



Use of hydrochemical and isotopic tracers to investigate the groundwater quality and recharge processes of the shared Iullemeden Aquifer System in the Sahel region (Western Africa)

K. Zouari¹ · R. Trabelsi¹ · Luis L. Araguás Araguás² · S. Hussaini³ · S. Rabe⁴ · A. Alassane⁵

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Abstract

The management of water resources across many parts of Africa is facing significant challenges in quantitative and qualitative terms. Meeting the increase in water demand, linked to population growth and economic development, is affected by the limited resources due to climate variability, often-inefficient management of water resources, and quality degradation caused by some polluting domestic and agricultural practices. This investigation of the Iullemeden Aquifer System (IAS), which is shared between Niger, Nigeria, Mali, Benin and Algeria, is based on the combined use of hydrochemical and environmental isotope methods. The aim is to improve understanding of the IAS hydrogeological functioning, including groundwater flow patterns, mechanisms of recharge, and vulnerability to pollution, as well as the interaction between surface waters and the most significant aquifer units. Assessment of groundwater recharge in this arid and semiarid area is complex due to the temporal and remarkable spatial variability of rainfall and recharge processes. In the IAS, groundwater recharge was highly heterogeneous, mainly occurring during the late Pleistocene and humid periods of the Holocene, particularly for the Continental Intercalary and Continental Terminal aquifers. The heterogeneity of groundwater residence time is reflected in the considerable spatial variation of $\delta^{18}\text{O}$, $\delta^2\text{H}$, ^3H , d-excess, ^{14}C and total dissolved solids (TDS) contents.

Keywords Arid regions · recharge · Palaeorecharge · Isotopes · Africa

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✉ K. Zouari
kamel.zouari@enis.tn

¹ National School of Engineers of Sfax, Geological Department, Laboratory of Radio-Analysis and Environment, University of Sfax, BP 1173, 3038 Sfax, Tunisia

² International Atomic Energy Agency, Isotope Hydrology Section, Wagramer Strasse 5, PO Box 1001400, Vienna, Austria

³ Nigeria Hydrological Services Agency (NIHSA), Plot 222, Shetimma Ali Monguno Crescent, Foundation Plaza, Abuja 900001, Nigeria

⁴ Ministry of Hydraulic and Environment, General Water Authority, BP 257, 8001 Niamey, Niger

⁵ University Abomey-Calavi, National Institute of Water, BP 4521, Cotonou, Benin

Introduction

Constituting one of the most important basins in the Sahel region, covering an area of ~525,000 km², the Iullemeden Aquifer System (IAS) is shared by Niger (82%), Nigeria and Mali, with minor areas in Algeria and Benin (OSS 2008, 2011a). The IAS extends for ~1,000 km from 10 to 19°N and about 980 km from 1 to 10°E (Fig. 1). It corresponds to the area of the “Middle Niger” River basin. The current population in this area is ~15 million, with 65% in Niger, 34% in Nigeria and less than 2% in Mali, Benin and Algeria.

The Iullemeden basin presents important water resources in the form of large rivers and lakes, vast wetlands, and widespread groundwater resources. The IAS is multilayered, including the Cretaceous Continental Intercalary sedimentary aquifer, overlain by Tertiary layers of Continental Terminal formations. It constitutes the primary perennial drinking water source and a strategic resource for the sustainable development of the concerned countries, estimated to be about 5,000 km³ (OSS 2011b). Due to the scarcity of

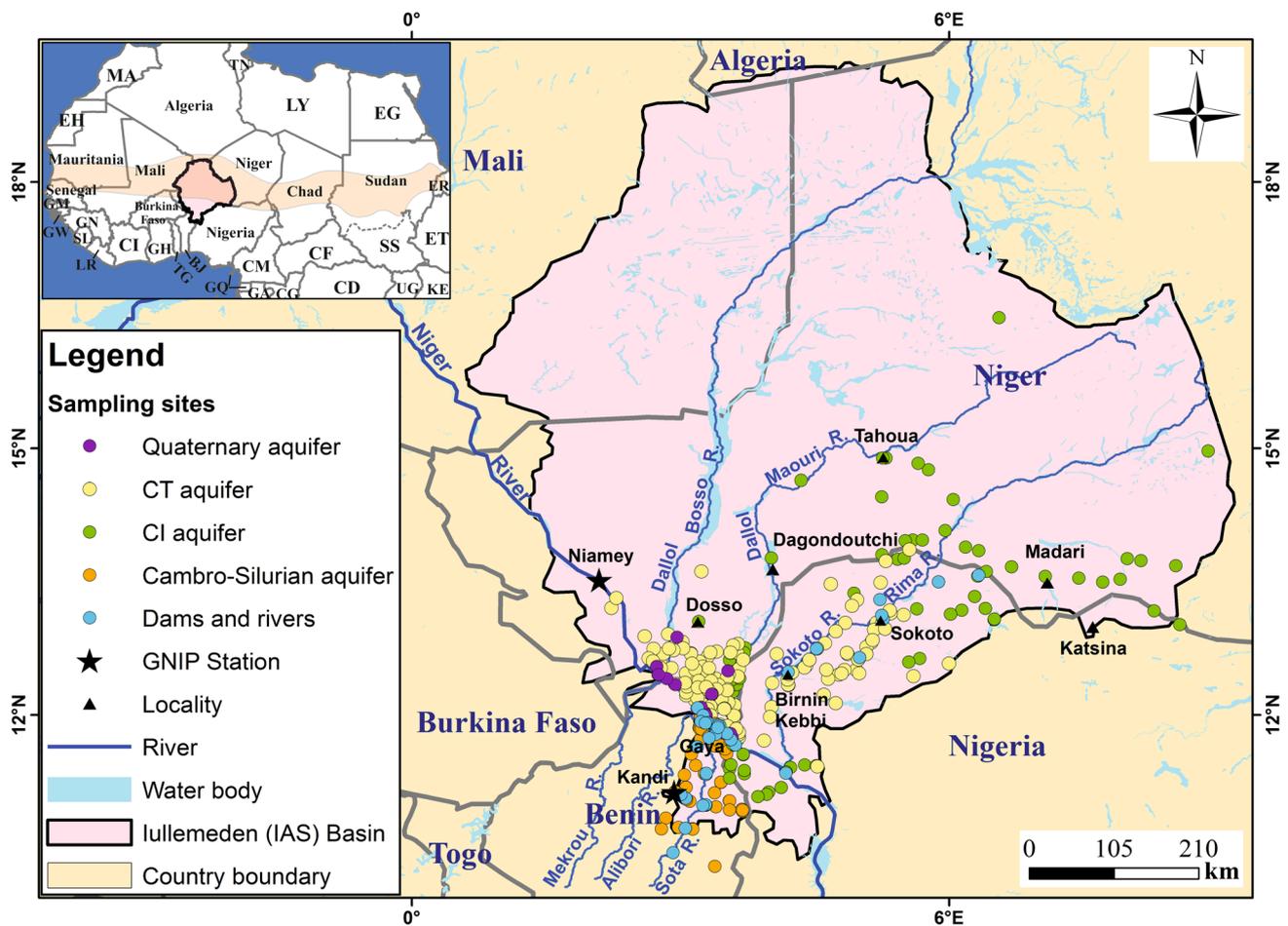


Fig. 1 Location map of the Iullemeden basin showing the locations of collected water samples

surface waters in the region, most of the available freshwater is found in transboundary aquifer systems; thus, the populations in the IAS basin depend more and more on groundwater as their primary source of water supply for domestic, industrial and agricultural demand. As a result, the number of wells and irrigation systems in shallow aquifers has increased during the last few decades with no or inadequate regulation and institutional arrangements for groundwater abstraction. This inappropriate governance has caused intensive exploitation, leading to a marked lowering of groundwater levels and a subsequent decline in the available water quantity. Moreover, one major problem with respect to the IAS is the assessment of recharge mechanisms and rates through (1) direct infiltration of rain, (2) lateral seepage of surface water, and (3) palaeorecharge by upward leakage from deep aquifers. In addition, the deterioration of groundwater quality has been highlighted around urban centres and areas of intensive agriculture (Girard and Hillaire-Marcel 1997). High nitrate contents and incipient salinization are significant water quality issues of growing concern (Girard and Hillaire-Marcel 1997). This situation is worsened by the

natural risks introduced by spatial and temporal variability of rainfall, combined with climate change, causing rising water scarcity and shrinking of some water bodies. In fact, this region is subject to alternating wet and dry seasons, and the area has been suffering from recurrent droughts since the early 1980s.

Previous isotopic studies have been performed in the Iullemeden basin to investigate the circulation dynamics of old groundwater, paleoclimate and groundwater flow patterns, and residence times in some aquifers (Andrews et al. 1994; Le Gal La Salle and Fontes, 1995; Beyerle et al. 2003; Kpegli et al. 2015). The present study is the first regional geochemical investigation that concerns the different aquifers of the Iullemeden basin.

The present work was carried out within the framework of an interregional project (RAF/7/011 and RAF/7/019) supported by the International Atomic Energy Agency (IAEA). The project aimed to better characterize the hydrogeology of the principal shared aquifers in the Sahel region, including the IAS, with the final aim of promoting the joint management of water resources by the concerned countries. This

investigation is based on the combined uses of hydrochemical and isotope hydrology methods (stable and radioactive, $\delta^{18}\text{O}$, $\delta^2\text{H}$, ^3H , $\delta^{13}\text{C}$, and ^{14}C). These geochemical tools have provided new hydrological information on the groundwater origin, surface-water/groundwater interaction, and recharge processes.

General setting of the Iullemeden aquifer system

The Iullemeden basin is characterized by a tropical arid and semiarid climate, with a long dry season and a short and irregular wet season from June to September (Le Gal La Salle et al. 2001). The rainfall distribution in the Iullemeden basin in Niger presents a marked strong gradient: less than 50 mm/year in the north to over 800 mm/year in the south. The positions of the average annual isohyets suggest four climatic zones: (1) the Saharan zone (less than 150 mm); (2) the nomadic Sahelian zone (between 150 and 300 mm/year); (3) the sedentary Sahelian zone (between 300 and 600 mm/year); and (4) the Sahelo-Sudanian zone (between 600 and 800 mm/year; OSS 2011a; IAEA 2017).

The Iullemeden basin is prone to extended droughts. A long-term reduction in rainfall is reflected in the displacement to the south of the 200 mm isohyet during the Sahel drought of 1970–1985. Consequently, the IAS is in one of the world regions most vulnerable to the impacts of extreme climate change, desertification and drought. These impacts are also expected to affect aquifer replenishment (OSS 2011a).

The sedimentary Iullemeden basin is bordered by the massif of Air to the north, the Adrar to the northwest and the Jos Plateau (in Nigeria) towards the south. In the east, along a line from the Jos Plateau to the Air chains, the basin is partly separated from the confined aquifers in the Chad Basin by the south–north Continental Dorsal. To the west, in Mali and Niger, the IAS is bounded by the Hamadien Sandstone and may be connected to the Tamesna extension basin at the west of the Adrar highlands through the Gao Trench. In the southwest, the basin limit follows the basement range along the Niger River (Fig. 2; IAEA 2017).

The IAS consists of sedimentary formations that range in age from the Cambrian–Ordovician to Tertiary and Quaternary (OSS 2011a; Figs. 2 and 3). The principal shared aquifers are the Continental Intercalary (CI) aquifer (Lower Cretaceous), the Continental Terminal (CT, Upper Cretaceous–Tertiary) and the Quaternary.

The CI aquifer is the most extensive multilayered aquifer system in the IAS basin. In Nigeria, this aquifer is composed of the Gundumi and Illo sedimentary formations of the Lower Cretaceous, which uncomfortably overlies the basement complex (Figs. 2 and 3). In both Mali and Niger,

from bottom to top, the CI aquifer includes the Tegama sandstone, Farak clays and the Continental Hamadien. The CI is continuous throughout the basin, subartesian in Nigeria, unconfined at its borders, and confined in Mali's central and western sectors. The main groundwater flow paths run from the northwest, north and northeast to the south. The main discharge of the CI is through the Niger River (OSS 2008).

The CT consists of alternating sands, sandstones, silts, and clay deposits (Beyerle et al. 2003) with frequent lateral and vertical changes of facies of the Tertiary sediments. In Niger, the CT is a multilayer aquifer system composed of three well-recognized sets of formations (Figs. 2 and 3; Guéro 2003):

1. The Continental Terminal 1 (CT1), which is a confined aquifer except for its outcropping part
2. The Continental Terminal 2 (CT2), a semiconfined aquifer characterized by radial convergent groundwater flow, along the NW–SE and NE–SW directions (OSS 2008, 2011a)
3. The Continental Terminal 3 (CT3), a mostly unconfined with the main drainage axes along the NW–SE and NE–SW directions

Impermeable sediments (mainly clayey sediments) separate the CT units from each other and the underlying CI aquifer (Andrews et al. 1994). Besides, this multilayer configuration of this aquifer disappears towards the discharge area in the southwest, close to the Niger River (Greigert 1966).

The groundwater main flow lines run from the northwest, north and northeast to the south. Discharge areas are the Niger and Goulbi Maradi rivers (OSS 2008). In Nigeria, the Continental Terminal aquifer is found in the Rima and Sokoto groups and the Gwandu Formation (Kogbe 1989; Fig. 2). The Rima Group (Upper Cretaceous) is subdivided into the Taloka, Dukamaje, and Wurno formations. The Dukamaje Formation is composed of shale units separating the two aquifer units of the underlying Taloka Formation from the overlying Wurno Formation. The Taloka Formation (Maastrichtian) is composed of semiconsolidated fine–medium-grained sand with a thickness of about 90 m. The Wurno aquifer is found at a depth of 80–120 m and consists of medium to coarse sand with a maximum thickness of 180 m. The Sokoto Group (Paleocene) consists of marine deposits with shales of the Dange Formation at the bottom and the limestone of the Kalambaina Formation at the top. The limestone formation of the Sokoto Group is the principal aquifer. The Gwandu Formation (Eocene–Miocene) comprises semiconsolidated fine to coarse sand and clay deposits. It is the best-known geological formation, where the four aquifer units have been identified. In the western part of the basin, the Gwandu Formation crops out over

Age			Niger		Nigeria		Benin	
			Group	Formation/ Lithology	Group	Formation/ Lithology	Group	Formation/ Lithology
Cenozoic	Quaternary	Holocene	Quaternary Aquifer	Alluvium deposits, dunes	Quaternary Aquifer	Alluvium deposits	Quaternary (Aquifer)	Alluvium deposits
		Pleistocene						
	Neogene	Pliocene	Continental Terminal CT3 Aquifer	Series of the clayey sandstone of the Middle Niger	Continental Terminal aquifer	Gwandu	Continental Terminal (not very productive aquifer)	Sand and claystones
			Aquitard					
		Miocene	Continental Terminal CT2 Aquifer	Clayey-sandy to lignite series				
			Aquitard					
	Paleogene	Oligocene	Continental Terminal CT1 Aquifer	Siderolithic series	Continental Terminal aquifer (Sokoto group)	Kalambaina Aquifer		
		Eocene	Aquitard					
		Paleocene	Marine Paleocene Aquitard	Sandstone, papyraceous schists				
Mesozoic	Cretaceous	Upper Cretaceous	Marine Cretaceous Aquitard	White sandstone	Continental Terminal aquifer (Rima Group)	Taloka Aquifer		
	Lower Cretaceous	Continental Intercalary (CI) Aquifer	Hamadian Continental, Clays of the Farak, Tégama sandstone	Intercalary/Hamadian Continental Aquifer	Gundumi & Illo Aquifer	Cretaceous Aquifer (Sende Formation)	Sandstones, siltstones and conglomerates	
Paleozoic	Silurian						Kandi - Aquifer	Siltstones and sandstones
	Ordovician							
	Cambrian						Were - Aquifer	Sandstones and conglomerates
Precambrian	Archean		Pan-African basement				Crystalline bedrock	Granite, Gneiss, Quartzites

Fig. 2 Litho-stratigraphy, showing the different geologic units of the IAS basin and indicating the aquifer layers

13,600 km² with sediments made up of interbedded semi-consolidated sand and clay units. Generally, the Gwandu and the Gundumi-Illo formations are the most prolific aquifers for groundwater development in the basin (Kogbe 1989).

In Benin's part of the Iullemeden basin, known as the Kandi basin, coarse sandstones of the Mio-Pliocene constitute the Continental Terminal, which corresponds to the surficial part of the Gwandu Formation (Kogbe 1989; Fig. 2). It is not a very productive aquifer in most parts of the Kandi basin since it is not a geologically and hydraulically continuous layer. However, the Continental Terminal appears to cover other sectors of the Iullemeden basin in

Niger and Nigeria. This aquifer is hydraulically continuous and exploitable in Niger and Nigeria.

The Cretaceous aquifer is represented by the Sende Formation composed of fluvial coarse sandstones, siltstones and pebble conglomerates (Konate et al. 2003). The Paleozoic aquifers (i.e the Cambro-Ordovician and the Terminal Ordovician-Silurian aquifers) are very developed in Benin. The Cambro-Ordovician aquifer, known as the Wèrè Formation, constitutes the main aquifer and is usually semiconfined but becomes unconfined in the western and southern areas of the basin, where it outcrops (Boukari 2007). The Terminal Ordovician-Silurian is the second aquifer and is identified as

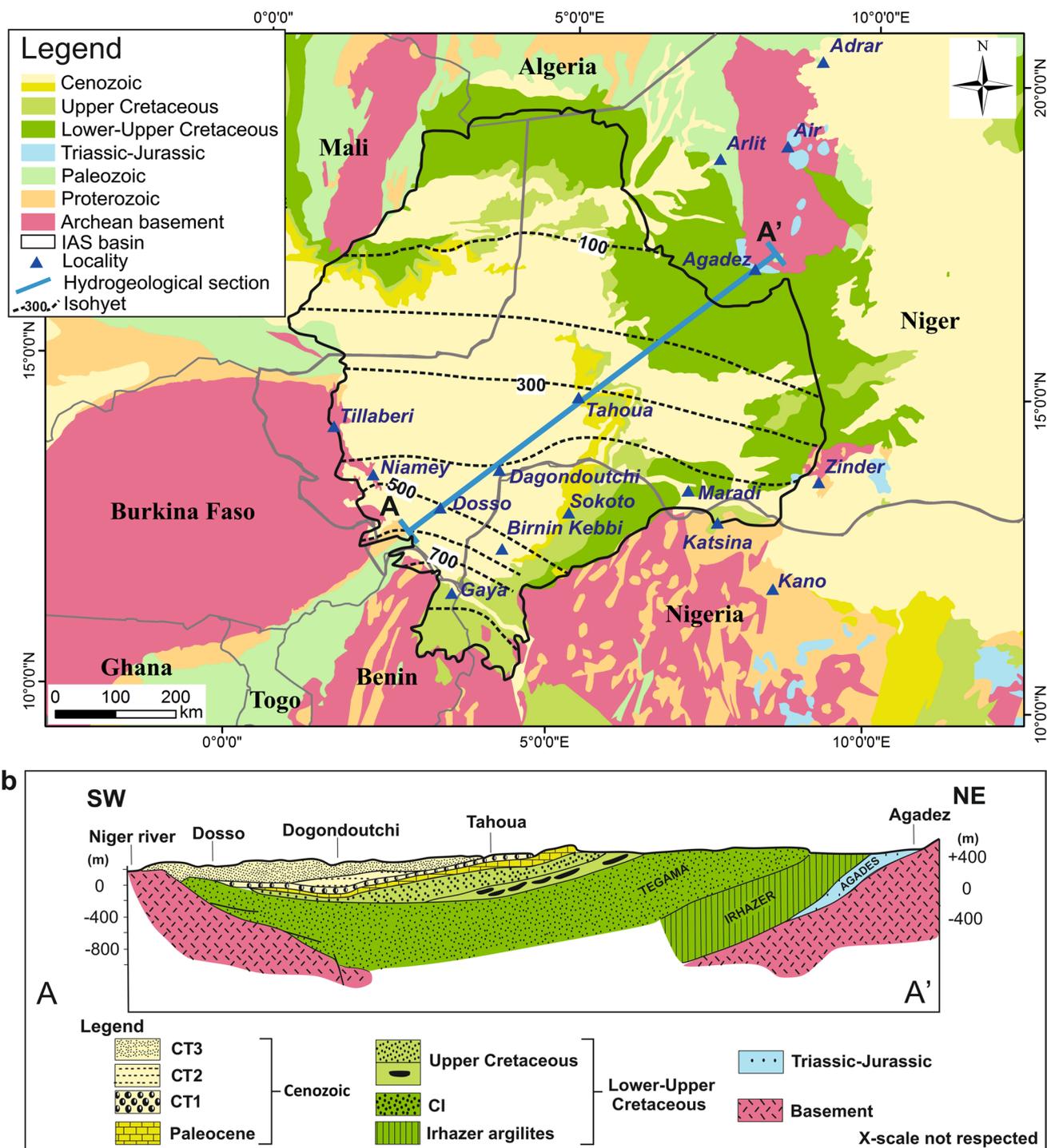


Fig. 3 a Geological map showing the spatial distribution of the annual rainfall and b the hydrogeological cross-section, of the Iullemeden basin (after Greigert 1966)

the Kandi Formation (Fig. 2). In Nigeria, the Paleozoic aquifer is formed by the complex lithologies of the basement, where groundwater occurs in the weathered decomposed portion, the fractured areas, and the portions associated with dykes. The recharge occurs mainly in the southern basin,

and groundwater flows towards the north, towards the Niger River valley, which constitutes the main natural discharge area (Kpegli et al. 2015).

In addition to the shared aquifers mentioned in the preceding, the Quaternary sediments constitute an important aquifer

mainly used for irrigation and farming. It is composed of alluvium deposits and is mainly found along the Niger River.

The IAS, mainly the CT and the CI, is the primary water supply source to the basin's inhabitants and plays a fundamental role in the sustainable management of groundwater resources. Much of the population lives in the CT area due to easy accessibility and good quality water. The annual groundwater abstraction in the basin has gradually increased from 50 million m³ in the 1970s to 180 million m³ in 2004, mainly due to population growth of about 6 million in 1970 to 15 million in 2004 and is expected to double by the year 2025 (OSS 2008). Unfortunately, many wells in the IAS used for domestic and irrigation activities, abstracting from different depths, were drilled without any permission. Even though water resources availability in the region is significant, there is evidence of limited recharge to both the CT and CI, as well as massive exploitation of the aquifer system. This situation has resulted in the marked decline of the piezometric heads, water quality degradation, and exacerbation of the undesired impacts of climate change and variability (OSS 2011a).

To improve understanding of the hydrogeological functioning of the IAS, the present investigation is based on the use of chemical and isotopic properties of the principal sedimentary aquifers, which are simplified as follows: the Cambro-Silurian, the CI, the CT, and the Quaternary.

Sampling and analytical methods

A total of 560 surface water and groundwater samples were collected in the IAS basin (in Niger, Nigeria, and Benin) during several sampling campaigns in the dry and humid seasons from 2013 to 2020 (Fig. 1). Samples collected are distributed as follows—60 samples from surface waters (Niger River, Alibori River, Sôta River, Rima River, Sokoto River and some reservoirs), 35 groundwater samples from the Quaternary aquifer, 210 samples from the CT aquifer, 108 samples from the CI aquifer, and 147 samples from the Cambro-Silurian aquifer. The samples were collected, after purging the wells using a pump or bailer, in polyethylene bottles which were rinsed first with water to be sampled several times and then filled to the top.

In addition, historical isotopic compositions of rainwater at Kandi and Niamey stations, located in Benin and Niger, respectively (Fig. 1), were compiled from the Global Network of Isotopes in Precipitation (GNIP), jointly run by the International Atomic Energy Agency (IAEA) and the World Meteorological Organization (WMO; IAEA/WMO 2006).

Field measurements, such as electrical conductivity (EC), water temperature (T), pH, alkalinity (Alk) and geographic coordinates, were carried out on site. Alkalinity was measured in the field using chemical colorimetric titration by a HACH digital titrator. EC, pH and Alk

were measured in the laboratory to confirm the accuracy and consistency of the field measurements. Hydrochemical analyses (a total of 436 points) of water samples collected during the sampling campaigns were conducted for major elements (Mg²⁺, Ca²⁺, Na⁺, K⁺, Cl⁻, SO₄²⁻, NO₃⁻) using high-performance liquid chromatography (HPLC WATERS), equipped with columns for both cations and anions, with overall detection limit for most ions of 0.04 mg/L. The HCO₃⁻ and CO₃²⁻ concentrations were measured by titration. All chemical analyses present an ionic balance in the range of 0 to ±5%. The total dissolved solids (TDS) refers to the total concentration of major ions dissolved in the water sample.

Stable isotope contents ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) analyses were performed on 539 water samples using the laser absorption spectrometer LGR DLT 100 with autosampler (Penna et al. 2010) and mass spectrometric methods by the CO₂ equilibration method for $\delta^{18}\text{O}$ (Epstein and Mayeda 1953) and the isotopic exchange method with H₂ gas for $\delta^2\text{H}$ (Coplen et al. 1991). Isotopic results are reported in delta units, i.e., per mil deviations (‰) versus the VSMOW reference material (Vienna Standard Mean Oceanic Water). The precision of the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ measurements were ±0.2 and ±1‰, respectively.

Tritium analyses were performed on 375 water samples using liquid scintillation counting (LSC) after electrolytic enrichment (Taylor 1976). ³H concentrations are expressed in tritium units (TU) with a detection limit of 0.2 TU. One TU is defined as one tritium atom in 10¹⁸ protium atoms.

Carbon isotope analyses (¹⁴C and $\delta^{13}\text{C}$) were performed on the dissolved inorganic carbon (DIC) on 51 samples at the Centre for Isotope Research (CIO) at the University of Groningen (Netherlands). The ¹⁴C content was analyzed by accelerator mass spectrometry (AMS) and expressed as percent modern carbon (pmC). Radiocarbon results were normalized using the reference material NIST oxalic acid (SRM 4990C), and a 'zero' point was obtained from CO₂ derived from ¹⁴C-free marble. Analytical uncertainty, reported as one standard deviation (1 SD) of replicate samples, was better than ±0.2 pmC (Dee et al. 2020). Carbon-13 measurements in dissolved inorganic carbon ($\delta^{13}\text{C}$ -DIC) were obtained from an aliquot of headspace CO₂ after acidification of water samples using isotope ratio mass spectrometry (IRMS) methods. $\delta^{13}\text{C}$ values were expressed in the delta notation as parts per thousand (‰) deviations from the international standard (Vienna Pee Dee-Belemnite, VPDB). The uncorrected $\delta^{13}\text{C}$ values were calculated relative to a high-purity research-grade CO₂ pulse and then normalized to the VPDB scale using laboratory reference materials. Analytical precision of the IRMS method for DIC was lower than ±0.2‰ for $\delta^{13}\text{C}$.

Chemical analyses and isotopic measurements ($\delta^{18}\text{O}$, $\delta^2\text{H}$, ³H) were mainly performed at the Radio-Analysis and Environment Laboratory (LRAE) of the National Engineering School of Sfax (Tunisia) and CNESTEN Centre

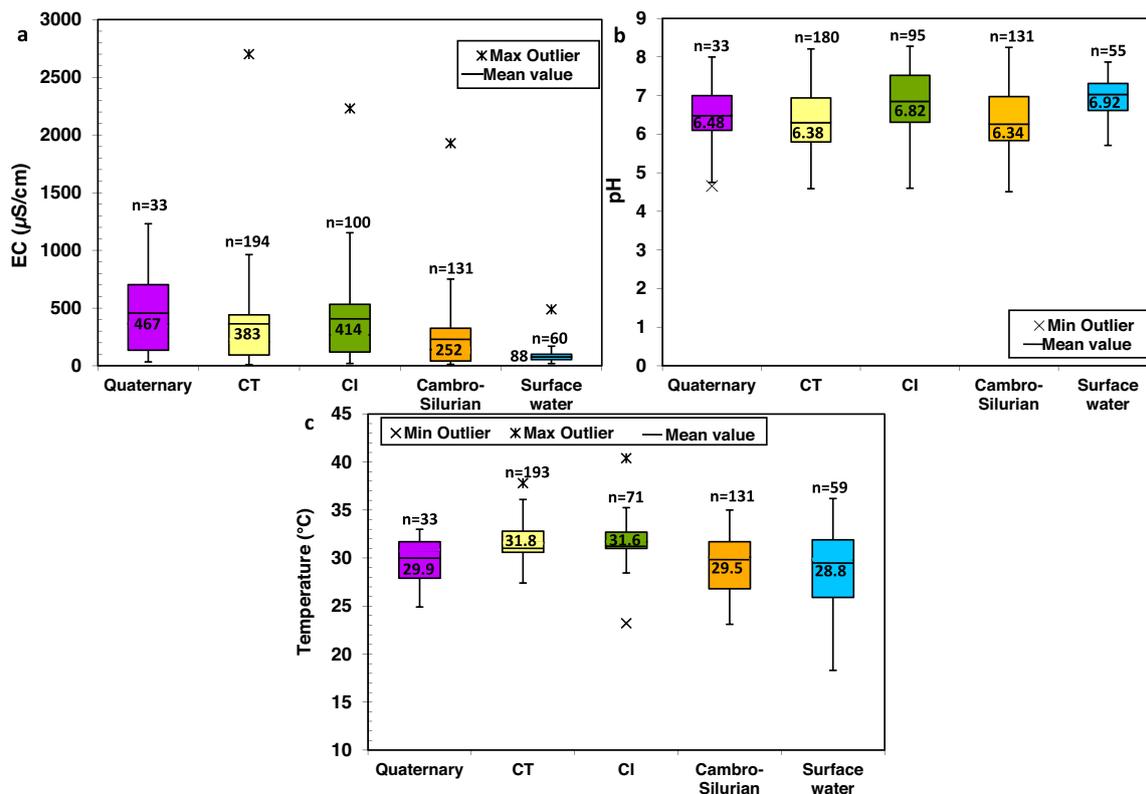


Fig. 4 Boxplot of physico-chemical parameters: **a** electrical conductivity (EC), **b** pH and **c** temperature

(Morocco). Some tritium analyses were performed at the Hydrosys Laboratory in Budapest (Hungary), while ^{14}C and ^{13}C analyses were carried out at the CIO Isotopic Centre, University of Groningen, The Netherlands.

Results

The results regarding the chemical and isotopic investigations are presented in the following sections and in the electronic supplementary material (ESM).

Chemical investigation

Physical-chemical parameters

Electrical conductivity (EC) of groundwater in the studied sites varied from 10 to 2,700 $\mu\text{S}/\text{cm}$ (Fig. 4a). Groundwater samples from the different geological formations do not present statistically significant differences in the EC in terms of both absolute values and variability. Groundwater samples from the CT, CI and Cambro–Silurian aquifers showed the lowest mineralization and slight variability with a mean value of EC of about 383, 414 and 252 $\mu\text{S}/\text{cm}$, respectively. However, the Quaternary aquifer is the most heterogeneous with the highest mean

value of 467 $\mu\text{S}/\text{cm}$. Water samples from rivers and dams were more homogenous, showing limited variability, with values ranging from 19 to 490 $\mu\text{S}/\text{cm}$, and a mean value of 88 $\mu\text{S}/\text{cm}$.

The pH values of water samples collected in the IAS ranged from 4.5 to 8.28, with most of them in the range of 6–7 (Fig. 4b). Both low and high pH waters were characteristics of different aquifers in the basin. The first group included the Quaternary, CT and Cambro–Silurian aquifers, which displayed slightly acidic pH values, while the second group was characterized by neutral and weakly alkaline pH values. The observed values of pH for surface waters varied from 5.71 and 7.87 units.

The temperature of surface waters and groundwaters in the IAS basin ranged from 18.3 to 36.2 $^{\circ}\text{C}$, and 23.1 to 40.4 $^{\circ}\text{C}$, respectively (Fig. 4c). The CI and CT aquifers presented the highest mean water temperatures with low variability. However, surface water was characterized by the lowest values with a mean value of about 28.8 $^{\circ}\text{C}$, which was consistent with the annual atmospheric temperatures in this region ranging between 27 and 30 $^{\circ}\text{C}$ (OSS 2011a).

Total dissolved solids (TDS)

The TDS values found in surface water and groundwater samples ranged from 10 to 1,860 mg/L. The lowest values were measured in surface waters, with the TDS values ranging from

18 to 445 mg/L (Fig. 5g). The spatial distribution maps of TDS in groundwater (Fig. 5) show a very heterogeneous variation without any dependency on the aquifer type. This indicates that the IAS basin is characterized by important heterogeneities and the TDS values are affected by different geochemical processes. The CT groundwater presented the largest variation in terms of mineralization, reflecting the effect of the heterogeneity and the multilayered structure of the aquifer (Fig. 5c). The Quaternary, CI and Cambro-Silurian groundwaters are more homogenous with a slight increase of salinity in some sectors (Fig. 5b, d, e).

Water types

The chemical composition of the analyzed water samples was plotted on Piper trilinear diagrams, presented in Fig. 6. Most groundwater samples were classified as calcium-magnesium-bicarbonate (Ca–Mg–HCO₃) and sodium-chloride (Na–Cl) water types. Except for a few samples from the CT and CI aquifers, which presented an evolution towards sulfate water type, all other samples can be explained as a mixture of bicarbonate and chloride water types. Surface waters were mostly of the bicarbonate water type.

The large spatial variability of the existence of several hydrochemical facies could be related to the physical and chemical weathering reaction of the mineral phases present in the aquifer's matrix.

Stable isotopes investigation

Isotopic composition of local precipitation

For this study, data on the stable isotopic composition of rainwater in the IAS basin were downloaded from the nearest GNIP (Global Network of Isotopes in Precipitation) sites: Kandi and Niamey stations (Fig. 1). The isotopic composition of rainwater at these two stations allowed for estimation of the long-term weighted mean values and assessment of the seasonal isotopic range of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ in rainfall over the IAS. At the Kandi station, in operation from 2014 to 2016, $\delta^{18}\text{O}$ values vary from -6.43 to 1.92‰ VSMOW, with a mean value of -3.47‰ , and from -37.0 to 23.8‰ VSMOW for $\delta^2\text{H}$, with -14.5‰ mean value. The largest isotopic variations were recorded at the Niamey station during the period 1992–2020 (IAEA-GNIP database) where the isotopic composition of rainwater samples ranged from -8.4 to 2.88‰ VSMOW for $\delta^{18}\text{O}$, with a mean long-term value of -4.00‰ , and between -59.0 and 28.8‰ VSMOW for $\delta^2\text{H}$, with a mean long-term value of about -21.2‰ .

The weighted least squares regression (PWLSR) (Hughes and Crawford 2012) of 69 pairs of monthly $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values at Niamey yielded the following local meteoric water line (LMWL) for the west of Niger: $\delta^2\text{H} = 7.38 \delta^{18}\text{O} + 7.38$ ($R = 0.97$). The values at Kandi basin defined a slightly different

LMWL for the southern part of the IAS basin: $\delta^2\text{H} = 7.43 \delta^{18}\text{O} + 11.04$ ($R = 0.98$; Fig. 7). These observations were consistent with the previous isotope investigations conducted in the region and in neighboring sectors in Mali and Niger (Gourcy et al. 2000; Taupin et al. 2002; Huneau et al. 2011).

Isotope composition of groundwater and surface water in the IAS basin

The analyzed groundwater samples from the investigated IAS aquifers showed that $\delta^{18}\text{O}$ ranged from -8.5 to -0.9‰ VSMOW and $\delta^2\text{H}$ from -60.0 to -10.3‰ VSMOW. Generally, the most negative isotopic values were recorded in the CI aquifer (between -8.5 and -2.69‰ for $\delta^{18}\text{O}$), the CT aquifer (between -7.84 and -1.8‰ for $\delta^{18}\text{O}$) and a little less for the Cambro-Silurian aquifer (between -6.53 and -0.90‰ for $\delta^{18}\text{O}$), whilst the more isotopically enriched values were found in the Quaternary aquifer (between -5.39 and -1.77‰ for $\delta^{18}\text{O}$) and surface-water samples (between -4.52 and 4.12‰ for $\delta^{18}\text{O}$).

The position of the various types of samples in the $\delta^{18}\text{O}/\delta^2\text{H}$ diagram (Fig. 8) showed a noticeable variability in their stable isotopic composition, which may be related to the interference of several processes related to different origins of water. The most depleted stable isotope contents are often interpreted as the result of the isotope altitude effect or the presence of fossil groundwater. In fact, it is well established that groundwaters that have been recharged under cooler climatic conditions present more depleted isotopic contents (Huneau et al. 2011; Trabelsi et al. 2020).

River, lake, and reservoir water samples exhibiting enriched isotope contents with respect to present-day long-term mean $\delta^{18}\text{O}$ values in precipitation are the result of isotopic fractionation accompanying the evaporation of open water bodies. Isotopically enriched groundwater highlights the importance of the surface water input to groundwater recharge particularly those to the Quaternary aquifer.

Most points falling close to the Global Meteoric Water Line (GMWL; Craig 1961) and between the depleted and the relatively enriched stable isotope end members, indicate that they originated from the infiltration of modern precipitation and/or the mixing of present-day rainfall and paleowaters (Fig. 8).

Radioactive isotopes investigation

Tritium contents of groundwater and surface water in the IAS basin

Surface waters of the IAS presented tritium contents varying from 0.6 to 5.6 TU. Tritium contents in groundwater samples ranged from undetected to 6.9 TU. Most water samples of the IAS basin showed significant tritium contents. About 60% of the water samples presented tritium contents

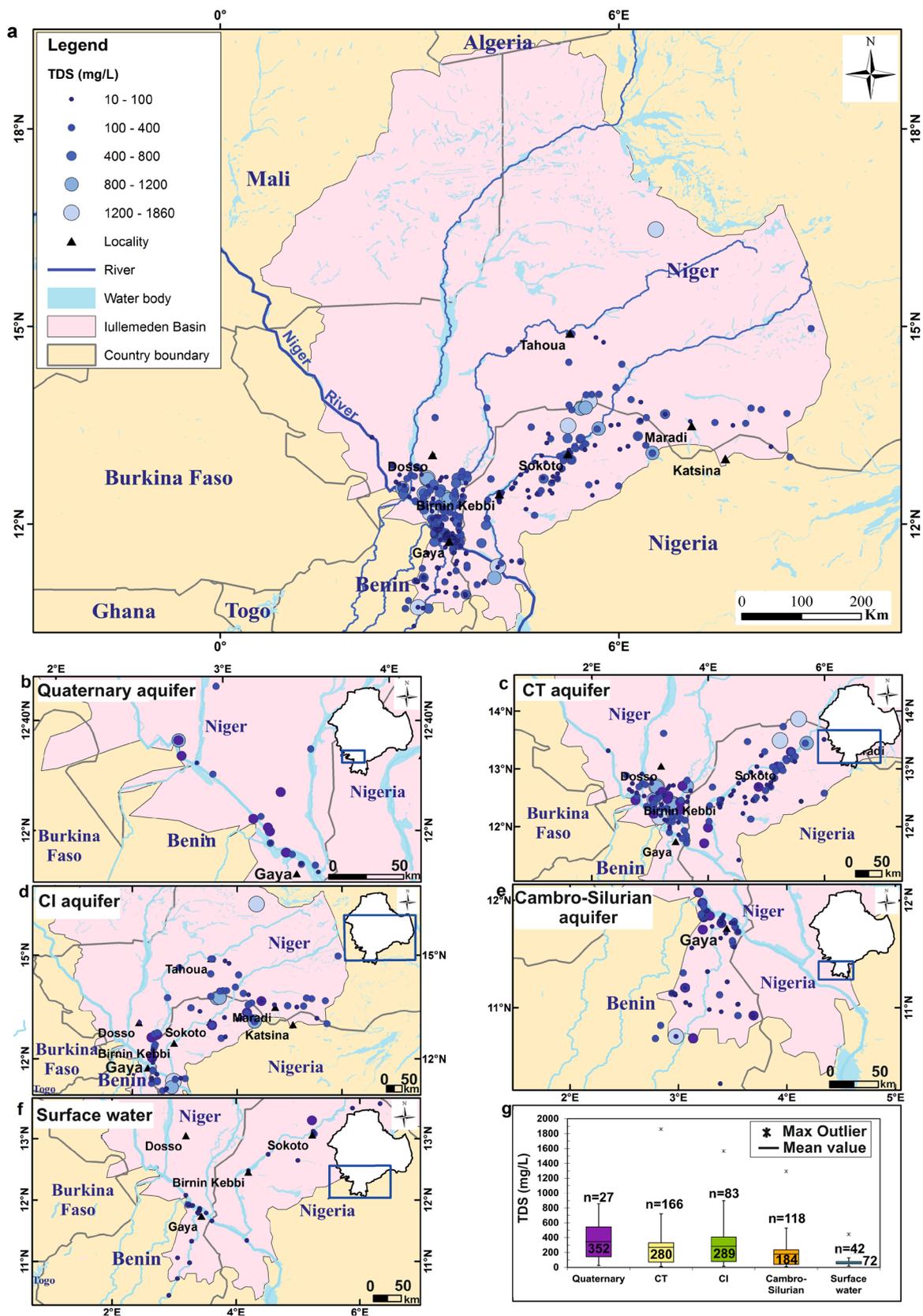


Fig. 5 Spatial distribution maps of total dissolved solids (TDS, mg/L) of the groundwater samples in **a** the IAS, **b** the Quaternary aquifer, **c** the CT aquifer, **d** the CI aquifer, and **e** the Cambro-Silurian aquifer. **f** TDS in the surface-water samples, **g** TDS boxplot

Fig. 6 Piper diagram for surface water and groundwater samples in the IAS basin

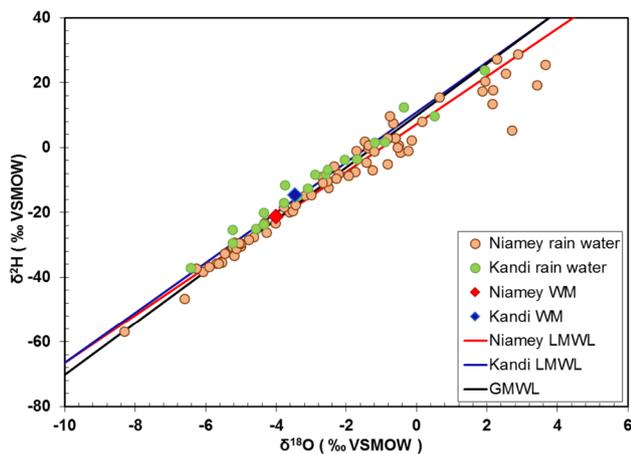
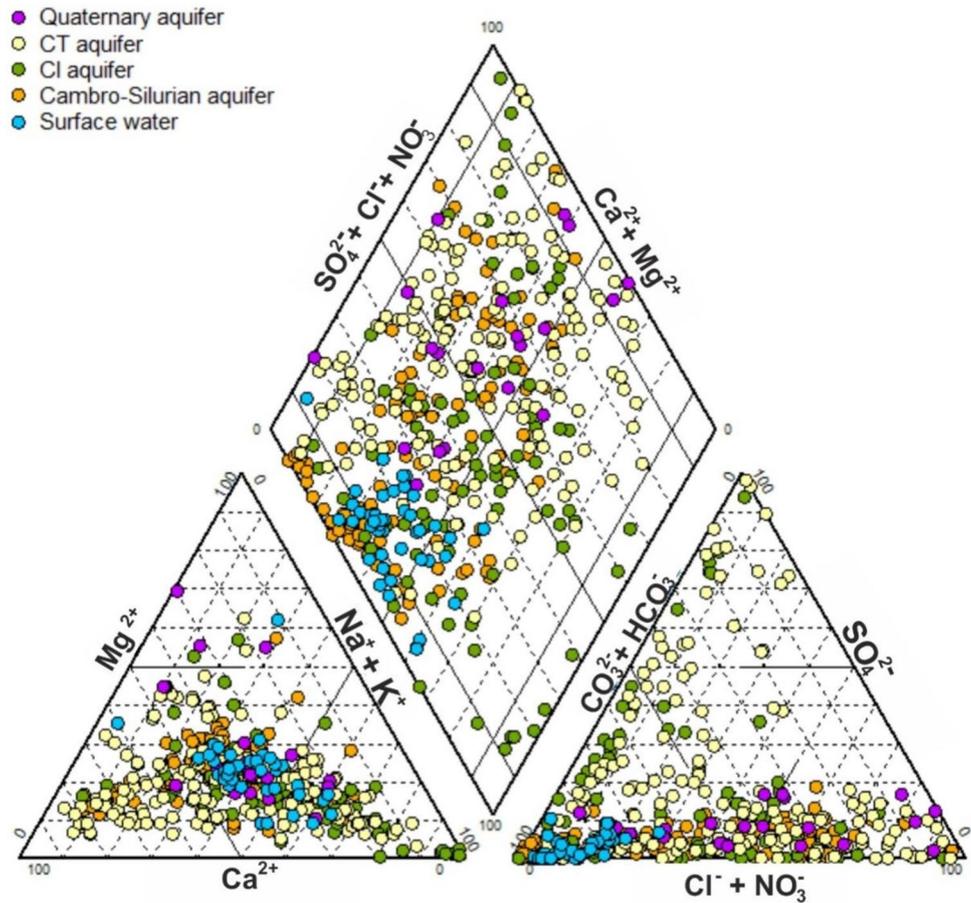


Fig. 7 Oxygen-18 vs deuterium diagram of rainfall at Niamey and Kandi stations, and showing the weighted mean annual composition of rain (WM), the Global Meteoric Water Line (GMWL) (Craig 1961) and the local meteoric water line (LMWL)

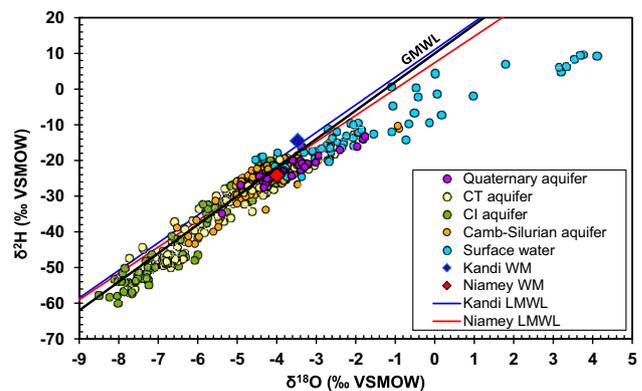


Fig. 8 $\delta^2\text{H}/\delta^{18}\text{O}$ relationship of surface water and groundwater samples in the IAS basin station, showing the weighted mean monthly composition of rain (WM), the local meteoric water line (LMWL) and the Global Meteoric Water Line (GMWL)

higher than 1 TU. In contrast, only 40% of the water samples showed tritium contents lower than 1 TU (Fig. 9a).

The samples of the CI aquifer showed a small range of variation, with the lowest mean tritium value of about 0.7 TU. Compared to the CI aquifer, the CT and the

Cambro-Silurian aquifers presented a higher variation in tritium activities, indicating the existence of both old and recent groundwater in the basin. The Quaternary, the CT and the Cambro-Silurian groundwaters showed the contribution of recent recharge, with an average tritium content higher than 2 TU; however, the CI aquifer showed the

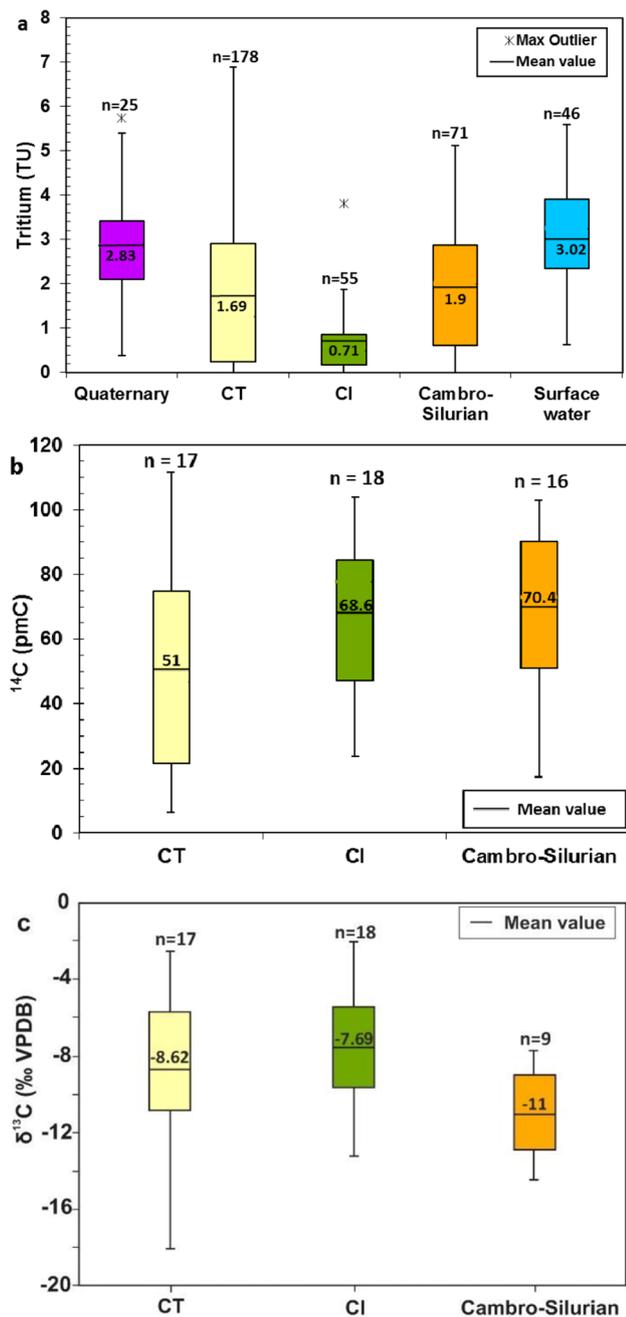


Fig. 9 Boxplots of **a** ^3H contents, **b** ^{14}C activity and **c** $\delta^{13}\text{C}$ contents for groundwater and surface water samples in the IAS basin

widespread presence of groundwater recharged before 1950, with tritium contents lower than 0.5 TU (Fig. 9a).

Carbon isotopes of groundwater in the IAS basin

The ^{14}C and $\delta^{13}\text{C}$ contents were measured on 51 groundwater samples. The ^{14}C activity of groundwater samples in the CT, the CI and the Cambro-Silurian aquifers ranged from

6.6 to 112 pmC (percent modern carbon), with a mean value of 51.0, 68.6 and 70.4 pmC, respectively (Fig. 9b).

The CT aquifer presented the highest variability in ^{14}C activities, reflecting different recharge periods and flow paths, leading to a great heterogeneity of the CT groundwater contained in the three levels (CT1, CT2 and CT3). The analyzed CI groundwater samples were located particularly in the Nigerian sector, where relatively significant carbon-14 activities were found, pointing to the existence of generalized recent recharge of the aquifer in this region (Fig. 9b).

Most groundwater samples collected in the Cambro-Silurian aquifer in Benin presented significant carbon-14 activities, ranging from 70 to 103 pmC, also reflecting the importance of recent groundwater recharge (Fig. 9b). Carbon-13 contents of groundwater samples of the IAS showed wide variations in the isotopic compositions, ranging from -18 to -2‰ VPDB, reflecting different sources of inorganic dissolved carbon in groundwater, such as the dissolution of the aquifer matrix and the soil gas CO_2 . Lower ^{13}C variability was observed in the Cambro-Silurian aquifer with a mean value of -11‰ , a value that reflects a relatively open system to soil CO_2 . The CI aquifer showed higher mean contents of $\delta^{13}\text{C}$ of about -7.7‰ , ranging from -13 to -2‰ vs VPDB (Fig. 9c).

The CT aquifer was also characterized by large variability in $\delta^{13}\text{C}$ contents, ranging from -18 to -2.5‰ vs VPDB. This situation was interpreted as the result of the evolution from an open system to soil CO_2 characterized by $\delta^{13}\text{C}$ ranging between -20 and -13‰ (Le Gal La Salle et al. 2001) to a closed system, marked by the interaction with the rock matrix with $\delta^{13}\text{C}$ close to 0‰ (Huneau et al. 2011).

Discussion

Recharge processes and groundwater residence time (^3H and ^{14}C)

Identifying the dominant recharge mechanism(s) is a key issue for properly assessing the sustainability and groundwater management in arid and semiarid regions. The present isotopic investigation, through the combined use of stable and radioactive isotopes, revealed the existence of different recharge processes in the IAS such as direct recharge, surface-water/groundwater interactions and paleo-recharge.

Direct recharge

Stable isotopes data showed that 65% of the IAS groundwaters are characterized by $\delta^{18}\text{O}$ ranging from -5 to -3‰ and $\delta^2\text{H}$ from -34 to -16‰ , which basically matches the stable isotope content of present-day rainfall in these arid and semiarid regions. Thus, most of the samples of IAS groundwater

plot along the GMWL confirming the existence of significant direct recharge by infiltration of rainwater (Fig. 8). The spatial distribution of oxygen-18 contents in the IAS (Fig. 10) revealed a rather heterogeneous distribution across the different types of IAS aquifer units. Most of the Cambro-Silurian groundwater samples showed an isotopic range similar to the isotopic range observed in present-day rainfall (Fig. 10e). The existence of significant present-day recharge is confirmed by tritium contents clearly higher than 1 TU for most of the analyzed samples (Fig. 9a). However, the recent recharge is very low in some sectors of the CI aquifer, particularly in the part of the IAS in Niger, where the aquifer depth is important. The recharge of the CI aquifer by direct infiltration of rainwater in Nigeria is highlighted by significant tritium values (Fig. 11d), oxygen-18 and deuterium contents ranging from -5 to -3‰ , and from -30 to -20‰ VSMOW, respectively (Fig. 10d), and most of the measured carbon-14 activities are systemically higher than 70 pmC (Fig. 9b).

Since the CT aquifer system consists of three separate levels with different lithologies and depths, the spatial distribution of oxygen-18 and tritium showed unusually high variability. The direct recharge of CT groundwater by infiltration of rainfall is more significant in the shallow (upper) aquifer level (CT3). This was confirmed by noticeable tritium contents (Fig. 11c) and high carbon-14 activities (Fig. 9b); additionally, high tritium contents were also measured in the Quaternary aquifer (Fig. 11b). Present-day recharge could be the result of direct infiltration of rainfall or bank filtration of river water. The interaction between surface water and groundwater will be discussed in the following.

Surface-water/groundwater interaction

The role of river waters and, more generally, surface waters contributing to the recharge of various aquifer systems in the Sahel region, was confirmed by different authors: Ketchemen (1992) in northern Cameroon, Kadjangaba (2007) for the N'Djamena region in Chad, Djoret (2000) and Abderamane (2012) for the Chari-Baguirmi sector in Chad, Raizi (2008) and Genthon et al. (2015) for the Diffa region in Niger.

However, surface-water contribution to groundwater recharge is progressively affected by the increasing aridity in the IAS basin, marked by a substantial decrease in rainfall and runoff and an increase in temperature registered during the last few decades (OSS 2011a). Consequently, the evaporation rate from open water bodies has increased, leading to a reduction in the net recharge to several groundwater systems.

The evaporation process was clearly identified in most samples collected from rivers and reservoirs, which are isotopically enriched with respect to rainfall, and plot below the GMWL along an evaporation trend (Fig. 8). Furthermore, groundwater in the Quaternary aquifers revealed the isotopic signature of evaporation, as shown by significantly enriched values in $\delta^{18}\text{O}$

and $\delta^2\text{H}$. This evaporation process could occur before the infiltration, during direct infiltration through the soil and unsaturated/weathered zone, and from direct open wells that are shallow and not covered. This process is also highlighted in some areas of the CT multilayer aquifer, particularly in CT3 in Niger.

The calculated deuterium-excess (d-excess) using Eq. (1) (Dansgaard 1964), also presented large variability, helping to identify different recharge modes (Fig. 12). Enriched stable isotope contents, coupled with small d-excess values, particularly for the CT3 and Quaternary groundwater samples, confirmed that these unconfined aquifers were affected by evaporation processes. The spatial distribution maps showed that the most enriched samples are located close to the Niger River, particularly in the southern part of the IAS basin (Fig. 10b,c).

$$d - \text{excess} = \delta^2\text{H} - 8 \times \delta^{18}\text{O} \quad (\text{Dansgaard 1964}) \quad (1)$$

The contribution of surface water to the recharge of the Quaternary aquifer was quantified in some wells close to the rivers, by using a simple isotopic mass balance equation, based on $\delta^{18}\text{O}$ contents, according to the following equations (Stichler et al. 1986; Mook 2006):

$$\delta^2\text{O}_{\text{Quat}} = X \delta^{18}\text{O}_{\text{riv}} + (1 - X) \delta^{18}\text{O}_{\text{rain}} \quad (2)$$

$$X = (\delta^2\text{O}_{\text{Quat}} - \delta^{18}\text{O}_{\text{rain}}) / (\delta^{18}\text{O}_{\text{riv}} - \delta^{18}\text{O}_{\text{rain}}) \quad (3)$$

where X represents the fractional contribution of river/surface water, $\delta^{18}\text{O}_{\text{riv}}$ represents the mean $\delta^{18}\text{O}$ of river samples, equal to -1.5‰ VSMOW, and $\delta^{18}\text{O}_{\text{rain}}$ represents the weighted mean $\delta^{18}\text{O}$ contents at Niamey station, equal to -4‰ VSMOW.

The calculated contributions are highly variable, ranging from ~ 2 to $\sim 89\%$. The higher fractions were found in the southern part of the basin, in the vicinity of the bed of the Niger River. The infiltration of river water, particularly during the flood period, leads to a decrease in the salinity of groundwater and to higher tritium activities in groundwater (Table 1; Fig. 13).

Paleo-recharge conditions

Stable isotopes are conservative tracers, reflecting the average climatic conditions during the recharge process (Gat 1971). Subsequently, these tracers deliver information on the recharge processes of groundwater and aid in answering questions concerning the residence time and origin of groundwater. Indeed, the residence time of groundwater in actively circulating systems may range from tens to many thousands of years and changing climate conditions could be recorded in palaeowaters.

Such climate changes are manifested by a shift in the stable isotope content of precipitation; the use of the isotopic method

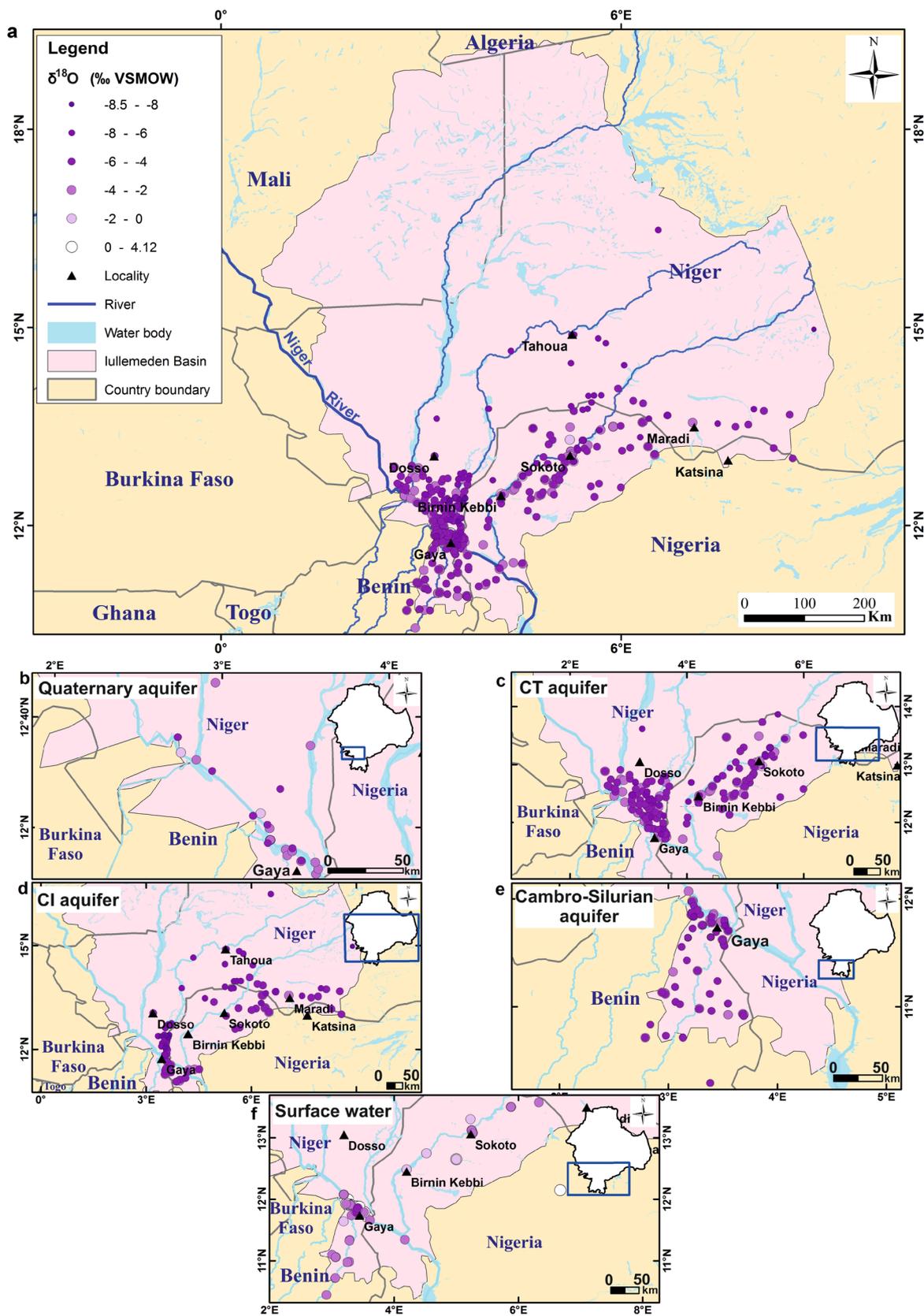


Fig. 10 Spatial distribution maps of $\delta^{18}\text{O}$ values (‰ VSMOW) of the groundwater samples in **a** the IAS, **b** the Quaternary aquifer, **c** the CT aquifer, **d** the CI aquifer, and **e** the Cambro-Silurian aquifer. **f** $\delta^{18}\text{O}$ values of surface water samples

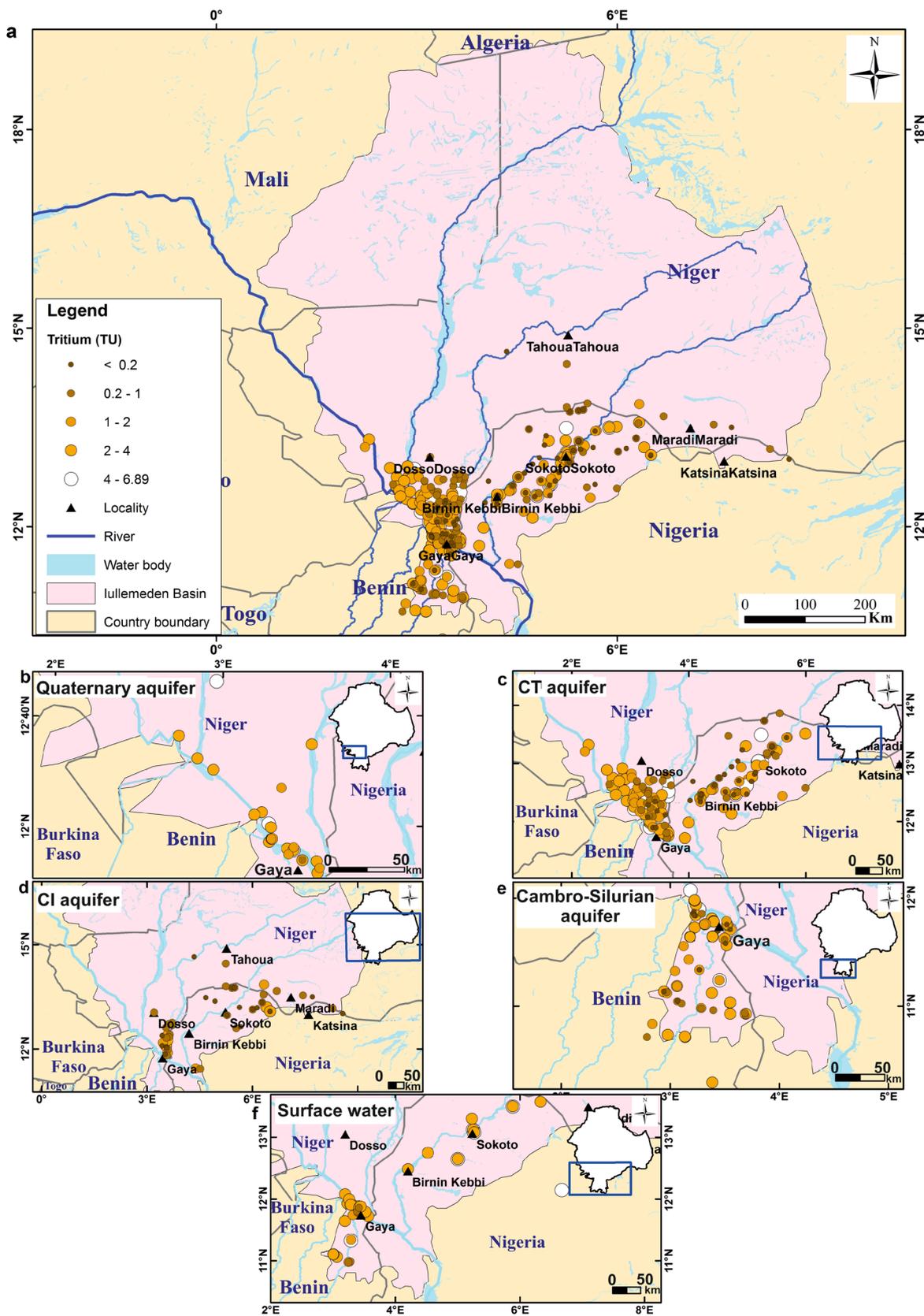


Fig. 11 Spatial distribution maps of ^3H contents (TU) of the groundwater samples in **a** the IAS, **b** the Quaternary aquifer, **c** the CT aquifer, **d** the CI aquifer, and **e** the Cambro-Silurian aquifer. **f** ^3H contents of the surface water samples

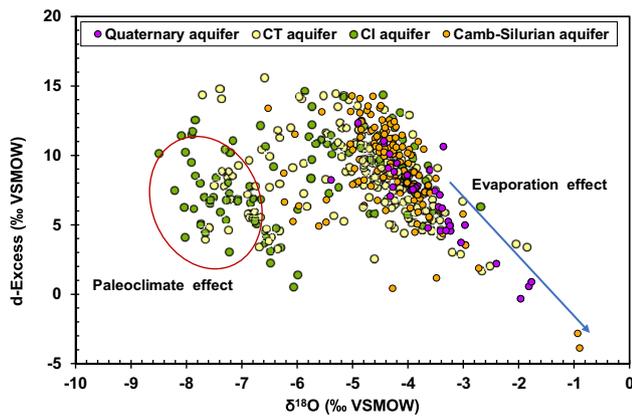


Fig. 12 Deuterium excess vs $\delta^{18}\text{O}$ (both in permil units) relationship of groundwater samples in the IAS basin

is then an important tool to distinguish early Holocene, late Holocene and Pleistocene waters that could circulate in the deeper confined parts of the aquifer (Darling et al. 1997).

The use of stable and radioactive tracers confirmed that a significant portion of groundwaters in the IAS basin is very old and recharged during past wet periods from the Holocene and the late Pleistocene (Beyerle et al. 2003; Andrews et al. 1994). Indeed, the $\delta^2\text{H}/\delta^{18}\text{O}$ relationship highlighted the

presence of about 18% of the total analyzed samples, presenting isotopically depleted waters ($\delta^{18}\text{O}$ ranging from -8.5 to -6.5 ‰, and $\delta^2\text{H}$ from -60 ‰ to -36 ‰ VSMOW (Fig. 8). This fraction of tritium-free groundwater was identified in some regions of CI and CT aquifers (Fig. 13).

To provide additional evidence of the presence of fossil groundwater in the IAS, radiocarbon activities in groundwater samples were used (Fig. 14). In the deeper sectors of the IAS, the CI aquifer, and in the Niger part, low radiocarbon activities were found, ranging from 24 to 60 pmC.

The heterogeneous spatial distribution observed in the stable isotope contents of the different aquifers is also observed in the distribution of d-excess. Considering the depleted stable isotopes composition of groundwater samples, particularly for the CI aquifer, a paleoclimate recharge with atmospheric circulation different from current conditions is suggested, as reflected by the d-excess values (Fig. 12).

Flow patterns (interaction between aquifers, recharge and discharge)

The use of environmental isotopes combined with other geochemical tracers helped derive new insights into the recharge processes of the IAS groundwater and helped to address some questions on groundwater flow paths.

Table 1 Calculated contribution of river water to the recharge of the Quaternary aquifer in the IAS basin

Sample name	Country	Latitude (DD)	Longitude (DD)	$\delta^{18}\text{O}$ (‰ VSMOW)	$\delta^2\text{H}$ (‰ VSMOW)	^3H (TU)	d-excess (‰ VSMOW)	TDS (mg/L)	Contribution of river water (%)
Q1	Niger	12.0853	3.2306	-1.77	-13.2	3.65	0.9	142	89.4
Q2	Benin	11.9238	3.2870	-1.82	-14.0	NA	0.6	98	87.3
Q3	Niger	12.4528	2.7539	-1.96	-16.1	NA	-0.33	434	81.4
Q4	Benin	11.9238	3.2870	-2.40	-17	1.60	2.2	241	63.9
Q5	Niger	11.9942	3.2850	-2.97	-18.8	3.30	4.97	703	41.2
Q6	Benin	11.7182	3.5624	-3.04	-20.6	2.48	3.72	NA	38.4
Q7	Benin	11.8653	3.3820	-3.23	-21.3	NA	4.5	631	30.8
Q8	Niger	12.4092	2.8461	-3.24	-21.0	3.42	4.9	83	30.3
Q9	Niger	11.7489	3.5756	-3.27	-20.9	1.88	5.2	96	29.3
Q10	Benin	11.9247	3.2913	-3.31	-21.9	2.86	4.58	NA	27.6
Q11	Niger	12.4942	3.5294	-3.36	-16.3	3.61	10.6	192	25.6
Q12	Niger	12.8736	2.9614	-3.38	-20.9	5.73	6.19	226	24.7
Q13	Benin	11.7952	3.4732	-3.41	-22.7	2.94	4.6	215	23.6
Q14	Benin	11.8653	3.3820	-3.42	-20.2	3.13	7.1	587	23.1
Q15	Benin	11.9247	3.2888	-3.45	-21.3	NA	6.3	123	22.0
Q16	Benin	11.9247	3.2888	-3.51	-20.7	3.03	7.4	146	19.6
Q17	Benin	11.8556	3.4191	-3.61	-20.0	4.36	8.9	350	15.4
Q18	Niger	11.7897	3.5575	-3.64	-24.3	2.88	4.8	151	14.5
Q19	Benin	11.7952	3.4732	-3.83	-22.8	NA	7.8	257	6.9
Q20	Benin	11.8556	3.4192	-3.86	-23.2	2.22	7.68	NA	5.6
Q21	Benin	11.8313	3.3907	-3.91	-23.7	1.92	7.58	NA	3.6
Q22	Benin	11.8556	3.4191	-3.94	-24.0	NA	7.5	388	2.3

NA not analyzed

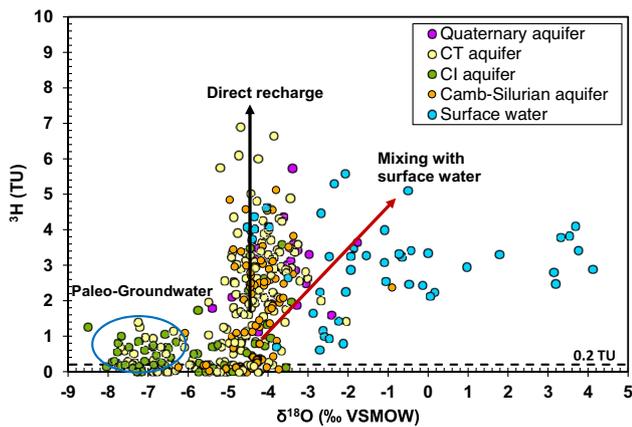
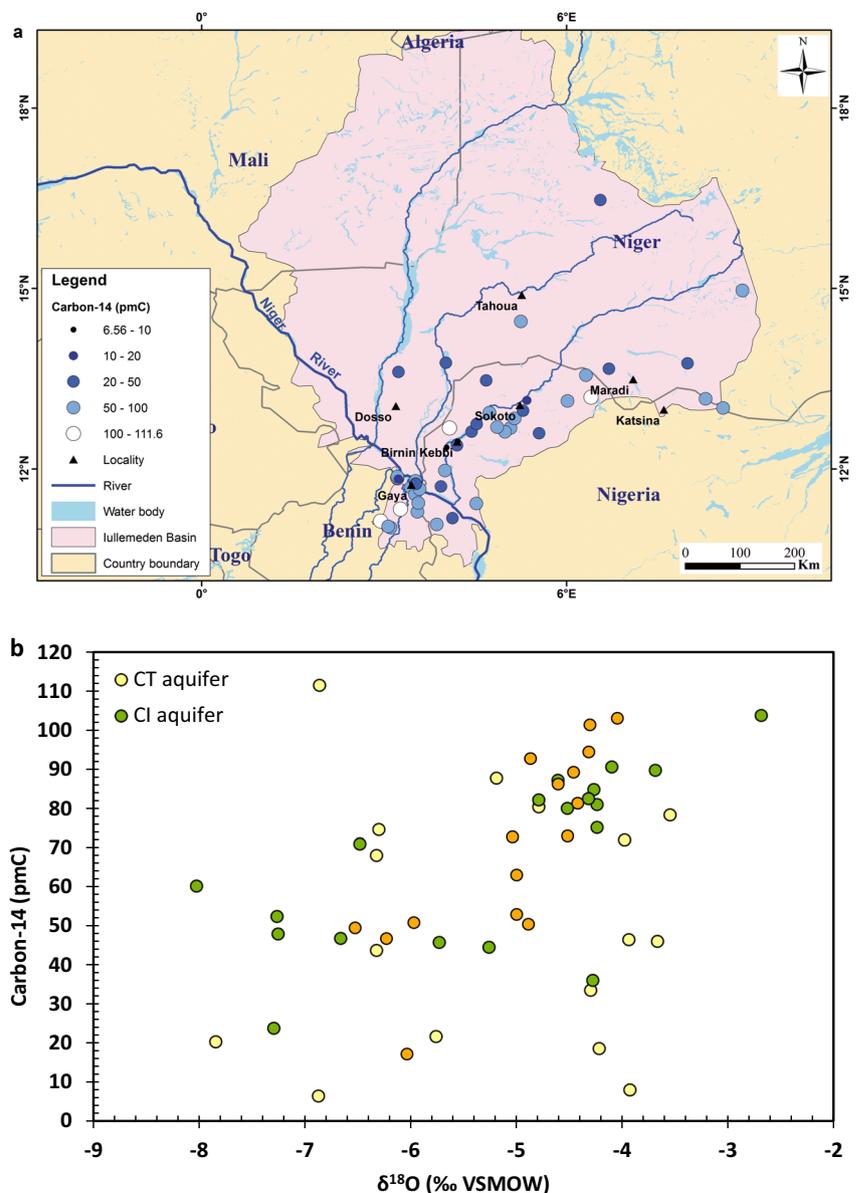


Fig. 13 $^3\text{H}/\delta^{18}\text{O}$ relationship of surface water and groundwater samples in the IAS basin

Fig. 14 a Spatial distribution map of ^{14}C activities (pmC) of all groundwater samples, **b** $^{14}\text{C}/\delta^{18}\text{O}$ relationship of the CT, CI and the Cambro-Silurian aquifers in the IAS basin



The CI aquifer, one of the main aquifers in the Iullemmeden basin, presented quite heterogeneous isotopes and hydrochemical contents, reflecting the highly variable hydrogeological proprieties and complex recharge and geochemical processes of the system. Unfortunately, due to security issues, the northern part of the basin (Malian part and the north of Niger) was not covered under this study. Only one point was sampled in this confined region of the CI in Niger, where the groundwater is tapped at depths exceeding 500 m. The high mineralization of this borehole, coupled with depleted stable isotope composition ($\delta^{18}\text{O}/\delta^2\text{H}$) and low ^{14}C activity, suggested that the groundwater in this sector is composed of very old water that has chemically evolved through enhanced water–rock interaction reactions.

In the Nigeria part, particularly in the Sokoto basin, the CI is the oldest Cretaceous formation overlaying the

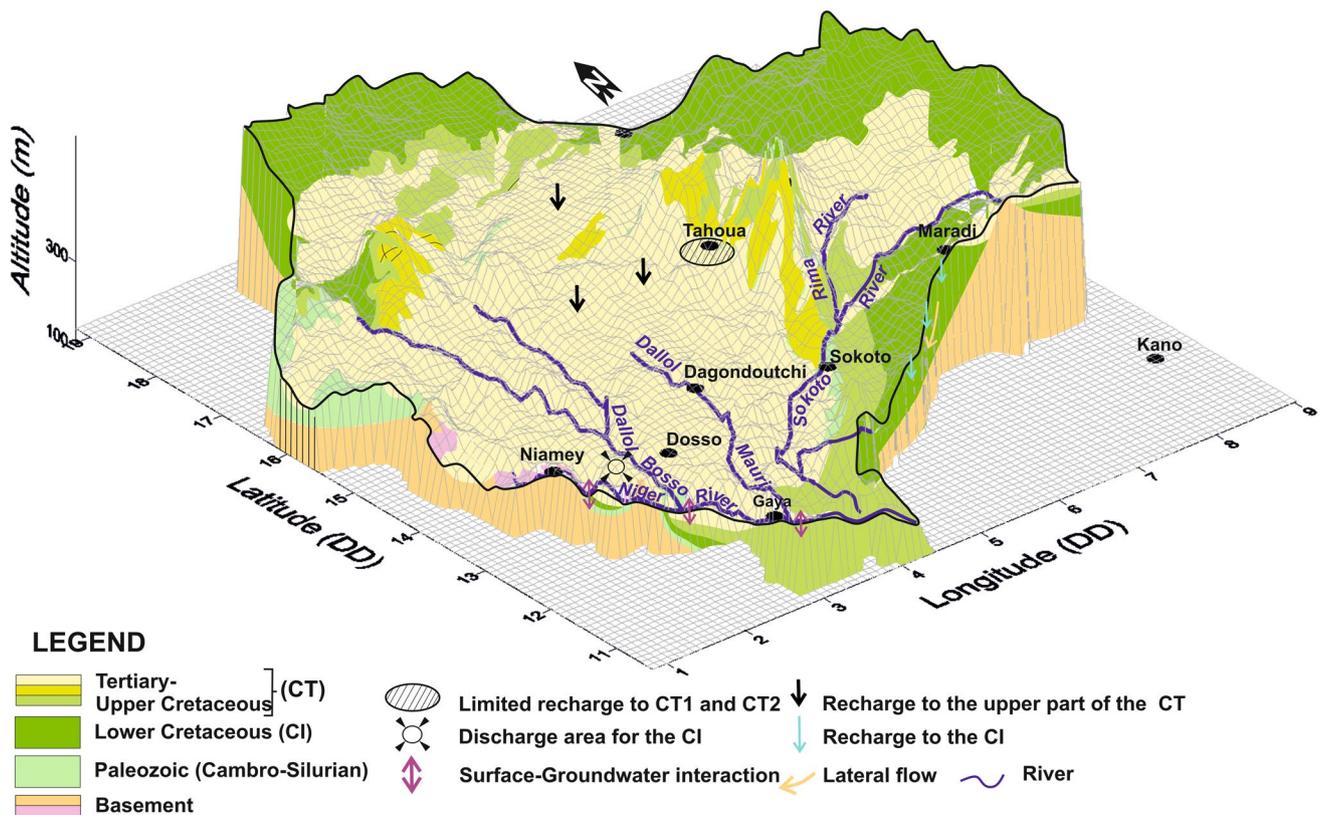


Fig. 15 Block diagram summarizing main groundwater flow patterns in the IAS basin

Precambrian basement complex rocks, according to the literature. Ideally, the waters from this aquifer are expected to be much older compared to any other waters of the multiaquifer systems of the basin. However, the radiocarbon data from the CI samples proved the presence of recent groundwater probably as a result of the locations of the sampled well in the eastern part of the basin characterized by the relatively shallow depth (80–100 m). As summarized in Fig. 15, the CI aquifer outcrops at the south-eastern border of the Iullemeden basin (in Nigeria) represent a preferential recharge zone; hence, the CI aquifer within the Nigerian part of the Iullemeden basin shows a high pmC ranging between 70 and 103‰ (Fig. 14a), which is indicative of groundwater being younger in the CI aquifer compared to samples from the CT, characterized by lower ^{14}C activity (36–45 pmC). The younger samples suggest possible meteoric input (renewal) due to their proximity to the transition zone, the basement complex outcrops, which could be their source of recharge. The relatively older water with lower ^{14}C activity in the CI indicates that limited recharge occurs far away and that the groundwater flows towards the centre of the basin. The stable isotopes of the CI range from -15.2 to -33.6 ‰ for $\delta^2\text{H}$ and from -2.7 and -5.7 ‰ for $\delta^{18}\text{O}$ showing significant homogeneity compared to the stable isotopes contents in the whole basin, which varies from -51.4 to 9.4 ‰ for $\delta^2\text{H}$

and from -7.8 to -0.4 ‰ for $\delta^{18}\text{O}$ (Fig. 10). The variation in the hydrochemical facies of CT and CI aquifers further suggests possible groundwater evolution which could have been influenced by rock–water interaction along its flow path, as well as its residence time.

In the Tahoua region (Niger), the isotopic signature of the CI groundwater was quite variable, with $\delta^{18}\text{O}$ contents ranging from -7.9 to -5.7 ‰. The few samples measured for tritium were generally below the detection limit, except for a sample with ^3H value of 1.7 TU. Also, the carbon-14 activities of two samples in this sector were 48 and 71 pmC (Figs. 10, 11 and 14a), reflecting a relatively limited recharge to the CI aquifer in this region.

In the southern part of the basin, in the Dosso region, the CI aquifer is shallow, and the groundwater level is close to the topographic surface, with some artesian boreholes. About 80% of the collected samples in this area showed depleted stable isotope contents ($\delta^{18}\text{O} < -7$ ‰ and $\delta^2\text{H} < -49$ ‰) with a mean tritium activity close to the detection limit. The chemical composition of these points was dominated by Na and Cl, indicating an advanced stage of water–rock interaction processes. The depleted stable isotope signature of these evolved groundwaters indicated a significant recharge occurring during the humid phases of the Holocene (Beyerle et al. 2003). This finding is consistent

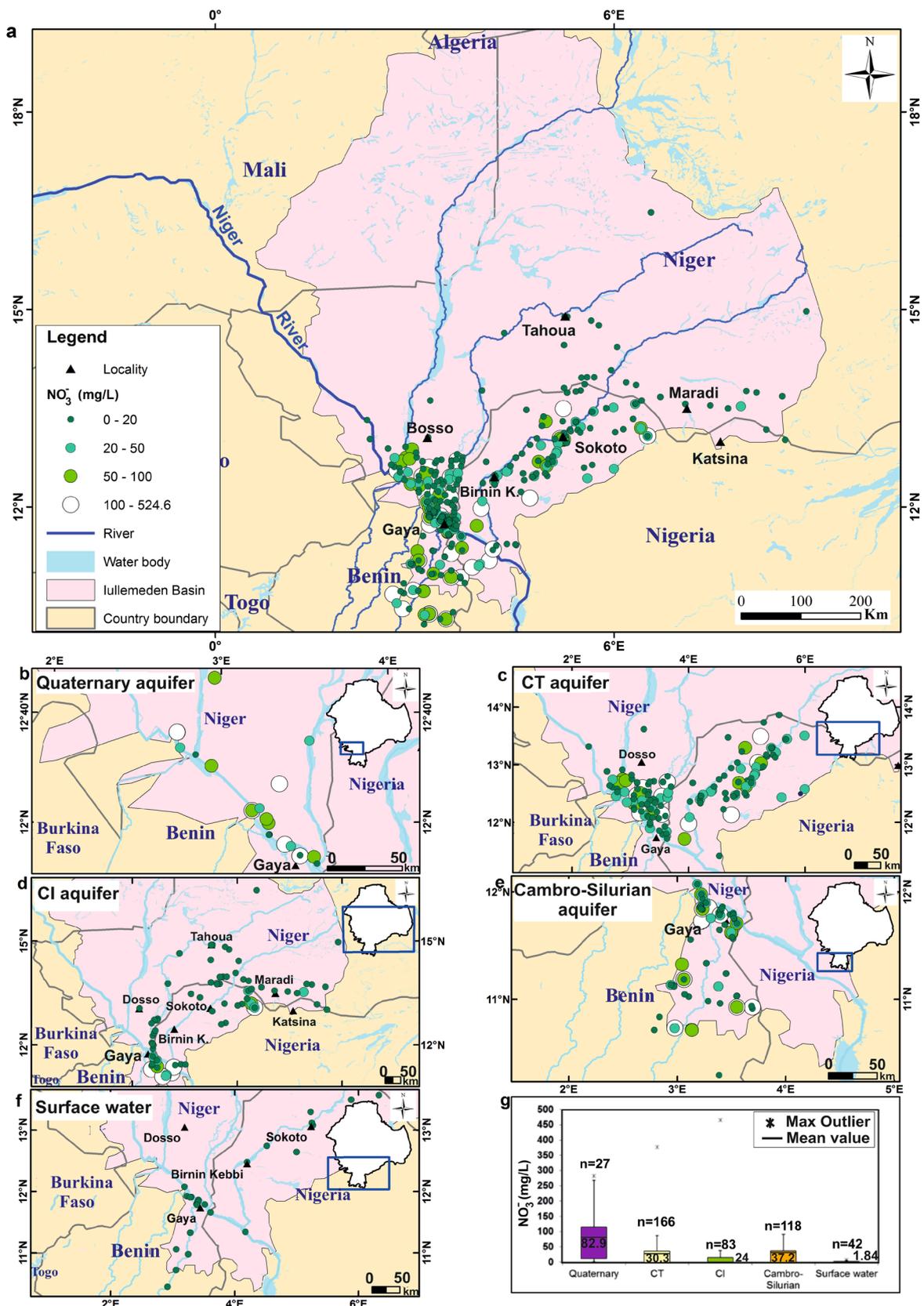


Fig. 16 Spatial distribution maps of NO₃⁻ concentrations (mg/L) of the groundwater samples in **a** the IAS, **b** Quaternary aquifer, **c** the CT aquifer, **d** the CI aquifer, and **e** the Cambro-Silurian aquifer. **f** NO₃⁻ concentrations in surface water samples; **g** NO₃⁻ boxplot

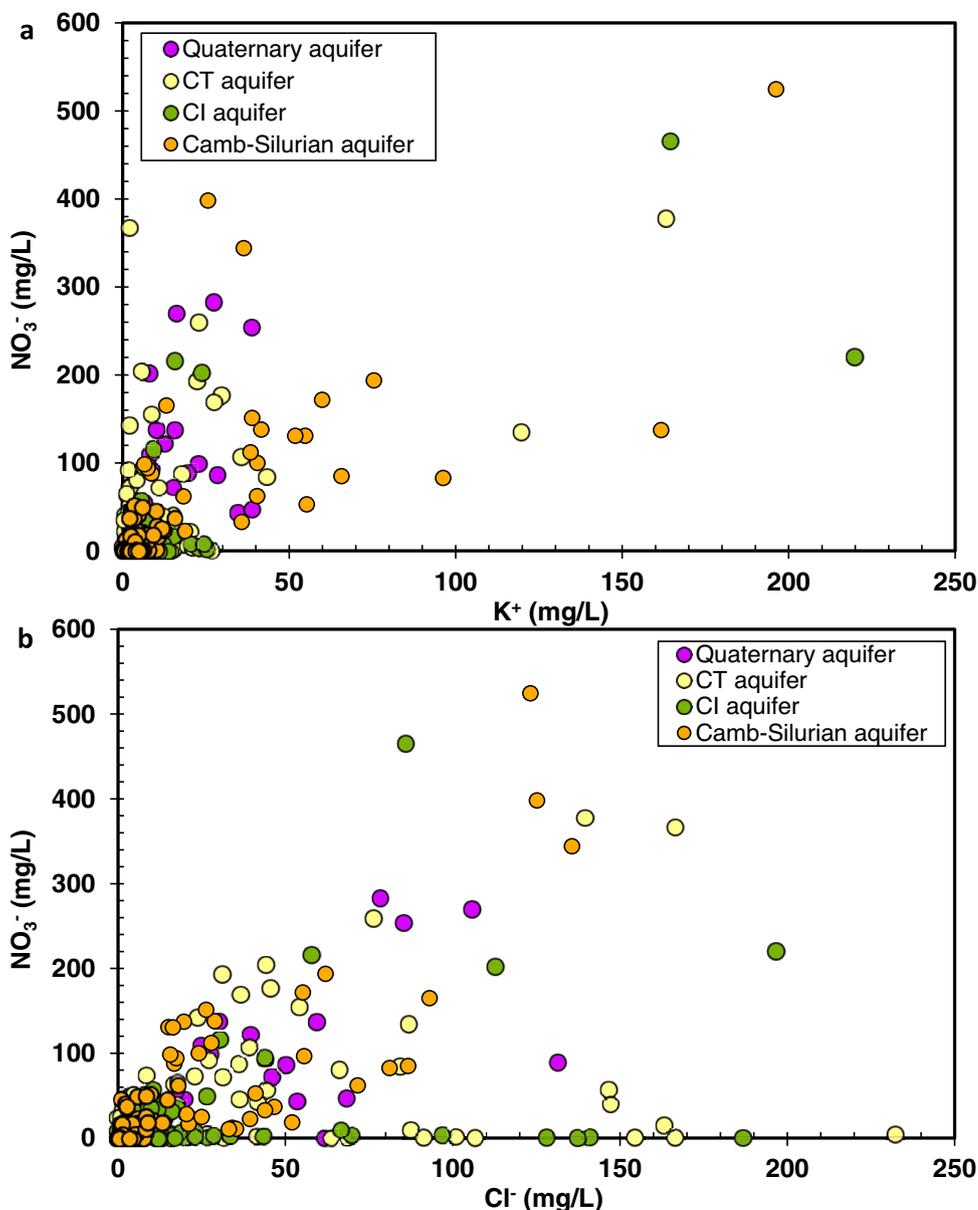
with the results and explanations proposed for northern Mali by Fontes et al. (1993), for Niger by Andrews et al. (1994) and Beyerle et al. (2003), and for a nearby basin in Burkina Faso by Huneau et al. (2011).

Nevertheless, only a few samples tapping the CI in this region and close to the Niger River presented enriched stable isotope contents, with significant tritium contents. This observation could indicate a local interaction between the CI groundwater and the surface waters (Fig. 15). Thus, the hydrodynamic functioning of the CI aquifer seems to be more complicated than the proposed groundwater flow model by previous investigations—i.e., a single recharge area and a single discharge point into the Niger River (OSS 2011a).

Compared to the CI aquifer, the CT is very heterogeneous since it consists of three separate aquifer levels with different

lithologies and depths. This is reflected in the high variability of stable and radioactive isotope contents. The CT3 in Niger and Gwandu Formation in Nigeria is characterized by the presence of recent recharge highlighted by significant tritium content and enriched $\delta^2\text{H}$ and $\delta^{18}\text{O}$ isotope compositions. This present-day recharge could be the result of the river-bank and rain infiltration; however, the deeper level of the CT aquifer system (CT1 in Niger and Rima Formation in Nigeria) presents depleted stable isotope composition with low tritium contents and ^{14}C activities. This isotope signature is concordant with the finding described by Le Gal La Salle et al. (1994) in the southwest of Niger, close to the Niamey region, where ^{14}C activities are close to the detection limit; thus, the recharge of the CT1 groundwater probably occurred during the late Pleistocene (above 25 kyr BP; Beyerle et al. 2003).

Fig. 17 a NO_3^-/K^+ and b $\text{NO}_3^-/\text{Cl}^-$ relationships of groundwater samples in the IAS basin



The CT2 (Sokoto Formation in Nigeria) groundwater shows depleted stable isotope values generally similar to those of CT1 (Fig. 14a,b). This is consistent with previous dating performed in the Niger part, where the CT2 groundwater age is between 6 and 15 kyr BP (Beyerle et al. 2003), in the confined region of this aquifer. For this reason, it is very hard to distinguish between CT2 and CT1 aquifers in some parts of the IAS basin where a vertical hydraulic communication could exist, as some wells contain mixed groundwaters from both aquifers. However, in the unconfined part of the CT2, the stable isotope composition is rather more enriched than that of the confined regions indicating a relatively recent recharge, which was evaluated to be around 6 kyr BP (Beyerle et al. 2003). Previous investigations (Beyerle et al. 2003) reveal that a mixing of groundwater between the CT2 and the CT3 occurs in some wells in Niger.

Vulnerability to pollution: anthropogenic fingerprint on groundwater

A relatively important phenomenon affecting the IAS groundwater is the strong anthropogenic fingerprint encountered in the basin. In fact, moderate to high nitrate concentrations were observed in some wells and boreholes across the IAS basin, which is particularly accurate for the most densely populated zones.

The spatial distribution map of nitrate concentrations of the different aquifers in the whole IAS basin showed that more than 80% of samples contain nitrate contents lower than 50 mg/L (the maximum recommended value for drinking water). However, ~52% of the samples collected in the Quaternary aquifer present significant local pollution by nitrate (Fig. 16), and are therefore unsuitable for drinking water purposes, with concentrations exceeding the statutory limits set by the World Health Organization (50 mg/L NO_3^- ; WHO 2022). High to moderate nitrate pollution was also found in the groundwaters of the Cambro-Silurian aquifers in about 20% of the samples. The CI and CT groundwaters are less affected by nitrate contamination and present nitrate contents below drinking water standards.

Several geochemical diagrams showing the relationships between NO_3^- , K^+ , and Cl^- (Fig. 17) were used to infer the sources of nitrate in groundwater. In the scatter plot of Fig. 17, high NO_3^- values coincide with elevated Cl^- concentrations and K^+ contents at a few sampling sites of the Quaternary, the Cambro-Silurian and the shallow aquifer of the CT. This observation suggests that such groundwaters have the same origin of NO_3^- , Cl^- and K^+ , i.e., the excessive use of fertilizers in the form of urea (N), NH_4Cl , NPK (N, P, K rich fertilizer) and farmyard manure on the land surface. In addition, due to agricultural activity, various other anthropogenic sources, such as industrial and domestic, might lead to high nitrate contents

in the groundwater. The use of isotope ^{15}N may shed light on the contribution of these sources of nitrate and the magnitude of the biogeochemical processes occurring in the IAS.

Concluding remarks

This study, based on the use of environmental isotopes ($\delta^{18}\text{O}$, $\delta^2\text{H}$, ^3H , $\delta^{13}\text{C}$, and ^{14}C) combined with other geochemical tracers and hydrological techniques, has provided new insights into the recharge processes throughout the Iullemmeden Aquifer System (IAS), helping to address some questions on groundwater flow paths. Previous investigations on the basin have dealt with local approaches on a fragmentary basis. For the first time, samples from three different neighbouring countries have been considered together.

The study revealed that there is a great groundwater potential with an overall good quality in the IAS, with some very local nitrate contamination issues, particularly in Benin and Nigeria. The major processes affecting groundwater quality and hydrodynamics are highlighted.

The use of environmental isotopes (stable and radioactive) has provided new major information on recharge and palaeorecharge conditions as well as groundwater flow in the studied part of the IAS. The palaeowater signature was detected in several deep groundwater samples, in particular in CI and CT aquifers. These aquifers were probably recharged under different climatic conditions during the late Pleistocene and early Holocene. It appears that most of the shallower units are recharged currently, as proved by their stable isotope composition, which is similar to the isotopic signature of rainwater, and also by their significant tritium contents and carbon 14 activities. Moreover, the stable isotope signatures highlighted the evaporation effect that has strongly marked the geochemical signature of water bodies and has been used to demonstrate the interaction between groundwater and surface water. Indeed, surface-water infiltration toward the CI aquifer was clearly identified locally around the Niger River.

Further, the applied isotopic approach revealed a preferential groundwater recharge area for the CI aquifer near the southeastern border of the IAS, which should be preserved from uncontrolled occupation and pollution. The main findings regarding groundwater flow in the IAS are summarised on a preliminary 3D block-diagram which can be improved in the future as long as further evidence regarding the IAS's flow pattern becomes available.

The scientific results obtained have significant implications for groundwater management in the Sahel region, especially in the IAS, and should be integrated into the Strategic Management Program of the region. Indeed, the water resources in the studied aquifers represent significant water reserves of good quality which need to be properly managed

and protected as high-quality resources and as part of integrated plans for the basin's future supplies. In this context, intensive care to be taken with respect to the risk of salinization and contamination by nitrate in some areas.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s10040-023-02744-0>.

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Declarations

Conflicts of interest No potential conflict of interest was reported by the authors

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