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Review

Global diagnosis of nitrate pollution in groundwater and review of removal technologies

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HIGHLIGHTS

- 272 regions worldwide are analyzed to a rigorous diagnosis of nitrate pollution.
- Groundwater bodies can be contaminated by NO_3^- (>50 ppm) along with other pollutants.
- Agriculture, industry, sewage, septic tanks & landfills are the main pollution sources.
- The catalytic reduction has high nitrate conversion (98–100%) and no waste generation.
- Nowadays, the number of groundwater treatment plants is still very limited.

GRAPHICAL ABSTRACT

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ABSTRACT

Clean water and sanitation for the world population is one of the most important challenges established by the Sustainable Development Goals of the United Nations since worldwide, one in three people do not have access to safe drinking water. Groundwater, one of the main sources of fresh water, has been considerably damaged by human activities. Nevertheless, while numerous plants are globally aimed at removing pollutants from surface waters, a much scarcer number of facilities have focused on groundwater remediation. Nowadays, there is increasing concern about the presence of nitrates (NO_3^-) in groundwaters as a consequence of the intensive use of fertilizers and other anthropogenic sources, such as sewage or industrial wastewater discharge. In this context, the selection and development of highly effective and low-cost solutions for the sustainable management of groundwater resources need to be addressed. Thus, this work collects data from the literature regarding the presence of nitrates in groundwater, and, simultaneously, it reviews the main alternatives available to remove NO_3^- from groundwater sources. A total of 292 sites have been analyzed categorized by continents, carefully discussing the possible origins of nitrate pollution. In addition, a discussion is carried out of the different technologies currently employed to treat groundwater, highlighting the progress made and the main challenges to be overcome. Finally, the review gathers the data available in the literature for nitrate treatment plants at full-scale.

Contents

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1. Introduction

The supply of high-quality drinking water constitutes the Sustainable Development Goal (SDG) 6 by 2030 established by United Nations General Assembly in 2015 (United Nations, 2016). However, water bodies are continuously facing a reduction in terms of quality and quantity. Besides, despite three-quarters of our planet is made up of water, only 2.5% is freshwater, distributed as follows: 68.7% in glaciers, 30.1% groundwater, and just 1.2% is surface water (USGS Water Science School, 2013). Recent estimations indicate that almost 50% of drinking water and approximately 40% of irrigation water come from aquifers. Thus, urgent actions are required to slow down the deterioration of water bodies (Díaz-Alcaide and Martínez-Santos, 2019; Majkić-Dursun et al., 2019) since, at present, around 66% of the world's population already suffers from severe water shortages at least one month a year (Mekonnen and Hoekstra, 2016). This is mainly due to the following factors:

i) An increase in the population growth from 5.3 to 7.7 billion between 1990 and 2020, and an expected growth to 9.7 billion by 2050 (United Nations, 2019)
ii) Pollution and degradation of available water resources due to increased industrial activity since the mid-18th century. In fact, this factor is responsible for human health issues, ecosystem damage and impacts on food production, economic activity and development (Bond et al., 2018), along with the discharge of untreated wastewater (80% of the total wastewater) (Bond et al., 2018; Daesslé et al., 2020)
iii) Climate change (Bond et al., 2018; Lorite et al., 2018) contributes to low rainfall and hence provokes water stress worldwide, especially in regions with desertic climates (Lorite et al., 2018; World Health Organization, 2012). Moreover, in recent years many countries have suffered chronic droughts, as well as irregular and violent rainfall leading to disastrous floods and drylands (Bouderbala, 2019; World Health Organization, 2012). In this respect, the World Health Organization (WHO) points out that the effects will be particularly acute in arid areas and areas with growing populations (World Health Organization, 2012). Also, and due to the lack of rainfall, crops require constant intensive irrigation and an increase in external agents, which in turn rise water extraction, with a consequent increase in the Water Stress Index (Lorite et al., 2018).

As summarized in Table 1, the WHO and the Food and Agriculture Organization of the United Nations (FAO) have established quality standards for drinking water and for irrigation. Accordingly, different thresholds are considered for nitrates (NO_3^-), calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), sulphate (SO_4^{2-}), chloride (Cl^-), bicarbonate (HCO_3^-) and fluoride (F^-), as well as conductivity, pH and total dissolved solids. It is important to note that FAO's values are stricter than WHO's recommendations in all the common parameters quantified.

In contrast, the inordinate use of fertilizers and pesticides in agriculture produces high number of pollutants and water degradation. Thus, fertilizers abuse as a source of nutrients and the excessive use of chemicals, such as insecticides and antibiotics, increase the pressure exerted on aquatic systems altering their autochthonous fauna and facilitating species invasions. In general, fertilizers are composed primarily of nitrogen, phosphorus, and potassium compounds, among others, to provide nutrients to plants and crops. As a result of the agriculture activities, such compounds end up in surface water and groundwater bodies, raising the concentration of nitrates in water, which are then returned to the soil through irrigation (Andreo-Martínez et al., 2020). In this sense and according to recent studies (Ayers and Westcot, 1985) nitrate concentrations above 22 mg/L (ppm) may affect sensitive crops (i.e. sugar beets or grapes), while other crops, such as maize, remain unaffected by concentrations below 132 mg/L. The risk of nitrate leaching is closely related to the excessive use of fertilizers, occurring after or during harvest and within the crop cycle. An elevated concentration may affect the production of several crops, which can lead to over-stimulation of growth, delayed maturity or poor quality. Nitrates can affect human health by inducing methemoglobinaemia, thyroid effects or cancer, and irreparable damage in the aquatic system, to the point of provoking fish die-offs (Martínez et al., 2017; Tokazhanov et al., 2020; World Health Organization, 2012).

Cost-effective and energy-efficient treatments are necessary for removing nitrates, and therefore, to contribute to achieve the SDG and the quality standards established by WHO and FAO organizations. This review aims to collect data relevant to discriminate the quality of groundwater bodies worldwide. In addition, the different technologies available so far to remove nitrates are analyzed, emphasizing their advantages and drawbacks. The work is also complemented with the most representative both pilot and full-scale plants installed with these technologies, with details on their location and their capacity and efficiency. Thus, this review brings

Table 1
Drinking water and irrigation water standards established by WHO and FAO, respectively.

Parameter	WHO (World Health Organization, 2012)	FAO (Misstear et al., 2017)
Nitrates (mg/L)	50	22
Calcium (mg/L)	300	
Magnesium (mg/L)	300	
Sodium (mg/L)	200	69
Potassium (mg/L)	12	
Sulphates (mg/L)	250	
Chloride (mg/L)	200	107
Bicarbonates (mg/L)		91.5
Fluorides (mg/L)	1.5	
Conductivity (μS/cm)	2500	
pH	6.5–8.5	
Total dissolved solids (mg/L)	600	450

together an updated diagnosis of nitrate pollution in groundwater bodies together with the analysis of the different alternative solutions for their removal.

2. Nitrate sources

Nitrate is considered the most widespread pollutant in groundwater induced by its high solubility in water and the difficulty to be fixed in the soil (Bhatnagar and Sillanpää, 2011). Although nitrogen compounds are present in the environment from natural sources e.g., coming from igneous rocks, atmospheric deposition and symbioses of some plants, along with cyanobacteria and some heterotrophs (Gutiérrez et al., 2018); the main problem of nitrates is the dramatic increase in recent years due to anthropogenic actions. On the other hand, the main anthropogenic sources of nitrate pollution are nitrogen-rich fertilizers used for agricultural purposes, discharge of poorly treated domestic, and industrial wastewaters, livestock manure, and leachate from landfill sites (Gutiérrez et al., 2018; Tokazhanov et al., 2020). In this sense, in the Mediterranean area, where agriculture is the predominant economic activity, nitrate pollution is mainly caused by the use of nitrogen-rich fertilizers. This issue affects shallow wells, due to their location close to the surface, where nitrate pollution occurs (Heaton et al., 2012; Re et al., 2017). In contrast, in developing countries the main source of nitrates in groundwater is the lack of adequate sanitation (Kapembo et al., 2016).

The nitrate cycle explains the generation of nitrate by natural pathways or by anthropogenic actions as shown in Fig. 1. Firstly, N_2 is leached and fixed in the soil by bacteria. Consequently, in a second step, the fixed nitrogen suffers ammonification, and the ammonia reacts and converts to nitrites or nitrates. These compounds, in turn, may leach to groundwater or suffer denitrification by some bacteria or plants and return to the atmosphere in the form of nitrogen gas (Gutiérrez et al., 2018).

Contrarily, many works highlighted a close relation between nitrates and chloride (Cl^-) concentration, caused by sewage infiltration (Rezaei et al., 2017; Vystavna et al., 2017). In this context, Vystavna et al., 2017 and Rezaei et al., 2017, have determined that denitrification processes occur when the ratio NO_3^-/Cl^- decreases. Su et al., 2020 reported that a high NO_3^-/Cl^- ratio may suggest pollution from fertilizers and rainwater, while low ratios correspond to manure pollution (Su et al., 2020). Moreover, when high NO_3^-/Cl^- molar ratios and high Cl^- concentrations are observed in groundwater, they are attributed to domestic sewage pollution (Su et al., 2020). Thus, areas of sewage inputs could be differentiated from the other farming inputs by Cl^- and NO_3^- concentrations.

Besides, the presence of nitrates due to the use of chemical fertilizers implies the co-existence of other ions in groundwater, such as F^-, SO_4^{2-}, and K^+ (Roy et al., 2020). In this regard, Varol and Şekerci (2018) reported that the use of certain chemical fertilizers may be the source of some heavy metals (i.e. arsenic) in groundwater (Varol and Şekerci, 2018). Mazhar and Ahmad (2020) concluded that the presence of cadmium (Cd^{2+}) and zinc (Zn^{2+}) is also due to the use of fertilizers (Mazhar and Ahmad, 2020). Moreover, *Escherichia coli*, Enterococci, and Total Coliforms are also present in groundwater when nitrate pollution is associated with domestic leaks, mainly latrines and septic tanks (Kapembo et al., 2016).

Regarding the strong seasonal fluctuations, two opposite effects could occur depending on the characteristics of the terrain: i) an increase in nitrate concentration during the rain period due to rainwater infiltration (Varol and Şekerci, 2018) and, ii) a decrease in NO_3^- concentration in the wet season because ions are diluted in the rainfall (Abu-alnaeem et al., 2018; Roy et al., 2020).

In terms of the interest aroused from the literature through studies, regarding groundwater and nitrates pollution, Fig. 2 shows the bibliometric graph of articles published on groundwater pollution and nitrate presence from the late 1960s to the present. As depicted in Fig. 2, the interest in the study of the health of groundwater has experienced an exponential growth since the early 80s. While in the 70–80 decade, the scientific community published an average of 100 articles per year, this number has steadily increased with 2742 articles published in 2020 in this field. In the case of the publications focused on the presence of nitrates in groundwater, the number of published articles has increased from 20 manuscripts published in 1990 to 280 articles published in 2021 highlighting the growing interest and concern regarding this topic.

Thus, groundwater health and specifically the presence of nitrates in groundwater is nowadays of great concern and is necessary to be thoroughly considered. In this framework, two different aspects need to be addressed: i) collecting the data reported so far, and ii) making a comparison of technological alternatives for nitrates removal to help in the decision-making process of future nitrate mitigation strategies.

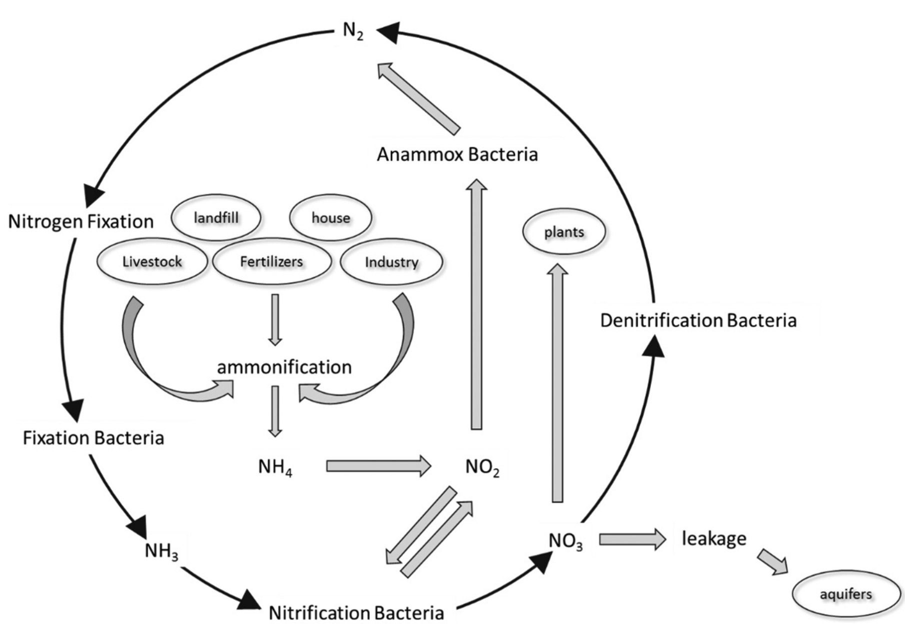

Fig. 1. Nitrogen cycle.

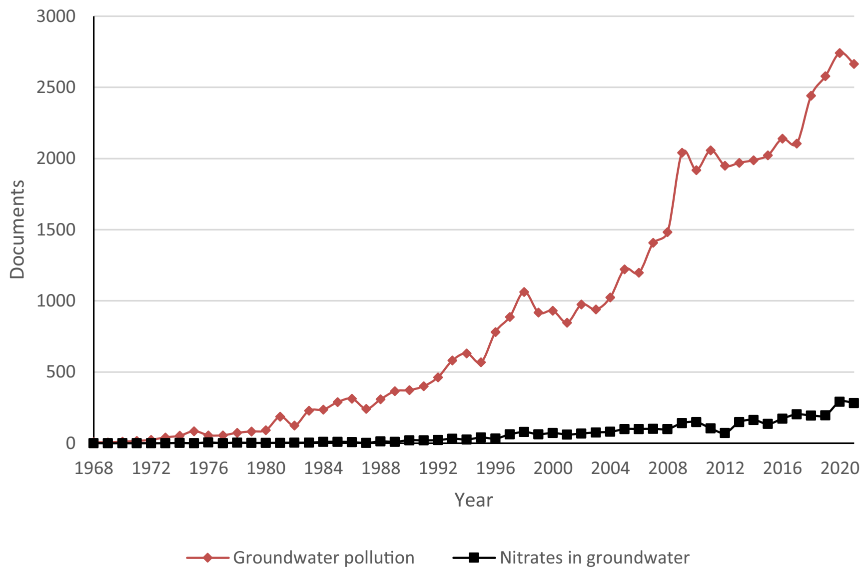

Fig. 2. Bibliometric analysis of articles published in the field of groundwater pollution and nitrate presence. (Scopus source, updated at 16th November 2021).

3. Nitrates in groundwater bodies

Groundwater hydrochemistry has been largely considered in several works, especially in the last few years (Ahmed et al., 2019; Awomeso et al., 2020; Bojarczuk et al., 2019; da Silva Peixoto et al., 2020; Devaraj et al., 2020; Erdogan et al., 2020; Gil-Márquez et al., 2019; Hepburn et al., 2020; Rodriguez-Espinosa et al., 2020). This discipline studies the main ions present in groundwater; which presence is mainly determined by the geology and mineralogical composition of the environment, the water residence time, and the rock-water interactions, among others (Islam et al., 2019). Numerous works have been reported worldwide, taking groundwater samples from wells, boreholes, and springs, that are mostly used for irrigation or domestic purposes.

Fig. 3 shows the extended concentration of nitrate differentiated with colours. Blue pushpins indicate that the mean value is under 50 ppm, the threshold established by WHO to guarantee good quality of groundwater. In contrast, yellow pushpins show the locations where, despite the average is accordance to WHO standards, some measurements are above 50 ppm. Orange colour reflects the groundwater bodies with more than 25% of the samples with nitrate concentration higher than 50 ppm, and finally, red colour pushpins indicate the locations where groundwater is highly polluted, and therefore the quality of this water is prohibited for utilization without previous treatment according to the standards established by the WHO. For a clearer visualization of the data, Table 2, summarizes the 292 points studied as a function of nitrate pollution. In this context, 94 samples were studied in Africa, 93 in Asia, 71 in Europe, and 34 in America. Although Oceania continent has reported an article in the field of groundwater health, it does not provide data about nitrates (Hepburn et al., 2020).

Regarding contaminated areas, 30 regions in Africa, 20 in Asia, and 9 in Europe are in a critical situation. Besides, Europe presents the highest percentage of regions polluted with more than 25% of samples above 50 ppm of nitrates. On the other hand, despite the data corresponding to Asia showing 45 regions without nitrate pollution, 19 areas present contamination in at least 25% of the samples. In this regard, despite the number of polluted regions in Asia is higher than in Europe, 19 and 16 respectively, in terms of percentage of polluted regions, Europe counts with more areas with concentrations of nitrates above 50 ppm. For its part, America only reports 5 regions with more than 25% of samples with NO_3^- concentration above WHO recommended value. Finally, the manuscripts addressing the African continent highlight 34 regions with average values below 50 ppm.

In the next sections, a discussion of the results will be held continent by continent, emphasizing the most striking values as well as the main source of nitrate contamination in that region. In addition, the location of nitrate pollution regions has been numbered in each figure from the lowest to the highest average value to facilitate the discussion. When the information was available, in addition to the average value, the minimum and maximum values reported in the literature have been included. The detailed references of each pushpin are included as supporting information (SI) which contains a complete characterization in terms of NO_3^-, Ca^{2+}, Mg^{2+}, Na^+, K^+, SO_4^{2-}, Cl^-, HCO_3^- of each point analyzed along the manuscript. For a clearer visualization of the data assessed among the continents, both the manuscript and the SI contain the maps and the graphics including average, maximum, and minimum values reported by the authors.

3.1. Asia

The Asian continent is the one that shows the greatest variability in the data, as can be seen in Fig. 4, which is divided in two parts. The first part includes all the regions studied (Fig. 4a), and the second one reports the mean, maximum and minimum values of each point (Fig. 4b). In the aforementioned figure, the data of 93 regions are collected. For the vast majority of the areas, the maximum value exceeds the WHO standard (50 ppm) despite the average value being within the standards. Specifically, in 19 regions, despite the mean value is in concordance to the standards recommended by WHO, more than 25% of the samples have values above 50 ppm.

A total of 20 regions located in India, Palestine, and Saudi Arabia, and other areas of China and Pakistan show a mean value above 50 ppm (Abu-alnaeem et al., 2018; Alslaibi et al., 2017; Devaraj et al., 2020; Feng et al., 2020; Jehan et al., 2019; Kumari and Rai, 2020; Musaed et al., 2020; Pant et al., 2020; Roy et al., 2020). In particular, as shown in Fig. 4, it is also possible to verify that 11 samples have maximum values above 300 ppm and 5 of them (points 81, 84, 86, 89, and 92, Fig. 4) have maximum values greater than 600 ppm. (Feng et al., 2020; Kumari and Rai, 2020; Musaed et al., 2020; Pant et al., 2020).

In India, there are numerous available samples, and the variability of the data is striking since while some of the areas studied have mean values below 50 ppm, other regions have averaged values between 59 and 215 mg/L (points 75, 76, 77, 79, 82, 84, 88 and 92, Fig. 4) (Devaraj et al., 2020; Pant et al., 2020; Roy et al., 2020). It is important to note that this last value is more than fourfold higher than the standard recommended by WHO. Birbhum (points 44 and 45, Fig. 4), Solapur (points 66

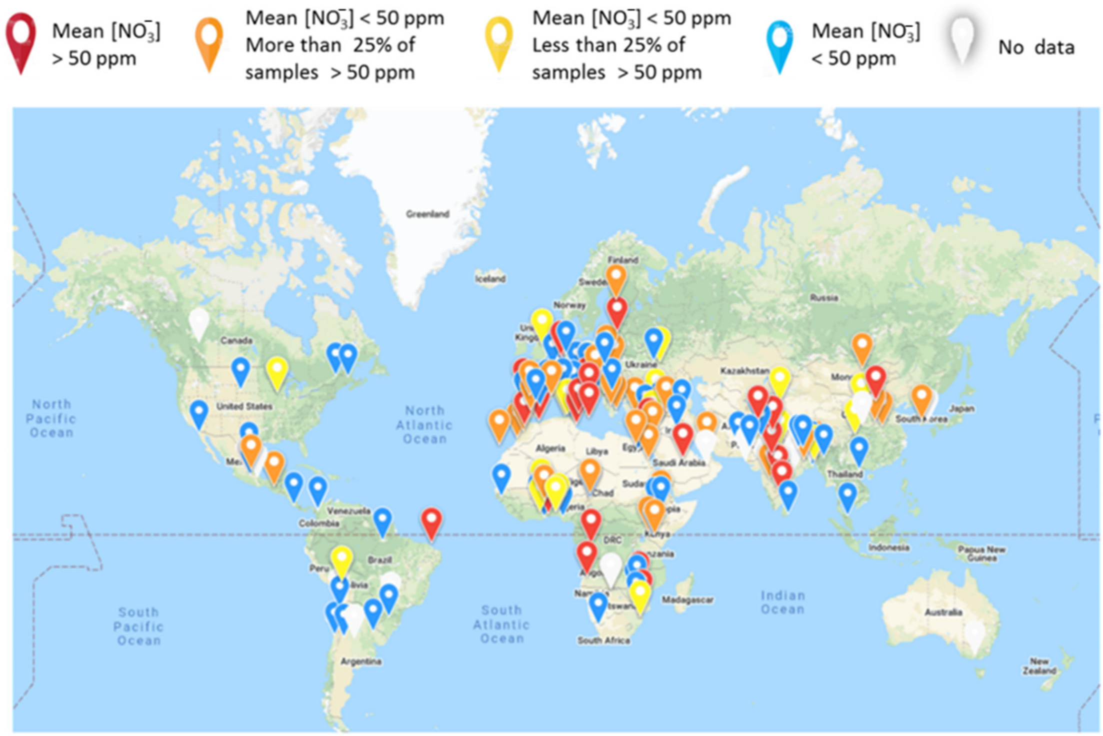

Fig. 3. Locations and nitrate concentration [NO_3^-] in groundwater bodies (Abboud, 2018; Abderamane et al., 2013; Abou Zakhem et al., 2017; Abou Zakhem and Hafez, 2015; Abu-alnaeem et al., 2018; Ahmed et al., 2019; Aladejana et al., 2020; Ali et al., 2019; Alslaibi et al., 2017; Anim-Gyampo et al., 2018; Ruiz-García et al., 2019; Ashun and Bansah, 2017; Asmael et al., 2015; Atikul Islam et al., 2017; Awomeso et al., 2020; Barbieri et al., 2019; Batsaikhan et al., 2018; Bello et al., 2019; Benkaddour et al., 2020; Benmarce and Khanchoul, 2019; Beyene et al., 2019; Bicalho et al., 2019; Blázquez-Pallí et al., 2019; Bojarczuk et al., 2019; Bon et al., 2020; Boudjana et al., 2019; Bouteraa et al., 2019; Bretzler et al., 2017; Bucci et al., 2017; Canora et al., 2019; Carasek et al., 2020; Carvalho et al., 2019; Celestino et al., 2019; Chandrajith et al., 2014; Charizopoulos et al., 2018; Costa et al., 2015; Ćuk et al., 2020; Daesslé et al., 2020; Danni et al., 2019; da Silva Peixoto et al., 2020; de Oca et al., 2019; Devaraj et al., 2020; di Lorenzo et al., 2012; Dippong et al., 2019; Egbi et al., 2019; el Gammal and Ibrahim, 2017; el Ghali et al., 2020; Elumalai et al., 2019; Erdogan et al., 2020; Erostate et al., 2018; Esteller et al., 2017; Fan et al., 2020; Feng et al., 2020; Ferchichi et al., 2018; Ferrante et al., 2018; Gamazo et al., 2018; Gamboa et al., 2019; Gevera and Mouri, 2018; Gil-Márquez et al., 2019; Giménez-Forcada et al., 2017; Gomez et al., 2019; Gorgij et al., 2019; Gromadzka et al., 2015; Heaton et al., 2012; Hossain and Patra, 2020; Islam et al., 2018, 2019; Ismail et al., 2019; Jehan et al., 2019; Kapembo et al., 2016; Karakuş, 2019; Karroum et al., 2017; Kattan, 2018; Kawo and Karuppannan, 2018; Khanoranga and Khalid, 2019; Kshetrimayum and Thokchom, 2017; Kumari and Rai, 2020; F. Liu et al., 2020; J. Liu et al., 2020; T. Liu et al., 2020; Loh et al., 2020; Loomer et al., 2019; Lorette et al., 2018; Majkić-Dursun et al., 2018; Masocha et al., 2019; Mazhar and Ahmad, 2020; Melki et al., 2019; Mendes et al., 2019; Mirčovski et al., 2018; Moni et al., 2019; Moratalla et al., 2009; Mostaza-Colado et al., 2018; Mouassa et al., 2020; Mudzielwana et al., 2020; Mukanga et al., 2016; Mukate et al., 2019; Musaed et al., 2020; Mushtaq et al., 2018; Muzenda et al., 2019; Ncibi et al., 2020; Nguyen et al., 2015; Nikolenko et al., 2019; Nyilitya et al., 2020; Ogrinc et al., 2019; Opoku et al., 2020; Ossa-Valencia and Betancur-Vargas, 2018; Owamah, 2020; Panno et al., 2019; Pant et al., 2020; Papazotos et al., 2019; Pedretti et al., 2019; Quenet et al., 2019; Quino-Lima et al., 2020; Rashid et al., 2020; Re et al., 2017; Rezaei et al., 2017; Rodriguez et al., 2020; Roy et al., 2020; Rufino et al., 2019; Sánchez-Gutiérrez et al., 2020; Sarker et al., 2018; Sedlazeck et al., 2017; Sefie et al., 2018; Silva et al., 2017; Slavinskienė and Jurevičius, 2016; Smedley et al., 2018; Snousy et al., 2020; Strauhal et al., 2016; Sunkari et al., 2019; Swift Bird et al., 2020; Talib et al., 2019; Taucare et al., 2020; Thakur et al., 2015; Tolera et al., 2020, 2017; Torres-Martínez et al., 2020; Tran et al., 2020; Tzoraki et al., 2018; Ujević Bošnjak et al., 2012; Varol and Şekerci, 2018; Vystavna et al., 2015; Yetiş et al., 2019; Zaki et al., 2019; Zango et al., 2019; Zhang et al., 2020).

and 71, Fig. 4), and Bareilly (points 31 and 51, Fig. 4), all of them located in India, they report mean values in the range between 7 and 47 mg/L. Nevertheless, these regions have also more than 25% of samples of nitrate concentration above 50 mg/L (Hossain and Patra, 2020; Mazhar and Ahmad, 2020; Mukate et al., 2019). Regarding the possible causes of contamination in this country, some authors highlight the poor maintenance of septic tanks and boreholes, sewage discharge, and fertilizers (Barbieri et al., 2019; Devaraj et al., 2020; Roy et al., 2020).

Contrarily, the mean values are between 50 and 220 mg/L in Palestine (points 80, 83, 85, 87, 90, 91, and 93, Fig. 4) and Saudi Arabia (point 86, Fig. 4), where in most cases, the authors attributed this fact to sewage disposal and farming inputs (Abu-alnaeem et al., 2018; Barbieri et al., 2019; Musaed et al., 2020). Palestine also shows mean values below 50 ppm in some regions (points 7, 8, 60, 73, and 74, Fig. 4) (Alslaibi et al., 2017).

In the Asian continent, the NO_3^- pollution is primarily due to septic tanks and agriculture inputs leakage. Moreover, many authors emphasize the high variability due to the strong influence that the rainy season has on the groundwater quality of this continent (Amanambu et al., 2020; Nguyen et al., 2015). In this continent, water scarcity is recognized as one of the most important concerns in many countries especially in the Middle

Table 2
Sources of nitrates categorized by continents.

Continent	Asia	America	Europe	Africa	Total
Number of regions with [NO_3^-] average > 50 ppm	20	1	9	30	60
Number of regions with [NO_3^-] average < 50 ppm and more than 25% of samples with NO_3^- concentration > 50 ppm	19	4	16	10	49
Number of regions with [NO_3^-] average < 50 ppm and less than 25% of samples with NO_3^- concentration > 50 ppm	9	2	6	7	24
Number regions with [NO_3^-] average < 50 ppm	45	27	39	34	145
Total number of regions	93	34	71	94	292

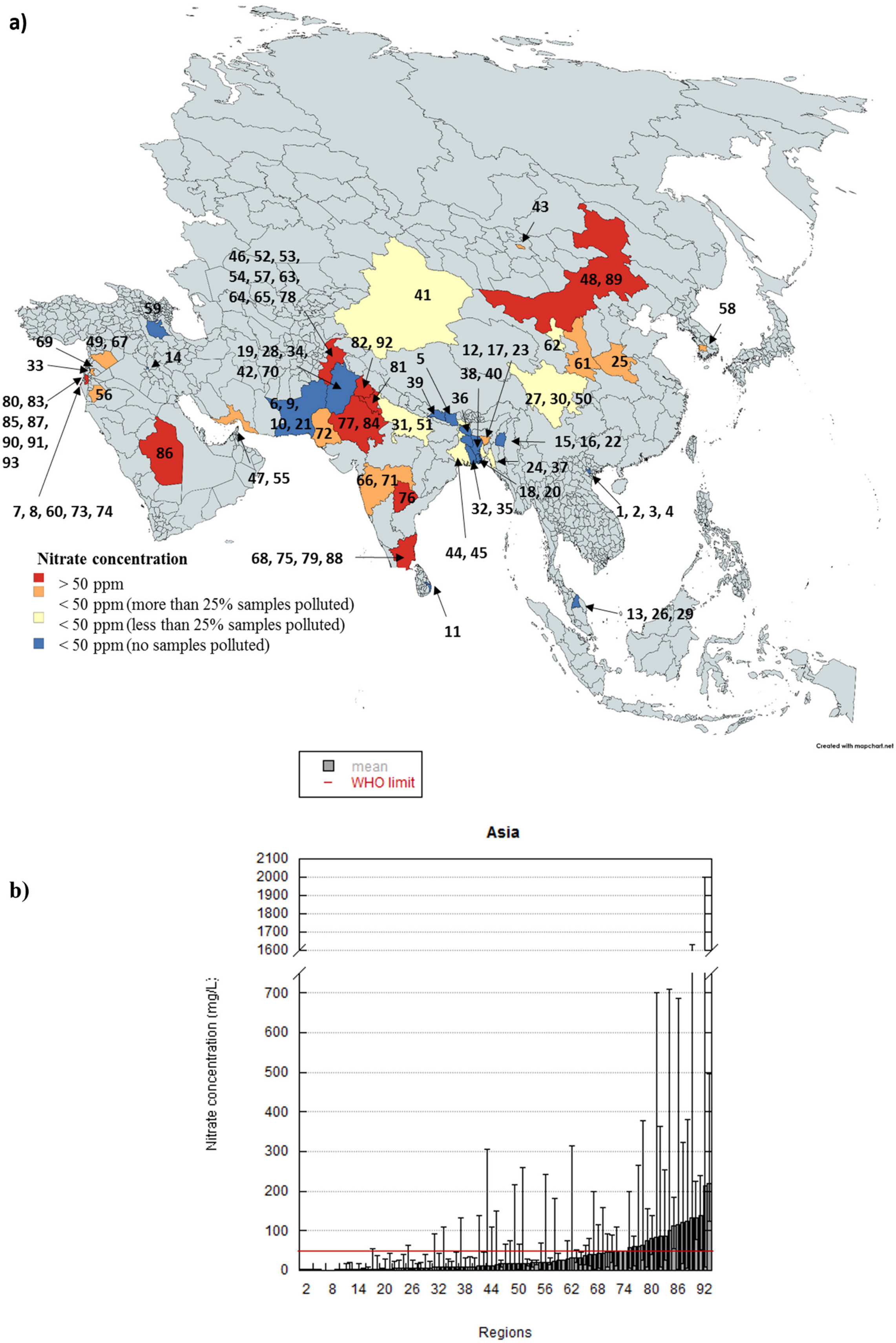

Fig. 4. a) Studied regions in Asia. b) Mean value and range of nitrate concentration in Asian groundwater bodies (Ahmed et al., 2019; Alslaibi et al., 2017; Asmael et al., 2015; Atikul Islam et al., 2017; Batsaikhan et al., 2018; Chandrajith et al., 2014; Costa et al., 2015; Fan et al., 2020; Islam et al., 2018, 2019; Ismail et al., 2019; Khanoranga and Khalid, 2019; Kshetrimayum and Thokchom, 2017; J. Liu et al., 2020; T. Liu et al., 2020; Mazhar and Ahmad, 2020; Moni et al., 2019; Mushtaq et al., 2018; Nguyen et al., 2015; Sarker et al., 2018; Sefie et al., 2018; Talib et al., 2019; Thakur et al., 2015). Detailed references are included in Table S1 of the Supplementary Information.

East, and consequently, the use of fertilizers is continuously growing, encouraging the presence of nitrates in groundwater (Alaei Shahmirzadi et al., 2018).

The different information collected is summarized in Table 3, which shows the source of nitrate pollution classified into five categories: septic tanks, sewage, agriculture, landfills, and industrial activities. Besides, three levels of risk have been determined and associated to the categories, low (▲), medium (■), or high (●) along the different countries considered in the study. Green colour has been established when the category is of "no concern" in the country, meaning there is no risk evidence so far. Medium risk has been considered when some regions in a country present concerning values of nitrate, and the category is considered as a "potential influence". Finally, the highest level of risk has been set up when any region in the country reports nitrate pollution, and the evidence indicates that this category has a "significant contribution".

Agriculture is the main activity that causes the high nitrate values in Saudi Arabia, Myanmar and Jordan (Abboud, 2018; Kshetrimayum and Thokchom, 2017; Musaed et al., 2020; Pincetti-Zúniga et al., 2020). Concerning nitrates and phosphates in groundwater in Sri Lanka, Bangladesh, Vietnam, and Nepal, recent works pointed to the agriculture inputs and latrines that penetrate the sandy aquifers as potential contamination activities (Chandrajith et al., 2014; Islam et al., 2018; Lee et al., 2018; McArthur et al., 2012; Nguyen et al., 2015; Thakur et al., 2015). In the case of Iraq and the Gaza coastal (Palestine) recent works reported a clear concern for agricultural fertilizers, industrial waste, and municipal sewage (Abu-alnaeem et al., 2018; Ismail et al., 2019; Mostaza-Colado et al., 2018). In contrast, the information available for Iran explains that in this country, the nitrate concentration is regularly low (Rezaei et al., 2017). Still, it can achieve abnormal values subsequently of draining or overflow from the farming areas together with human or livestock wastes. On the other hand, poor living conditions and unsuitable sanitary and sewage disposal practices constitute an additional source of NO_3^- pollution in developing countries such as Mongolia (Batsaikhan et al., 2018). Finally, despite Malaysia has NO_3^- values within the WHO recommended standards, the impact of intensive human activities and deforestation in certain areas has provoked an increase in nitrate contamination (Sefie et al., 2018).

Table 3
Main causes of nitrate pollution in Asia.

	Septic tanks	Sewage	Agriculture	Landfills	Industrial
Asia	●	●	●	▲	■
Bangladesh	■	▲	■	▲	▲
China	●	●	●	▲	●
India	●	●	●	▲	▲
Iran	▲	▲	■	▲	▲
Iraq	▲	▲	■	■	■
Jordan	■	■	■	▲	▲
Malaysia	▲	■	■	▲	▲
Mongolia	■	■	▲	▲	▲
Myanmar	▲	▲	■	▲	▲
Nepal	▲	■	■	▲	■
Pakistan	●	▲	●	▲	■
Palestine	▲	●	●	▲	■
Saudi Arabia	▲	▲	●	▲	▲
South Korea	▲	▲	■	▲	■
Sri Lanka	■	▲	■	▲	▲
Syria	▲	●	■	▲	■
Vietnam	■	■	■	▲	▲

Risk of contamination: (▲) low, (■) medium, (●) high.

In a different way, the high values of nitrates in different regions (e.g., Pakistan) are attributed to the solubility of minerals, such as feldspar, biotite, muscovite, calcite, and dolomite (Jehan et al., 2019; Khanoranga and Khalid, 2019). In this line, Khanoranga and Khalid (2019) highlighted that the composition of Balochistan province area groundwater is greatly influenced by the geological composition and anthropogenic activities like agricultural occupations and brick kiln factories carried out in the vicinity of the study area (Khanoranga and Khalid, 2019).

China, for its part, has different hypotheses to justify the increment of nitrates in groundwater. For example, in a recent article, high values are defined as non-point sources of contamination (Feng et al., 2020). In particular, Feng et al. (2020) emphasized the risk associated with irrigation of farmland employing sewage, and excessive nitrogen fertilizer usage. In this regard, Tolera et al. (2017) and Kim and Park (2016) reported groundwater contamination by nitrate from non-point sources (application of fertilizers and animal wastes) as a serious concern in South Korea (Kim and Park, 2016; Tolera et al., 2017).

3.2. America

In this section, the different regions of the American continent are discussed, where 34 areas are collected. Fig. 5 illustrates all the regions studied in this continent (Fig. 5a) and the mean values sorted by size, including the minimum and maximum data (Fig. 5b). Although up to 11 countries have been studied in this continent, only one of the references reported an average value of nitrates higher than 50 ppm (point 33, Fig. 5). This region is Fortaleza (Brazil) which informed a concentration above WHO's limits, with a mean value of 51.3 mg/L (da Silva Peixoto et al., 2020). In this way, the high values of nitrates in Brazil are referred mainly to septic tanks and sewage discharge and to a lesser extent to agriculture inputs and industrial wastes (da Silva Bellettini et al., 2019; da Silva Peixoto et al., 2020; Rezende et al., 2019; Suhogusoff et al., 2019). Besides, da Silva Peixoto et al. (2020) attributed this contamination to two different sources: (i) sewage discharge and (ii) low natural recharge from precipitation (da Silva Peixoto et al., 2020). In contrast, da Silva Bellettini et al. (2019) reported that the coal mining industry without proper wastewater treatment contributed to high nitrates concentration in the Carboniferous region (Brazil) (da Silva Bellettini et al., 2019).

Most of the sources are represented in blue colour, indicating no pollution in terms of nitrates, highlighting several areas of Mexico, Chile, Costa Rica, the USA, Colombia, and Argentina which have the maximum values within the WHO limits (points 1–16, Fig. 5). Nevertheless, in other regions located in Mexico, such as Mezquital and Toluca Valley, the mean values are in the range 2–36 mg/L; but more than 25% of the samples analyzed report values above 50 mg/L of NO_3^- (points 14, 16, 18 and 31, Fig. 5) (de Oca et al., 2019; Esteller et al., 2017).

Besides, in two regions located in Illinois (United States of America) (point 24, Fig. 5) and Bolivia (point 29, Fig. 5), although some measurements reflect a certain degree of contamination, less than 25% of the values exceed 50 ppm (Panno et al., 2019; Quino-Lima et al., 2020). Specifically, in the case of Bolivia, as illustrated in Fig. 5, although the average value fulfills the standard, some samples are above 200 mg/L, provoking a cause of great concern in this region (point 29, Fig. 5) (Quino-Lima et al., 2020). The authors attribute the contamination to human activities, such as insufficient treatment of urban wastes and intensive agriculture (Quino-Lima et al., 2020).

Regarding the main causes of nitrate pollution, Table 4 summarizes the different risks associated with the different activities analyzed. For example, in Argentina, human activities have affected some areas since some

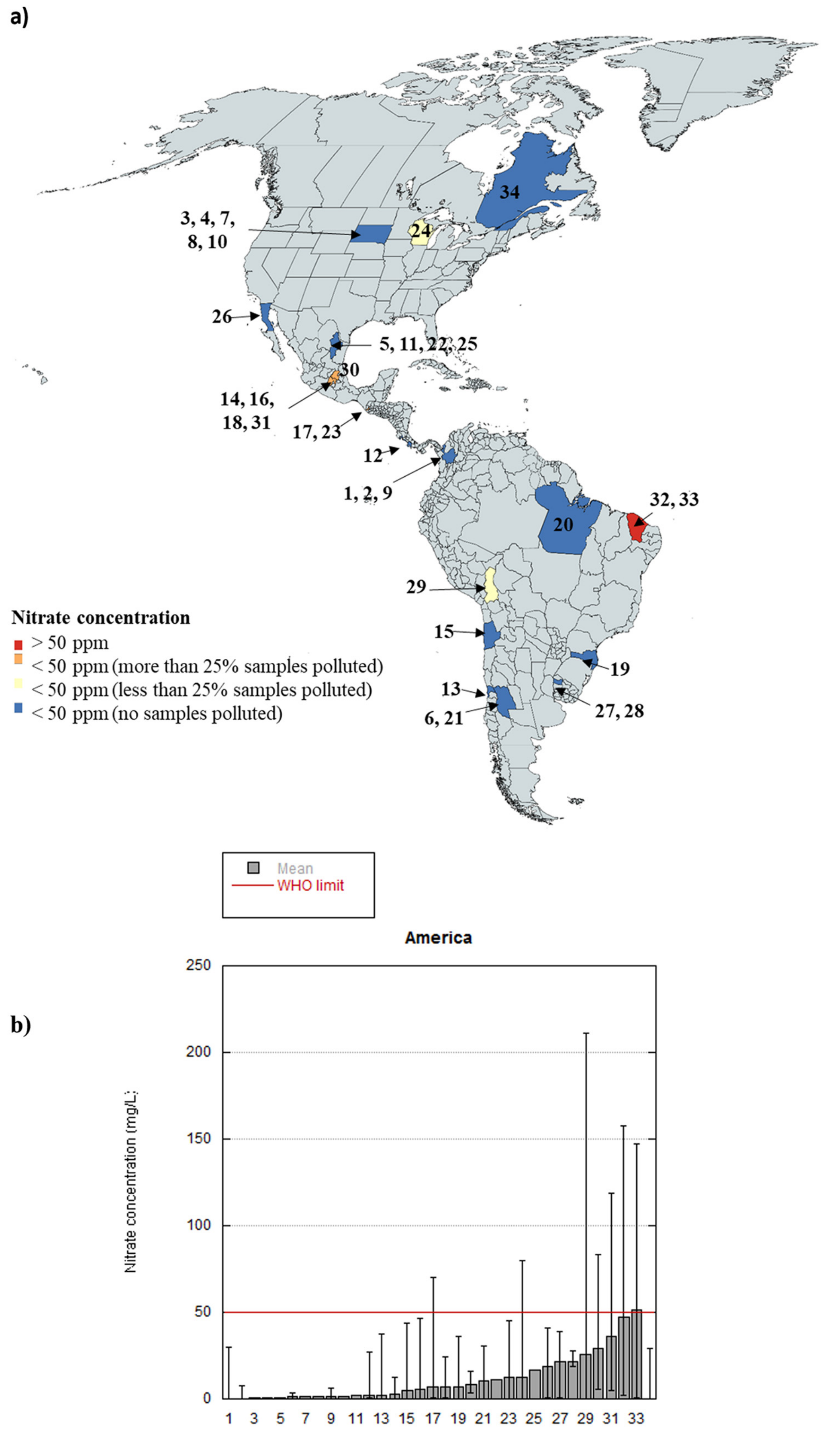

Fig. 5. a) Regions studied in America. b) Mean value and range of nitrate concentration in American groundwater bodies (Bucci et al., 2017; Carasek et al., 2020; Celestino et al., 2019; Daesslé et al., 2020; da Silva Peixoto et al., 2020; de Oca et al., 2019; Esteller et al., 2017; Gamazo et al., 2018; Gamboa et al., 2019; Gomez et al., 2019; Juliana et al., 2018; Loomer et al., 2019; Mendes et al., 2019; Ossa-Valencia and Betancur-Vargas, 2018; Panno et al., 2019; Quino-Lima et al., 2020; Sánchez-Gutiérrez et al., 2020; Swift Bird et al., 2020; Taucare et al., 2020; Torres-Martínez et al., 2020). Detailed references are incorporated in Table S3 of the Supplementary Information.

Table 4
Nitrate pollution in America.

	Septic tanks	Sewage	Agriculture	Landfills	Industrial
America	■	■	●	▲	■
Argentina	▲	■	■	▲	■
Bolivia	▲	■	●	▲	▲
Brazil	●	●	■	▲	■
Canada	▲	▲	●	▲	■
Chile	▲	▲	■	▲	▲
Colombia	■	■	■	■	■
Costa Rica	■	▲	▲	▲	▲
Guatemala	▲	■	■	▲	▲
Mexico	▲	●	●	▲	●
Uruguay	■	■	■	■	▲
USA	■	●	■	▲	▲

Risk of contamination: (▲) low, (■) medium, (●) high.

aquifers are increasing the groundwater abstraction rates and the return velocity of the sewage or the industrial discharge with poor treatment (Isla et al., 2018; Lupi et al., 2019). On the other hand, while in Chile, nowadays, agricultural practices are the major cause of concern for nitrates pollution (Sánchez-Gutiérrez et al., 2020), in Colombia, groundwater at the shallow levels present anthropogenic contamination from agricultural activities, livestock, poor management of solid waste, and the lack of a sewer system in certain regions (Ossa-Valencia and Betancur-Vargas, 2018). Finally, in Guatemala, the risk is associated with fertilizers and urban wastewater (Bucci et al., 2017).

In the specific case of the USA, Ward et al. (2018) point out that there are many private wells in the United States of America that are not regulated by the Environmental Protection Agency (EPA) (Ward et al., 2018) and consequently no pollutant areas are reported. In fact, these authors estimated that 2% of public-supply wells and 6% of private wells exceeded the maximum contaminant level (MCL) for nitrate in public drinking water supplies in the United States. MCL is situated as 10 $mg{\cdot}L^{-1}$ as nitrate-nitrogen, which is approximately equivalent to the WHO guideline of 50 $mg{\cdot}L^{-1}$. Moreover, an assessment carried out in the United States between 1991 and 2003 demonstrated that nitrate concentrations were highest in shallow groundwater beneath agricultural land use or areas with well-drained soils and oxic geochemical conditions. In contrast, lower values were found in deep groundwater when these water bodies were older since the anthropogenic sources had not already caused a significant impact. In this context, The United States Geological Survey (USGS), a scientific agency of the government, developed a robust model, based on nationwide data, to estimate the risk of nitrate contamination in shallow groundwater across the United States. This model integrates nitrogen inputs and aquifer vulnerability using Geographic Information System (GIS) technology ("Groundwater Quality", 2019). Nitrogen inputs include commercial fertilizer and manure application rates, atmospheric contributions, and population densities.

Aquifer vulnerability is represented by soil-drainage characteristics and the extent to which woodlands are interspersed with cropland. Fig. 6, extracted from the USGS, represents the areas with the highest risk of contamination from both natural and human-induced sources ("Groundwater Quality", 2019).

3.3. Europe

In the European continent, a total of 71 regions have been collected and discussed. Despite the mean value being under the WHO limit in many areas, 22 regions have maximum values above 50 ppm. In more detail, 16 of the studied regions have more than 25% of samples with NO_3^- values above 50 $mg{\cdot}L^{-1}$. Fig. 7 depicts the different areas studied (Fig. 7a) and

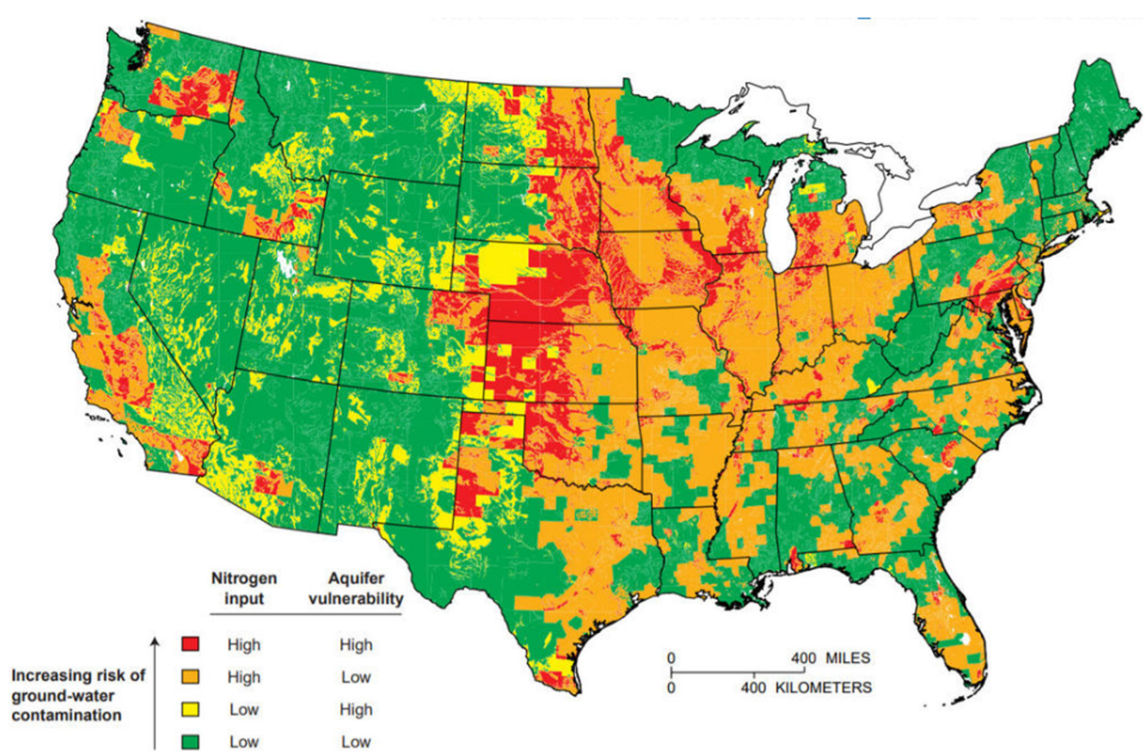

Fig. 6. Risk of nitrate pollution in groundwater aquifers in USA.
Adapted from ("Groundwater Quality", 2019).

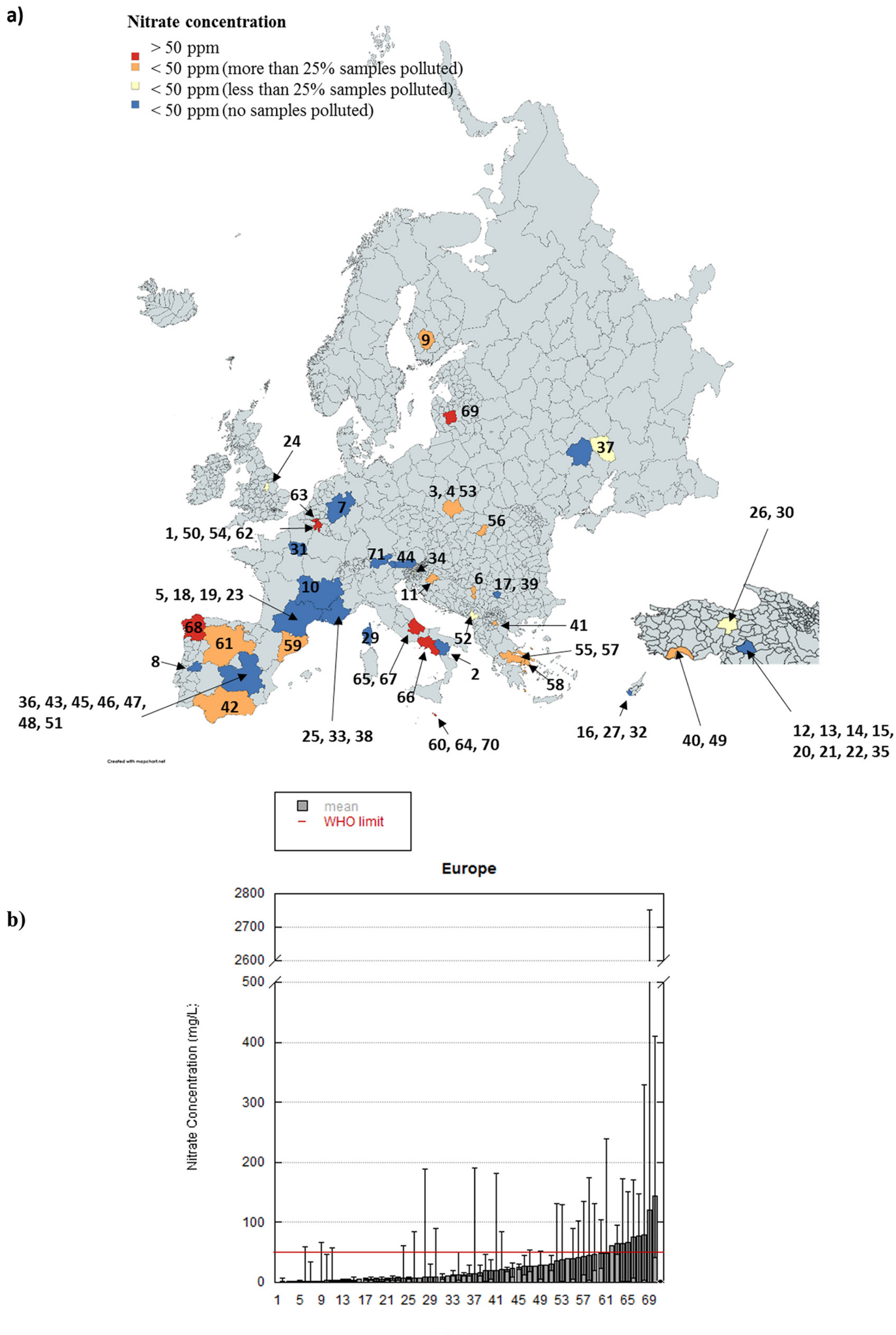

Fig. 7. a) Distribution of nitrate polluted zones in Europe. b) Nitrate concentration in European groundwater bodies (Ruiz-García et al., 2019; Barbieri et al., 2019; Bicalho et al., 2019; Blázquez-Pallí et al., 2019; Bojarczuk et al., 2019; Boudjana et al., 2019; Canora et al., 2019; Carvalho et al., 2019; Charizopoulos et al., 2018; Ćuk et al., 2020; di Lorenzo et al., 2012; Dippong et al., 2019; Erostate et al., 2018; Gil-Márquez et al., 2019; Giménez-Forcada et al., 2017; Gromadzka et al., 2015; Heaton et al., 2012; Karakuş, 2019; Lorette et al., 2018; Majkić-Dursun et al., 2018; Mirčovski et al., 2018; Moratalla et al., 2009; Mostaza-Colado et al., 2018; Nikolenko et al., 2019; Ogrinc et al., 2019; Papazotos et al., 2019; Pedretti et al., 2019; Quenet et al., 2019; Rodriguez et al., 2020; Rufino et al., 2019; Sedlazeck et al., 2017; Slavinskienė and Jurevičius, 2016; Smedley et al., 2018; Strauhal et al., 2016; Tran et al., 2020; Tzoraki et al., 2018; Ujević Bošnjak et al., 2012; Varol and Şekerci, 2018; Vystavna et al., 2015; Yetiş et al., 2019). References are detailed in Table S5 of the Supplementary Information.

the mean, maximum and minimum values of each point (Fig. 7b). As it is drawn in the mentioned figure, the values of Italy (points 65–67, Fig. 7), Malta (points 60, 64 and 70, Fig. 7), Lithuania (point 69, Fig. 7), Belgium (points 7 and 62, Fig. 7) and Spain (point 68, Fig. 7), contain high concentration of nitrates, with a mean value between 72 ± 6 mg/L, 104 ± 41 mg/L, 120 mg/L, 62 ± 3 mg/L, and 80 mg/L, respectively (Boudjana et al., 2019; di Lorenzo et al., 2012; Heaton et al., 2012; Nikolenko et al., 2019; Rodriguez et al., 2020; Slavinskienė and Jurevičius, 2016). In addition, two additional regions in Duero (Spain) (point 61, Fig. 7) and Malta (point 70, Fig. 7) also have maximum concentration values of nitrates of high concern since they exceed 200 ppm. In the case of Italy, the major concerns in terms of activities that cause higher nitrate concentration are synthetic fertilizers, mainly ammonium salts, and runoff processes. Nevertheless, groundwater in Italy is often locally affected by denitrification processes that reduce the nitrate content, offering values consistent with legal standards. Regarding the high values reported, Malta mineral fertilizers, animal and human sewage waste, and soil cultivation are the main sources of contamination (Heaton et al., 2012). Contrarily, Lithuania has reported the maximum worldwide value of nitrate concentration, 2753 mg/L (point 69, Fig. 7) (Slavinskienė and Jurevičius, 2016). This high concentration was measured in a groundwater body close to open hydrogeological systems, where some landfills are located (Slavinskienė and Jurevičius, 2016).

In Belgium, the concerning values of nitrates are attributed to poor treatment of household sewage and manure (Nikolenko et al., 2019). In this line, a recent report from the European Commission to the council and the European parliament claimed that a high percentage of groundwater monitored currently shows high NO_3^- concentration, above the maximum 50 ppm, in Malta, Germany, Luxemburg, Spain, Portugal, and Belgium (Flanders region).

In contrast, points from 1 to 61 present mean values below 50 ppm. In this sense, some countries, such as Finland (point 9, Fig. 7), Croatia (point 11, Fig. 7), Serbia (points 17 and 39, Fig. 7), Spain (point 29, Fig. 7), Macedonia (point 41, Fig. 7), Poland (point 53, Fig. 7), Greece (points 55, 57 and 58, Fig. 7), and Romania (point 56, Fig. 7) have mean values below 50 mg/L (Bojarczuk et al., 2019; Charizopoulos et al., 2018; Ćuk et al., 2020; Dippong et al., 2019; Mirčovski et al., 2018; Ogrinc et al., 2019; Papazotos et al., 2019; Pedretti et al., 2019; Ujević Bošnjak et al., 2012). Nevertheless, there are areas of special concern where more than a quarter of the collected samples are polluted. Additionally, Turkey (point 49, Fig. 7), Ukraine (point 37, Fig. 7), United Kingdom (point 24, Fig. 7), Slovenia (point 34, Fig. 7) and Albania (point 52, Fig. 7) report samples with nitrate concentration above 50 mg/L (Barbieri et al., 2015; Ogrinc et al., 2019; Smedley et al., 2018; Varol and Şekerci, 2018; Vystavna et al., 2015).

As in previous continents, Table 5 shows the risk of nitrate pollution due to the five different activities analyzed in this work. In Europe, the primary source of nitrates is leaching of fertilizers, the excess of livestock, and municipal human wastes (Heaton et al., 2012; Rodriguez et al., 2020). For example, in Spain, France, and Macedonia, intensive agriculture using NPK fertilizers and the manure used as fertilizer are the main sources of high nitrate levels (Giménez-Forcada et al., 2017; Mirčovski et al., 2018; Nofal et al., 2019; Rodriguez et al., 2020). A similar situation is found in Greece, where the upper values of nitrates are related to intensive agriculture and nitrogen fertilizers (Vasileiou et al., 2019). Regarding Romania, high concentrations have been found in areas with pig farms and grapevine cultures (Dippong et al., 2019). In this way, many authors pointed to agricultural activity, poor sewage treatment, and manure as the main anthropogenic sources of pollution in Turkey, Ukraine, the United Kingdom, Slovenia, and Albania (Barbieri et al., 2015; Ogrinc et al., 2019; Smedley et al., 2018; Varol and Şekerci, 2018; Vystavna et al., 2015).

While the discharge of municipal sewage and organic-mineral fertilization of agricultural land are the main contributors to the changes in the quality of shallow groundwater in Poland (Bojarczuk et al., 2019), in Croatia and Cyprus the contamination of groundwater excluded any urban or industrial source but underlined the risk of agriculture inputs (Nikolaou et al., 2020; Tzoraki et al., 2018; Ujević Bošnjak et al., 2012). Finally, previous works focused on Serbia, remarked some anthropogenic activities of special concern, nitrogen-based fertilizers and manure, and to a larger extent, mixing of sewage and septic tank effluents (Majkić-Dursun et al., 2019, 2018).

In the line with the conclusions reported in previous works and collected through this work, the European Environment Agency (EEA) has developed a map highlighting the percentage of groundwater body areas that present bad chemical status due to nitrate and total nitrogen input from organic and inorganic fertilizers (EEA, 2012). Fig. 8 shows the areas and the risk associated. Northern Europe countries (Norway, Sweden, Estonia, Latvia, and Lithuania) present good quality of groundwater in terms of nitrates presence. However, in general, regions of central Europe (France, Germany, Poland among others) have a remarkable percentage of areas with a risk of up to 10% of groundwater with poor chemical status due to nitrates. Moreover, some areas in the Czech Republic, Slovenia, Netherlands, and Spain present a percentage between 70% and 90% classified as bad quality, which is considered a concerning problem to be solved in the near future (EEA, 2012).

Table 5
Pollution sources in Europe.

	Septic tanks	Sewage	Agriculture	Landfills	Industrial
Europe	▲	●	●	■	■
Albania	▲	●	●	▲	▲
Belgium	▲	■	●	▲	▲
Croatia	▲	▲	■	▲	▲
Cyprus	▲	▲	■	▲	▲
Finland	▲	▲	■	▲	▲
France	▲	▲	■	▲	▲
Germany	▲	▲	■	▲	▲
Greece	▲	▲	●	▲	▲
Italy	▲	●	●	▲	■
Lithuania	▲	▲	■	●	▲
Macedonia	▲	■	●	▲	■
Malta	▲	▲	●	●	▲
Poland	▲	▲	●	▲	■
Portugal	▲	▲	■	▲	▲
Romania	▲	▲	●	▲	▲
Serbia	▲	▲	■	▲	▲
Slovenia	▲	■	●	▲	▲
Spain	▲	■	●	▲	■
Turkey	▲	●	●	▲	■
Ukraine	▲	●	●	▲	▲
United Kingdom	▲	■	●	▲	▲

Risk of contamination: (▲) low, (■) medium, (●) high.

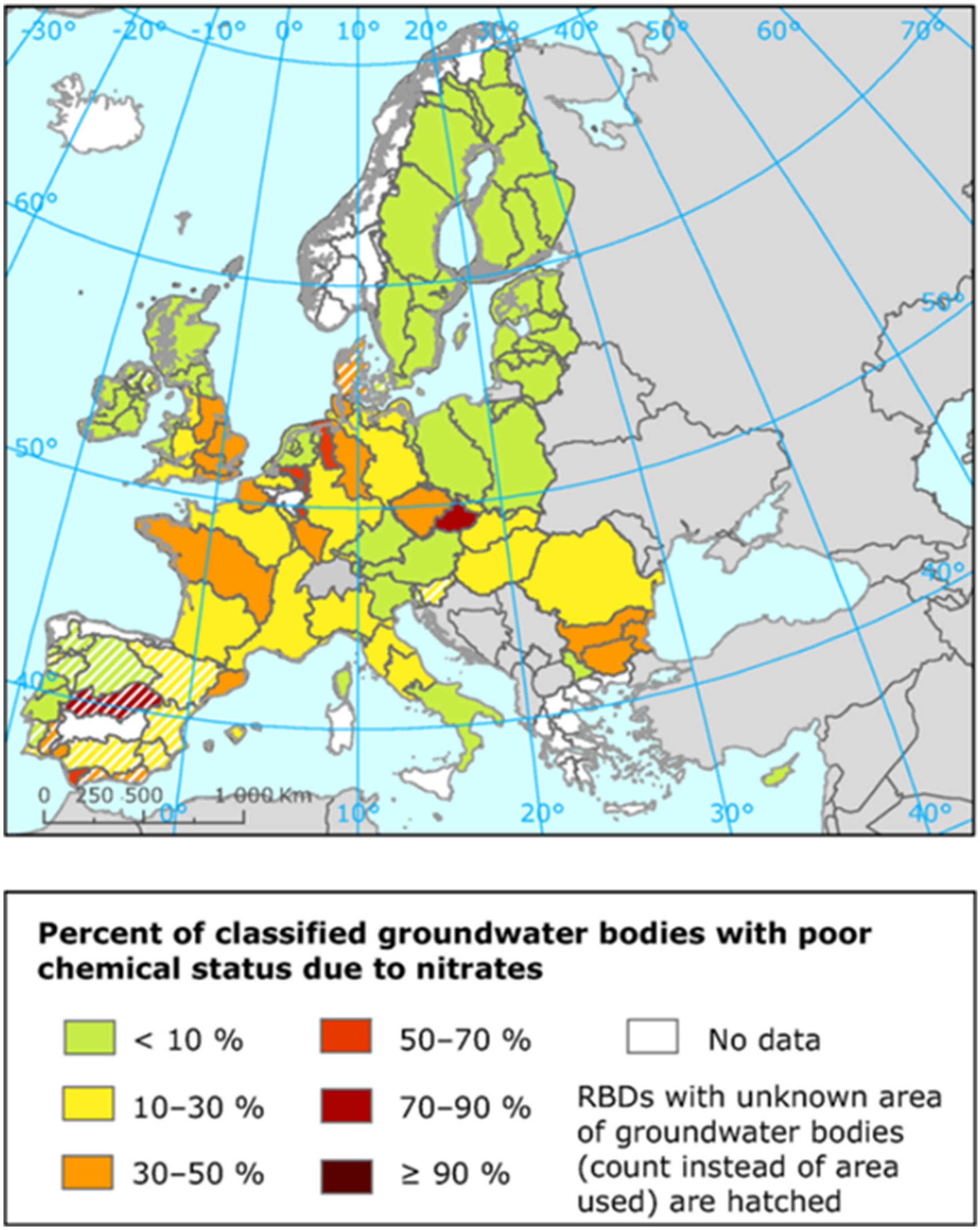

Fig. 8. Map developed by EEA classifying the quality of groundwater (EEA, 2012).

3.4. Africa

Finally, this section debates the African continent, which presents the most concerning situation since one-third of the data reported mean values of nitrate concentration in groundwater above the WHO threshold. In this continent, 94 points were collected and analyzed, as illustrated in Fig. 9a; the data reported so far in 15 sites did not allow to obtain average values (Fig. 9b). In this regard, Africa has several countries in which groundwater has been contaminated by nitrate, with mean values ranging from 52 to 776 mg/L (see Fig. 9). For example, in some regions in the Democratic Republic of the Congo, Mozambique, and Zimbabwe, mean values are between 189 and 775 mg/L (points 68–81, Fig. 9), 89–172 mg/L (points 64 and 67, Fig. 9), and 72 mg/L (point 59, Fig. 9) respectively (Barbieri et al., 2019; Kapembo et al., 2016; Muzenda et al., 2019). Previous works focused on Mozambique and Zimbabwe mainly attributed the high mean values to filtration and leaks from septic tanks and latrines (Arsénio et al., 2018; Barbieri et al., 2019; Muzenda et al., 2019). In this sense, but in a more dramatic way, the Democratic Republic of the Congo reports the most concerning situation worldwide, since there is poor sanitation and people make use of contaminated water for irrigation, domestic and drinking purposes. For example, in the commune of Bumbu (Democratic Republic of the Congo), above 75% of people have no access to safe water, and the water sources are shallow wells that are located near latrines (Kapembo et al., 2016).

Moreover, Angola (point 63, Fig. 9) and Grand Yaéré, Cameroon (point 51, Fig. 9) reported average values of 83 and 50 ppm achieving maximum values of 132 and 646 mg/L respectively (Bello et al., 2019; Silva et al., 2017). In

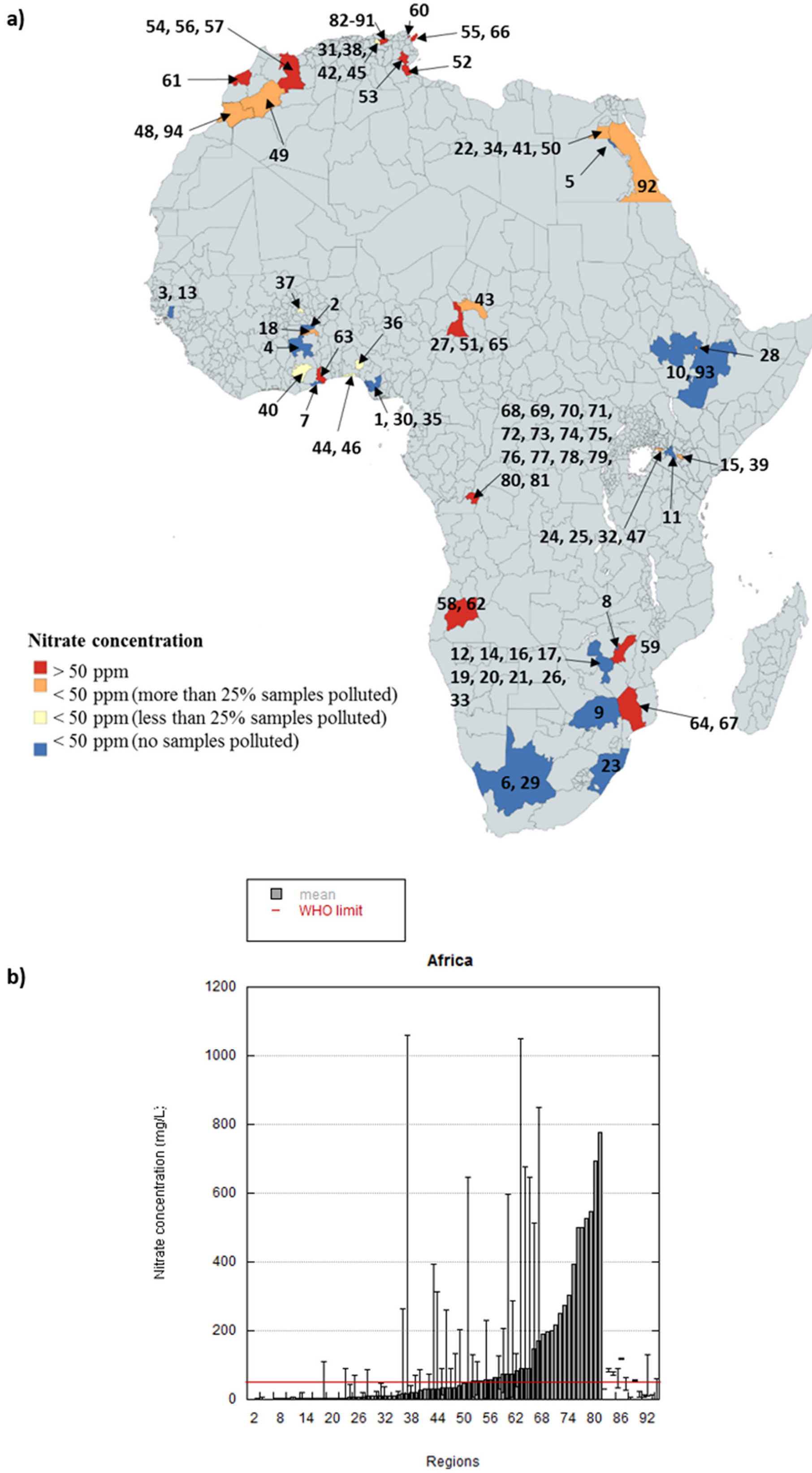

Fig. 9. a) Regions studied in Africa. b) Nitrate concentration in African groundwater bodies (Abderamane et al., 2013; Aladejana et al., 2020; Anim-Gyampo et al., 2018; Ashun and Bansah, 2017; Awomeso et al., 2020; Bello et al., 2019; Benkaddour et al., 2020; Benmarce and Khanchoul, 2019; Beyene et al., 2019; Bon et al., 2020; Bouteraa et al., 2019; Bretzler et al., 2017; Danni et al., 2019; Egbi et al., 2019; el Gammal and Ibrahim, 2017; el Ghali et al., 2020; Elumalai et al., 2019; Erdogan et al., 2020; Ferchichi et al., 2018; Ferrante et al., 2018; Gevera and Mouri, 2018; Kapembo et al., 2016; Karroum et al., 2017; Kawo and Karuppannan, 2018; Loh et al., 2020; Masocha et al., 2019; Melki et al., 2019; Mouassa et al., 2020; Mudzielwana et al., 2020; Mukanga et al., 2016; Muzenda et al., 2019; Ncibi et al., 2020; Nyilitya et al., 2020; Opoku et al., 2020; Owamah, 2020; Re et al., 2017; Silva et al., 2017; Snousy et al., 2020; Sunkari et al., 2019; Tolera et al., 2020; Zaki et al., 2019; Zango et al., 2019). Detailed references appear in Table S7 of the Supplementary Information.

these two countries, the NO_3^- pollution is attributed to latrines and livestock waste (Bello et al., 2019; Silva et al., 2017). Firstly, Silva et al. (2017) reported the presence of nitrate probably due to the direct discharge of nitrogen-rich and untreated domestic effluents in the high-porosity soil in Angola. Supporting this argument, in Cameroon, Bello et al. (2019) found NO_3^- evolution independently of K^+, thus, excluding the use of agricultural fertilizers as potential source and therefore, attributing the presence of nitrates in this region to oxidation-reduction reactions of organic matter associated with septic-tank effluents, animal or plant production.

Morocco presents nitrate concentrations between 33 and 74 mg/L (points 48 and 61, Fig. 9) (Benkaddour et al., 2020) and the main source of nitrate pollution, is similar to the different areas studied in Europe; the overuse of fertilizers, pesticides, manure, and intensive agriculture and irrigation (Benkaddour et al., 2020).

In the case of Ghana, South Tongu and Ada East region (point 63, Fig. 9) and Tunisia (points 52, 53, 55, 60 and 66, Fig. 9) the mean values are 89 mg/L and 100 ± 38 mg/L, respectively (Egbi et al., 2019; Ferchichi et al., 2018; Melki et al., 2019; Ncibi et al., 2020; Re et al., 2017) because of the intensive agricultural activities and discharge of domestic and industrial sewage (Egbi et al., 2019; Ferchichi et al., 2018; Re et al., 2017).

Other countries with contaminated sites in the African continent are South Africa (point 23, Fig. 9), Kenya (points 25, 39 and 47, Fig. 9), Ethiopia (point 28, Fig. 9), Nigeria (points 36, 44 and 46, Fig. 9) and Chad (point 43, Fig. 9) (Abderamane et al., 2013; Aladejana et al., 2020; Ashun and Bansah, 2017; Awomeso et al., 2020; Elumalai et al., 2019; Kawo and Karuppannan, 2018; Nyilitya et al., 2020).

In the specific case of Algeria, there are two areas with a mean value under 50 ppm, but some samples contain nitrate concentrations higher than this value (points 42 and 45, Fig. 9) (Bouteraa et al., 2019; Mouassa et al., 2020). Nevertheless, several points only reported mean values, which are in most cases above 50 mg/L (points 82–91, Fig. 9) (Benmarce and Khanchoul, 2019). The presence of nitrates was attributed as in Kenya to the intensive agriculture and to livestock activity in this area (Benmarce and Khanchoul, 2019; Mouassa et al., 2020; Nyilitya et al., 2020). In the same line, Kawo and Karuppannan (2018) pointed out the presence on nitrates in central Ethiopia due to agricultural inputs, waste disposal site and several industries (Kawo and Karuppannan, 2018).

Finally, in the case of Nigeria, NO_3^- comes from weathering of bedrocks, leachate from septic tanks and dumpsites, runoff of materials, hardness, nutrients from agricultural lands, and chlorine pollution (Awomeso et al., 2020). In summary, Table 6 collects the different activities studied in this work as a function of the risk in each country.

Table 6
Nitrate risk pollution in Africa.

	Septic tanks	Sewage	Agriculture	Landfills	Industrial
Africa	●	●	●	●	■
Algeria	▲	■	●	■	■
Angola	●	●	■	●	■
Cameroon	●	■	●	■	■
Chad	■	■	■	■	▲
Democratic Republic of the Congo	●	■	●	●	■
Egypt	▲	■	■	▲	▲
Ethiopia	■	■	■	▲	■
Ghana	■	●	■	▲	●
Kenya	■	■	■	▲	▲
Morocco	▲	●	●	▲	●
Mozambique	●	■	■	■	■
Nigeria	■	■	■	▲	■
South Africa	■	■	■	▲	▲
Tunisia	▲	■	●	▲	●
Zimbabwe	●	■	■	■	▲

Risk of contamination: (▲) low, (■) medium, (●) high.

3.5. Summarize of nitrate pollution worldwide

In view of the analysis of nitrate pollution, continent-by-continent, it can be concluded that nitrate pollution is a global problem. In this context, if we pay attention to Fig. 10, and according to the analysis made country-by-country, most of the regions studied in the Mediterranean area have worried values and therefore is one of the most polluted areas worldwide in terms of nitrate presence in groundwaters. To have a global vision and to appreciate this fact, Fig. 10 represents the regions studied in Europe and North Africa, and as can be seen, regarding NO_3^- concentration, most of the areas collected show orange or red colour implying that more than 25% of the samples report values above 50 ppm (orange) or mean values higher than 50 $mg{\cdot}L^{-1}$ (red), which is the value established by WHO to determine the polluted areas.

Besides, apart from the five categories of contamination studied through this manuscript, different catastrophic events that occurred during the past decades have changed the presence of nitrates in groundwater in the affected areas. For example, the groundwater around the Union Carbide factory in Bhopal has been contaminated with excessive levels of nitrate, chloride, and heavy metals ("Water Contamination Crisis - Bhopal's Second Disaster - The Bhopal medical appeal", 2021). In the case of Beirut, in 2020, 2750 tons of ammonium nitrate inadequate storage caused an explosion creating an important number of pollutants being released to the environment. Compared with a similar incident in Tianjin (China), cofferdams and cement encasements should be built to prevent nitrates leakage to soil and groundwater, or nitrate contamination is likely to be distributed in groundwater in five years (ur Rehman et al., 2021). Regarding the Chernobyl accident, two regions of Belarus assessed the hazard of nitrates since two regions reported similar radiation values but, the incidence of pediatric thyroid cancer was different, being the main difference the nitrate contamination in drinking water, 40 vs. 185 ppm (Drozd et al., 2018).

Regarding the existing regulation of nitrates worldwide, the world health organization developed a report in 2015 using data from members of the WHO International Network of Drinking-water Regulators, WHO regional and country office contacts, through internet searches or purchased from the relevant standards organizations considering 104 countries and territories. Nitrates was one of only three inorganic parameters with a value set by all countries and territories. For example, at the European level, there exists a Directive (2006/18/EC) based on the protection of groundwater against pollution and deterioration, fixing as quality standards a maximum of 50 $mg{\cdot}L^{-1}$ of nitrates concentration. Besides, in 1991, the EU introduced the Nitrates Directive (91/676/EEC), which aimed to reduce water pollution caused or induced by nitrate from agricultural sources. This Directive also defines Vulnerable Nitrate Zones where it is necessary to establish and implement action programs to reduce water pollution from nitrogen compounds. In the case of the United States of America, the Environmental Protection Agency (EPA) has established a limit for nitrates and nitrites in drinking water (10 parts per million (ppm) for nitrates and 1 ppm for nitrites), and the Food and Drug Administration (FDA) has recommended limits for nitrates and nitrites in bottled water (same thresholds as the EPA) and foodstuffs (sodium nitrate lower than 500 ppm in the finished meat product, sodium nitrite below 200 ppm in the finished meat product and potassium nitrate as curing agent in the processing of cod roe in an amount less than 200 ppm in the finished roe).

This manuscript contains in-depth analysis developed using more than 272 regions worldwide, despite the focus on specific areas, which could not necessarily apply to more extensive zones, allows us to conclude: i) the

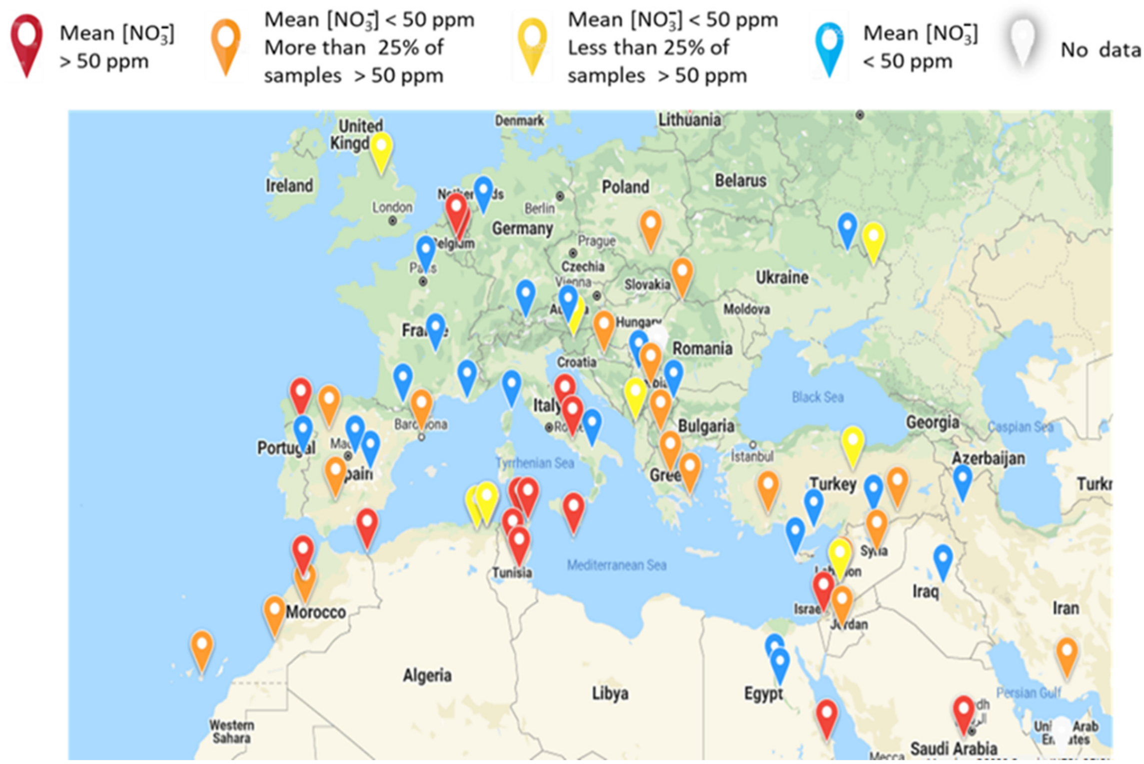

Fig. 10. Location and nitrate concentration in groundwater bodies in the Mediterranean area. (Abboud, 2018; Abou Zakhem et al., 2017; Abou Zakhem and Hafez, 2015; Abualnaeem et al., 2018; Alslaibi et al., 2017; Ruiz-García et al., 2019; Asmael et al., 2015; Benmarce and Khanchoul, 2019; Bicalho et al., 2019; Blázquez-Pallí et al., 2019; Bojarczuk et al., 2019; Boudjana et al., 2019; Bouteraa et al., 2019; Canora et al., 2019; Carvalho et al., 2019; Charizopoulos et al., 2018; Ćuk et al., 2020; Danni et al., 2019; di Lorenzo et al., 2012; Dippong et al., 2019; el Gammal and Ibrahim, 2017; el Ghali et al., 2020; Erostate et al., 2018; Ferchichi et al., 2018; Gil-Márquez et al., 2019; Giménez-Forcada et al., 2017; Gorgij et al., 2019; Heaton et al., 2012; Karakuş, 2019; Karroum et al., 2017; Kattan, 2018; Lorette et al., 2018; Majkić-Dursun et al., 2018; Melki et al., 2019; Mirčovski et al., 2018; Moratalla et al., 2009; Mostaza-Colado et al., 2018; Mouassa et al., 2020; Ncibi et al., 2020; Nikolenko et al., 2019; Papazotos et al., 2019; Quenet et al., 2019; Re et al., 2017; Rezaei et al., 2017; Rodriguez et al., 2020; Rufino et al., 2019; Sedlazeck et al., 2017; Smedley et al., 2018; Snousy et al., 2020; Strauhal et al., 2016; Tran et al., 2020; Tzoraki et al., 2018; Ujević Bošnjak et al., 2012; Varol and Şekerci, 2018; Vystavna et al., 2015; Yetiş et al., 2019; Zaki et al., 2019).

primary anthropogenic sources of nitrate are the use of fertilizers and the discharge of non-treated domestic and industrial wastes, ii) at many points, the nitrate contamination is associated with other contamination by fluorine, sulphates, potash, phosphates and some heavy metals such as zinc, cadmium or arsenic, and iii) nitrate pollution is mainly concentrated in the Mediterranean area and some regions of South Asia, where agriculture is an essential source of income but also the presence of NO_3^- is a big concern in some developing African countries, due to the lack of adequate sanitation.

Also, this pollution, far from being solved, shows an increasing trend that demands drastic and urgent solutions to prevent this problem. Thus, immediate actions are required before natural water bodies are entirely spoiled, especially in developing countries with water shortages. Although there are different sources of nitrates pollution in groundwater bodies, the challenge of water remediation can be facilitated by using similar technologies that enable the transfer of knowledge and inter-site adaptation.

Table 7
Different technologies available to remove pollutants present in water bodies.

Technology	Substances to be removed	Ref
Precipitation and coagulation	Phosphorous, fluoride, arsenic, ferrocyanide, and heavy metals.	(Ballinas et al., 2004; Sharma and Bhattacharya, 2017)
Softening	Hardness ions	(Sharma and Bhattacharya, 2017)
Distillation	Toxic chemicals, heavy metals, bacteria, viruses or parasites.	(Sharma and Bhattacharya, 2017)
Adsorption	Eliminate compounds that add either colour, taste, or odor to water, such as VOCs, chlorine, heavy metals, organics.	(Sharma and Bhattacharya, 2017)
Ion exchange	Anions or cations based on resin type.	(Sharma and Bhattacharya, 2017)
Reverse osmosis	Salts, pesticides, microbes.	(Sharma and Bhattacharya, 2017; Yang et al., 2019)
Electrodialysis	Dissolved ionic particles.	(Akhter et al., 2018; Sharma and Bhattacharya, 2017)
Catalytic processes	Organic compounds, formic acid or nitrates.	(Guate et al., 2019; Rezvani et al., 2019; Sharma and Bhattacharya, 2017; Tokazhanov et al., 2020)
Bioremediation	Heavy metals, sediments, pathogens, or dissolved organic chemicals.	(Kozyatnyk and Klymenko, 2016; Sharma and Bhattacharya, 2017)
Magnetic separation	Oil, suspended solids and some ions with difficulties to coagulate.	(Sharma and Bhattacharya, 2017)
Disinfection (UV or chlorine)	Bacteria and viruses.	(Sharma and Bhattacharya, 2017)
Active carbon filtration	Dissolved organic carbon	(Sharma and Bhattacharya, 2017)
Filtration (UF, NF, gravel, sand…)	Suspended solids, dyes or organic matter	(Rashid et al., 2021)
Oxidation (mechanical, thermal, with ozone)	Colour, odor, organic compounds, and inorganic compounds	(Radu and Racoviteanu, 2021)
Clarification	Total suspended solids (TSS)	(Ortiz et al., 2015)
Aeration/air stripping	Transferring of volatile components of a liquid into an air flow	(Radu and Racoviteanu, 2021)

4. Alternatives to nitrate removal

As mentioned above, apart from nitrates, groundwater could contain different ions which must be removed before use. In this sense, nowadays, numerous water treatment technologies have been developed, according to the pollutants, infrastructure, affordability, and acceptability (Hosseini et al., 2016; Sharma and Bhattacharya, 2017). In fact, the most common technologies to remove pollutants are summarized in Table 7.

Currently, there are numerous technologies implemented at the industrial scale to remove different pollutants from water, and particularly, to treat surface waters. But, there is a scarce number of demonstration plants in the world dedicated to groundwater treatment. On this basis, Table 8 summarizes the facilities dedicated to this purpose as well as their daily capacity and the pollutants removed by these techniques. If we take as a basis, a daily treatment flow greater than 1000 m^3, we find that there are only 13 plants implemented in the world: five in the USA, five in Italy, and one in Australia, India and Poland, respectively. Regarding the processes carried out in these plants, reverse osmosis (RO), filtration, and ultrafiltration (UF) are the most popular technologies employed in the United States of America. Besides, Italy has numerous plants located all over the country, able to remove a wide variety of pollutants. On the other hand, iron, manganese, and inorganic compounds stand out among the contaminants that are also removed. Also, the pilot situated in Australia has installed thermal

Table 8
Characteristics of groundwater treatment plants.

		Region	Process	Capacity (m^3/day)	Pollutants removed	Opening year	Ref
Asia	India		biofilm reactor, flocculation, precipitation, deferrization, lamella separator, gravel filter, filtration activated carbon, sludge treatment, thickener, chamber filter press	528	BTEX, MIBK, chlorinated compounds, phenols, nitroaromatics, anilines, pesticides, hydrocarbons	2011	("Züblin Umwelttechnik GmbH - Groundwater Treatment Plant", 2021)
		New Delhi	Aeration, lime softening, coagulation, flocculation, clarification, pH adjust, UF, disinfection (UV or chlorine)	4536	bicarbonate hardness removal, suspended particles, colloids and harmful microorganisms	n. a.	(Pillai et al., 2019)
	Saudi Arabia	Buraydah, Qassim region	Coagulation, UF, Sand filtration, RO		TDS, Fe, Mn	n. a.	(Haider, 2017)
America	Canada	White Rock	Pre-oxidation with ozone, filtration, adsorption		Mn, As	2019	("Water Treatment Plant | White Rock, BC", 2021)
	Ecuador		RO	336		2013	("Industrial Reverse Osmosis Equipment 87,000 GPD - Ecuador - Pure Aqua, Inc.", 2020)
	Haiti		RO, activated carbon	216	Salts	n. a.	("Groundwater Treatment Plant 57000 GPD - Haiti - Pure Aqua, Inc.", 2021)
	USA	Jordan, Utah	RO, UV	26,496	TDS and Chemical impurities	2011	("Southwest Groundwater Treatment Plant - Flatiron", 2021)
		Lary Lane, Exeter NH	Filtration, chemical compounds for co-precipitation, pH adjustment and disinfection	5904	Fe, Mn, As	2015	("Lary Lane Ground Water Treatment Plant | Town of Exeter New Hampshire Official Website", 2020)
		Jacksonville (North Carolina)	NF, bio scrubbing system, disinfection, pH adjust	15,142		2011	("City Water Plant | Jacksonville, NC - Official Website", 2021)
		Scottsdale (Arizona)	RO, airstripping technology		Trichloroethylene	In construction	("City of Scottsdale - City Construction Projects - Thomas Groundwater Treatment Facility and Improvements to the Central Groundwater Treatment Facility", 2021)
		Santa Mónica (California)	Pretreatment, RO, aeration	32,160	Mehtyl tert-butyl ether	2010	("Santa Monica Public Works - Santa Monica Water Treatment Plant", 2021)
		Davie (Florida)	Ion exchange RO, aeration, disinfection	37,848		n. a. n. a.	("Water Treatment | Davie, FL", 2020)
		Orrington (Maine)	UF, granular activated carbon	312	Hg	2012	("Groundwater Treatment Plant | beyondholtrachem", 2021)
Europe	Italy	Porto Marghera	Filtration, activated carbon	1320	Heavy metals, organic chlorinated compounds and aromatic compounds	n. a.	
		Porto Torres	Physico-chemical treatment, filtration, activated carbon	1200	Heavy metals, organic chlorinated compounds and aromatic compounds	n. a.	
		Assemini	Ion exchange	1320	Heavy metals, organic chlorinated compounds and aromatic compounds	n. a.	("Trattamento Acque di Falda", 2021)
		Portovesme	Physico-chemical treatment, activated carbon	9600	Heavy metals, inorganic compounds and aromatic compounds	n. a.	
		San Gavino Monreale	Physico-chemical treatment, activated carbon	864	Heavy metals, organic compounds and inorganic compounds (sulphates)	n. a.	
		Porto Torres	Mechanical oxidation, UF	4800		n. a.	
	Poland	Drzenin	Aeration, filtration	1080	Fe, Mn	1977	(Jakubaszek, 2019)
Oceania	Australia	Botany	Air stripping, thermal oxidizer, filtration, biological aerated filters, granular activated carbon, filtration, RO	6000	chlorinated hydrocarbons	2006	(Orica, 2020)

oxidizers within other processes and the Botany plant removes chlorinated hydrocarbons (Orica, 2020). The Orrington plant (Maine, United States of America), although it has a small daily treatment capacity, is worth to mention because the main element that is removed is mercury ("Groundwater Treatment Plant | beyondholtrachem", 2021).

Among the different technologies available to remove nitrates, no one is fully implemented or stands out clearly from the others. Table 9 summarizes the main advantages and drawbacks of the technologies existing in each type of approach. In this way, despite the scarcity of data regarding nitrate removal values, great efforts are being made at the laboratory level to find out the optimal figures of merit for each technology with associated strengths and weaknesses.

There are two foremost approaches, i) the separation of nitrates from the water (reverse osmosis, ion exchange, and electrodialysis) and ii) the transformation of nitrates into harmless nitrogen gas (biological denitrification, and catalytic methods). Regarding the first alternative, the separation requires a second step to remove, concentrate or neutralize the target compounds.

In the case of reverse osmosis, the membranes that are commonly employed are polyamide and cellulose triacetate (Hosseini et al., 2016). Schoeman and Steyn (2003) achieved, approximately, 98% nitrate removal using this membrane technology (Schoeman and Steyn, 2003). Contrarily, Richards et al. (2010), studied the influence of pH to remove nitrates concluding that this parameter does not affect the retention of nitrates but suffers a decrease in the presence of Na^+ due to the screening effects (Richards et al., 2010). Regarding the materials employed in ion exchange, Purolite A 520E has been considered the most effective ion exchange resin to remove nitrates (Samatya et al., 2006).

Membrane technology, along with ion exchange, has less efficiency than the catalytic process in the degradation of pollutants but has lower costs, automatization is more accessible, and has fewer control needs. For its part, adsorption has been analyzed by several authors and the different adsorbents studied are carbon-based sorbents, natural sorbents (i.e. zeolite or clays), agricultural wastes (i.e. sugar bagasse), industrial wastes (i.e. red mud), biosorbents (i.e. bamboo power) or miscellaneous sorbents (i.e. double layered hydroxides, silica or alumina). In this sense, double layered hydroxides or modified chitosan report higher uptake of nitrate than conventional adsorbents such as carbon-based sorbents (carbon nanotubes, activated carbon) or natural sorbents (clay, zeolite) (Bhatnagar and Sillanpää, 2011). Soías-Viciana et al. (2008) removed nitrates from water using calcinated hydrotalcite-type compounds. On the other hand, Chatterjee et al. (2009) studied chitosan as adsorbent to remove nitrates. They achieved an adsorption capacity of 104 mg/g using crosslinked chitosan beans while the reported adsorption capacity of normal chitosan beans is of 90.7 mg/g (Chatterjee et al., 2009). In the case of electrodialysis, Aliaskari and Schäfer (2020) studied this process to remove salinity, nitrates, fluoride and arsenic. They concluded that the removal of contaminants followed the order, nitrate = salinity > fluoride > arsenic. These authors, obtained a high nitrate removal with low electrical potential while arsenic

Table 9
Advantages and drawbacks of the main technologies for nitrates removal.

	Technology	Advantages	Drawbacks	Ref
Separation	Reverse osmosis	- Compact equipment - Continuous operation is possible - Not necessary post-treatments	- Fouling (to reduce it, sulfuric acid and sodium hexametaphosphate)	(Archna et al., 2012; Häyrynen et al., 2009; Schoeman and Steyn, 2003; Sharma and Bhattacharya, 2017; Tokazhanov et al., 2020)
	Ion exchange	- Simplicity - Effectiveness - Selectivity - Recovery - Relatively low cost	- Lower affinity of resins to nitrates with respect to sulphates	(Kabay et al., 2007a, 2007b; Primo et al., 2009; Samatya et al., 2007, 2006; Sharma and Bhattacharya, 2017; Tokazhanov et al., 2020)
	Adsorption	- Ease of operation - Simplicity of design - Remove different types of contaminants, organics and inorganics	- Necessity of take into account some factors: i) initial nitrate concentrations, ii) other ions present in water, iii) adsorbent quantity, iv) water pH, v) operation and maintenance and vi) temperature	(Bhatnagar and Sillanpää, 2011; Sharma and Bhattacharya, 2017)
	Electrodialysis	- Higher taxes of recovery - May remove contaminants and desalinate simultaneously - Environmentally friendly technology	- Necessary to consider time, temperature, flow rate, and voltage to optimize - Process - Efficiency loss due to the fouling and scaling - The need for a pretreatment remineralization requirements	(Aliaskari and Schäfer, 2020; el Midaoui et al., 2002; Sharma and Bhattacharya, 2017)
Transformation	Biological denitrification	- Economical - Environmentally friendly technology	- High levels of nitrate concentration may be difficult to reduce - Long time - Bacteria sludge - High pH requirements - Low selectivity - Higher energy requirements	(Chu and Wang, 2013; Ghafari et al., 2008; Martínez et al., 2017; Sharma and Bhattacharya, 2017; Tokazhanov et al., 2020; Xu et al., 2018; Yang et al., 2019)
	Catalytic reduction	- Nitrate conversion of 98–100% - No waste	- Formation of ammonia - High operational costs	(Ghosh et al., 2017; Guate et al., 2019; Liu et al., 2012; Marchesini et al., 2019; Martínez et al., 2017; Pizarro et al., 2018; Sharma and Bhattacharya, 2017; Tokazhanov et al., 2020; Zhou et al., 2017)
	Electrocatalytic reduction	- Versatile - Scalable	- Requirement of certain conductivity	(Akbari et al., 2020; Garcia-Segura et al., 2018; Martínez et al., 2017; Sanjuán et al., 2020; Tokazhanov et al., 2020; Weber et al., 2019)
	Photocatalytic method	- High selectivity	- Formation of nitrite and ammonium	(Kozyatnyk and Klymenko, 2016; Sharma and Bhattacharya, 2017; Zhang et al., 2005)

elimination increases with high electrical potential (Aliaskari and Schäfer, 2020).

However, the use of techniques to transform nitrates to N_2, has the advantage of no waste production. Biological denitrification can be heterotrophic or autotrophic. On the one hand, heterotrophic bacteria need the presence of organic compounds or hydrocarbons as energy and carbon source. On the other hand, autotrophic bacteria use inorganic compounds, usually carbon dioxide as carbon source, and electron donors like hydrogen or reduced sulfur compounds as energy source. In this context, autotrophic denitrification using hydrogen as electron donor is called hydrogenotrophic denitrification (Ghafari et al., 2008). Lee and Rittmann (2000) developed a hollow fiber membrane biofilm reactor capable of removing nitrate using hydrogen as electron donor (Lee and Rittmann, 2000). Zhou et al. (2017) developed a hollow fiber membrane reactor combining biofilm with a palladium catalyst achieving a reduction of nitrates faster than using only biofilm or only the palladium catalyst and they reported a selectivity toward nitrogen gas (Zhou et al., 2017). In contrast, catalytic reduction employs a bimetallic catalyst consisting of a noble metal and a transition metal. These catalysts are usually composed of palladium or platinum and copper, tin or indium. The reaction occurs with hydrogen as reducing agent and carbon dioxide is commonly used as buffer to control pH (Martínez et al., 2017). Although this method achieves high nitrate degradation yields, the chemical catalytic process is not implemented yet at full-scale. Currently, Marchesini et al. (2019) compared $PdIn/SiO_2$ and $PdIn/Al_2O_3$ catalysts to reduce nitrates from real water, that is, in the presence of other anions such as sulphate, carbonate or chloride. They concluded that for $PdIn/SiO_2$ the presence of other anions decreases the nitrate reduction yield and the selectivity toward nitrogen gas while for $PdIn/Al_2O_3$, the presence of other anions only diminishes the selectivity toward nitrogen gas (Marchesini et al., 2019). Control of pH has been studied by Pizarro et al. (2018) using CO_2 as buffer gas. They achieved 100% of conversion using $PdIn/Al_2O_3$ catalyst with and without CO_2 buffer but the selectivity toward ammonia was 34% and 92% respectively (Pizarro et al., 2018). Additionally, zero-valent iron (ZVI) has been used as catalyst too. ZVI, in presence of nitrates, is oxidized to form Fe^{2+}/Fe^{3+} and nitrates are reduced to form nitrites, nitrogen or ammonia (Dominguez et al., 2018; Martínez et al., 2017). Ghosh et al. (2017) achieved 72–100% of nitrate reduction with a selectivity higher than 99% toward ammonia (Ghosh et al., 2017), while Liu et al. (2012) reached 90% nitrate reduction with nitrogen gas as the main product, together with 20% selectivity toward ammonia and a small number of nitrites (Liu et al., 2012).

The electrocatalytic reduction of nitrates to nitrogen or ammonia is another alternative. The reaction occurs in the cathodic surface which acts as a catalyst too. The main configurations to remove nitrates using this technology are single chamber cell, where cathode and anode are in the same compartment and in contact with the electrolyte, and dual-chamber cell, where a membrane separates the electrodes (Martínez et al., 2017). Several authors have researched this process to eliminate nitrates. Weber et al. (2019) achieved 99% and 90% of nitrate conversion with 81% and 78% of selectivity toward nitrogen gas using CuSn and CuPd as cathode, respectively (Weber et al., 2019). Akbari et al. (2020) secured 90% of nitrate conversion with a nitrogen selectivity of 88.8% employing Fe^0/Fe_3O_4 over nickel foam as cathode (Akbari et al., 2020). However, the photocatalytic method to remove nitrates is still in its infancy due to the formation of nitrite and ammonium as byproducts; promising results have been published by Zhang et al. (2005) who reported 100% selectivity toward nitrogen gas with Ag/TiO_2 with fine Ag cluster catalyst (Zhang et al., 2005).

Regarding transformation techniques, catalytic reduction presents more advantages than biological denitrification such as high nitrates removal rates. However, biological denitrification has full-scale installations in contrast to catalytic reduction and this technology does not produce ammonia (Rezvani et al., 2019). Only a pilot plant located in Borrasa (Spain) has been currently reported in the literature ("Breakthrough in Nitrate-polluted Water Treatment", 2021).

Regarding pilot plants for nitrates removal based on separation or transformation technologies, there are several plants developed throughout the world. Nevertheless, these plants, in many cases, are only dedicated to the treatment of surface waters. Table 10, shows the main pilot or full-scale plants installed to treat nitrates. This table collects the data reported in the bibliography for nitrate treatment plants; in many cases, other compounds are removed at the same time. In view of the results, it is important to highlight, i) the treatment capacity of the plants offers high variability, from 18 m^3 per day in Spain using the electrodialysis reversal technology to 38,400 m^3 in Germany employing biological denitrification, ii) the lack of data regarding the nitrate removal yield, iii) the high number of plants installed in the USA based on the ion exchange technology and, iv) Asia and Africa, with large regions contaminated by nitrates, only have two plants dedicated to nitrates removal. On the other hand, it is worth to mention the high differences in terms of capacity per day of reverse osmosis plants.

5. Conclusions

This review collects the quality of different groundwater bodies throughout the world in terms of nitrates concentration and reports the treatment options both for this compound and for other critical pollutants, identifying the pilot plants that are currently in operation.

After analyzing a total of up to 292 points from 146 works reported in the literature, some general conclusions can be drawn: i) The presence of nitrates in groundwater is continuously growing, and in consequence, urgent measures are required to avoid the degradation of these water bodies. ii) Different areas worldwide present values higher than the WHO standards (50 ppm). iii) Nitrate pollution coming from agriculture and, specifically, from the use of fertilizers and pesticides is a common denominator on all continents. iv) In Asia, there is significant variability in the concentration of NO_3^- because of the strong influence of the rainy season and the poor treatment of septic tanks. v) In addition to contamination by fertilizers, aquifers in America suffer from wastewater discharged without adequate treatment. vi) In Europe, landfills, industrial activities, and sewage have an outstanding contribution to nitrate presence in groundwater. vii) In Africa, apart from the different reasons previously summarized, pollution is mostly due to the poor health conditions in many countries (e.g., the presence of septic tanks without treatment).

Nowadays, there is a high number of plants dedicated to surface water treatment, nevertheless, the number of plants for groundwater treatment is still very limited. Regarding the different treatments to remove nitrates, continue research is necessary since, although there are many technologies available, scarce applications have reached the pilot plant scale. Also, between all the technologies that are currently under development, there is not a clear line because all present clear advantages and disadvantages. In the case of reverse osmosis, commonly used to remove pollutants from surface water, the degradation of the membranes by fouling and the fact that nitrate is not converted to nitrogen prevents it from being the most promising technology. On the other hand, apart from reverse osmosis, biological denitrification and ion exchange are the main treatments implemented at full-scale to nitrate removal. However, these technologies have the drawback of the difficult nitrate removal at high concentration. Catalytic nitrate reduction, which still needs further research, is a promising technology for the destruction of nitrates due to the high conversion close to 98–100%, and no waste generation. Thus, despite there is still a long way to achieve full groundwater remediation, the efforts which are currently being performed will promote short-term responses.

Table 10
Nitrate treatment plants at full-scale.

	Location	Capacity (m^3/day)	Nitrates removed (mg/L)	Opening year	Ref.
Reverse osmosis	Bakersfield (California)	654	75–84	n. a.	(Jensen et al., 2014)
	Brighton (Colorado)	25,075	49–89	n. a.	
	Arlington Desalter, Riverside (California)	24,982	44–89	n. a.	
	Yemen	169		n. a.	(Shams, 2010)
	France	120		n. a.	
	Zava-Giyani (South Africa)	54.5		n. a.	
	Milan (Italy) (13 sites)	169–1352		n. a.	
	Pomaire (Chile)	518		2015	("Reverse osmosis dwtp for nitrate removal, pomaire, chile – wes chile – water & energy solutions", 2021)
Ion exchange	Ellsworth (Minnesota)	178		1994	
	Clear Lake (Minnesota)	178		1995	
	Edgerton (Minnesota)	519		2002	
	Adrian (Minnesota)	488		1998	
	McCook (Nebraska)	25,741	> 125	2006	
	McFarland (California)	3785	60	1983	
	McFarland (California)	3785	64	1987	(Jensen et al., 2014)
	La Crescenta (California)	10,221	70–100	1987	
	Grover City (California)	8706	80–130	n. a.	
	Des Moines (Iowa)	37,854	>55	1992	
	Glendale (Arizona)	37,854	177	2010	
	Indian Hills (Colorado)	272.5	53–71	2009	
	Avondale (Arizona)	10,357		2003	
	Salinas (California)	16,353		2002	
	East Valley, San Bernardino Highland (California)	5451		2003	
	Yucca Valley (California)	13,627		2002	(Shams, 2010)
	Pomona (California)	5451		Late 1990s	
	- (California)	10,902		n. a.	
	Colina (Chile)	630		n. a.	("Nitrate removal dwtp, colina, chile – wes chile – water & energy solutions", 2020)
Electrodialysis	Spain	18	80	n. a.	(Jensen et al., 2014)
Selective reversal	Israel	1690	84–89	n. a.	(Jensen et al., 2014)
Biological denitrification	Rialto (California)	10,901	17–19	n. a.	
	Riverside (California)	9102	44–89	n. a.	
	Obersiebenbrunn (Austria)	4320		n. a.	
	Chateau Landon (France)	1248		n. a.	
	Champfleur (France)	1680		n. a.	
	Issoudun (France)	14,000		n. a.	
	Bourg les valences (France)	10,800		n. a.	
	Nord Sarthe (France)	7200		n. a.	
	Dreux-Vernouillet (France)	16,800		n. a.	
	Niort (France)	48,000		n. a.	
	Thouard (France)	20,400		n. a.	(Jensen et al., 2014)
	Vernoy (France)	4800		n. a.	
	Hanau (Germany)	1200		n. a.	
	Neuss (Germany)	3600		n. a.	
	Frankfurt Airport (Germany)	7680		n. a.	
	Aschaffenburg (Germany)	38,400		n. a.	
	Föhr Island (Germany)	2160		n. a.	
	Albanacci (Italy)	9120		n. a.	
	Czestochowa (Poland)	12,000		n. a.	
	Rancho Cucamonga (California)	3600		n. a.	
	Coyle (Oklahoma)	300		n. a.	
	Falset (Spain)	120		2018	(Alvarez and Murría, 2020)
	Republic (Washington)	272		2006	
	Republic (Washington)	32,7		2005	
	Cajamarca (Perú)	1199		2003	(Reinsel, 2014)
	Lead (Dakota del Sur)	2180		1997	
	Nye (Montana)	13,082		1995	
	Langenfeld and Monheim (Germany)			n. a.	
	Monchengladback (Germany)			n. a.	
	Vsetaty (Czech Republic)			n. a.	
	Drosing (Austria)			n. a.	(Shams, 2010)
	Montferland (the Netherlands)			n. a.	
	Blankaart (Belgium)			n. a.	
Electrocatalytic reduction	Borrassà (Spain)	100		2017	("Breakthrough in Nitrate-polluted Water Treatment", 2021)

CRediT authorship contribution statement

E. Abascal: Formal Analysis, Investigation, Writing - Original Draft
L. Gómez-Coma: Methodology, Investigation, Writing - Original Draft
I. Ortiz: Writing - Review & Editing, Supervision, Funding acquisition
A. Ortiz: Conceptualization, Investigation, Writing - Review & Editing, Supervision, Project administration, Funding acquisition

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.152233.

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