Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.922285

54 (2015) 3511–3522
June

Taylor & Francis
Taylor & Francis Group

Characteristics of hydrochemical variations and contaminant load during rainfall in an acid mine drainage-impacted watershed, Korea

G. Pak[a], U. Hong[b], M. Jung[b], H. Kim[b], K. Han[b], K.J.B. Mallari[b], S. Kim[a,b], Y. Kim[a,b], J. Yoon[a,b,*]

[a]*Department of Environmental Engineering, Korea University, Sejong 339-700, Korea, emails: gjpak00@korea.ac.kr (G. Pak), ub1905ub@korea.ac.kr (S. Kim), kimyo@korea.ac.kr (Y. Kim), jyyoon@korea.ac.kr (J. Yoon)*
[b]*Program in Environmental Technology and Policy, Korea University, Sejong 339-700, Korea, emails: firebugs@korea.ac.kr (U. Hong), inver@korea.ac.kr (M. Jung), tinymons@korea.ac.kr (H. Kim), rudwls1009@korea.ac.kr (K. Han), kristinemallari@korea.ac.kr (K.J.B. Mallari)*

Received 15 January 2014; Accepted 14 March 2014

ABSTRACT

Acid mine drainage (AMD) refers to the outflow of acidic water from usually abandoned metal mines. Since AMD is closely related to the rainfall–runoff process, it is important to monitor rainfall events for its characterization. This work aims to study the characteristics of hydrochemical variations of metals during rainfall in the Geopung Mine Watershed, Korea. Eleven dry-weather and five rainfall event monitorings were conducted in 2010 at two locations (GP-1 and GP-2). At GP-1, concentrations significantly increased during the fifth event, which was the largest one (As from 0.0003 to 0.0046 mg/l; Cd from 0.0114 to 0.2188 mg/l; Cu from 0.0206 to 3.1512 mg/l; Pb from 0.0006 to 0.1857 mg/l; Zn from 0.5823 to 15.9194 mg/l; Ni from 0.0011 to 0.0294 mg/l). Similar though less-marked trend of increase in metal concentration was found at GP-2 during the fifth rainfall event. As for the load, as much as 62–97% of the annual metal loads were accounted for by rainfall at GP-1 and GP-2. This pattern implies that most of the metals in AMD are brought into the stream by rainfall.

Keywords: Acid mine drainage; Hydrochemical variation; Metal load; Rainfall

1. Introduction

Acid mine drainage (AMD) refers to the outflow of acidic water from abandoned metal mines or coal mines. The pollution from an abandoned mine is usually generated from AMD which is related to waste rock and tailings [1–3]. Waste rock is the residue crystal after separating useful minerals from the raw ore. Most of the crystal is derelict after the separation because of the poor quality of the minerals. Tailings are mine wastes that have the size of sand or silt particles separated from the crystal body during the separation process [4,5]. Approximately 2,500 metal, coal, and nonmetal mines had been developed in Korea. However, 80% of them had been shut down without treatment or remediation, leaving them as the

*Corresponding author.

Presented at the 5th IWA-ASPIRE Conference, 8–12 September 2013, Daejeon, Korea

potential sources of contamination with waste rock and tailings being the principal wastes [6].

Waste rock and tailings that are created at the mine site contain sulfide minerals called pyrite. If sulfide minerals are neglected on the ground without appropriate measures, exposure to air and water (rainwater, surface water, or ground water) will cause oxidation of metal sulfides within the surrounding rock and overburden which generates acidity [7]. AMD also involves the migration of metals from mine wastes into ground water or surface water. Many studies have been conducted to estimate the potential effect caused by the migration of metals and found that the neglected mine waste can act as a potential pollution source and constantly affect the ecosystem [8].

Traditional approaches to the assessment of the polluted metal mine sites have been to quantify the contamination of point source, such as waste rock and tailings area [9–11]. However, recent studies focus on the fate and transport of heavy metals in rivers [12–16]. Though useful, these studies have focused on metal migration during dry period. On the other hand, some researches also have been done to investigate the fate of metals in rivers during rainfall when AMD can become an important factor in metal migration [17–19]. However, those researches were limited in terms of both number of storm events sampled (two events or less) and locations (single location only).

In this study, metals in river water were continuously monitored during dry- and wet-weather conditions to investigate the hydrochemical variations of metal contaminants at multiple locations in the stream reach. In addition, event and annual metal loadings were estimated to understand relative importance of dry- and wet-weather conditions in the export of metals from the watershed.

2. Site description

The study area is Geopung Mine Watershed, a tributary of Geum River Basin located in Chungbuk Province (Fig. 1). Geopung mine, mined for copper and zinc, was shut down in 1998 and has been identified as the area with the highest pollution level of metals among the mine areas of the Geum River Basin.

The basin has a total area of 24.8 km^2 and is characterized by mountains and hills. Jangyoun Stream, a small stream with a length of 10.5 km, drains the basin. The Jangyoun Stream eventually drains to the Geum River via the Bocheong Stream. The downstream reach of Jangyoun Stream near mine site is heavily affected by AMD, evidenced by yellow-orange solids precipitated on the bottom sediments colloquially known as yellow boy.

The sampling points are shown in Fig. 1 and have been marked as GP-1 and GP-2. GP-1 is the influent point from mine source area to Jangyoun Stream and it receives direct inflow of AMD by rainfall. GP-2 is the outlet of the watershed, which is approximately 1 km downstream of the Geopung mine area.

3. Methodology

Samples for water quality and discharge were taken at two points (GP-1 and GP-2) of the Jangyoun Stream (Fig. 2) during the dry and wet weathers. At GP-1 and GP-2, water quality samples were analyzed for the concentrations of metals, and their loadings were calculated.

Surface water samples were collected in 1 L LDPE bottles and the pretreatment was performed with the use of nitric acid. Bottles were washed in 10% (v/v) nitric acid prior to sampling. Samples were pumped by an air pump vacuum system. Before taking a sample, the equipment was purged with river water to avoid sample cross-contamination. The metal pollutants considered in this study are As, Cd, Cu, Pb, Hg, Ni, and Zn, which were selected because they are classified in the Korean Warning & Countermeasure Standards as major elements. Metals in surface water were analyzed via inductively coupled plasma with mass spectrometry (ICP-MS, Agilent Technologies). In the case of Hg, hydride/vapor generation coupled to ICP-MS was used. The DO, pH, EC, TDS, and ORP were measured with a portable probe (Xian 1000) during the sampling.

The discharge data were taken with the USGS Type AA Current Meter model 6200 and USGS Pygmy Meter model 6205. The measurement was performed before and during the rainfall event. The frequency of the measurement was every two, three, or six hours depending on the stream depth variation during the rainfall. Sampling for water quality was simultaneously done with the discharge measurement. Finally, the metal loadings were calculated based on the concentration and discharge data collected.

4. Results and discussion

4.1. Rainfall and discharge variation

A total of 11 measurements were taken for discharge from January to November 2010 each month during the dry weather at GP-1 and GP-2 stations. In addition, five rainfall events were also monitored for discharge during July–August 2010 at these two

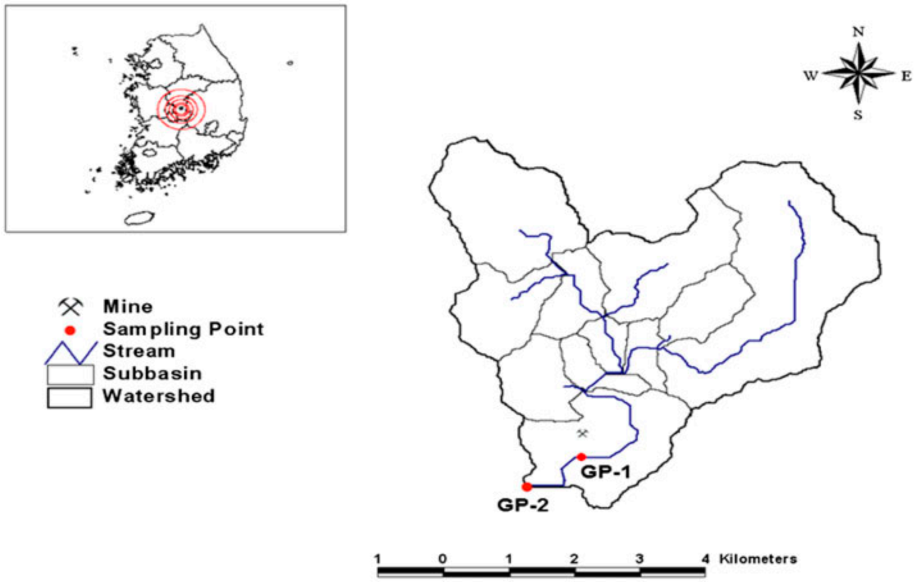

Fig. 1. Plan view of Geopung Mine Watershed.

Fig. 2. Satellite view of the study area showing monitoring points.

stations. Table 1 shows the measured discharge during dry and wet weather. In case of GP-1, there were instances when there was no water collected due to dry conditions but the discharge typically ranged between 0.003 and 0.018 m^3/s, while the discharge measured at the stream outlet GP-2 was between 0.025 and 0.387 m^3/s during the dry weather. The rainfall depths for five rainfall events monitored varied from 19 to 207 mm. During rainfall, the discharge measurements at GP-1 ranged from no flow to 0.105 m^3/s, while at GP-2 ranged from 0.013 to 4.433 m^3/s. Fig. 3 shows the hydrographs at GP-1 and GP-2 during the study period as well as the daily rainfall recorded in the catchment. For both GP-1 (area influent point from mine source to Jangyoun Stream) and GP-2 (outlet of Jangyoun Stream Watershed), they appeared to have sharp increase and fall in discharge since Jangyoun Stream drains a very small area.

4.2. Hydrochemical variation

Table 2 shows the statistics of the hydrochemical data during dry and wet weather at GP-1 and GP-2 stations. Pollutant statistics for the dry weather was derived from 11 monthly monitorings, whereas statistics for wet weather was derived from five rainfall events. Figs. 4 and 5 show the hydrochemical variations during wet weather at points GP-1 and GP-2, respectively. In the figures, metal concentrations were in general found to react sensitively to the total rainfall. At GP-1, concentrations significantly increased during the fifth event, which was the largest one (As from 0.0003 to 0.0046 mg/l; Cd from 0.0114 to 0.2188 mg/l; Cu from 0.0206 to 3.1512 mg/l; Pb from 0.0006 to 0.1857 mg/l; Zn from 0.5823 to 15.9194 mg/l; Ni from 0.0011 to 0.0294 mg/l). Similar though less-marked trend of increase in metal concentration was found at GP-2 during the fifth rainfall event. This shows that most of the metals found in the stream during rainfall event are due to AMD from nearby mine area. It was also seen that metals flowing into the stream during initial rainfall eventually decrease in concentration by the effect of dilution.

4.3. Pollutant loads

Table 3 shows the metal loadings during dry and wet weather at GP-1 and GP-2 stations. The loading of

Table 1
The measured discharge during dry and wet weather

	Dry weather		Wet weather				
	Discharge (cms)					Maximum Discharge (cms)	
Time	GP-1	GP-2	Event	Duration (h)	Rainfall (mm)	GP-1	GP-2
Jan.	NF	0.025	1	11	44.5	0.0287	1.3292
Feb.	NF	0.387	2	29	46.5	0.0486	2.4899
Mar.	NF	0.098	3	20	19.0	0.0099	0.6366
Apr.	0.018	0.206	4	30	20.5	0.0108	0.8264
May	NF	0.240	5	159	207.5	0.1054	4.4331
Jun.	NF	NF					
Jul.	0.012	0.095					
Aug.	0.012	0.176					
Sep.	0.011	0.113					
Oct.	0.003	0.031					
Nov.	NF	0.033					

Note: NF = no flow.

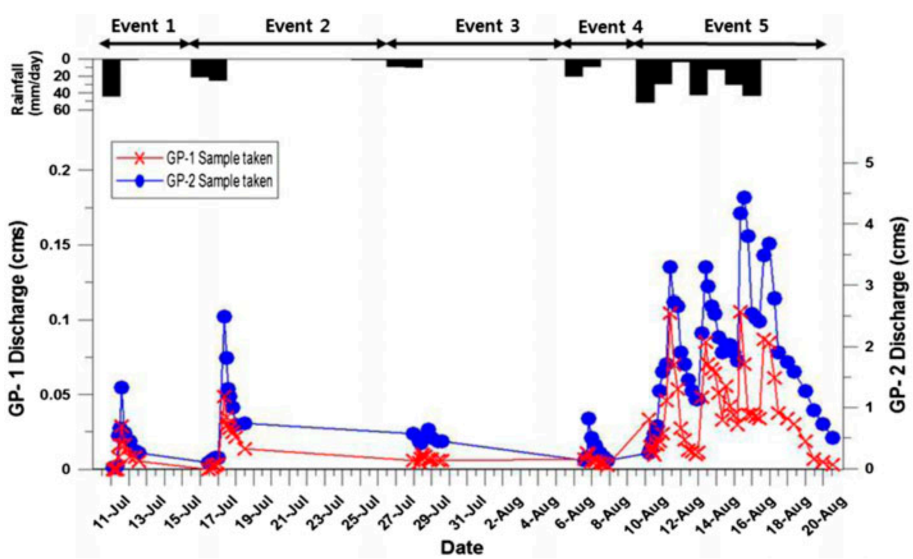

Fig. 3. Hydrographs at sampling points GP-1 and GP-2 during wet weather.

each dissolved metal during the dry weather was calculated by simply multiplying the discharge and concentration of each element measured once a month. As for the loadings during wet weather, the maximum loading was chosen, which is the maximum of the product of discharge and concentration during the storm event.

IIt can be seen from Table 3 that maximum loadings during events were significantly larger than the loadings during the driest month (October). At GP-1, the Cd loading was 410 times, Cu loading was 690 times, Pb loading was 430 times, Zn loading was 200 times, and Ni loading was 150 times higher during maximum rainfall (Event 5) compared to October loadings. At GP-2, the As loading was 600 times, Cd loading was 120 times, Cu loading was 590 times, Pb loading was 4,940 times, Zn loading was 60 times, and Ni loading was 740 times

Table 2
Basic statistics of analyzed metal concentrations during dry and wet weather (mg/l)

Station	Period	Metal elements	*n*	Mean	Median	Standard deviation	Minimum	Maximum
GP-1	Dry weather	As	11	0.0005	0.0005	0.0004	0.0001	0.0011
		Cd	11	0.0865	0.0662	0.0814	0.0065	0.2833
		Cu	11	0.9575	0.8276	0.8208	0.0612	2.5540
		Pb	11	0.0057	0.0049	0.0040	0.0028	0.0137
		Hg	11	ND	ND	ND	ND	ND
		Zn	11	8.3092	5.7219	7.2305	0.6385	21.9840
		Ni	11	0.0065	0.0049	0.0048	0.0021	0.0166
	Wet weather	As	84	0.0009	0.0007	0.0007	0.0003	0.0046
		Cd	84	0.0926	0.0798	0.0531	0.0114	0.2188
		Cu	84	1.0571	1.0884	0.9116	0.0206	3.1512
		Pb	84	0.0187	0.0091	0.0338	0.0006	0.1857
		Hg	84	ND	ND	ND	ND	ND
		Zn	84	4.0546	3.8850	2.7839	0.5823	15.9194
		Ni	84	0.0074	0.0064	0.0053	0.0011	0.0294
GP-2	Dry weather	As	11	0.0003	0.0002	0.0003	0.0001	0.0009
		Cd	11	0.0018	0.0019	0.0009	0.0013	0.0035
		Cu	11	0.0139	0.0131	0.0095	0.0048	0.0371
		Pb	11	0.0003	0.0002	0.0004	0.0002	0.0015
		Hg	11	ND	ND	ND	ND	ND
		Zn	11	0.1566	0.1427	0.0834	0.0939	0.3239
		Ni	11	0.0005	0.0005	0.0002	0.0003	0.0009
	Wet weather	As	84	0.0005	0.0005	0.0002	0.0002	0.0013
		Cd	84	0.0027	0.0019	0.0038	0.0006	0.0335
		Cu	84	0.0208	0.0205	0.0113	0.0009	0.0650
		Pb	84	0.0037	0.0022	0.0055	0.0001	0.0370
		Hg	84	ND	ND	ND	ND	ND
		Zn	84	0.1422	0.1112	0.1394	0.0039	1.2206
		Ni	84	0.0010	0.0007	0.0010	0.0001	0.0059

Note: ND = not detected.

higher during maximum rainfall (Event 5) compared to October loadings. Hence, the increase in stream contaminant loadings can be attributed to AMD generated during wet weather. Figs. 6 and 7 show the variations in metal loadings during wet weather at GP-1 and GP-2, respectively. Metal loadings were also found to be sensitive to total rainfall similar to metal concentrations at GP-1 and GP-2.

Annual metal load for each metal element were estimated based on the results of dry- and wet-weather monitoring. The sum of metal load based on 11 dry weather monitorings on a monthly basis and the load during the five monitored rainfall events (330 mm) can provide rough estimate for annual metal load. However, five rainfall events monitored during rainy season will not represent wet load for the whole year. For this reason, annual loads were estimated by considering all of the rainfall (1,029 mm) during the whole year at the study watershed. To this end, rainfall vs. metal load relationship was constructed based on the regression analysis of five monitored rainfall events during July–August 2010. Annual metal loads during rainfall were then recalculated by summing up the equivalent loadings of each rainfall event that occurred during the whole year, derived from the regression equation. Table 4 shows the estimated annual metal loads. The calculation showed that the metal loads during rainfall, with regard to As, Cd, Cu, Pb, Zn, and Ni, accounted for at least over 81% of the total for the whole year at GP-1, and at least over 60% at GP-2.

The estimated metal loads understandably lack accuracy as it was based on the extrapolation from the regression between load and discharge, rather than on actual measurements. However, the results can be interpreted that most of the metal loads in the stream occur during the wet weather.

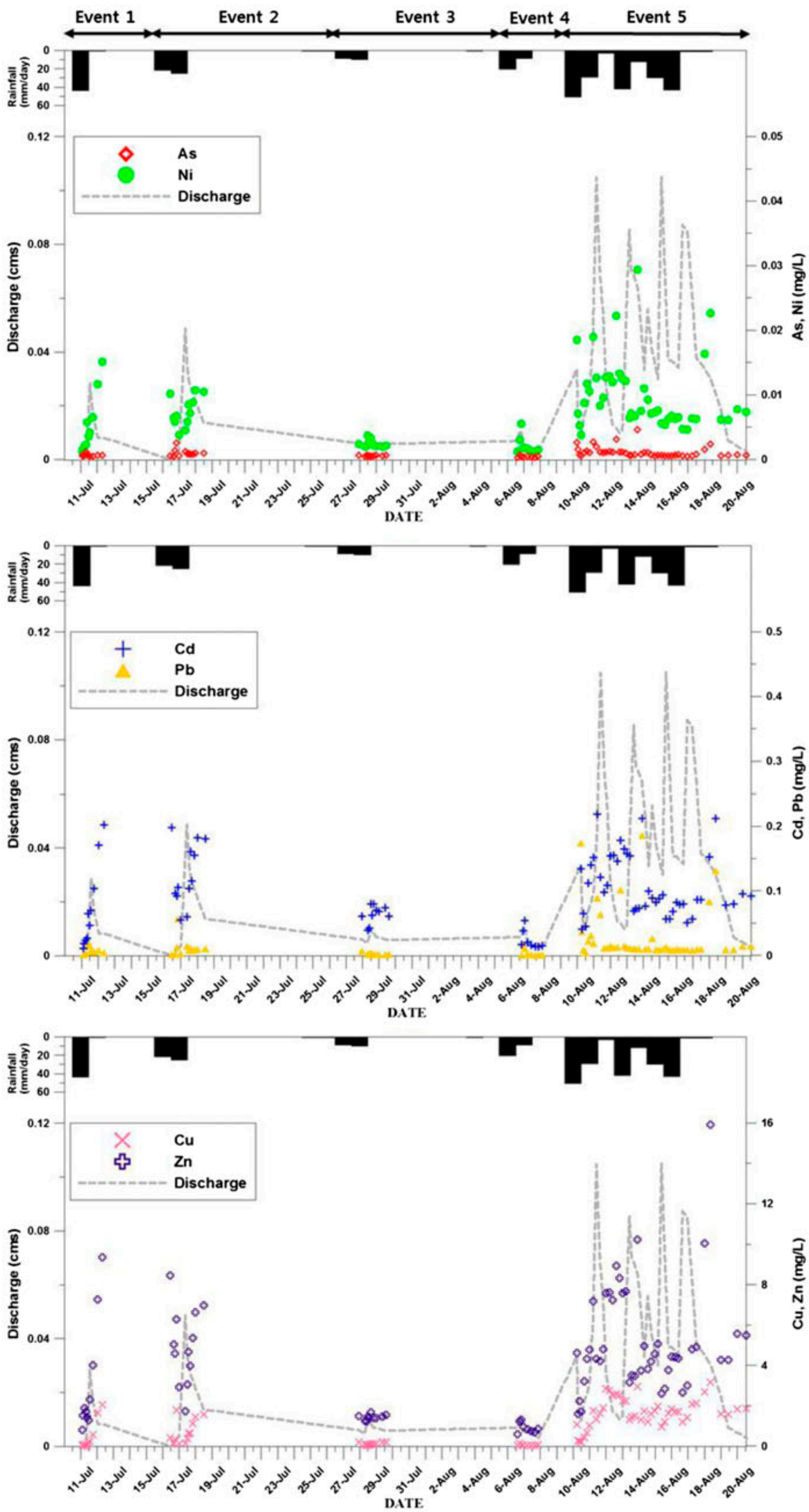

Fig. 4. Variation of dissolved metal concentrations during wet weather at GP-1.

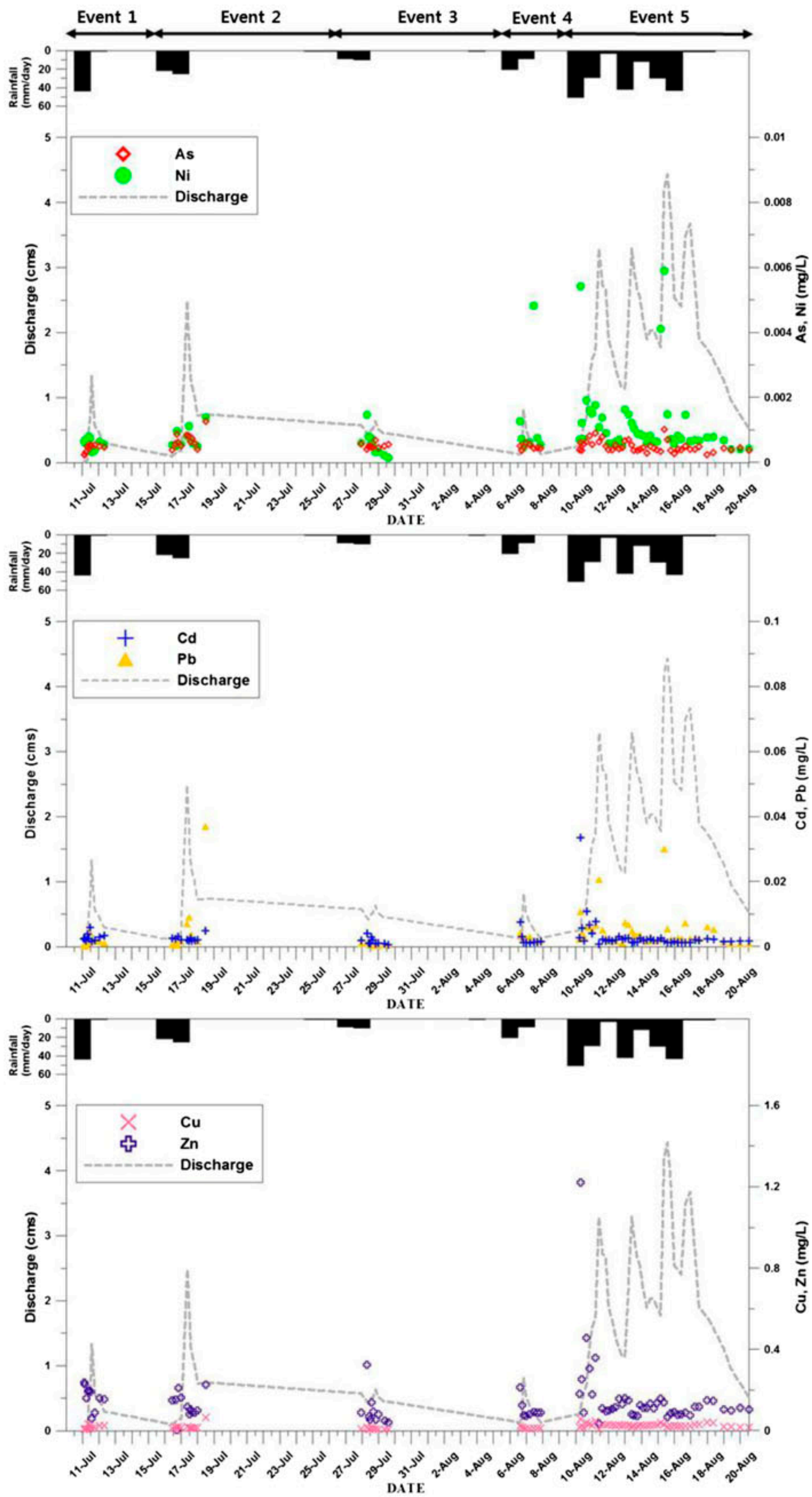

Fig. 5. Variation of dissolved metal concentrations during wet weather at GP-2.

Table 3
Comparison of metal loadings during dry and wet weather at GP-1 and GP-2

Station	Period	Month	As (kg/d)	Cd	Cu	Pb	Zn	Ni
GP-1	Dry weather	Jan	NF	NF	NF	NF	NF	NF
		Feb	NF	NF	NF	NF	NF	NF
		Mar	NF	NF	NF	NF	NF	NF
		Apr	0.0017	0.0675	0.8204	0.0045	5.6516	0.0071
		May	NF	NF	NF	NF	NF	NF
		Jun	NF	NF	NF	NF	NF	NF
		Jul	0.0011	0.2668	2.3842	0.0110	15.7656	0.0164
		Aug	0.0013	0.1255	1.1710	0.0073	8.4222	0.0085
		Sep	0.0012	0.1095	1.1576	0.0032	7.1598	0.0074
		Oct	ND	0.0017	0.0159	0.0007	0.1655	0.0005
		Nov	NF	NF	NF	NF	NF	NF
	Wet weather (Event maximum loading)	Event 1	0.0005	0.1427	1.4463	0.0074	6.5705	0.0106
		Event 2	0.0033	0.3027	1.8717	0.0366	9.3632	0.0191
		Event 3	0.0011	0.2668	2.3841	0.0110	15.7656	0.0164
		Event 4	0.0004	0.0356	0.1007	0.0038	0.8000	0.0014
		Event 5	0.0096	0.6994	10.921	0.3015	32.8960	0.0747
GP-2	Dry weather	Jan	0.0003	0.0026	0.0151	ND	0.3	0.0008
		Feb	0.0064	0.0419	0.4371	ND	4.4	0.0182
		Mar	0.0016	0.0240	0.1107	ND	2.1	0.0039
		Apr	0.0054	0.0302	0.2656	0.0081	2.5	0.0111
		May	0.0039	0.0386	0.7702	0.0062	3.3	0.0090
		Jun	NF	NF	NF	NF	NF	NF
		Jul	0.0053	0.0177	0.0751	0.0031	0.9	0.0039
		Aug	0.0062	0.0228	0.2744	0.0231	1.4	0.0091
		Sep	0.0087	0.0215	0.1911	0.0033	1.4	0.0031
		Oct	0.0003	0.0038	0.0129	0.0006	0.5	0.0010
		Nov	0.0005	0.0069	0.0255	0.0005	0.7	0.0019
	Wet weather (event maximum loading)	Event 1	0.0005	0.0024	0.0181	0.0021	0.1105	0.0004
		Event 2	0.1075	0.3084	4.1848	2.3832	15.0170	0.1119
		Event 3	0.0053	0.0177	0.0751	0.0031	0.8956	0.0039
		Event 4	0.0282	0.0917	0.5038	0.0458	6.3090	0.0326
		Event 5	0.1782	0.4388	7.5882	2.9629	29.6720	0.7424

Notes: NF = no flow; ND = not detected.

4.4. Comparison with other AMD sites

Several studies have performed wet-weather monitoring in mine areas to find out the characteristics of AMD [14,17,20,21]. These studies recognized metal release by flood events as an important physicochemical process that is used for the control of AMD. The report on metals in the abandoned Beatson Mine in Alaska [20] shows that the concentrations of Al, Fe, Cu, and Pb increased with increasing rainfall. As for the North Fork Clear Creek [14], storm events resulted in both increases and decreases in dissolved metal concentrations. Decreases were said to be most likely due to dilution of the stream water with storm water, while increases were said to be due to flushing of AMD sources. Results from the Rio Tinto study [17] show that the river transported approximately 1,000 times more water during rainfall than during the dry season and between 140 (Na) and 8,200 (Pb) times more dissolved elements, highlighting the extreme seasonality of water flow and pollutant transport, and the key role of flood events. Based on data from three mine sites in the United States and other sites outside the US, Nordstrom [21] observed that concentrations of Cu and Zn increased temporarily as the mine discharges increased in flow during rainstorm events. Overall, previous studies show the similar results with our study in that, following a rainfall event, metal concentrations and loadings significantly increase due to wash off of AMD sources.

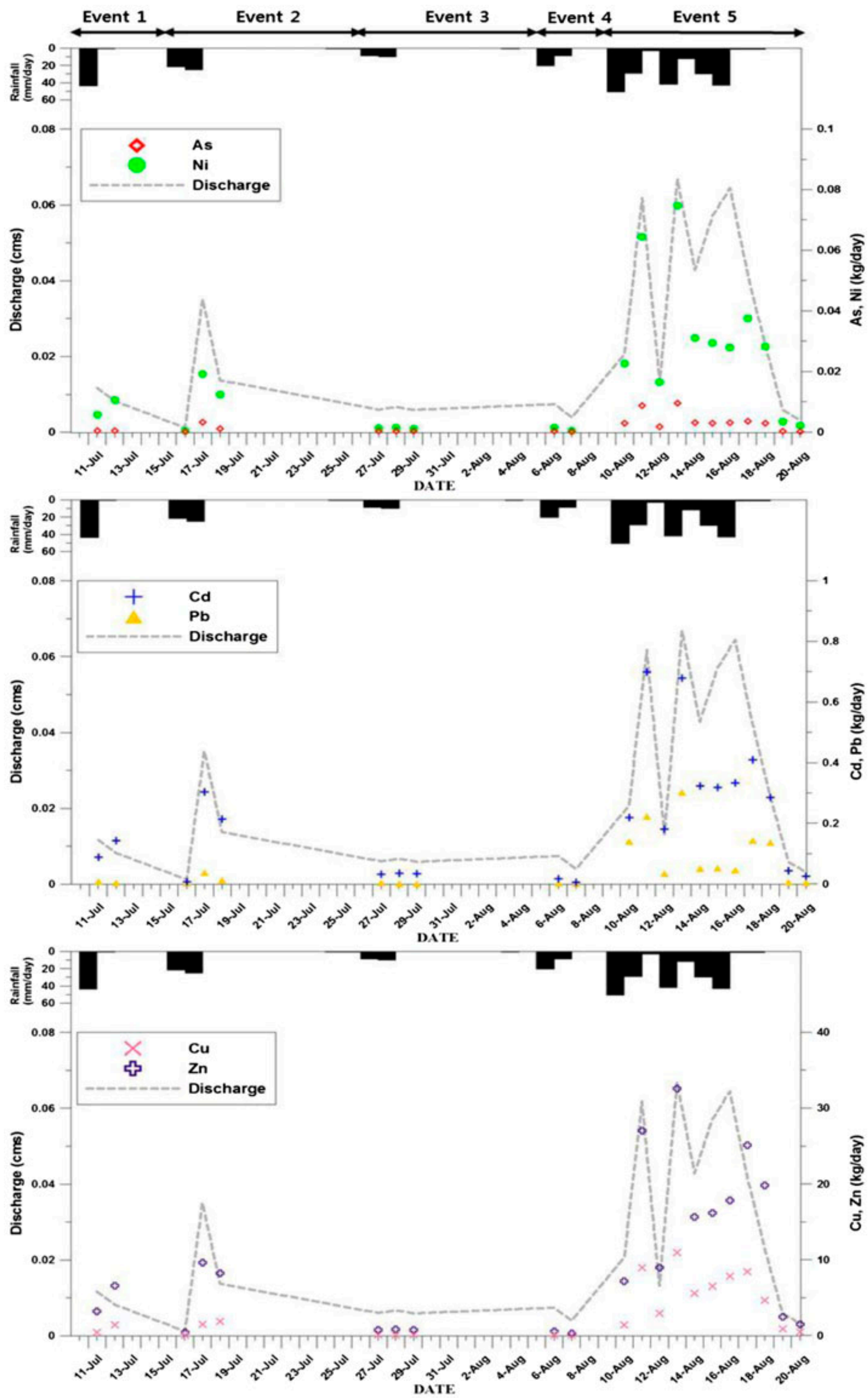

Fig. 6. Variations of dissolved metal loadings during rainfall at GP-1.

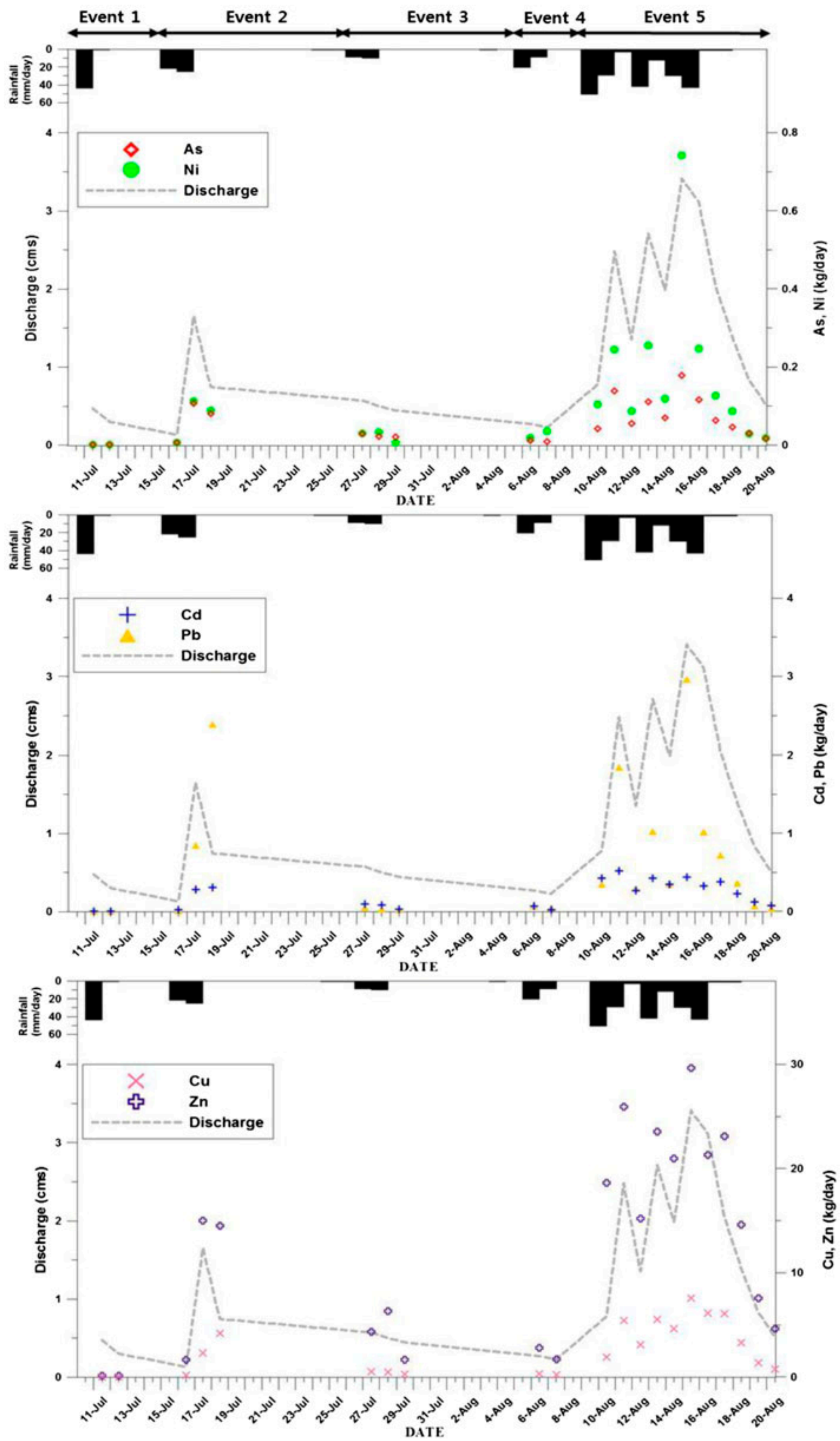

Fig. 7. Variations of dissolved metal loadings during rainfall at GP-2.

Table 4
Estimated annual metal load at GP-1 and GP-2 (1,029 mm rainfall)

Monitoring point	Period	As	Cd	Cu	Pb	Zn	Ni
GP-1	Annual (kg)	0.18	16.63	231.3	4.02	819.61	1.43
	Dry weather (%)	11.55	14.06	9.70	2.69	18.37	11.44
	Wet weather (%)	88.45	85.94	90.30	97.31	81.63	88.56
	Annual (kg)	5.03	21.52	246.84	42.2	1384.86	9.77
GP-2	Dry weather	23.37	29.79	26.98	3.26	38.43	19.32
	Wet weather	76.63	70.21	73.02	96.74	61.57	80.68

5. Conclusions

In this study, Geopung Mine Watershed was selected for investigation because of the presence of a highly polluted abandoned metal mine in the area. The characteristics of hydrochemical variations and contaminant loadings in AMD and receiving stream were analyzed.

As indicated from the results, metal concentrations are sensitively affected by total rainfall in that maximum concentrations during rainfall were significantly higher than before rainfall at points GP-1 and GP-2. Results of estimation for metal loadings also show how loadings evidently increased during the rainfall events. From this, it is understood as well that pollutants detected in the stream come from AMD with contaminants washed off from the mine area during rainfall.

The results of this study have important consequences for remediation efforts in abandoned mine areas. Although remediation measures consider the lithologic condition of a site [22,23], most of them are designed without taking into account the total rainfall, which could have an influence on the metal concentrations and loadings. However, based on the results of this study, the total rainfall is an important parameter that should be considered in determining metal loadings for the design of remediation systems. Long-term and high-frequency monitoring protocol used in this study can provide necessary support for such design improvement.

Acknowledgments

This research was financially supported by the National Institute of Environmental Research and Guem-River Environment Research Center. This research was also supported in part by a grant from Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2013R1A1A4A01007676) and by a grant (13CRTI-B052117-01) from Regional Technology Innovation Program (RTIP) and another grant from Advanced Water Management Research Program funded by Ministry of Land, Infrastructure and Transport of Korean government.

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