Evaluation of risk to groundwater due to extractive waste in abandoned mine site: Case study of Gorno, NW Italy

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Extractive waste (EW) from abandoned mines can pose serious pressure to natural water systems. The harmful effects of EW such as deterioration of water sources by allowing leaching of potentially toxic elements (PTE) into groundwater make it necessary to carry out careful, scientific and comprehensive studies on this subject. The present research used risk analysis approach to study the effect of presence of PTE to groundwater in abandoned mine site in Gorno, Lombardy (NW Italy). Results indicated that the groundwater was at risk due to Cd and Zn when point of compliance (POC) was at zero m and decreased to no risk at distance of 1500 m from waste dumps.
Keywords: *mining waste, risk analysis, abandoned mine, extractive waste, groundwater, hydrogeology.*

Valutazione del rischio per le acque sotterranee a causa dei rifiuti minerari in una miniera abbandonata: caso studio di Gorno, NW Italia. *I rifiuti estrattivi (EW) provenienti da miniere abbandonate possono rappresentare un serio problema per sistemi idrici naturali. Gli effetti dannosi dei EW, come il deterioramento delle risorse idriche derivanti dalla lisciviazione di elementi potenzialmente tossici verso le acque sotterranee, rendono indispensabile effettuare studi accurati su questo argomento. La presente ricerca ha utilizzato un approccio di analisi del rischio per studiare l'effetto della presenza di PTE nelle acque sotterranee in un sito minerario abbandonato a Gorno, in Lombardia. I risultati hanno indicato che le acque sotterranee sono a rischio per Zn e Cd nel caso di un punto di conformità (POC) pari a zero mentre si riduce zero a una distanza di 1500 m dalla discarica di rifiuti.*
Parole chiave: *rifiuti minerari, analisi del rischio, miniera abbandonata, rifiuti di estrazione, acque sotterranee, idrogeologia.*

1. Introduction

Italy has a long history of mining activities. Classical authors have pointed out that some of the mining sites were in operation since Phoenician-Punic times in Sardinia region (Caro *et al.*, 2013). The mining industry contributed strongly to economy of the Sardinia region from Roman times. The mining was carried out for producing metals (Sardinia), mercury and iron (Tuscany), talc, asbestos, and mixed metallic sulphides (north of Italy). Most of the mining activities in Italy are closed now, leaving behind a legacy of about 3000 abandoned mines (APAT, 2006).

These abandoned mine sites, consisting of mining waste (also referred as extractive waste) provide obvious sources of contamination in the surface environment. As, it acts as source of potentially toxic metallic and metalloid elements to the earth's surface environment for a long time (Hudson-Edwards and Edwards, 2005).

Potentially toxic elements (PTE) are of major concern because of their persistent and bio-accumulative nature which may pose threat to groundwater (GW) in the vicinity of abandoned mine areas (Abraham and Susan, 2017). The present study focuses on calculating risk to GW due to extractive waste (EW) in abandoned mine site of Gorno, Lombardy (NW Italy).

2. Methodology

2.1. Study site

The Gorno mining district is located within the Seriana, Riso and Brembana valleys (Lombardy, Northern Italy). It belongs to the Alpine type zinc-lead-silver stratabound ore deposits, associated with the middle Triassic carbonatic series, as shown in Figure 1. The mineralization (Zn-Pb ± Ag ± baryte ± fluorite) mostly occurs within the "Metallifero" (i.e., "ore-bearing") formation of upper Ladinic – lower Carnian age (Rodeghiero and Vailati, 1977; Jadoul *et al.*, 2012).

Important minerals present in the area are sphalerite (ZnS) and galena (PbS) (average Zn/Pb ratio= 5:1), with minor pyrite (FeS_2), marcasite (FeS_2), chalcopyrite ($CuFeS_2$) and argentite (Ag_2S). The dominant gangue minerals are calcite, dolomite and quartz (± ankerite). The industrial exploitation for Zn and Pb started in the 1837 and continued until 1982.

In the investigated site, Riso creek represent the main watercourse of the Riso valley, which flows to reach the Serio river, the main water artery of the Val Seriana. From the hydrogeological point of view, the area is located in recharge area of Nossana spring (Gattinoni & Francani, 2010). This is a very important spring, used for the water supply of Bergamo city. The groundwater circulation takes place along discontinuity planes (tectonic lines, cracking/fracturing or stratification/schistosity) and karst cavities. Due to the presence of dolomites and limestones outcrops, groundwater flows in fractured karst media with high permeability and high hydraulic conductivity, thus transferring contaminants away from source at high speed (De Luca *et al.*, 2019).

2.2. Sampling and analysis of extractive waste

Site investigation was performed to collect information about waste typology and location, in order to ensure that the facilities were suitable for characterisation and sampling. The sampling site at Gorno, consisted of different WR facilities. One of the important waste facility is located in Mt. Arera, where sampling was conducted. The WR dumps in Arera are spread uniformly with thickness of approximately 2 m.

The location and number of sampling points are often site-specific, however a systematic sampling strategy was adopted in order to obtain representative data of the whole waste facility. Consequently at the site, the WR material was sampled using hand shovel and a hammer (where necessary). Each sample (8-10 kg) was collected in an area of 2m × 2m, after cleaning from organic residues. In total 10 samples of WR were collected at the site (fig. 1). The waste rock samples collected from the site were digested using *aqua – regia* and were analysed for PTE concentrations using Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES). The detailed methodology followed for

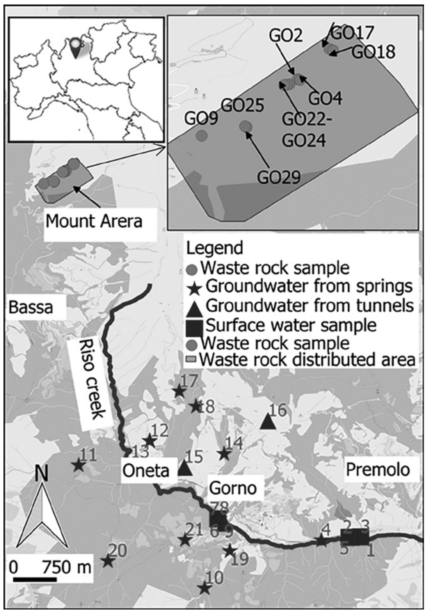

Fig. 1. Geomorphological setting and sample locations.
Inquadramento geomorfologico e ubicazione dei campioni.

the measurement has been reported in Mehta *et al.* (2018).

2.3. Sampling and analysis of water sources

In Gorno two sampling campaigns were conducted in September and October 2016. Groundwater samples were collected from springs and mine tunnels. Surface water (SW) was sampled in Riso creek and other creeks. During the two sampling campaigns, a total of 17 GW samples and 4 SW samples were collected. It should be noted that, due to limited site access and the lack of spring and wells in the area, the water samples were not collected near to WR dumps in Arera, but at distance varying from 3500 m-8000 m from the dumps.

The physical-chemical parameters, like temperature, pH, and electrical conductivity (EC), were measured *in situ* for all samples. Water samples were measured for total alkalinity (sum of CO_3^{2-} + HCO_3^-), bicarbonate and carbonate alkalinity using potentiometric method. Anions (NO_2^-, F^-,SO_4^{2-},-NO_3^-,Cl^-) were measured using 761 Compact IC Metrohm Ion chromatography The major and minor metal cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, P, Al, Ag, Fe, Hg, Ni, Mn, B, Ti, Mo, Sb, As, Be, Cd, Co, Cr (total), Cr (VI), Pb, Cu, Se, Tl, Zn, Sn were measured using ICP-OES. The detailed methodology followed for the measurement has been reported in Mehta *et al.* (2018).

2.4. Risk analysis

The potential risks to GW due to the presence of contaminants in WR were evaluated using Risk – Net software, in accordance with the provisions of Risk Based Corrective Action (RBCA) (ASTM, 1995; ASTM, 2015). It indicates that sites should be managed to have low and acceptable risk levels rather than bringing them to pristine levels. The permissible limits for the chemical elements for the risk calculations were taken from the Italian Legislative Decree 152/06 and risk analysis guidelines (Ministero dell'Ambiente e della tutela del territorio, 2006; APAT, 2008).

The risk was calculated under following conditions: (1) site characteristics and exposure parameters at Gorno, and (2) considering the point of compliance (POC) at varying distance from the WR. To account for the most conservative risk analysis results, the following aspects were considered for source, receptor and pathway:

1) Source: the contaminated WR dumps were considered as source. The concentrations of contaminants at source (CRS) were considered as 95% upper confidence limits for the concentrations of PTE at fractions <20 mm of WR.
2) Receptor: the superficial aquifers present in detritic cover were considered as receptor. Consequently, the karstic au-

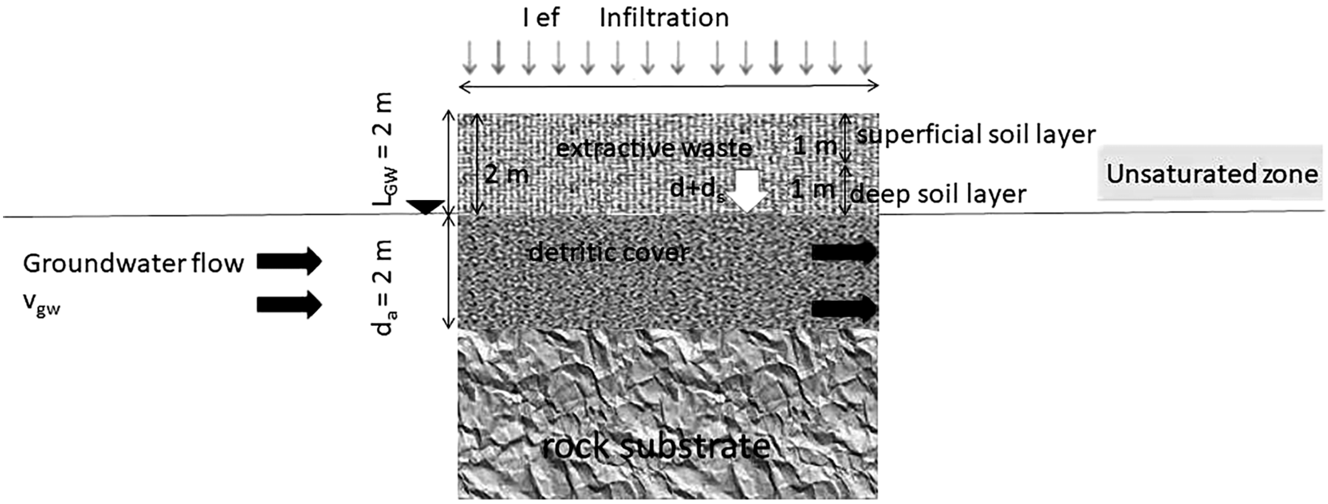

Fig. 2. Schematization of the modelled scenario for risk analysis.
Schema dello scenario modellizzato per l'analisi di rischio.

qifer, that is present below detritic cover, was not taken into account in the risk analysis procedure (fig. 2).
3) Pathway: the contaminants considered at site are inorganic in nature, hence, the only migration pathway of contaminants from the WR was leaching to the underlying water.

2.5. Site characteristics

Observations collected from the general site setting, field surveys, and previous hydrogeological and climatic studies led to the development of the site characteristics used in the risk analysis (tab. 1). The study site is almost completely covered by WR at Gorno. These characteristics are explained in detail as follows.

- The natural soil has been replaced by WR in a rectangular manner, with 900 m and 450 m as the dimensions of sides.
- The total thickness of the on-site waste deposits was 2 m, thus leading to superficial soil with a depth of 1 m and deep soil with a depth of 1 m in the waste distributed area, as shown in Figure 2.
- The groundwater flow through the detritic cover and WR was considered to be predominant. The depth to the GW level from the top was 2 m, and thus the thickness of the unsaturated zone was 1.9 m, considering that a thickness of 0.1 m became saturated due to capillary action.
- The hydraulic gradient was approximately equal to the slope

Tab. 1. Unsaturated zone, saturated zone and outside environment properties at Gorno.
Zona insatura, zona satura e caratteristiche ambientali esterne (Gorno).

Parameter		Units	Gorno
$L_{s(SS)}$	Depth of top of source in the surface soil relative to the surface level	m	0
$L_{s(SP)}$	Depth of the top of the source in the deep soil relative to the surface level	m	1
d	Thickness of source of contamination in the surface soil (unsaturated)	m	1
d_s	Thickness of the source of contamination in the deep soil (unsaturated)	m	1
L_{GW}	Depth of the groundwater level (total from the top) (phreatic level)	m	2
h_v	Thickness of the unsaturated zone	m	1.9
pH	pH		7.72
I_{ef}	Effective infiltration	cm/year	49.6
P	Precipitation	cm/year	166
W	Extension of the source in the direction of groundwater flow	m	900
S_w	Extension of the source in the direction perpendicular to the groundwater flow	m	450
d_a	Aquifer thickness	m	2
K_{sat}	Hydraulic conductivity of saturated soil	m/s	50×10^{-4}
i	Hydraulic gradient		0.3
v_{gw}	Darcy's velocity	m/s	1.5×10^{-3}
v_e	Average effective rate in the aquifer	m/s	4.25×10^{-3}
$\theta_{e\,sat}$	Effective porosity of the ground in the saturated zone		0.353
POC	Distance of the receptor (off-site) (DAF)	m	0
δ_{gw}	Thickness of the mixing zone in the aquifer	m	2
LDF	Dilution factor in groundwater	calculated	213

of the mountains and was thus 0.30.

- Out of all the types of soil in the software, the physical properties of the WR within the fractions less than 20 mm most closely resembled those of sand. Thus, the effective porosity of the source, volumetric air content and volumetric water content of sand were used.
- The infiltration capacity was measured considering an average precipitation of 166 cm at Gorno (the average annual precipitation for the period of 1961-1990 for the site).

2.6. Risk calculation

To evaluate the risks to GW the concentrations expected in the groundwater at the POC were compared with drinking water quality criteria set by legislation (Ministero dell'ambiente e della tutela del territorio, 2006). This is one of the approaches most widely adopted in European countries (Di Gianfilippo *et al.*, 2018). The risks to GW due to elements being leached from the soil are calculated using Eq. (1), (2) and (3) by calculating $R_{SS.\,LF}$ and $R_{DS.\,LF}$. There is presence of risk to GW due to a contaminant if the summation of $R_{SS.\,LF}$ and $R_{DS.\,LF}$ i.e. RGW exceeds 1, with the potential risks increasing with increase in RGW.

$$RGW = R_{SS\cdot LF} + R_{DS\cdot LF} \quad (1)$$

$$R_{SS\cdot LF} = \frac{CRS \cdot LF_{SS}}{DAF \cdot CSC \cdot 10\text{–}3\ mg/\mu g} \quad (2)$$

and

$$R_{DS\cdot LF} = \frac{CRS \cdot LF_{DS}}{DAF \cdot CSC \cdot 10\text{–}3\ mg/\mu g} \quad (3)$$

Where

RGW = Risk to groundwater

$R_{SS\cdot LF}$ = Risk to groundwater due to the contaminant in superficial soil,

$R_{DS\cdot LF}$ = Risk to groundwater due to the contaminant in deep soil,

CRS = Concentration of contaminant at source (mg/kg),

LF_{SS} = Leaching in groundwater from the contaminant in superficial soil,

LF_{DS} = Leaching in groundwater from the contaminant in deep soil,

DAF = Factor of dilution in groundwater (for on-site exposure, DAF =1), and

CSC = Permissible limit of a particular element in groundwater (Ministero dell'ambiente e della tutela del territorio, 2006, legislative decree 152/06).

2.7. Risk calculation with variation in point of compliance

To assess, wider span of possible scenario conditions, potential risks to GW were calculated at varying distance of POC from source of contamination. The effect due to superficial soil layer and sub soil layer, were analysed. This was done, to see the effect of distance on the GW quality. This is a key aspect to consider, as the variation of POC can lead to changes in concentration of contaminants due to attenuation during transport from the source to the underlying groundwater. The dilution attenuation factor (DAF) in groundwater (Eq. 2 and 3) to be considered will change with the placement of POC. The DAF takes into account the dispersive phenomenon in all directions (i.e. x, y, z).

3. Results and Discussion

3.1. Analysis of extractive waste

The concentration of PTE in waste rock samples and pH values are shown in Table 2. The pH values were found to be in slightly alkaline range varying from 7.4 to 7.9, with an average of 7.7. The results also indicated that Zn was present at very high levels, with an average concentration of 37,193 mg/kg, whereas the concentrations varied from 179 mg/kg to 98,706 mg/kg. Cadmium was found to vary from

Tab. 2. Potentially toxic elements concentrations (mg/kg) in WR samples with sizes less than 20 mm from Gorno.

Concentrazioni di elementi potenzialmente tossici (mg/kg) in campioni WR con dimensioni inferiori a 20 mm (Gorno).

Sample	pH	Sb	As	Be	Cd	Co	Cr	Ni	Pb	Cu	Tl	V	Zn
Limit 1		**10**	**20**	**2**	**2**	**20**	**150**	**120**	**100**	**120**	**1**	**90**	**150**
Limit 2		**30**	**50**	**10**	**15**	**250**	**800**	**500**	**1000**	**600**	**10**	**250**	**1500**
GO2	7.9	7.5	9.9	0.1	**28.1**	0.4	1.4	1	6	21	0.4	15.1	**12,169**
GO4	7.8	**11.2**	10.0	0.2	**133.0**	0.5	2.3	1	9	50	0.6	21.0	**50,678**
GO9	7.8	3.6	15.2	0.5	**21.2**	1.2	3.1	3	26	13	**1.2**	12.7	**12,669**
GO17	7.8	4.5	**23.7**	0.4	**28.6**	0.3	1.6	1	9	15	0.6	9.5	**14,505**
GO18	7.8	1.6	3.7	0.0	**18.8**	0.1	0.5	0	2	9	0.2	2.5	**6,626**
GO22	7.8	**16**	**37.2**	0.5	**167.3**	1.3	4.1	4	12	77	**1.4**	29.9	**76,243**
GO23	7.7	**32.3**	**45.1**	0.8	**210.8**	3.1	9.4	7	24	110	**3.8**	54.9	**98,706**
GO24	7.7	**49.9**	**28.6**	0.4	**167.9**	1.4	4.4	3	40	91	**1.5**	30.5	**69,077**
GO25	7.5	8.0	15.6	0.2	**37.3**	0.5	1.8	2	11	17	0.7	24.7	176
GO29	7.4	8.3	16.2	0.3	**59.0**	0.7	2.7	3	17	25	0.8	20.7	**31,081**

* Legislative limits currently adopted in Italy for PTE concentrations in the soil (Ministero dell'ambiente e della tutela del territorio, 2006), limit 1 is intended for green and residential areas, while 2 for commercial and industrial areas.

19 mg/kg to 211 mg/kg, with an average concentration of 87 mg/kg. High concentrations of Zn and Cd, can be explained by the fact that the site is rich in Zn as the exploitation activities also were performed for Zn.

Antimony was found to vary from 2 mg/kg to 50 mg/kg, with an average concentration of 14 mg/kg. Arsenic was found to vary from 4 mg/kg to 45 mg/kg, with an average concentration of 20 mg/kg. It should be noted that the samples from the site were analysed for their concentrations of PTE in size fractions of less than 2 mm. However, the concentrations used in risk analysis were calculated from fractions of less than 20 mm following risk analysis guidelines (Ministero dell'ambiente e della tutela del territorio, 2006; APAT, 2008).

Tab. 3. Physical-chemical properties of the water samples. Limit: Legislative limits adopted in Italy for PTE concentrations in water (Ministero dell'ambiente e della tutela del territorio, 2006).
Proprietà fisico-chimiche dei campiodi d'acqua. Limite: limiti legislativi adottati in Italia per le concentrazioni di PTE nell'acqua (Ministero dell'ambiente e della tutela del territorio, 2006).

Sample	Type	pH	Temp	EC	Cd	Zn
Unit			°C	µS/ cm	µg/l	µg/l
Limit					5	3000
1	GW	7.9	18	770	0,3	38.2
2	SW	8.45	16.4	320	<0,2	14.8
3	SW	8.52	25.3	348	0,2	19.4
4	GW	7.78	21.2	232	<0,2	<1.0
5	SW	8.35	16	326	<0,2	7.6
6	GW	8.35	12.6	335	<0,2	<1.0
7	SW	8.47	14.2	308	<0,2	<1.0
8	GW	8.31	14.3	275	<0,2	<1.0
9	GW	7.89	11.3	380	<0,2	<1.0
10	GW	7.39	10	289	<0,2	<1.0
11	GW	7.95	10.2	289	<0,2	<1.0
12	GW	8.07	11	289	<0,2	<1.0
13	GW	8.35	10.6	259	3,1	260
14	GW	8.27	10	258	<0,2	<1.0
15	GW	8.24	10.05	299	1,0	327
16	GW	8.21	8.2	244	0,3	25.8
17	GW	7.92	11.1	190	0,3	69.0
18	GW	8.03	9.8	277	<0,2	<1.0
19	GW	7.89	11.9	247	<0,2	<1.0
20	GW	8.31	8.3	256	<0,2	<1.0
21	GW	8.22	11.3	256	<0,2	<1.0

3.2. Analysis of water samples

The results of the analyses performed on water samples are shown in Table 3. With regard to physical parameters, GW from mine tunnel showed temperature varying from 8.2°C and 12.6°C. Surface water showed a wide range of temperature, between 14.2°C and 25.3°C (registered in a secondary creek near Ponte Nossa). The pH values varied from 7.39 and 8.40 in GW. In SW the values of pH were quite constant from 8.40 to 8.52 and were in alkaline range which could be due to presence of carbonate minerals like calcite and dolomite.

The surface water samples collected at Gorno showed no parameter exceeding the limits, according to Italian Law Decree 152/06. Although there was presence of mine activities for the extraction of sphalerite and calamine, water samples showed no contamination of Zn in both groundwater and SW (fig. 1). The SW samples collected in the Riso creek in the lower part of the valley shows concentration of Zn, respectively 14.8 µg/l and 7.6 µg/l. The water samples also showed no contamination due to Cd. Table 3, gives the values of Cd and Zn only, as the major elements present in waste rock were Cd and Zn (mentioned in Section 3.1). These could potentially leach to GW and cause contamination. However, the concentrations of other parameters can be found in Mehta (2019).

The absence of contamination in water samples (GW and rivers) could be due to several concomitant factors: 1) higher pH of groundwater, which facilitates the precipitation of PTE, 2) presence of sampled water sources at distance from the WR dumps, which leads to attenuation of contaminants due to dispersion and transfer phenomena, and 3) increased flow velocity in the karst limestone rocks.

3.3. Risk analysis

For POC positioned in the groundwater under the source of contamination, the GW was at risk due to the presence of Cd and Zn (tab. 4). This can be attributed to the fact that the WR were rich in Cd and Zn. Moreover, the potential risks to groundwater due to Cd and Zn were very similar in values. The possible reason could be that Cd concentrations in WR were associated with Zn concentrations. This happens due to strong geochemical associations between these two metals because of similar physical and chemical properties. Similar phenomenon has been observed in lead-zinc mines in Upper Silesia (Poland) and Zawar (India) (Ullrich *et al.*, 1999; Anju and Banerjee, 2011). Metals like Be, Co, Cr and Se were present in very low concentrations compared to the permissible limit of metals in GW in Italy and thus causing low level of potential risks.

3.4. Risk analysis with variation in POC

The total risks to groundwater due to Zn at POC placed at zero distance was 2.16 (0.72 due to superficial layer and 1.44 due to deep layer). The potential risks due to Cd, it was observed that the value was 2.44 for groundwater under the source of contamination. The total risks to GW decreased to 1 due to both Cd and Zn at about 1500 m, i.e. at POC = 1500 m. Thus there was no risk to GW, for aquifers at distance equal to and/or greater than 1500 m from WR. The detailed values of potential risks to GW due to superficial soil layer and deep soil layer can be found in Figure 3.

Tab. 4. Risks to groundwater in Gorno. CRS is concentration of contaminant at source. There is presence of risk when RGW (Risk to GW) exceeds 1.

Rischi relativi alle falde acquifere (Gorno). CRS è la concentrazione di contaminante alla fonte. Esiste una presenza di rischio quando RGW (Risk to GW) supera 1.

		Superficial layer	Deep layer
Contaminant	CRS (mg/kg)	Risk to GW	Risk to GW
Antimony	30.38	0.32	0.63
Arsenic	28.09	0.22	0.45
Beryllium	0.47	3.49×10^{-4}	6.98×10^{-4}
Cadmium	130.20	**0.814**	**1.63**
Cobalt	1.46	0.001	0.003
Chromium	4.60	1.2×10^{-7}	2.40×10^{-7}
Nickel	3.67	0.006	0.01
Lead	22.22	0.006	0.012
Copper	83.08	0.006	0.01
Selenium	0.02	9.32×10^{-4}	1.86×10^{-3}
Thallium	1.72	0.03	0.06
Zinc	5,70,79.00	**0.72**	**1.44**

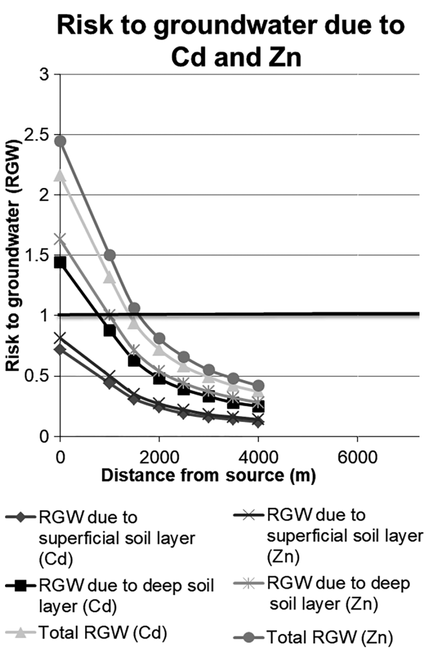

Fig. 3. Graph showing variation of RGW due to Cd and Zn with respect to distance of POC from source of contamination at Gorno.

Grafico della variazione di RGW dovuta a Cd e Zn rispetto alla distanza di POC dalla sorgente di contaminazione (Gorno).

3.5. Possible solutions

The risk analysis calculations using the software shows presence of potential risks at POC = 0 m, according to the used parameters in most conservative cases for concentration of contaminants and position of aquifers. However, it is difficult to confirm these results at site, as the aquifers sampled in the study showed no contamination. It should be noted that, the water samples were collected away from WR dumps and not directly beneath the dumps, due to limited site access.

The future efforts and intervention activities to reduce any potential risks to GW can include: (1) placing of low permeability layer on WR dumps to reduce the leaching of contaminants, (2) transporting the WR and reusing it, such as for recovery of raw materials, and (3) use of signage and boards in the area to communicate to the local public, about presence of elements in WR. These activities can be carried out on the basis of the interests of local public and government and using cost-benefit and life-cycle analysis (Mehta *et al.*, 2018; Dino *et al.*, 2018; Mehta 2019).

4. Conclusions

Risk analysis studies can be used to determine potential risks to GW. To calculate risks to GW elemental analyses of WR were performed. The WR samples at Gorno indicated that Zn was present at very high levels, with an average concentration of 37193 mg/kg, whereas the concentrations varied from 179 mg/kg to 98,706 mg/kg. Cadmium was found to vary from 19 mg/kg to 211 mg/kg, with an average concentration of 87 mg/kg.

Risk analysis studies at Gorno depicted that GW was at risk due to Cd and Zn contamination for POC at zero m and there was no risk to groundwater at POC placed at distance equal to and/or greater than 1500 m from WR dumps. These risk analysis calculations were performed according to the parameters in most conservative cases for concentration of contaminants and position of aquifers. In reality, the sampled GW in the area collected not directly beneath the dumps, did not show any contamination, possibly due to dilution of contaminants.

The studies noticing the effect of distance of POC provides necessary information about the extent of the impact on water sources due to waste and thus can provide guidance towards application of mitigation measures to source of contamination. Factoring in the natural attenuation of contaminants during transfer, leads to more robust risk analysis calculations. This risk analysis approach can form the basis towards understanding, evaluating and assessing GW contamination.

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