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# An Overview of Priority Pollutants in Selected Coal Mine Discharges in Europe

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**Abstract** Coal mine water discharges in several European countries were investigated as part of the European Commission's MANAGER project. The emphasis of the project was identification of priority pollutants and potential remedial approaches. The main identified priority pollutants were sulphate (all countries) and iron (all countries except Greece). High concentrations of chloride (particularly in Germany and Poland) were associated with discharge of saline mine waters linked to the presence of fossil sea water; these mine waters also had high boron concentrations, in contrast to chloride-rich waters in UK that are linked to recent sea water inflow. Concentrations of trace metals vary among countries, but radium is an important contaminant in barium-rich waters with low sulphate concentrations, essentially in Poland. Concentrations of trace metals and metalloids were generally low because of their relative scarcity in coal strata and adsorption onto ferric oxides and hydroxides, but they still often exceeded the environmental quality thresholds.

**Keywords:** coal basin, pollution, database, emission limits.

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## Introduction

During coal mining, groundwater is pumped in order to lower the water table and permit exploitation. After abandonment and cessation of pumping, the natural rise of the water table inundates or floods the mined voids (Banks et al. 2010; Wolkersdorfer and Bowell 2004). This flooding, also called “rebound”, can create a mine aquifer, as connections between the subsurface and surface waters, often used for drinking water, are re-established (Younger et al. 2002). Also, during mining, the strata are exposed to an oxidizing environment, but the return to pre-mining water table levels restores low-oxygen conditions. Flushing of oxidized minerals can then release contaminants (Kopřiva et al. 2005), which may pollute the local groundwater and surface waters (Gzyl and Banks 2007). Concentrations of dissolved major ions in untreated mine discharge are frequently very high (Cravotta 2008a; Cravotta and Brady 2015).

Contamination of mine water has been studied as part of the European MANAGER project, which was partly funded by the Research Fund for Coal and Steel (RFCS), to assess “Management of mine water discharges to mitigate environmental risks during the post-mining period”. Mitigation of the environmental risks of mine water on aquatic ecosystems involves decreasing the discharge of contaminants. The main aim of the MANAGER project was to develop and implement cost-effective, sustainable, and innovative mine water treatment technologies to mitigate environmental risks after mine closure. This paper presents the results of the first part of the project, which focused on identifying mine water contaminants that cause the greatest environmental risks.

The main goals of this stage of the project were:

- a) to gather information about mine water contaminants in various European coalfields in a coherent and comprehensive database;
- b) to select mine water contaminants that pose risks to aquatic ecosystems, taking into consideration local conditions and the objectives of the Water Framework Directive.

The study included numerous European coalfields from the 6 countries participating in the MANAGER project, i.e. France, Poland, Germany, Spain, Greece, and the United Kingdom (UK) (Fig. 1). A total of 97 representative coal mine waters were investigated and more than 26,000 analysis or physicochemical measurements were gathered.

**Figure 1 will be placed near here during the printing process**

## Review of Selected Coal Mine Waters in Europe

### FRANCE

INERIS, the French national institute for risk assessment, prepared a water composition database for the French coalfields, using data supplied by “Charbonnages de France” (the former French coal authority), GEODERIS (the French public interest group for risk management related to post-mining), INERIS files, and from scientific publications. The database contains 81 discharges with 450 flow measurements and 274 water analyses.

Coal mining began in France in the 13<sup>th</sup> century and ended in 2004 (Bouzenot et al 2010). During this period, 4,700 Mt of coal were extracted from nine major coal basins: most of them were in the Nord-Pas de Calais and the Lorraine basins, which respectively extend to Belgium and Germany. About half of all the French coal basins are now flooded and most of them discharge to surface waters. The main pollutants are sulphate and iron (Fabriol 2005).

The coal mine aquifers contain about  $3 \times 10^9$  m<sup>3</sup> of mine water and their total outflow is estimated to exceed 120 Mm<sup>3</sup>/a, which is equivalent to about 6% of the total river flow in France. 95% of the coal mine waters flow into rivers, while 4% flows into lakes, and 1% flows directly into the sea.

### GERMANY

Coal mining in Germany is very diverse, comprising both lignite and hard (anthracite and bituminous) coal. Whereas opencast lignite mining (not discussed in this paper) will continue post-2018, the last active hard coal mining will cease in 2018. The German hard coal measures extend east to west, following the Rhenohercynian and Saxothuringian strike, and extend into Belgium and France in the west and Poland in the east.

The remaining active mines can be found in the Ruhr Area (Rheinisch-Westfälisches Steinkohlerevier) and at Ibbenbüren. As in many other coal mining areas, the mines have varied hydraulic status; mines currently being flooded are located close to mines that have already been flooded and are discharging water. There is still no discharge during the flooding because pumping is temporarily being maintained. Furthermore, the groundwater level is not uniform; many abandoned and partially flooded mines in the Ruhr Area have a temporary water level below -600 m b.s.l. This means that most of the German coal mines aquifers have not reached equilibrium conditions, which would allow for extrapolation of long term discharge flow rates and quality. Instead, an “unsaturated zone” exists above the mining water table, where oxidative reactions play an important role, and the potential for post-flooding mobilization of oxidation products is still considerable.

The Ruhr Area is the largest of the German coal fields and its discharges exceed the sum of all of the other German hard coal mining areas. Pumping rates in this area has ranged between 48 and 54 Mm<sup>3</sup>·a<sup>-1</sup> during the last decade. However, its main environmental impact is related to the flow rate of the receiving water course. Therefore, future dewatering of the Ruhr Area plans within in frame of mine closure and cessation of pumping activities for withdrawal from some actual discharges into local rivers (Emscher, Ruhr, and Lippe), and installation of an underground drainage system via mining voids to a major discharge into the Rhine River, which can provide high dilution rates (Fischer 2015).

### GREECE

Many coal basins exist in Greece: the major coal mining areas are found in or near Ptolemais, Florina, Drama, Elassona, Megalopolis, Ioannina, and Komnina (Koukoulas and Koukoulas 1995). Power generation in Greece relies mainly on lignite combustion (about 50% of power generation). The major generating stations are situated in Ptolemais-Kozani, Amyntaio, Florina, and Megalopoli, and are owned by the Public Power Corporation (PPC). Data concerning mine water discharges were obtained from the PPC Environmental Protection and Land Reclamation Sector Determination agency, and relate to the Ptolemais and Meliti opencast lignite mines. Water

samples were collected and analysed from wells, pumping stations, and streams. The main contaminants were As, Cd, Cr, Ni, Zn, Pb, Hg, and SO<sub>4</sub>.

#### POLAND

Information on the impact of mine water discharges on the water environment in Poland is dispersed and not readily available. Under the framework of this project, multi-year data, including basic hydrochemical parameters, specific contaminants, and priority substances in 15 mine water discharges, were gathered by the Central Mining Institute in Katowice.

The Upper Silesian Coal Basin (USCB) is located in southern Poland and is one of the largest hard coal basins in the world (Rozkowski 1993). Hard coal has been mined in the USCB since about 1540. Recently (2014), there were 29 active hard coal mines, 25 abandoned coal mines still being dewatered, and one abandoned coal mine where dewatering has stopped and which consequently has been partially flooded. The amount of groundwater being pumped from the coal mines totals 209 Mm<sup>3</sup>/a. Mine dewatering is the main cause of the USCB's hydrochemical regime disturbances. Mine waters from these mines are typically discharged into tributaries of the upper Wisła (Vistula) and upper Odra (Oder) rivers (Janson et al. 2009). These mine waters are generally alkaline, but have elevated concentrations of sodium, chloride, and sulphate (Pluta and Zuber 1995). Radium is also often present in waters with low sulphate concentrations (Chalupnik and Wysocka 2008).

In the USCB, about 480 million metric tons of mine waste is stored in approximately 160 waste heaps (Szczepańska and Twardowska 1999; GUS 2015). Research carried out within the MANAGER project revealed that some of them may be considered to be “hot spots,” releasing acidic runoff waters with high concentrations of sulphate, iron, aluminium, zinc, and nickel into the groundwater (Bauerek et al. 2014). Quantitative data concerning this phenomenon is sparse, although the data from the eastern part of Silesia shows that the environmental impact of contaminated runoff waters is locally important, and that it increases substantially during warmer periods of the year, when heavy rains occur (Bauerek et al. 2013). Some of the waste heap discharge waters are alkaline, but they frequently have high concentrations of sulphate, ammonia, and metals (Sracek et al. 2010).

#### SPAIN

In central Asturias, a large coal field has been mined since the early 19<sup>th</sup> century in several exploitation units (Ojeda 1985). Most of the strata of the Central Carboniferous River basin can be considered effectively impermeable, except for some intensely fractured siliciclastic and carbonate rocks, through which infiltration takes place (Martos et al. 2008). There are more than 73 coal shafts in this area, and more than 300 mines and 2000 mine adits in this mountainous area (Fandos et al. 2004). Currently only four collieries remain in operation in two coal fields (San Nicolas and Santiago Colliery in the Caudal Field, and Maria Luisa and Carrio Colliery in the Nalón Field).

Coal has been mined in Spain for more than 200 years, generating a system called “anthropogenic aquifer,” which is still being dewatered by several wells (Pendás and Loredó 2006). The low sulphur content in the coal, as well as the abundant carbonate materials, which act as a buffer, reduces the problem of acidic and ferruginous water. However, concentrations of sulphate, bicarbonate, calcium, magnesium, and other ions can be elevated (Martos et al. 2007). Hunosa, a state-owned coal company, is responsible for the mine water discharges at locations where pumping stations are installed. The total outflow exceeds 37 Mm<sup>3</sup>/a for 15 discharge points, all of which are being monitored. In addition, four old mine water discharges have been periodically monitored.

#### UNITED KINGDOM

Underground and opencast mining is ongoing in the UK, but on a considerably reduced scale when compared with historic mining rates in the 19<sup>th</sup> and 20<sup>th</sup> centuries. The Coal Authority manages the effects of past coal mining, including mine water pollution, and has been regularly monitoring mine water levels and quality across the UK for over 20 years (Crooks and Thorn 2016). Most of the mine waters in the UK are circumneutral in pH and ferruginous, and range from fresh to hypersaline, depending on coal field geology, geometry, and mining history. Iron and, to a lesser extent, manganese are the principal metal contaminants in UK mine waters. Other metal and metalloid contaminants are typically only present at relatively low concentrations.

The Coal Authority operates 70 mine water treatment sites throughout Britain, of which 53 remediate existing mine water discharges and 17 treat water on-site to prevent new discharges of contaminated water (Lawrence

2015). Together, the 70 mine water treatment sites prevent over 4,000 tons per year of iron from discharging into watercourses, and so improve and protect the UK's rivers and streams. Some water treatment operations also exist to protect important drinking water aquifers.

## Materials and Methods

The first step for selecting priority substances for mine water was to develop a coherent and comprehensive methodology and the tools for gathering, storing, and analysing mine water contaminant data from different coalfields in Europe. This consisted of the following activities:

- Gathering available information, from all partners, about contaminants in rivers and mine water discharges from different coalfields in Europe, and collecting additional samples where data were lacking;
- Listing of 58 major and minor ions, metallic trace elements, and physicochemical parameters, based on all of the measured or analysed factors;
- assigning the following attributes to each discharge point:
  - individual name,
  - information about geographical coordinates, country, coal district, name of mine, type of mining and mine status, and quantity and altitude of the discharges,
  - name, code, and mile of river or water body where a discharge was localized.
- Standardization of units for each parameter (e.g. nitrogen compounds can be expressed in mg<sub>N</sub>/L or mg<sub>NO<sub>3</sub></sub>/L, depending on the country);
- Replacement of concentrations under limits of detection (LOD) by systematically applying LOD/2; this was necessary because the studied countries had different LOD and/or the LOD may have varied over time;
- Performing a basic statistical evaluation based on multi-year observations for each mine water discharge point;
- Gathering information about emission limit values (ELV) and acceptable status ranges, according to local regulations introduced by the Water Framework Directive (WFD) as environmental quality standards (EQS).

The next step was to analyse the information gathered from the different coalfields to identify the important contaminants from the point of view of the WFD objectives. Water parameters that could cause environmental risks under specific local conditions, and contaminant concentrations in discharges that exceeded local pollutant emission limits, were also selected as mine water priority substances (MWPS). Selection of MWPS was based on two multi-year average contaminant concentrations (MACC) criteria, where a mine water discharge was causing a significant deterioration in water body status:

- The MACC of a specific contaminant in mine water discharges was exceeding an established ELV or EQS, based on local regulations or the WFD, regardless of initial water quality;
- The MACC of a specific contaminant in mine water discharges was not exceeding established ELV or EQS, but the water body quality exceeded one or more of these thresholds downstream of the mine water discharge point, based on local regulations or the WFD.

## Results and Discussion

The main goals were achieved. A coherent and comprehensive database for storing and analysing mine water contaminant data from the different European coalfields was developed, and mine water priority substances that pose the risks to the aquatic ecosystems were identified. The database contains:

- Information about MACC at 97 mine water discharge points,
- ELV for 62 discharge points and EQS regarding the WFD,
- A list of mine water priority substances for each country, and
- Substantiation of the selected mine water priorities.

Contaminants were identified that can cause environmental risks, such as deterioration of river ecosystems, when their concentrations exceed ELV for European coal mine waters (Table 1). Sulphate and iron were identified as priority substances in almost all of the countries, while several metallic and metalloid trace elements (e.g. As, Cr, Zn) were problematic only in some. High concentrations of chloride (and sodium, not shown) were linked to the Na-Cl brines associated with coal mining in Germany, Poland, and UK. High  $^{226}\text{Ra}^{2+}$  concentrations in Germany and Poland were associated with high Ba/low sulphate water.

**Table 1 will be placed near here during the printing process**

Box/whisker plots for the principal priority substances shown in Table 1 can be observed in Figure 2. Concentrations values and basic statistical parameters are provided in Table 2. More than 26,000 analyses were gathered for 97 mines spread over the studied countries, ranging from 5 to 26 by country. These mines represent a total discharge of about 220 m<sup>3</sup>/s, i.e. almost 7,000 Mm<sup>3</sup>/a.

**Figure 2 and Table 2 will be placed near here during the printing process**

The pH was always higher or equal to 6.0, with an average of 6.50 (Table 2). Sulphate (Fig. 2a) averaged 713 mg/L and iron (Fig. 2b) averaged 20.9 mg/L, for all of the country's mine discharges. These elements are produced by the oxidation of sulphides like pyrite (Blowes et al. 2003), which produces 1 mol of Fe<sup>2+</sup> and 2 mol of SO<sub>4</sub><sup>2-</sup> for each mol of pyrite that is oxidized. The reaction also produces two mol of H<sup>+</sup>, which can react with carbonate rock, such as calcite and dolomite, if any is present in the nearby strata.

Furthermore, the released Fe<sup>2+</sup> is oxidized and precipitates at pH > 3.0 as ferric hydroxide, which is also an acid-producing reaction (Langmuir 1997). Meanwhile, the neutralization reactions can produce rather high concentrations of Ca<sup>2+</sup> (Fig. 2a, Table 2), which in this study averaged 384 mg L<sup>-1</sup>, and Mg<sup>2+</sup> (Fig. 2a), which averaged 169 mg L<sup>-1</sup>. The Ca<sup>2+</sup> may precipitate with sulphate in neutralized waters as gypsum, CaSO<sub>4</sub>·2H<sub>2</sub>O, but sulphate in neutral pH mine waters generally behave in a conservative way because saturation with respect to gypsum is not typically reached. Jarosite, KFe<sub>3</sub>(SO<sub>4</sub>)<sub>2</sub>(OH)<sub>6</sub>, precipitates in mine waters at a pH < 3.0 (Langmuir 1997; Blowes et al. 2003) and, thus, cannot control sulphate concentrations in the generally circumneutral studied mine discharges.

Manganese was also present in most of the mine discharges (Fig. 2b), with an average value of 3.69 mg/L.

Chloride together with sodium (not shown), averaged 2,747 mg/L, and was most commonly associated with the coal deposits in Germany, Poland, and the UK (Fig. 2a). In Germany and Poland, these major ions come from fossil sea water but in the UK, they are probably due to sea water inflow into submarine mine adits (Younger et al. 2002). Chloride behaves conservatively in mine discharges and its concentrations generally decreases only by dilution and dispersion (Motyka and Postawa 2000). Boron (Fig. 2b), which averaged 0.96 mg/L, is also linked to fossil sea water in Germany and Poland.

Lead (Fig. 2c) was present in all of the mine discharges except for Spain at relatively high concentrations (averaging 54.2 µg/L), compared to its EQS (1.2 µg/L<sup>-1</sup>). Cadmium was also present in several of the mine discharges, presumably as a consequence of dissolution of carbonates. Its average concentration (5.5 µg/L) was also high relative to its EQS, which depends on water hardness; the EQS for Cd varies from less than 0.08 µg/L if the hardness (as CaCO<sub>3</sub>) is < 40 mg/L to 0.25 µg/L if the hardness exceeds 200 mg/L. Typically, there is no solubility control on these contaminant concentrations but they can be attenuated by adsorption on ferric precipitates (Cravotta et al. 2008).

Significant concentrations of Hg (Fig. 2c) occur in coal mine discharges in several countries, averaging 0.3 µg/L, which exceeds its EQS (0.05 µg/L), which means that many of the mine waters exceed this threshold.

High concentrations of Ni, As, and Cr (Fig. 2c) were less common and occur mostly in mine discharges in the UK, France, and Poland. Their respective average concentrations and EQS were 37.4 µg/L for Ni (EQS: 4.0 µg/L<sup>-1</sup>), 18.6 µg/L for As (EQS: 4.2 µg/L), and 8.3 µg/L for Cr (EQS: 3.4 µg/L<sup>-1</sup>). Generally, there is no solubility control on the concentrations of these elements, but they also are adsorbed on precipitated ferric phases (reaction 3) at about neutral pH (Cravotta 2008b; Sracek et al. 2011) because desorption of oxyanions occurs at higher, alkaline pH, i.e. for As at pH above 8.2 (Ravenscroft et al. 2009). These trace elements do not represent a global environmental problem for coal mine water.

Zn was also found in mine water in Poland and Germany with sometimes very high concentrations (respective maxima at 17,800 and 25,000 µg/L), but there is no EQS for this element. Just like Ni, Zn occurs as a cation and there is generally no solubility control on its concentrations in mine discharge.

Se can be an important issue in coal mine water (Vesper et al. 2008; Yudovich and Ketris 2006). However, it was only analysed in coal mine waters of Greece and Spain with an average concentration of 3.5 µg/L. Again, there is no EQS for this element.

Finally,  $^{226}\text{Ra}^{2+}$  (Fig. 2c), which averaged 1,190 µg/L, is of particular interest, but it was present only in mine water discharges in Germany and Poland with significant differences (respective maxima were 7,300 and 244 µg/L). In these waters, sulphate has been reduced by reactive organic matter (Appelo and Postma 2005) and so there is no solubility control of  $^{226}\text{Ra}^{2+}$ . In high sulphate waters,  $^{226}\text{Ra}^{2+}$  co-precipitates with gypsum and/or barite (Chalupnik and Wysocka 2008). Relatively high concentrations of Ba (Fig. 2b), respectively averaging 26.1 and 5.1 mg/L, also occur in low sulphate waters. High Ra and Ba concentrations in low sulphate waters are consistent with findings of Cravotta (2008b) coal mine discharges in Pennsylvania.

## Conclusions

More than 26,000 mine water analyses were gathered from 97 mines from the countries involved in the project, i.e. France, Greece, Germany, Poland, Spain, and the UK. In each country, the amount of water stored in coal mine aquifers totals over  $10^9$  m<sup>3</sup>, and the combined mine water outflow (natural in the case of abandoned mines or artificial, due to pumping, in the case of active mines) is around several hundred Mm<sup>3</sup> per year. All the studied mines represent a total discharge of about 7,000 Mm<sup>3</sup>/a.

The quantitative influence of coal mine water discharges is therefore substantial, and evaluating their environmental impact on surface and underground water is important. The coal mine water quality evaluation notes a first group of pollutants, represented by SO<sub>4</sub> and Fe, which were shared by all countries; a second group of frequently observed pollutants (Mn, Cd, Pb); and a third group, that is specific to certain mine waters (Cl, B, Ba, As, Cr, Ni, Zn, Hg, Ra). Some discharges show substantial mineralization, frequently accompanied by high concentrations of metallic or metalloid trace elements, and sometimes by radium and barium, typically in low sulphate waters. Chloride and boron have a special status because they are linked to the impact of fossil sea water brines. The remaining trace metals generally were present at low concentrations, but since their EQS are very low, they often exceed these thresholds.

The selection of mine water priority substances was based on a comparison of river water quality standards with mine water quality data. This means that there is no certainty that the mine discharges of selected substances will cause deterioration of river status or adversely affect aquatic ecosystems. The mine water quality database that we developed identified the contaminants that are of primary concern from the point of view of achieving WFD objectives, and that should be considered during environmental risk assessment.

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### **Figure Captions**

**Fig. 1:** Location of the studied coalfields in Europe: a) Major elements; b) Minor elements; c) Trace elements

**Fig. 2:** Concentrations of priority pollutants in mine waters; coloured boxes are the interval from the 1st quartile (Q1) to the 3rd quartile (Q3) and represent the median in the form of a horizontal black line; the vertical black bars correspond to the limits above which the values are considered abnormal, i.e.  $Q1 - 1.5(Q3 - Q1)$  for the lower limit and  $Q3 + 1.5(Q3 - Q1)$  for the upper limit.

**Table 1.** Global statistics of priority pollutants. EQS (Environmental Quality Standards) are EQS-AA (Annual Average) for all trace elements but PNEC (predicted no effect concentrations) for As and Cr (under European Directive 2013/39/UE). No. = number of samples.

	Major ions (mg/L)					Minor ions (mg/L)				Trace elements (µg/L)								Bq/L	
	pH	Ca	Mg	Cl	SO <sub>4</sub>	Fe	Mn	B	Ba	As	Cd	Cr	Pb	Ni	Zn	Hg	Se	Ra	
No.	2583	2470	2419	2972	2848	2538	2507	1977	1637	245	1269	846	1702	1758	1348	236	30	91	
Min.	6.0	1	1	1	0	0.01	0.01	0.01	0.01	bdl	bdl	bdl	bdl	bdl	bdl	0.1	0.3	0.01	
Max.	8.9	5070	1320	71400	4680	330	77.0	13.0	5800	900	130	155	2280	421	25000	2.5	10	7.30	
Ave.	6.5	384	169	2756	713	20.9	3.69	0.96	9.77	18.6	4.5	8.3	54.2	37.4	440	0.3	3.5	1.19	
EQS	6-9									4.2	*	3.4	1.2	4.0	7.8	0.05			

\* depending on water hardness: from <0.08 µg/L if hardness (as CaCO<sub>3</sub>) < 40 mg/L to 0.25 µg/L if hardness ≥ 200 mg/L; bdl = below detection limit

**Table 2.** Statistics of priority pollutants by country; Number (No.) represents the number of water samples analysed in the mines referenced in each country; “bdl” is below detection limit)

Country		Major ions (mg/L)				Minor ions (mg/L)				Trace elements (µg/L)								Bq/L	
		Ca	Mg	Cl	SO <sub>4</sub>	Fe	Mn	B	Ba	As	Cd	Cr	Pb	Ni	Zn	Hg	Se	Ra	
Greece	No.	60	60	60	60	60	60	0	0	60	60	60	60	60	60	60	10	0	
	Min.	44	15	6	0	0.03	0.01			0.2	0.2	1.7	4.0	5.0	0.5	bdl	10.0		
	Max.	184	49	78	467	1.21	0.10			3.2	1.3	28.0	12.0	24.0	40.0	bdl	10.0		
	Ave.	94	28	16	144	0.18	0.03			1.1	1.0	3.9	5.1	8.7	6.8	bdl	10.0		
Spain	No.	67	67	0	65	65	63	40	0	40	40	40	40	40	0	40	20	0	
	Min.	46	8		24	0.01	0.01	0.01		0.3	0.1	0.3	0.3	bdl		bdl	0.3		
	Max.	319	100		1747	9.05	0.92	0.19		9.4	0.1	0.6	4.8	2.9		bdl	0.4		
	Ave.	129	53		416	1.87	0.26	0.07		0.6	0.1	0.3	0.4	1.4		bdl	0.3		
Poland	No.	231	231	606	606	243	213	21	44	0	198	198	200	199	201	15	0	10	
	Min.	1	21	41	119	0.01	0.01	2.23	0.12	bdl	bdl	0.6	0.4	0.3	0.3	0.1		0.060	
	Max.	1062	707	36072	2868	39.0	8.27	4.25	7.70	bdl	130	155	560	421	17800	2.5		0.244	
	Ave.	263	162	7579	783	3.59	1.30	3.19	5.06	bdl	9.3	18.9	60.8	43.3	287	2.3		0.160	
United Kingdom	No.	1427	1378	1427	1225	1313	1427	1340	986	53	385	0	848	900	484	0	0	0	
	Min.	1	1	6	3	bdl	0.03	0.01	0.01	1.0	0.3	bdl	1.0	10.0	5.0				
	Max.	1920	1180	29100	4680	91.5	77.0	7.36	0.06	900	2.6	bdl	2280	260	7840				
	Ave.	247	129	2296	944	34.2	5.58	0.73	0.03	21.9	7.3	bdl	85.5	53.1	40.4				
Germany	No.	601	601	795	792	758	650	571	604	41	557	522	531	531	572	104	0	81	
	Min.	3	7	61	3	0.01	0.01	0.23	0.03	1.0	0.1	2.0	0.5	2.5	5.0	0.1		0.010	
	Max.	5070	1320	70400	1890	330	28.0	13.0	580	10.0	40.0	130	1000	240	25000	0.6		7.30	
	Ave.	841	291	12597	353	7.97	1.19	1.51	26.1	5.0	1.7	5.3	13.7	16.0	899	0.2		1.32	
France	No.	84	82	84	100	99	94	5	3	51	29	26	23	28	31	17	0	0	
	Min.	30	3	1	159	0.05	0.02	0.18	0.06	2.5	0.3	0.3	bdl	1.9	0.2	0.1			
	Max.	472	453	278	3010	43.8	9.11	1.01	0.11	347	25.0	57.2	50.0	32.0	500	1.0			
	Ave.	195	156	64	839	10.5	2.50	0.52	0.09	60.5	4.2	10.2	4.9	10.5	42.3	0.3			

bdl = below detection limit

Fig 1

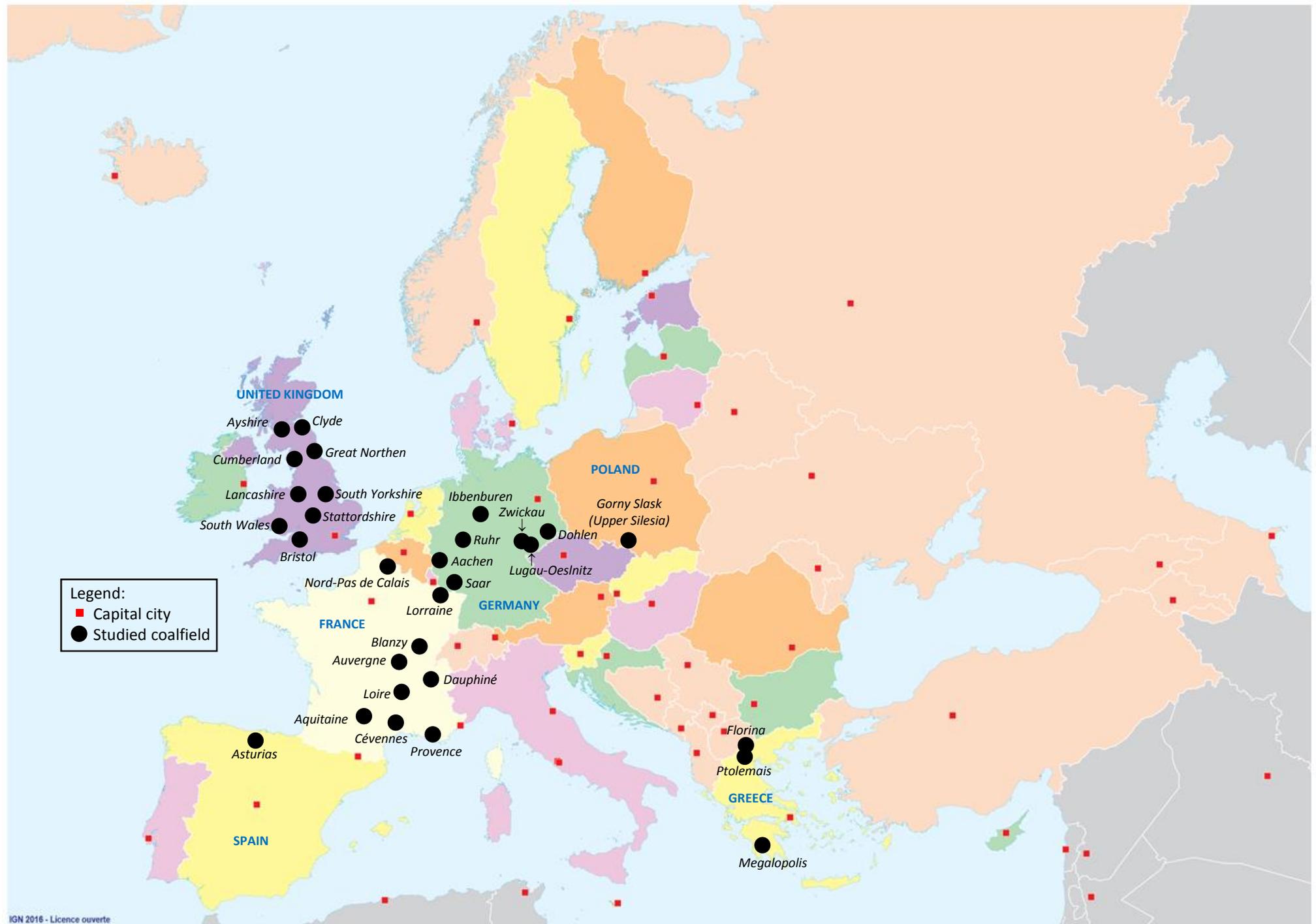


Fig 2a

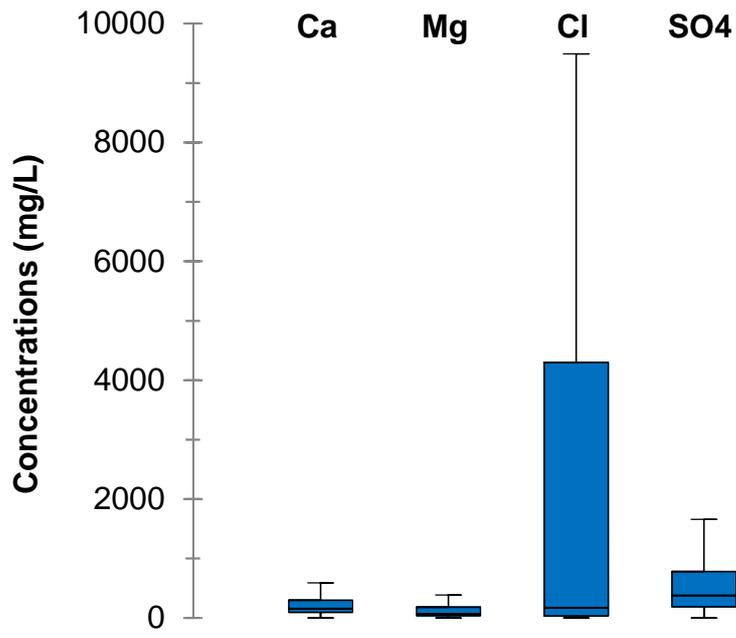


Fig 2b

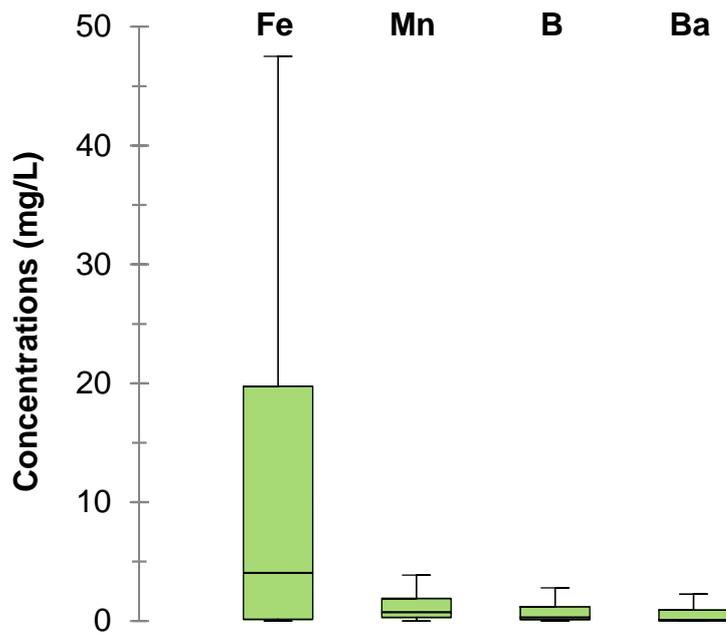


Fig 2c

