

Geochemistry, isotopic composition (delta O-18, delta H-2, Sr-87/Sr-86, Nd-143/Nd-144) in the groundwater of French Guiana as indicators of their origin, interrelations Géochimie et compositions isotopiques (delta O-18, delta H-2, Sr-87/Sr-86, Nd-143/Nd-144) des eaux souterraines de Guyane comme indicateurs de leur origine et interrelations

Philippe Négrel, Emmanuelle Petelet-Giraud

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1	Geochemistry and isotopic composition ($\delta^{18}O$, $\delta^{2}H$, ${}^{87}Sr/{}^{86}Sr$, ${}^{143}Nd/{}^{144}Nd$) in
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3	interrelations
4	
5	Géochimie et compositions isotopiques (δ^{18} O, δ^{2} H, 87 Sr/ 86 Sr, 143 Nd/ 144 Nd) des
6	eaux souterraines de Guyane comme indicateurs de leur origine et
7	interrelations
8 9 10 11 12 13 14 15	Philippe NÉGREL, Emmanuelle PETELET-GIRAUD B.R.G.M., Avenue C. Guillemin, BP 6009, 45060, Orléans Cedex 02, France, e-mail: <u>p.negrel@brgm.fr</u> ; e.petelet@brgm.fr
14 15 16 17 18 19 20 21 22 23 24 25	Abstract The current use of untreated river water for drinking purposes by the population of French Guiana has important impacts on public health. Consequently, groundwater is of major importance as a possible alternative drinking water supply to reduce these impacts. Since French Guiana belongs to the Guyana Shield, sustainable water management can be expected to depend increasingly on water from fissured aquifers in hard rocks. Groundwater samples were collected from shallow drill holes in the densely populated coastal area, and deeper wells in the basement (around Cayenne and along the Maroni and Oyapock rivers). This study reports on major and trace elements for which Na ⁺ and Ca ²⁺ excess with regard to Cl reflect the role of water-rock interaction, as well as Sr and Nd isotopes that reflect the role of the different lithologies. δ^{18} O and δ D in waters give constraints on the water cycle (recharge and evaporation processes).
26 27 28 29 30 31 32 33 34 35 36 37	Résumé L'utilisation d'eau de surface non traitée comme eau de boisson par la population guyanaise a des effets importants en termes de santé publique. En conséquence, les ressources en eaux souterraines sont d'une importance majeure comme alternative pour l'alimentation en eau potable afin de réduire les impacts sur la santé publique. La Guyane française faisant partie du bouclier guyanais, les ressources en eau souterraine sont probablement contenues dans les roches fracturées. Des eaux souterraines ont été prélevées dans des forages peu profonds dans les zones de forte densité de population dans la zone côtière et dans des ouvrages plus profonds dans les zones de socle (proche de Cayenne et le long des fleuves Maroni et Oyapock). Cette étude présente la géochimie des éléments majeurs et traces pour lesquels les excès de Na ⁺ et Ca ²⁺ par rapport au Cl reflètent le rôle des interactions eau-roche tandis que les isotopes du Sr et Nd montrent le rôle des différentes lithologies drainées. Les isotopes de la molécule d'eau (δ^{18} O et δ D) tracent le cycle de l'eau, précisant la recharge et les processus d'évaporation.
38 39	Keywords: French Guiana, Groundwater, Strontium isotopes, Neodymium isotopes

40 Mots clés : Guyane française, eau souterraine, isotopes du strontium, isotopes du néodyme

42 **1. Introduction**

Groundwater resources are of the utmost importance in French Guiana for water supply because the use of untreated river water for drinking by the inland population induces important health impacts. Since French Guiana belongs to the Guyana Shield, sustainable water management can be expected to depend increasingly on water from fissured aquifers, e.g. mainly hard rocks (crystalline, metamorphic and volcanic rocks). The exploitation of such fissured (heterogeneous and anisotropic) systems has to be linked to the characterisation of aquifer structure and functioning, aquifer heterogeneities and relationships with surface water (Négrel and Lachassagne, 2000; Négrel et al., 2002).

51 Present day research has to focus on increasing use of existing geochemical tools (such as Sr-Nd isotopes as well 52 as lead isotopes) dedicated to elucidate the structure and functioning of the different compartments of hard rock 53 aquifers, i.e. overlying sediments, when they do exist, weathered cover alterites, weathered-fissured zone, 54 fractured hard rock (Steinmann and Stille, 2006; Négrel, 2006). In particular, research will have to deal with the 55 identification of the relative signature of groundwater circulations in the alterites and in the underlying 56 weathered-fissured zone. This zone, with efficient porosities ranging from 3 to 20% in the alterites and from 0.5 57 to 2% in the weathered-fissured zone, may contain most of the groundwater reserve (since the efficient porosity 58 of fresh bedrock is often less than 0.01%). This will also help to identify the role of these different 59 hydrogeological compartments, both under natural and pumping conditions, and in the framework of surface-60 groundwater relationships. These methods give better understanding of the alterites and underlying weathered-61 fissured zone as hydrogeophysics, e.g. use of advanced geophysical methods to understand the interaction 62 between geology and fluid flow in the subsurface, did (Auken et al., 2009; Sailhac et al., 2009).

63 Since a sufficient number of boreholes have been drilled in French Guiana (Fig. 1), ongoing research is now 64 focusing on the geochemistry of ground waters for which the database will serve to build up a referential for this 65 region and will be valorised in conjunction with geologic, hydrodynamic, etc. data, as one main objective will be 66 to better define the functioning of French Guiana hard rock aquifers.

67

68692. General conditions in French Guiana

70 French Guiana covers 10% of the Guyana Shield, which represents the northern extension of the Amazonian 71 Platform (Edmond et al., 1995; Deckart et al., 2005). The Guyana Shield comprises three rock complexes: the 72 Imataca Archaen gneiss (3.4-2.7 Ga), the Lower Proterozoic volcano-sedimentary terrains and granite-gneiss 73 rocks (2.3-1.9 Ga), and the Middle Proterozoic continental deposits and magmatic rocks (1.9-1.5 Ga). With 74 regard to its weathered substratum, which is composed only of Lower Proterozoic (2.5-1.9 Ga) igneous and 75 metamorphic rocks, French Guiana is similar to the Guyana Shield drained by the Orinoco (Edmond et al., 76 1995). The geology of French Guiana can be summarized as four different units (Gruau et al., 1985; Deckart et 77 al., 2005) with the Cayenne Island series, the Lower Paramaca (hereafter referred to as unit P) of mainly 78 metavolcanic rocks and rare sediments, the Upper Paramaca (hereafter referred to as unit S) and Orapu 79 comprising schists, mica schists, quartzites, conglomerates, metagraywackes, metasiltites and rare metavolcanic 80 rocks and finally plutonic intrusions of gabbro-diorite, granite and granodiorite from the "Guyana plutonism", 81 and granitoid, granodiorite and tonalite from the "Caribbean plutonism".

82 The climatic and geodynamic conditions over the Guyana Shield since the Cretaceous have induced the 83 extensive development of 50 to 100 m thick alterites masking the substratum (Driscoll and Karner, 1994). The 84 climate is humid tropical, with two rainy seasons and two dry seasons and the total annual effective rainfall is 85 around 1000 mm.y⁻¹. Most of the groundwater resource is located in the crystalline bedrock, typical of those of 86 tropical hard rock settings. The alluvia have high clay content, mainly due to the nature of the sediments in 87 tropical settings and also due to local weathering, they are relatively thin (mainly less than 5 m) and are of 88 reduced lateral extent; they therefore account for only a small volume of groundwater. The alterites, which 89 constitute the upper compartment of the bedrock aquifers, have an effective porosity of a few percent and thus a 90 water storage function.

91

92 93 **3. Sampling procedures and analytical methods**

94 Groundwater samples (Fig.1) were collected 1) along the coastal area bordering the Atlantic Ocean from shallow 95 drill holes in the extensive sandy-argillaceous terrane, mainly Holocene in age, (GWA/alt-sed in Table 1), which 96 is the only densely populated area in French Guiana (Négrel et al., 2002), 2) along the Maroni and Oyapock 97 River catchments (Négrel and Lachassagne, 2000) from shallow wells in the alluvia (GWA/alt-sed in Table 1). 98 Groundwater samples were also collected from deep wells in the basement (Fig. 1), MM4, CAR1 and MKT2 99 around Cayenne (GWA/bas in Table 1) from which groundwater is pumped from bedrock fractures, deep wells 100 (GWA/bas in Table 1) in the basement along the two main rivers Maroni and Oyapock (F1, 2, M1, 3bis, 4, 5, 101 L1bis, 2, CR1, 2) and more inland (Ni5).

- 102 Groundwater were filtered on site through 0.2 µm acetate cellulose filters and stored in pre-cleaned 103 polypropylene bottles. The samples for cation and isotope measurements were acidified to pH 2 with ultrapure 104 HNO₃, and one bottle of each sample (not acidified) was kept for anion determination. Electrical conductivity, 105 water temperature and pH were measured in the field with a conductivity meter standardized to 20 °C and with a 106 combined electrode and a pH-meter regularly calibrated using two standard buffers. Chemical analysis of the 107 water samples was carried out by capillary ion electrophoresis for major cations and anions, by ICP-MS for Sr, 108 Sm and Nd and HCl titration and Gran's method for the total alkalinity. Precision ranged between ± 5 and 10% 109 for the determination of major and trace elements measurements. Classical methods for the separation by 110 exchange column (cation for Sr, cation and HDEHP reverse chromatography for Nd) and isotope analysis 111 (Finnigan MAT 262 multiple collector mass spectrometer) were used for Sr and Nd isotope measurements 112 (Négrel and Lachassagne, 1997; Négrel et al., 2000). The reproducibility of ⁸⁷Sr/⁸⁶Sr measurement was tested by 113 duplicate analyses of the NBS 987 standard (mean value 0.710227 ± 17.10^{-6} , 2σ , n = 70) of the La Jolla 114 international standard for Nd (143 Nd/ 144 Nd of 0.511826 ± 11.10⁻⁶, 2 σ , n = 33). The 143 Nd/ 144 Nd ratios are 115 expressed as $\epsilon Nd(0)$, which represents the deviation in parts per 10⁴ (ϵ unit) from ¹⁴³Nd/¹⁴⁴Nd in a chondritic 116 reservoir with a present day CHUR value of 0.512636. Data are presented in Table 2.
- 117

118
1194. Results and Discussion

120 4.1. Chemical characterisation of waters

121
122 The Total Dissolved Solids (TDS) fluctuates from 10 up to 166 mg.L⁻¹ in the groundwater from the extensive
123 sandy-argillaceous terrane and from the alluvia (hereafter referred to as GWA/alt-sed) and from 46 to 218 mg.L⁻¹

124 in the groundwater from the basement (hereafter referred to as GWA/bas). The water chemistry shows a large 125 variation in major element contents (Table 2). Chloride content in the GWA/alt-sed groundwater ranges from 50 126 to 4241 μ mol.L⁻¹ and from 45 to 457 μ mol.L⁻¹ in the GWA/bas groundwater. Though Cl⁻ ions do not have any 127 significant lithological origin, since French Guiana is evaporite free, Cl⁻ ions in groundwater may originate 128 mainly from rainfall recharge (sea salts, Gaillardet et al., 1997), and to a lesser extent from human activity 129 (domestic sewage, fertilisers, etc). Chloride often behaves conservatively through the hydrological cycle and is 130 used as an atmospheric-input reference element in many hydrosystems (Gaillardet et al., 1997). Like Cl⁻, Na⁺ 131 shows a wide range of contents and, when compared to Cl⁻ in Figure 2a, Na concentrations in most surface and 132 ground waters plot above the seawater dilution line (SWDL), indicating Na⁺ excess. Most of the surface and 133 ground waters from the GWA/alt-sed plot between the SWDL and the line with a molar Cl/Na ratio of around 134 0.5. Three GWA/bas ground waters (CR1, CR2 and F1-oct-98) plot with a molar Cl/Na ratio of 0.7, the 135 hydrogeological data suggest that the alterite compartment contributes significantly to the well flow rate. The 136 remaining GWA/bas ground waters plot with a molar Cl/Na ratio of 0.2. This divergence from the seawater 137 dilution line reflects a large Na enrichment that is mainly related to water-rock interaction. Ca²⁺ concentrations 138 show a large range in groundwater from about 5-10 to 999 μ mol.L⁻¹ and fluctuate also largely in rain- and 139 surface waters when compared to Cl⁻ in Figure 2b (all samples plot above the SWDL, with a Cl/Ca ratio of 1.42 140 for the surface waters indicative of a Ca^{2+} excess). The groundwater from the GWA/alt-sed also display a Ca^{2+} 141 excess and plot with a molar Cl/Ca ratio ranging between that of SWDL and the ratio 1.42, four of them plot 142 with a molar Cl/Ca ratio higher than 1.42. The GWA/bas groundwater plot with a molar Cl/Ca ratio ranging 143 between 1.42 and 0.05, reflecting the larger Ca^{2+} excess. Some of them plot with a molar Cl/Ca ratio of around 144 1.42, close to the groundwater from the GWA/alt-sed, reflecting the fact that water partly originates from the 145 alterite compartment. Similarly, K⁺, Mg²⁺ and Sr²⁺ excess are observed when plotted versus Cl⁻, (not shown), 146 reflecting the weathering of aluminosilicates.

147 The positive correlation between HCO₃ and the sum of cations in groundwater samples (Σ^+ , $r^2 = 0.55$, n = 38) 148 clearly indicates that the cations released by weathering are balanced by the alkalinity, in good agreement with 149 observations in other South American basins influenced by similar climatic conditions (Gaillardet et al., 1997; 150 Freyssinet and Farah, 2000). The bicarbonate increase during weathering originates from the atmospheric/soil 151 CO₂, which implies a positive correlation between HCO₃ and pH in groundwater samples ($r^2 = 0.66$, n = 38) 152 related to the water-rock interaction.

153

154 155 4.2. Investigating groundwater recharge through $\delta^2 H$ and $\delta^{18} O$ isotopic signature

156 Variations in the stable-isotope composition in a catchment's water balance are mainly caused by natural 157 variations in the isotopic composition of rainfall and through mixing with pre-existing water and the influence of 158 evaporation (Kendall and McDonnell, 1998). The stable isotopes $\delta^2 H$ and $\delta^{18}O$ of all rain samples collected in 159 French Guiana (Négrel et al., 1997) defines the local meteoric water line (LMWL), as illustrated in Figure 3a. 160 The δ^{18} O and δ^{2} H in GWA/alt-sed ground waters fall in the range -2.1 to -3.7% for δ^{18} O and -7.9 to -19.2% for 161 δ^2 H and in the range -2.9 to -4.0% for δ^{18} O and -12.3 to -18.4% for δ^2 H for the GWA/bas ground waters, in 162

close agreement with the range of surface water (Négrel and Lachassagne 2000).

- 163 The δ^{18} O and δ^{2} H relationships are illustrated in Figure 3a and b for rainwater (regression line and 95%) 164 confidence range), global meteoric water line (GMWL; $\delta^2 H = 8 \delta^{18} O + 10$, Craig, 1961), surface waters and 165 groundwater. Surface waters plot close to the LMWL in Figure 3b and, as demonstrated by Négrel and 166 Lachassagne (2000), most show a significant shift (close to 1.4‰) to the right of this line as result of evaporation 167 processes. Most of the ground waters from GWA/alt-sed clearly plot close to the local and global lines. Some of 168 these ground waters are displaced below the LMWL. This could be due either to evaporation prior or during 169 infiltration or, as suggested by Boronina et al. (2005), because of partial evaporation from soils and dilution by 170 subsequent recharge. GWA/bas groundwater also plot close to the LMWL and GMWL, reflecting a meteoric 171 origin and a lack of significant evaporation during recharge or oxygen isotope exchanges between water and the 172 rock matrix.
- 173 Infiltrating rainwater shows a mean weighted signatures for δ^{18} O and δ^{2} H of -4.51‰ and -24.23‰ respectively, 174 corresponding to 1790.4 mm of rainfall (Négrel et al., 1997). However, mean rainwater δ-values cannot explain 175 groundwater recharge as less negative values occur in many samples (Fig. 3b). Thus recharge from precipitation 176 events occurs in significant amount at least during two periods The first period was September to January and 177 corresponds to the rainy season with 496 mm of rainfall (27.7% of the total annual rainfall, mean weighted rain 178 signatures input led to δ^{18} O and δ^{2} H of around -2.45‰ and -7.58‰, respectively). The second period occurred in 179 March-April with 249 mm rainfall, which corresponds to the middle of the rainy season (13.9% of the total 180 annual rainfall, mean weighted rain input led to δ^{18} O and δ^{2} H signatures of around -3.10% and -11.11%, 181 respectively). These two rainy periods should constitute the recharge period that would explain the least negative 182 δ -values in the ground waters. It is worth noting that part of the δ^{18} O and δ^2 H signatures in the groundwater 183 should reflect the recharge occurring during May to July. During this period, the rainfall was 943.3 mm, (52.7% 184 of the total annual rainfall) and the mean weighted rain input led to δ^{18} O and δ^{2} H signatures of around -4.24‰ 185 and -20.89%, respectively. This period of rain and thus recharge may explain the most negative δ -values in the 186 groundwater samples. This suggests different stages of groundwater recharge with isotope signatures in close 187 connection with the range in rainwater.
- 188

4.3. ⁸⁷Sr/⁸⁶Sr as a proxy of weathering processes 190

Sr isotope studies of rivers and lakes have shown that variations in ⁸⁷Sr/⁸⁶Sr and Sr contents are caused by
 mixing waters with different ⁸⁷Sr/⁸⁶Sr ratios and Sr contents, each of them reflecting water-rock interaction with
 different rock types (Oliver et al., 2003; Grove et al., 2003; Chung et al., 2009).

194 All the surface- and groundwater samples from French Guiana are plotted in a ⁸⁷Sr/⁸⁶Sr vs. 1/Sr diagram 195 (Fig. 4a), which is classically used to evaluate two-component mixing and end-member water compositions. 196 This figure indicates the existence of at least three end-members. The surface waters from the Maroni catchment, 197 reported from Négrel and Lachassagne, 2000, plot along the mixing trend between the end-members 198 corresponding to the drainage of the Unit P metavolcanic rocks (low ⁸⁷Sr/⁸⁶Sr ratio, highest [Sr]) and that of the 199 Unit S meta-sedimentary lithology (highest ⁸⁷Sr/⁸⁶Sr ratio, intermediate [Sr]). The low ⁸⁷Sr/⁸⁶Sr ratio would 200 reflect the weathering of rocks such as basalt and amphibolite that are known to impart a low ⁸⁷Sr/⁸⁶Sr ratio to 201 the waters (Louvat and Allègre, 1997; Dessert et al., 2001) while the high ⁸⁷Sr/⁸⁶Sr ratio would be related to the 202 weathering of schists and micaschists, which deliver higher ⁸⁷Sr/⁸⁶Sr ratios to the water (Aubert et al., 2002). 203 This compares to the results obtained on rivers draining "undifferentiated Proterozoic rocks" of the Guyana 204 Shield (Edmond et al., 1995), which also lie on the mixing trend between the S and P end-members. The third 205 end-member (intermediate ⁸⁷Sr/⁸⁶Sr ratio, low [Sr]) could correspond to the drainage of plutonic granitoid 206 intrusions ($\delta\eta$). In the Maroni catchment, the shift of some surface waters to the right of the mixing trend 207 between S and P end-members could reflect the input into the main stream of tributaries draining weathered 208 granitoids. This is also illustrated by some rivers from the GWA/alt-sed (Négrel et al., 2002) that plot in the field 209 δη in the Figure 4a. Some ground waters from the same area plot very close to the mixing trend between S and P 210 end-members but most of them plot with a low ⁸⁷Sr/⁸⁶Sr ratio, reflecting the influence of the drainage of volcanic 211 rocks of the Lower Paramaca (Unit P). It is worth noting that some points of groundwater are shifted with a 212 higher 1/Sr ratio agreeing with the rainwater samples. Therefore it may be concluded that the shift reflects a 213 large rainwater input for the Sr budget compared to the Sr released by water-rock interaction.

214 The GWA/bas groundwaters lie on the straight line corresponding to a mixing between waters that have 215 interacted with rocks from Unit P and S. Most ground waters display the lowest ⁸⁷Sr/⁸⁶Sr ratio, reflecting the 216 interaction with rocks having a low Sr isotopic ratio. However, the location of the deep ground waters from 217 Grand Santi (F1, F2) is not consistent with an interaction with volcanic rocks of the Unit P because the boreholes 218 were drilled within granitoids. One way to explain the similarity between the drainage of volcanic rocks (Unit P) 219 and ground waters that have interacted with granitoids is to consider that the latter are influenced by the 220 weathering of low Rb-rich Sr phases, which would impart a low ⁸⁷Sr/⁸⁶Sr ratio to the waters. Weathering of low 221 Rb-rich Sr phases and more constraints on the ⁸⁷Sr/⁸⁶Sr ratio variations may be tested through a diagram 222 87 Sr/ 86 Sr versus Ca/Na ratio (Fig. 4b). The use of a cation ratio rather than absolute concentrations alone, avoids 223 variations due to dilution or concentration effects (BenOthmann et al., 1997, Chung et al., 2009). As previously 224 detailed, when Ca is compared to Cl content (Fig. 2b), all ground waters are indicative of a Ca^{2+} excess, more 225 marked in the ground waters from the basement, as well as for Mg, while the Na enrichment is less. Thus, Ca/Na 226 appears to be a good indicator of weathering processes in waters from French Guiana. The surface water from 227 the Maroni catchment does not define a clear hyperbola that binary mixing should lead to (Langmuir et al., 228 1978). They appear rather to be scattered along a main trend showing a large fluctuation in the ⁸⁷Sr/⁸⁶Sr ratio 229 with a weak Ca/Na range. This makes possible to define a first water-rock interaction process that corresponds to 230 surficial weathering of unit P and S. Surficial weathering of Unit S shows a larger ⁸⁷Sr/⁸⁶Sr ratio without changes 231 in the Ca/Na ratio. Rainwater field (RW in Fig. 4) shows a low Ca/Na ratio and a ⁸⁷Sr/⁸⁶Sr ratio around 0.710. 232 Two other trends can be defined with a weak fluctuation in the 87 Sr/ 86 Sr ratio within a change of the Ca/Na ratio. 233 The first trend corresponds to a water-rock interaction process from the surficial to the deep weathering of 234 granitoids. The deeper weathering of granitoids reflected in this trend implies that the weathering of Ca-bearing 235 phases (e.g. plagioclases and/or calcite, Pett-Ridge et al., 2009) is greater in sample MM4 than in sample CRM2. 236 The second trend corresponds to a water-rock interaction process from the surficial to the deep weathering of the 237 unit P with a weak fluctuation of the ⁸⁷Sr/⁸⁶Sr ratio (around 0.704) associated with a large increase in the Ca/Na 238 ratio. As for the first water-rock interaction processes, the shift towards a relatively high Ca/Na ratios can be 239 explained by more extensive weathering of Ca-bearing phases with a low ⁸⁷Sr/⁸⁶Sr ratio, such as plagioclase 240 and/or calcite. High Ca/Na ratios generally relate to carbonate weathering (Gaillardet et al., 1997), which is also 241 accompanied by increase in the Mg/Sr and Ca/Sr ratios (Rengarajan et al., 2009). The increase in the Ca/Na 242 ratios during the weathering of the unit P is accompanied by an increase of the Ca/Sr ratios (up to 500)

- 243 notwithstanding that this value is the lowermost value of carbonate weathering (Meybeck, 1986). On the other
- 244 hand, large Ca/Na ratios were observed during basalt weathering (up to 3, Dessert et al. 2001; Raiber et al., 2009; 245
- Rengarajan et al., 2009). Such large Ca/Na ratios were accompanied by Ca/Sr ratio around 500 (Dessert et 246 al.2001). Thus we can conclude that the trend in the GWA/bas ground waters from the unit P may reflect a more
- 247 intense weathering of Ca-bearing phases like plagioclases as no evidence of calcite has been reported in basalts.
- 248
- Using the ⁸⁷Sr/⁸⁶Sr and Ca/Na ratios of the extreme samples of the surficial and deep weathering of the unit P, 249 which can be considered as end-members, a mixing line was calculated and reported in Figure 4b. The first end-250 member corresponds to the surficial weathering with a ⁸⁷Sr/⁸⁶Sr and Ca/Na ratios of around 0.703 and 0.4. The 251 latter represents the value classically used for the weathering of silicate rocks (Gaillardet et al., 1997; Louvat and 252 Allègre, 1997; Rengarajan et al., 2009). The second end-member shows a slightly higher ⁸⁷Sr/⁸⁶Sr ratio (i.e. 253 0.705) and a greater Ca/Na ratio (i.e. 5). The calculation led to a divergence from the surficial weathering end-254 member of around 10% for F5, 25% for F2 and M4 and 50-60% for L1bis and L2.
- 255

256 257 4.4. Nd contents and Nd isotopes: implications for water-rock interactions

258 Since the development of the method by ICP-MS to determine the dissolved REE concentrations (Stetzenbach et 259 al. 1994, Johannesson et al. 1995), many studies deal with the behaviour of the REE in continental waters 260 (Gaillardet et al., 1997; Viers et al., 2000; Andersson et al., 2001; Janssen and Verweij, 2003 and references 261 therein) but, in contrast, neodymium isotopes have not been extensively used in hydrogeological studies. Some 262 data are available on the compositions of river water (Goldstein and Jacobsen, 1987; Tricca et al., 1999; 263 Andersson et al., 2001; Steinmann and Stille, 2006) and very little information is available for saline waters 264 (Négrel 2006) and ground waters (Tricca et al., 1999; Viers and Wasserburg, 2004). The contents of Sm and Nd, 265 as well as for the other dissolved REEs vary greatly. The dissolved Nd content in the surface waters varies from 266 20 to 102 ng.L⁻¹ with a low TDS (10-20 mg.L⁻¹), in agreement with values reported by other studies (Amazon, 267 Gaillardet et al., 1997; Cameroon, Viers et al., 2000, Viers and Wasserburg, 2004) but broadly independent of 268 other parameters such as total dissolved solids and pH. In the groundwater from the GWA/alt-sed, the dissolved 269 Nd content varies from 39 to 752 ng.L⁻¹ together with a broad range in the TDS (10 up to 166 mg.L⁻¹). In the 270 GWA/bas groundwater, the dissolved Nd contents are lower and vary from 3 to 63 ng.L⁻¹ with TDS values 271 ranging from 46 to 218 mg.L⁻¹. These ranges agree with other studies in groundwater (Oliva et al., 1999; Viers 272 and Wasserburg, 2004). As usually stated in terrestrial surface processes and in granite-gneiss rocks, Sm displays 273 similar behaviour to Nd (Nd = $4.9 \times \text{Sm} + 6.5$, $r^2 = 0.98$, n = 30), because of the similarity in their physical and 274 chemical properties (Goldstein and Jacobsen, 1987; Gaillardet et al., 1997; Viers and Wasserburg, 2004). The 275 isotopic composition of dissolved Nd in surface and ground waters from French Guiana ranges from ¹⁴³Nd/¹⁴⁴Nd 276 $= 0.511377 \pm 9.10^{-6}$ to 0.512162 ± 8.10^{-6} corresponding to a range of ϵ Nd(0) from -9.2 to -24.6. The lowest 277 ɛNd(0) are observed in the GWA/alt-sed groundwater whereas the highest are for the GWA/bas groundwater. 278 The small differences between $\varepsilon Nd(0)$ values for both the dissolved and suspended phases found within the same 279 rivers suggests that the Nd isotopic composition can be used as an indicator of the weathered parent rock 280 (Goldstein and Jacobsen, 1987, 1988; Tricca et al., 1999). Mixing processes and water-rock interaction have 281 both contributed to the observed $\varepsilon Nd(0)$ (Négrel et al., 2001).

282 The similarity between the ¹⁴³Nd/¹⁴⁴Nd ratios in waters and related bedrock can be postulated now according to 283 the recent works by Aubert et al. (2001). Comparing the Nd and Sr isotopic composition in minerals and waters, 284 they demonstrated that the Sr and Nd isotopic characteristics of the major mineral phases of a granite clearly 285 shows the strong influence of plagioclase and phosphate minerals (e.g. apatite) on the isotopic composition of 286 the spring and stream waters. This was confirmed on other granite massif (France) by Négrel (2006). Thus, 287 comparison of the ɛNd(0) values with the corresponding Sm/Nd ratios (e.g. ¹⁴⁷Sm/¹⁴⁴Nd ratios represented with 288 the propagate errors) yields more information about the origin of the REE in the waters from French Guiana 289 (Fig. 5). With the exception of the samples GUY99-11 and F1, they are positively correlated ($\epsilon Nd(0) = -43.3 +$ 290 233.8 x 147 Sm/ 144 Nd; r² = 0.64; n = 16) with low 147 Sm/ 144 Nd ratios for the river water and rather high ratios for 291 the ground waters.

- 292 Most of the ground waters have ¹⁴⁷Sm/¹⁴⁴Nd ratios significantly higher than the average continental crust value 293 of 0.105 (Allègre and Lewin, 1989). The ɛNd(0) vs. the ¹⁴⁷Sm/¹⁴⁴Nd ratio in the surface water and GWA/bas 294 groundwater, illustrated in Figure 5, are consistent with that of the parent rocks (Gruau et al., 1985; Delor et al., 295 2001). If the $\varepsilon Nd(0)$ is mostly controlled by the weathering of plagioclase and phosphate phases, as suggested by 296 Aubert et al. (2001) and Négrel (2006), they should be shifted towards higher ɛNd(0) and Sm/Nd ratios and thus 297 would plot on the right of the parent rocks field, which is not observed. The $\epsilon Nd(0)$ vs. the ¹⁴⁷Sm/¹⁴⁴Nd ratio for 298 the groundwater from the GWA/alt-sed plot between the values measured in the parent rocks and that of 299 suspended matter from the Amazon Basin (Goldstein et al., 1984; Allègre et al., 1996). This suggests for some 300 groundwater a possible influence of sedimentary deposits in the coastal area that originate from the Amazon. The 301 other groundwater from the same area agree with the field of parent rocks, suggesting that Nd originates from the 302 weathering of the bedrock.
- 303

5. Summary and perspectives

We report the dissolved concentrations of major and trace elements, stable isotopes (O and D), strontium (⁸⁷Sr/⁸⁶Sr) and neodymium isotopes (¹⁴³Nd/¹⁴⁴Nd) in ground waters and surface waters in French Guiana. Groundwater samples were collected from (i) shallow drill holes in this coastal area, which is the only densely populated area in French Guiana, and (ii) deeper wells in the basement from which groundwater is pumped from bedrock fractures.

311 Major cations show excess due to water-rock interaction. Comparing δ^{18} O and δ^2 H reveals that most ground 312 waters agree with both local and global meteoric water lines without evaporation impacts. The ⁸⁷Sr/⁸⁶Sr ratios 313 indicate the existence of at least three end-members that corresponds respectively to the drainage of 314 metavolcanic rocks, meta-sedimentary lithology and plutonic granitoid intrusions. The ⁸⁷Sr/⁸⁶Sr and Ca/Na ratios 315 yield evidence of increase in the weathering of metavolcanic rocks with the larger divergence between surficial 316 and deep weathering of about 60%. The $\varepsilon Nd(0)$ in some ground waters reveals a possible influence of 317 sedimentary deposits in the coastal area that originate from the Amazon and on the other hand, some ground 318 waters plot in agreement with the field of parent rocks, suggesting that Nd originates from the weathering of the 319 bedrock. This geochemical and isotopic approach to the groundwater in French Guiana has allowed the origin 320 and complex relationships between the different compartments of the hard-rock aquifers to be more clearly

321 defined.

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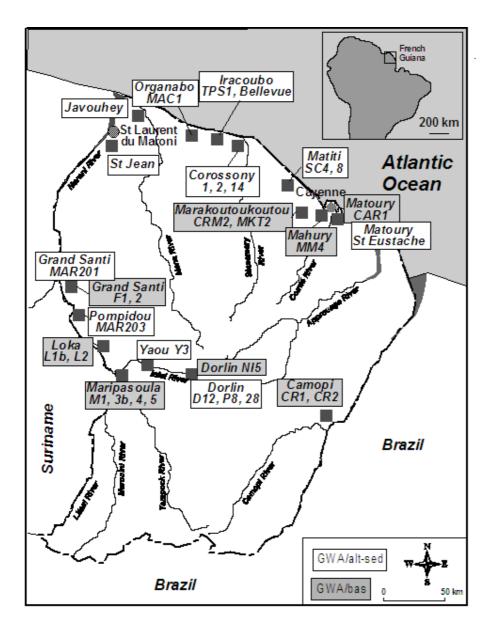
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- 327

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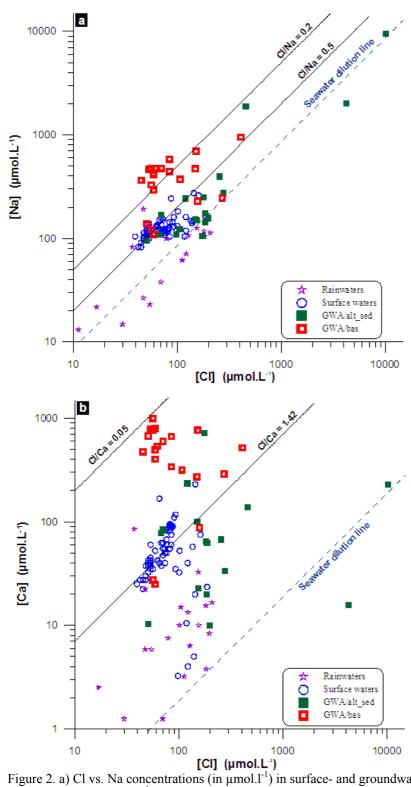
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436 437 438 439 Figure 1. Map showing the site locations in French Guiana where groundwaters were sampled and analysed for

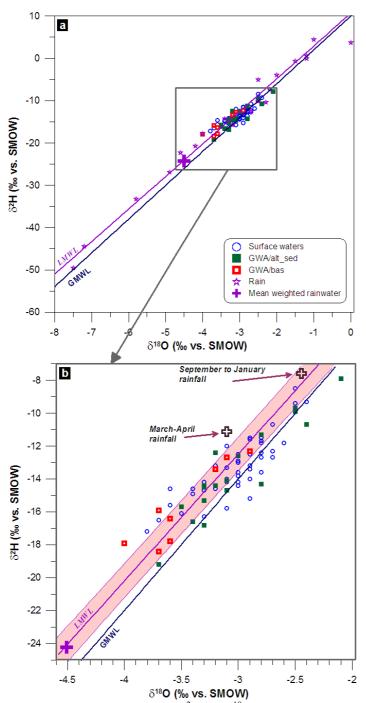
- major ions and isotope systematics.
- 440 Figure 1. Carte de localisation des sites d'échantillonage des eaux souterraines en Guyane pour analyses des 441 ions majeurs et des isotopes.



442 [CI] (μmol.L⁻¹)
 443 Figure 2. a) Cl vs. Na concentrations (in μmol.l⁻¹) in surface- and groundwaters from French Guiana. b) vs. Ca
 444 concentrations (in μmol.l⁻¹) in surface- and groundwaters from French Guiana. Rainwater data are from Négrel
 445 et al. (1997) and surface water data are from Négrel and Lachassagne (2000).

447 Figure 2. a) Concentrations des eaux de surface et souterraines de Guyane en Cl et Na (en μ mol. l^{-1}). b) Cl vs. 448 Ca (en μ mol. l^{-1}) dans les eaux de surface et souterraines de Guyane. Les données des eaux de pluies sont

448 *Ca* (en μ mol. Γ^{1}) dans les eaux de surface et souterraines de Guyane. Les données des eaux de pluies sont extraites de Négrel et al. (1997); celles des eaux de surface sont extraites de Négrel et Lachassagne (2000).



450 451

Figure 3. Relationships between δ^2 H and δ^{18} O (‰ vs. SMOW Standard Mean Ocean Water) in surface- and 452 groundwaters collected from French Guiana (a). Rainwater data, e.g. Local Meteoric Water Line LMWL, i.e. 10 453 monthly accumulations and 3 individual rain events in Cayenne (Négrel et al., 1997) and 6 rain events in the 454 Maroni catchment (Négrel and Lachassagne 2000) are shown together with the Global Meteoric Water Line 455 (GMWL: Craig, 1961); see text for origin of additional data. The big cross indicates the mean weighted rain 456 input. In the extended view (b), the regression line and error bar envelope are indicated for the LMWL.

458 Figure 3. Relations entre $\delta^2 H$ et $\delta^{18}O$ (‰ vs. SMOW Standard Mean Ocean Water) dans les eaux de surface et 459 souterraines de Guyane (a). Les données des pluies, e.g. droite météorique locale (LMWL, i.e. 10 échantillons

460 mensuels intégrés, 3 pluies individuelles à Cayenne (Négrel et al., 1997) et 6 pluies individuelles dans le bassin

461 versant du Maroni (Négrel and Lachassagne 2000) sont illustrées avec la droite météorique mondiale (Global

462 Meteoric Water Line GMWL: Craig, 1961). Les croix sur le schéma indiquent les valeurs moyennes pondérées

463 des pluies. Dans la vue étendue (b), sont indiquées la droitre de régression et les enveloppes d'incertitudes pour

464 la roite météorique locale LMWL.

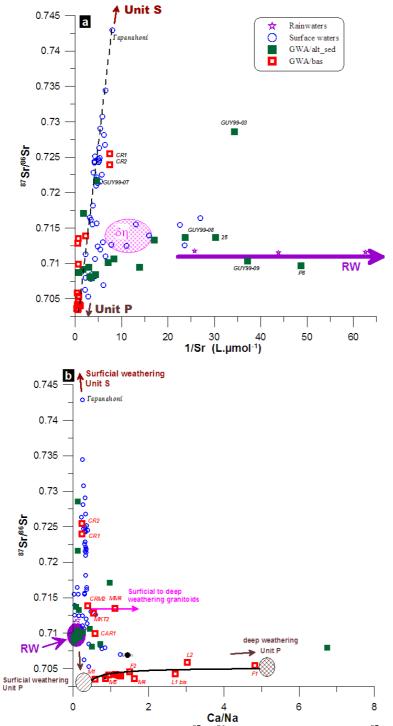




Figure 4. Relationship between a) ⁸⁷Sr/⁸⁶Sr ratios and 1/Sr and b) ⁸⁷Sr/⁸⁶Sr ratios and Ca/Na ratios in surface-467 and groundwaters collected from French Guiana. Rainwater data are from Négrel et al. (1997) and surface water 468 data are from Négrel and Lachassagne (2000). Unit S: schists, micaschists, quartzites, conglomerates, 469 metagrauwackes, metasiltites. Unit P: meta-volcanic rocks (basalt, amphibolite) and rare sediments. $\delta\eta$: gabbro-470 dioritic intrusions, granit and granodiorite.

472 Figure 4. Relation entre a) rapports ⁸⁷Sr/⁸⁶Sr et 1/Sr et b) rapports ⁸⁷Sr/⁸⁶Sr et rapports Ca/Na dans les eaux de 473 surface et souterraines de Guyane. Les données des eaux de pluies sont extraites de Négrel et al. (1997); celles

474 des eaux de surface sont extraites de Négrel et Lachassagne (2000). Unit S: schistes, micaschistes, quartzites,

475 conglomérats, métagrauwackes, métasiltites. Unit P: roches méta-volcaniques (basalte, amphibolite) et rare

476 sédiments. $\delta \eta$: intrusions gabbro-dioritiques, granite et granodiorite.

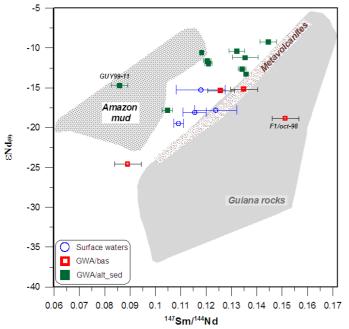




Figure 5. Plot of ɛNd(0) vs. ¹⁴⁷Sm/¹⁴⁴Nd in surface- and groundwaters collected from French Guiana. Bedrock whole rocks ɛNd(0) fields are from Gruau at al. (1985); Delor et al. (2001) and Deckart et al. (2005) and the Amazon suspended matter data are from Allègre et al. (1996).

Figure 5. Diagramme entre $\varepsilon Nd(0)$ vs. ¹⁴⁷Sm/¹⁴⁴Nd dans les eaux de surface et souterraines de Guyane. Les données des roches totales sont extraites de Gruau at al. (1985); Delor et al. (2001) et Deckart et al. (2005). Les données des matières en suspensions de l'Amazone sont extraites d'Allègre et al. (1996).

Name	Туре	Reference	Well type	lithology						
Grand Santi	GWA/alt-sed	MAR201	well in the village (5m)	sand, clays, alluvial deposits						
Pompidou	GWA/alt-sed	MAR203	well in the village (5m)	sand, clays, alluvial deposits						
Grand Santi	GWA/alt-sed	MAR201	well in the village (5m)	sand, clays, alluvial deposits						
Yaou 3	GWA/alt-sed	Y 3	spring from saprolite	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Dorlin14	GWA/alt-sed	D14	spring from saprolite	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Inini	GWA/alt-sed	25	spring	sand, clays, alluvial deposits						
Inini	GWA/alt-sed	P8	shallow drill hole (2m)	sand, clays, alluvial deposits						
St Eustache	GWA/alt-sed	GUY99-03	spring from saprolite	granitoide, migmatites, Caribbean plutonism γ						
Corossony 1	GWA/alt-sed	GUY99-04	piezometer (10m)	sand, clays developped on granitoides Caribbean plutonism γ						
Corossony 2	GWA/alt-sed	GUY99-05	piezometer (10m)	sand, clays developped on granitoides Caribbean plutonism γ						
Corossony 14	GWA/alt-sed	GUY99-06	piezometer (10m)	sand, clays developped on granitoides Caribbean plutonism γ						
TPS1	GWA/alt-sed	GUY99-07	drill hole (20m)	sand, clays developped on granitoides Caribbean plutonism γ						
Bellevue	GWA/alt-sed	GUY99-08	drill hole (20m)	sand, clays developped on granitoides Caribbean plutonism γ						
MAC1	GWA/alt-sed	GUY99-09	drill hole (20m)	sand, clays developped on granitoides Caribbean plutonism γ						
St Jean	GWA/alt-sed	GUY99-10	well in the village (5m)	sand, clays, alluvial deposits						
Javouhey	GWA/alt-sed	GUY99-11	drill hole	sand, clays, littoral deposits						
SC8	GWA/alt-sed	GUY99-12	drill hole (20m)	sand, clays, littoral deposits						
SC4	GWA/alt-sed	GUY99-13	drill hole (20m)	sand, clays, littoral deposits						
Camopi	GWA/bas	CR1	drill hole (30m)	migmatites						
Camopi	GWA/bas	CR1 CR2	drill hole (20m)	migmatites						
Grand Santi	GWA/bas GWA/bas	F1	drill hole (50m)	migmatites						
Grand Santi	GWA/bas GWA/bas	F1 F2	drill hole (60m)	migmatites						
Grand Santi Grand Santi	GWA/bas GWA/bas	F∠ F1/1h	drill hole (50m)	migmatites						
Grand Santi	GWA/bas	F1/11 F1/72h	drill hole (50m)	migmatites						
Grand Santi	GWA/bas GWA/bas	F2/72h	drill hole (60m)	migmatites						
Grand Santi	GWA/bas GWA/bas	F2/720	drill hole (50m)	migmatites						
Grand Santi	GWA/bas	F1 F2	drill hole (60m)	migmatites						
Maripasoula	GWA/bas	г∠ M1	drill hole (70m)	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Maripasoula	GWA/bas	M3 bis	drill hole (65m)	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Maripasoula	GWA/bas	M4	drill hole (60m)	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
	GWA/bas	M5	. ,							
Maripasoula Loka	GWA/bas GWA/bas	L1 bis	drill hole (75m) drill hole (50m)	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
		L1 DIS L2	. ,	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Loka	GWA/bas		drill hole (50m)	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Dorlin	GWA/bas	Ni 5	artesian drill hole (1m3/h)	meta-volcanic rocks (basalt, amphibolite) Lower Paramaca P						
Marakoutoukoutou	GWA/bas	CRM2	drill hole (90m)	granitoide, Caribbean plutonism						
MM4	GWA/bas	GUY99-01	drill hole (100m)	diorites						
CAR1	GWA/bas	GUY99-02	drill hole (80m)	Leptyno-amphibolite lle de Cayenne series						
Marakoutoukoutou	GWA/bas	MKT2	drill hole (100m)	granitoide, Caribbean plutonism						

Table 1. Groundwater typology, well type and lithology in the extensive sandy-argillaceous terrane from the
coastal area and from shallow wells in the alluvia along the Maroni and Oyapock River catchments (GWA/altsed) and from deep wells in the basement (GWA/bas).

Table 1. Typologie des eaux souterraines, type d'ouvrage et lithologie dans la zone argillo-sableuse de la zone
cotière et des aquifères superficiels des bassins versants du Maroni et de l'Oyapock (GWA/alt-sed) et des
ouvrages profonds dans la zone de socle (GWA/bas).

Name	Туре	Reference	EC	т	pН	Ca	Na	Mg	к	CI	SO₄	NO ₃	HCO3	Sr	⁸⁷ Sr/ ⁸⁶ Sr	Eps Nd (0)	147Sm/144Nd	Nd	Sm	8 ² H	δ ¹⁸ Ο
			µS/cm	°C						µmol∕								ng/l	ng/l		
Grand Santi	GWA/alt-sed	MAR201	39.5	28.5	5.8	85	170	21	13	70	2	10	391	0.321	0.708130	-	-	-	-	-15.7	-3.5
Pompidou	GWA/alt-sed	MAR203	46.4	19.7	5.99	65	248	8	21	180	2	31	200	0.142	0.710147	-	-	-	-	-16.8	-3.3
Grand Santi	GWA/alt-sed	MAR201	46.4	27.8	5	100	152	33	49	149	3	119	300	0.571		-	-	-	-	-16.6	-3.4
Yaou 3	GWA/alt-sed	Y 3	58	24.3	6.05	78	109	177	24	67.6	14	dl	505	0.231	0.708453	-	-	-	-	-14.5	-3.3
Dorlin14	GWA/alt-sed	D14	44	23.6	6.94	62	142	116	27	185.9	35	2	205	0.120	0.710646	-	-	-	-	-15.3	-3.3
Inini	GWA/alt-sed	25	40.7	25.6	5.4	10	157	58	10	197	32	6	38	0.033	0.713692	-	-	-	-	-19.2	-3.7
Inini	GWA/alt-sed	P8	21.1	30.7	5.13	5	109	8	13	99	7	3	19	0.021	0.709707	-	-	-	-	-14.3	-2.8
St Eustache	GWA/alt-sed	GUY99-03	39	25.7	4.48	20	175	40	12	185	12	28	-	0.029	0.728571	-12.64	0.1342	123	27.3	-12.4	-3.2
Corossony 1	GWA/alt-sed	GUY99-04	113	33.3	6.19	67	396	86	44	254	9	4	770	0.342	0.709466	-12.00	0.1209	130	26	-10.7	-2.4
Corossony 2	GWA/alt-sed	GUY99-05	67	30.3	5.19	16	2009	49	35	4241	10	dl	210	0.072	0.709461	-10.42	0.1321	65	14.2	-7.9	-2.1
Corossony 14	GWA/alt-sed	GUY99-06	298	29.8	7	228	9468	457	95	10174	70	dl	2680	1.335	0.708714	-11.63	0.1205	134	26.7	-14.4	-3.2
TPS1	GWA/alt-sed	GUY99-07	84	27.7	5.63	34	273	112	58	276	25	dl	410	0.215	0.721651	-13.28	0.1358	297	66.7	-14.7	-3.1
Bellevue	GWA/alt-sed	GUY99-08	27	26.7	4.53	10	123	28	16	106	5	10	150	0.042	0.713712	-11.22	0.1354	39	8.7	-12.6	-3
MAC1	GWA/alt-sed	GUY99-09	23	29.2	4.4	10	95	18	9	51	4	3	70	0.027	0.710370	-17.83	0.1048	96	16.7	-14.1	-3.1
St Jean	GWA/alt-sed	GUY99-10	98	26.2	6.31	235	243	50	104	120	21	dl	740	0.566	0.717093	-				-9.9	-2.5
Javouhey	GWA/alt-sed	GUY99-11	166	28.5	6.56	717	106	46	39	175	3	84	1310	0.284	0.707984	-14.71	0.0858	60	8.6	-11.3	-2.8
SC8	GWA/alt-sed	GUY99-12	281	29.4	6.64	139	1886	280	96	457	14	dl	2160	0.612	0.709211	-10.59	0.1182	752	147	-14.4	-3.3
SC4	GWA/alt-sed	GUY99-13	39	29	5.01	23	150	35	23	154	9	dl	110	0.059	0.713286	-9.25	0.1446	62	14.8	-9.7	-2.5
											-										
Camopi	GWA/bas	CR1		-		28	122	33	33	56	5	18	200	0.134	0.723978			-		-12.3	-2.9
Camopi	GWA/bas	CR2	-	-		25	109	25	26	59	5	19	150	0.134	0.725469	-		-	-	-12.7	-3.1
Grand Santi	GWA/bas	F1	146.9	26.4	6.6	475	365	188	18	45	1	dl	1750	1.027		-	0.1172	4	0.9	-16.4	-3.6
Grand Santi	GWA/bas	F2	198.5	26.2	6.93	783	465	208	21	54	7	dl	2200	1.484	-	-	0.1587	8	2.0	-17.8	-3.6
Grand Santi	GWA/bas	F1/1h	181	27.1	6.88	403	426	200		59	4	dl	1754	0.913	0.704100	-	-			-	-
Grand Santi	GWA/bas	F1/72h	148	26.9	6.38	535	478	254	-	62	6	dl	2197	1.142	0.704200	-		-	-	-	
Grand Santi	GWA/bas	F2/72h	196	26.5	6.83	600	474	225	-	70	7	dl	2295	1.142	0.703985	-		-	-	-	
Grand Santi	GWA/bas	F1	176		7.16	674	140	189	19	50.7	20	dl	1767	1.244	0.705429	-18.82	0.1512	3	0.7		
Grand Santi	GWA/bas	F2	215		6.95	668	445	238	32	84.5	11	2	2231	1.507	0.704584	-	0.1012				
Maripasoula	GWA/bas	M1	210		0.00	341	581	190	23	84.5	13	dl	1539	1.758	0.703520		0.1109	9	1.7	-	-
Maripasoula	GWA/bas	M3 bis				498	410	597	18	59.2	9	dl	2543	2.055	0.704046		0.0807	18	2.4		
Maripasoula	GWA/bas	M4				772	476	498	20	56.3	10	2	2979	3.174	0.703624		0.0676	8	0.9	-	_
Maripasoula	GWA/bas	M5	-	-	-	317	372	377	28	107.0	6	15	1708	1.689	0.703596		0.0850	25	3.5	-	-
Loka	GWA/bas		-	-	-	796	294	288	12	59.2	8	dl	2354	1.975	0.703390	-15.33	0.1257	25	1.6	-	-
Loka	GWA/bas	L1 bis L2	-	-	-	999	294 329	200 310	29	59.2 56.3	11	dl	2354	2.192	0.705883		0.1257	0 19	3.3	-	-
		LZ Ni 5	400	-	6.22	999 290	329 245		29	270.4	87	3		2.192	0.705665	-	0.0430		2.4	-	-
Dorlin	GWA/bas		186	24.9				415					1302			-	0.0430	34		-18.4	-3.7
Marakoutoukoutou	GWA/bas	CRM2	62	-	5.58	88	230	45	19	157	17	64 5	415	0.459	0.713903	-	-	2	-	-12.7	-3.1
MM4	GWA/bas	GUY99-01	275	26.6	7.18	766	693	312	49	151	32		2560	1.507	0.713512	-15.16	0.1348	5	1.1	-17.9	-4
CAR1	GWA/bas	GUY99-02	156	26	6.14	272	471	264	93	149	23	dl	1330	1.666	0.709933	-24.56	0.0890	6	0.8	-15.9	-3.7
Marakoutoukoutou	GWA/bas	MKT2	237	25.6	6.62	518	952	175	82	408	89	2	1885	1.723	0.712861	-	-	-	-	-13.4	-3.2

Table 2. Major field parameters, major and trace elements, and isotopic data (87 Sr/ 86 Sr, δ^{18} O, δ^{2} H and ϵ Nd(0)) in the groundwaters of French Guiana.

Table 2. Paramètres de terrain, éléments majeurs et traces et données isotopiques (${}^{87}Sr/{}^{86}Sr$, $\delta^{18}O$, $\delta^{1}H$ et $\varepsilon Nd(0)$) dans les eaux souterraines de Guyane.

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