

SPECIAL ISSUE ARTICLE

Impacts of mining and smelting activities on environment and landscape degradation—Slovenian case studies

Gorazd Žibret  | Mateja Gosar  | Miloš Miler  | Jasminka Alijagić 

Geological Survey of Slovenia, Department of Mineral Resources and Environmental Geochemistry, Ljubljana, Slovenia

Correspondence

G. Žibret, Geological Survey of Slovenia, Dimičeva ulica 14, SI-1000 Ljubljana, Slovenia.
Email: gorazd.zibret@geo-zs.si

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Abstract

This paper provides an overview of the physical and chemical landscape changes that have occurred at four reference sites due to historical mining and smelting activities within Slovenia, and their comparison with similar sites around the World. Literature review has been made with the intention to identify major pollutant sources, its dispersion control factors, and effects. The four reference sites are Idrija, with more than 500-year Hg mining and ore smelting history, the Meža Valley, also with a 500-year Pb–Zn mining and smelting history, the Celje area where Zn was smelted for 100 years and the Drava River alluvial plain, which is contaminated because of historical Pb–Zn mining upstream. Based on the comparison between the four abovementioned reference sites and similar sites around the world that are situated in different landscapes and climates, we identified major sources of contamination, which are the erosion of mine and ore processing wastes, and atmospheric emissions of metal-containing particles from smelters. In the first case, major control factors are rainfall pattern and river gradient, controlling erosion and sediment deposition patterns. In the second case, the prevailing control factors are topography and the dominant wind directions.

KEYWORDS

alluvial sediments, mining, physical and chemical degradation, soil, urban sediments

1 | INTRODUCTION

The earliest global traces of the impacts of mining on the environment, reconstructed from environmental archives, such as ice cores, peat bogs, lake and marine sediments, can be dated back to around 6500 BC. The first recorded global impact of mining was assigned to the Roman period due to Pb-smelting (Marx, Rashid, & Stromsoe, 2016). Mining and ore processing are amongst the most important impetus of human development and are regarded as the second worst global polluters today (Blacksmith Institute, 2018).

Mining and ore processing cause different changes to the landscape (Larondelle & Haase, 2012). The most important physical

landscape changes are deforestation and vegetation removal, changes in relief, deposition of mining wastes (spoil and tailings), construction of supporting infrastructure, increased erosion rates, suspended materials in surface water systems, and increased rate of soil and rock instability. Chemical changes are caused by the dispersion of extracted materials or chemical agents used in mining or ore processing (flotation, extraction, etc.), which lead to changes in the chemical composition of the natural environment. Although minerals are naturally present in the environment due to outcrop weathering and erosion, mining and related activities can produce increased levels of certain elements in the environment that exceed natural levels up to 1,000-times (Gosar, Šajn, & Teršič, 2016; Gosar & Žibret, 2011).

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Besides anthropogenic and geological factors, landscape and climate characteristics can also influence the rate and dispersion of pollutants. Dominant wind patterns control dispersion of particulate matter, precipitation rates might control material stability, and the river gradient controls erosion and deposition patterns.

The objectives of this study are to identify dominant factors, which affect the dispersion of the pollution, caused by mining and ore processing activities. This is done by comparing four historic mining sites in Slovenia with similar contaminated sites, situated in different climate and

topographic conditions (Figure 1; Table 1). Contaminant dispersion pathways and receptors are identified, and effects are linked to the landscape characteristics (i.e., topography and climate). The four reference sites are situated in the tectonically active junction between the Alps and Dinarides. The climate is warm-summer humid continental (Dfb; all climate symbols are according to the classification described in Peel et al., 2007), with the annual precipitation rates between 2,500 mm (Idrija) and 1,200 mm (Celje). Temperature inversion is the common weather phenomenon in the cooler part of the year.

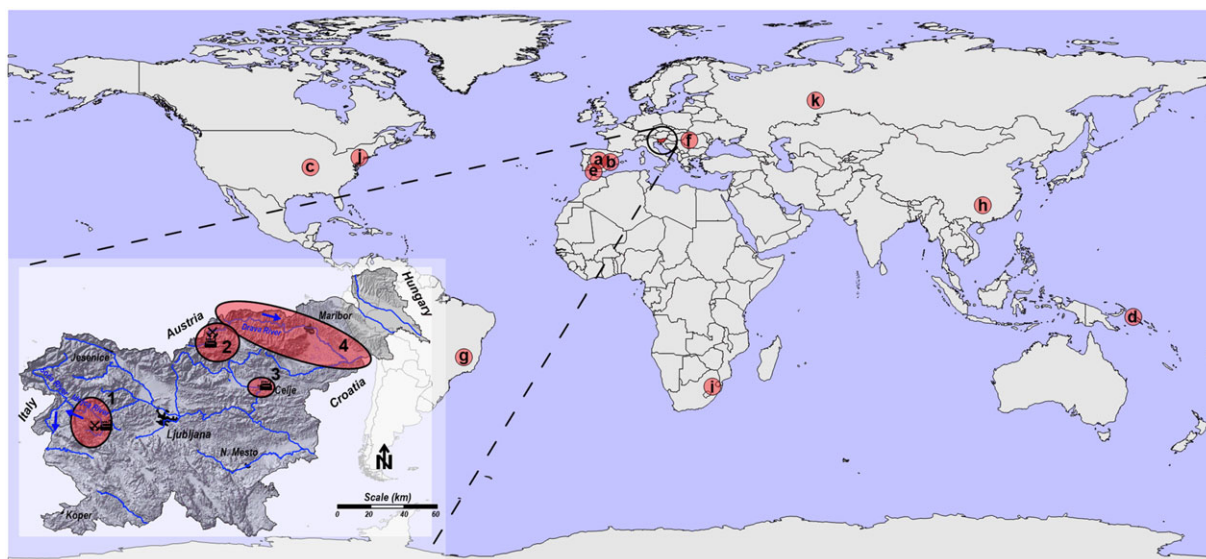


FIGURE 1 Four reference regions in Slovenia (figure in lower left), where the effects of historic mining and ore processing are clearly expressed: 1—The Idrija area; 2—The Mežica area; 3—The Celje area; 4—Drava River Valley; and similar contaminated sites around the world: a—Almadén; b—Cartagena-La Union; c—The Big River; d—Kawerong-Jaba; e—Aznalcollar; f—Baia Mare; g—Minas Gerais; h—Magu; i—eMalahleni; j—Palmerton; k—Karabash [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 1 Brief characteristics of the study areas

ID ^a	Locality	Country	Type of activity	Major contamination dispersion process ^b	Topography	Climate ^c
1	The Idrija area	SI	Hg mining, smelting	a, b, c	Valley	Dfb
2	The Mežica area	SI	Pb–Zn mining, smelting	a, b	Valley	Dfb
3	The Celje area	SI	Zn smelting	b	Basin	Dfb
4	Drava River Valley	SI	Pb mining upstream	a	Alluvial floodplains	Dfb
a	Almadén	ES	Hg mining, smelting	a, b	Flatland	BSk
b	Cartagena-La Union	ES	Pb–Zn–Fe mining	a	Flatland	BSn
c	The Big River	US	Pb mining	a	Alluvial floodplains	Dfa
d	Kawerong-Jaba	PG	Cu mining	c	Valley, alluvial floodplains	Af
e	Aznalcollar	ES	Polymetallic mining	d	Valley, alluvial floodplains	Csa
f	Baia Mare	RO	Au mining	d	Valley, alluvial floodplains	Dfb
g	Minas Gerais	BR	Fe mining	d	Valley, alluvial floodplains	Aw
h	Magu	CN	Zn smelting	b	Valley	Cfa
i	eMalahleni	ZA	Coal mining, smelting	b	Flatland	Cwb
j	Palmerton	US	Zn smelting	b	Valley	Dfa
k	Karabash	RU	Zn smelting	b	Flatland	Dfc

^aLocality code, as in Figure 1.

^bMajor contamination dispersion process: a—runoff and water erosion; b—atmospheric emissions of particulate matter; c—dumping of tailings or ore smelting wastes to the river channel; and d—tailing dam collapse.

^cKöppen climate classification codes (Peel, Finlayson, & McMahon, 2007).

Reference sites are located in Slovenia, EU (Figure 1): the Hg mining Idrija region; the Mežica Pb–Zn mining and smelting region; the town of Celje with Zn smelting history and the Drava River Valley with historical Pb mines in its catchment. In the discussion chapter, these four reference sites are compared with other similar sites (Figure 1), with the intention to detect possible influences of landscape characteristics on the contamination dispersion

2 | STUDY SITES

2.1 | Idrija Hg mine

In the town of Idrija (Figure 1), mercury ore was mined and processed for five centuries until 1995. These activities caused an unprecedented legacy of mercury contamination in the wider Idrija area (Teršič & Gosar, 2012; Gosar et al., 2016; Table 2). Sediments of the rivers Idrijca and Soča/Isonzo, which drain the contaminated area, are enriched with mercury, and they transport large amounts of Hg-loaded material towards the Gulf of Trieste in the northern Adriatic Sea. This results in the Gulf of Trieste being one of the most Hg contaminated areas in the whole Mediterranean region (Covelli, Petranich, Langone, Emili, & Acquavita, 2017; Kotnik et al., 2015). The influence of mercury originating from the Idrija ore deposit was also determined in the middle of Adriatic Sea (Foucher, Ogrinc, & Hintelmann, 2009), almost 500 km away.

A special characteristic of the town of Idrija is that it is built directly over the ore deposit and mine. The 4,000 m² large ore outcrop contains on average 3,800 mg kg⁻¹ (Mlakar & Čar, 2009). Natural migration of mercury into the wider surroundings of the deposit is insignificant because large anthropogenic impacts dominate the Idrija town (Bavec & Gosar, 2016).

Mercury was distributed throughout the surrounding areas by atmospheric emissions from ore smelting facilities. Gaseous and particulate matter emissions were the major cause of creating the geochemical halo around Idrija, where the areas of highest Hg levels in soil are limited to the valley floor, and not on the surrounding hills (Gosar et al., 2006). Models of the spatial distribution of mercury in Slovenian soils show that the influence of atmospheric Hg emissions from Idrija can be detected on a regional scale (Gosar et al., 2016; Figure 2).

Atmospheric mercury levels in Idrija were reported to be extraordinarily high at the times of ore smelting. In 1971–1972, mercury concentrations up to 30,000 ng m⁻³ were reported, and daily atmospheric mercury emissions were estimated at 20 kg (Kavčič, 1974). Measurements in 1994 (Gosar, Pirc, & Bidovec, 1997) showed much lower but still rather high mercury concentrations. Concentrations above 300 ng Hg m⁻³ were observed near the smelting plant and mine ventilation shaft. The results of various studies showed rapid variation of mercury concentrations in air depending mostly upon changing weather conditions (Gosar, Pirc, & Bidovec, 1997; Kotnik, Horvat, & Dizdarevič, 2005). After the production of mercury stopped in 1995, the conditions improved (Kotnik et al., 2005).

Ore smelting waste dumps were found to be the major source of river sediment contamination. Since 1652, when the first smelter building was built in Idrija, the mercury rich waste products were

deposited along the Idrijca River. The main reason for the complex spatial distribution of smelting residue dumps in Idrija and its surroundings are changes in smelting techniques over the centuries, continuously increasing quantities of processed ore accompanied by decreasing mercury content. Čar (1998) defined the locations of historic waste dumps containing mercury. Hg-binding forms in these dumps depend on the efficiency of the smelting technique and the predominant Hg species in the processed ore. In older dumps (deposited in 18th and 19th centuries), the predominant Hg species is cinnabar (crystallised Mercury (II) sulphide, HgS), whereas in dumps of the 20th century the amount of cinnabar in the material decreased due to the higher efficiency of the smelting process and the use of ore containing mostly native Hg. Leaching tests showed that although lower total Hg concentrations are found in the younger dumps, their long-term risk potential is higher than that of the older ones, which contain mostly immobile cinnabar (Biester et al., 1999).

From 1868 to 1977, most smelting remains were dumped directly into the Idrijca River. Because of its high average gradient of 4.35 m km⁻¹ between Idrija and Most na Soči, the Idrijca River carried the material downstream to the Soča River and Adriatic Sea. For this reason, the Idrijca River sediments have high mercury contents (Biester et al., 2000; Gosar, 2008; Gosar, Pirc, Šajn, et al., 1997; Gosar & Žibret, 2011). During high water levels, Hg-rich material was deposited on the floodplains in the lower part of the Idrijca and Soča Valley (Figure 3). These sediments represent a large accumulation of Hg-enriched sediments (Gosar & Žibret, 2011). It was estimated that about 2,000 tons of mercury are stored in the Idrijca River alluvial sediments (Žibret & Gosar, 2006).

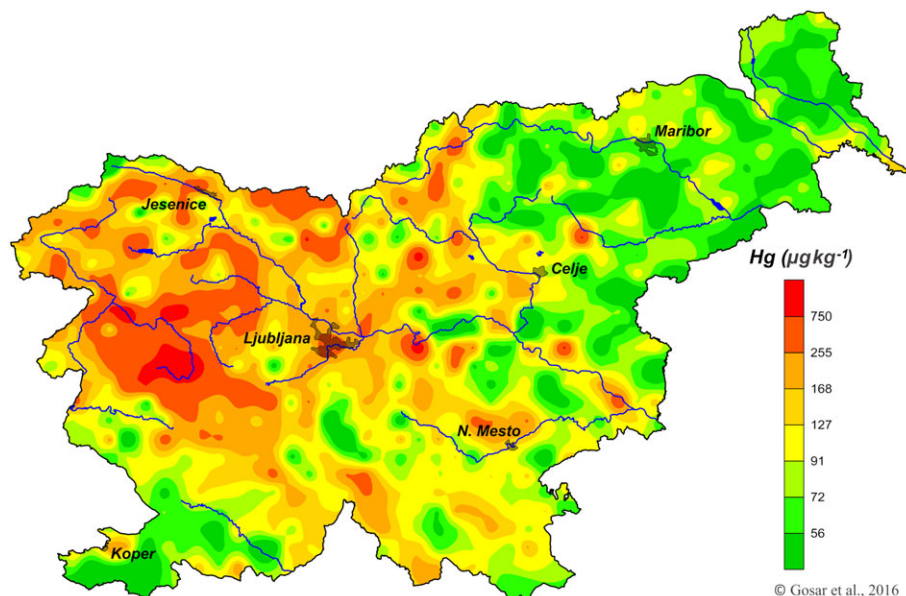
During the first 150 years of mercury production, numerous smaller ore smelting sites existed in the woods around Idrija. Detailed geochemical investigation of these sites proved their significance for environmental contamination (Teršič et al., 2014). The soils at these sites are highly contaminated with Hg, showing Hg loads into the percentage range. The investigations proved that Hg-loaded materials are still present at the smelting sites and are being intensively eroded and transported downstream during periods of high water levels. Investigation of the Hg load in the Idrijca River sediments shows that the historic smelting sites still remain an important source of Hg-contaminated material today and one of the primary concerns for persistent Hg release into the aquatic ecosystem (Gosar & Teršič, 2015; Teršič et al., 2014).

2.2 | Mežica Pb–Zn mine

The Mežica Pb–Zn underground mine lies in the upper part of narrow Meža Valley, located between steep hills and mountains at 480 m asl and extends over an area of 64 km². The Meža Valley has been characterised by more than 500 years of Pb–Zn mining and Pb-smelting. About 1 million tons of Pb, 0.5 million tons of Zn, and minor amounts of molybdenum were produced from 19 million tons of Pb–Zn ore and 80,000 tons of wulfenite (Rečnik, Zavašnik, & Fajmut-Štručl, 2014). Until the first half of the 20th century, the environmental impacts were limited to the areas surrounding the small ore processing facilities. After the introduction of rotary furnaces in 1958, the production increased dramatically. However, this plant was also

TABLE 2 Identified sources, pathways, and receptors in the case of the Idrija Hg mine, and its effects on the landscapes and possible effects on humans

Source	Contaminant	Pathway	Receptor	Range and influenced area	Changes in landscapes	Change type	Impacts on humans	References
Ore outcrop	Hg in air, soil, sediments	Air, soil, sediments	Vegetation, biota	Approximately 4,000 m ²	Contaminated soil, sediments	Chemical	Potential intake via air, soil, crops	Malakar & Čar, 2009
Hg smelting factories (two locations)	Hg contaminated air emissions, dust particles, and gaseous Hg(O) in fine and ultrafine aerosols	Air Precipitation Runoff	Vegetation and buildings All surfaces: soil, streets, vegetation Alluvial sediments	Approximately 19 km ² critically polluted Influenced area > 160 km ²		Chemical Physical	Potential intake of Hg via contaminated soil	Gosar, Pirc, & Bidovec, 1997; Gosar, Šajn, & Biester, 2006; Gosar et al., 2016
Smelting waste heaps	Hg contaminated sediment and soil	Erosion of waste heaps	Aquatic ecosystems and groundwater Fish	>100 km—alluvial and marine sediments	Contaminated soil	Chemical	Potential intake of Hg via contaminated sediments and floodplain soil Hg in river fishes Hg in marine biota which is also food for people	Biester, Gosar, & Müller, 1999; Gosar et al., 1997; Žibret & Gosar, 2006; Gosar, 2008; Malakar & Čar, 2009
Dumping of smelting waste into Idrija (ending in 1977)	Hg contaminated sediment, particulate matter in rivers	Suspended particulate matter in rivers	Alluvial sediments Marine sediments	>100 km—alluvial and marine sediments in Gulf of Trieste	Changes in river regime and ecology Polluted marine sediments	Physical Chemical	Hg in marine biota which is also food for people	All literature above and the following: Biester, Gosar, & Covelli, 2000; Gosar & Žibret, 2011; Kotnik et al., 2015; Covelli et al., 2016
Historic smelting sites	Hg contaminated soil, sediments, air	Air, soil, sediment, water	Living beings at this locations Vegetation Groundwater Streams draining these areas	Approximately 21 × 1–2 km ² Stream sediments, draining these areas	Soil Sediments Water contamination	Chemical	Potential intake of Hg from contaminated soil and sediments	Teršič, Biester, & Gosar, 2014; Gosar & Teršič, 2015



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FIGURE 2 Distribution of mercury in Slovenian soil based on soil sampling in a 5×5 km grid confirms the significant contribution of mining and ore processing in the Idrija mercury mine to the Hg contents in soil at the regional scale (after Gosar et al., 2016) [Colour figure can be viewed at wileyonlinelibrary.com]



FIGURE 3 Idrija River Valley is very picturesque, but sediments at that particular location contain more than 300 mg kg^{-1} Hg in fine fraction ($<0.040 \text{ mm}$; photo: M. Gosar) [Colour figure can be viewed at wileyonlinelibrary.com]

used for recycling of Pb-based waste and Pb-acid batteries. Following closure of the Pb–Zn mine in 1995, only recycling of Pb-containing waste and production of refined Pb and Pb-alloys continued at the site.

The past Pb-ore smelting was considered the major source of SO_2 and potentially toxic elements (PTEs) in the Meža Valley. The highest airborne concentrations of SO_2 were measured in 1977 in the uppermost part of the Meža Valley where they reached 235 µg m^{-3} (Kladnik, 2009). Afterwards, it gradually decreased to below 20 µg m^{-3} in 2001. Because the upper Meža Valley is very narrow and located between steep hills and mountains, aerial transport plays an important role in the dispersion of gaseous and solid pollutants up and downstream the valley. High SO_2 emissions and associated acid rain in the past severely damaged forests, mostly coniferous, in the area around the Pb-ore smelter below 800 m asl (Figure 4). The extent of the damaged forest in the late 1970s reached 3,200 ha, of which 280 ha were completely deforested. As a consequence, soil from deforested areas was completely eroded. Although primary Pb-ore smelting was abandoned and the emissions of SO_2 were reduced, about 651 ha of forest (Figure 5a) still remains damaged (Kladnik, 2009; Table 3).

The highest annual average Pb concentration in airborne particulate matter in upper Meža Valley was measured in 1972, reaching up to 37 µg m^{-3} , which caused contamination of various environmental compartments (Table 3), mostly soil, indoor dust and partly stream sediments. Studies documented that topsoil around the past Pb smelter in the Meža Valley is polluted, but contamination is more or less limited to the valley, and does not extend much to the surrounding hills (Šajn, 2006; Šajn & Gosar, 2004; Vreča et al., 2001), thus covering a surface of about 23 km^2 (Figure 5b). The topsoil in upper Meža Valley contains high mean levels of Pb (410 mg kg^{-1}) and Zn (400 mg kg^{-1} ; Šajn & Gosar, 2004) whereas in garden soils even higher mean levels of Pb (over $2,300 \text{ mg kg}^{-1}$) and Zn (over $1,500 \text{ mg kg}^{-1}$) were measured. Due to high contents of PTEs, soils are degraded, rendering them less fertile and unsuitable for cultivation of crops (Finžgar & Leštan, 2008). Investigation of attic dust, which is a good indicator of past air pollution and dispersion of pollutants, revealed that the influences of Pb-smelting and Pb-recycling cover about 30 km^2



FIGURE 4 Degraded landscape around Pb mine and smelter in Žerjav, Meža Valley, photo taken around 1990 (unknown author) [Colour figure can be viewed at wileyonlinelibrary.com]

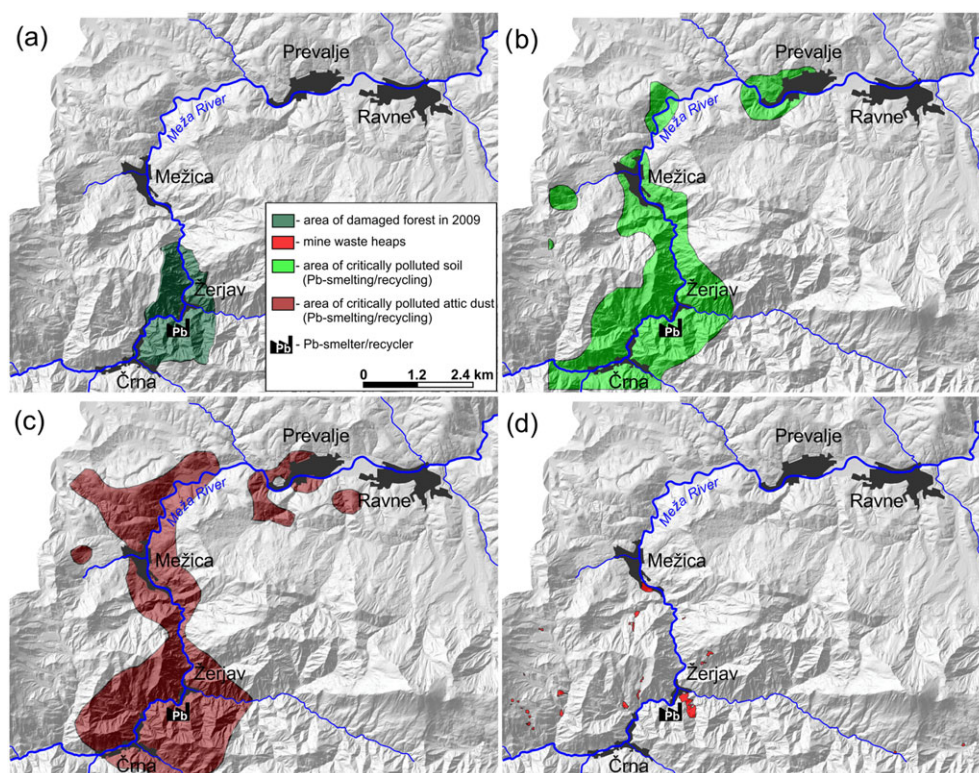


FIGURE 5 Polluted areas in Mežica (after Šajn & Gosar, 2004; Šajn, 2006; Kladnik, 2009, and references therein): (a) area of damaged forest in 2009 due to past Pb-smelting; (b) area of critically polluted soil due to Pb–Zn mining, Pb-smelting/recycling between Črna and Prevalje; (c) area of critically polluted attic dust due to Pb-smelting/recycling (Črna–Prevalje); (d) areas covered by Pb–Zn mine spoil heaps [Colour figure can be viewed at wileyonlinelibrary.com]

(Figure 5c). Mean levels of Pb, Zn, and Cd in attic dust were 3,700, 1,100, and 15 mg kg⁻¹, respectively (Šajn, 2006).

Due to exposure to contaminated crops, soil, and indoor dust, inhabitants of the upper Meža Valley had extremely high Pb blood concentrations, reaching a median value of over 400 µg l⁻¹ before 1978. In 2007, the median Pb levels in blood of children were still above 110 µg l⁻¹ (Jež & Leštan, 2015), which exceeded maximum permitted value for Pb concentration in blood, set at 100 µg l⁻¹ (OG RS, 2007).

Recent investigation of snow deposits revealed that about 85% of PTE-bearing particles derive from present-day Pb-recycling (Miller & Gosar, 2013). However, they represent only about 8.5% of all deposited particles. Pb-recycling has therefore little influence on the environmental pollution, and historic environmental burdens are still the important sources of Pb-enriched particles in the atmosphere.

Approximately 7.4 million m³ of mine waste are dispersed over an area of about 60 km² along the upper Meža Valley. Spoils are present as waste heaps in narrow valleys, on steep slopes above small streams

TABLE 3 Identified sources, pathways, and receptors in the case of the Mežica Pb–Zn mine, Pb–Zn smelter and ironworks, and their effects on the landscapes and possible effects on humans

Source	Contaminant	Pathway	Receptor	Range and influenced area	Changes in landscapes	Change type	Impacts on humans	Reference
Past Pb-ore smelting	SO ₂	Air	Vegetation	Approximately 30 km ²	Air pollution	Physical	Respiratory problems	Vreča, Pirc, & Šajin, 2001; Šajin, 2001; Šajin & Gosar, 2004; Šajin, 2006; Kladnik, 2009; Miller & Gosar, 2013; Jež & Leštan, 2015
Present-day Pb-acid battery	Dust enriched with Pb, Zn, Mo, Cd, As, Ag, Cu, Sb, Sn, Tl	Precipitation runoff	Soil	critically polluted (attic dust)	Historical deforestation	Chemical	PTE intake via food, soil, dust ingestion	
Pb-containing waste recycling			Snow deposits	Damaged forest (3,200 ha late 1970s; 651 ha in 2009)	Soil contamination		High metal levels in blood	
			Interiors of buildings	Complete deforestation (280 ha in 1972)	Road dust			
			Stream sediments		Household dust contamination			
					Changes in biological properties of soil			
Mine and ore processing waste heaps and tailings	Mine waste material and flotation tailings containing Pb, Zn, Cd, As, Mo	Waste material remobilisation (sliding, erosion, washing out)	Surface water	Waste heaps covering a total surface of approximately 0.5 km ² (7.4 million m ³ of contaminated material)	Changes in geomorphology	Physical	Landslide hazard	Šajin & Gosar, 2004; Budkovič, Šajin, Gosar, & Fux, 2006; Kladnik, 2009; Miller & Gosar, 2012; Miller & Gosar, 2013
		Soil dispersion from spoil heap surfaces	Stream sediments	affected area	Changes in stream flow regime	Chemical	Potential PTE intake via contaminated soil, sediment, fish	
		Dispersion of contaminated materials by construction works	Soil	Stream sediments contaminated in the length of approximately 30 km	Contamination of surface water			
		Direct discharge into streams (flotation tailings)			Influence on stream biota			
					Sediment contamination			
					Soil contamination			

Note. PTE: potentially toxic element.

and in abandoned mine shafts covering a total area of approximately 0.5 km² (Budkovič et al., 2006; Figure 5d). Some mine spoil heaps are considered unstable due to their surface topography and the underlying ground and are also subjected to significant runoff erosion and remobilisation of waste material.

Within spoil heaps mine waste material is chemically relatively stable. However, its washing out into streams may cause leaching and mobilisation of PTEs into the fluvial environment. Investigation of stream sediments in the Meža River Valley (Gosar & Miler, 2011; Miler & Gosar, 2012) showed that stream sediments are contaminated for a length of approximately 30 km downstream, mainly due to the steep stream gradient, which causes erosion of the polluted sediments. The Meža River sediments contain high median values of Pb (1,100 mg kg⁻¹), Zn (1,240 mg kg⁻¹), Cd (7 mg kg⁻¹), As (13 mg kg⁻¹), and Mo (23 mg kg⁻¹).

Between 1914 and 1979, 150,000 tons of flotation tailings were discharged every year directly into Meža River (Kladnik, 2009). As a consequence, the water in the Meža River contained over 200 mg l⁻¹ Pb, which completely destroyed all stream biota. After 1979, flotation tailings were deposited in mine shafts, and the water quality began to improve.

2.3 | Celje Zn smelter

Celje is a central Slovenian town (Figure 1) of approximately 50,000 inhabitants that is a well-known historic industrial centre. Smelting of imported Zn ore occurred between 1873 to 1970. The consequence of this is that Celje had major problems with air pollution, mainly because of SO₂ and PTE-containing dust emissions from the Zn-smelting plant. The first documented evidence of environmental contamination within the Celje region is from 1933. A report from the town forestry office states that about two-thirds of all trees in the area were badly damaged (Domitrovič-Uranjek, 1990). Between October 1967 and September 1968, 19 days had an average daily SO₂ concentration above 500 µg m⁻³, whereas maximum hourly SO₂ concentrations exceeded 2,000 µg m⁻³. The survey also detected high content of Pb (8.3 µg m⁻³) and Zn (up to 58.1 µg m⁻³) in the air. Daily averages in the vicinity of plant reached even 2–5 mg m⁻³ between the years 1973 and 1974 (Domitrovič-Uranjek, 1990; Resman, Verhovnik, & Hrašovec, 1975). The forestry survey from 1972 showed that forest damage covered an area of 142 km², and within this area, 2.5 km² of the forest had been completely destroyed by atmospheric pollution and associated acid rain (Špes, 1978). In 1980s, an ecological restoration plan of Celje basin took place, as a result of increased environmental awareness in Slovenia (Polajnar-Horvat, Smrekar, & Zorn,

2014). Decrease of airborne SO₂ concentrations between 1981 and 1989 was partly the result of stricter environmental controls (Leskošek, Mašat, Eržen, & Uršič, 1998), enforced by the restoration plan, and partly due to the economic crisis at the end of 1980s.

The study of Lobnik et al. (1989) revealed a geochemical Pb, Zn, Cd, and As anomaly covering an area of 32 km² within the densely populated town of Celje. The urban sediment study done within the territory of Slovenia by Šajn (1999) revealed that the levels of Zn, Cd, and Pb in urban dusts (house, attic, and street dust) of Celje town exceeded average Slovenian urban levels by more than 15 times. The studies of Žibret (2002) and Šajn (2005) revealed a large anomaly (Figure 6) of increased Pb, Zn, and Cd levels in soil and attic dust, where Zn levels reached 56,000 and 8,500 mg kg⁻¹; Cd levels 456 and 59 mg kg⁻¹ and Pb levels 6,900 and 1,500 mg kg⁻¹ in attic dust and soil, respectively. The size of the area where levels of potential toxic metals exceed Slovenian permitted values is 17.7 km² and this area covers the densely populated area of Celje. Compared with other cities globally, Celje has one of the highest levels of Zn and Cd in the soil (Ajmone-Marsan & Biasioli, 2010). The study of Bešter, Lobnik, Eržen, Kastelec, and Zupan (2013) highlighted the intake of potential toxic elements into the human body by the ingestion of root vegetables (potatoes, carrot, and beetroot), grown on contaminated land.

The study of Žibret and Šajn (2008) determined that the range influenced by the historic Zn-smelting plant was between 14 and 52 km for Zn in attic dust and between 9 and 14 km for Zn in soils. The geochemical assessment of house and street dust (Žibret & Rokavec, 2010) revealed that high levels of Pb, Zn, and Cd can be principally attributed to past environmental burdens, but other sources of metals are also present (nearby Štore ironworks and the chemical plant). Despite installation of dust filters in the nearby Štore ironworks in 2005, the urban dust survey of 2008 identified a new source of PTE contaminated dust within the densely populated region: construction works on the old brownfield areas, where pyrometallurgical waste from the Zn smelting plant had been deposited (Žibret, 2012, 2013; Table 4). Further study also revealed that this waste pit represents significant future risk, as extremely high levels of pollutants were measured at this site (Voglar & Leštan, 2010).

2.4 | The Drava River Valley

The Drava River (730 km) is the fourth largest Danube tributary. The Drava drains the Southern side of the Alps to the Danube and the Black Sea. Geomorphological characteristics divide the Drava basin

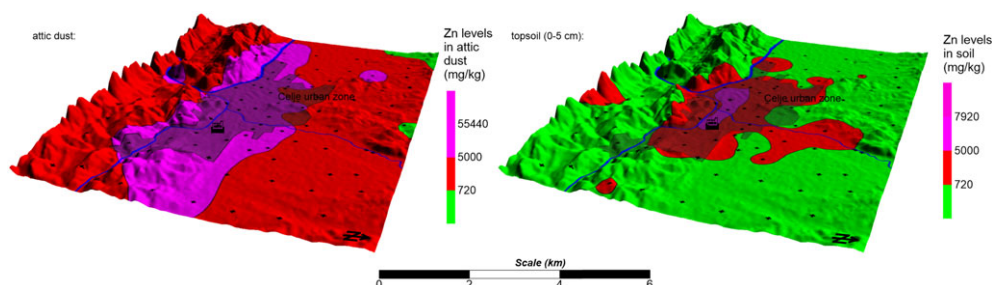


FIGURE 6 Geochemical anomaly caused by 100 years of Zn smelting in Celje. Sampling points are indicated by black crosses (data after Žibret, 2002) [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 4 Identified sources, pathways, and receptors in the case of the Celje Zn smelter and ironworks, and their effects on the landscapes and possible effects on humans

Source	Contaminant	Pathway	Receptor	Range and influenced area	Changes in landscapes	Change type	Impacts on humans	References
Zn ore smelter	SO ₂ Dust enriched with Pb, Zn, Cd, As	Air	All surfaces: soil, street dust Vegetation Alluvial sediments	Approximately 17 km ² critically polluted Influences in soil detected 15 km away	Historical deforestation Soil contamination Street dust contamination Changes in biological properties of soil	Physical Chemical	Respiratory problems PTE intake via food or soil ingestion Increased PTE content in house dust	Lobnik et al., 1989; Žibret, 2002; Šajn, 2005; Žibret & Šajn, 2008; Bešter et al., 2013
Tailings Tar dump	Pb, Zn, Cd, As Organic contaminants	Groundwater Dust and soil dispersion from tailings Dispersion of contaminated materials by construction works	Groundwater; Alluvial sediments downstream; Street and house dust nearby	Approximately 2 km ² Alluvial sediments not influenced due to river embankment	Depression fill Different flood regime	Physical Chemical	Potential intake of PTEs via contaminated soil	Žibret, 2012; Voglar & Leštan, 2010; Žibret, 2013

Note: PTE: potentially toxic element.

into two regions: Alpine and Pannonian (Šajn, Halamić, Peh, Galović, & Alijagić, 2011).

The recent floodplains reflect anthropogenic geochemical signatures, whereas the river terrace represents the historical geochemical signature of deposited material, and so can be used to differentiate between anthropogenic and natural processes as well as for the evaluation of trace element levels in soils and sediments before the industrial revolution (Halamić, Galović, & Šparica, 2003).

Historically, the Drava catchment area (42,240 km²) has been an important mining and smelting region. The industry underwent significant growth during the Middle Ages, achieving peak development in the middle of the last century. The most important Pb–Zn mines in the wider Alpine area with ancillary facilities for metal production are located in the catchment area (from approximately 100 to several hundred kilometres upstream): Cave del Predil and Salafossa in Italy, Bleiberg-Kreuth in Austria, and Mežica in Slovenia. The ore from the four Pb–Zn deposits, where the production stopped until the late 1980s, have important economic significance and have so far provided more than 8 million tons of Pb and Zn in last 700 years (Cerny, 1989).

Geochemical signatures in the soil, stream, and overbank sediments have been continuously recorded over the past two decades along the Drava Valley (Figure 7), which is an important agricultural area, as well as the source of drinking water. The floodplain area is contaminated with PTEs, especially Pb, Zn, and Cd (Halamić et al., 2003; Šajn et al., 2011), despite no obvious pollution sources are present in the region, while Pb, Zn, and Cd levels in terraces are comparable with the natural background. This means that PTE-bearing fine grained particles were transported several hundred kilometres before deposition. High levels of PTEs are found covering an area of approximately 88 km² of the floodplains, representing the largest contaminated area in Slovenia; an area larger than all other similar contaminated areas combined (Table 5).

Šajn et al. (2011) discovered that the highest levels of Pb, Zn, and Cd in soils developed on floodplains of the Alpine region are located in deeper soil horizons (maximum levels are 3,251 mg kg⁻¹, 1,152 mg kg⁻¹, and 17.4 mg kg⁻¹ for Zn, Pb, and Cd, respectively), whereas higher levels of Zn, Pb, and Cd in soils of the Pannonian region are detected in upper soil horizons (maximum levels are 2,466 mg kg⁻¹, 1,053 mg kg⁻¹, and 13.8 mg kg⁻¹ for Zn, Pb, and Cd, respectively). This shows that Drava River erosional and depositional patterns play an important role in setting the contamination distribution patterns. In the upper part of the river course (Alpine region), there is no evidence of recent sedimentation of coarse PTE-containing material, whereas the opposite trend is seen in the Pannonian region, where the resedimentation of PTE-containing fractions still occurs.

3 | DISCUSSION

Despite many potential sources of contamination, caused by mining and ore processing, the two most profound ones that are identified and investigated are erosion of mine and ore processing waste, and deposition of atmospheric particles from smelters.

The most important recognised source of contaminants, accounting the contaminant's quantities and size of contaminated area, are

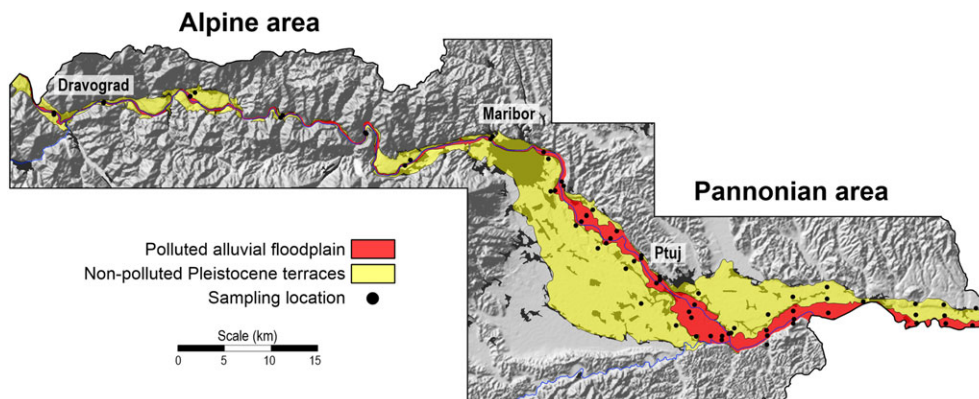


FIGURE 7 A generalised map of the Drava River alluvial plains in the Alpine and Pannonian regions in Slovenia (after Šajn et al., 2011) [Colour figure can be viewed at wileyonlinelibrary.com]

erosion and the transportation of mine waste and ore processing waste by water (Figure 8a). Contaminant receptors are alluvial and marine sediments, as is in the case of the Idrija and Mežica. Due to relatively large gradients of Idrija and Meža Rivers, contaminated materials are eroded and transported downstream. In Idrija, flash flooding during thunderstorms is a common weather phenomenon, allowing the miners to continuously dump mining and ore processing waste directly into the river channel. In contrast, Almadén Hg mining district in Spain, where Hg was produced for at least 2,000 years, has cold semiarid climate (BSk) with less than 500 mm annual precipitation. Despite similar stream gradients in Idrija, Mežica, and Almadén, the physical remobilisation by aqueous transport in Almadén is less important due to a much lower amount of rainfall (Higuera et al., 2006). A similar contaminated site is Pb–Zn–Fe mining district Cartagena-La Union (SE Spain), where ore has been mined for over 1,500 years until 1992 in open-pit and underground mines. Mines are located on the Mediterranean coast at about 400 m asl. This area has arid climate (BSh) with dry and hot summers, with an average annual rainfall of 275 mm (Conesa & Schulin, 2010). Despite a dry climate, streams in the area are extremely polluted along their entire length because of the erosion of mine waste deposits. Due to short but intense rainfalls and steep gradients, Pb contents in stream sediments in La Union district are by 10-times higher than in Meža River sediments (Brotons, Díaz, Sarriá, & Serrato, 2010). It is clear that not only the quantity of rainfall and river gradient but also the precipitation distribution pattern plays a crucial role in contaminated material dispersion by water.

Metal containing particles can be transported over large distances by water, as is in the case of Idrija area, where effects of mining are recorded in the marine sediments as far as 500 km away from the source (Foucher et al., 2009), or several hundred kilometres away from the mines as is in the case of Drava River. In both cases, geomorphological factors (i.e., past or present floodplain and terraces), rather than the distance from the source, determine the level of contamination (Gosar & Žibret, 2011; Šajn et al., 2011). Similar patterns can be observed in the case of The Big River (catchment area of 2,500 km²), which drains the Old Lead Belt mining district (SE Missouri). Channel and floodplain deposits are the most contaminated and act as storage of the transported mine tailings material, where three quarters of contaminated channel sediment is located within 25 km of the mining area, while floodplain contains 16% of the

contaminated material (Pavlovsky, Lecce, Owen, & Martin, 2017). Another example is the Kawerong–Jaba River system in Papua New Guinea, where the waste rocks and tailings have been continuously dumped in the river. Although the majority of tailings were transported into the sea, deposition patterns reveal that tailings were deposited mainly in the overbank sediments and in the area where river enters lowlands and the river gradient is reduced. The areas of lowest deposition are in confined, steeper sections, and in the swampy lowlands (Brown, 1974). Examples show that river erosion and deposition patterns, controlled by river gradient and geomorphological factors, also play crucial role in the contamination distribution pattern.

Two relatively recent environmental disasters that resulted in simultaneous releases of large quantities of contaminated material during mine waste dam collapses lead to tightening European legislation on mining waste management: Aznalcollar accident in Spain (year 1998) and Baia Mare spill in Romania (year 2000). One of the most recent environmental disasters of such type is the collapse of mining wastes dam in Minas Gerais, Brazil, which happened in 2015. All of them produced negative environmental impacts as far as 1,000 km away from the source.

The second most important identified contamination dispersion process is atmospheric emissions from smelters and ore processing facilities, and the transportation of contaminants by air (Figure 8b). The main receptors are soils, which accumulate metals. Topography plays an important role in the dispersion patterns at reference sites. Žerjav Pb and Idrija Hg smelters are located in narrow valleys, so deposition of airborne particulate matter is mostly located in the valleys. In the town of Celje, contamination pattern follows the dominant wind direction and orography, because the hills prevent the dispersion of contaminants southwards. The contamination range in Celje area is between 9 and 14 km for soils. This value is comparable with the Zn smelting district of Magu in China where Zn smelting has been taking place for the last 10 years. Zn smelter impacts in soils are recorded up to 10 km away (Yang et al., 2009). However, the maximum Pb, Zn, and Cd levels in topsoil are lower than in Celje area (8,500, 1,500 and 59 mg kg⁻¹ for the Celje area and 508, 97, and 6 mg kg⁻¹ for the Magu town, respectively).

An example of recent emissions from vanadium smelter is eMalahleni highveld area, Republic of South Africa, where the effects of chromium and vanadium smelters to the street dust composition, as

TABLE 5 Identified sources, pathways, and receptors in the case of the Drava River alluvial plain contamination due to past mining activities, and their effects on the landscapes and possible effects on humans

Source	Contaminant	Pathway	Receptor	Range and influenced area	Changes in landscapes	Change type	Impacts on humans	References
Remote Pb–Zn ore mining upstream	Pb, Zn, Cd, As	River sediments Agricultural soil Groundwater	Agricultural soil Natural riverine habitats Vegetation	Entire Drava River Valley (about 88 km ² in Slovenia)	Agricultural soil contamination	Physical Chemical	PTE intake via food from contaminated soil or water	Halamić et al., 2003; Šajh et al., 2011
Remote ore processing upstream		Aquatic ecosystems	River organisms					

Note. PTE: potentially toxic element.

a consequence of atmospheric particle deposition, is recorded up to 20 km away (Žibret, Van Tonder, & Žibret, 2013). No orographic barriers are present in this case. Another historic Zn smelting district is located in a narrow valley in Palmerton, PA, USA. In this case study, the effects on soils are recorded between 12 and 39 km away, depending on the upwind or downwind direction from the smelter, with maximum Zn, Pb, and Cd levels in topsoil reaching 135,000, 2,000, and 1,750 mg kg⁻¹, respectively (Buchauer, 1973). The study of lead isotopic composition in the most contaminated O2 soil horizon shows that the primary source of lead is the 80-year long Zn smelting tradition in the area (Ketterer, Lowry, Simon, Humphries, & Novotnak, 2001). Dominant impacts of wind direction to the contamination dispersion from Zn smelter has been measured in Karabash town, Russia, which has a long smelting tradition dating back to 1910. Total suspended particulate matter in wet and dry periods varied between 8 and 156 µg m⁻³, depending on the location of the measurement station. Particles collected upwind contain mainly Si and Fe minerals, while particles downwind of the smelter contain Pb and Zn minerals, which were also between 83% and 100% in the respirable size range (Williamson et al., 2004). SO₂ emissions from Karabash smelter, as in the Celje and Mežica cases, have caused significant damage to the vegetation in the distance up to 6 km away. Maximum hourly levels in Karabash were measured up to 20,000 µg m⁻³ (Udachin et al., 2003), whereas in Celje more than 2,000 µg m⁻³. The presented cases illustrate that the topography plays an important role in the dispersion of the contamination in case of smelters located in narrow valleys, while dominant wind direction is more important in cases of flat areas. Impacts on soils are typically measured up to 20 km away from the smelter, and the maximum metal levels in soil are defined by the total emissions from the smelter.

Both contamination dispersion types have different effects on the pollution distribution in the soil or sediment profile. In the case of atmospheric dispersion of contamination only the upper 15 cm of soil profile is usually affected (Buchauer, 1973). On the contrary, in the case of water transport, complex contamination patterns in the soil and alluvial sediment profile are recorded, and deeper layers can also be affected (Gosar & Žibret, 2011; Pavlowsky et al., 2017), thus making potential clean-up operations more challenging.

Contaminants released by mining activities can have negative effects on the ecosystem and humans. Most commonly reported impacts are effects on wildlife and vegetation (Brown, 1974; Covelli et al., 2017; Kladnik, 2009; Špes, 1978), metal uptake by plants and crops, especially by vegetables with edible roots, such as carrots (Bešter et al., 2013; Roy & McDonald, 2015) and increased levels of metals in blood and urine (Jež & Leštan, 2015; Zhang et al., 2012). Metal levels in crops in such areas can exceed reference metal levels by more than 50 times (Zhang et al., 2012).

Presented cases show that mining and ore processing industry has a high potential to produce large-scale impacts on the environment. Major potentially toxic substances dispersion processes are runoff erosion of tailings and ore smelting wastes, and particulate matter emission from ore processing facilities. Poor environmental practices in the past resulted in large contaminated areas, where contaminated flood plains and soils present a reservoir for future dispersion of pollution. It must also be noted that it was very difficult to quantitatively compare contaminated sites, because almost every author used their

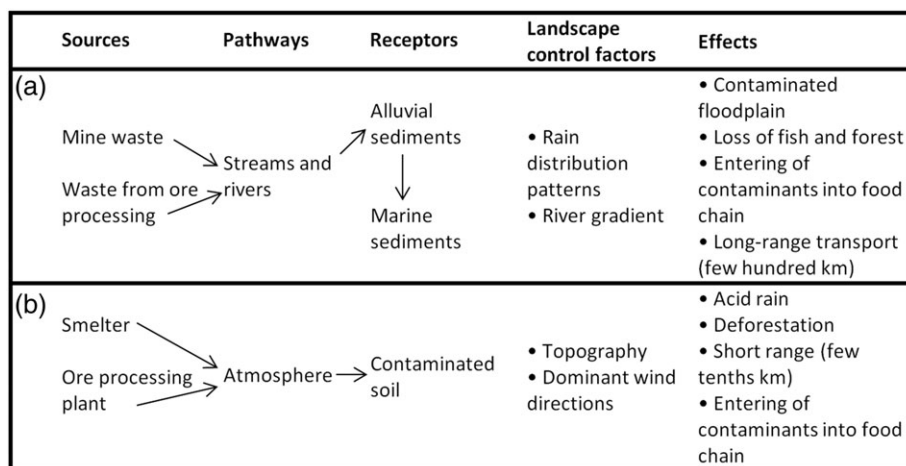


FIGURE 8 Dominant sources, pathways, receptors, landscape control factors, and effects of contaminant dispersion due to historical mining and ore processing

own sampling, analytical, and data interpretation methodology. Thus, the comparison in this paper is based more on qualitative assessment. It is important to know past environmental burdens, caused by metal pollution, which can help to better utilize contaminated areas and also utilize prevent further spreading of contamination today. On a global scale, such considerations can help to prevent dispersion of harmful substances from current and future mining operations around the globe.

4 | CONCLUSIONS

This study focuses on the impacts of mining and ore processing activities on the environment. Four reference study areas are presented and compared with similar sites around the world in order to identify most important sources of contamination, main transport mechanisms of pollutants, receptors, major landscape control factors, and effects. The most important identified contamination sources, accounting the contaminant quantities and size of the contaminated areas, are mining and ore processing wastes, and their runoff erosion. Receptors are floodplains and marine sediments, whereas contamination distribution pattern is dominantly controlled by precipitation and river erosional and depositional patterns. Impacts of such contamination dispersion pathway can be detected several hundred kilometres from the source. Contaminated floodplains, loss of vegetation and fish, and entering of contaminants into the food chain are commonly reported effects. The second most important contamination associated with ore processing is the emission of metal-containing particulate matter and SO₂ and their atmospheric dispersion from smelting facilities. Most important factors affecting contamination distribution are dominant wind direction and topography. Effects in such cases are contaminated soils up to 20 km away from the smelter, loss of vegetation and entry of metals into the food chain. Such studies are important not only for our understanding of the impact of present day mining and ore processing on the environment and landscape but also to allow better understanding of how the environment responds to these changes, allowing us to identify their long-term impacts and how historical changes influence the present-day biosphere and humans.

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ORCID

Gorazd Žibret  <http://orcid.org/0000-0002-9957-1895>

Mateja Gosar  <http://orcid.org/0000-0003-2767-9394>

Miloš Miler  <http://orcid.org/0000-0001-5217-1643>

Jasminka Alijagić  <http://orcid.org/0000-0003-3125-3353>

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