Determining nitrate pollution sources in the Kabul Plain aquifer (Afghanistan) using stable isotopes and Bayesian stable isotope mixing model

Abdulhalim Zaryab[ab], Hamid Reza Nassery[a*], Kay Knoeller[c], Farshad Alijani[a], Eddy Minet[d]

[a]Department of Minerals and Groundwater Resources, Faculty of Earth Sciences, Shahid Beheshti University, Evin Ave, Tehran, Iran

[b]Engineering Geology and Hydrogeology, Faculty of Geology and Mines, Kabul Polytechnic University, District 5, Kabul, Afghanistan

[c]Department Catchment Hydrology Helmholtz-Centre for Environmental Research – UFZ, D-06120 Halle, Germany

[d]Environmental Protection Agency (EPA), Dublin, Ireland

*Corresponding author, E-mail address: h-nassery@sbu.ac.ir (H.R. Nassery), Phone: (+98)-90-2983-2399

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*Corresponding author, E-mail address: h-nassery@sbu.ac.ir (H.R. Nassery), Phone: (+98)-90-2983-2399

Abstract

The Kabul urban aquifer (Afghanistan), which is the main source of drinking water for Kabul city's inhabitants, is highly vulnerable to anthropogenic pollution. In this study, the geochemistry of major ions (including reactive nitrogen species such as NO_3^-, NO_2^-, and NH_4^+) and stable isotope ratios ($\delta^{15}N$-NO_3^-, $\delta^{18}O$-NO_3^-, $\delta^{18}O$-H_2O, and δ^2H-H_2O) of surface and groundwater samples from the Kabul Plain were analyzed over two sampling periods (dry and wet seasons). A Bayesian stable isotope mixing model (SIMMR) was also employed to trace potential nitrate sources, transformation processes and proportional contributions of nitrate sources in the Kabul aquifer. The plotting of $\delta^{15}N$-NO_3 against $\delta^{18}O$- NO_3 ($\delta^{15}N$-NO_3 and $\delta^{18}O$- NO_3 values ranged from +4.8 to +25.4‰ and from -11.7 to +18.6‰, respectively) suggests that NO_3 primarily originated from the nitrification of sewage, and not from artificial fertilizer. High Cl^- concentrations and low NO_3^-/Cl^- ratios supported the assumption that sewage is the dominant nitrate source. The results indicated that denitrification did not influence the NO_3 isotopic composition in the Kabul aquifer. The SIMMR model suggest that nitrate in the dry season originated mainly from sewage ~81%, followed by soil organic N 10.5%, and chemical fertilizer 8.5%. In the wet season, sewage ~87.5%, soil organic N 6.7%, and chemical fertilizer 5.8% were the main NO_3 sources in the Kabul aquifer. Effective land management measures should be made to improve the sewage collection system in the Kabul Plain.

Keywords: Groundwater, Stable nitrate isotope, Nitrate, Bayesian stable isotope mixing model, Nitrogen transformation, Kabul

1. Introduction

The contamination of groundwater and surface water by nitrate (NO_3^-) in urban and agricultural areas is a serious issue (Mayer et al., 2002; Voss et al., 2006; Kendall et al., 2007; Kaushal et al., 2011; Xu et al., 2015; Matiatos, 2016; Luu et al, 2020; Ye et al., 2021). From an environmental perspective, discharge of

contaminated groundwater into surface water bodies can contribute to eutrophic conditions in lakes, streams, estuaries and the coastal zone. From a health perspective, a high level of NO_3−N (> 10 mg L^{-1}) in drinking water may result in blue baby syndrome in infants (methemoglobinemia) and also may contributing factor for some types of cancer (Fan and Steinberg, 1996; Gulis et al., 2002; Fewtrell, 2004; Ward et al., 2005; Pastén-Zapata et al., 2013; Nikolenko et al., 2017). While natural levels of nitrate in aquatic systems occurs at concentrations generally no higher than 2.5 mg L^{-1} as NO_3−N (Panno et al., 2006), higher NO_3 concentrations are often the result of anthropogenic activities associated with excessive use of chemical fertilizers and manure by agriculture, sewage waste, and/or urban solid waste landfills (Kendall, 1998; Chang et al., 2002; Kendall et al., 2007; Xue et al., 2009; Pastén-Zapata et al., 2014; Kim et al., 2015; Popescu et al., 2015; Nikolenko et al., 2017; Mingming et al., 2018; Li et al., 2018; Soto et al., 2018; Ogrinc et al., 2018; Nejatijahromi et al., 2019; Zhang et al., 2020). Other factors may also affect nitrate concentrations such as temporal variability of rainfall, hydrogeological conditions and land cover (Exner-Kittridge et al., 2016; Suchy et al., 2018; Sebestyen et al., 2019; Romanelli et al., 2020).

Groundwater is a major source of freshwater worldwide and its use as drinking water in cities is increasing. Reducing NO_3 pollution in groundwater is needed to improve, safe water usage, but to do so, it is important to accurately identify the origins of NO_3 and quantify the contribution of nitrate sources.

Some NO_3 sources exhibit characteristic $^{15}N/^{14}N$ and $^{18}O/^{16}O$ ratios (expressed as $\delta^{15}N$-NO_3 and $\delta^{18}O$-NO_3^-), which can provide valuable information to track the origin of nitrate contamination in aquatic environments (Böttcher et al., 1990; Wasssenaar, 1995; Chang et al., 2002; Kendall et al., 2007; Xue et al., 2009; Nestler et al., 2011; Kim et al., 2015; Jung et al., 2020). Typically, chemical fertilizers and precipitation show the lowest $\delta^{15}N$-NO_3 values, followed by soil-derived nitrate with intermediate values, and then nitrate derived from manure and sewage with the highest $\delta^{15}N$-NO_3 values (Kendall et al., 2007, Xue et al., 2009). Over the last three decades, the addition of $\delta^{18}O$- NO_3 has been successfully used by some to improve the identification of nitrate sources as well as nitrate attenuation processes in aquatic environments (Böttcher et al., 1990; Wassenaar, 1995; Aravena and Robertson, 1998; Kendall et al.,

2007; Savard et al., 2009; Liu et al., 2013; Ding et al., 2014; Griffiths et al., 2016; Yi et al., 2017; Li et al., 2018; Zendehbad et al., 2019; Zhao et al., 2019; Weitzman et al., 2021). However, δ values can overlap between nitrate sources, which can complicate data interpretation. Furthermore, isotopic fractionation during the nitrate transformation processes, including nitrification and denitrification, can also alter the original isotopic compositions of nitrate sources (Kendall et al., 2007; Xue et al., 2009; Yi et al., 2017; Minet et al., 2017; Zhu et al., 2019; Zheng et al., 2020; Yang et al., 2020; Blarasin et al., 2020). Recently, the identification of nitrate sources was considerably improved by combining the dual stable isotope approach with hydrochemical and land use data (Pastén-Zapata et al., 2013; Minet et al., 2017; Biddau et al., 2018; Nejatijahromi et al., 2019, Wu et al., 2020).

Stable isotopes have also been used to quantify NO_3^- sources in aquatic systems. Early attempts involved a dual isotopic linear mixing model based on mass balance (Phillips and Koch, 2002; Deutsch et al., 2006). Nevertheless, this method presumed no significant isotopic fractionations from transformation processes and was limited to track no more than three sources (Parnell et al., 2010). Parnell et al. (2010) later implemented a Bayesian stable isotope mixing model in the software package SIAR (Stable Isotope Analysis in R). The Bayesian stable isotope mixing model was employed by Xue et al. (2012) to quantify nitrate sources for the first time. The SIAR model estimates the probability distribution of the proportional contributions based on the Bayesian approach and quantitively identifies more than three nitrate sources, which overcomes earlier limitations of the linear isotopic mixing model (Parnell et al., 2010; Xue et al., 2012; Matiatos, 2016; Meghdadi and Javar, 2018; Li et al., 2019). During the past decade, other studies have combined stable isotopes in nitrate with a SIAR approach to identify the proportional contributions of NO_3^- sources in aquatic environments (Korth et al., 2014; Yang and Toor, 2016; Jin et al., 2019; Torres-Martínez et al., 2020). However, limitations remain as the SIAR approach rarely considers the denitrification process and related fractionation (Xia et al., 2017; Meghdadi and Javar, 2017). To overcome these issues, Bayesian stable isotope mixing models (SIMMs) were developed (Parnell et al., 2013; Zhao et al., 2019) and used in a few recent studies that also combine hydrochemical

and land use data to more accurately identify and quantify the nitrate sources in aquatic systems (Zhang et al., 2020; Romanelli et al., 2020, Re et al., 2021; Torres-Martínez et al., 2021).

The study area, Kabul city, is located in southeast Afghanistan with a population of about 5.38 million inhabitants. Over the last two decades, it has experienced rapid unplanned urbanization. Groundwater is the only source of drinking water but its quality has long deteriorated due to (i) lack of centralized sewerage system, (ii) intensive fertilizer application and/or (iii) the direct discharge of solid waste in surface water. Some studies have been conducted before on groundwater quality of the Kabul Basin aquifer (Houben et al., 2005; Broshears et al., 2005; Mack et al., 2010; Saffi, 2019; Jawadi et al., 2020) and Houben et al., (2005) reported worryingly elevated nitrate concentrations (>350 mg L^{-1}). The determination of nitrate sources of nitrate and transformation processes were however less considered. This study aims to identify and quantify nitrate sources and associated biogeochemical processes using stable isotope ratios and a stable isotope mixing model (SIMMR) combined with land-use, hydrogeological and hydrochemical data.

2. Material and Methods

2.1. Study area description

The study area of Kabul city, the capital and the largest city in Afghanistan with an estimated population of approximately 5.38 million (NSIA, 2021) is located in the Kabul Plain in the center of the Kabul province in eastern Afghanistan. It covers an area of 280 km^2 at an elevation of about 1800 m above sea level. (Fig. 1). The Plain is enclosed by low but quite steep mountain ranges, and it is divided into western and eastern plains by the Asmai and Share Darwaza mountains. Three major seasonal rivers flow through it: Kabul, Logar and Paghman (Fig.1). The western plain is drained from west to east by the upper Kabul and Paghman rivers and the eastern plain is drained by the Kabul and Logar rivers. The Kabul Plain is characterized by a semiarid climate with an average annual precipitation of 315 mm. The air temperature ranges from an average monthly low in January of -7°C to an average monthly high in July of +32°C. The rainy season, accounting for 68% of the annual rainfall, runs from January to April. The mountains

surrounding the Kabul Plain are mainly underlain by a variety of metamorphic rocks, which include, schist, gneiss and amphibolite, and sedimentary rocks, including limestone and dolomite (Bohannon, 2010). The study area of Kabul city consists of an accumulation of terrestrial and lacustrine sediments, mainly of the Quaternary and Neogene ages.

The main land-uses in the Kabul Plain are residential (54.2% of the total area), followed by agriculture (20%), transportation (9.7%) and vacant plots (8.7%) (Fig. 2). The area under cultivation in the Kabul Plain covers about 56 km^2. Agricultural land is mainly in the southwest of the Eastern Plain. The major crops of the Kabul Plain are wheat, potato, maize, millet, cucumber, vegetables and fruits. The chemical fertilizers used in the Kabul Plain ((Ministry of Agriculture, Irrigation and Livestock of Afghanistan, personal communication) are commonly imported by local suppliers from Pakistan, Iran, Turkey and China and their compositions are: urea ($CO(NH_2)_2$), ammonium sulfate $(NH_4)_2SO_4$) and diammonium phosphate ($(NH_4)_2HPO_4$). Besides chemical fertilizers, manures are also applied to crops. Chemical fertilizers are generally applied at the beginning (April) and in the middle of the growing season (June). Soil erosion is not a considerable process in the Kabul Plain (Safi et al., 2017). Based on land use information, potential sources of nitrate pollution in the plain are therefore anthropogenic and include sewage (pit latrines, septic tanks), intensive use of fertilizers (chemical fertilizers, manure and sewage sludge), solid waste landfill sites, solid waste disposal in the rivers and channels and presence of local cemeteries. It is important to note that there is currently no centralized sewerage system in Kabul city. Sewage collection and wastewater treatment plants operate only a small scale, particularly in areas like new townships (Paiman and Noori, 2019).

From a hydrogeological perspective, the Kabul Plain is thus divided into Quaternary and Neogene aquifers. The Quaternary aquifer, which was described in detail by Houben et al., (2009), is generally considered shallow and divided into three locally important interconnected Quaternary unconfined aquifers. It mainly consists of gravel and sandy beds covered by loess clay and silts beds with a thickness of up to a few meters. The thickness of this shallow aquifer can be up to 80 m. The hydraulic conductivity ranged from 4 to 112 m day^{-1} (Houben et al., 2009). The Neogene aquifer, on the other hand, is deep and

semi-confined. It has a thickness that ranges between 30 and 600 m and is part of an aquitard/aquiclude (Houben et al., 2009; JICA, 2011; Landell Mills, 2020). The Neogene deep aquifer is composed of stiff clay with unconsolidated sand, gravel and conglomerates with a transmissivity ranging from 2 to 27 m^2 day^{-1} (Landell Mills, 2020).

The major sources of groundwater recharge are infiltration of river channels, precipitation, and agricultural infiltration (water used for irrigation is supplied by rivers and groundwater), while groundwater discharge is largely accounted for by pumping withdrawals for drinking, agricultural and industrial uses. Due to over-abstraction, groundwater now flows toward the central part of the western plain, whereas it used to be directed from west and south-east toward the east (Brohears et al., 2005). The groundwater flow direction in the eastern plain is from south and west toward the east and from the central part of the plain to the northwest. It can be noted that with the recent and rapid urbanization of Kabul city, groundwater abstraction has increased dramatically and the groundwater table depth in the Plain has declined rapidly (-1.7 m/year on average) (Zaryab et al., 2017).

2.2. Sampling and analyses

After consideration of the different land uses, potential sources of pollution, and hydrogeological conditions, twenty-nine wells were selected for groundwater sampling, (Fig. 2). These water wells range in depth between 20 to 120 m (Table S1). Water samples were collected during two sampling campaigns conducted in the dry (November 2020) and in the wet seasons (April 2021). Before collecting the samples, each well was purged for up to 10 min, until all field parameters had stabilized. Other water samples that were collected included river water (Paghman, Kabul, and Logar Rivers), mixed wastewater and irrigation return flows. Field measurements, recorded with a pre-calibrated hand-held meter (Multi 340i, WTW, Germany), included pH, dissolved oxygen (DO), temperature and electrical conductivity (EC). Each water sample was split into three aliquots that were analyzed for either nitrogenous nutrients (NO_3^-, NO_2^-, NH_4^+), major ions (Ca^{2+}, Mg^{2+}, Na^+, K^+, HCO_3^-, Cl^-, SO_4^{2-}) or stable isotope signatures in nitrate ($\delta^{15}N\text{-}NO_3^-$, $\delta^{18}O\text{-}NO_3^-$). In addition, thirty-two groundwater and surface-water samples collected

during the wet season were analyzed for stable isotope signatures in water only (δ^2H-H_2O and δ^1H-H_2O) and $^{18}O/^{16}O$ ratios in H_2O). Nutrient concentrations were measured with a spectrophotometer (DR 5000, Hach, USA) on samples collection day at the Green Tech Laboratory in Kabul. Aliquots for major ions analysis were shipped to the Regional Water Organization Laboratory of Tehran (Iran). The measurement of parameters other than bicarbonate (HCO_3^-) was carried out by ion chromatography (S 151 IC, Sykam, Germany), as per APHA (2017). HCO_3^- concentrations were determined using the titration method outlined by Stumm and Morgan (2012). Some duplicates and field blanks were also collected and analyzed for quality assurance purposes. The reliability of all the chemical analyses was checked by ionic charge balance, with a threshold error of 5 % set to accept results. All the gathered data, including in situ measurements, ion concentrations, and isotopes ratios are presented in Table S1. The statistical summary of the data is listed in Table 2.

Aliquots for stable isotopes analysis in nitrate were filtered on-site into 60 mL HDPE bottles with 0.22 µm cellulose filters, which aimed at removing particulate matter and most microbial activity that could cause some isotopic fractionation. Samples were kept refrigerated until they were shipped to the Helmholtz Centre for Environmental Research-UFZ (Germany). Dual stable isotope analysis (i.e. $^{15}N/^{14}N$ and $^{18}O/^{16}O$ ratios in NO_3^-, referred to as $\delta^{15}N$-NO_3 and $\delta^{18}O$-NO_3^-, respectively) were carried out in duplicate for each sample on a Gasbenchll/delta V plus combination (Thermo Scientific, USA) using the denitrifier method for a simultaneous determination of $\delta^{15}N$ and $\delta^{18}O$ in the measuring gas N_2O, produced by controlled reduction of sample nitrate (Sigman et al., 2001; Casciotti et al., 2002). Nitrogen and oxygen stable isotope results are reported in delta (δ) notation ($\delta_{sample} = [(R_{sample}/R_{standard}) - 1] \times 1000$) as part per thousand (‰) deviation relative to the standards AIR for nitrogen and Vienna Standard Mean Ocean Water (VSMOW) for oxygen, where R is the ratio of the heavy to light isotope (i.e. $^{15}N/^{14}N$ and $^{18}O/^{16}O$). The standard deviation of the described analytical measurement was ±0.4 ‰ for $\delta^{15}N$ and ±1.6 ‰ for $\delta^{18}O$. Stable isotope results in Section 3 represent the mean value of duplicated measurements for each sample.

Aliquots for stable isotopes analysis in water were also filtered on-site into 60 HDPE bottles with 0.22 μm cellulose filters and shipped to the Helmholtz Centre for Environmental Research-UFZ. Dual stable isotope analyses ($^{2}H/^{1}H$ and $^{18}O/^{16}O$ ratios in $H^{2}O$, referred to as $\delta^{2}H$-H_2O and $\delta^{18}O$-H_2O, respectively) were carried out using laser cavity ring-down spectroscopy (Picarro L2120-I, Santa Clara, USA) without further treatment of the water samples. Results were reported in δ values relative to VSMOW. Samples were normalized to the VSMOW scale using replicate (20x) analysis of internal standards calibrated to VSMOW and Standard Light Antarctic Precipitation (SLAP) certified reference materials. The analytical uncertainty of the $\delta^{18}O$ measurement was ±0.1 ‰, for hydrogen isotope analyses, an analytical error of ±0.8 ‰ has to be considered.

ArcGIS tools were employed to represent the spatial variations of nitrate concentrations and values of $\delta^{15}N$-NO_3^-, $\delta^{18}O$-NO_3^-. AqQA software was used to characterize of hydrochemical facies of the water samples. All the scatter diagrams were created using Microsoft Excel 2019.

2.3. Estimation of contributions from different nitrate pollution sources

The proportional contributions of the different NO_3 sources in the water samples were calculated using a Bayesian Stable Isotope Mixing Model (SIMMR) package for R, and an updated version of the Bayesian isotope mixing model named SIAR. The methodology employed in the SIMMR model has been described in detail by Parnell et al. (2013) and Parnell (2016). This approach can incorporate sources of uncertainty, isotope fractionation, and multiple NO_3 sources (Parnell et al., 2013; Zhao et al., 2019; Herms et al., 2021). The SIAR model is an open-source software package for R and uses the Markov chain Monte Carlo method to estimate possible source proportions. By defining, a set of N mixture measurements on j stable isotopes with k source contributors are expressed as follows:

$$X_{ij} = \sum_{k=1}^{K} P_k(S_{jk} + C_{jk}) + \varepsilon_{ij}$$

$$S_{jk} \sim N(\mu_{jk}, \omega_{jk}^2)$$

$$C_{jk} \sim N(\lambda jk, \tau_{jk}^2)$$

$$\varepsilon_{jk} \sim N(0, \sigma_j^2)$$

where X_{ij} is the isotope value j of the mixture i, in which i = l, 2, 3,..., N and j = 1, 2, 3, …, J; S_{jk} is the source value k based on isotope j (k = 1, 2, 3, …, K) and is normally distributed with the mean μ_{jk} and the standard deviation ω_{jk}; P_k is the proportion of source k, as estimated by the SIAR model; C_{jk} is the fractionation factor for isotope j on source k and is normally distributed with mean λ_{jk} and standard deviation τ_{jk}; and ε_{ij} is the residual error, representing the additional unquantified variation between individual mixtures and is normally distributed with a mean of 0 and a standard deviation σ_j.

The SIMMR model was conducted in the SIMMR package of the R Programming Language (version 4.1) in the RStudio environment. The different isotopic signatures of nitrate sources employed in the SIMMR were obtained through a literature review and local end-members (Xue et al., 2009; Nikolenko et al., 2018; Zhang et al., 2018; Torres-Martínez et al., 2020). These values are listed in Table 1.

3. Results and discussion

3.1. Hydrochemistry

The in-situ measurements, chemical parameters, and isotopic ratios measured in the groundwater and river water samples collected from the Kabul Plain are presented in Table S1 of the supplementary material. pH in both seasons was very similar and slightly alkaline. It ranged from 7.0 to 8.1 (mean value of 7.6) in the dry season (November 2020), and from 7.1 to 8.0 (mean of 7.6) in the wet season (April 2021). EC ranged from 618 to 17830 μS cm^{-1} (mean of 1640 μS cm^{-1}) in the dry season, and from 458 to 16430 μS cm^{-1} in the wet season (mean of 1430 μS cm^{-1}). The elevated EC values were encountered in the lacustrine evaporite deposits (Zaryab et al., 2021). The temperature of water samples ranged from 6ºC to 17.8ºC (mean value of 15.4ºC) in the dry season and from 12.7ºC to 18ºC (mean value: 16.2ºC) in the wet season. Dissolved oxygen (DO) concentration varied from 2.1 to 9.6 mg L^{-1} (mean 5.3 mg L^{-1}) in the dry season, and from 1.9 to 8.3 mg L^{-1} (mean 4.8 mg L^{-1}) in the wet season, indicating aerobic conditions in the aquifer. The elevated DO contents in the dry and wet seasons, could possibly be due to the presence of thousands of water wells in the Kabul Plain with long screens and excessive pumping from these wells.

In most groundwater samples of the western plain, the dominant cations were $Ca^{2+} > Mg^{2+} > Na^{+} > K^{+}$, whereas the dominant anions were $HCO_3 > Cl > SO_4^{2-}$. In comparison, Mg^{2+} was the dominant cation in most groundwater samples of the eastern aquifer. High concentrations of Mg^{2+} might be due to the dissolution of dolomite (Zaryab et al., 2021). Major ion concentrations of groundwater samples were plotted in a Piper diagram (Fig. 3) to characterize hydrochemical dominant groundwater types in the Kabul aquifer. As shown in Fig. 3a and 3b, groundwater collected in both November 2020 and April 2021 displays similar patterns. Based on the Piper diagram, the dominant type of groundwater in the western plain is Ca-HCO_3, followed by Ca-(Mg, Na)-HCO_3. In the eastern plain, the dominant type of water is Mg-HCO_3, followed by Mg-(Na, Ca)-HCO_3(Cl, SO_4), Na-SO_4, and Na-Cl water types. Except for sampling well W5, all groundwater samples in the western plain displayed freshwater facies. On the other hand, most of the groundwater samples in the eastern plain demonstrate brackish water facies.

3.2. Isotopic compositions of water

The isotopic composition was used to assess the interactions between groundwater and rivers to identify the origins of water in the study area (Cook, 2013; Clark, 2015). Values of δ^2H-H_2O and $\delta^{18}O$-H_2O in groundwater and river water samples from the Kabul Plain are presented in Table S1 of the supplementary material. The values of δ^2H-H_2O and $\delta^{18}O$-H_2O in groundwater ranged from -73.4‰ to -30.3‰ (mean of -52.2‰) and -10.59‰ to -6.39‰ (mean of -8.31‰), respectively. The values of δ^2H-H_2O and $\delta^{18}O$-H_2O in the rivers varied from -53.9‰ to -43.3‰ and -8.50‰ to –7.37‰, respectively. As shown in Fig. 4, most of the water samples fall above the global meteoric water line ($\delta^2H = 8.13\delta^{18}O + 10.8$) (Rozanski et al., 1993) and to below the local meteoric water line ($\delta^2H = 7.325\delta^{18}O + 12.37$), which is based on IAEA data (1962-1989) reported by Houben et al. (2005). Generally, the deviation of the local meteoric water line from the global meteoric water line is controlled by an element-specific fractionation of the stable water isotopes along the vapor mass trajectory due to (1) latitudinal effect (lower delta values at increasing latitude), (2) continental effect (lower delta values the more inland), (3) altitude effect (lower delta values at higher altitude, (4) seasonal effect (lower delta values in winter in temperate

regions) and (5) amount effect (lower delta values during heavy storms (Mook and de Vries, 2000). The deviation of the water isotopic values of the groundwater from the local meteoric water line may be due to the occurrence of evaporation. This means that groundwater is isotopically enriched with respect to local precipitation due to evaporation. Therefore, the local rainfall after evaporation is the major source of groundwater in the Kabul Plain. Fig. 4 shows that except for water samples W4, W15 and W24 all groundwater samples followed the same pattern. The water well W4 experienced a rapid water table decline in recent years due to low permeability of subsurface soil (Meldebekova et al., 2020). It reveals that low $\delta^{18}O$ and $\delta^{2}H$ values in water well W4 reflect long groundwater residence times and recharge during colder climates in the past. It should be mentioned that the water table in the northwest of the study area also dramatically declined in recent years due to the low permeability of the aquifer (Landell Mills, 2020), but the groundwater flow direction (Fig. 1) indicates that groundwater mixing occurred in the mentioned area. Water wells W15 and W24 are drilled within the lacustrine evaporite deposits. Therefore, isotopically-depleted groundwater sample W15 and enrichment in $\delta^{18}O$ and $\delta^{2}H$ of groundwater sample W24 might be because of high salinity in groundwater and exchange with rock minerals, respectively (Clark, 2015; Domenico and Schwartz, 1990). The isotopic compositions of groundwater clearly reflected the rate of recharge from rivers and rainfall and the effects of evaporation in the Kabul aquifer. In general, the $\delta^{2}H$-H_2O and $\delta^{18}O$-H_2O values indicated that most groundwater in the Kabul aquifer was recharged under similar climatic conditions.

The river water sample from Paghman River (PR) plots on the local meteoric water line (LMWL), while two other river water samples (Kabul River (KR) and Logar River (LR)) plot in a more isotopically enriched range similar to the groundwater samples. It suggests that Kabul and Logar rivers evaporated along their flow directions, because before entering the Kabul Plain, they flow long distances compared to the Paghman River. In the meantime, Fig. 4 indicates that the Kabul River originates from a higher altitude because this river was more depleted in $\delta^{18}O$ and $\delta^{2}H$ than Paghman and Logar rivers. The isotopic compositions of rivers indicate that the rivers also are the main source of groundwater in the Kabul Plain.

3.3. Concentrations and isotopic compositions of nitrate

The concentrations and isotopic compositions of nitrate in water samples of the Kabul Plain are listed in Table S1 (no isotopic values for well W20). The $\delta^{15}N\text{-}NO_3$ values in groundwater samples from 28 different locations in the Kabul aquifer ranged from 4.8‰ to 20.8‰ (mean of 10.2‰) in the dry season (November 2020) and from 6.9‰ to 25.4‰ (mean of 12.8‰) in the wet season (April 2021). The $\delta^{18}O\text{-}NO_3$ values varied from -11.7‰ to 18.6‰ (mean of 7.2‰) in the dry season and from -3.4‰ to 11.5‰ (mean of 4‰) in the wet season. The $\delta^{15}N\text{-}NO_3$ in rivers' water varied between7.7‰ and 10.2‰, whereas $\delta^{18}O\text{-}NO_3$ varied between -3.4‰ and 5.3‰. The $\delta^{18}O\text{-}NO_3$ spanned a wide range (-11.7 to 18.6‰) in the dry season and relatively narrowed (-3.4 to 11.5‰) in the wet season (Table S1). The observed wide range of $\delta^{18}O\text{-}NO_3$ values in the dry season might be due to the application of chemical fertilizers in agricultural lands and the mixing of water with different NO_3 loads. The $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ values of mixed wastewaters and mixed irrigation water ranged from 14.4‰ to 13.6‰ and 11‰ to -4.1‰, respectively. The high $\delta^{15}N\text{-}NO_3$ value in the mixed irrigation water sample could be due to using sewage sludge as fertilizer in agricultural areas. The NO_3 concentrations spanned a wide range of values, from 4 to 120.4 mg L^{-1} in the dry season (mean value 13.7 mg L^{-1}) and from 4.9 to 59 mg L^{-1} in the wet season (mean of 17.71 mg L^{-1}). All nitrate concentrations were above (and often widely above) the threshold value for anthropogenic activity (2.5 mg L^{-1}) (Panno et al., 2006; Ogrinc et al., 2019), which can most likely be linked to human activities in the study area (Broshears et al., 2005; Houben et al, 2008). The high $\delta^{15}N\text{-}NO_3$ values suggest that urban sewage was a major source of nitrate in the area (Kendall, 1998; Mayer et al., 2002; Xue et al., 2009, Xu et al., 2016). It is worth mentioning that nitrate concentrations in the groundwater of the Kabul aquifer decreased in comparison to the results reported in previous studies (Broshears et al., 2005; Houben, et al., 2005). It might be due to a significant decline in groundwater tables in densely populated areas (Khair Khana and Dashti Barchi). The nitrite concentrations ranged from 0.01 to 2.8 mg L^{-1} and from 0.01 to 0.36 mg L^{-1} with average values of 0.22 and 0.03 mg L^{-1} in the dry and with the season, respectively (Table S1).

3.4. Spatial variations of nitrate concentrations and isotope values

The spatial variations in the concentration of NO_3, $\delta^{15}N$-NO_3 and $\delta^{18}O$-NO_3 values in groundwater of the Kabul aquifer for both dry and wet seasons are illustrated in Fig. 5. The highest observed concentration of nitrate in the dry season was 120.4 mg L^{-1} in well W12, while the lowest observed concentration of nitrate was 4 mg L^{-1} in well W20. The highest and lowest concentrations of nitrate (58.9 and 4.87 mg L^{-1}) were detected at wells W27 and W7, respectively. The field survey reveals that sewage waste may have caused high concentrations of nitrate in groundwater wells W12 and W27. Elevated nitrate (50.9 mg L^{-1}) in well W1 in the dry season might be associated with agricultural activities and the presence of a local cemetery. The NO_3 concentrations were generally higher during the wet season compared to the dry season in the study area. However, NO_3 concentration in well W12 considerably decreased in the wet season compared to its concentration in the dry season. In general, the spatial distribution of nitrate concentrations in the Kabul aquifer in the dry and wet seasons does not reveal any clear picture. About 70% of the groundwater samples showed nitrate concentrations exceeding 2.5 mg L^{-1} as NO_3−N, the threshold considered as a result of anthropogenic activity (Panno et al., 2006), and about 6% of the groundwater samples showed nitrate concentrations above the threshold value (50 mg L^{-1}) as set by the WHO (WHO, 2017). This highlights the significant influence of anthropogenic NO_3 inputs on groundwater quality in the Kabul aquifer. Decreasing nitrate concentration and increasing $\delta^{15}N$-NO_3 value in the northern part of the study area in the wet season may be associated with volatilization of ^{15}N-depleted ammonia and mixing processes (Kendall et al., 2007; Biddau et al., 2019). The low NO_3 concentration in sample W12 could be also due to low NH_4^+ concentrations (Fadhullah et al., 2019). The high $\delta^{15}N$-NO_3 values (>10‰ in average) were detected in most collected groundwater samples in both dry and wet seasons, suggesting that sewage and to a lesser extent manure inputs were probably responsible for the high nitrogen isotope ratios of nitrate in the urban aquifer of the Kabul city. Overall, higher $\delta^{15}N$-NO_3 values were typically detected in the wet season. It could be explained by volatilization processes that may affect sewage and NH_4^+ fertilizer and mixing processes (Shalev et al., 2015; Biddau et

al., 2019; Torres-Martínez et al., 2021). Generally, higher $\delta^{18}O\text{-}NO_3$ values were observed in the dry season compared with the wet season. This pattern may be associated with evaporation in the dry season (Kendall et al., 2007; Shalev et al., 2015). From this, it can be concluded that the isotopic composition of nitrate in the study area is to an extent impacted by the hydrological regime.

3.5. Using dual isotopes to identify nitrate sources

In this study, $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ values (Fig. 6) were employed to identify the dominant potential sources of nitrate in the Kabul aquifer (Kendall et al., 2007). Almost all groundwater samples plotted in manure/sewage waste area, which suggests that sewage and septic wastes, and to lesser extent manure are the main contributors of nitrate in the plain with significant urban land use (Fig. 6). Almost all groundwater samples have $\delta^{15}N\text{-}NO_3$ values above +7‰ in the dry and wet seasons, which suggests that most of the nitrate in the Kabul aquifer is derived from sewage sources (Fig. 6). Chemical fertilizers did not appear to be a considerable contributor, although $\delta^{15}N\text{-}NO_3$ values below +7‰ indicate a mixture of sewage waste and soil N in wells W3 and W4, and sewage and ammonium fertilizer in well W21. It is worth mentioning that a mixture of nitrate from different sources is in good agreement with land-use data (Fig. 2), where different N source types coexist. Therefore, NO_3 in the Kabul Plain should primarily be influenced by the mixing of at least three sources (Sewage, soil organic N and chemical fertilizers). The NO_3^-/Cl molar ratios and $\delta^{15}N\text{-}NO_3$ data also were used to identify the major sources of nitrate in the Kabul Plain (Yue et al., 2014). According to Fig.7b, most of the water samples in both seasons displayed relatively high NO_3^-/Cl molar ratios with $\delta^{15}N\text{-}NO_3$ values greater than 8‰, suggesting that NO_3 was originated mainly from nitrification sewage and septic wastes.

3.6. Biogeochemical processes controlling nitrate fate

Besides the influence of various nitrate sources that have characteristics of isotopic compositions, the isotopic composition of groundwater nitrate is also affected by biogeochemical processes. A theoretical $\delta^{18}O\text{-}NO_3$ value produced by microbial nitrification in an aerobic environment was determined using the

$\delta^{18}O$-H_2O values measured for the surface and groundwater in the Kabul Plain (Table S1 in the supplementary material), assuming $\delta^{18}O$ of atmospheric O_2 (23.5‰) based on the following equation (Anderson and Hooper, 1983, Kendall et al., 2007).

$$\delta^{18}O\text{-}NO_3 = 2/3(\delta^{18}O\text{-}H_2O) + 1/3(\delta^{18}O\text{-air})$$

For simplification of the equation, we did not consider any potential oxygen isotope exchange of intermediates with the ambient water during nitrification as proposed by Casciotti and Buchwald (2012). As shown in Fig. 7a, the calculated oxygen isotopic compositions for most samples in the wet season plot are close to the theoretical nitrification trend, which suggests that nitrification is the main process forming dissolved nitrate in the Kabul Plain. The DO concentrations also confirm that the nitrification process could be the major source of nitrate in the Kabul Plain (Table S1). For the other groundwater samples, the $\delta^{18}O$-NO_3 values plot far from the theoretical nitrification line, suggesting that other processes are also involved (Fig. 7a). Some samples (LR, W25, W9, and W6) showed slightly lower $\delta^{18}O$-NO_3 values than the theoretical nitrification line. This observation might be explained by oxygen isotope exchange between intermediates during nitrification and ambient H_2O, which could alter the oxygen isotopic composition of nitrate in soil and lead to low values of $\delta^{18}O$-NO_3 in water (Kool et al., 2011; Wexler et al., 2012). A few samples tended to exhibit slightly higher $\delta^{18}O$-NO_3 values than to the theoretical nitrification line, which could be attributed to the incorporation of oxygen from water with high $\delta^{18}O$-H_2O values (because of evaporation of soil waters or bacterial respiration) (Yue et al., 2014; Biddau et al., 2019; Torres-Martínez et al., 2021). Moreover, Fig. 7a shows that $\delta^{18}O$-NO_3 values for water samples of the Kabul Plain are far below the atmospheric oxygen line characteristic for atmospheric nitrate, suggesting that nitrification is the main process forming dissolved nitrate in the Kabul aquifer (Meghdadi and Javar, 2018, Yang et al., 2020; Torres-Martínez et al., 2021).

The $\delta^{15}N$-NO_3 values used in combination with of NO_3^-/Cl molar ratios were also employed to investigate microbial processes in the Kabul Plain. As shown in Fig. 7b, most samples from the dry and wet seasons plot in a range characteristic of nitrification, indicating that nitrification is the main process forming

dissolved nitrate in the Kabul aquifer (Table S1). Two samples (KR and W21) revealed medium NO_3^-/Cl molar ratios with $\delta^{15}N\text{-}NO_3$ values lower than 5.5‰, suggesting that NO_3 may be derived from ammonium fertilizer in the dry season, which was also confirmed by land-use information (Fig. 2). A few samples in both dry and wet seasons displayed medium to high NO_3^-/Cl molar ratios with $\delta^{15}N\text{-}NO_3$ values between 5‰ and 8‰, suggesting that NO_3 was primarily derived from soil organic N (Fig. 7b) (Torres-Martínez et al., 2021). Most of the samples in both dry and wet seasons exhibited relatively high NO_3^-/Cl molar ratios with $\delta^{15}N\text{-}NO_3$ values greater than 8‰, indicating that NO_3 was affected directly by sewage and septic wastes (Fig. 7b). Moreover, some samples in the central and north-west of the eastern plain showed relatively low NO_3^-/Cl molar ratios with $\delta^{15}N\text{-}NO_3$ values greater than 15‰ (Fig. 7b), suggesting that NO_3 may originate from sewage waste and also that ammonia volatilization and mixing processes might have occurred in the mentioned areas (Kendall et al., 2007; Widory et al., 2013; Biddau et al., 2019; Torres-Martínez et al., 2021).

Further analysis of the potential sources of nitrate in the Kabul aquifer depends on the presence/absence of denitrification. Denitrification causes a significant reduction of NO_3 concentrations and increases $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ values of the residual NO_3 (Böttcher et al., 1990; Rivett et al., 2008; Hocaoglu et al., 2011). Regression analyses reveal that $\delta^{15}N\text{-}NO_3$ had a weak positive relationship with $\delta^{18}O\text{-}NO_3$ in the dry season ($R^2 = 0.16$, $p < 0.05$, slope = 0.65, n = 28) and in the wet season ($R^2 = 0.22$, $p < 0.05$, slope = 0.46, n = 31) (Fig. 6). The slope of the linear regression line for the dry season was within the reported ranges of denitrification (0.5 – 1) (Mayer et al., 2002; Kendall et al., 2007), however, the water samples were not plotted along the 2:1 or 1:1 denitrification line. Therefore, the volatilization and mixing processes might be responsible for the slight isotopic enrichment of $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ during the dry season (Biddau et al., 2019, Yue et al., 2014). Overall, denitrification processes did not have a significant impact on the nitrate isotopic composition in the Kabul Plain in the dry and wet seasons (Ding et al., 2014). DO concentrations (Table S1) also confirm the limitation of denitrification processes in the Kabul aquifer (Orginc et al., 2019). However, a recent study by Utom et al. (2020) indicated that the

denitrification process does not always happen under strictly anoxic conditions, as it is commonly assumed.

Moreover, the relationships of $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ versus $1/[NO_3^-]$ were employed to further assess the mixing and denitrification processes that may have occurred in the Kabul aquifer. The Keeling plots (Fig. 8a-b) displayed concentrations of NO_3 versus $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$, respectively in the dry and wet seasons. The $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ values of water samples showed only weak correlations with the inverse NO_3 concentrations ($R^2 = 0.005$, $p > 0.05$ and $R^2 = 0.35$, $p < 0.05$) and ($R^2 = 0.012$, $p > 0.05$ and $R^2 = 0.035$, $p > 0.05$) in the dry and wet seasons, respectively. The plots for water samples exhibited that $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ were negatively correlated with $1/[NO_3^-]$ confirming the limitation of denitrification processes in the Kabul aquifer. Weak negative relationships (Fig 8a-b) could be explained as the mixing of various nitrate sources such as sewage with soil organic nitrogen and chemical fertilizer (Romanelli et al., 2020). Overall, the data suggested that denitrification did not significantly influence the nitrate dynamics in the Kabul aquifer in the dry and wet seasons.

3.7. Quantification of the dominant sources of nitrate by SIMMR model

In order to quantitatively estimate the proportional contributions of various potential nitrate sources to the Kabul Plain, the Bayesian stable isotope mixing model (SIMM) in R with the SIMMR package was employed. Two isotopes (J=2) ($\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3^-$) and three potential sources of nitrate (K=3) (sewage and manure (S&M), soil organic N (SN) and chemical fertilizers (CF)) were employed. In this study, denitrification is not thought to have a major impact on the isotopic composition of NO_3^-, thus the fractionation factor is considered zero (Fadhullah et al., 2019; Zhang et al., 2020). According to the SIMMR, the contributions of the three potential NO_3^- sources were quantified, suggesting a considerable variability in the different seasons and areas (Table S1 and Fig. 9a and 9b). The $\delta^{15}N\text{-}NO_3$ and $\delta^{18}O\text{-}NO_3$ values of the nitrate sources were based on the relevant literatures (Xue et al., 2009; Nikolenko et al., 2018; Torres-Martínez et al., 2020). Based on the SIMMR output results, sewage had the highest contribution (~81%), followed by soil organic nitrogen (10.5%), and chemical fertilizer (8.5%) in the dry

season. In the wet season, sewage (~87.5%), soil organic N (6.7%), and chemical fertilizer (5.8%) contributed in a similar proportion to the nitrate pool in the Kabul aquifer. The SIMMR results suggest that sewage was the dominant source of NO_3 in the Kabul aquifer. It reflects the influence of rapid urbanization in the Kabul Plain. The contribution of NO_3 from urban sewage to the Kabul aquifer is expected to increase due to the growing population, lack of sewerage collection system and wastewater treatment plants in the city. The sources of NO_3 estimated by SIMMR were quite reasonable and in good agreement with qualitative analysis of dual nitrate isotopes of nitrate and in-situ observations.

4. Conclusion

An integrated approach that combined hydrogeochemical data, stable isotope signatures of nitrate and water, and a stable isotope mixing model (SIMMR) were employed to determine potential sources of nitrate, biogeochemical processes and seasonally proportional contributions of nitrate sources in the Kabul aquifer. The results indicated that NO_3 concentrations in the Kabul aquifer varied from place to place in urban land use and were significantly influenced by localized anthropogenic activities. The results of water stable isotope analyses indicated that rainfall and river water after evaporation were the major source of groundwater in the Kabul Plain. The dual stable isotope signatures of nitrate revealed that nitrification of sewage and human wastes was the main nitrate pollution source in the aquifer, which was confirmed by hydrogeochemical data. Nitrification was identified as a main biogeochemical process in the aquifer. The spatio-temporal variability in the proportional contribution of nitrate sources was determined by a Bayesian stable isotope mixing model (SIMMR). In this respect, sewage was the main contributor of groundwater nitrate in the Kabul aquifer in the dry and wet seasons followed by soil organic nitrogen and chemical fertilizers. This is the first detailed nitrate pollution study in Afghanistan employing an integrated approach. The combined approach of isotopic the composition of nitrate, hydrochemical and land use data and SIMMR model allowed a more reliable assessment of the nitrate sources, biogeochemical processes and nitrate source apportionment in groundwater of the Kabul aquifer. Continuous rehabilitation of existing sewerage collection systems and constructing decentralized on-site

treatment systems in new townships are important ways to reduce NO_3 pollution in the Kabul aquifer. A long-term monitoring of groundwater quality in the Kabul aquifer is strongly advised.

CRediT authorship contribution statement

Abdulhalim Zaryab: Conceptualization, Methodology, Investigation, Software, Data curation, Formal analysis, Writing – original draft, Writing – reviewing & editing, Visualization. **Hamid Reza Nassery:** Conceptualization, Methodology, Formal analysis, Supervision, Validation, Writing – reviewing & editing. **Kay Knoeller:** Conceptualization, Methodology, Resources, Supervision, Validation, Writing – reviewing & editing. **Farshad Alijani:** Conceptualization, Methodology, Software, Formal analysis, Visualization, Validation, Writing – reviewing and editing. **Eddy Minet:** Conceptualization, Methodology, Visualization, Validation, Writing – reviewing and editing.

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Appendix: Supplementary material

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Table 1. Summary of $\delta^{15}N\text{-}NO_3^-$, $\delta^{18}O\text{-}NO_3$ isotopic signatures, from some possible end-members employed in the SIMMR model (Zhang et al., 2018; Torres-Mrtínez et al., 2020; Torres-Mrtínez et al., 2021)

Source	**Dry season** $\delta^{15}N\text{-}NO_3$ **Mean ± SD**	**Wet season** $\delta^{18}O\text{-}NO_3$ **Mean ± SD**	**Dry season** $\delta^{15}N\text{-}NO_3$ **Mean ± SD**	**Wet season** $\delta^{18}O\text{-}NO_3$ **Mean ± SD**
Sewage and Manure (S&M)	12.20 ± 3.53	4.87 ± 1.87	13.2 ± 3.73	4.38 ± 3.72
Soil Organic Nitrogen (SN)	3.98 ± 1.95	2.51 ± 1.41	3.98 ± 1.82	2.47 ± 1.41
Chemical fertilizer (CF)	0.90 ± 2.00	2.80 ± 1.70	0.90 ± 2.0	2.60 ± 1.90

Table 2. Statistical summary of the physical, chemical, and isotopic results on sampling point in the Kabul Plain

Parameter	Unit	Dry season (November 2020)				Wet season (April 2021)			
		Min	Mean	Max	Std Dev	Min	Mean	Max	Std Dev
pH	-	7.00	7.60	8.10	0.23	7.10	5.26	8.30	1.59
T	ºC	6.00	15.40	17.80	2.10	12.70	16.15	18.00	1.47
DO	mg/L	2.10	5.23	9.60	1.88	1.90	5.26	8.30	1.59
EC	µS/cm	618.0	1640.0	16400	3827.60	458.0	1429.50	16430	3653.90
Ca^{2+}	mg/L	20.50	79.45	727.30	131.10	22.40	57.30	365.00	68.55
Mg^{2+}	mg/L	12.30	82.40	738.00	153.10	11.40	57.00	520.20	130.60
Na^{+}	mg/L	20.50	94.15	3014.0	672.85	15.56	71.68	2374.80	540.58
K^{+}	mg/L	2.16	7.05	74.00	15.40	2.10	5.58	85.80	18.44
HCO_3^-	mg/L	183.0	429.9	840.0	170.60	183.0	393.45	802.76	163.87
Cl^-	mg/L	7.30	135	3997	838.60	6.75	89.29	3163.40	622.77

SO_4^{2+}	mg/L	19.80	114	4573	1074.23	16.32	102.24	4582.10	955.85
NO_3^-	mg/L	3.98	13.7	120.41	21.30	4.87	17.71	58.88	13.24
NO_2^-	mg/L	0.01	0.20	2.80	0.55	0.01	0.03	0.36	0.11
$\delta^{15}N\text{-}NO_3^-$	‰AIR	4.80	10.20	20.80	3.98	6.90	12.00	25.40	4.28
$\delta^{18}O\text{-}NO_3^-$	‰VSMOW	-11.70	7.20	18.60	6.41	-3.40	4.00	11.50	3.55
$\delta^{2}H\text{-}H_2O$	‰VSMOW					-73.40	-52.00	-30.30	7.67
$\delta^{18}O\text{-}H_2O$	‰VSMOW					-10.59	-8.31	-6.39	0.90
NH_4^+	mg/L	0.01	0.04	53.05	9.83	0.00	0.60	109.80	63.09

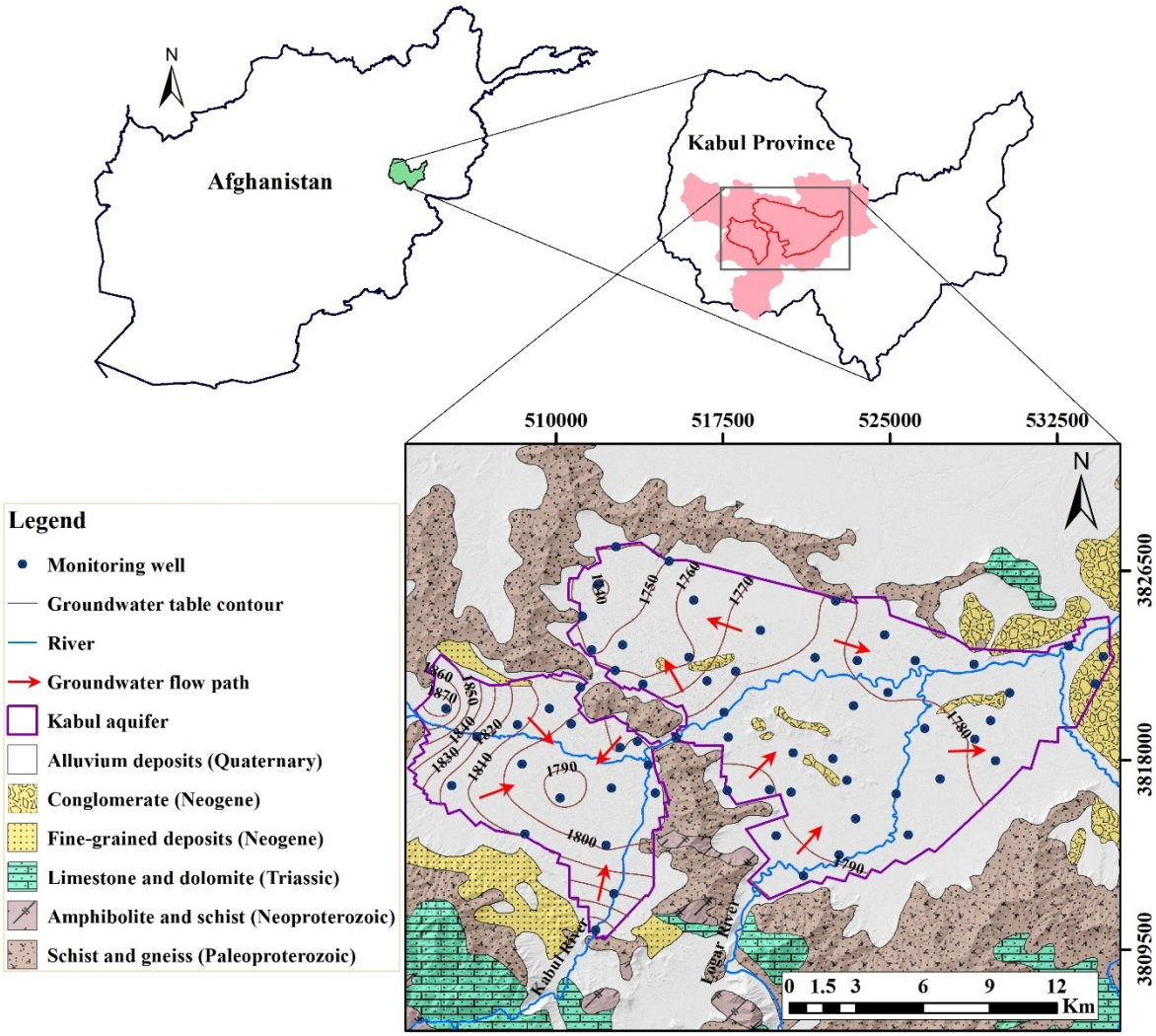


Fig. 1. Location of Kabul aquifer with simplified geological map (Houben et al 2009; Bohannon 2010). Groundwater table contours (August 2020) and flow directions (red arrows) are shown. The labeled contour lines represent the elevation of the groundwater table.

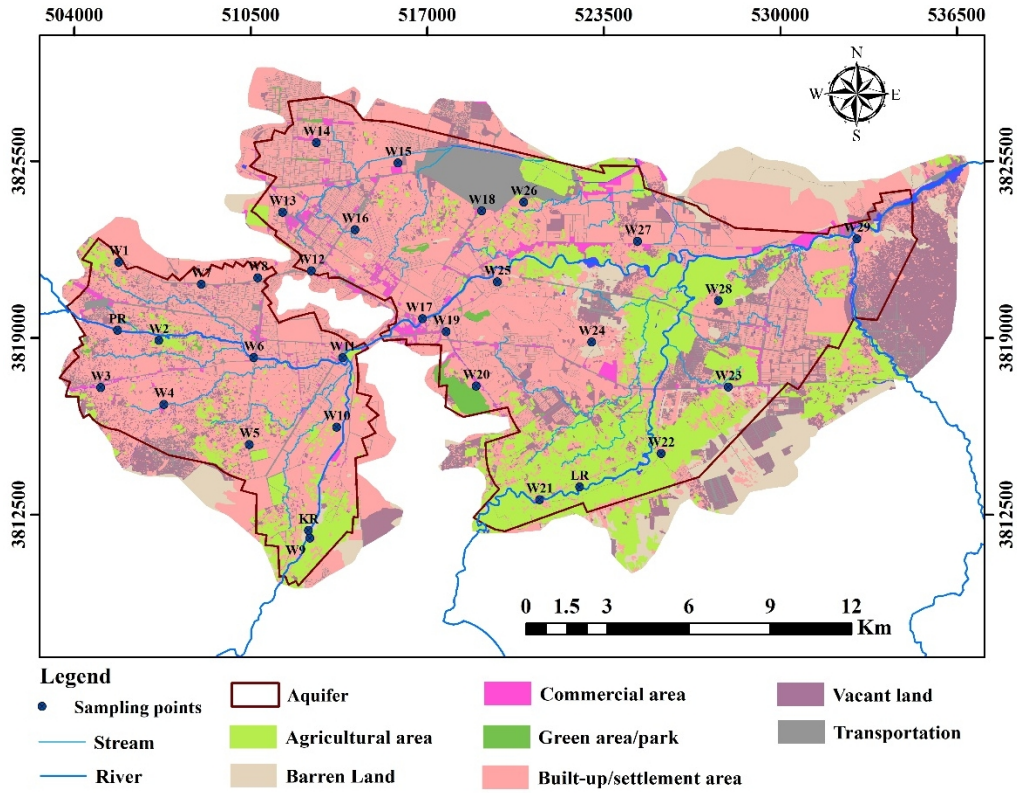


Fig. 2. Land use map of the study area (Kabul city, Afghanistan) and locations of sampling points.

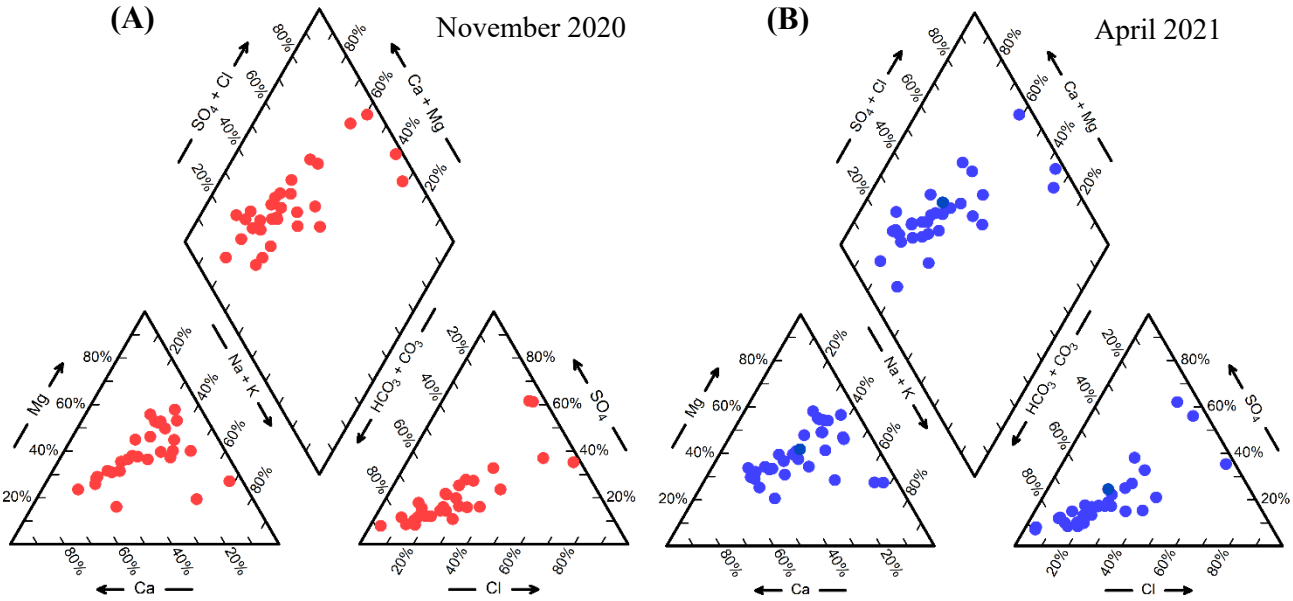


Fig. 3. Piper plot showing the chemical compositions of groundwater from the Kabul aquifer in (A) November 2020 and (B) April 2021

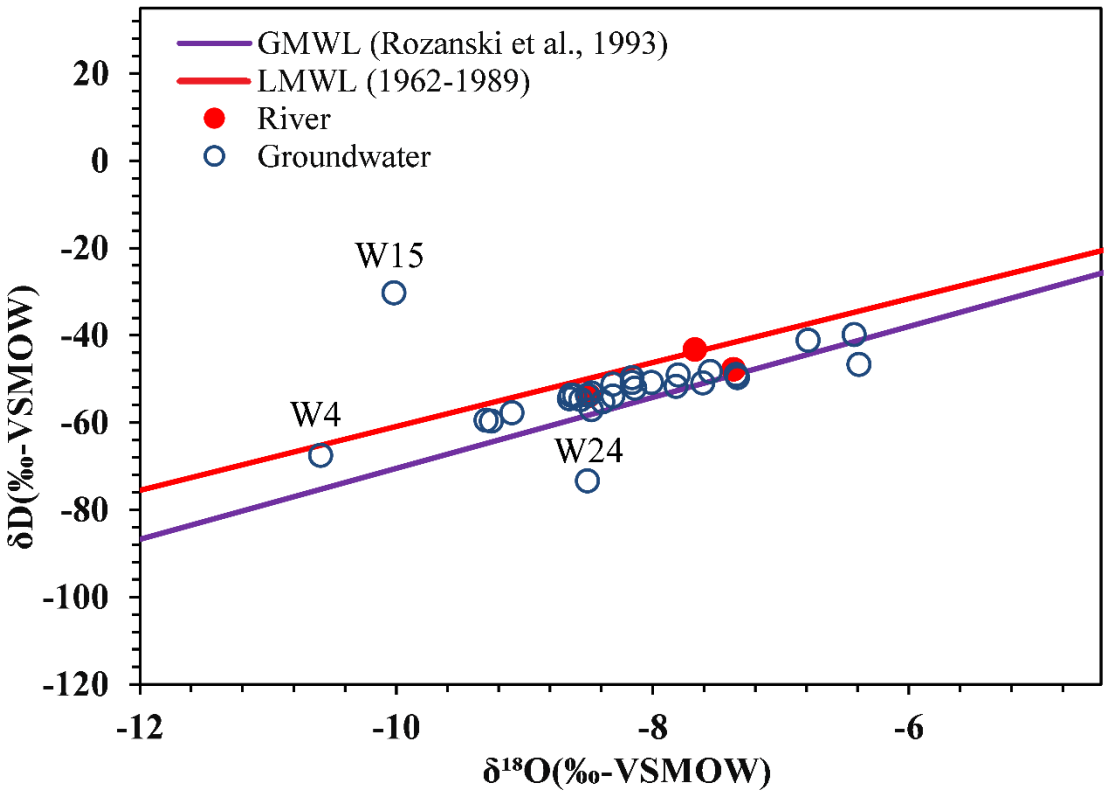


Fig. 4. δ^2H-H_2O vs $\delta^{18}O$-H_2O plot for the water samples (groundwater and river water) collected from the Kabul Plain in November 2020 and April 2021.

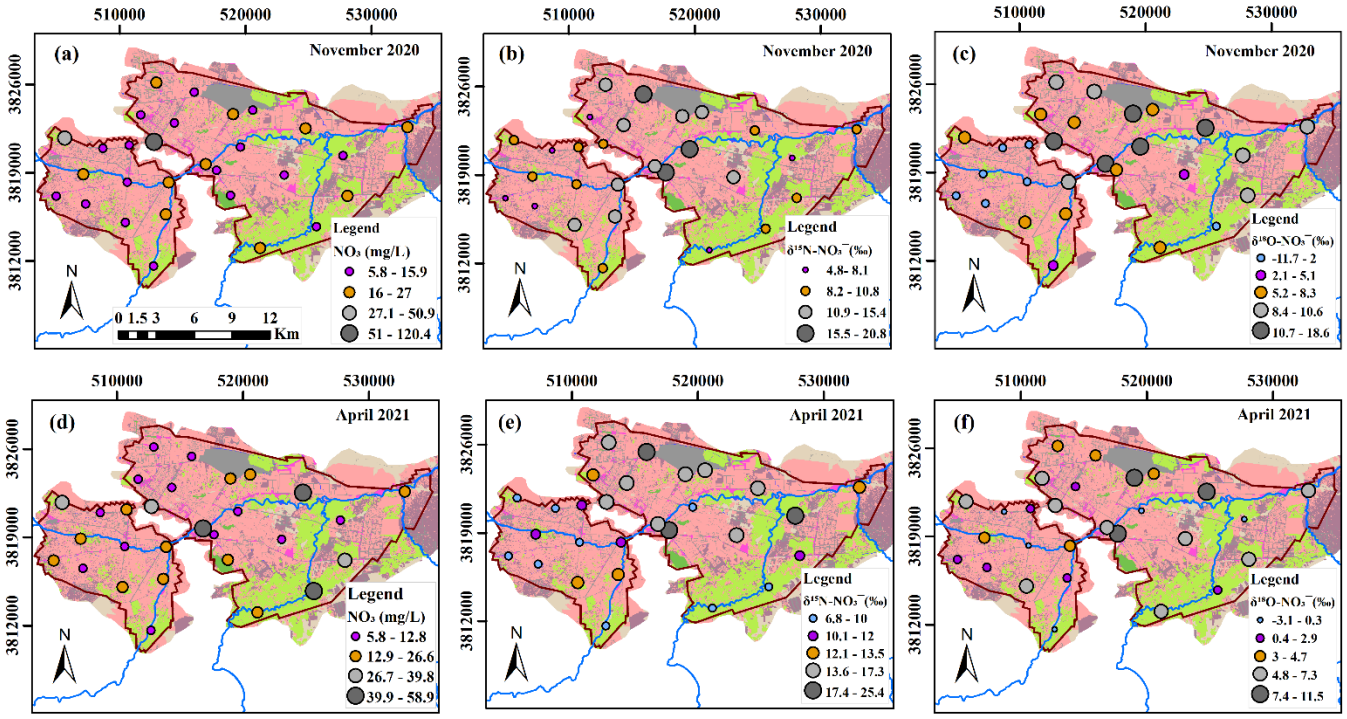


Fig. 5. Spatial variations of (a and d) nitrate concentrations, (b and e) $\delta^{15}N\text{-}NO_3^-$ and (c and f) $\delta^{18}O\text{-}NO_3^-$ values of groundwater samples from the Kabul aquifer collected in the dry season (November 2020) and wet season (April 2021)

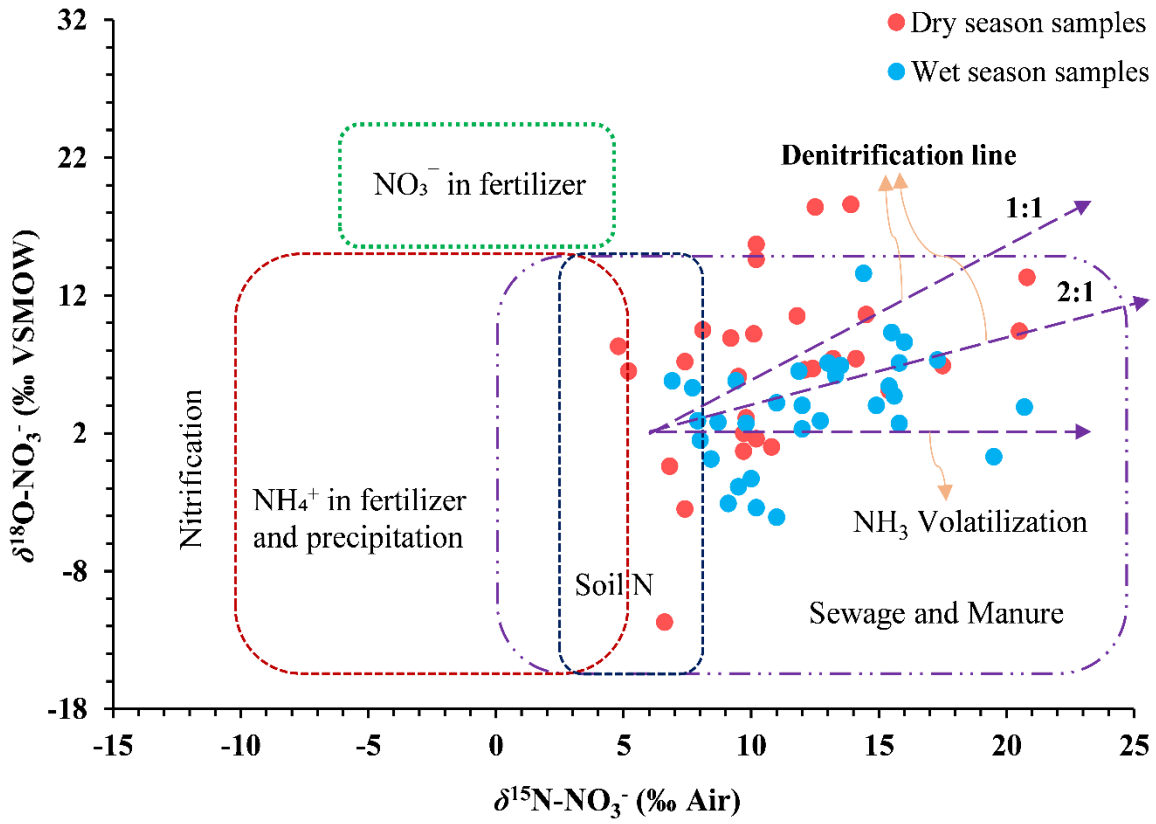


Fig. 6. $\delta^{15}N\text{-}NO_3$ versus $\delta^{18}O\text{-}NO_3$ in water samples from the Kabul Plain. The typical ranges of isotopic compositions are derived from Kendall et al. (2007).

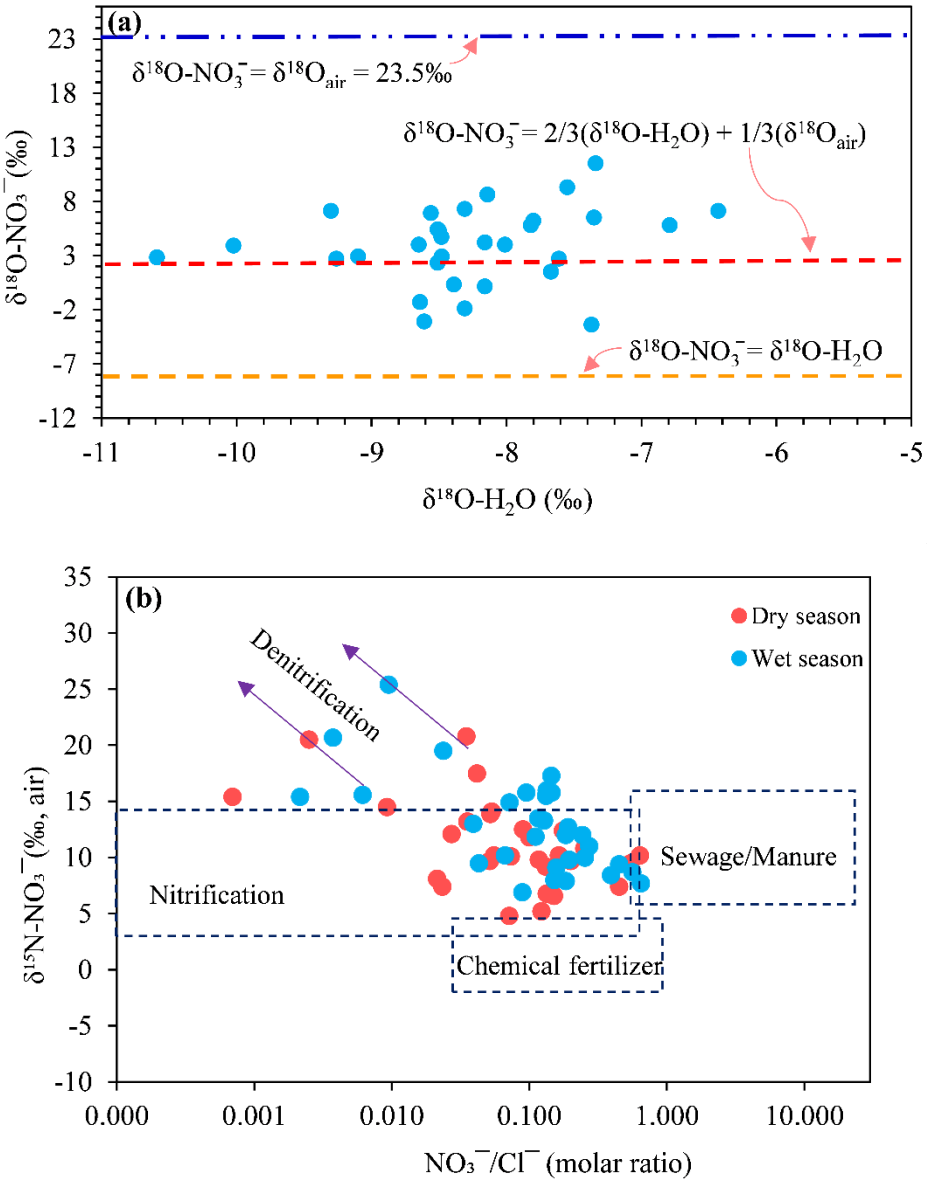
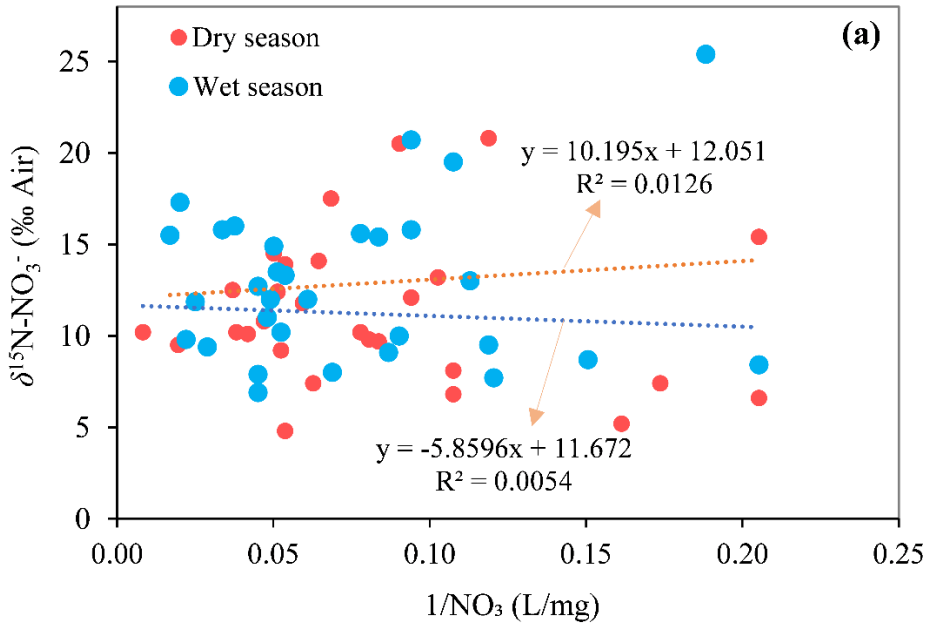


Fig 7. Diagrams for identification of nitrification and denitrification processes. (a) Shows $\delta^{18}O$-H_2O versus $\delta^{18}O$-NO_3 in water samples from the Kabul Plain, there lines represent the theory line in different conditions and (b) Presents $\delta^{15}N$-NO_3 versus NO_3^-/Cl molar ratio in collected water samples from the Kabul Plain.



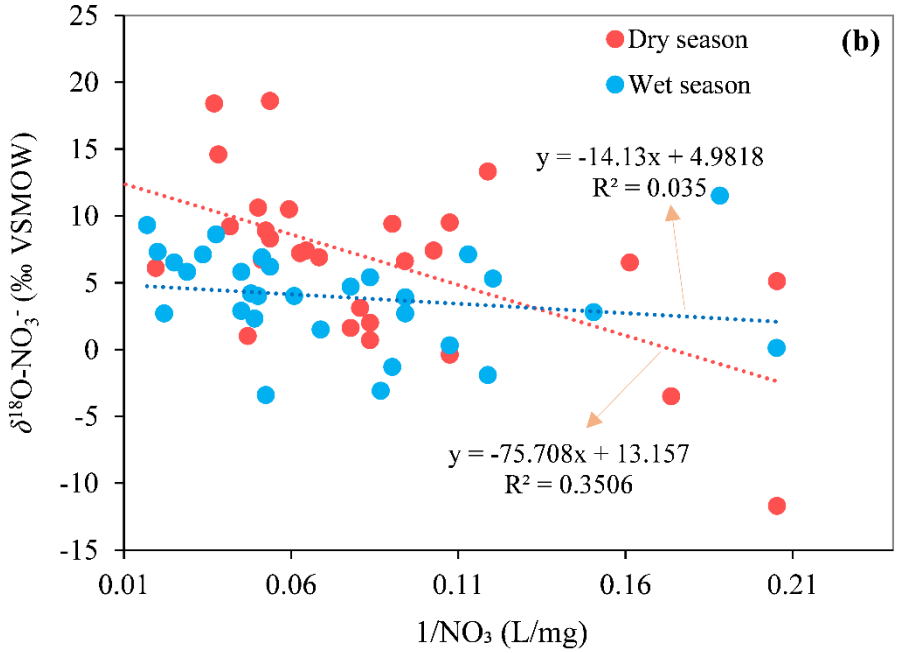
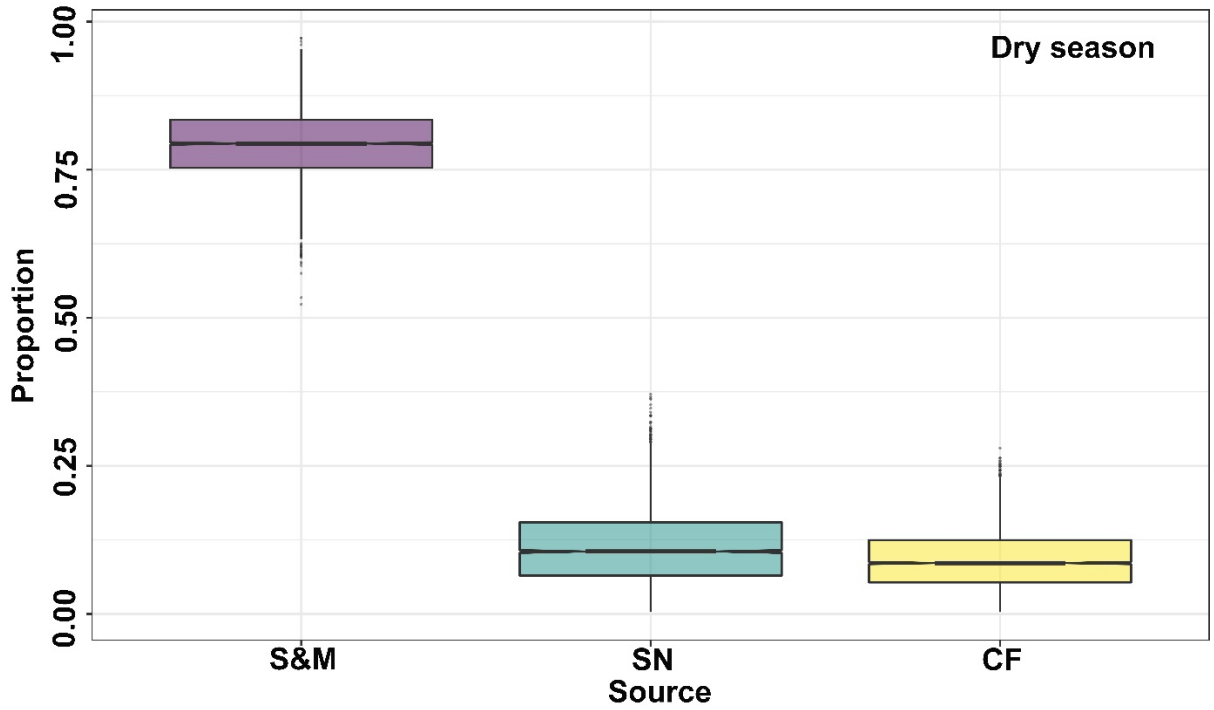


Fig. 8. Relationship between (a) δ^{15}N-NO_3 and (b) δ^{18}O-NO_3 versus 1/ [NO_3^-] in water samples from the Kabul Plain



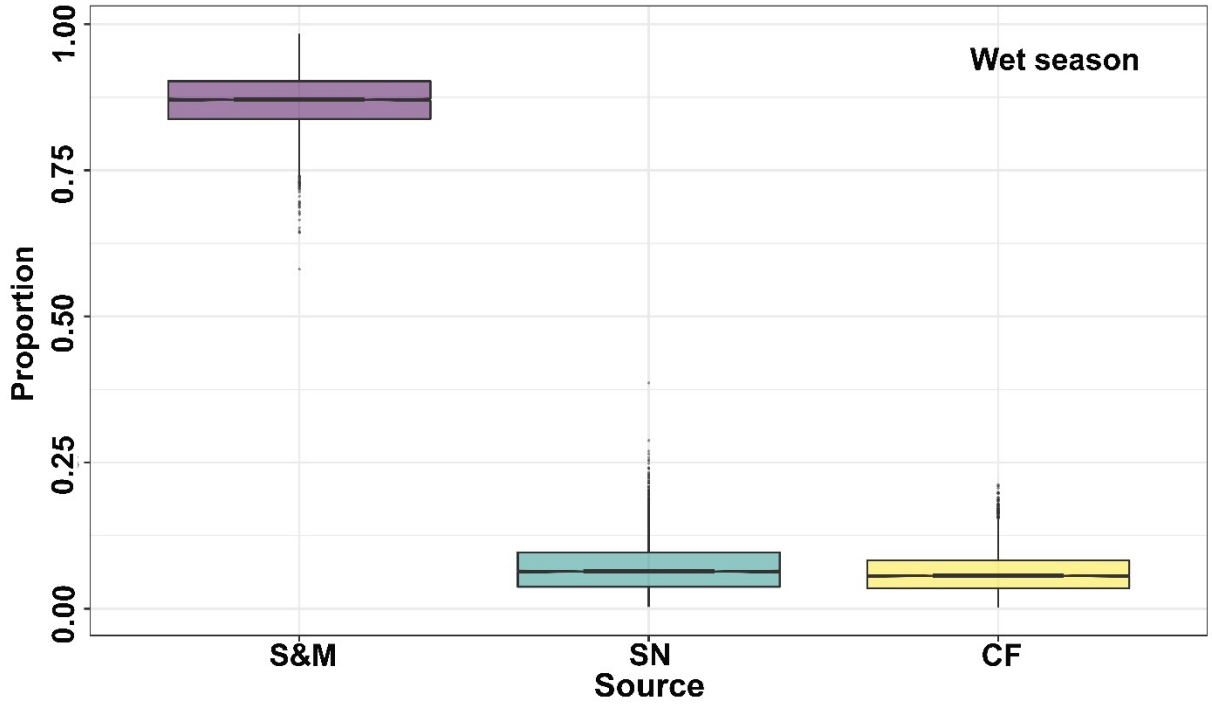


Fig 9. Seasonally proportional contributions of three protentional sources of NO_3^- in the Kabul aquifer estimated using the SIMMR model (in percentage). Boxplots illustrate the credible interval of 50%

Graphical abstract

Highlights

- Isotopic analysis was combined with land-use and hydrochemical data to identify the main sources of nitrate in the Kabul aquifer.
- High Cl^- concentrations and low NO_3^-/Cl^- ratios indicated that sewage was the dominant nitrate source in the Kabul aquifer.
- Nitrate processes in the groundwater were mainly derived from nitrification, volatilization.
- Bayesian stable isotope mixing model: Highest contribution by sewage.