HIGH-RESOLUTION (PIXE) ANALYSES OF CARBONATE DEPOSITS IN A ROMAN AQUEDUCT (FRÉJUS, SE FRANCE): IMPLICATIONS FOR THE STUDY OF PALAEOHYDROLOGICAL VARIABILITY AND WATER RESOURCES MANAGEMENT IN SOUTHERN GAUL DURING THE ROMAN PERIOD*

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Urban water was supplied to the Roman city of Forum Julii (Fréjus, southeastern France) for at least 200 years by a 39.4 km long aqueduct, operating in AD 50. Two perennial springs were successively collected, the Foux and the Siagnole, located at the outlet of Triassic and Jurassic karstic reservoirs, respectively. In this study, we performed high-resolution PIXE (Particle-Induced X-ray Emission) measurements of Ca, Si, Fe and Sr concentrations on selected cross-sections of laminated carbonate sampled along the sidewalls, before and after the connection of the two collection channels. Seasonal variations of water composition, suspended sediment load and discharge are recorded by alternating clear and dark layers, and can be traced by their Sr and Fe contents. On the basis of an annual bimodal high-discharge regime for the two karstic reservoirs, the concentration measurements allow the derivation of a chronological record (117.5 years) of water supply and maintenance activities for a part of the operational period of the aqueduct. The water level in the channel was mainly controlled by the extent of carbonate deposition and by dredging, repair and maintenance operations rather than by the past hydrological regime of the two springs.

KEYWORDS: AQUEDUCT, FORUM JULII, CARBONATE DEPOSITS, PIXE ANALYSES, WATER RESOURCE MANAGEMENT, ROMAN PERIOD

INTRODUCTION

Urban water supply to the Gallo-Roman city of Forum Julii (Fréjus, southeastern France) was accomplished by building a 39.4 km long aqueduct collecting the perennial Foux and Siagnole springs, located in the hinterland, north-east of the city. At the decline of Forum Julii, the
aqueduct, devoted to public monuments, baths and fountains, was abandoned and part of its structure was dismantled (i.e., water intakes for irrigation and mills in medieval times, stone recovery). The channel fed by the Siagnole spring in the remotest part of the aqueduct was repaired and partially reconstructed during the 19th century and is still used for water distribution (c. 11 × 10^6 m^3 yr^−1; Gébara 2002a). The discovery in 1988 of a first channel collecting the Foux spring, 24 km north of Forum Julii (Michel et al. 1994) renewed interest in the study of the water resource management ability of the builders. As comparisons with present-day conditions are possible using palaeo-environmental data covering this period (e.g., Jorda and Provansal 1996; Holzhauser 1997; Mann and Jones 2003; Holzhauser et al. 2005; Ollivier et al. 2006), derivation of information on the past hydrological regime of the springs during the period of Roman settlement in southern Gaul was also expected. The reasons for building a two-step water collection system are complex and are attributed to an increasing need for additional water resources, linked to the expansion of Forum Julii and the building of new public baths and fountains during the second half of the first century AD.

A means of answering this question is provided by the study of Ca-carbonate deposits (also called travertine—Fouke et al. 2000; Pentecost 2005) accumulated along the internal sidewalls of the aqueduct channels (Adolphe 1973; Fabre and Fiches 1986; Bonté 1999; Blanc 2000; Guendon and Vaudour 2000; Joseph et al. 2000; Bobée 2002; Dubar 2006; Carlut et al. 2009). These deposits are organized as speleothems (cave deposits) with the succession of alternating clear and dark millimetric to sub-millimetric laminae (Baker et al. 1993; Genty and Quinif 1996; Genty et al. 1997; Frisia et al. 2000). Although the calcification mechanism remains uncertain, some characteristics of laminated-type carbonates, such as organic matter content, colour, porosity or chemical and stable-isotope compositions, are currently used as palaeohydrological proxies, providing estimates of climatic parameters such as temperature and precipitation intensity (Gascoyne 1983; Shopov et al. 1994; Rousseau et al. 1995; Brook et al. 1999; Fairchild et al. 2001; Genty et al. 2001a, 2002, 2003; Polyak and Asmeron 2001; Baker et al. 2002; Burns et al. 2002; Frisia et al. 2003; Niggemann et al. 2003; Tooth and Fairchild 2003; Wang et al. 2005; McDonald et al. 2007; Zhang et al. 2008). In contrast to cave deposits, the clear layers of carbonate deposits in aqueducts are usually related to periods of high water discharge (‘rainy seasons’), whereas the dark layers are rather attributed to low water stages (‘dry seasons’) (Guendon et al. 1994, 2002; Bobée 2002). However, they may not only record seasonal changes in water composition but also fingerprint single hydrological events (flash floods). If cave deposits result from water infiltration through the overlying soils and rocks, roman aqueducts in southeastern France collect water at the outlet of perennial springs and convey regulated flows within underground channels. The Roman builders developed ‘skilful’ technologies to control discharge, including upstream retention tanks, steep channel sections to dissipate flow kinetic energy, culverts or spillways for the release of excess storm water, distribution tanks and hydraulic cement application on the sidewalls (Hodge 1992; Chanson 2008). A common attribute between cave and aqueduct deposits is the occurrence of dark layers enriched in organic matter and clay, oxide–hydroxide minerals or even charcoals (Baker et al. 1997; Ramseyer et al. 1997; Genty et al. 2001b; Perrette et al. 2008; Carlut et al. 2009). These inputs may also record temporary damage due to root penetration or local collapses of the tunnel structure. The water supply must be interrupted to repair, clean and dredge the channels. Therefore, the overall carbonate deposits provide an intricate record that accounts both for natural spring discharge variations and periods of water shortage for maintenance activities.

The aims of this study were: (1) to perform high-resolution (100–200 μm) concentration measurements of selected elements of laminated carbonate deposits of the Roman aqueduct of
Forum Julii; and (2) to discuss the meaning and the reliability of these measurements for hydrological and climatic reconstructions. Two sites were selected. The first one is located in the upstream part of the aqueduct and corresponds to carbonate deposits solely in connection with the remotest (Siagnole) spring. The second one accounts for the first collected proximal spring (Foux) and involves the additional contribution of the Siagnole. Better constraints on the chronology of water collection and on the modalities of water management, using geochemical fingerprinting of water supply by carbonate deposits, are also expected from comparisons between these two locations.

THE PHYSIOGRAPHIC SETTING OF THE FORUM JULII AQUEDUCT

The archaeological setting

From the foundation of the city (in around 50 BC) until the middle of the first century AD, all of the domestic quarters, public baths, fountains and water reservoirs of Forum Julii were fed by local water resources; that is, from collection and storage of runoff and rainwater, and from wells. Excavations and observations carried out during the 19th century provide evidence for an organized sewerage system, completed by numerous cisterns and wells (Gébara 2002b). Towards the middle of the first century AD, major changes took place with the development of public facilities such as monumental fountains (nymphaeum) and baths. Street fountains were built and an additional sewerage system was implemented. This additional demand coincided with the construction of a 24 km long aqueduct, conveying water by gravity from the Foux spring to a collection tank (castellum divisorum), located at the highest position of the city (Fig. 1). However, accurate dating to the precise year is not possible. The construction of the aqueduct most probably took place after AD 20–25, when an ultimate local groundwater collection system was achieved (the ‘Porte d’Orée’ drain; Fig. 1). According to the chronology of building operations in Forum Julii (i.e., the ‘Clos de la Tour’ sewer system and the ‘Moulin à Vent’ nymphaeum; Fig. 1) and to the occurrence of carbonate deposits in pipes and channels, water distribution by the aqueduct was fully operational in AD 50 (Gébara 2002b).

During the following years, a 15.4 km long extension of the former aqueduct to the north was built to collect the Siagnole spring (Guendon et al. 1994, 2002). Several monuments (i.e., the ‘Porte d’Orée’ nymphaeum, the cardo maximus fountain and the ‘Clos Saint Antoine’ baths; Fig. 1) bear witness to a continuous and enhanced water supply operating in AD 70. The dating of a newly excavated fountain (the ‘Mangin’ fountain; Pasqualini et al. 2006) confirms this fact. During the second century AD, a major thermal establishment was constructed (the ‘Porte d’Orée’ baths; Fig. 1), benefiting from water distribution in Forum Julii (Gébara 2002b). Pipes were installed in streets (the ‘Clos de la Tour’; Fig. 1) and repairs were carried out for two aqueduct bridges and one of the public baths. The final interruption of the water supply by the aqueduct may only be estimated, as being between AD 250, when the repairs were carried out, and c. AD 350, when most of the buildings and monuments located near the ramparts (including the castellum divisorium) were no longer used.

The excavations show that sections of the Forum Julii aqueduct were generally built in covered trenches, with a concrete U-shaped internal channel about 0.70 m wide and 1.30 m deep (caementicium, a hydraulic cement made of a mixture of quicklime, sand and crushed bricks as described by the Roman architect Vitruvius in De architectura, Book 8), surmounted by vertical sidewalls and covered with a vaulted roof (Fig. 1; Michel 2002). A fine finishing layer of waterproof mortar was added to the coarse caementicium. Direct access for maintenance or
Figure 1  (a) A simplified geological map of the hinterland of Fréjus, with the locations of the main sites cited in the text (modified from Gébara et al. 2002b). (b1–b4) Sketches of the main channel types (modified from Michel 2002). (c) A schematic map of the main hydraulic structures, buildings and monuments devoted to water cults of the antique Forum Julii (Fréjus), with the location of the aqueduct (modified from Gébara 2002a).
inspection purposes was realized through vertical shafts. The slope of the aqueduct channel gradually increases from 0.13% (1.3 m km\(^{-1}\)) near the Siagnole spring to 8.20% 10 km downstream (Fig. 1). Between this latter location and the connection with the Foux channel, the slope decreases to 1.52%. The crossing of valleys required the construction of numerous bridges on arches along the course of the aqueduct and tunnels were dug for three different sections (Gébara and Michel 2002).

The geological setting

The Siagnole and Foux springs collect groundwater from contrasting geological formations, the Jurassic karst plateau located between the Audibergue massif and the Artuby valley (Julian and Nicod 1984; Etienne 1987) and the Triassic formations of the Fayence–Callian depression (Michel et al. 1994; Gébara et al. 2002), respectively. The Roman aqueduct crosses three main geological formations (Fig. 1), which are, from north to south: (1) the Provençal terrace, between the Siagnole spring and the Saint-Cassien lake; (2) the Carboniferous formations of the Reyran valley; and (3) the Permian formation of the Bas-Argens. The Provençal terrace is composed of Jurassic limestones and dolomites in its northern part and of Triassic limestones, dolomites, marls, clays and gypsum (Keuper formation) in its central part. The Permian metamorphic series are characterized by volcanic (rhyolite) and meta-sedimentary (pelites, sandstones, tuffs and conglomerates) formations. The major lithological contrast is the boundary between the Jurassic–Triassic limestones and the Carboniferous–Permian rocks (Bordet and Mennessier 1966; Toutin-Morin and Crévola 1994). The antique city of Forum Julii and its harbour were set up on Pliocene and Quaternary alluvial deposits overlying Permian bedrocks. Therefore, the carbonate deposits in the pipes and channels of the city are not linked to local groundwater collection and transport. Their occurrence indicates water collection from the karstic springs of the hinterland.

The hydrological setting

The present-day rainfall and Siagnole river discharge data at Mons have been downloaded from the MétéoFrance (http://www.meteofrance.fr) and DIREN (Direction Régionale de l’Environnement, http://www.hydro.eaufrance.fr) websites. Average monthly precipitation and discharge records have been calculated using the two databases (1998–2007, 220 measurements each). Although the precipitation record is not directly linked to the discharge of the Siagnole spring, which drains a large watershed, its temporal variation should provide a first-order estimate of seasonal outflow for the springs located in the region. The transport of water to Forum Julii takes place within covered channels or underground galleries (Fig. 1), and water temperature change is not the major process controlling carbonate deposition. Only a limited increase, from 12°C at the spring outlet to 12.5°C 500 m below (the Roche-Taillée site; Fig. 1), has been measured in July 2003 in the covered section of the modern channel.

MATERIALS AND METHODS

Sample collection

Chemical analyses were carried out on carbonate deposits sampled, before and after the connection of the Siagnole and the Foux channels, at the Vallon de la Route site (site 6, Fig. 1) and the Les Adrets site (site 7, Fig. 1), respectively. The former site is located 500 m before the confluent.
ence. Only carbonate deposits are preserved. They now crop out at ground level (Fig. 2), with a maximum thickness of 12.2 cm and a major unconformity at 4.6 cm from the sidewall.

Les Adrets (also called Pra Bousquet) is located in the Reyran valley, c. 8 km below the Siagnole–Foux connection (Fig. 1). The sample, collected on the upper part of the sidewall, has a total thickness of 19.3 cm (Fig. 2) and displays remarkable regressive layers (at 4.1–6.0 cm and 9.6–12.5 cm from the sidewall) and transgressive layers (at 6.0–9.6 cm and 12.5–19.3 cm from the sidewall), with no major unconformity. The first deposits only linked to water supply by the Foux spring are missing for the studied section, but have been described for the Boson and Jaumin sites (Fig. 1; Guendon et al. 2002), with which cross-correlations could be made (see discussion). There is also evidence that part of the deposits has been removed by dredging the sidewall accumulations, but this operation was carried out above the studied section (Fig. 2).

Spectral analyses of hydrological data

Spectral analyses of precipitation and discharge data were carried out with the Arand software package (Howell 2001), following the method of Blackman and Tukey (1958). The rainfall database was sampled at constant 1 month intervals and linearly de-trended before analysis. The procedure consists of computing the autocorrelation matrix of the data and in applying a Fourier transform (80% confidence interval, number of lags calculated by dividing the number of values by 3, 0% pre-whitening).

High-resolution PIXE analyses of carbonate deposits

All measurements were carried out along linear profiles oriented in the growth axis of the carbonate laminae, using the AGLAE accelerator facility (Accélérateur Grand Louvre Analyse Elémentaire—Calligaro et al. 2004; Salomon et al. 2008) of the C2RMF (Centre de Recherche et de Restauration des Musées de France) in Paris. Particle-induced X-ray emission (PIXE) analyses were performed on polished sections to reduce beam deviations due to surface roughness. X-ray emissions were measured using two Si (Li) detectors (Calligaro et al. 2000). The
distance between two measuring spots was set at 100 μm for the Vallon de la Route sample and 200 μm for the Les Adrets sample. Raw spectral data were recorded in ‘counts per second’ converted to concentration units—that is, ppm (parts per million equivalent to mg kg⁻¹)—using the GUPIX software package (Maxwell et al. 1995).

RESULTS

Present-day precipitation and discharge data

The distribution of monthly precipitation at Mons and monthly discharge of the Siagnole between 1989 and 2007 (19-year record) is displayed in Figure 3. The data are characterized by high inter-annual variability, with a mean annual precipitation (±1σ) of 925 ±269 mm yr⁻¹ and a mean annual discharge (± 1σ) of 478 ± 209 mm yr⁻¹.

Most years display two distinguishable ‘rainy seasons’, although the values bear high standard variations (high inter-annual variability). The two rainfall maxima averaged (± 1σ) 92.7 ± 64.6 mm and 132.0 ± 90.7 mm, for April and October, respectively (Fig. 3). The ‘dry seasons’ are characterized by rainfall minima in February (44.0 ± 42.5 mm) and July (36.8 ± 30.9 mm). The average discharge values of the Siagnole river are (± 1σ) 30.8 ± 26.2 mm and 3.4 ± 6.7 mm for March and August (the ‘dry seasons’), respectively, and rise to 47.1 ± 37.2 mm and 84.4 ± 77.4 mm for May and November (the ‘rainy seasons’), respectively. Spectral analyses of rainfall and discharge data provided prominent 6 and 12 month periods, consistent with a bimodal seasonal frequency (Fig. 3). Due to recent urbanization and enhanced groundwater collection in the Fayence–Callian region, the Foux spring only delivers intermittent and very low outflows. We assumed that its past and present-day discharge distributions were equivalent to that of the present-day Siagnole spring. This assumption is supported by studies that reveal comparable climate conditions between the present-day and Gallo-Roman periods (for references, see the Introduction), although alternating low and high flood regimes connected to land degradation processes have been reported along the Rhône valley (i.e., Salvador et al. 2002; Van der Leeuw and ARCHAEOMEDES research team, 2005).

PIXE analyses of carbonate deposits

The composition of the carbonate deposits is inherited both from the chemical composition of each spring and from the occurrence of detrital components trapped within the carbonate deposits. The Foux spring provides gypsum-saturated groundwater, whereas the Siagnole spring involves diluted Ca-bicarbonate waters (Guendon et al. 1994). Carbonate layers with higher average Sr concentrations highlight major contributions of the Foux spring. Their incorporation and/or exchange with flowing water during carbonate crystallization have been extensively described for other travertine deposits (e.g., Gascoyne 1983; Tesoriero and Pankow 1996; Verheyden et al. 2000; Huang and Fairchild 2001; Huang et al. 2001). The distribution of the Ca, Si, Fe and Sr concentrations is displayed in Figure 4 for Vallon de la Route and in Figure 5 for Les Adrets (average concentrations in Table 1).

The average PIXE concentrations are consistent with previous measurements for both sites (Guendon et al. 1994). The carbonate deposits of the Vallon de la Route are composed of alternating clear and dark laminated deposits, with no significant change in thickness (Fig. 6). Within a representative section, the thickness averaged (±1σ) 310 ± 200 μm and 220 ± 130 μm for clear and dark laminae, respectively (262 ± 167 μm for all laminae, 175 measurements).
Figure 3  (a) A 19-year record (1989–2007) of the precipitation at Mons and the water discharge of the Siagnole and (b) average monthly discharges and precipitations (standard deviation is ± 1σ; dark arrows outline years characterized by two marked rainy seasons). (c) A plot of spectral density versus frequency (or period) for the monthly precipitation at Mons; and (d) a plot of spectral density versus frequency (or period) for the monthly discharge of the Siagnole (CI refers to confidence interval).
No clear relationship links Ca, the major constituent of carbonate deposits, and other elements. However, several Ca minima are phased with high Fe, Sr and Si concentration peaks (Fig. 4). These relative drops correspond to more porous carbonate layers, either accumulating solid suspended matter or incorporating higher dissolved loads during calcification. Fe and Si behave in a similar way but not systematically, and opposite trends are also displayed for Sr and Si (Fig. 7). A given clear and porous lamina is enriched in Sr with respect to the thin and dark laminae located on both sides, but the extent of the chemical discrimination is variable along the section. The difference between dark and clear laminae is best highlighted by the ratio of the Sr to Fe concentrations (Fig. 8).

In contrast to the Vallon de la Route, the thickness of the laminae at Les Adrets increases thoroughly from the sidewall to the centre of the channel (Fig. 6). The first laminae have a low thickness (550 ± 340 μm, 28 measurements, 2.0–3.5 cm from the sidewall) with respect to the ultimate laminae (1760 ± 760 μm, 37 measurements 13.0–19.3 cm from the sidewall). These differences in size also correspond to changes in Sr content, which increases from 228 ± 41 ppm...
(2.0–3.5 cm, 75 measurements; Fig. 6) to 561 ± 86 ppm for the last 6.3 cm (13.0–19.3 cm, 323 measurements; Fig. 6). The former average concentration, although higher, approaches the value obtained for the Vallon de la Route (104 ± 32 ppm; Table 1), reflecting a major water supply by the Siagnole spring. In contrast to Sr, the Si and Fe concentrations do not display any increasing trend along the studied section (Fig. 5). The difference between dark and clear laminae is outlined by changes in Si and Fe concentrations, as for Vallon de la Route, but the average concentrations are significantly higher; that is, 8767 ± 1642 ppm versus 4179 ± 1357 ppm for Si (Table 1). Although carbonate deposition at Les Adrets involves collection of waters supplied by the two springs, the thick and clear laminae still provide higher Sr/Fe than the thin and dark laminae, as shown for Vallon de la Route (Fig. 8).

Figure 5  Plots of Ca, Fe, Si and Sr concentrations and Sr/Fe versus distance from the sidewall measured by PIXE for the Les Adrets carbonate deposits. Sr concentrations are also reported for the Boson (empty squares) and Jaumin (black squares) carbonate deposits. The spot resolution is 30 μm, with 200 μm intervals between each measurement. A sample photograph is displayed above the plots (Rg, regressive layers; Tr, transgressive layers).
Table 1  Average concentrations for selected carbonate deposits

<table>
<thead>
<tr>
<th>Element</th>
<th>Vallon de la Route, PIXE (this study)</th>
<th>Vallon de la Route (Guendon et al. 1994)</th>
<th>Les Adrets, PIXE (this study)</th>
<th>Boson (Guendon et al. 1994)</th>
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<tr>
<td></td>
<td>Mean*</td>
<td>σ*</td>
<td>Mean</td>
<td>σ*</td>
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<tr>
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<td>41 917</td>
<td>391 678</td>
<td>4 150</td>
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<tr>
<td>Si</td>
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<td>Fe</td>
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<td>428</td>
<td>752</td>
<td>153</td>
</tr>
<tr>
<td>Sr</td>
<td>104</td>
<td>32</td>
<td>104</td>
<td>8</td>
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</table>

*Means and standard deviations of 1223 measurements for the Vallon de la Route site and 973 measurements for the Les Adrets site.

Figure 6  Plots of lamina thickness and Sr concentration versus distance from the sidewall for (a) a selected section of the Vallon de la Route carbonate deposits and (b) the carbonate deposits from Les Adrets.
DISCUSSION AND INTERPRETATION

The hydrological significance of laminated carbonate deposits of aqueducts

Deposition of Ca carbonates in the channel of the Forum Julii aqueduct is controlled by degassing of surfacing CO$_2$-rich groundwater, originating from two large karstic systems with contrasting geological bedrocks. Excess water with respect to the maximum capacity of the aqueduct was routed to the rivers by the collection systems (Gébara and Michel 2002). Any physical, chemical and biological process that enhances CO$_2$ degassing in the aqueduct—that is, slope breakdown or increases in sidewall rugosity, temperature and/or biological activity—will favour carbonate deposition (Stumm and Morgan 1996). Over time, this process will also reduce water velocity and ultimately discharge in the collection system by increasing friction with the sidewalls. There is evidence that the Roman engineers were aware of the problem, as shown by the application along the sidewalls of a fine-grained hydraulic cement to reduce turbulence and allow easier removal of the carbonate deposits. However, changes in the aqueduct slope—in particular, before the connection with the Foux channel (0.13–8.20%)—indicate that the role of turbulence and degassing processes was not fully understood. Discharge calculations were based on reference sections of the aqueduct channel (Bonnin 1984) rather than on the mathematical product of velocity and wet section area. The direct link between carbonate deposition and water discharge is, however, puzzling. Deposition models based on a thin stagnant water layer at the surface of travertine predict an increasing rate, with a decreasing dissolved CO$_2$ concentration and an increasing layer thickness (Dreybolt 1988; Dreybolt and Buchmann 1991). The process is thoroughly enhanced by turbulence in the overlying water column. Therefore, an increase in the
deposition rate is expected with an increase in the discharge, and vice versa. However, evidence for positive as well as for negative links between carbonate accumulation and water discharge has been reported for locations where water evaporates or substrates lead to erosion of the previous deposits (Kano et al. 1999; Drysdale et al. 2003; Ihlenfield et al. 2003). The carbonates accumulated in the aqueduct channel only display small ‘erosion marks’ in their bottom parts (Guendon et al. 2002), and the balance between carbonate deposition and post-depositional erosion may only partly explain the accumulation pattern. As spring water collection was regulated, the progressive increase in thickness of the laminae after the connection of the Foux and Siagnole channels (Fig. 6) reflects additional water collection but does not mean major changes in the past hydrological regimes of the karstic reservoirs. The best explanation is that the accumulation of carbonates progressively modified the sidewall roughness and enhanced turbulence. Transgressive layers tend to reflect the rise of the water level due to a progressive reduction of the channel section by carbonate deposits rather than an increase of spring outflow (Figs 2 and 5). Using height measurements of carbonate deposits along sidewalls, characteristic friction coefficients (Bailhache 1979) and slope measurements, Guendon et al. (2002) could determine internal wet sections, from which water discharge estimates could be derived (180–250 l s$^{-1}$ and

Figure 8 Plots of Sr/Fe versus carbonate length for selected sections of (a) the Vallon de la Route carbonate deposits and (b) the Les Adrets carbonate deposits. The grey and white bands correspond to the dark and clear laminae, respectively.
60–280 l s\(^{-1}\), for the Foux and the Siagnole, respectively). When the water collection system was fully operational, the total discharge exceeded 500 l s\(^{-1}\) beyond the confluence, for a maximum capacity of 477–822 l s\(^{-1}\). Water supply by the remote Siagnole spring was thoroughly reduced or interrupted several times during the following years, either for maintenance or repair activities. Several regressive layers account for these operations (Figs 2 and 5) and some of the high Fe and Si concentration peaks (Figs 4 and 5) may fingerprint local damage to the tunnel structure. These interpretations are confirmed by the rupturing and abandonment of a primitive conduct at La Roche Taillée (site 3, Fig. 1). Towards the end of the collection period, the total supply to *Forum Julii* decreased to 240–170 l s\(^{-1}\), with a major contribution from the Foux, shown by the persistently high Sr concentrations.

The PIXE analyses revealed that carbonate laminae might be described by their chemical composition; in particular, by Sr/Fe ratios. Trace elements such as Sr enable discrimination between the types of laminae, as reported for other modern and archaeological carbonates (e.g., Carlut et al. 2009). For a unique water supply (Vallon de la Route), the dark and compact laminae are enriched in Fe and Si with respect to the clear and porous laminae due to transport of detrital particles and their incorporation within carbonate deposits. Such conditions occur either during low water stages by settling of suspended matter that is usually exported further downstream in the aqueduct channel or following periods of enhanced infiltration of soil water linked to damage in the tunnel structure. The dark laminae were formed during these periods. When low precipitation conditions prevail in the watershed—that is, during the ‘dry seasons’ (winter and summer)—the water levels within the karst reservoirs are low. They are only refilled by enhanced water infiltration during the ‘rainy seasons’ (spring and autumn) or after seasonal snow-melting in the watershed. Pre-existing water may be flushed out during these periods (e.g., Lastennet 1994; Baker et al. 1997; Plagnes and Bakalowicz 2002). The geochemical composition of spring waters is, however, controlled by complex types of hydrological behaviour (Ford and Williams 2007) and varying fractions of the rainfall input will contribute to either slow or quick outflow generation (e.g., Pinault et al. 2001; Aquilina et al. 2003). High dissolved load fluxes are expected when outflow involves the saturated zones of karstic reservoirs and displaces large amounts of pre-existing water. In contrast, dilution by rainwater after major storms or flash floods should provide diluted waters (Dreiss 1989; Petelet-Giraud and Negrel 2007; Maréchal et al. 2008). Clear and porous laminae with a high Sr content (and a low Fe content) were deposited during periods of flush flow, most probably at the beginning of the rainy seasons. In contrast, a rapid transfer of rainwater will supply spring water with a lower mineralization status (a lower Sr content), due to more limited contact with the bedrocks of the karst reservoirs. This situation should correspond to the deposition of intermediate clear–dark layers. On the whole, the Sr/Fe ratios (Fig. 8) provide a first-order fingerprint of clear and dark laminae that covers a large range of hydrological situations. The mixing of saturated groundwater collected by the Foux spring with diluted Siagnole water at Les Adrets (Guendon et al. 1994) should control the composition and the thickness of the laminae (Fig. 6). Higher Sr concentrations (with respect to Vallon de la Route) fingerprint a higher water contribution of the Foux spring end-member in the mix. In contrast, a high contribution of the Siagnole end-member lowers the Sr concentration. This process is superimposed with the one due to seasonal discharge variations on the basis of synchronous behaviour for the two karstic systems.

**The derivation of a water supply record from the carbonate deposits of Forum Julii**

Assuming a synchronous bimodal high discharge regime for the two springs, one hydrological year should correspond to two ‘rainy’ and two ‘dry’ periods. On the basis of high concentrations
for high discharges (the clear laminae of the rainy periods) and low concentrations for low discharges (the dark laminae of the dry periods), two maxima and two minima should occur every year. A total of 470 maxima and minima could thus be extracted from the overall data, providing 117.5 years (470/4) of carbonate deposition, equivalent to 1.64 mm yr$^{-1}$ with respect to the total length of the sample (193/117.5 mm yr$^{-1}$; Fig. 5). Nearly equivalent results could be derived from Sr/Fe peak counts (114 years).

Because the first laminae are missing for Les Adrets, the carbonate deposits should not provide a continuous record for the entire period of water supply to Forum Julii. A plot of Sr concentration versus distance from the sidewall allows comparison with other sites (Boson and Jaumin; Figs 1 and 5). Except for the first laminae, the overall compositions follow the same trend towards the centre of the aqueduct channel and are linked to the relative contribution of the two springs, fingerprinted by variable Sr concentration levels. The use of the palaeo-discharge model of Guendon et al. (2002) strengthens this approach. Several periods, labelled P1–P6 in Figure 9, have been distinguished. During the first period (P1), water was solely supplied by the Foux spring, as shown by high Sr concentrations in the carbonate deposits (559 ± 8 and 533 ± 6 ppm for Boson and Jaumin, respectively; Fig. 5 and Guendon et al. 2002). Additional collection of Siagnole water thoroughly increased the discharge from about 2501 s$^{-1}$ to 5301 s$^{-1}$ during the second period (P2) and matched, c. 25 years later, the extensive use of water for monuments in Forum Julii. Deposition of the first laminae was initiated during this period of mixed water supply. The increasing Sr content of the carbonate deposits (from 118 to 506 ppm; Fig. 5) of the third period (P3) reflects an erratic and progressively decreasing contribution of the Siagnole. The end of this increasing trend may be placed at the limit with the following period (P4), when water collection from the Siagnole spring was interrupted. The P4 boundaries are highlighted by two Sr
minima, at 118 ppm and 494 ppm, respectively (Fig. 9), between which a relatively constant high Sr concentration (563 ± 72 ppm) reveals a unique contribution of Foux spring water. Intermittent water collection from the Siagnole proceeded during the following period (P5), as shown by highly variable Sr concentrations for the last carbonate layers. According to our interpretation, the last period (P6) is not represented, most probably because carbonate deposits were dredged or collapsed within the channel. Because the P4 period can be positioned within the Sr concentration curve, 117.5 years of carbonate deposition may be placed on the palaeo-discharge diagram, on the basis of two Sr maxima and two Sr minima per year (Fig. 9). The scaled carbonate record provides a period of water supply spanning from +41 years (c. ad 91) to +155 years (c. ad 209), with respect to the onset of water distribution in Forum Julii (c. ad 50; Gébara 2002b). Relating these periods to the structural characteristics of carbonate deposits supports this chronological record. The first set of regressive–transgressive layers, linked to water-level variations, provides evidence for repeated maintenance operations during the third period (P3), related either to damage caused by excess water discharge or to warping by carbonate deposits (Guendon et al. 2002). Traces of repair and channel dredging have been spotted in the carbonate deposits at Jaumin (+54 years), Esquine (+69 years) and in the Reyran valley (+77 years) using the same counting technique. The second set of regressive–transgressive layers observed during the fourth period (P4, +119 to +139 years) is linked to the collapse of the channel structure during this period (Roche-Taillée site; Fig. 1; also Guendon et al. 2002). Water supply to Forum Julii by the aqueduct lasted for several more years (+181 to +218 years), as fingerprinted by carbonate deposits of other sites (Guendon et al. 2002). Although the corresponding carbonate deposits were not recovered at Les Adrets, excavations performed in Forum Julii support a reduced but persistent water supply until c. ad 350 (Gébara 2002b).

CONCLUDING REMARKS

In order to determine whether or not carbonate deposits of antique aqueducts are suitable for palaeohydrological studies, high-resolution (100–200 μm) PIXE measurements of Ca, Si, Sr and Fe concentrations were carried out on carbonate deposits of the Roman aqueduct of Forum Julii (Fréjus, southeastern France), which collected water from two perennial springs. The thick and clear laminae of the carbonate layers are enriched in Sr and depleted in Fe and Si with respect to the thin and dark laminae. The former tend to characterize high discharges and high dissolved loads that correspond to long periods of contact between groundwater and carbonate rocks of the geological basements and flush flows from the karstic reservoirs during the ‘rainy seasons’. The latter tend to reflect low water stages (with low dissolved loads) typical of ‘dry seasons’ and/or enhanced detrital inputs due to damage in the channel structure. The overall geochemical record of alternating dark and clear laminae covers a large range of hydrological situations that involves periods of water breakdown due to maintenance or repair activities. Using a simple assumption on the past hydrological regime of the karstic reservoirs, the Sr concentration measurements indicate that the carbonate sequences may provide a chronological record of collection and mixing of water originating from the two springs. Derivation of a time scale was also possible, but only for a part of the water supply period to Forum Julii (117.5 years), due to the absence, removal or collapse in the aqueduct channel of the first and last deposits. Therefore, the information that can be derived from aqueduct carbonate deposits must be considered with great care and may not be easily converted to efficient palaeohydrological proxies as in the case of cave

1 According to Gébara (2002b), the building of the aqueduct took place in ad 20–25 and it was operational from ad 50.

deposits. The extension of the former Forum Julii aqueduct connecting the proximal Foux spring to the remote Siagnole spring most likely corresponded to a concomitant expansion of the antique city, as revealed by the succession of water monuments, fountains, thermal baths, water pipes and sewerage networks operating between the middle of the first century AD and the end of the third century AD.

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