nitrate-contaminated groundwater below agricultural areas and topics such as managed aquifer recharge with nitrate-containing water. The second are studies conducted in nature and agricultural areas. It is striking that before the legislation came into force, there were few publications on nitrate contamination, which indicates that the legislation on nitrate contamination was not primarily a response to scientific publications. The period 2000–2012 yielded the most publications on nitrate contamination per annum. Scientific attention was thus greatest during that period, covering both soil leaching and aquifer chemistry. Thereafter, the number of scientific publications on nitrate contamination of groundwater in the Netherlands decreased, yet the environmental problem of nitrate contamination did not disappear despite the legislative framework present.

It is worth noting that some topics have many research motives and that the motives also shift over time in response to societal and technological developments, which is particularly the case for research on the presence of saline and brackish groundwater in deeper aquifers (Fig. 2). It also holds for methane chemistry: methane-rich groundwater was studied as a fossil fuel resource more than a century ago, but nowadays it is also studied in relation to potential leakage from deep oil and gas wells.

A few remarks can be made from an international perspective. The Netherlands was among the first countries to pay attention to MAR (Dillon et al. 2019). The interest was driven by the salinisation of the dune aquifers from which groundwater was abstracted to supply drinking water to Amsterdam and other large cities in western Netherlands. Somewhat related, it is worth noting that the papers by Rutten (1949) and Foster (1950) on the role of cation exchange to explain the Na–HCO_3 groundwater type as present in coastal aquifers were published in close succession presumably without knowing about each other.

Edmunds (2009) has dated the emergence of hydrogeochemical research to after World War II, noting in particular the appearance of hydrogeochemical papers in the journal *Geochimica et Cosmochimica Acta* and the publications by Hem (1959) and Garrels and Christ (1964). However, as the present overview has demonstrated, hydrogeochemical studies were being conducted in the Netherlands before World War II: hydrogeochemical research methods that go beyond reporting water analyses were employed as early as ~1900 (Lorié 1899; Van der Sleen 1912). V.I. Vernadsky also used hydrogeochemical methods to study natural water in the 1930s, publishing his findings in Russian (Edmunds and Bogush 2012). In mainland Europe, scientific publishing in the national language or in French or German was much more common in the past, so any attempt to chart the global history of hydrogeochemical research requires a multilingual approach.

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