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Multiple environmental tracers for a better understanding of water flux in a wetland area (La Bassée, France)

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Abstract

Understanding water exchange between groundwater and streams, or groundwater/surface-water relationships, is of primary importance for solving conflicts related to water use and for restoring water ecosystems. To this end, we tested a combination of classic geochemical tools and isotopic tools on the Bassée study site, located in the alluvial plain of the River Seine, to see whether they are relevant for tracing multiple and complex groundwater/surface-water relationships. The Ca/Sr ratio associated with Sr isotopes shows contrasted values and suggests that infiltration of surface water to groundwater increases when approaching the Seine. Furthermore, stable isotopes of the water molecule indicate that water from gravel-pit lakes may infiltrate into groundwater. Tritium and CFC tools confirmed surface-water influence on the Alluvial and Chalk aquifers. This geochemical approach, tested on the Bassée site, clearly demonstrates the need of using various geochemical tools for describing groundwater/surface-water relationships, and can be conclusively addressed to other case studies for helping decision makers in their management of natural water resources.

Keywords
δ¹⁸O, δ²H, ⁸⁷Sr/⁸⁶Sr, ³H, CFC, SF₆, wetland, groundwater, La Bassée, France

1. INTRODUCTION

In studies of hydrological systems over the past few years, new environmental tracers have been successfully applied, such as strontium, boron and sulphate isotopes, as well as CFCs and SF₆ (e.g. Vengosh et al., 2002; Négrel and Pauwels, 2004; Négrel and Petelet, 2005; Gooddy et al., 2006). New-generation inductively-coupled plasma mass spectrometers (ICP-MS) have allowed the development of much more precise isotopic measurements for various elements and a wider use of these tools (e.g. Luais et al., 1997; Marechal et al., 1999; Halicz et al., 1999; Rehkamper and Halliday, 1999; Galy et al., 2001; Rouxel et al., 2002; Beard et
al., 2003; Cardinal et al., 2003; Wombacher et al., 2003). This has provided larger means of studying hydrological and environmental processes.

Recently, the combined use of various geochemical tracers has been successfully applied for obtaining a better understanding of hydrological systems (Millot et al., 2007; Vengosh et al., 2007; Bouchaou et al., 2009; Ma et al., 2010) as well as of the interaction between groundwater and surface water (Oxtobee and Novakowski, 2002; Lamontagne et al., 2005; Darling et al., 2010). In Millot et al. (2007), the combination of Li, B, Sr and Nd isotopic systems highlights the complexity of studying geothermal reservoirs, as well as the fact that using only one isotopic tool could lead to an incomplete interpretation of the origin of water. For this reason, the use of a multi-tracer approach is relevant and should provide additional information for the characterization of natural waters in relation to their source.

Langman and Ellis (2009), investigating the source of water, showed the usefulness of a multi-isotope approach in a complex aquifer subject to natural and anthropogenic influences. The use of traditional geochemical tracers commonly is insufficient for explaining the source of water in deeper aquifers and the interactions between aquifers; in this case, additional tools such as δ¹¹B and ⁸⁷Sr/⁸⁶Sr permit a reinterpretation of the isotopic composition of water, leading to a conclusive revision of the hydrological model.

The complementary information these tracers provide, generally confirms the initially established conceptual hydrogeological model. But, in some cases all tracers provide the same information, rendering the use of various tools to be less than cost effective.

Occasionally, the information obtained from various geochemical tracers appears to be contradictory. Though, at first sight, his situation might be considered as a weakness of the multi-tool geochemical approach, it commonly reveals the strength of the method. Understanding a system can initially seem complicated, requiring reappraisal of the initial hypothesis; the use of multiple methods then is the only way of by-passing pre-established hydrogeological models and of proposing new models that consider the full complexity of hydrological systems.

In the present study, we proposed to use various tracers for evaluating the interconnection between aquifers and the possible relationships between surface water and groundwater in a flooded plain. These include tracers of the water cycle (δ²H and δ¹⁸O), of water-rock interactions (⁸⁷Sr/⁸⁶Sr), of residence time and transfer mode (CFCs and SF₁₆), and of anthropogenic impact (³H).

La Bassée lies south-east of Paris in the alluvial plain of the River Seine. In this area, hydrological flux between surface- and groundwaters is expected to be complex due to the presence of several interconnected aquifers below wetlands, ponds, oxbow lakes, and rivers. The Seine has been channelled since decades and natural flooding is quite reduced in the area. The Bassée site is the last alluvial plain in the Paris Basin exploited for gravel (Baron
and Piketti, 2001). The intensive sand and gravel extraction throughout the alluvial plain since 40 years has created large borrow-pits that, at the end of the exploitation period, are filled by ground- and rain-water. The Bassée site is also the major wetland area of the Ile-de-France region, labelled ‘Natura 2000’, and part of a Europe-wide network of sites tasked with the preservation of our natural heritage. It is one of the 87 wetlands of major importance in France and governmental action plans were established in order to protect the surface water covering 20% of the plain (http://natura2000.environnement.gouv.fr/sites/FR1112002.html).

The groundwater here is of great importance as a drinking-water supply and is considered as a reservoir to be protected for the Paris metropolitan-area water supply (SDAGE, Management Plan of Seine-Normandy basin). The study area covers 23 km² of the downstream part of the alluvial plain. Previous hydrogeological studies (Mégnien, 1965; Sogreah, 2004; Armines, 2005) using classical hydrogeological investigation methods (water-level monitoring, geology, geophysics, geotechnics, and modelling) left uncertainties on the magnitude of the relations between deep and alluvial aquifers, on the possible hydraulic connections between deep aquifers and the river, and on the extent of the relationships between the River Seine and shallow aquifers.

The scope of this study was to improve the hydrological understanding of the system through the use of multiple geochemical tools, in order to arrive at a better protection of this floodplain from qualitative and quantitative points of view. To this end, three different types of water relationships had to be evaluated in the alluvial plain: Chalk and Alluvial groundwater connections, groundwater (Chalk and Alluvial) and river water relationships, and the relation between other surface waters (oxbow lakes, ponds, gravel pit lakes) and groundwater. The alluvial plain in the past having been subject to periodic flooding by the Seine and today being intensively exploited for sand and gravel extraction, the existing surface water is of particular ecological and hydrological importance in the studied area.

2. CHARACTERISTICS OF THE BASSÉE SITE

The Bassée site, south-east of Paris, is of major importance for the drinking-water supply of Paris and surroundings. The site lies in the large Seine floodplain, delimited upstream by Bray-sur-Seine and to the west by the Montereau-Fault-Yonne at the junction between the rivers Seine and Yonne (Fig. 1). In view of its numerous ponds, canals and river arms, the Bassée plain was included in the “Natura 2000” program.

Sand and gravel exploitation and agriculture are the main economic activities in the area. Three aquifers are interconnected: i) the Alluvial aquifer of the River Seine located within the whole studied area, ii) the mainly unconfined Chalk aquifer below the alluvial one, and iii) the Tertiary calcareous formations in the northern part of the area (Fig. 2). The Alluvial and Chalk aquifers discharge into the Seine (Weng et al., 1999; Vernoux et al., 2004). Groundwater
within the Alluvial aquifer circulates following the Seine-flow direction, while groundwater flow in the Chalk aquifer is south-east to north-west. This geometry induces most probably a connection between the Chalk and Alluvial aquifers, with direct or indirect (by drainage) flow of the Chalk aquifer into the Alluvial layers. The very low regional slope created various meanders that have led to an important and heterogeneous Alluvial aquifer. The water-level map (Mégnien, 1965) indicates hydraulic continuity between these two aquifers. The Chalk aquifer is considered as a major component of Alluvial aquifer recharge.

In the northern part of the study area, the chalk formations are becoming less permeable and the groundwater is sometimes in semi-oxic or anoxic conditions. The Tertiary aquifer is located only in the northern part of the Seine floodplain and is a multilayer calcareous and marly calcareous aquifer, which locally may recharge the alluvial layers. Thus, recharge of the Alluvial aquifer occurs directly from precipitation, or through the other aquifers of the studied site. Natural Seine channels and spaces left by sand and gravel pits form numerous overflowing areas. During high-water periods, river water infiltration may be possible.

It is estimated that about 100 lakes were formed by sand and gravel extraction in the lowest part of the Bassée plain. After closure of the gravel pits, the space left is filled with low-permeability material such as clay and silt from recent alluvial deposits, and rapidly reaches equilibrium with the Alluvial and Chalk groundwater. These gravel-pit lakes are believed to have very low surface-groundwater exchanges (Schanen et al., 1998).

3. MATERIAL AND METHODS

3.1 Sampling procedures

Two sampling campaigns were carried out in June 2007 during the high water period and in November 2007 during a low water period. Sampled were 7 locations in the Chalk (noted Sc), and 12 locations in the alluvial aquifers (noted Sa), the Seine upstream at Bray-sur-Seine and downstream just before the confluence of the Rivers Seine and Yonne. Sampling was realised using 6" piezometers drilled in 2002. The complete technical description (total depth, depth to groundwater, lithology, depth to screens,...) is available and allowed to select only the piezometers exploiting a unic geological formation (Chalk or Alluvial). In addition, two gravel-pit lakes at La Tombe and Bazoches-lès-Bray and a well located at La-Chapelle-sur-Oreuse in the Chalk aquifer upstream from the Bassée alluvial plain were sampled in November 2007 (Fig. 1). A complementary sampling campaign took place in July 2008. Tritium analyses were carried out on water of all groundwater points sampled in 2007 and the Seine at Bray-sur-Seine.

The analyses of major chemical elements and stable water isotopes ($\delta^2$H, $\delta^{18}$O, $^3$H, $^{87}$Sr/$^{86}$Sr, were completed by the determination of apparent groundwater ages using CFCs
and SF$_6$.

The boreholes were sampled after pumping at least three purge volumes and stabilizing the chemical groundwater parameters, such as pH and conductivity. Samples for cation and $^{87}$Sr/$^{86}$Sr determinations were kept in polyethylene bottles after acidification with nitric acid. Raw water samples of 50 ml and 1 litre in HDPE bottles were used for $\delta^2$H, $\delta^{18}$O and $^3$H analyses. Glass sampling bottles of 500 ml were used for CFC and SF$_6$ analyses (IAEA, 2006).

3.2. Analytical methods

Concentrations were measured on water samples by inductively coupled plasma-atomic emission spectrometry (ICP-AES) for Ca$^{2+}$, Na$^+$, K$^+$ and Mg$^{2+}$, and by ion chromatography for Cl$^-$, NO$_3^-$, SO$_4^{2-}$ and PO$_4^{3-}$ (uncertainty for both methods 5-10%). Anion analyses were done by ion chromatography (model DX120, Dionex®), cations by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES; Ultima-2 model, Jobin Yvon®). The accuracy of both techniques was around 5-10% depending upon the concentration.

Sr concentrations were measured by inductively coupled plasma mass spectrometry (ICP-MS) (uncertainty 5%). Alkalinity was determined by HCl titration and Gran’s method. Strontium for isotopic analysis was separated using an ion-exchange column (Sr-Spec resin).

Procedural blank levels were lower than 0.5 ng. The $^{87}$Sr/$^{86}$Sr ratio was measured using a Finnigan MAT 262 multiple collector mass spectrometer. Sr isotopic ratios were normalized to $^{87}$Sr/$^{86}$Sr=0.1194. Average uncertainties on individual $^{87}$Sr/$^{86}$Sr measurements were 8.10$^{-6}$ (at 2σ level). International reference NIST-NBS987 measurements yielded $^{87}$Sr/$^{86}$Sr=0.710238±22 (2σ; N=18). Isotopic ratios of both O and H are given in per mil (‰) versus VSMOW (Vienna Standard Mean Ocean Water) reference material in the usual δ-scale defined as $\delta_{\text{sample}}$‰ = $\{(R_{\text{sample}}/R_{\text{standard}})-1\} \times 1000$, where R is the $^2$H/$^1$H and $^{18}$O/$^{16}$O atomic ratios. O and H isotopic composition of water was measured with a Finnigan MAT 252 mass spectrometer with an uncertainty of $\pm 0.1$‰ and $\pm 0.8$‰ for $\delta^{18}$O and δ$^2$H, respectively.

Samples for tritium activity measurements were collected in one litre polyethylene bottles. Analyses were done by direct liquid scintillation counting. The detection limit was 10 TU (Tritium Unit with 1 TU equal to 1 tritium atom in 10$^{18}$ hydrogen atoms). All isotopic analyses were carried out by the BRGM Geochemistry Laboratory.

Samples for CFC and SF$_6$ were collected in 500 mL glass bottles. To avoid any air contamination, each bottle was kept in a water-filled metal box. A simplified sample collection method described by IAEA (2006) was applied. The analyses were performed by gas chromatography with an electron-capture detector after pre-concentration using the purge-and-trap technique developed by Spurenstofflabor, Wachenheim (Germany). The detection limit was close to 10$^{-4}$ pmol, allowing the measurement of CFC concentrations down to
0.01 pmol L\(^{-1}\) and SF\(_6\) concentrations to 0.1 fmol L\(^{-1}\). The reproducibility was about ±5% for water samples.

4. RESULTS

4.1 Chemical composition of groundwater

Physico-chemical parameters (Table 1) indicate that groundwater conditions in the northern part of the studied area, on the right bank of the Seine, are mainly semi-oxic. Eh is <200 mV NHE (normal hydrogen electrode) for six wells in the Alluvial and Chalk aquifers. In addition, the NO\(_3\) concentration below the detection limit indicating possible denitrification for various points compatible with semi-oxic conditions observed in part of the aquifer. Surface- and groundwaters are of the Ca-HCO\(_3\) type (Fig. 3). Water from one well, Sa26, is of the Ca-SO\(_4\) type. Water in well Sc13 has very low SO\(_4\) concentrations, representing the more anoxic conditions and possible sulphate reduction.

Binary diagrams (Fig. 4a) confirm an excess of sulphate compared to chloride from rainfall origin. All surface- and groundwaters fall above the theoretical dilution line for sea water. Sulphate may originates from evaporates, but another possible source is gypsum for soil improvement, or fertilizers. In Figure 4b some points are also clearly enriched in chloride compared to sodium ions. As local rocks are evaporite free, potential chloride sources are rainfall and Manmade ones. Around Paris, Cl concentrations in precipitation are close to 0.15 mol/L (Mégnien, 1979). Considering an average enrichment factor (F) estimated by Rainfall\(_{\text{mm yearly average}}\)/(R-Evapotranspiration process\(_{\text{mm yearly average}}\)) of 1.5, chloride concentrations up to 0.23 mmol/L are from rainfall origin.

The anthropogenic impact on surface- and groundwater is important in the Bassée plain where agriculture is the main economical activity, even if gravel extraction gains on cultivated lands. On a NO\(_3\) vs. Cl diagram (Fig. 5) both groundwater from shallow (Alluvial) and deep (Chalk) aquifers present nitrate concentrations over 25 mg/L (0.4 mmol/L) and up to 83.1 mg/L (1.34 mmol/L; Sa21). Nitrate concentrations below the detection limit of 0.5 mg/L and chloride contents over 0.23 mmol indicate possible denitrification processes for these waters in wells Sa26, Sc09, Sc13, Sc16, SC22, SC4H, Sa16 and Sa38 (only for the November campaign). In addition, the gravel-pit lakes have very low nitrate concentrations (<0.5 and 2.8 mg/L or <0.01 and 0.05 mmol/L).

Intense pressure from agriculture is also reflected in groundwater by the presence of four pesticides in 116 analysed samples, which were detected at concentrations above the detection limits. These pesticides and degradation molecules are atrazine (max. 0.091 mg/L in the Voulzie River), deethylatrazine (max. 0.3 mg/L in Sc09), glyphosate (max. 5.59 mg/L in Sa37) and aminomethylphosphonic acid, AMPA (max. 2.52 mg/L in Sa21). Atrazine has not been used in this area since 2003. Glyphosate has both agricultural and non agricultural uses, which makes its origin difficult to identify.
Both x vs. y and Piper diagrams highlight a similar chemistry for both Alluvial and Chalk groundwaters. In addition, the chemistry of the Seine and Voulzie is not very different from that of groundwater.

4.2 Strontium isotopes

In the Bassée flood plain area, Ca vs. Sr shows a good correlation whereas the NO$_3$ vs. Sr correlation is weak, indicating a dominant lithological origin for Sr (Figs. 6a and 6b). Sr and the $^{87}$Sr/$^{86}$Sr ratio may therefore be controlled only by water-rock interaction in the Bassée flood plain, as anthropogenic input of Sr can be neglected.

The strontium-isotope range observed in this study agrees with values expected for Jurassic carbonate rock ($^{87}$Sr/$^{86}$Sr = 0.7067 to 0.7078) and Cretaceous Chalk ($^{87}$Sr/$^{86}$Sr = 0.7072 to 0.7080) (Koepnick et al., 1985, 1990). Furthermore, values measured in this study for the Chalk aquifer are compatible with those acquired by Négrel and Petelet-Giraud (2005) for this aquifer in the Somme Basin, north of Paris. Nevertheless, values measured in this study are globally higher than those documented in the previously cited article. This result implies the contribution of other Sr sources in addition to carbonate dissolution. The hypothesis of a strontium contribution from anthropogenic origin was rejected because of the low Sr concentrations observed (<8 μmol/L) and the lack of significant correlation between Sr concentrations and nitrate (Fig. 6b) or chloride concentrations (not shown here). On a $^{87}$Sr/$^{86}$Sr vs. 1/Sr (Fig. 7a) diagram, samples plot globally in a triangle. In this classic diagram, such a distribution suggests that, at least, three sources of dissolved Sr must explain the ground- and surface-water composition. One end-member should display low $^{87}$Sr/$^{86}$Sr values and high Sr concentrations (End-member 1). Another end-member is expected to have high $^{87}$Sr/$^{86}$Sr values and intermediate Sr concentrations (End-member 2). A third end-member would display low $^{87}$Sr/$^{86}$Sr values and low Sr concentrations (End-member 3). End-member 1 displays a Sr concentration and Sr-isotope composition close to the composition of the Sa26 sampling point ($^{87}$Sr/$^{86}$Sr = 0.70792 and 0.70796; 7.51 and 7.19 μmol/L) and compatible with a Sr contribution from carbonate dissolution. End-member 2 is close to the Sc22 and Sa37 sampling points, both of which display the highest $^{87}$Sr/$^{86}$Sr and Ca/Sr ratios measured in this study (Fig. 8). The Ca/Sr ratio was demonstrated elsewhere to be a good tool for tracing contrasting lithological origins (Brenot et al., 2008). As there is no correlation between $^{87}$Sr/$^{86}$Sr (or for the Ca/Sr ratio) and the NO$_3$ concentration (index of anthropogenic origin), End-member 2 most probably has a lithological origin. Nevertheless, the $^{87}$Sr/$^{86}$Sr and Ca/Sr ratios measured for the Sc22 and Sa37 sampling points (defining End-member 2) are much higher than expected values for local carbonate rock (as discussed above). A similar deviation towards a radiogenic composition (higher $^{87}$Sr/$^{86}$Sr values) was earlier observed locally in the Seine Basin (Roy et al., 1999; Brenot et al., 2008), and for the Chalk aquifer in
the Somme Basin (Négrel and Petelet-Giraud, 2005). In these cases, the authors suggested that radiogenic Sr might derive in part by thin marly-clayey beds in carbonate rock through ion exchange or desorption of Sr, or by dissolution of the potential dolomitic part of carbonate rock with a more radiogenic isotopic signature. End-member 3 is close to the composition of Seine water, water from gravel-pit lakes and the Sa48 and Sc13 sampling points. Furthermore, the Sr-isotope composition of Seine water (Roy, 1996) falls between 0.70777 and 0.70793, close to the composition expected for End-member 3. Thus, End-member 3 could potentially reflect a contribution of dissolved elements in surface water. Water in the Voulzie River has a different Sr isotopic signature compared to that of the Seine (Fig. 7) and water from gravel-pit lakes, which could be explained by significant differences in water-rock interaction. Indeed, the Voulzie River flows through Early Tertiary layers composed of clay and organic matter, whereas the Seine above the Bassée floodplain mainly flows over carbonate rock (Roy et al., 1999). The Chalk aquifer sampled upstream also has a quite different Sr-isotope signature compared to the Chalk aquifer sampled in the Bassée floodplain (Fig. 7).

On a $^{87}$Sr/$^{86}$Sr vs. Ca/Sr diagram (Fig. 7b), a positive correlation is seen between $^{87}$Sr/$^{86}$Sr values and the Ca/Sr ratio for all sampling points. The lowest Sr-isotope composition and Ca/Sr ratio in this study are in the Seine ($^{87}$Sr/$^{86}$Sr=0.70793 to 0.70778; Ca/Sr=424 to 964), whereas the highest Sr-isotope compositions and Ca/Sr ratios occur in groundwater at the Sc22 and Sa37 sampling points ($^{87}$Sr/$^{86}$Sr=0.70832 to 0.70858; Ca/Sr=1103 to 1253).

Based on only $^{87}$Sr/$^{86}$Sr values, and 1/Sr and Ca/Sr ratios (Fig. 7), one cannot discriminate the groundwater of the Alluvial and Chalk aquifers sampled in the Bassée floodplain.

4.3 Tritium and CFC data

The trends of CFCs and SF$_6$ used as input functions in air correspond to those of the Mace Head (Ireland) station from the ALE/GAGE/AGAGE program (available from cdic.esd.ornl.gov/ftp/ale_gage_Agage/; Prinn et al., 2000), the closest survey station to the Bassée site. Apparent groundwater ages were evaluated by using Excel® programs for CFC developed by the International Atomic Energy Agency (IAEA, 2006). The recharge date was determined by comparing the calculated partial pressure of CFCs and SF$_6$ in solubility equilibrium with the water sample, with historical CFC concentrations in local air. The equilibrium concentration of CFCs in groundwater depends on the air pressure (i.e. altitude) and temperature in the saturated zone. Recharge temperatures used are the temperature of groundwater at high-water-stage sampling time. The mean altitude of recharge is close to 55 m, the average altitude of the alluvial plain. The uncertainty in recharge temperatures and recharge elevations is respectively maximum 1°C and 50 meters.

CFC-11, CFC-12, CFC-113, and SF$_6$ were systematically measured for groundwater dating. For five sampling points, Sa26, Sa38, Sa48, Sc16 and ScH4, semi-anoxic conditions most
probably caused a degradation of CFCs (Horneman et al., 2008), in which case SF6 was then used for dating the water, this element being less sensitive to microbial degradation. All water samples could be dated at least once. Determination of apparent water ages using CFC and SF6 (Plummer and Busenberg, 2000) was done by applying a binary mixing model considering old water before 1950 (CFC-free) and young water with an age between 1985 and Present. This model was given the best results in “age-dating” (example for CFC-11 and CFC-113 given in Fig.8). Young-water mixing fractions ranged from 0.20 to 0.99. Results from the two sampling campaigns are quite similar. The uncertainty in recharge temperature estimation (about 1°C what is the annual variability of groundwater temperature) is leading to an uncertainty in mixing ratio of a maximum of 5%. Anthropogenic contamination was observed in few samples, mainly for CFC-12 and SF6 possibly due to the proximity of large urban areas (Oster et al., 1996; Santella et al., 2008).

The dissolved gases and age-dating interpretation show a significant differentiation between the Chalk and Alluvial aquifers. Only two wells (<20%) located in the Alluvial aquifer (Sa5, Sa20) have a percentage of young water below or equal to 50%, whereas five deep wells (>71%) tapping the Chalk aquifer (Sc11, Sc16, Sc22, ScH3, ScH4) show a percentage of young water below 50%. Surprisingly, wells Sc13 and Sc09 in the Chalk aquifer mainly contain young water, and a direct influence of surface water on these points is expected. Being far from the River Seine, the influence of ponds or gravel-pit lakes is supposed. The Alluvial groundwater in 75% of the wells is represented by water with a present (1985-2007) component below 80%. Therefore, the older and deeper groundwater component is significant, confirming the initial conceptual hydrogeological model of the Chalk aquifer discharging into the upper aquifer layers.

A few kilometres upstream from the Bassée floodplain, the nuclear power plant at Nogent-sur-Seine regularly has released tritium into the Seine since its start-up in 1988-1989. Tritium can thus be used as a radionuclide tracer (Sanchez-Cabeza and Pujol, 1999) of the infiltration of the Seine to groundwater since then. In the Seine at Bray-sur-Seine, the tritium concentration measured during the project is quite variable, from 21 TU in July 2008 up to 573 TU in April 2008, reflecting the variable release from the power plant (3.1 to 11.7 TBq/month for the period August 2006-August 2008, data provided by EDF, the operator). By comparison, the tritium values for Orléans precipitation (closest station at about 120 km from the Bassée site) have on average below 6 TU since 1999 (IAEA/WMO, 2009).

Tritium values measured in groundwater are very high (>60 TU) for Sc13, Sa41, Sa48 and Sa61, and high (>15 TU) for Sc09, indicating a more or less important influence of surface water on the Chalk and Alluvial aquifers at these points.

4.4. Stable isotopes
The δ²H and δ¹⁸O isotope composition of groundwater is -47 to -25‰ and -7.1 to -2.4‰, respectively (Table 1). In the δ²H vs. δ¹⁸O diagram (Fig. 9), most of the water falls on the local meteorological line (LML) represented by Orléans station (IAEA/WMO, 2009) with no significant difference between the Alluvial and Chalk aquifers and the River Seine. The average annual composition of δ²H and δ¹⁸O of the Seine water measured from oct.74 to sep.76 at Poses (upstream Paris, 200 km far from La Bassée; Conrad et al., 1978) is falling very close to the 1996-2010 weighted mean of precipitation collected at Orléans.

The most isotopically depleted groundwater is that sampled in the Chalk aquifer upstream from the Bassée area, which thus can be considered as the Chalk aquifer without influence by surface and/or alluvial water. Another groundwater sample taken in 2009 at 120 km west of the Bassée area in the Chalk aquifer of Paris Basin, has a similar isotopic content as that of the Chalk water sampled at La-Chapelle-sur-Oreuse. River water from the Seine or the Voulzie shows more enriched values. All other samples may be considered to be a mix between “pure” groundwater and surface (rainfall and/or river) water from different periods, and therefore with different isotope contents. Variation of the Seine isotopic composition is about 1 ‰ (Conrad et al., 1978).

Some points in the Alluvial (Sa26, Sa38) and Chalk (Sc13, Sc16) aquifers fall on a line with a slope of 4.3 that may correspond to isotopic fractionation caused by evaporation (evaporation lines have a slope between 3.5 and 6 as function of local atmospheric humidity; Gat, 1981). To test this hypothesis, we calculated the evaporation line with as starting point the average weighted δ¹⁸O and δ²H values of precipitation in Orléans for the period 1999-2009, and which would explain the compositions of samples Sa26, Sa38 (Alluvial aquifer) and Sc13, Sc16 (Chalk aquifer). The calculations show that the two gravel-pit lakes sampled in November 2007 have a stable-isotope composition falling onto the same evaporation line and display the most fractionated isotopic composition from all samples in the Bassée floodplain; here, water renewal is limited and therefore evaporation is expected to be high. Though to a smaller extent, river water is also evaporated and falls on the same evaporation line. This clearly indicates that some sampling points in groundwater integrate evaporated water that could be provided either by gravel-pit lakes or by river water.

5. DISCUSSION

5.1. Groundwater-river relationships

Existing models (Mégnien, 1979; Armines, 2005), consider that the Chalk aquifer supplies water to the Alluvial aquifer, which then discharges into the Seine. More specific studies (Chabart et al., 1992) have shown that, locally and occasionally, the River Seine can influence the Alluvial aquifer.
Based on only the major element concentrations, the geochemical composition of groundwater and surface water cannot be discriminated. Other geochemical tools are thus needed to reach a better understanding of the potential relationships between rivers and groundwater.

Sr isotopes are known (Négrel and Petelet-Giraud, 2005) to be an interesting tool for groundwater-surface water relationships. In the Bassée site the Sr ratio contrast observed between groundwater and surface water confirms its potential use in this context.

The spatial distribution of sampling points in the Bassée floodplain allows the construction of two north-south profiles perpendicular to the Seine. Along these profiles, the Ca/Sr ratio and $^{87}$Sr/$^{86}$Sr values (Fig. 10) show a significant decrease, with the lowest values close to the Seine for the high- and the low-water-stage sampling campaigns. Knowing the Ca/Sr ratio and $^{87}$Sr/$^{86}$Sr values of the Seine water, the concomitant decrease of these two geochemical tracers suggests an increasing contribution of Seine water to the groundwater close to the riverbanks. Furthermore, the Ca/Sr ratio (1139) and the Sr isotopic signature ($^{87}$Sr/$^{86}$Sr=0.70812) of Chalk aquifer groundwater collected at “La Chapelle”, 10 km away to the north (Fig. 1), are significantly higher than the signatures measured for groundwater collected closed to the Seine (Sa48, Sa41, ScH3, Sa26, Sa21, Sa20 and Sc16). The Ca/Sr ratio and Sr isotopic compositions thus clearly show a recharge of the aquifers by surface water during the high- and low-water stages. Surface water contribution seems to be of the same importance for both Alluvial and Chalk aquifers. River-water infiltration to groundwater is possible close to the river banks, and around ponds and gravel-pit lakes.

The sampling points located upstream and far from the Seine (Sc22, Sa37, Sc09, Sa61 and Sa65) have the highest Ca/Sr ratio (930 to 1134) and $^{87}$Sr/$^{86}$Sr (0.70817 to 0.70835) compared to the Seine water composition. At the opposite, sampling points located downstream of the Bassée alluvial plain and close to the Seine (Sc11, ScH3 and Sa02) have a Ca/Sr (876 to 916) close to the ratio measured at the upstream Seine station (not all points fall onto the N-S profiles and therefore cannot be seen in Figure 10).

Tritium-content data show that very high (>60 TU) values for Sc13, Sa41, Sa48 and Sa61 and high (>15) values for Sc09 indicate a direct influence of river water on the Chalk and Alluvial aquifers at these points. For well Sc13, located far from the main stream but close to a large flooded area, many of the ponds and gravel-pit lakes are most probably fed directly by the River Seine and the canal parallelo to the Seine. Tritium content then indicates the input of river water into groundwater via a residence period in ponds and gravel-pit lakes. A quantitative estimation is not possible as tritium input varies throughout the year.

### 5.2 Relationship between groundwater and gravel-pit lakes

Numerous gravel-pit lakes or natural ponds, swamps, oxbow lakes, and other backwaters exist in the flood plain. Chemical data and $^{87}$Sr/$^{86}$Sr ratios do not show significant differences...
between water from the aquifers and such surface waters. This was expected, as such waters mostly consist of groundwater.

In these water bodies, water renewal is limited and evaporation is quite high. Evaporation of water before infiltration was highlighted for some wells using the $\delta^2$H and $\delta^{18}$O isotope composition.

Most of the lakes were created by gravel extraction, causing a change in the hydraulic properties of the Alluvial aquifer. It is expected that these artificial holes also facilitate the infiltration of surface water down to the deeper parts of the aquifers. The high permeability of the walls suggests that water will mainly infiltrate horizontally into both the Chalk and Alluvial aquifers.

Water sampled at Sc13 in the Chalk aquifer is the most intensively evaporated. Due to the strong variability in H and O isotope signatures of the gravel-pit lakes (only two of them were sampled), it is possible that fractionation is even stronger in other superficial water bodies. The Sc13 water may result from a mixture of surface water with a more fractionated isotopic signature (higher H and O isotopic signatures) than the two gravel-pit lakes sampled in this study or the groundwater. The infiltration of surface water to the aquifer is also clearly shown at sampling points Sa26, Sa38, Sc13 and Sc16 (Fig. 9).

Dissolved gases (CFC-11, CFC-12, CFC-113, SF$_6$) and age-dating interpretation give quite heterogeneous results. Piezometers Sc13 and Sc09 in the Chalk aquifer mainly contain young water, and a direct influence of surface water on these points is thus expected. Being far from the Seine, the influence of ponds or gravel-pit lakes is most likely for these two wells. However, the recharge functioning is quite complex for Sc09 located in the eastern part of the study area, far from the Seine and any flooded area. No explanation could be found to explain the geochemical parameters of groundwater at this point.

5.3. Impact of the water dynamics on groundwater quality

An understanding of the hydraulic connections in the Seine floodplain may help in understanding groundwater contamination. Borrow pits were initiated in the area in the 1970s and rapidly increase in number. This extraction activity does not seem to have a direct qualitative impact on the groundwater. Agriculture today occupies 40% of the Bassée plain as against 46% in 1976 (Fustec et al., 2001). The dominant crops are cereals (wheat, maize, barley, rape, sunflower) and sugar beets, and diffuse pollution is expected at the study area.

Groundwater contamination can be quite high in the left bank of the Seine. Nitrate concentrations measured in June and November 2007 were, for 9 wells of the 19 sampled, over 17 mg/L and up to 83.1 mg/L (Sa21), confirming an anthropogenic impact. For 5 wells (4 in the Chalk and 1 in the Alluvial aquifer), reducing conditions may have led to denitrification. Existing data (www.eaufrance.ades.fr) underlines the importance of contamination of the Alluvial aquifer. Actually, 40% of the wells in the Bassée Alluvial aquifer
in the past showed triazine contamination at least once. Pesticide (mainly atrazine and DEA) and nitrate contamination have also been detected in wells tapping the Chalk aquifer. For the Water Framework Directive, both groundwater bodies (FRHG006 Alluvial aquifer on the Bassée site and FRHG209 Chalk aquifer) were classified as having a poor chemical status. The percentage of young groundwater was compared to nitrate concentrations (Fig. 10). A positive correlation was observed between the percentage of young water and nitrate concentrations for water in aerobic conditions, circulating in the Alluvial and Chalk aquifers. For most sampling points, the more important is the influence of the young groundwater, the higher are the nitrate concentrations (Fig. 11), whereby the “youngest” groundwater dates from 1985 up to 2007. In captive parts of the aquifer or around wetlands, the nitrate concentrations are below the detection limit (0.5 mg/L), most probably due to denitrification processes (in grey on Fig. 11). Seine water upstream and downstream from the study area has an average nitrate concentration lower than groundwater (<0.5 mg/L to 35.4 mg/L). For Sa61, Sa41, Sa48 relatively low nitrate concentrations compared to the apparent CFC ages may be explained by surface water influence (Fig. 11). In fact, the very high tritium content in groundwater from these three wells indicates a stronger influence of the Seine on groundwater, an influence that was also felt in Sc13 and Sc09. At the latter two points, a low redox potential suggests possible denitrification processes. In Sc09, the nitrate concentration in November 2007 (74.2 mg/L) and the redox potential (311 mv NHE) are higher than during the high water period (<0.5 mg/L and 345 mV NHE, respectively). As other chemical elements have similar concentrations for the high and low water sampling campaigns, denitrification is the most probable process for explaining major changes for only the NO₃ concentration.

The two sampled gravel-pit lakes have nitrate concentrations of 2.8 and <0.5 mg/L. Low nitrate concentrations can be explained by dilution (from rainfall and/or rivers), adsorption by plants and denitrification processes, as was earlier demonstrated in riparian wetlands upstream from the present study area (Bendjoudi et al., 2002). Concentrations of other chemical elements are similar or lower than in river water. Dilution by rainfall input is probably the most important factor, as Redox conditions may not allow denitrification processes and there are no or very few plants in the gravel-pit lakes. Previous work (Fuster et al., 2001) showed that silt deposits at the bottom of the gravel-pit lakes greatly reduce groundwater exchanges. Evaporation and recharge by precipitation therefore are dominating factors of the hydraulic balance of these specific surface-water bodies. Infiltration of water from gravel-pit lakes has a positive influence on groundwater quality. As for river water, the infiltration of water from gravel pits reduces, at least locally, the nitrate concentration in wells upstream from these artificial lakes.

6. CONCLUSIONS
From the geochemical tools the following conclusions may be drawn:

- No systematic chemical differentiation exists between the Chalk and Alluvial aquifer waters. This confirms that the conceptual hydrogeological model is correct at a regional scale, which considers that Chalk groundwater supplies water to the Alluvial aquifer. However, at the local scale more complex hydraulic connections exist between the different water bodies, as was highlighted by using isotope tools and age-dating. Both the Chalk and the Alluvial aquifers may be influenced by surface water.

- Age-dating highlighted the presence of young, i.e. less than 10 years old, groundwater in the wells Sc09 and Sc13 tapping the Chalk aquifer.

- The influence of Seine water, direct or indirect through the alluvial deposits, was shown using tritium and Sr isotope data for Sa41, Sa48 and Sa61.

- The impact on the aquifer of water stored in the gravel-pit lakes or other natural shallow lakes, ponds or swamps, was highlighted by using stable-isotope tools. The evaporation effect was clear on groundwater collected in wells Sc13, Sc16, Sa26 and Sa38. For Sc13, both a surface-water influence by the Seine through alluvial deposits and by infiltration from gravel-pit lakes is possible.

- $\delta^2$H and $\delta^{18}$O, tracers of water-rock interaction processes ($^{87}$Sr/$^{86}$Sr, Ca/Sr) and of water origin ($^3$H) differentiated the role of the River Seine, gravel-pit lakes (and most probably other superficial water bodies) in local groundwater recharge. The geochemical approach provided a spatial characterization of the origin of water and dissolved elements from surface and groundwater bodies.

- The geochemical approach tested on this site clearly demonstrates the efficiency of geochemical tools for describing the relationships between groundwater, river water and lake water. In addition, it underlined the need of using multiple tools for understanding a complex water system.

- The approach described in this paper can be usefully applied to other case studies, and will help decision makers in the context of natural water management.

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Figure and table captions

Figure 1 - Map showing the studied area and location of sampling points

Figure 2 - N-S synthetic geological profile through the Bassée area

Figure 3 - Piper diagram for ground- and surface waters sampled in June and November 2007 in the Bassée floodplain and in one well upstream in the Chalk aquifer

Figure 4 - SO$_4$ vs. Cl and Na vs. Cl for surface and groundwater sampled in the Bassée plain in June and November 2007 and groundwater sampled upstream (Chalk aquifer)

Figure 5 - NO$_3$ vs. Cl for surface and groundwater sampled at the Bassée plain in June and November 2007 and groundwater sampled upstream (Chalk aquifer)

Figure 6 - Ca vs. Sr and NO$_3$ vs. Sr for surface and groundwater sampled at the Bassée plain in June and November 2007 and groundwater sampled upstream (Chalk aquifer)

Figure 7 - $^{87}$Sr/$^{86}$Sr vs. 1/Sr (molar) and $^{87}$Sr/$^{86}$Sr vs. Ca/Sr (molar) for surface and groundwater sampled in the Bassée plain in June and November 2007 and in an upstream well

Figure 8 - CFC-11 vs. CFC-113 (in pptv) for groundwater collected in the La Bassée site in June and November 2007 and piston flow, exponential and binaxy mixing of young water (recharged in 1985 and 2005) with CFC-free water reference lines

Figure 9 - $\delta^{2}$H vs. $\delta^{18}$O for surface and groundwater collected in the Bassée site in June and November 2007; local meteoric line and mean weighted values for precipitations at Orléans (1996-2010), mean of Seine water at Poses (1974-1976) and calculated evaporation line

Figure 10 - $^{87}$Sr/$^{86}$Sr vs. distance and Sr/Ca vs. distance for groundwater collected in June and November 2007 along the two north-south profiles

Figure 11 - NO$_3$ vs. % of young groundwater for groundwater sampled in June and November 2007 in the Bassée alluvial plain

Table 1 - Analytical results for June and November 2007 sampling campaign
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<td>12/06/2007</td>
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<td>12/06/2007</td>
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Figure 2: Synthetic geological profile of the La Bassée area
Figure 3: Piper diagram for ground and surface water sampled in June and November 2007 in the La Bassée floodplain and one well groundwater upstream (chalk aquifer)
Figure 4: SO$_4$ vs. Cl and Na vs. Cl for surface and groundwater sampled at the La Bassée plain in June and November 2007 and groundwater sampled upstream (Chalk aquifer)
Figure 5: NO$_3$ vs. Cl for surface and groundwater sampled at the La Bassée plain in June and November 2007 and groundwater sampled upstream (chalk Aquifer)
Figure 6: Ca vs. Sr and NO$_3$ vs. Sr for surface and groundwater sampled at the La Bassée plain in June and November 2007 and groundwater sampled upstream (Chalk aquifer).
Figure 7: $^{87}$Sr/$^{86}$Sr vs. 1/Sr (molar) and $^{87}$Sr/$^{86}$Sr vs. Ca/Sr (molar) for surface and groundwater sampled at La Bassée plain in June and November 2007 and one upstream well.
Figure 8: CFC-11 vs. CFC-113 (in pptv) for groundwater collected in the La Bassée site in June and November 2007 and piston flow, exponential and binary mixing of young water (recharged in 1985 and 2005) with CFC-free water reference lines.
Figure 9: $\delta^2H$ vs $\delta^{18}O$ for surface and groundwater collected in the La Bassée site in June and November 2007; local meteoric line and mean weighted values for precipitations at Orléans (1996-2010), mean of Seine water at Poses (1974-1976) and calculated evaporation line.
Figure 10: $^{87}$Sr/$^{86}$Sr vs. distance and Sr/Ca vs. distance for the groundwater collected in June and November 2007 following the two North-South profiles.
Figure 11: NO₃ vs. % of young groundwater for groundwater sampled in June and November 2007 in the La Bassée alluvial plain.