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Isotopic characteristics of the Garonne River and its tributaries

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The Garonne is the largest river in the south-west of France, and its drainage basin stretches between the Pyrénées and the Massif Central mountains. Until now, no water stable isotope study has been performed on the whole Garonne river basin which is composed of different geological substrata, and where the water resources are limited during the dry summer period. This study focuses on the Garonne river and its tributaries from the Pyrénées foothill upstream to its confluence with the Lot River downstream. The aim of the study is to determine the origins of the surface waters using their chemical and stable isotopic compositions ($^{18}$O, D and $^{13}$C), to better understand their circulation within the drainage basin and to assess the anthropogenic influences. The Garonne displays a specific $^{18}$O seasonal effect, and keeps its Pyrénéan characteristics until its confluence with the Tarn River. The difference in the dissolved inorganic carbon (DIC) comes mainly from the change in lithology between the Pyrénées and the Massif Central mountains. Agriculture activity is only detected in the small tributaries.

With a mean annual discharge of $630 \text{ m}^3/\text{s}$, the Garonne is the third greatest French river by water volume after the Rhone and the Loire. Without the Dordogne tributary, the Garonne basin covers 10% of the territory of France, i.e. 55 400 km$^2$. The source of the Garonne is in the central Pyrénées and it flows northwards before receiving tributaries from the Massif Central and then turning westwards in the direction of the Atlantic Ocean. Even if the Atlantic rainfalls are dominant, there is a Mediterranean influence in the eastern part of the basin as a result of the Autan wind blowing from the south-east through the Lauragais opening.

The 478 km long Garonne river rises on the Spanish side of the Pyrénées and flows through the limestone of the Tuca Blanc of Pomérol before re-emerging in the Val dera Artiga above the Aran Valley.\textsuperscript{1} It follows the Aran Valley northwards into France, and then receives the Pique tributary (river length 33 km, basin area: 325 km$^2$). Near Mazères de Neste it receives its more westerly tributary, the Neste (75 km, 906 km$^2$). Further downstream as it leaves the Pyrénées foothills, it is joined by the Salat (70 km, 1570 km$^2$). Just before the city of Toulouse it is joined by the last and most important Pyrénéan tributary, the Ariège (150 km, 3450 km$^2$). All these Pyrénéan tributaries have their sources close to the Franco-Spanish border at elevations ranging between 2800 and 3200 m, where some high valleys receive as much as 1500–2000 mm of rain per year. At Toulouse (Portet gauging station), the inter-annual mean (1910–2009) Garonne discharge is around 188 m$^3$/s.

After Toulouse, the main low-altitude tributaries with low discharges come from the Lannemezan plateau on the west. This is also the driest part of the whole Garonne basin with a mean annual rainfall of 600 mm and, thus, the river depends completely on the upstream water. First the Save (137 km, 1105 km$^2$) joins the Garonne at Grenade and then the Gimone (136 km, 827 km$^2$). After the confluence with the Tarn (an easterly tributary) the third Lannemezan tributary is the Gers (176 km, 1190 km$^2$) which joins the Garonne at Layrac and the last is the Baise (180 km, 2910 km$^2$). The total mean discharge of these four rivers does not exceed 30 m$^3$/s. The Massif Central tributaries then bring high discharge and a more constant water level to the Garonne network. The first Massif Central tributary is the Tarn (375 km, 9100 km$^2$), which has its source near Mount Lozère (1699 m) and a discharge of 210 m$^3$/s, including that from the Aveyron (59 m$^3$/s). The Aveyron River itself (292 km, 5170 km$^2$) joins the Tarn just less than 20 km before the Garonne-Tarn junction. The last tributary, the Lot (491 km), has a wide drainage basin (9170 km$^2$), and also has its source in Mount Lozère quite close to that of the Tarn. The Truyère, one of the tributaries of the Lot, flows in a more northerly direction and it receives water from the old volcanic peak of the Plomb du

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Cantal. The Lot joins the Garonne at Aiguillon and brings a mean discharge of 144 m$^3$/s.

The water isotopes ($^{18}$O and D) are useful tools for the study of river basins.$^{2-5}$ Apart from our own work,$^{6-8}$ there have been very few studies of the Garonne Basin. Some studies on Pyrénées thermal springs were, however, published in the 1970s and 1990s.$^{9-11}$

The $\delta^{13}$C value of riverine dissolved inorganic carbon (DIC) is used as a tracer of carbon origins: biogenic CO$_2$ produced by soil organic matter decay and roots respiration, dissolution of carbonate rocks and atmospheric CO$_2$. Contrary to what is found with $^{18}$O and D, carbon isotopic signatures are not conserved from the soils to the river systems because different biogeochemical processes in the river itself, such as photosynthesis/respiration$^{30}$ and CO$_2$ evasion,$^{31,32}$ can affect the $\delta^{13}$C.

In contrast to what is known for other large river basins of the world, no isotopic data on water and carbon exists for the whole Garonne river basin. The main objectives of this study are first to get some basic data on the DIC carbon isotopes and the $\delta^{18}$O of water and for deuterium during the period 1910–2009, whereas the other sampling periods were relatively dry (50 m$^3$/s$^{-1}$ during the period 1910–2009), whereas the other sampling periods were relatively dry (50 m$^3$/s$^{-1}$ and 156 m$^3$/s$^{-1}$, respectively, for September 2004 and March 2007) compared with the inter-annual means (85 m$^3$/s$^{-1}$ and 218 m$^3$/s$^{-1}$, respectively), i.e. 41% and 28% lower, respectively. In addition, all the Garonne $^{18}$O data collected between 2000 and 2007 just after Toulouse were compiled with the view of obtaining precise information about any seasonal effects (see Fig. 4(a)).

**EXPERIMENTAL**

**Field sites**

Seventeen sampling sites were selected over the whole basin (see Fig. 1): 7 along the Garonne River itself, 3 on tributaries from the Pyrénées, i.e. the Neste, Salat and Ariège Rivers, 4 on tributaries coming from the Lannemezan Plateau, i.e. the Save, Gimone, Gers and Baise Rivers, and 3 on tributaries from the Massif Central mountains, i.e. the Tarn, the Aveyron and the Lot Rivers.

In order to assess the seasonal variations, and the influence of local rains, three sampling series were undertaken during February 2004, September 2004 and March 2007. The first of the series had an average monthly discharge (179 m$^3$/s$^{-1}$ at the Portet gauging station on the Garonne River) slightly (14%) lower than the inter-annual February mean (207 m$^3$/s$^{-1}$ during the period 1910–2009), whereas the other sampling periods were relatively dry (50 m$^3$/s$^{-1}$ and 156 m$^3$/s$^{-1}$, respectively, for September 2004 and March 2007) compared with the inter-annual means (85 m$^3$/s$^{-1}$ and 218 m$^3$/s$^{-1}$, respectively), i.e. 41% and 28% lower, respectively. In addition, all the Garonne $^{18}$O data collected between 2000 and 2007 just after Toulouse were compiled with the view of obtaining precise information about any seasonal effects (see Fig. 4(a)).

**Stable isotopes from water**

The river waters were sampled in their middle stream during the low water period, i.e. in February–March for the winter period, and September for the summer period. All quotas of the water were put into capped 10-mL Exetainer vials (Labco Ltd., High Wycombe, UK).

To measure the $^{18}$O values of the water samples, the samples and the internal standards were flushed off-line with a gas mixture of 2% CO$_2$ in helium. The Exetainer vials were then left to equilibrate at either 25.0°C or room temperature for a minimum of 18 h. The analytical precision of the measurements was ±0.15 per mil (%o).

To measure the $^2$H values of the samples, Hokko beads were added to the Exetainer vials containing the water samples and the internal standards before the vials were flushed off-line with a gas mixture of 2% H$_2$ in helium. The Exetainer vials were left to equilibrate at either 25.0°C or room temperature for a minimum of 1.5 h. The analytical precision of the measurements was ±2.0‰.

Analysis of the samples was carried out using a Gasbench + Deltaplus XP isotope ratio mass spectrometer (Thermo Fisher Scientific, Waltham, MA, USA) at the G.G. Hatch Isotope Laboratories, University of Ottawa, Ontario, Canada.

The equipment was calibrated by using two water standards that are traceable to the primary reference standards V-SMOW2 (Vienna-Standard Mean Ocean Water) and V-SLAP2 (Vienna-Standard Light Antarctic Precipitation) distributed by the IAEA, Vienna, Austria. A third traceable water standard was analysed alongside the samples to check the accuracy of the data.

The results are expressed in per mil on the V-SMOW/SLAP scale, for oxygen:

$$\delta^{18}\text{O}_{\text{V-SMOW2}}(\%o) = \left(\frac{^{18}\text{O}_{\text{s}}}{^{16}\text{O}_{\text{s}}} \right)_{\text{s}} \left(\frac{^{18}\text{O}_{\text{standard}}}{^{16}\text{O}_{\text{standard}}} \right) - 1) \times 1000;$$

and for deuterium:

$$\delta^{2}\text{H}_{\text{V-SMOW2}}(\%o) = \left(\frac{^2\text{H}_{\text{s}}}{^1\text{H}_{\text{s}}} \right)_{\text{s}} \left(\frac{^2\text{H}_{\text{standard}}}{^1\text{H}_{\text{standard}}} \right) - 1 \times 1000.$$

For the calculation of the deuterium excess, the equation for the Global Meteoric Water Line defined by Craig was used.$^{33}$

**Carbon stable isotopes**

Water samples were filtered in the field through a 0.45 µm Millipore filter (Millipore, Billerica, MA, USA). The samples were collected in 125 mL polyethylene bottles, carefully sealed taking care that no trapped air remained in contact with the sample, and stored at a temperature between 0 and 5°C prior to the extraction. To extract the DIC,$^{34}$ an aliquot of the sample (10–15 mL, depending of the alkalinity) was acidified inside a vacuum line with concentrated phosphoric acid. The evolved CO$_2$ was purified and trapped with liquid nitrogen in a glass tube. The analyses were run on a 602 VG Optima Mass spectrometer (Isoprime Ltd., Cheddle Hulme, UK) at the Centre de Geochimie de Surface (CCS), Strasbourg, France. The resulting analytical precision of the measurements was ±0.2‰.

The results are reported as $\delta$ values with reference to Vienna Pee Dee Belemnite (VPDB):$^{35}$

$$\delta^{13}\text{C}_{\text{VPDB}}(\%o) = \left(\frac{^{13}\text{C}_{\text{s}}}{^{12}\text{C}_{\text{s}}} \right)_{\text{s}} \left(\frac{^{13}\text{C}_{\text{standard}}}{^{12}\text{C}_{\text{standard}}} \right) - 1) \times 1000.$$
RESULTS AND DISCUSSION

The different water pools

Three different water pools can be seen from the $\delta^{18}$O isotopic characteristics reported in Table 1 and shown in Fig. 2. The first pool corresponds to the higher Pyrénées water. The only glacier area melting in the Garonne Basin can be found for the Neste River, Pic Long (3192 m) and Munia (3133 m), and for the upper Garonne River itself, Perdiguère (3222 m) and Aneto (3404 m). Effectively, the most negative values of $\delta^{18}$O are found for these two rivers, respectively $-10.0\%$ and $-10.25\%$. The other Pyrénéan rivers such as the Salat and Ariège are only fed by neve and snow, and display slightly less negative values ($-9.23$ to $-8.75\%$). This area provides the main inflow of water to the mid-Garonne Valley before the inflow of the second important pool from the Massif Central river group.

The Tarn and Aveyron Rivers nearly double the discharge of the Garonne with a mean input of 207 m$^3$/s. Together with the Lot River, these rivers, which come from moderate
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<th>Table 1. $\delta^{18}$O values for the 17 sampling points over the 3 dates</th>
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**Figure 2.** $\delta^{18}$O values versus conductivity for the 2007 data. The ellipses represent the different water pools: the upper Garonne River and tributaries with more negative isotope values and the smaller conductivity, the Massif Central tributaries (Tarn, Aveyron and Lot) with higher isotopes values and moderate conductivity, and the Lannemezan Plateau tributaries (downstream) with the higher isotopes values and conductivity. The upper stream of these four rivers is on the Garonne line, certainly due by the income of the Neste Canal. The downstream Garonne sites are located at the right end of the line and correspond to the mixing of these different water pools.
altitudes (775 to 1815 m), display lower $\delta^{18}O$ values: $-8.29$ to $-6.6\%o$, with an even less negative value for the Aveyron in September: $-5.7\%o$.

The third water pool is formed by the four rivers coming from the Lannemezan Plateau with a mean altitude of around 600 m. For these rivers the $\delta^{18}O$ values would expected to be in the range of $-7.5$ to $-6.5\%o$ due to the lower altitude origin of the water as measured in the local wells. However, for irrigation purpose, these rivers can be fed by the Neste Canal, bringing Pyrénées water, which displays high altitude heavy isotope depletion with values as low as $-10.3\%o$. Thus, for the upper part of these rivers, the water isotopic signature involves a mixing of these water sources with value ranging between $-8.65$ and $-7.64\%o$. Downstream, the evaporation process and the inflow of local rainfall and groundwater discharge give rise, in general, to less negative $^{18}O$ values: from $-6.97$ to $-5.22\%o$. Even in September 2004, the Neste water contribution could be detected at the end of the Gers River with a value of $-9.64\%o$.

Table 2 gives the deuterium amount and the calculated D excess for all the sampling undertaken, while Fig. 3 reports $\delta^{18}O$ versus $\delta^2H$ only for the March 2007 period. In this figure, the dotted line represents the GWML with a slope of $8\%o$; a proportion of the sampling points from 2007 are above this line, and this is also the case for 2004 (data not shown). It can also be seen in Table 2 that many rivers display a D excess above 10\%o and this could suggest an influence of Mediterranean rainfall inputs characterised by a higher D excess of 14\%o, although none of the eastern rivers such as the Tarn are implicated in this shift. According the IAEA rainfall data over Toulouse, the local water meteoric line (LWML) presents an equation of $y = 7.2x + 5.7$, and with these values all the experimental points are located beneath this line. It thus seems certain that all the rainfall originates from the Pyrénées (Causses) for the Massif Central side.
Atlantic Ocean, and that the Mediterranean inputs are quantitatively negligible.

Krimissa et al.\textsuperscript{11} found a LWML relation of $y = 7.7x + 9.9$ for the East-Pyréenées spring, and only the more eastern samples exhibit a high deuterium excess (14.2\%), thus showing Mediterranean rainfall input.

Most of the samples from the upstream Garonne and the upper Lannemezan rivers are close to the GWML, and this is also the case for the Garonne samples for September 2004, showing that there is no important evaporation process. On the contrary, for the downstream Lannemezan rivers, a shift is clearly seen in Fig. 3, and there are many low D excess values shown in Table 2, suggesting that evaporation has taken place. The calculation of the slopes between upstream and downstream for these four rivers gives effectively lower values with values ranging from 6.7 for the Baise River, to 5.7 for the Gimone River, 5.6 for the Gers River and 3.6 for the Save River. The arrow in Fig. 3 shows this mean lower slope. The evaporation is also confirmed by the water concentration as seen by the high conductivity, and the anthropogenic influence by the high nitrate (28–49 mg/L) and chlorine (30–50 mg/L) levels. The Tarn River is quite close to the GWML line, whereas the Aveyron and Lot Rivers are shifted slightly downwards, also suggesting evaporation. In February 2004, the Aveyron and the Tarn also displayed low D excess values. The consequence is that after the arrival of these Lannemezan and Massif Central rivers, the Garonne loses its altitude characteristics with strong negative $^{18}$O values, and begins to incorporate evaporated water. The specific value found in Lamagistère with a D excess value of 1.78 can be generated by the evaporation and heating of the Nuclear power station at Golfech, working at full capacity during this cold March month.

**Seasonal variations**

In Fig. 4(a) for the Garonne River sampled a little downstream of Toulouse, a slight seasonal effect (around 0.5 units for the mean value) is seen, with the more depleted $^{18}$O values being observed in April–June with the snow melt, and the more enriched values at the end of summer with the local rainfall inputs during the river low water period. In the Pyrénées, the glacier system is too small (5 km$^2$) to present a typical glacial profile such as that for the Rhine River, which displays more negative $^{18}$O values in August. The Pyrénées profile could then be more properly called a snow-firn profile.

Figure 4(b) reports the $^{18}$O variations between the winter low water (February 2004) and the summer low water (September 2004). The solid line represents no variation ($y = x$). The three ellipses correspond to three water pools: 1 – the upper Garonne Basin, ellipse 2 to the Massif Central rivers plus the two Garonne downstream sites: Mas d’Agenais (MA) and Lamagistère, and ellipse 3 to the Lannemezan Plateau rivers.
Dissolved inorganic carbon

The isotopic composition of the DIC was only investigated over two sampling missions: September 2004 and March 2007, and the results of the second campaign have not been completed. All the available data are given in Table 3.

The $\delta^{13}C_{\text{DIC}}$ values show a small range from $\sim 7.4\%$ to $\sim 11.6\%$, with the majority falling between $\sim 9\%$ and $\sim 10.6\%$. The samples with the lightest $\delta^{13}C_{\text{DIC}}$ are those from the Lannemezan Plateau rivers and the Tarn and Lot Rivers. Along the Garonne fluvial continuum (from Valcabrère to Mas d’Agenais), a small decrease in $\delta^{13}C_{\text{DIC}}$ is observed from $\sim 8.9\%$ to $\sim 9.8\%$ in March 2004. This trend is the inverse of that observed for other large river basins, like the Saint Lawrence,\textsuperscript{29} the Danube (Pawelke and Veizer),\textsuperscript{22} the Patagonian rivers,\textsuperscript{14} or the Nyong River (Brunet et al., in press). In these large river basins, a progressive enrichment in $^{13}C$ is generally observed, caused by degassing of CO\textsubscript{2} to the atmosphere. In the case of the Garonne River, this process is not highlighted by the $\delta^{13}C_{\text{DIC}}$ values, probably due to the contribution of the tributaries which supply DIC with more negative $\delta^{13}C$ values.

The differences in the DIC isotopic composition in the different tributaries can be attributed to the regional lithology. In the upper basin, limestones and dolomites are dominant, whereas the lithology of the Lannemezan Plateau rivers is dominated by molasses, consisting of conglomerate, marl and shale, detritical sediments supplied by the erosion of the Pyrénées mountains. For the tributaries draining from the Massif Central Mountains, the lithology is a mixture of carbonate rocks, granites and volcanic rocks. The DIC resulting from carbonate dissolution has a $\delta^{13}C$ on average of about $0\%$ \textsuperscript{37} Dandurand et al.\textsuperscript{38} report carbonate rocks with a $\delta^{13}C$ between $+2.9\%$ and $+3.9\%$ in the Ariège region, between the Pyrenean plateau and the Lannemezan Plateau tributaries, and the Lot, the Tarn and its main tributary, the Aveyron, which have higher $\delta^{13}C_{\text{DIC}}$ due to carbonate rock and a low $\delta^{18}O_{\text{H2O}}$ due to snow melting, and from the Massif Central and the Lannemezan Plateau rivers, there is a general decreasing trend between $\delta^{13}C_{\text{DIC}}$ and $\delta^{18}O_{\text{H2O}}$ showing a progressive mixing of waters from the Pyrénées with a high $\delta^{13}C_{\text{DIC}}$ due to carbonate rock and a low $\delta^{18}O_{\text{H2O}}$ due to snow melting, and from the Massif Central and the Lannemezan Plateau with lower $\delta^{13}C_{\text{DIC}}$ (particularity for the Lannemezan Plateau rivers) due to Miocene sedimentary deposits and higher $\delta^{18}O_{\text{H2O}}$ due to evaporation processes and low altitude rainfall inputs. It can be clearly seen in Fig. 5(a) that the Garonne River at lower stations (Lamagistère and Mas d’Agenais) is a mixing of three main sources: the upper Garonne River and the Pyrenean tributaries, the Lannemezan Plateau tributaries, and the Lot, the Tarn and its main tributary, the Aveyron, which have higher $\delta^{13}C_{\text{DIC}}$ values during September 2004 (very dry month: $-41\%$ compared with the inter-annual monthly mean) than during March 2007. This can be explained by a very important contribution from the ground waters draining important carbonate reservoirs in the upper Tarn and Aveyron drainage basins during the driest period.
Along its fluvial continuum, the Garonne River displays a small decrease in the Baïse Rivers, with higher conductivity and nitrate concentrations, and lower D excess. The refilling of these rivers in the upper basin by the Neste Chenal is also indicated by the most negative δ18O values.

Agriculture activity is mainly detected in the small tributaries on the left side (Gascogne region), from the Save to the Baise Rivers, with higher conductivity and nitrate concentrations, and lower D excess. The refilling of these rivers in the upper basin by the Neste Chenal is also indicated by the most negative δ18O values.

Along its fluvial continuum, the Garonne River displays a small decrease in δ13C, contrary to what is found for other large rivers.

The δ13C values in the Garonne Basin reflect mainly the DIC contribution resulting from dissolution of carbonate rocks, with some regional variations.

CONCLUSIONS

This study should contribute to a better understanding of the carbon and water origins, but also of the carbon and water balance over the whole Garonne Basin. Such a study has also shown the necessity of coupling water and carbon cycles at the scale of a large river basin such as the Garonne.

The main results obtained in this study are:

- The Garonne River keeps its upstream water isotopic characteristic far beyond the city of Toulouse, until its junction with the first Massif Central tributaries, the Tarn and Aveyron Rivers.
- A seasonal effect is found with the most depleted values obtained in March–April due to the snow melting, and lesser ones in August corresponding to the warmer low altitude rainfall.
- Agriculture activity is mainly detected in the small tributaries on the left side (Gascogne region), from the Save to the Baise Rivers, with higher conductivity and nitrate concentrations, and lower D excess. The refilling of these rivers in the upper basin by the Neste Chenal is also indicated by the most negative δ18O values.
- Along its fluvial continuum, the Garonne River displays a small decrease in δ13C, contrary to what is found for other large rivers.
- The δ13C values in the Garonne Basin reflect mainly the DIC contribution resulting from dissolution of carbonate rocks, with some regional variations.

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