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# **The environmental legacy of historic Pb-Zn-Ag-Au mining in river basins of the southern edge of the Massif Central (France)**

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## **Abstract**

The main rivers (Aude, Orb, Hérault) that discharge into the Gulf of Lions and the west bank tributaries of the Rhône River including the Gardon have former non-ferrous metal mines in their upper drainage basin. Using unpublished data and data from the literature, this study provides an integrated overview of the contamination of water and sediment along the continent-sea continuum and of its impacts on the biota and on human health. In the upper part of these basins, water and stream sediments are enriched in metal(-oids) compared to median European concentrations. Arsenic is the main contaminant in the rivers Aude and Gardon d'Anduze, Sb in the Orb and Gardon d'Alès, Tl in the Hérault river. A rapid reduction in dissolved and particulate concentrations was systematically observed along the river due to dilution and precipitation.

The high concentrations of metal(-oid)s observed suggest that former mining activity still represents a potential threat for the environment, but the lack of high temporal resolution monitoring, especially during Mediterranean floods, prevents accurate assessment of metal fluxes from these rivers to the Mediterranean Sea. Studies dedicated to impacts on human

health are too rare, given that studies have shown a higher rate of arsenic specific cancer near Salsigne mine in the Aude River basin, and cases of saturnism in children in the upper Hérault River basin. These studies underline the need to take environmental-health issues into consideration not only in these watersheds but around the entire Mediterranean basin, which harbors numerous metalliferous ores that have been mined for millennia.

**Keywords:** metals; metalloids; historical mining legacy; Mediterranean continent-sea continuum

## 1. Introduction

Mining of metalliferous mineral deposits has left large quantities of sulfide-rich tailings, heaps of slags and polluted soils. These wastes are often responsible for metal(-loid) pollution of water and soils that can persist for decades or even centuries after the mines close (Hudson-Edwards et al., 2011) and have harmful effects on fauna and on human health (Bindler et al., 2009; Grattan et al., 2003; Monna et al., 2011).

Acid mine drainage (AMD), which contains high concentrations of dissolved metal(-loids) is released when sulfidic wastes present in the tailings come into contact with oxygenated water. Precipitation of Fe and Al oxides and neutralization of AMD generally takes place downstream from mine sites, resulting in the transfer of the metal(-loids) to the particulate phase (Berger et al., 1988; Casiot et al., 2003; Chapman et al., 1983; Egal et al., 2010). In rivers, metal(-loids) are transported in dissolved form, or more often in particulate form (Macklin et al., 2006 and references therein).

Rivers can transport mining contaminants great distances from their source regions. For example metal(-loid) contamination of the Tinto and Odiel Rivers that drain the former mines in the Iberian Pyrite Belt is transferred by marine currents to the Mediterranean Sea through the Strait of Gibraltar (Elbaz-Poulichet et al., 2001). Cd originating from a former Zn refining plant in the Lot River basin (France) is transported along the Garonne-Gironde river-estuary system (Audry et al., 2004; Elbaz-Poulichet et al., 1987) and then by coastal currents along the Atlantic coast, where contamination of oysters has been reported more than 400 km from the source (Baudrimont et al., 2005; Boutier et al., 2000). In the Danube River, the dispersal of sediment-associated metals occurs hundreds of kilometers downstream from past or present mines, resulting in the transfer of contaminants across international borders (Bird et al., 2010).

Mine contamination persists long after mining activity has ceased (Byrne et al., 2012; Johnson and Hallberg, 2005). For example, in France the imprint of Gallic (~ca. 300 B.C. to ca. 20 A.D.) and medieval (~ca. 1000-1300 A.D.) mining activity is still visible in peat bog sediments and soils (Baron et al., 2006, 2005). In Jordan, plants, animals and sediments in the vicinity of one of the main copper mining and smelting centers of the Nabatean, Roman and Byzantine periods are still heavily contaminated (Pyatt et al., 2000). In the UK, in France and elsewhere, metal released during the peak mining period in the 19<sup>th</sup> and early 20<sup>th</sup> centuries is still a source of pollution for the rivers draining the former orefields (Hudson-Edwards et al., 1997; Macklin et al., 1997; Resongles et al., 2014).

Polymetallic ores for the production of Ag, Au, Cu, Fe, Pb, Sb, Zn.etc., have been heavily worked in Mediterranean regions since pre-Roman times. Metal ore exploitation was particularly intensive in the 19<sup>th</sup>–20<sup>th</sup> centuries in the northwestern Mediterranean basin where large amounts of wastes were generated. Although most of the mines are now abandoned, they still present a threat to the environment and human health that is poorly assessed in France and elsewhere in the Mediterranean basin today (Doumas et al., 2016)

In southern France, the border of the Massif Central has been an important mining area of non-ferrous (Pb-Zn-Ag-Au) metals since at least the medieval period and probably even earlier (Roman era). Mining activity intensified beginning at the end of the 19<sup>th</sup> century, then declined in the second half of the 20<sup>th</sup> century. The vast majority of metal mines closed in the 1960s except the Salsigne gold mine, which closed in 2004.

The major streams discharging into the Mediterranean Sea (the Aude, the Orb, the Herault rivers) and the right bank tributaries of the Rhone River such as the Gardon River, have former metal(-loid) mining sites in their watershed. In the last 20 years, a substantial number of studies highlighting the metal contamination levels and impacts on different environmental compartments of these river basins, have been published by researchers from different

scientific communities (geochemists, microbiologists, plant biologists, epidemiologists). However, the results are dispersed in the journals of these different communities. What is more, each publication deals with a single basin and often with a single system (water, sediments, soils, biota or humans). In this context, it is difficult to have an integrated overview of the environmental impacts of the intensive past mining activity in these river basins.

The aim of this study was to inventory published data on metal(-loid) contamination caused by former non-ferrous mining activity in different environmental reservoirs (water, sediments) of the Orb, Aude, Hérault and Gardon river basins and its impacts on biota and on human health. Implementation of the EU Water and Marine Strategy Framework Directives requires a good knowledge of the sources of metal pollution and their fluxes to rivers and to the sea. This study was also carried out to see if the available data would enable the assessment of metal(-loid) fluxes transported by mine impacted rivers to the Gulf of Lions.

To this end, data published in peer-reviewed journals on waters, sediments, biota and health were compiled. Unpublished data on water and sediments from the Hydrosocieties Laboratory (Montpellier, France) were also included. The elements considered are Zn, As, Cd, Sb, Tl and Pb, which were either exploited as metals of economic interest (Zn, Pb) or present as by-products (As, Cd, Sb, Tl) in the ores.

## **2. River basins studied**

The main geographic and hydrologic features of the basins studied are summarized in Table 1. Details on major metal(-loid)s mines on these basins are listed in Table 2. More data on former mines can be found in the French Geological Survey database (<http://sigminesfrance.brgm.fr/>).

### *The Aude river basin*

The Aude river basin covers an area of 4,840 km<sup>2</sup> (Fig.1). The estimated mean annual discharge at the river mouth is 50 m<sup>3</sup>s<sup>-1</sup> but marked seasonal and inter-annual variations are observed related to the Mediterranean climate, which alternates between long periods of drought and devastating rainfall events like the one that occurred in the Aude watershed in 1999. During this event, the discharge of the Aude river 100 km from its mouth reached 3,000 m<sup>3</sup>s<sup>-1</sup> at peak discharge (Gaume et al., 2004).

The former Salsigne gold mine was located in the watershed of the Orbiel River, a tributary of the Aude River. The mine, which was the biggest gold mine in Europe, closed in 2004 after 100 years of operation during which 100 t of gold were produced. Between 1873 and 1910, the mine also produced 25% of the world's As.

After the mine closed, the tailings were confined using waterproof geo-membranes or phyto-stabilized. An AMD treatment plant was set up in the framework of the European project Difpolmine (EU Life 02 ENV/F000291 demonstration Project, 2002-2005).

### *The Orb river basin*

The Orb river basin (Fig.1) covers an area of 1,585 km<sup>2</sup>. The river has a mean discharge of 26 m<sup>3</sup>s<sup>-1</sup> at its mouth. Its hydrological regime is characterized by severe low water stages, as low as 3 m<sup>3</sup>s<sup>-1</sup> in summer, and brief but disastrous flood events mainly in autumn when water discharges can reach 1,100 m<sup>3</sup>s<sup>-1</sup>. Several dams have been built on the Orb River for the production of electricity and/or for irrigation. The biggest, the Mont d'Orb lake (or Avene lake) dam, was built in 1964 in the upper valley as a reservoir for irrigation of the lower valley. The dam is also used to produce electricity and incidentally the artificial lake, which controls 8% of surface area of the the Orb watershed, acts as a flood clipper dam.

The watershed harbors coal ores that were exploited until the 1980s along with former metal-(loid) mines (Fig.1). The Bournac Sb mine produced 600 t of Sb between 1908 and 1920. The mine tailings are drained by a small creek that flows into Sebestrières Creek, a tributary of the Mont d'Orb Lake. A former Pb-Zn-Ag mine (the Rabasse mine) is also present in the Orb watershed. Exploitation of the mine began in the Roman era. The mine closed in 1954, leaving behind former ore treatment facilities and tailing heaps containing As, Pb and Zn, that were partially embedded in the Mont d'Orb lake.

#### *The Hérault river basin*

The Hérault River drains a watershed of 2,500 km<sup>2</sup>. Its mean yearly water discharge at the mouth is 43.7 m<sup>3</sup>s<sup>-1</sup>. Notable deviations from this mean are observed. During summer, the mean water discharge is 7.5 m<sup>3</sup>s<sup>-1</sup> but during Mediterranean rainfall events, extreme values of up to 2,800 m<sup>3</sup>s<sup>-1</sup> have been reported (Bournet et al., 2005). Located in the upper Hérault watershed (Fig. 1), the former Malines Pb-Zn-Ag mine was already exploited in ancient times and in the Middle Ages. Modern production was mainly of Pb and Zn, but stopped in 1991. The mine left tailings stored behind a dam. The AMD generated by the oxygenation of the sulfides contained in the tailings are treated by addition of lime. However the presence of pyrite sand in the effluent at the base of the tailings was observed during field surveys. Ore treatment has also left soils near the Avinière Zn mine polluted by As, Pb, Zn and Cd. In 2006, the soil was removed from several gardens located on the left bank of the Vis River and stored in the tailings stock of the former Malines mine.

#### *The Gardon river basin*

The Gardon River (Fig.1) is a tributary of the Rhone River. It is 144 km long and drains an area of 2,014 km<sup>2</sup>. The hydrological functioning of the river is characterized by a severe low

water stage ( $1.7 \text{ m}^3\text{s}^{-1}$ ) mainly during summer, and extreme flood events like the one in September 2002 when discharge probably reached  $6,600 \text{ m}^3\text{s}^{-1}$  (Banque Hydro webpage, 2017; Neppel, 2004).

Coal was intensely exploited in the basin until 1985. Concerning metals, Ag was already being extracted in the upper watershed in the Roman era. In the Middle Age, the ores were exploited for Ag and Pb. Intensive modern production started in the middle of the 19<sup>th</sup> century but declined after 1960. During this period, about 4 Mt of pyrite, 85,000 t of Zn, 50,000 t of Pb and 2,570 t of Sb were produced in the Gardon watershed, leaving several million tons of wastes close to the ore extraction and processing sites (Basol webpage, 2017).

### **3. Material and methods**

Both published and unpublished data were used in this study. Details on the method used for sample treatment and Zn, As, Cd, Sb, Tl and Pb determination are provided in Supplementary Material S1. Sampling sites for unpublished data are shown in Fig.1.

In the Aude basin, stream water and sediment samples were collected between 2002 and 2011. The sampling frequency ranged from one to four depending on the site and on the type of sample (water or sediment). In the Orb basin, water and suspended particulate matter (SPM) were sampled during a single survey in 2004. In the Hérault basin, between March 2004 and April 2005, stream water and sediments were sampled in four to nine surveys depending on the site.

Trace metal-(loids) are natural components of the Earth's crust where their natural abundance ranks in the following order: Zn, Pb, As, Tl, Sb, Cd (Taylor and McLennan, 1985). They are released into water through erosion and weathering. In order to minimize the variations in concentrations related to their different natural abundance, and also to assess the degree of contamination compared to a reference, median concentrations were normalized to

FOREGS (FOREGS Geochemical Baseline Database) median concentrations in European stream water and stream sediment (Table S5) (De Vos et al., 2006).

## 4. Results

### 4.1. Water contamination

Mean and median dissolved concentrations of metal(-oids) in the surface waters of the rivers basins studied are listed in Table S1 (Aude), S2 (Orb), S3 (Herauld), S4 (Gardon).

Median concentrations normalized to FOREGS median concentrations in European streams are presented in Fig. 2.

In the Aude River watershed, dissolved metal concentrations in mine drainage exceeded FOREGS average values by a factor of 22 (Sb) to 21 300 (As). Upstream from the mining area in the Orbiel River (site O0), normalized concentrations ranged between 1.5 (Cd, Sb) and 11 (As). Downstream from the mining area, concentrations were lower than those of FOREGS by a factor of 0.5 for Zn, similar for Cd and Pb and significantly higher by a factor of 3.5 for Sb, 17 for Tl and 35 for As. In the Aude River at site A0 located downstream from the junction with the Orbiel River, a significant enrichment factor (factor of 6) was observed for As whereas other elements showed no (Zn) or moderate (Cd, Sb, Tl, Pb) enrichments.

In the Orb watershed in the vicinity of the former Bournac Sb mine at site S0 located upstream from the tailings stock, most elements except Tl and Pb were in excess by a factor of 3.4 (Zn) to 43 (Sb) compared to FOREGS concentrations. Downstream from the tailings stock, enrichments of all elements increased and reached values ranging from 3.1 (Tl) and 232 (Sb) at site S0.8. In the Orb River at site O0 located upstream from the Mont d'Orb lake, all elements were depleted compared to FOREGS median values except Tl, which was slightly enriched by a factor of 2. In the Mont d'Orb lake, only Pb values did not exceed the FOREGS

values. At site O3-a located near the ore processing plant of the former Rabasse mine, enrichments comprised between 2.1 (Tl) and 9.4 (Sb) were observed. On the opposite bank of the lake (site O3-b), enrichments were generally lower. At the exit of the lake (site O5) and in the Orb River downstream (site O8) persisting enrichments were detected for Cd (4), As (7) and Sb (7.5)

In the Hérault watershed, drainage water from the Malines mines tailings stock (sites C0-a, b, c and d) displayed high enrichments (up to 6,552 for Tl, 935 for Zn, 305 for Cd, 43 for Pb and Sb) compared to FOREGS values, whereas no significant enrichment in As was detected. In the Crenze River (site C1, C2), which receives the mine drainage water, normalized concentrations ranged between 3.01 (As) and 3,485 (Tl) pointing to serious Tl contamination (concentrations reached  $17.4 \mu\text{g.l}^{-1}$ ). Although lower, the normalized concentrations of the other elements were still high, ranging from 4 to 380 for Cd and Zn. In the Vis River (Fig.2), normalized concentrations (comprised between 3.15 for Sb and 32.5 for Tl) were higher at site V1 than at site V0 except for As, reflecting the inputs from the Crenze River. Only As was relatively depleted (normalized concentration=0.52). The confluence with the Vis River also resulted in an increase in the concentrations of most elements in the Hérault River. Immediately downstream from the confluence with the Vis River (site H0.185), normalized concentrations in the Hérault River were around 5 for all elements except Tl (16). These results, which show an increase in the concentrations of Pb in the Hérault River downstream from the confluence with the Vis River, confirm those obtained by Petelet et al., (1998). Further downstream, normalized concentrations of Zn, Pb and Tl decreased along the Hérault River to reach 1.9 for Zn and Pb and 6.7 for Tl at the most downstream sites, probably as a result of dilution and/or adsorption onto particles. The concentrations of other elements did not vary significantly.

In the Gardon watershed, median concentrations of metals and metalloids in the AMD of the former Carnoules mine exceeded those of the FOREGS by a factor of 726 (Sb) to 227,200 (As) in the source of Reigous Creek (site R0) located at the base of the tailings stock. Along Reigous Creek, the concentrations of As, Sb and Tl decreased by a factor of 1000 (As), 48 (Sb) and 30 (Tl). The decrease in the concentrations of the other elements (Zn, Cd and Pb) was less (factor of 2). In the Amous River upstream from the junction with Reigous Creek (site A0), element studied were enriched by a factor of 2.2 (Sb), 3.2 (Pb), 8 (Cd), 15 (Tl) and 18 (Zn) which reflects an enriched geochemical background. At site A3, downstream from the confluence with Reigous Creek, median FOREGS values increase for all element with enrichment ranging from 9.1 (Sb) to 161 (Tl). This shows that despite the strong dilution and precipitation processes that occur during the mixing of the two rivers (Egal et al., 2010), the water of the Amous river is still highly polluted by effluents from the former Carnoules mine.

Along the Gardon d'Ales (GAL), the median concentrations of Cd, Tl and Sb exceeded those of the FOREGS by a factor of more than 3. The highest enrichment in Sb (up to 161 times the FOREGS value) was measured in the upper part of the river (site GAL7.7) during the low water stage, reflecting the impact of former Sb mines as stated by Resongles et al., (2015). Higher normalized concentrations of Tl and Cd were observed downstream from site GAL20, pointing to a source in the lower basin near the town of Ales.

Along the Gardon d'Anduze (GAN), median normalized concentrations were generally lower than those in the GAL except for As which was enriched by a factor of up to 11 compared to FOREGS values. During the low water stage, in addition to As, only Sb was enriched by a factor of 4.2 in the lower basin. During the high water stage, enrichments of Cd and Tl also occur in the lower basin probably linked to the former Pb-Zn mines located in the vicinity of Ales (Fig.1).

In the Gardon River (G), downstream from the confluence of the two GAN and GAL Rivers, Zn and Pb concentrations were similar to or slightly higher (by a factor of 2) than the FOREGS concentrations. Cd, As, Sb and Tl (the latter in the high water stage) were higher than FOREGS values. The higher concentrations of Cd, Sb and Tl in the G river compared to the GAN indicate that the main source of these elements is the GAL sub-basin, whereas the main source of As is the GAN sub-basin, (probably the former Carnoules Pb-Zn mine).

#### 4.2. Sediment contamination

Concentrations in the suspended particulate matter (SPM) or in the sediments of the basins studied here are listed in Table S6 (Orb), S7 (Herauld), S8 (Gardon). Differences in the concentrations normalized to FOREGS values as a function of the distance they travelled in the rivers are presented in Fig. 3.

In the Orb watershed, SPM displayed the highest enrichments (from 2.8 for Tl to 5130 for As) compared to FOREGS values in Sebestrières Creek at site S0.4 downstream from the former Sb mine at Bournac. In the Mont d'Orb lake, SPM normalized concentrations were either depleted (Sb) compared to average FOREGS values or similar (Tl) to moderately (factor 3.4 to 12) enriched (Cd, As, Sb) compared to average FOREGS values. The maximum enrichment (12 for As) in the vicinity of the former ore processing site (site O3-a) on the left bank of the lake although high reflected a marked reduction in SPM contamination in the lake compared to that in Sebestrières Creek. Downstream from Mont d'Orb lake (site O5), Sb, As and Cd were enriched, with maximum normalized concentrations ranging from 4.1 (Pb) to 16 (As). Tl and Sb normalized concentrations were similar to FOREGS values.

In the upper Hérault basin, the highest enrichments (from 10 for As to 908 for Zn) were observed in the of mine drainage sediments at the base of the tailing stock (sites C0-a to C0-d). Although lower, the concentrations in the sediments of the Crenze River downstream are

still significantly polluted with enrichments ranging from 7.3 for As and 120 for Cd at site V1.5, upstream from the confluence with the Vis River. Upstream from the confluence with the Crenze River (site V0) the Vis River showed no or only moderate enrichments (from 1 for As to 6.8 for Cd). The junction with the Crenze River resulted in an increase in metal(-loid) enrichments of between 7.3 (As) and 120 (Cd) at site V1.5 located just upstream from the junction of the Vis and the Herault rivers. The Herault River upstream from the mine district is characterized by relatively moderate enrichments (from 2.2 to 6.5 for As and Cd respectively). The confluence with the Vis River increased enrichments of the Herault River sediment in most studied metals with the exception of As. Particular enrichment affected Zn, Cd and Pb, which reached respectively, 12, 10 and 8 at site H1.85. From station H40 on, enrichments decreased. At site H67 (67 km from the Malines mine district) enrichments ranged from 1.7 (Tl) to 5 (Cd).

In the Gardon watershed, the concentrations in stream sediment reported by Resongles et al., 2015, reflect metal(-loid) enrichments in the Gardon d'Ales with normalized concentrations ranging from 2 (Cd) to 74 (Sb). In the Gardon d'Anduze, except for As, lower normalized concentrations (from 0.92 for Cd to 4.7 for Sb) than those measured in the Gardon d'Ales were observed. Downstream from the confluence of the two Gardon Rivers (site G0), normalized Zn, Cd, Sb, Tl and Pb concentrations were higher than in the Gardon d'Anduze, suggesting that the main source of these elements is the Gardon d'Ales. On the other hand, the Gardon d'Anduze is the main source of both particulate and dissolved As, probably linked to the former Carnoules mine.

## **5. Discussion**

### **5.1. Appraisal**

All the concentrations of trace elements measured revealed significant multi-contamination of both the dissolved and particulate phase of the water in the river basins studied. As contamination predominated in the Aude and Gardon d'Anduze basins, whereas in the Hérault basin, Tl and Cd or Zn contamination predominated in the dissolved and sediment phases, respectively. In the Orb basin, the water was mainly contaminated by Sb whereas the SPM were enriched in As compared to FOREGS values. In the Gardon d'Ales, Sb predominated in the SPM and in the water, except near the confluence with the Gardon d'Anduze (site GAL37.5) where Tl became the most enriched element in the water. Interestingly although Zn and Pb ores were exploited both in the Malines mining district and in Carnoules mine, the main contaminants in the Hérault and Gardon Rivers draining these two mines differed, pointing to site specificities.

Downstream from mining areas, contamination of surface water decreased due to natural attenuation processes such as dilution, or the transfer of elements from the dissolved to the particulate phase due to adsorption or (co)-precipitation. For example, in Reigous Creek (Gardon d'Anduze) the decrease in dissolved Tl concentrations was attributed to dilution (Casiot et al., 2011) whereas both dilution and precipitation with Fe-oxides were proposed for As (Casiot et al., 2003; Egal et al., 2010) and Sb (Resongles et al., 2013). Particle sedimentation in lakes such as the Mont d'Orb lake, or in water reservoirs in the Hérault basin, also probably contributed to the observed reduction in contamination in the particulate phase downstream.

## 5.2. Impact on biota

Literature on the impacts of this contamination on wild life in our study area is rare. Following the remediation of the former gold mining site of Salsigne (from 1996 to 2006), a study was carried out to assess the effects of remediation actions on As transfers from soils to small mammals (Drouhot et al., 2014). For this purpose, wood mice (*Apodemus sylvaticus*), the Algerian mice (*Mus spretus*), common voles (*Microtus arvalis*), and the greater white-toothed shrews (*Crocidura russula*) were collected from zones that had undergone three different levels of remediation treatment, and from a control site. As concentrations were measured in the soil and in the animals' stomach contents, livers, kidneys, and lungs.

The results of that study revealed no correlations between soil contamination and As accumulation. In fact, despite the decrease in arsenic concentrations in the remediated soils, both wood mice and Algerian mice were subject to higher oral exposure to As in remediated zones than in the control area, probably because factors such as the habitat and diet preferences of the animals or their mobility play a significant role in As transfers. However, the study did show a weak relationship between the mass of liver/lung/kidney and As levels, suggesting possible histological alteration due to As exposure.

In the Gardon basin, the accumulation of metal(-loids) in fish (*Leuciscus cephalus*) was studied by Casiot et al., (2009) in the Amous River upstream and downstream from its confluence with Reigous Creek. As, Pb and Tl were shown to bioaccumulate in liver tissue (As, Pb) or otoliths (Tl). Bioaccumulation of metals or As was not detected in muscle tissue.

## 5.3. Impact on human health

A study of cancer mortality in the period 1968 to 1994 was carried around in the Aude basin in the vicinity of the former Salsigne gold mine by (Dondon et al., 2005). For that study, the authors used an exposure zone comprising 260 km<sup>2</sup> including all the communes

surrounding the former mining site of la Combe du Saut (Fig.1). The control area included all communes located within 20 km of Salsigne, excluding those selected in the exposed area and the large cities (Carcassonne, Aussillon and Mazamet). The control area had a total of 39923 inhabitants, and the exposure zone 8207. Cancer mortality observed in the two areas was compared to overall cancer mortality in France. The resulting standardized mortality ratios (SMR) showed significant excess mortality due to all types of cancer (SMR ratio=1.1), lung cancer (SMR ratio 1.8), pharynx cancer (SMR ratio 2.1) in the whole population, and due to digestive system cancer (SMR ratio 1.3) among women compared to that observed in the control area. The results were the same when miners' deaths were excluded. The authors concluded that the excess mortality caused by cancer in the exposure area was probably due to environmental contamination.

In the town of Saint Laurent le Minier (Herauld basin), 41 children aged from 6 months to 17 years and 3 pregnant or breast feeding mothers were screened for Pb poisoning (Cicchelero, 2005). The objective of this study was to assess the prevalence of saturnism and the impact of the pollution on children's health. Among the 41 children, 80% were below  $50 \mu\text{g L}^{-1}$ , the standard threshold of Pb in the blood. The study also revealed Pb concentrations higher than  $50 \mu\text{g L}^{-1}$  in the blood of five children aged from six months to 17 years, two of whom presented Pb concentrations  $>100 \mu\text{g L}^{-1}$ . One of the breast feeding women had a concentration of Pb of  $55 \mu\text{g L}^{-1}$  in her blood.

In the Gardon d'Anduze basin, a survey of blood metal concentrations was also carried out in several communes in 2016. The official results have not yet been made available to the public.

#### 5.4. Contribution to metal(-loid) fluxes to the Mediterranean Sea

Dilution processes, adsorption, precipitation processes and retention in lake and water reservoir water help reduce the metal(-loid) concentrations downstream from former mines, but the pollution is still there, accumulated in fluvial deposits and can be remobilized during flood events (Coulthard and Macklin, 2003; Foulds et al., 2014; Macklin et al., 1997). Table 3 compared the published particulate metal(-loid) fluxes in the rivers we studied with those in the River Rhone. On an annual basis, the fluxes of the Rhone (more than 90%) dominated inputs to the Gulf of Lions by far. During the 2003 flood event, the total particulate fluxes of the Aude, Orb and Herault rivers represented between 8% (Zn) and 10.6% (Pb) of the fluxes in the Rhone River, which also receives water from rivers like the Gardon River that are affected by former mines. However these annual fluxes were calculated based on only one or two sample concentrations measured in the Aude, Orb and Herault Rivers (Radakovitch et al., 2008). This is largely insufficient for flux calculations owing to the hydrological variability in the Mediterranean context, which is characterized by sudden, intense and devastating flood events during which large quantities of metal(-loids) can be mobilized especially in the particulate phase. For example, during a moderate flood of the Gardon River (maximum river discharge  $800\text{m}^3\text{ s}^{-1}$ ), Resongles et al., (2015) calculated particulate fluxes of metal(-loids) of 5,214 kg (Zn), 1,915 (As), 24 kg (Cd), 94 kg (Sb), 38 kg (Tl) and 2,000kg (Pb) in 24 hr.

Nevertheless, the particulate Zn, Cd, Sb and Pb fluxes exported by the Rhone River at Arles (Table 3) suffice to highlight the importance of its western bank tributaries, which drain former mines. The influence of the Gardon River is indirectly reflected by the higher metal (-loid) fluxes observed in 2002 when the contribution of this river to the flood of the Rhone River represented 60% of the Rhone discharge compared to 2001 and 2003 when the flood came originated from the northern tributaries. During the exceptional flood of the Gardon River (discharge reached  $6000\text{ m}^3\text{ s}^{-1}$ ) in September 2002, Ollivier et al., (2006) showed that

the imprint of As and Sb pollution caused by former mining activity in the Gardon basin was visible at the mouth of the Rhone River.

Concerning impacts on the Mediterranean Sea, moderate Pb, Zn and Cd enrichments in nearshore sediments were measured at the mouth of the rivers studied compared to concentrations measured at offshore sites (Radakovitch et al., 2008; Roussiez et al., 2006).

## **6. Conclusion**

The main French coastal rivers discharging into the Mediterranean Sea west of the Rhone River and the right bank tributaries of the Rhone River drain former non-ferrous metal mines in their upper basins.

In these basins, waters, sediments, and suspended matter are significantly enriched in metal(s) or metalloid(s) compared to European median FOREGS concentrations (De Vos et al., 2006). Contamination involves many elements, but the main contaminant differs with the basin. Arsenic is the main contaminant in the waters of the Aude and Gardon d'Ales Rivers, Sb in the Orb and Gardon d'Anduze, Tl in the Hérault. As is also the main contaminant in the particulate phase (SPM or sediments) in the Aude and Gardon d'Anduze, whereas Zn and Cd are the main contaminants of the particulate phase in the Hérault basin.

The results of this study showed that contamination was higher in the upper basins in the vicinity of former mines. Contamination decreased strongly and rapidly along the rivers due to dilution by uncontaminated river inputs. In the dissolved phase, a fraction of the decrease in the concentration was also due to precipitation and to the adsorption of metal(-loids) onto the particulate phase. Dams, which favor sedimentation, may also trap contaminated sediment and hence help reduce contamination of the particulate phase.

The impacts of the contamination on the biota are poorly documented. Published studies on its impacts on human health are also rare, but increased mortality due to arsenic specific cancer around the mine of Salsigne in the Aude basin and two cases of saturnism were reported in children in Saint Laurent le Minier. Further epidemiological studies are needed to better assess the impacts of contamination on populations living in the former mining districts.

Concerning the contribution of mine impacted rivers to the fluxes of metal(-loid)s to the Mediterranean Sea, one of the main conclusions of our study is the lack of data. Data are generally lacking on Mediterranean flood events, which supply the majority of water and sediments to the Mediterranean Sea. Further studies including systematic monitoring of metal(-loid) concentrations and discharges are required to establish more realistic budgets for the metal(-loid) inputs from rivers flowing from the Cevennes mountains to the Mediterranean Sea. Better assessment of these fluxes is needed not only in France but around the whole Mediterranean basin, where many metalliferous ores have been mined, some for millennia. The quantification of these fluxes is particularly important as stricter regulations concerning metal discharges to rivers and coastal areas are implemented. The final objective being to achieve the good ecological status of these water masses.

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## Figure captions

Figure 1: Map of the study area showing the locations of sampling sites. A, B, C, D, E, F are the main mines (see Table 2).

Figure 2: Dissolved metal(-loid) concentrations in the Aude Basin (a), the Orb basin (b), the Herault basin (c), in the Gardon River during the high water stage (HWS) (d) and during the low water stage (LWS) (e) and in Reigous Creek and the Amous River draining the former Pb-Zn Carnoules mine (Gardon d'Anduze basin). Data on concentrations in the Gardon basin are from Resongles et al., (2015); data for Reigous Creek and the Amous river are from the OSU-OREME website: <http://data.oreme.org/observation/pollumine>. Concentrations ( $\mu\text{g L}^{-1}$ ) are normalized to FOREGS median concentrations ( $\mu\text{g L}^{-1}$ ) in European stream water.

Figure 3: Median metal(-loid) concentrations in SPM from the Orb watershed (a), in sediments from Herault watershed (b), in SPM from the Gardon basin (c). Concentrations ( $\mu\text{g g}^{-1}$ ) are normalized to FOREGS median concentrations ( $\mu\text{g g}^{-1}$ ) in European stream sediments.

River basin	Area of drainage basin (km <sup>2</sup> )	River length (km)	Mean river discharge (m <sup>3</sup> s <sup>-1</sup> )	Minimum-Maximum river discharge (66 years)	Maximum instantaneous river discharge
Aude	5327	224	40.6	8-74	1850
Orb	1442	136	26	5.3-38.9	1700
Herauld	2550	147.6	41.9	2.8-25.1	2800 (1)
Gardon	1100	127.3	na	na	6600 (2)

Table 1: Main geographical and hydrological features of the basins and streams or rivers studied. Data are from the *Eau France* database (<http://www.eaufrance.fr/>)

River basin	Major metal(-loid) mines	Position	Main (accessory) metal produced	Metal (T)	Date of mine closure	Wastes	Receiving river body	Remediation
Aude	La Loubatière	2°15'18.35" E 43°24'18.22" N	Pb (Ag, Zn)	48000t (Pb)	1962	565 000t	Le Linon	nd
	Salsigne	2°21'18.27" E 43°20'18.51" N	Au	100t (Au)	1998	6Mt	Orbiel	Partial
Orb	Bournac	3°3'17.52" E 43°47'56.00" N	Sb				Sebestrières	no data
	La Rabasse	3°4'46.00" E 43°46'34.37" N	Pb (Ag, Au, Zn)	4800t (Pb)	1954	790000t	Orb (Mont d'Orb Lake)	Remediated
Herauld	Les Malines	3°37'23.01" E 43°55'27.86" N	Zn (Ag, Ba, Pb)	1Mt (Zn)	1991	13.7Mt	Crenze	Partial
	Les Avinières	3°39'51.62" E 43°55'55.31" N	Zn	40000t (Zn)	na	na	Vis	Partial
Gardon	Carnoullès (A)	4°0'8.01" E 44°7'2.92" N	Pb (Ag)	30000t (Pb)	1963	1.3Mt	Reigous	Planned
	La Croix de Pallière (B)	3°56'22.25 E 44°2'43.61 N	Zn (Ag Cd Ge Pb)	80000t (Zn)	1981	296000t	Ourne, Aigues-Mortes	Under study
	La Felgerette (Collet de Dèze) (C)	3°11'22.50 E 7°9'39.91 N	Sb (As)	2570t (As)	1948	40000	Ravin des Bernes	nd
	Le Soulier (St Martin de Valgalgue) (D)	4°52'46.53 E 61°55'26.26 N	Pyrite (F Pb Zn)	4Mt (pyrite)	1962	1.2Mt	Gardon of Alès, Grableux	nd
	Landas (E)	4°7'15.72" E 44°12'24.08" N	Zn (F pyrite)	4300t (Pyrite)	na	na	Avène	nd
	Saint Jean du Pin (F)	3°49'56.26" E 44°49'56.26" N	Zn (pyrite)	1000t (Zn)	na	na	Alzon	nd

Table 2: Main former metal(-loid) mines in the river basins studied. Wastes= Exploitation+Processing of wastes. Data are from the BRGM database (<http://sigminesfrance.brgm.fr/>). A, B, C, D, E, F see Fig.1

	Zn	As	Cd	Sb	Tl	Pb
<b>Annual Fluxes (1)</b>						
Aude	20.2		0.077			5.5
Orb	11.1		0.039			5.3
Herauld	27.2		0.13			10.8
Rhone	1292		4.5			356
Total (Aude,Orb, Herauld)/Total (Aude, Orb, Herauld, Rhone) rivers (%)	4.3		5.2			5.7
<b>Fluxes during the 2003 floods</b>						
Aude (2)						
2003 flood (8 days)	7.2		0.028			1.9
Orb (2)						
2003 flood (8 days)	4.1		0.014			1.8
Herauld (2)						
2003 flood (8 days)	28.1		0.136			11.1
	39.4		0.178			14.8
Rhone (3)						
2003 flood (12 days)	494		1.75	5.17		140
Total (Aude,Orb, Herauld)/Total (Aude, Orb, Herauld, Rhone) rivers (%)	8		10			11
Gardon (4)						
2013 flood (24h)	5.2	0.204	0.024	0.194	0.038	2.9

Table 3: Particulate metal(-loid) fluxes transported by the Rhone, Aude, Orb, and Herauld Rivers into the Mediterranean Sea. Fluxes for the Gardon river during the 2013 flood were measured at site G0. Data are from (1) Radakovitch et al., (2008), (2) Roussiez et al., (2012), (3) Ollivier et al., (2011), (4) Resongles et al., (2015)

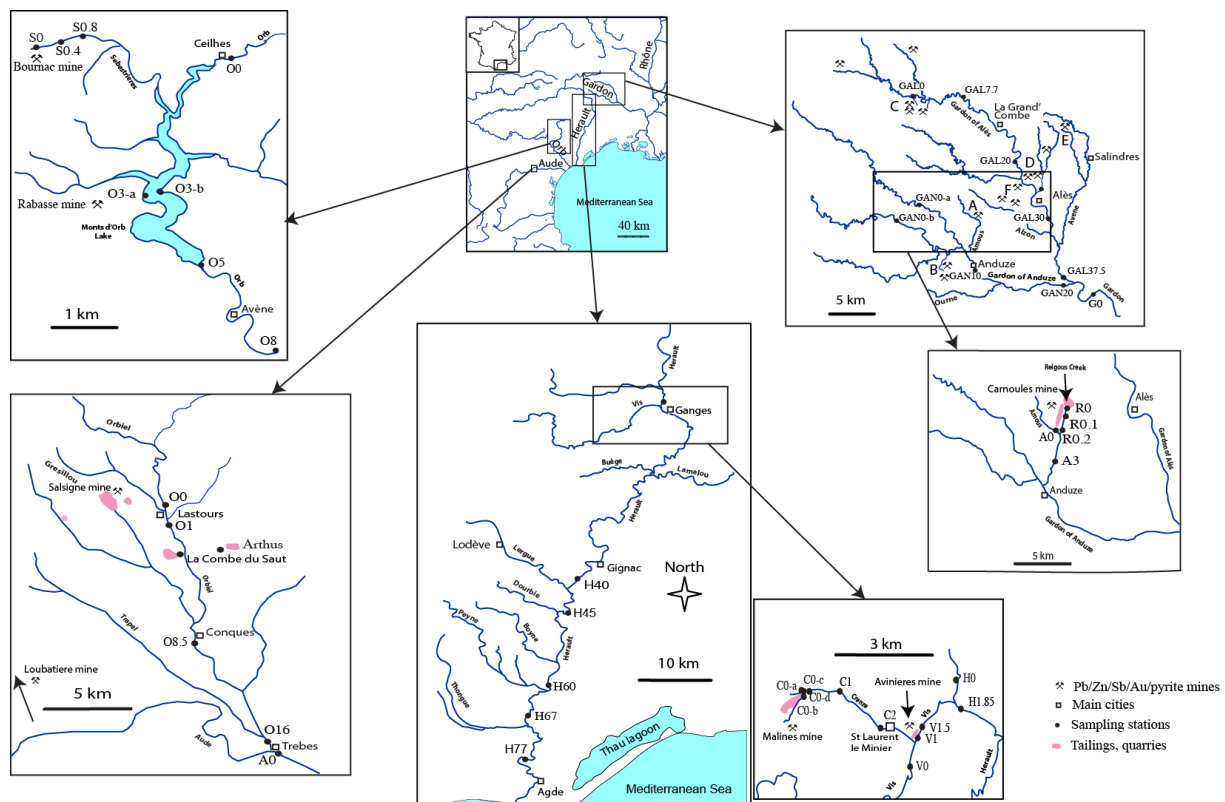


Figure 1

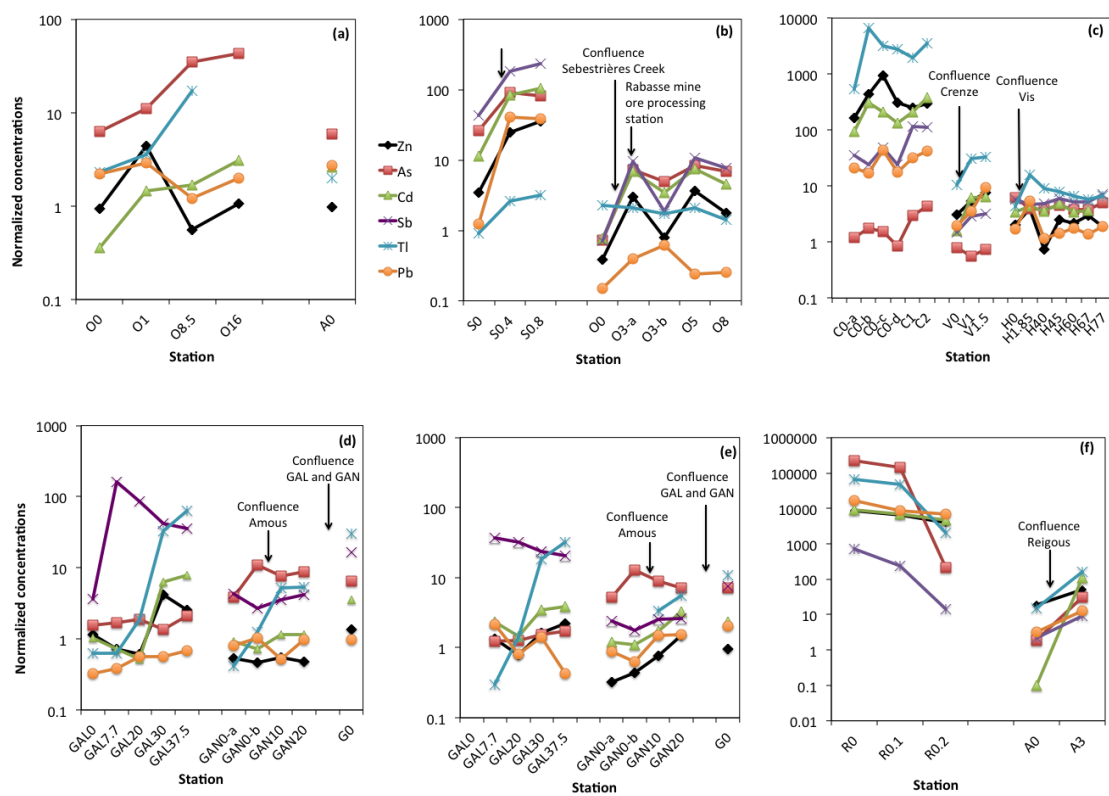


Figure 2

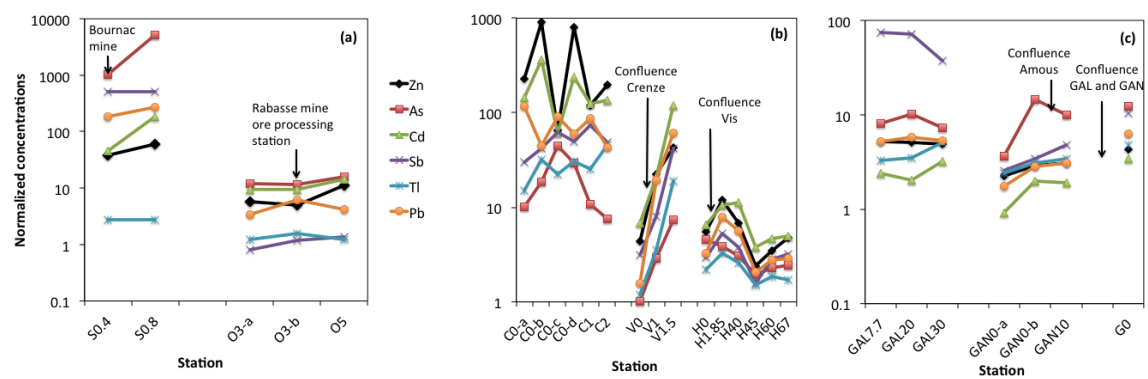


Figure 3