Physicochem. Probl. Miner. Process. 49(2), 2013, 621−629

www.minproc.pwr.wroc.pl/journal/

Physicochemical Problems of Mineral Processing

ISSN 1643-1049 (print)
ISSN 2084-4735 (online)

Received January 17, 2013; reviewed; accepted April 14, 2013

REMOVAL OF HAZARDOUS AIR POLLUTANTS BASED ON COMMERCIAL COAL PREPARATION PLANT DATA

Gulhan OZBAYOGLU

Faculty of Engineering, Atilim University, Incek, Golbasi, Ankara, Turkey, e-mail:gulhan@atilim.edu.tr

Abstract: This paper investigates the concentration, distribution, and rejection of hazardous air pollutants, specifically identified by the US Clean Air Act Amendments of 1990, based on commercial coal preparation plant data obtained on-site. The samples were collected from the products of the different cleaning circuits of the operating plant. The concentrations of twelve potentially hazardous trace elements, including As, Cd, Co, Cr, Hg, Mn, Ni, Pb, Se, Th and U in those samples were determined. Compared with the average concentration of the trace elements in Turkish coals, the run-of-mine coal fed to the existing plant appears to contain higher concentrations of Cd, Hg, Mn, Th and V. However, the concentrations of As, Cd, Cr, Mn, Se, Th, U and V of the run-of-mine coal are above the world averages. Cd, Cr, Hg, Mn, Pb and Th concentrations of run-of-mine coal were easily removed at commercial coal preparation plant refuse in the range of 51.8% to 77.4 %, while only a small reduction was achieved for U and V as they were concentrated in clean coals. The present study reveals that conventional coal preparation technologies could significantly reduce hazardous air pollutants concentrations in coal.

Key words: *hazardous air pollutants, coal preparation, Turkish lignites, trace elements rejection, dense medium separation*

Introduction

General

Many of the potentially hazardous air pollutants (HAP's for short) occur as trace elements in run-of-mine coals. The growing awareness of environmental issues has increased the attention on these elements (Coal Trading Blog, 2011; Gürdal, 2008; Pesek et al., 2005; Swaine, 1998). As coal will continue to be a major energy source to generate electricity worldwide, the trace elements which may cause hazardous emissions upon combustion of the coal have also become an increasingly important concern for those involved in the industry. The US Clean Air Act Amendments of 1990 specifically identifies As, Be, Cd, Cr, Co, Hg, Mn, Ni, Pb, Sb, Se and U as potential HAP's (Demir et al., 1998). These trace elements are known to be toxic and

http://dx.doi.org/10.5277/ppmp130221

have adverse effects on humans, plants and animals. Table 1 summarizes the average concentrations of a number of trace elements in Turkish coal samples compared against world averages (Tuncali et al., 2002).

Table 1. Potential hazardous air pollutants in Turkish lignites and the world (ppm)

Trace Elements	As	Be	Cd	Co	Cr	Hg	Mn	Ni	Pb	Se	Th	U	V
Average	53	1	1	9.4	109	0.1	123	126	12	2	6	13	87
Minimum	2	0.2	0.01	1	7	0.03	5	3	0.1	0.1	1	0.4	6
Maximum	686	7	45	55	580	0.7	691	1700	286	26	29	132	287
World average	10	2	0.5	5	20	-	70	20	20	1	4	2	40

As seen in Table 1, the concentration of these trace elements have a wide range of variation, and their amounts vary considerably from seam to seam and, in some cases, even within the same seam. During combustion at electrical utilities, trace elements may be released to the atmosphere as solid compounds with the fly-ash and, in the vapor phase, with the flue-gas (Yasushi et al., 2003; Esenlik et al., 2006; Zevenhoven et al., 2005; Ward, 2002). To control toxic air emissions, post-combustion and/or pre-combustion methods can be used (Vejahati et al., 2010; Gupta, 2006; Vassilev et al., 2001; Akers et al., 2000). Elements such as Sb, Be, Cd, Co, Pb and Mn are believed to be removed with fly-ash by applying post-combustion methods – that is, by electrostatic precipitators. On the other hand, trace elements such as As, Cl, Hg and Se have the potential to volatilize /vaporize and, consequently, cannot be controlled as effectively by conventional post-combustion techniques. However, coal cleaning, when applied before combustion to remove ash in coal, is effective in reducing the level of most of the trace elements, but the degree of removal of ash and trace elements are dependent on the coal, the degree of liberation of the trace elements bearing mineral matters, and the cleaning method of coal itself (Davidson, 1998). Although, physical cleaning may provide limited improvement in the quality of run-of-mine coal, it has been considered an economical and effective technique in minimizing environmental problems (Dangyu et al., 2007; Weng and Peng, 2003; Wang et al., 2009).

Unfortunately, there are limited information concerning of trace elements removal during coal washing in the world (Wang et al., 2009; Wang et al., 2003). Similarly, very limited data is available on the subject of trace elements removal from Turkish coals by coal preparation techniques (Özbayoğlu, 2010; Özbayoğlu, 2011).

As stated before, coal preparation is used commercially to improve the quality of coal by removing ash and sulfur. During the removing of ash, the trace elements which are associated with inorganic matter would be rejected autogenously, since there is a general relationship between overall ash removal and trace elements

rejection. Therefore, the objective of this investigation is to determine the HAP's partitioning and rejections that would be achieved in different coal cleaning circuits of an existing coal preparation plant in Turkey. Although, Imbat coal preparation plant was chosen for this study, trace elements reduction would be expected similarly in different ranges in the other coal preparation plants depending on coal, modes and occurrences of trace elements and cleaning methods used.

Coal preparation plants in Turkey

Coal is the main raw material to generate power for electricity in Turkey with a total reserve of about 12.88 petagrams (Pg or billion tons), of which amount 11.56 Pg (billion tons) is lignite and 1.32 Pg is bituminous coal. The Turkish coal sector produces both hard coal and lignite primarily used for power generation. Currently, there are around 45 coal preparation plants in operation in Turkey, with capacities ranging from 50 to 1000 mega grams per hour (tph), 12 to treat bituminous coal, and the rest to cover about 40% of the country's lignite production. Most of the lignite coal preparation plants are based on dense medium separation facilities. Static (drums, baths, vessels) and dynamic heavy medium (cyclones) equipment are used to treat coarse coal (+18 mm) and fine coal (0.5–18 mm), respectively. In this investigation, both the distribution and the possibility of the removal of trace elements are addressed in different cleaning circuits of the commercial Imbat coal preparation plant, whose flowsheet has been simplified in Fig. 1. This plant has a capacity of 350 Mg per hour, and it treats the coal coming from underground operations.

As seen from the flow sheet, the run-of-mine coal is crushed to 50 mm top-size before processing and, then, screened through 18 mm to separate the coal into coarse (+18 mm) and fine (–18 mm) size fractions. Drewboy is used for cleaning coarse size coal into two steps. The first Drewboy cleans the coal at a density of 1.80 g/cm^3 to remove coarse shale, followed by the second Drewboy working at 1.55 g/cm^3 density to produce clean coal and middling. Magnetite is used in the preparation of dense medium. The fine pieces (–18 mm) are deslimed at about 0.5 mm, and the 0.5–18 mm fraction is cleaned in dense medium cyclones at 1.55 g/ cm^3. The clean coals are screened through 10 mm and 0.5 mm screens to produce clean coals in sizes of 10–18 mm and 0.5–10 mm according to the market specifications. The –0.5mm-size coal is passed through hydrocyclones to recover the 0.1–0.5mm fraction and to remove the slime. Dewatering of the coarse and fine coal fractions is done in vibrating and screen-bowl centrifuges. Eventually, the products of +18 mm middling and hydrocyclones U/F are mixed together to be burnt in power plants.

There, at the coal preparation plant, the main problem is the clay, which clogs the openings of the screen surfaces causing an inefficient screening and mixing of clay particles to the bath of Drewboy. This condition has a diverse impact on both the performance and the capacity of the plant.

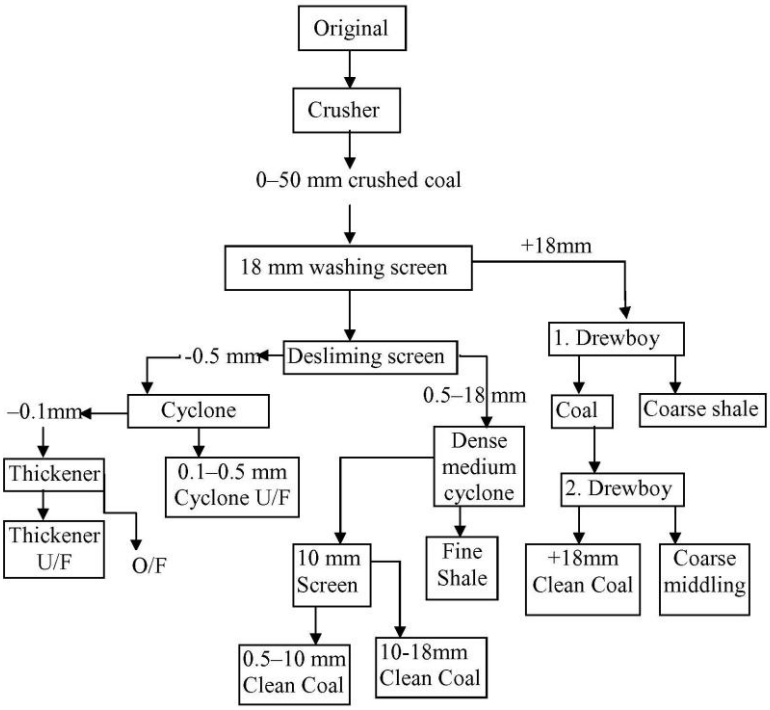

Fig. 1. Simplified flow sheet of Imbat Coal Preparation Plant

Materials and method

Systematic samples were collected from the run-of-mine coal (r.o.m.), the coarse and fine clean coals, coarse shale and middling of Drewboy and fine shale of dense medium cyclones, and the U/F of hydrocyclones and that of the tailing pond of the Imbat Coal Preparation Plant. The samples obtained were collected incrementally within 60-minute intervals in one day from the cleaning equipments. The trace elements analysis was carried out on dried representative samples of the mentioned products using the XRF and ICP-OES techniques.

For each trace element, the enrichment ratio – the ratio of the grade of concentrate (products) to that of the run-of-mine coal – was calculated. Also, the contents and distributions of trace elements in each cleaning circuit were determined. Finally, the rejection of trace elements was calculated.

Results and discussions

The proximate analysis and trace elements (hazardous air pollutants) contents of the run-of-mine coal and plant products were determined as illustrated in Tables 2 and 3.

Table 2. Proximate analysis of the samples (on dry basis)

Samples	Weight, %	Ash , %	Volatile Matter, %	Low Heat Value, MJ/kg	Total sulfur, %	Sulfur in ash %
+18mm clean coal	17.16	17.56	40.25	23.58	1.16	0.60
10–18mm clean coal	4.46	15.44	40.93	24.37	1.16	0.55
0.5–10mm clean coal	14.75	13.60	41.20	25.21	1.19	0.52
0.1–0.5mm cyclone U/F	2.92	30.94	37.13	18.59	1.05	0.74
Coarse middling	9.82	34.75	38.21	16.27	0.61	0.54
Coarse shale	28.67	62.70	36.98	1.49	1.03	0.23
Fine shale	17.96	64.16	35.92	1.54	0.24	0.57
Thickener U/F	4.26	44.53	25.24	6.17	0.96	0.43
Feed (run-of-mine)	100.00	42.54	38.08	11.90	0.69	0.46

Table 3. Trace elements contents of the samples (ppm)

Trace elements	As	Cd	Co	Cr	Hg	Mn	Ni	Pb	Se	Th	U	V
+18 mm clean coal	45.1	2.0	3.0	26.8	0.7	149.8	14.6	7.9	1.2	3.8	18.1	104.8
10–18 mm clean coal	46.7	2.0	3.0	27.5	1.0	71.3	15.0	7.4	1.1	3.8	17.8	132.1
0.5–10mm clean coal	51.2	0.5	3.0	30.8	1.0	58.7	16.5	8.3	1.1	4.1	18.6	165.2
0.1–0.5mm cyclone U/F	45.0	2.0	3.0	36.8	1.0	128.1	19.0	15.4	1.2	6.2	15.1	134.7
Coarse middling	44.8	2.0	5.4	35.2	0.6	50.9	16.5	12.9	1.1	5.6	14.5	117.6
Coarse shale	31.2	2.0	3.0	41.3	1.0	360.4	12.8	12.3	1.1	8.3	3.8	45.4
Fine shale	23.7	2.0	3.0	37.2	1.0	349.1	15.6	12.8	0.9	7.0	3.4	53.9
Thickener U/F	30.1	2.0	5.8	51.6	0.5	180.3	23.4	18.6	1.0	14.6	14.3	103.6
Feed (run-of-mine)	37.6	1.8	3.3	35.6	0.9	214.5	15.2	11.2	1.1	6.4	10.8	90.8

According to the results, the ash content of the run-of-mine coal treated in the conventional Imbat Coal Preparation Plant is high with a concentration level of

42.54% because of the underground operations. However, its total sulfur content is very low (0.69%).

The ash content of +18 mm clean coal is high as 17.56% due to the mixing and floating of undispersed clay particles with the coal. The ash content decreases with the decrease in coal size. When these three clean coals are mixed together, the combined products will have a weight percentage of 36.37% with an average ash content of 15.69% and average low heat value of 24.34 MJ/kg according to metallurgical balance calculations. By similar calculation, the total plant shale, which is a mixture of coarse and fine shale, will have 46.63% weight with a total ash removal 69.4%.

It can be observed that, among the hazardous air pollutants of the run-of-mine coal, Cd, Mn, Th, V and Hg contents are above the Turkish coal averages; yet, Co, Cr, and Ni contents are much lower. On the other hand, As, Cd, Cr, Mn, Se, Th, U and V contents of the run-of-mine coal are above the world averages as seen in Table 1. The enrichment ratios of the trace elements detected within the plant products are tabulated in Table 4.

As seen in Table 4, U and V enrichments almost appear in clean coal samples, while Mn and Th are enriched in the shale. Also, 0.1–0.5 mm slimes collected in U/F of cyclone and slimes precipitated in the thickener U/F are enriched in Cd, Cr, Ni, Pb, U and V. Among all the examined products, the highest enrichment is found for Th at 2.28, and the second highest enrichment for Co at 1.76, at the thickener U/F.

Table 4. Enrichment Ratio of the products

	As	Cd	Co	Cr	Hg	Mn	Ni	Pb	Se	Th	U	V
+ 18 mm clean coal	1.20	1.11	0.91	0.75	0.78	0.70	0.96	0.71	1.09	0.59	1.68	1.15
10–18 mm clean coal	1.24	1.11	0.91	0.77	1.11	0.33	0.99	0.66	1.00	0.59	1.65	1.45
0.5–10mm clean coal	1.36	0.28	0.91	0.87	1.11	0.27	1.09	0.74	1.00	0.64	1.72	1.82
0.1–0.5mm cyclone U/F	1.20	1.11	0.91	1.03	1.11	0.60	1.25	1.38	1.09	0.97	1.40	1.48
Coarse middling	1.19	1.11	1.64	0.99	0.67	0.24	1.09	1.15	1.00	0.87	1.34	1.30
Coarse shale	0.83	1.11	0.91	1.16	1.11	1.68	0.84	1.10	1.00	1.30	0.35	0.50
Fine shale	0.63	1.11	0.91	1.04	1.11	1.63	1.02	1.14	0.82	1.09	0.31	0.59
Thickener U/F	0.80	1.11	1.76	1.45	0.56	0.84	1.54	1.66	0.91	2.28	1.32	1.14

The trace elements contents and distributions of the plant products are calculated, and the extents of the removal of hazardous air pollutants are indicated in Table 5. Here, the trace elements retained in the clean coals and the ones which can be

removed with the shale can be seen. Summarized in the bottom row are the total amount of clean coal produced and the total amount of hazardous air pollutants rejection with the shale.

Table 5. Trace elements rejections (%) in different products

Trace elements	As	Cd	Co	Cr	Hg	Mn	Ni	Pb	Se	Th	U	V
+18mm clean coal	20.6	19.3	15.3	12.9	13.5	5.7	16.4	12.1	19.1	10.2	28.7	19.8
10–18mm clean coal	5.5	5.0	4.0	3.4	5.0	1.2	4.4	2.9	4.5	2.6	7.3	6.5
0.5–10mm clean coal	20.1	4.2	13.2	12.8	16.6	3.5	16.0	10.9	15.0	9.4	25.4	26.8
Middling*	15.2	14.3	18.4	12.7	9.9	8.6	14.2	15.3	13.3	11.4	17.2	17.0
Coarse shale	23.8	32.2	25.6	33.2	32.3	48.2	24.1	31.4	29.2	37.1	10.1	14.3
Fine shale	11.3	20.2	16.1	18.8	20.2	29.2	18.4	20.4	15.0	19.6	5.7	10.7
Thickener U/F	3.4	4.8	7.4	6.2	2.4	3.6	6.5	7.0	3.9	9.7	5.6	4.9
Total clean coals**	46.2	28.5	32.5	29.1	35.1	10.4	36.8	25.9	38.6	22.2	61.4	53.1
Total refuse***	35.1	52.4	41.7	52.0	52.5	77.4	42.5	51.8	44.2	56.7	15.8	25.0

* Mixture of coarse middling and cyclone U/F

** Mixture of +18mm, 10–18mm, 0.5-10mm clean coals

*** Mixture of coarse and fine shales

Parallel to the achievement in Table 4, Table 5 showed that among the twelve trace elements examined, six of them – namely, Cd, Cr, Hg, Mn, Pb and Th – were rejected between 51.8% and 77.4 % together with the refuse. These trace elements demonstrate affinity to the inorganic matter, and are concentrated in the sink products. On the other hand, 61.4% of U and 53.1% of V retained in the clean coal, because U and V have a strong association with organic matter and is difficult to remove by coal preparation methods.

It can be concluded that in a conventional coal preparation plant, without any advanced size reductions, it is possible to minimize the harmful effects of hazardous air pollutants once the ash is removed. Although advanced coal cleaning processes are often more successful in removing a higher proportion of trace elements, the economics may not be attractive in that they are relatively expensive and can result in rather wet and fine coal product, thus making it difficult to handle and transport.

Conclusions

The run-of-mine coal treated at Imbat Coal Preparation Plant contains potential hazardous air pollutants – namely, As, Cd, Co, Cr, Hg, Mn, Ni, Pb, Se, Th, U and V –

identified by the US Clean Air Act Amendments of 1990. These trace elements are known to be toxic and have adverse effects on humans, plants, and animals.

The conventional Imbat Coal Preparation Plant with a capacity of 350 Mg per hour, cleans the coal obtained from underground coal operations containing 42.54% ash, 38.08% volatile matter and 0.69% total sulfur in dry basis with 11.90MJ/kg low heating value. The trace elements of As, Cd, Cr, Mn, Se, Th, U and V within the run-of-mine coal are above the world averages while Cd, Mn, Th,V and Hg grades are above the Turkish coal averages. The plant produces 36.37% by weight total clean coal with 15.69% ash and 24.34 MJ/kg low heat value. The total ash rejection from the plant is 69.4%.

The trace elements in the run-of-mine coal, namely Cd, Cr, Hg, Mn, Pb and Th, all of which are associated with the inorganic portion of the coal were easily removed at Imbat Coal Preparation Plant. The highest rejection was obtained for Mn (77.4%), which was more than the plant ash rejection.

During the 69.34% of ash removal from coal, six trace elements mentioned above are rejected within a range of 51.8% to 77.4 % with the ash. However, 15.8% and 25.0% reductions were achieved for U and V, respectively due to their strong association with organic matrix.

Although conventional coal preparation processes in use today may not make up for a complete removal of any trace elements, it was proven that they are effective in reducing the concentration of many trace elements without any further size reduction, especially those present at relatively high concentrations. For further rejection, it is necessary to reduce the top size of the feed and to grind middling to enhance the liberation of trace elements as well as to improve the cleaning circuits by applying advanced coal cleaning technologies. Advanced coal cleaning technologies may offer better trace elements removal than conventional cleaning.

Acknowledgements

The author would like to thank TKI-ELI-Soma Chemical Laboratories staff to provide operational data and carry out the chemical analyses of the plant products.

References

AKERS D.J., ALUKO M.E., EKECHUKWU K.N., LEBOWITZ H.E., 2000, *Process for removal of hazardous air pollutants from coal*, US Patent No: 6156281.

COAL TRADING BLOG 2011, *Significance of trace elements in coal. An overview*, http://bestcoaltrading.blogspot.com/2011/significance-of-trace-elements-in-coal.htmp.

DANGYU S., YONG Q, JUNYING, Z., WENFENG, W, CHUGUANG, Z., 2007, *Concentration and distribution of trace elements in some coals from Northern China*, International Journal of Coal Geology, Vol. 69, Issue 3, 179–191.

DAVIDSON R., 1998, *Coal cleaning to remove trace elements. A review*, Coal Preparation, Vol.19, 159–176.

DEMIR I., RUCH R.R., DAMBERGER H.H., HARVEY R.D., STEELE J.D., HO K.K., 1998, *Environmentally critical elements in channel and cleaned samples of Illinois coals*, Fuel, Vol. 77, Issues 1–2, 95–107.

ESENLIK S., KARAYIGIT A.İ., BULUT Y., QUEROL X., ALASTUEY A., FONT O., 2006, *Element behaviour during combustion in coal-fired Orhaneli power plant*, Bursa-Turkey, Geologica acta, vol. 4, Issue 4, 439–449.

GUPTA R., 2006, *Advanced coal characterization: A review*, Presented at the Sino-Australia Symposium on Advanced Coal Utilization Technology, July 12–14, Wuhan, China.

GURDAL G., 2008, *Geochemistry of trace elements in Çan coal (Miocene)*, Çanakkale, Turkey, International Journal of Coal Geology, vol. 74, Issue 1,3, 28–40.

OZBAYOGLU G., 2011, *Partitioning of Major and trace elements of a Turkish lignite with size and density*, Physicochemical Problems of Mineral Processing, 47, 5160.

OZBAYOGLU G. 2010, *Potential of removing trace elements from a Turkish lignite*, International Journal of Coal Preparation and Utilization, 30, 322–333.

PESEK J., BENCKO V., SYKOROVA I., VASICEK M., MICHNA O., MARTINEK K., (2005), *Some trace elements in coal of the Czech Republic, environment and health protection implications*, Cent.Eur.J.Public Health, 13(3).153–158.

SWAINE D.J., 1998, *Trace elements during the mining and beneficiation of coal*, Coal Preparation, Vol. 19, 177–193.

TUNCALI E., CIFTCI B., YAVUZ N., TOPRAK S., KOKER A., GENCER Z., AYCIK H., SAHIN N., 2002, *Chemical and Technological Properties of Turkish Lignites*, General Directorate of Mineral Research and Exploration (M.T.A.), Ankara, Turkey.

WANG W., QIN Y., SONG D., 2003, *Cleaning potential of hazardous elements during coal washing, Journal of Fuel Chemistry and Technology*, DOI: cnki:ISSN:0253-2409.0.2003-04-002.

WANG W., QIN Y., WANG J., LI J., 2009, *Partitioning of hazardous trace elements during coal preparation*, The 6th. International Conference on Mining Science and Technology, Procedia Earth and Planetary Science 1, 838–844.

WARD C.R., (2002), *Analysis and significance of mineral matter in coal seams*, International Journal of Coal Geology, 50, 135–168.

WENG L., PENG S., 2003, *Effectiveness of coal preparation in removing hazardous trace elements in air*, Environmental Science and Technology, DOI: CNKI:SUN:FJKS. 0.2003-01-003.

VASSILEV S.V., ESKENAZY G.M., VASSILEVE C.G., 2001, *Behaviour of elelements and minerals during preparation and combustion of the Pernik coal*, Bulgaria, Fuel Processing Techology, Vol.72, Issue 2, 103–129.

VEJAHATI F., XU Z., GUPTA R., 2010, *Trace elements in coal: Associations with coal and minerals and their behavior during coal utilization – A review*, Fuel 89, 904–911.

YASUSHI S., KUNIHISA S., MIO O., EIICHI K., MASAHIKO M., 2003, *The release behavior of trace elements from coal during high temperature process*, Nippon Enerugi Gakkai Sekitan Kagaku Happyo Ronbunshu, vol.40. 136–137 (Japanese).

ZEVENHOVEN R., SAVOLAHTI J., VERHOVEN L., 2005, *Partitioning of mercury and other trace elements from coal and waste-derived fuels during fluidized bed pyrolysis*, 18th International Conference on Fluidized Bed Combustion, Toronto, Ontario, Canada.