Research papers

Roman road pollution assessed by elemental and lead isotope geochemistry in East Belgium

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ABSTRACT

The ability of inorganic geochemistry to record environmental change and especially human impact has been evidenced by several studies across Europe, especially in peat, where it is possible to record the impact of agriculture, mining and other industries. However, despite the numerous investigations on the impact of ancient human activities such as ore mining and smelting, little attention has been paid to geochemistry as a tool to solve problems of palaeopollution in the surroundings of archaeological sites. This paper presents geochemical evidence of the impact of a possible early Roman road built in SE Belgian peatland. Increased Zn and Pb concentrations suggest that Pb–Zn ores were transported on the road. Lead isotope analyses suggest that these ores are locally derived, being compatible with those found in the nearby Pb–Zn ore deposits from East Belgium. Present results provide direct evidence that East Belgian Pb–Zn ores were already being mined during Roman times, i.e. earlier than previously suspected (i.e. 14th century) and that Zn appears to be relatively immobile here. On a broader scale, it also demonstrates that such an early road already had an impact on the environment in terms of metal pollution. This paper enlarges on the range of possibilities offered by geochemistry in the field of geoarchaeology.

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1. Introduction

The Hautes Fagnes plateau (SE-Belgium) consists of a large and discontinuous wetland made of fen, mire and rarely bogs (Fig. 1A). It has provided a key site in studies investigating human occupation and environmental impact in the region since the Celtic civilisation (Remy, 1981; Damblon, 1994; Graillet, 1998; De Vleeschouwer et al., 2007). Interestingly, this area is also located nearby Pb–Zn ores that are known to have been mined since the 14th century (Dejonghe and Ladeuze, 1993).

The Hautes Fagnes plateau is crossed by a 6-km cobbled road, in a NW–SE direction (Fig. 1A), which is now partly covered by peat. Historical (Remy, 1981; Graillet, 1998) and palynological (Dalemans and Streel, 1986) data suggest that it was used as a thoroughfare up to the 13th century although the age of its construction, function and its intended route have always been a matter of debate (e.g. Corbiau, 1981a,b; Dalemans and Streel, 1986).

The cobbled road crossing the Hautes Fagnes is recorded in several historical texts and the archaeological remains were discovered in 1768 (see Nekrassoff, 1993). The first detailed description of the road was made by Bastin (1934), who discovered a multi-layered wooden structure which was thought to form the foundation for the cobblestones on the waterlogged peat (Fig. 1B). Such a structure is not found where the road is crossing non-peaty
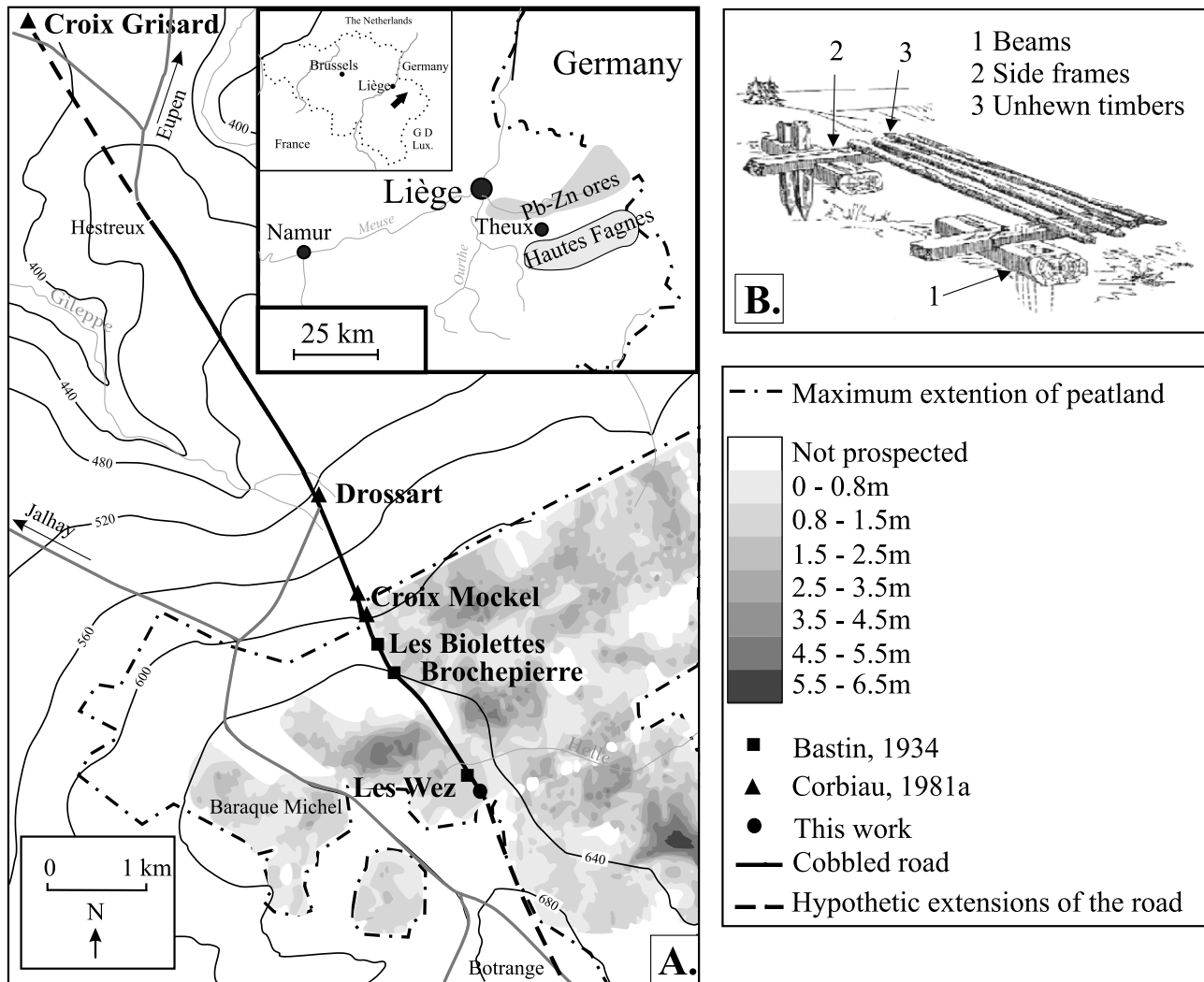
Fig. 1. (A) Location map of the Hautes Fagnes plateau, the Pb–Zn ores of E-Belgium (Cauet et al., 1982; Dejonghe, 1998), the cobbled road and the various excavations (after Corbiau, 1981a). Peat thickness reported from Wastiaux and Schumacker (2003). (B) Sketch of the findings from Bastin (1934) redrawn by Mertens (1957).

sediments (Corbiau, 1981a) as this kind of wood basement is not necessary to support the road when the substratum is not waterlogged. From base to top, this bearing beech-wooden structure was composed of three parts: (1) a set of beams perpendicular to the road, (2) girder/side frame parallel to the road and fitted into the beams, (3) an assembly of unhewn timbers lying perpendicular to the road. The cobbling overlying the wooden foundations is composed of quartzite blocks from the Revin group (Cambrian), a rock common as erratic blocks on the Hautes Fagnes plateau. Finer sediments of variable grain-size ranging from clay to fine gravel are scattered between the cobbles. Given these particular structural and compositional characteristics, although without any age-dating control, Bastin (1934) attributed a Roman origin to the road. Much later, Corbiau (1981a) carried out three new excavations in the northern part of the road where it was built across a very thin peat layer or on non-peaty sediment (Fig. 1A). The alder-wooden structure at these excavations was either missing or consisted of a single unhewn timbers assembly under the cobblestones. Wood fragments were radiocarbon dated to a period between the 5th and the 8th centuries AD (Damb-lon, 1994). Therefore, Corbiau (1981a) advocated a Merovingian age for this part of the road. This younger age was not contradictory with the estimation of Bastin (1934), as Corbiau (1981a) mentioned that these new excavations could be non-contemporaneous from those previously studied. Three wood fragments kept from the excavations of Bastin (1934) were later radiocarbon dated and also provided contradictory age-dating of BC 170, BC 248 (Lewalle, 1963) and AD 550 ± 50 (Corbiau, 1981a) that were never discussed. Subsequently, Dalemans and Streel (1986) did a palynological investigation of four cores from the southernmost site studied by Bastin (Fig. 1A) in an attempt to constrain the age-dating of the road construction. They assigned the first impact of the road to small eroded quartzite fragments found in the peat. Using this proxy for the lowest level of the road, these authors compared the pollen record with that of Persch (1950) and estimated the building of the road to have occurred around AD 700 (Dalemans and Streel, 1986).

The possible destination of the road was also thoroughly discussed. In the middle of the 19th century the cobbled road was considered to be part of a Roman road

linking the cities of Maastricht and Trier (Corbiau, 1981b). Later on, it was associated to a Roman road called *Via Mansuerisca*, which has been cited in historical texts since 670 AD (Corbiau, 1981b). Several tracks were proposed to link the cobbled road to Trier or to the Köln-Trier Roman road. However, as stated by Corbiau (1981b) these tracks are still hypothetical. Systematic archaeological investigations are needed as despite these numerous studies, the age of the road and its purpose and destination, which may be crucial factors in determining its age, are still under debate.

Moreover, of wider interest, it is well known that the Romans built the first extensive road web across Europe. First built for military purposes, these roads were further developed for trade purposes (Chevallier, 1972). In Belgium, the Roman road system was extended *ca.* 20 BC, mainly for the defence of northern borders against possible Barbarian invasions, but also for political purposes and for trade between the different cities (Corbiau, 2006). At the Roman Empire apogee, roads covered 150,000 km (Chevallier, 1972) throughout Europe. The environmental impacts provoked by such an intense and modern road web has however never been investigated.

Roman road engineers generally avoided any wetlands, river overbank and inundation plains (Chevallier, 1972). For these reasons, to the authors' knowledge, apart from one other site in Austria (*via Claudia Augusta* near Lermoos city), the archaeological site described here is unique for the Roman Empire. Indeed, due to its construction in a peatland, the road is very well preserved. Moreover, it offers a unique opportunity to study possible pollution that could be recorded in the surrounding peat accumulation.

In order to answer the various historical, geographical and environmental questions mentioned above, the geochemical and Pb isotopic records in two peat columns taken alongside the cobbled road have been studied. These analyses are accompanied by radiocarbon dates. Local problems discussed in the present study are (1) documenting the purpose of such a road by placing it in a geographical and historical context, (2) providing new clues to better constrain the age of its construction and (3) providing an age of possible early Pb–Zn mining in Belgium. On a broader scale, this study also (1) assesses the environmental impact of such early roads and (2) discusses Pb and Zn mobility in peat.

2. Using geochemistry in geoarchaeology

Elemental geochemistry and Pb isotope analyses have been successfully used for investigations of peat sequences to reconstruct pollution chronologies in Europe (e.g. Brännvall et al., 1999; Martínez-Cortizas et al., 2002; Novak et al., 2003; Shotyk et al., 2003; Le Roux et al., 2004a). Contaminant metals, particularly Pb and Zn, linked to mining and smelting activities, have been monitored at various European sites including West Germany, (e.g. Kempter and Frenzel, 1999), Switzerland, (e.g. Shotyk et al., 2003), France, (e.g. Baron et al., 2005) and East-Belgium, (e.g. Sonke et al., 2002). In some cases, early pollution could be evidenced, especially if the polluting source was in the vicinity of the study site, (e.g. West et al., 1997; Mighall et al., 2002; Baron et al., 2005). Previous research dealing with pollution histories in Belgium have been carried out in ombrotrophic bogs from the Hautes Fagnes plateau, SE-Belgium (Kempter, 1996; Gérard, 2004; Goormaghtigh, 2005; De Vleeschouwer et al., 2007) and peaty sediments (Sonke et al., 2002). These studies showed that anthropogenic impact dated back to early Roman times.

In the specific field of geoarchaeology, little attention has been given to the potential of tracing past pollution using geochemistry (Mighall et al., 2006; Nriagu, 1996). However, previous studies have successfully demonstrated the potential of this approach, (e.g. Elmaleh et al., 2007; Grattan et al., 2007; Baron et al., 2005; Le Roux et al., 2004b). To the authors' knowledge, this study constitutes the first investigation of environmental impact due to road use during the Roman Empire. It provides a good example of how geochemistry can be used as a tool to broaden knowledge in the role that early civilisations played on the environment.

3. Materials and methods

3.1. Coring and subsampling

In July 2004, an archaeological excavation was carried out by the "Direction de l'Archéologie du Ministère de la Région Wallonne" (Corbiau, 2005) a few tens of metres from the original site studied by Bastin (1934). Fig. 2A shows a cross-section of the excavation. The 30-cm thick peat covering the road was removed and a 1.2-m deep trench was dug in the southern section of the road, perpendicularly to its direction. The revealed road structure (Fig. 2A) was similar to the section previously studied by Bastin (1934). The peatland is developed on a slight slope. It is now partly fed by small eastward surface streams.

Two peat columns (CR1, downhill and CR2, uphill; Fig. 2B) were collected in the trench on each side of the road using plastic U-boxes (20 cm × 10 cm × 6 cm). CR1 and CR2 measure approximately 103 cm and 125 cm long, respectively. An undisturbed 2 cm-thick slice was cut along the side of each block and impregnated with resin. The peat blocks were then cut into 1-cm thick slices with plastic tools. Note that 0 cm is at the bottom of the cores.

As there is surface runoff, any possible environmental impact of the road should be more accurately recorded down slope. Therefore, most of the results were obtained on CR1.

3.2. Thin sections

Thin sections were made using the method described in De Vleeschouwer et al. (2008). The peat slices were freeze-dried under high vacuum (6–8 mbar) and impregnated with resin under low vacuum (700–1000 mbar). Polymerisation was achieved in a drying-oven (40 °C, 72 h) and then under laboratory conditions (100 h). Blocks were then polished and thin sections were prepared.
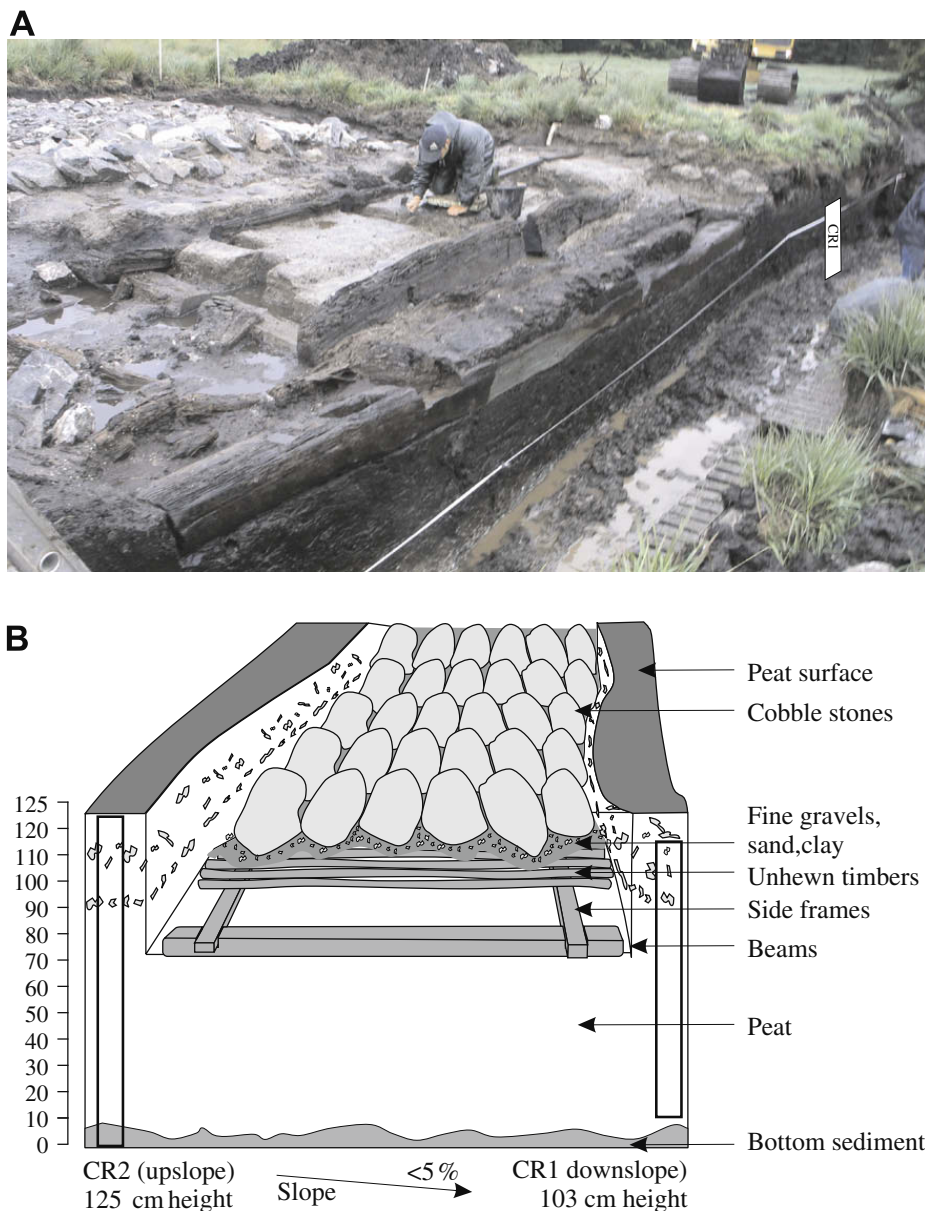
Fig. 2. (A) Picture of the excavation in course of study. The position of CR1 is shown. (B) Schematic drawing of the two peat sections (CR1 and CR2) taken aside the road. Wooden structure after Corbiau (2005). Vertical scale is exaggerated five times. CR1 is 103 cm long and is located downhill from the road section and CR2 is 125 cm long and is located uphill from the road section.

3.3. Geochemistry

One-cm thick slices of peat were freeze-dried and milled in an agate jar pulverisette (400 rpm, 1 h). Pellets were prepared by pressing 3 g of peat powder (200 kN/cm^2). The following elements were analysed using X-ray fluorescence (ARL 9400 XP, URPGE, ULg): Si, Al, Ca, Na, K, Zn and Pb. Thirty-nine and 46 samples were analyzed in CR1 and CR2, respectively. International reference material such as coals (CLB-1, NIST 1632b), lichen (BCR-482), pond sediments (NIES-2), tea leaves (NIES-7), exhaust particles (NIES-8) and marine sediments (JSD-2, JSD-3, SGR 44) were used as working standards for trace elements. In-house standards, made by mixing together a low ash peat with Merck quality oxides or carbonates for each of the major element, were used for the calibration of major elements. The resulting mix was then progressively diluted to obtain

five aliquots of different concentrations. Their respective concentrations were previously measured by inductively coupled plasma atomic emission spectrometry (ICP-AES) at the "Musée Royal d'Afrique Centrale" (MRAC) in Tervuren (Belgium) together with the low ash peat after ashing and acid digestion in a clean laboratory (De Vleeschouwer, 2007). Repeated measurements ($n = 20$) allowed the estimation of reproducibility and accuracy (Table 1).

3.4. Lead isotopes

Twelve samples from CR 1 and five samples from CR 2 (see Table 2 for depths) were prepared in a laminar flow clean air cabinet using the following protocol: 100 mg of peat powder were digested in 30 mL open Savillex® beakers (95 °C, 72 h) using successive additions of H_2O_2 30% p.a. and HNO_3 14 N cc sub in a proportion of 2:1.

Table 1
Selected elemental geochemistry in CR1 and CR2 cores

CR1

	Si	Al	Na	K	Zn	Pb
Reprod. (%)	99	99	97	84	96	87
Acc (%)	3	6	29	21	20	6
LD	8.1	1.7	2.4	0.1	1.0	4.9
LSQ	16.3	3.3	5	0.21	1.9	9.8
LQ	27.1	5.6	8	0.35	3.2	16.4
Height (cm)						
102	28.4	9.9	1.1	1.3	223	329
100.5	17.6	9.1	0.8	1.3	280	299
99.5	33.0	10.2	1.1	1.2	299	134
98.5	25.4	9.6	1.0	1.2	414	77
97.5	25.2	9.1	0.9	1.1	466	63
96.5	24.7	8.2	0.8	1.0	333	43
95.5	19.3	7.2	0.7	0.9	397	37
94.5	11.6	5.0	0.5	0.7	588	41
93.5	1.82	3.1	0.26	0.17	792	42
92.5	1.55	2.8	0.24	0.14	672	36
91.5	1.36	2.7	0.24	0.11	689	31
90.5	1.24	2.6	0.24	0.11	626	28
89.5	1.98	2.8	0.25	0.18	540	29
88.5	5.7	3.5	0.34	0.42	522	29
87.5	20.9	7.6	0.8	1.1	233	14.4
86.5	13.6	5.4	0.6	0.8	390	20
85.5	6.7	4.0	0.4	0.5	517	33
84.5	9.0	4.1	0.4	0.6	420	28
83.5	18.2	5.4	0.7	0.9	339	23
82.5	28.4	7.5	0.9	1.0	164	14.6
81.5	33.2	6.7	0.7	0.8	114	10.9
80.5	2.28	2.4	0.25	0.15	712	40
79.5	1.52	2.3	0.26	0.09	931	66
78.5	1.11	2.1	0.24	0.06	778	55
77.5	0.88	1.7	0.24	0.031	1037	65
76.5	1.13	1.9	0.24	0.06	1034	77
75.5	1.13	1.8	<LD	0.06	457	46
74.5	1.45	1.9	<LD	0.09	251	48
73.5	1.92	1.9	<LD	0.13	158	40
72.5	2.27	1.9	<LD	0.17	109	39
71.5	1.94	1.8	<LD	0.14	84	43
70.5	1.66	1.65	<LD	0.11	49	41
60.5	0.95	1.34	<LD	0.045	<LD	10.7
50.5	1.43	1.46	<LD	0.10	<LD	6.0
40.5	<LD	1.23	<LD	<LD	<LD	13.7
30.5	1.06	1.64	<LD	0.037	<LD	6.2
20.5	1.88	2.4	<LD	0.18	<LD	7.3
9.5	4.2	3.6	0.28	0.38	<LD	7.2
0.5	14.8	8.9	0.8	1.2	<LD	6.0

CR2

	Si	Al	Na	K	Zn	Pb
Reprod. (%)	99	99	97	84	96	87
Acc (%)	3	6	29	21	20	6
LD	8.1	1.7	2.4	0.1	1.0	4.9
LSQ	16.3	3.3	5	0.21	1.9	9.8
LQ	27.1	5.6	8	0.35	3.2	16.4
Height (cm)						
124.5	8.8	4.0	0.4	0.6	640	455
123.5	8.4	4.0	0.4	0.6	940	578
122.5	10.8	4.7	0.5	0.7	786	479
121.5	12.9	5.3	0.5	0.8	427	312
119.5	12.7	4.9	0.5	0.7	203	132
118.5	10.7	4.4	0.4	0.6	199	104
117.5	7.7	4.0	0.4	0.46	174	86
116.5	5.9	3.5	0.33	0.35	162	66
114.5	5.2	3.3	0.29	0.31	132	58
113.5	6.5	3.4	0.32	0.38	104	50
112.5	5.8	3.3	0.30	0.36	87	46
111.5	11.4	3.9	0.29	0.5	46	26
109.5	2.0	2.2	<LD	0.11	76	31
108.5	1.6	2.0	<LD	0.08	73	29
107.5	2.0	2.1	<LD	0.13	68	28
106.5	5.2	3.0	0.4	0.44	52	21
104.5	1.5	1.67	<LD	0.08	45	15.8
103.5	1.1	1.44	<LD	0.042	39	16.6
102.5	1.1	1.40	<LD	0.043	53	14.5
101.5	1.2	1.38	0.34	0.06	27	12.9
99.5	1.2	1.29	<LD	0.05	9	8.1
98.5	1.3	1.27	<LD	0.05	9	8.1
97.5	1.5	1.43	<LD	0.05	20	4.9
96.5	1.1	1.33	<LD	0.035	8	11.8
95.5	1.2	1.38	<LD	0.039	9	10.5
94.5	1.2	1.37	<LD	0.040	<LD	6.8
93.5	1.1	1.32	<LD	0.023	<LD	11.8
92.5	1.1	1.35	<LD	0.024	<LD	11.2
91.5	1.1	1.34	<LD	0.021	<LD	8.9
90.5	1.0	1.29	<LD	0.013	<LD	14.0
89.5	<LD	1.24	<LD	<LD	<LD	11.4
88.5	0.9	1.28	<LD	<LD	<LD	7.9
87.5	<LD	1.24	<LD	<LD	<LD	10.9
86.5	<LD	1.18	<LD	<LD	<LD	14.5
85.5	<LD	1.12	<LD	<LD	<LD	7.3
84.5	<LD	1.19	<LD	<LD	<LD	9.1
83.5	<LD	1.32	<LD	0.019	<LD	17
80.5	<LD	1.24	<LD	<LD	<LD	15.2
70.5	<LD	1.23	<LD	0.011	<LD	13.2
60.5	<LD	1.39	<LD	<LD	<LD	19
50.5	0.9	2.0	<LD	0.014	<LD	27
40.5	2.9	2.7	<LD	0.28	<LD	25
30.5	2.1	2.6	<LD	0.19	<LD	10.2
20.5	4.6	4.3	0.32	0.5	<LD	10.3
10.5	31.8	24	2.1	2.3	<LD	16.6
0.5	34	25	2.3	2.3	5	18

Results are given in mg/g for major elements and in μg/g for trace elements in dry peat, respectively. Values in italics, underlined and in bold are respectively qualitative (3σ–6σ), semi-quantitative (6σ–9σ) and quantitative (>9σ). LD, LSQ and LQ mean respectively limit of detection, limit of semi-quantification and limit of quantification.

Samples were then dried, HF 24 N suprapur and HNO_3 14 N cc sub were added in a proportion of 10:1 and the beakers were closed and heated (130 °C, 72 h). After drying, 2 mL of HCl 6.8 N were added and the solutions were slowly evaporated at 75 °C. The dried residues were finally dissolved in 0.5 mL HBr 0.8 N. The Pb separation was achieved by successive HBr and HCl additions on pre-conditioned columns filled with an anionic resin (AG1X8). The eluted pure Pb solution was evaporated and stored. Before isotopic analysis, the Pb elution was re-dissolved in 100 μL of HNO_3 14 N cc sub, evaporated and finally dissolved in 1.5 mL of HNO_3 0.05 N.

Lead isotopes were measured using a Nu-plasma multi collector-inductively coupled plasma-mass spectrometer (MC-ICP-MS) at the 'Département des Sciences de la Terre et de l'Environnement' (DSTE – Université Libre de Bruxelles).

Table 2
Pb-isotopic ratios in selected samples from CR1 and CR2

Height (cm)	$^{208}Pb/^{204}Pb$	2sd	$^{207}Pb/^{204}Pb$	2sd	$^{206}Pb/^{204}Pb$	2sd	$^{208}Pb/^{206}Pb$	2sd	$^{207}Pb/^{206}Pb$	2sd
CR1										
100.5[*]	38.1746	0.0169	15.6117	0.0067	18.1879	0.0079	2.09887	0.00037	0.85830	0.00012
100.5	38.1807	0.0154	15.6139	0.0057	18.1916	0.0064	2.09874	0.00045	0.85825	0.00013
98.5	38.3991	0.0177	15.6288	0.0068	18.4255	0.0067	2.08402	0.00036	0.84815	0.00011
93.5	38.3355	0.0178	15.6260	0.0062	18.3709	0.0067	2.08673	0.00037	0.85053	0.00010
85.5	38.3457	0.0152	15.6279	0.0061	18.3682	0.0066	2.08764	0.00044	0.85076	0.00012
81.5	38.6681	0.0208	15.6603	0.0079	18.7404	0.0081	2.06340	0.00004	0.83561	0.00012
80.5	38.3528	0.0160	15.6248	0.0058	18.3868	0.0060	2.08591	0.00038	0.84924	0.00011
77.5	38.3120	0.0106	15.6236	0.0041	18.3299	0.0053	2.09010	0.00029	0.85230	0.00008
75.5	38.3506	0.0169	15.6252	0.0061	18.3657	0.0074	2.08813	0.00045	0.85073	0.00011
70.5	38.3883	0.0149	15.6268	0.0057	18.3984	0.0072	2.08641	0.00035	0.84928	0.00011
60.5	38.4817	0.0180	15.6387	0.0067	18.4532	0.0072	2.08542	0.00034	0.84744	0.00011
40.5	38.7448	0.0290	15.6745	0.0079	18.8368	0.0181	2.05707	0.00072	0.83216	0.00017
9.5	38.9589	0.0153	15.7012	0.0057	19.1661	0.0071	2.03270	0.00038	0.81917	0.00011
CR2										
123.5	38.0000	0.0098	15.5952	0.0039	18.0354	0.0053	2.10686	0.00020	0.86468	0.00006
109.5[**]	38.3886	0.0105	15.6212	0.0038	18.4096	0.0035	2.08510	0.00022	0.84852	0.00005
109.5	38.3879	0.0126	15.6212	0.0053	18.4111	0.0049	2.08495	0.00020	0.84848	0.00007
102.5	38.4798	0.0084	15.6297	0.0032	18.4918	0.0053	2.08085	0.00023	0.84521	0.00008
96.5	38.6165	0.0124	15.6488	0.0042	18.6344	0.0049	2.07228	0.00021	0.83979	0.00007
50.5	38.8087	0.0113	15.6872	0.0036	19.0550	0.0043	2.03662	0.00027	0.82325	0.00007

"sd" refers to standard deviation. Duplicate is denoted by [*] while replicate is denoted by [**].

As an internal isotopic standard, a Tl solution was added to each sample and standard to monitor and correct for mass dependent isotopic fractionation. Whilst the samples were characterized by a large variability in Pb concentrations, the sample solutions were prepared to obtain a beam intensity in the Axial collector (^{204}Pb) of a minimum of 100 mV and a Pb/Tl ratio of ~5, matching the Pb and Tl concentrations of the NBS981 standard (200 ppb in Pb, with 50 ppb in Tl). The NBS981 standard was measured several times before each analytical session and between each two samples. Forty-one analyses of the standard were performed and gave the following mean values: $^{208}Pb/^{204}Pb$ = 36.7131 ± 0.0071 (2SD), $^{207}Pb/^{204}Pb$ = 15.4959 ± 0.0027 (2SD), $^{206}Pb/^{204}Pb$ = 16.9392 ± 0.0028 (2SD), which are in good agreement with long term laboratory values (*n* = 1000, $^{208}Pb/^{204}Pb$ = 36.7130 ± 0.012 (2SD), $^{207}Pb/^{204}Pb$ = 15.4950 ± 0.004 (2SD), $^{206}Pb/^{204}Pb$ = 16.9393 ± 0.0044 (2SD), and the MC-ICP-MS values of Weis et al. (2006). These values are also in agreement with TIMS triple-spike values previously published by Galer and Abouchami (1998). Although the NBS981 standard results were within the error of the triple-spike values after online correction for instrumental mass bias by Tl addition, the sample results were further corrected by the sample-standard bracketing method (as described by White et al., 2000 and Weis et al., 2006) to circumvent any instrumental drift during the analytical session. A duplicate (i.e. second sample treated the same way) of CR1 (100–101 cm) and a replicate (i.e. second analysis of the same sample) of CR2 (109–110 cm) were also measured to check the reproducibility (Table 2).

3.5. Radiocarbon dating

Six samples of bulk peat powder from CR1 were ^{14}C-dated. Samples were either 1 cm thick (10–11 cm, 80–81 cm and 82–83 cm), or 2 cm thick, i.e. a mix of two 1 cm-thick samples (74–76 cm, 76–78 cm and 78-80 cm). "Poz" samples were measured by acceleration mass spectrometry (1.5 SDH-Pelletron Model "Compact Carbon A.M.S.") at Poznan Radiocarbon Laboratory (Poland) after pre-treatment using an acid–alkali–acid washing sequence in order to remove humic/fulvic acids and bacterial CO_2 (Speranza et al., 2000). "GrN" samples were measured by conventional counting at Center for Isotope Research in Groningen (The Netherlands).

4. Results

4.1. Selected major elements (Si, Al, Na, K)

In both CR1 and CR2, concentrations of Si, Al and K display similar patterns (Table 1). In CR1, concentrations decrease from the base of the section (Si = 148 mg/g, Al = 89 mg/g, Na = 8 mg/g and K = 12 mg/g) to 30 cm (Si = 10.6 mg/g, Al = 16.4 mg/g, Na < DL and K = 0.37 mg/g) and are low between 30 and 80 cm. Between 80 and 103 cm, concentrations fluctuate from low (Si = 12.4 mg/g, Al = 24 mg/g, Na = 2.4 mg/g and K = 1.1 mg/g) to high values (Si = 332 mg/g, Al = 102 mg/g, Na = 11 mg/g and K = 13 mg/g). In CR2, elemental concentrations decrease between the bottom sample (Si = 340 mg/g, Al = 250 mg/g and K = 23 mg/g) and 50 cm (Si = 9 mg/g, Al = 2 mg/g and K = 0.14 mg/g). Values are low and stable between 50 cm and 105 cm, then increase towards 122 cm and finally decrease towards the surface. Most of the Na values are below the detection limit in CR2.

4.2. Selected contaminant metals

In CR1, Zn and Pb profiles display a similar pattern (Table 1). Between 0 and 70 cm, Zn is below the detection limit while Pb concentrations are low (6 µg/g < Pb < 13.7 µg/g). A significant amount of Zn occurs at 70 cm (49 µg/g) while the Pb content increases to reach 41 µg/g at 70 cm.

Table 3
The ^{14}C dates on 6 samples from CR1. Three samples (74-76 cm, 76-78 cm and 78-80 cm height) are a mix of two 1 cm-thick slices

Height (cm)	^{14}C date	Uncertainty	Calibrated ages Oxcal (Bronk Ramsey, 2001) version 3.10 (2005)	Calibrated ages Bpeat wiggle-matching (Blaauw and Christen, 2005)	Lab no.
82–83	1625	30	**AD 350–540**	–	Poz-11552
80–81	1765	30	AD 130–380	**AD 152–328**	Poz-11378
78–80	1810	60	AD 70–380	**AD 131–258**	GrN-29298
76–78	1860	50	AD 50–320	**AD 86–214**	GrN-29297
74–76	1930	40	BC 40–AD 210	**AD 19–200**	GrN-29296
10–11	4580	50	**BC 3510–3090**	–	GrN-29473

The calibrated age intervals in bold are used in the discussion.

From 70 cm to 78 cm, concentrations of both elements increase drastically (41 µg/g < Pb < 77 µg/g, 49 µg/g < Zn < 1037 µg/g). Concentrations then decrease towards 81.5 cm (Pb = 10.9 µg/g, Zn = 114 µg/g) and peak twice, at 85.5 cm (Pb = 33 µg/g, Zn = 517 µg/g) and 93.5 cm (Pb = 42 µg/g, Zn = 792 µg/g), respectively. From 98 cm to the surface, the Zn concentration decreases while that of Pb increases strongly. In CR2, Pb concentrations vary between 6.8 and 30.6 µg/g from 0 cm to 110 cm. From 111 cm, Pb values increase and reach around 600 µg/g at 123.5 cm. Zinc is below the detection limit between 10 and 95 cm, except in the bottom sample (5 µg/g). Then Zn concentrations increase from 111 cm to 123.5 cm to reach 940 µg/g at 123 cm. The Zn profile also displays three peaks at 97.5 cm (20 µg/g), 102.5 cm (53 µg/g) and 109.5 cm (76 µg/g), respectively.

4.3. Lead isotopes

Table 2 summarises results of Pb isotope analyses. In CR1 (Table 2), $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ isotopic ratios decrease from the bottom (respectively, 38.9589 ± 0.0153, 15.7012 ± 0.0057 and 19.1661 ± 0.0071) to 70.5 cm (respectively, 38.3883 ± 0.0149, 15.6268 ± 0.0057 and 18.3984 ± 0.0072). From 70.5 cm to 98.5 cm values are stable (respectively, between 38.3120 ± 0.0106 and 38.3991 ± 0.0177, between 15.6236 ± 0.0041 and 15.6288 ± 0.0068 and between 18.3299 ± 0.0053 and 18.4255 ± 0.0067) except in sample 81.5 cm which displays higher values (respectively, 38.6681 ± 0.0208, 15.6603 ± 0.0079 and 18.7404 ± 0.0081). The sample located near the surface presents the lowest values (respectively, 38.1746 ± 0.0169, 15.6117 ± 0.0067 and 18.1879 ± 0.0079).

In CR2, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ isotopic ratios decrease from 50 cm (respectively, 38.8087 ± 0.0113, 15.6872 ± 0.0036 and 19.0550 ± 0.0043) towards the surface (38.0000 ± 0.0098, 15.5952 ± 0.0039 and 18.0354 ± 0.0053).

4.4. Radiocarbon dates

Results were calibrated using *Oxcal* 3.10 (Bronk Ramsey, 2001) and four selected dates (74–76, 76–78, 78–80 and 80–81 cm) were wiggle-matched using a Bayesian approach developed for peat in the calibration tool *Bpeat* (Blaauw and Christen, 2005). The data set is coherent even if intervals are partly overlapping in the *Bpeat*-calibrated samples (Table 3). This overlap is due to (1) the continuity of sampling for ^{14}C dating and (2) the high compaction of the peat making a 1 cm-thick slice to represent several tens of years. Therefore, there is an overlap which is however chronologically ordered. For example, sample GrN29296 (74–76 cm) displays the oldest probablility age of AD 19 and the youngest probability of AD 200. The directly above sample GrN29297 (76–78 cm height) displays the oldest probablility age of AD 86 (i.e. younger than AD 19) and the youngest probability of AD 214 (i.e. younger than AD 200). The same goes between GrN29297 (76–78 cm height) and GrN29298 (78–80 cm height), and between GrN29298 (78–80 cm height) and Poz11378 (80–81 cm height). *Bcal* Bayesian approach (Buck et al., 1996) was also tested (results not reported here) but did not improve the results from *Bpeat*. Calibration using *Bpeat* provided smaller intervals. In the following discussion, these *Bpeat*-calibrated age dates will therefore be used together with the *Oxcal*-calibrated age dates of the two other samples.

5. Discussion

5.1. Environmental impact

5.1.1. Building and use of the road

Geochemical analyses provide clear documentation not only about the impact of the road on the surrounding environment, but also about one of its possible purposes, namely ore transport. Sudden increases in Si concentrations in both peat columns show a direct impact of the building and the regular use of the road. The first significant Si concentrations occur at 81.5 and 106.5 cm height, respectively in CR1 and CR2 (Fig. 3). The Al, Na and K profiles also follow the same trend. Careful observation of thin sections from these depth intervals reveals a strong contamination of the peat by numerous Revinian quartzite particles used to cobble the road. This material is therefore assumed to have been produced during the construction and the use of the road. Absolute concentration discrepancies are explained by the water stream causing preferential
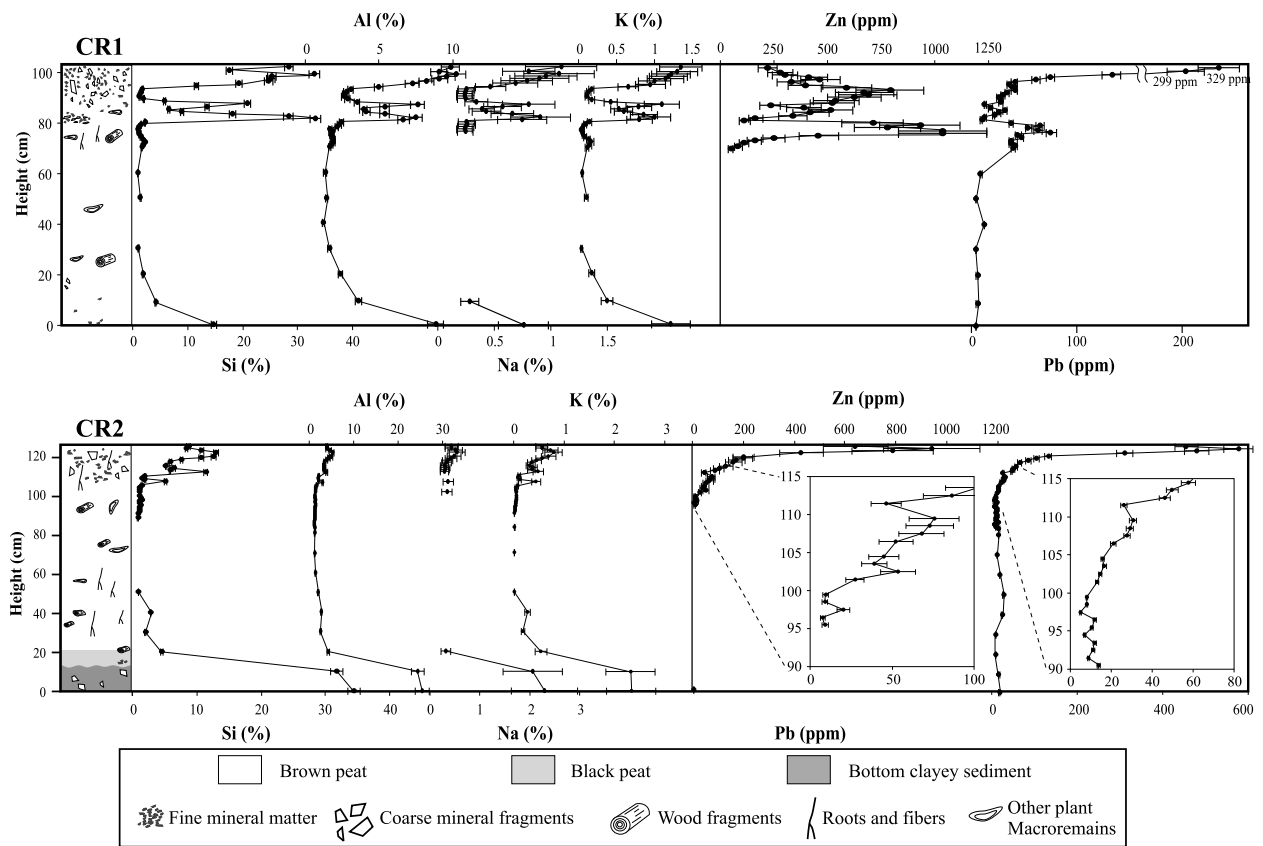
Fig. 3. Selected elemental concentrations (major + trace elements) in CR1 and CR2. Lithological information is based on microscopic observation on thin sections.
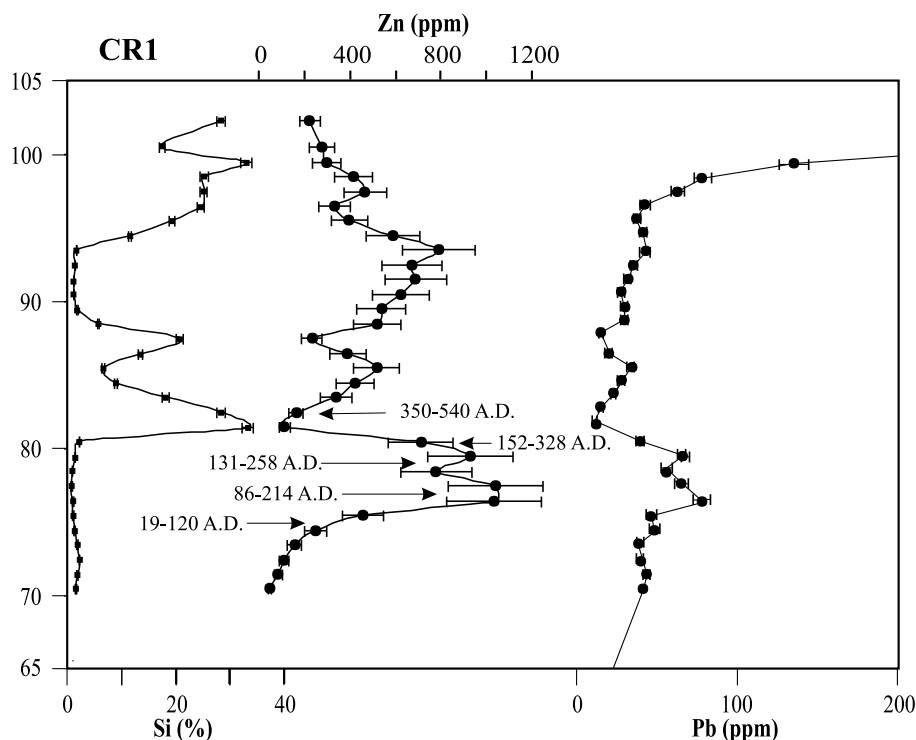
Fig. 4. The ^{14}C based chronology performed on the CR1 profile (see text for details).

accumulation to the eastern side of the road. The sample below (80.5 cm) and the sample above (82.5 cm) the Si peak were respectively dated at AD 152–328 and AD 350–540 (Fig. 4). The youngest date indicates that the road was already cobbled before AD 350–540. The oldest date must be interpreted cautiously due to the random mode of particle deposition and the lack of information about the time span between the cobbling and the first particle accumulation. However, it seems reasonable to assume that this accumulation started as soon as the cobbling was realized, i.e. during or after AD 152–328.

Higher Pb and Zn concentrations suggest a strong local pollution possibly beginning between AD 19 and AD 120 and peaking between AD 86 and AD 328. (Fig. 4). Particular levels (between 81 cm and 84 cm, and between 86 cm and 88 cm height, respectively) display low Zn and Pb content due to the important amount of quartzite fragments, increasing the Si signal and diluting the Pb and Zn concentrations. In Belgium, Pb and Zn concentrations have been previously measured in a core from a nearby ombrotrophic peat bog (De Vleeschouwer et al., 2007). This record displays quite similar Pb concentrations (40 µg/g < Pb < 100 µg/g) during the same time span, possibly reflecting the Pb pollution found here. However, Zn concentrations are often very low (8 µg/g < Zn < 35 µg/g) or below the detection limit, excepted in AD 1950 ^{210}Pb-dated peat layers where Zn peaks at 250 µg/g. This value is still 4 times less than the one found beside the cobbled road. A study carried out by Kempter (1996) in another nearby ombrotrophic bog showed no particular enrichment in Zn or Pb during the first five centuries AD. It can therefore be concluded that the peat aside the cobbled road has recorded a very local Pb and Zn pollution. Such pollution is likely to be from the transport of Pb–Zn-rich material on the road.

5.1.2. Sources of pollution

Lead isotopes are used to discriminate between possible Pb sources, especially at the highly Pb–Zn-polluted levels. Lead isotopic data define two trend lines (Fig. 5). The right (i.e. higher [206]Pb/[207]Pb and lower [208]Pb/[206]Pb ratios) trend-line is defined by the Verviers Pb–Zn ores (Pasteels et al., 1980; Cauet et al., 1982; Dejonghe, 1998) and a crustal (i.e. non anthropogenic) source. This crustal source differs significantly from the mean upper continental crust values defined by several authors (Asmerom and Jacobsen, 1993; Hemming and McLennan, 2001; Millot et al., 2004). It fits rather better with the Belgian pre-industrial background values proposed by Sonke et al. (2002) and De Vleeschouwer et al. (2007). The left trendline is defined by the Verviers Pb–Zn ores and the recent (1994-1999) urban aerosols from nearby Germany and The Netherlands (Bollhöfer and Rosman, 2001).

In section CR1, samples from the lower part of the peat column (9.5 cm and 40.5 cm) show that the main inorganic inputs to these layers derive from the regional soil and rock erosion as their isotopic signatures are in good agreement with the Belgian pre-industrial background. Samples situated in the highly Zn- and Pb-contaminated interval (i.e. 60–99 cm, *n* = 8) are all centred upon the isotopic field of the Pb–Zn ores of the Verviers syncline, located 15 km northwards of the excavation site. This observation is in good agreement with the hypothesis that this road was used for ore transport. The sample from 81.5 cm shows a lower [208]Pb/[206]Pb ratio and a higher [206]Pb/[207]Pb ratio that can be explained by the occurrence of quartzite fragments
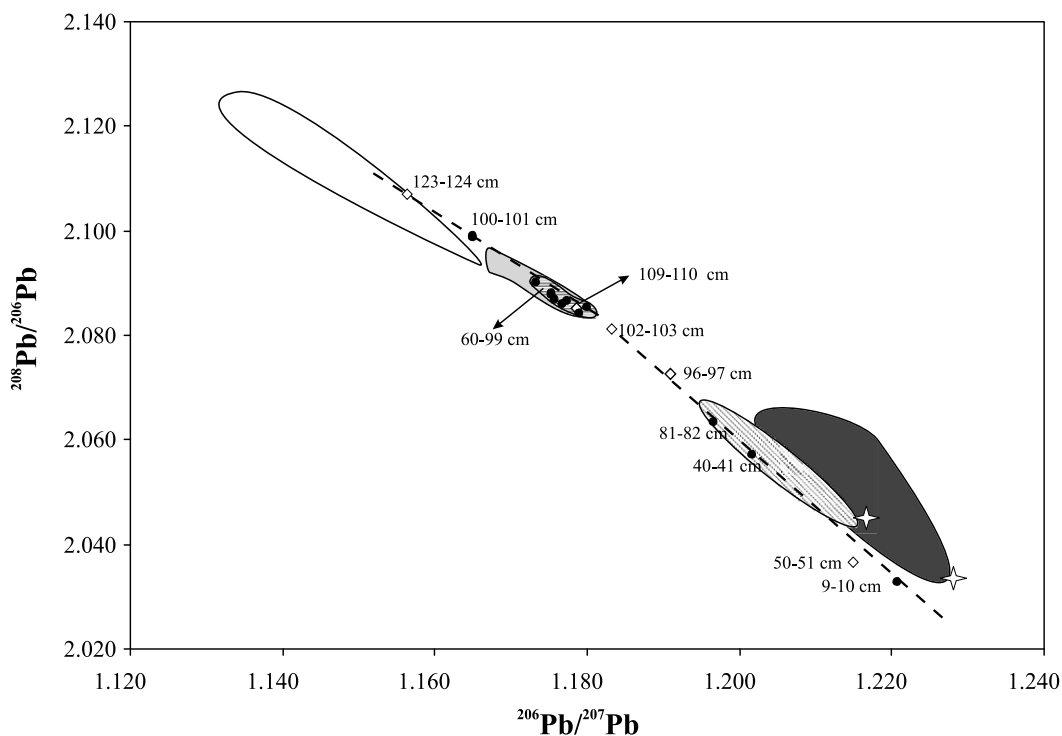
Fig. 5. $^{208}Pb/^{206}Pb$ vs. $^{206}Pb/^{207}Pb$ isotopic biplot. Black dots and white diamonds are samples from CR1 and CR2, respectively. The error ranges are smaller than the symbols. Horizontal dashed field encompasses samples of CR1 between 60–99 cm height (i.e. eight samples with very high Zn and high Pb contents). White field (13 points) represents the modern aerosols from Belgium, nearby Germany and The Netherlands (Bollhöfer and Rosman, 2001). Light grey field (18 points) represents the nearby (less than 10 km) Pb–Zn ores from the Verviers syncline (Cauet et al., 1982; Dejonghe, 1998). Dark grey field (4 points) represents the Upper Continental Crust (Millot et al., 2004; Hemming and McLennan, 2001; Grousset et al., 1994; Asmerom and Jacobsen, 1993). Oblique dashed area represents the Belgian pre-industrial background proposed by Sonke et al. (2002). White stars are the local pre-industrial background in Belgian peat proposed by De Vleeschouwer et al. (2007).

(with a crustal Pb isotopic signature). The uppermost peat sample (100.5 cm) displays a signature in good agreement with modern aerosols.

Samples from CR2 display the same trend. Samples at 50.5 cm and 96.5 cm height show isotopic ratios similar to the Belgian pre-industrial background. Samples at 102.5 cm and 109.5 cm (i.e. samples with Pb–Zn contamination) display Pb isotopic ratios in good agreement with the Verviers Pb–Zn ores field. This again favours the hypothesis of ore transport on the road. The uppermost (i.e. recent) sample shows Pb isotopic ratios similar to modern aerosols.

5.1.3. Post-depositional mobility of contaminant metals

The archaeological findings presented here are for the area located down slope on a peatland where surface run-off was observed during the sampling. Punctual inorganic inputs are thus likely. The question of whether these phenomena could affect the geochemistry of the peat must therefore be assessed.

Investigations of pollution history in peat are usually achieved in ombrotophic bogs, (e.g. West et al., 1997; Shotyk et al., 1998; Martínez-Cortizas et al., 1997, 2002; Nieminen et al., 2002; Mighall et al., 2002; Le Roux et al., 2004a; De Vleeschouwer et al., 2007). However, a few studies have demonstrated the potential of minerotrophic peat to record past geochemical changes, either natural or anthropogenic, (e.g. West et al., 1997; Shotyk, 2002; Baron et al., 2005). In addition, the knowledge of local pol-

lution sources is of high interest to accurately reconstruct global contaminant metal pollution (Baron et al., 2005). Lead has been used as an atmospheric pollution tracer in several geochemical studies in peat bogs, (e.g. Martínez-Cortizas et al., 1997; Shotyk et al., 1998; Mighall et al., 2002). Most authors have considered Pb as being immobile in ombrotophic peat (Shotyk et al., 1998, 2000; Mighall et al., 2002; Martínez-Cortizas et al., 1997; Jones and Hao, 1993) and in some cases also in minerotrophic peat (Shotyk, 2002). Elevated amounts of Pb were therefore considered as contemporaneous of the level that records it.

Zinc mobility in peat is however still under debate. Its distribution is linked, among others, to bioaccumulation, water table fluctuation and pH conditions (Shotyk, 1988). Zinc distribution can also be linked to the type of vegetation as different parts of the same plant can concentrate Zn in different ways (Livett et al., 1979). Zinc seems to be more mobile than Pb and therefore more likely to be leached (Livett et al., 1979). Recently, Weiss et al. (2007) have demonstrated Zn mobility in peat cores from Finnish natural and polluted sites. They suggest that Zn is affected by multiple biogeochemical processes. They therefore recommand that these processes have to be taken into account before using Zn as a tracer of pollution. However, even more recently, Sonke et al. (2008) have found deep Zn and Pb peaks in organic sediment from Belgium. They correlate these peaks with the well known history of local Zn and Pb smelting. They concluded, conversely to Weiss et al. (2007) that Zn is not subject to vertical migration.

Despite this debate, Zn is regularly used to trace pollution history linked to mining activities, but explanations depend on the context. Mighall et al. (2002) studied pollution history linked to the Pb–Zn ore mining in a blanket peat. In their study, Pb concentrations were in good agreement with the mining activities history. Zinc concentrations could therefore also reflect mining activities. However, the authors suggested caution as the data showed possible re-mobilization which may have blured the actual boundaries of deposition. West et al. (1997) studied the pollution history linked to the mining of various (including Pb and Zn) ores. Concentration patterns and enrichment factors were interpreted as reflecting the regional pollution but elemental mobility was not discussed. Jones and Hao (1993) measured major and trace element concentrations in peat. The resulting statistical treatment showed that Zn and Pb were strongly correlated. They showed the same pattern, most likely indicating anthropogenic impact, and were at their initial level of deposition. In this peat bog, Zn was not mobilized. From this brief overview, it could be concluded that Pb is the best indicator and that Zn can be a good second element to trace past pollution.

In the peat columns studied here, post-depositional mobilization of Pb and Zn is unlikely. Indeed, samples with high Zn and Pb content (CR1 between 60 and 99 cm excluding the 80.5 cm sample) fit within a narrow range of Pb isotopic ratios (Fig. 5). Moreover, all these ratios fall within the field of Verviers syncline Pb–Zn ores. If any mobilization of Pb from samples above the first Si peak

had occurred, a progressive decrease of Pb and Zn content down-core would have been expected, together with a mixed Pb isotopic signature between Verviers ores and Belgian pre-industrial background. This is clearly not observed here. The stability of Pb isotopic ratios has already been used by Baron et al. (2005) as an argument to attest to Pb immobility in fen (i.e. minerotrophic) peat in France. Moreover, Pb and Zn display a high degree of correlation ($R^2 = 0.88$) in CR1 between 76 and 94 cm. This suggests a common origin and deposition process for Pb and Zn. It also favours the immobility of both elements.

5.1.4. Early mining activities and purpose of the road

In conjunction with more conventional road uses, e.g. civilian and military movement, trade routes, etc. geochemical investigations carried out here provide clear evidence for the road being used for the transport of local Pb and Zn ores from the Verviers syncline. These ores are known to have been mined from the 14th century (Dejonghe and Ladeuze, 1993). Although the extraction of Pb–Zn ores of the Verviers syncline during Roman or even Celtic times is mentioned in the literature (Dejonghe, 1998; Ladeuze et al., 1991), it only refers to Plini (1953) who vaguely mentioned that an ore called *Cadmia* was extracted in "*Germania*". No more precise location was mentioned. In addition, there is no archaeological, palynological or geochemical evidence attesting to an early mining of Pb and Zn or smelting activities in East Belgium. However, various archaeological artefacts found in and around the Hautes Fagnes area (Gilot, 1977; Dumont, 1979, 1980;
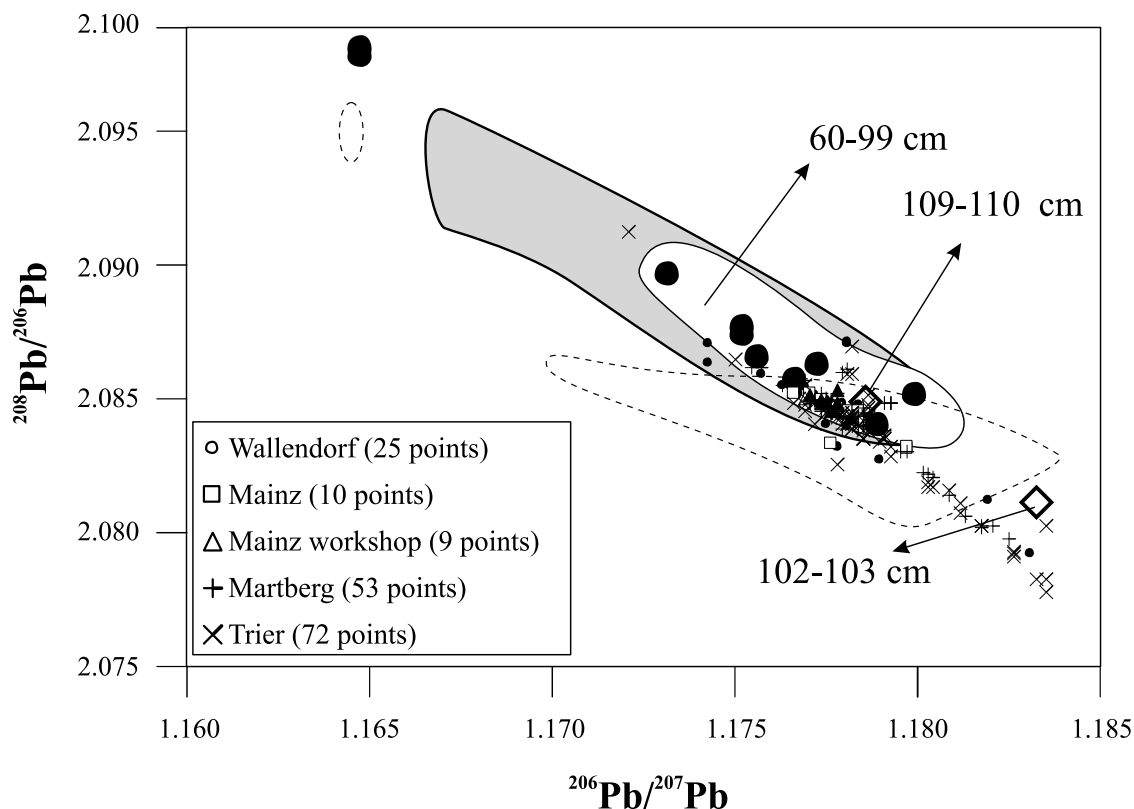
Fig. 6. ^{208}Pb/^{206}Pb *vs.* ^{206}Pb/^{207}Pb isotopic biplot showing data from CR1 (black circles) and CR2 (white diamonds) compared to isotopic signatures of Verviers Pb–Zn ores (grey shaded area), German Eifel Pb ores (dashed line areas, after Durali-Mueller et al., 2007) and German Pb artefacts (enclosed symbols) from various locations (after Durali-Mueller et al., 2007). The error ranges are smaller than the symbols.

Remy, 1981; Cauuet, 2005), like alluvial mounds remaining from Au prospecting (ranging from BC 120 – AD 140 to BC 400– 200), graves, coins and remains of cities and villages, attest to an early occupation. The increasing anthropogenic impact of local population is also recorded by increases in *Plantago lanceolata* and cereal pollen in dated peat cores (Damblon, 1994; Gotjé et al., 1990).

The dataset presented here, especially the strong Pb isotopic similarity between contaminated peat layers and local ores, may therefore represent the first evidence for ore mining during early Roman to Merovingian times. Together with the high elemental concentrations in Pb and Zn, it can also be assumed that these ores were transported on the road across the Hautes Fagnes.

As mentioned in Section 1, Trier has already been considered to be the destination of the road. This city is known to have been a Late Roman capital, a major centre of coinage (Sutherland, 1967), a key crossroad and a fluvial point. Numerous Pb artefacts have been found in archaeological excavations in this area. Several of them have been investigated for their Pb isotopic ratios by Durali-Mueller et al. (2007), together with artefacts from other nearby locations (Mainz, Wallendorf, Martberg) west to the Rhine river. Lead isotopic signatures of these artefacts correspond to the signatures of German Pb ores. However, Pb isotopic ra-

tios of these artefacts also show an important overlap with the data presented here and with the isotopic signatures of Verviers Pb–Zn ores (Fig. 6). As strong Pb–Zn pollution is encountered in the peat near the road during late Roman times, Belgian Pb–Zn ores must have therefore been mined and transported towards Germany and may also have used in the fabrication of various Pb objects in the Trier area. Moreover, several Cu ore deposits were also found around this city (Fig. 7). Romans are known to have made some of their coins and other objects with brass (Halleux, 2005), which is a Zn–Cu alloy. Assembling all this information, it seems legitimate to propose that Zn and Pb ores were mined in the Verviers syncline and transported through the Hautes Fagnes towards Trier.

As Pb and Zn peaks are considered to reflect ore transport, the time interval of AD 86–328 provides a minimum age-date for this transport and its impact on the peat. It fits clearly within Roman times (see Fig. 4) and is in good agreement with the suggestion developed above. However, the road was probably not cobbled before AD 152–328 (see Section 5.1.1). Such a discrepancy between ore transport (attested by the first Pb–Zn contamination) and the cobbling (attested by the first Si contamination) may be due either to the random quartzite deposition mode, or to an effective ore transport on an early non-cobbled road. How-
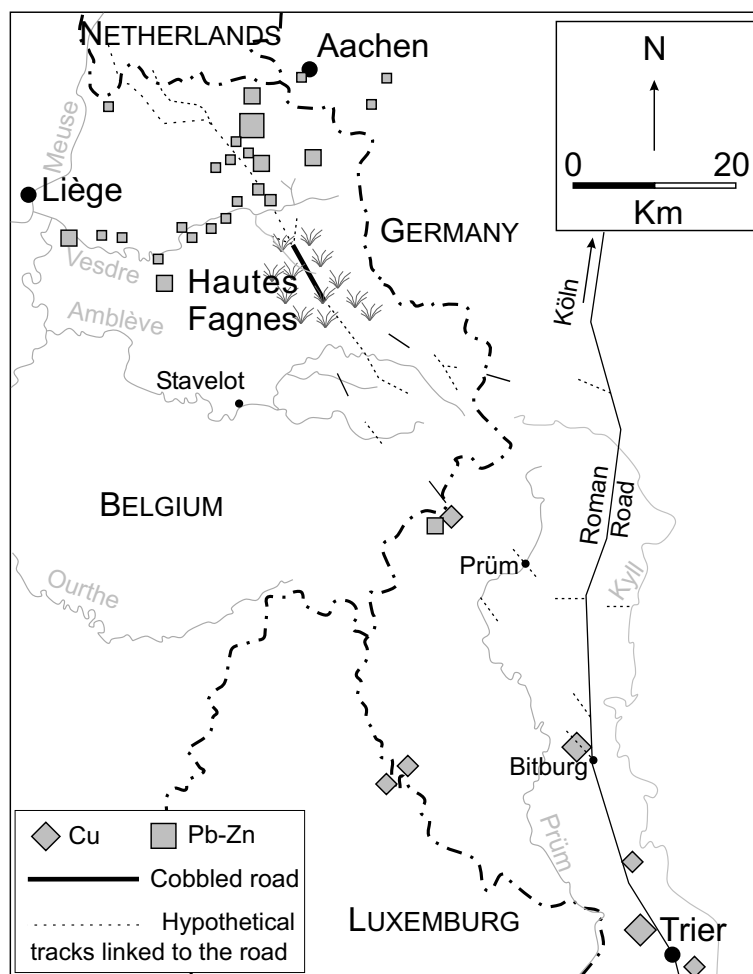

Fig. 7. The cobbled road in the historical and geographical context. Principal Cu, Zn and Pb ores are also reported. (modified after Corbiau, 1981b and Permingeat et al., 1964).

ever, due to the large overlap between each age interval, the present data do not allow distinguishing between the two hypotheses. Archaeological investigations are still ongoing and might provide additional support for the chronology of the road construction.

6. Conclusions

The geochemical investigation of peat columns collected alongside an ancient cobbled road in SE-Belgium documents the road's impact on the local environment but also provides new perspectives for future geoarchaeological research in general.

At a local scale, the investigation shows that: (1) high contents of Si, Al, Na and K reflect early peat pollution when cobbles and finer material were eroded during the building and use of the road; (2) the precise date of the road cobbling is still unclear. However, the occurrence of mineral particles in the peat evidence the presence of the cobbled road just before AD 350–540 and probably not before AD 152–328; (3) the mineral pollution is also preceded by a Pb–Zn pollution. Lead isotopic ratios reveal that the road served, among other things, to transport Pb–Zn ores from nearby Verviers Pb–Zn ore deposits.

It is therefore concluded that the onset of ore transport started during the early Roman times. This time frame is in good agreement with other archaeological evidence of Roman activities in the area. Geochemical findings from this study also support that the Belgian Pb–Zn ores from Verviers syncline could have been mined as early as during early Roman times. It is therefore suggested that in similar studies, the local isotopic source should be taken into consideration when dealing with past pollution reconstruction for this time period.

More generally, Zn is apparently immobile in the profiles, due to its similar behaviour to Pb. Moreover, the preservation of an ancient road and its environmental impact in a peatland also provides the unique opportunity to directly trace palaeopollution close to an archaeological site using geochemistry. More interest should be taken on similar ancient roads or other archaeological sites, in terms of their possible impact on the immediate environment.

This study demonstrates how geochemistry can be successfully applied to an archaeological problem. Geochemistry can indeed be used as a tool to investigate small scale changes in peat columns and to provide conclusion in terms of ore mining, routes and trade, and therefore to give insights to solve historical problems.

Acknowledgements

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