

Investigation of Organic and Inorganic Contaminants in Water Sources around Elbistan Lignite Beds

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Keywords	Abstract
Kahramanmaraş	The household and industrial use, as well as mining of coal, pose various environmental and health risks
Lignite	including lung and kidney diseases such as Balkan Endemic Nephropathy (BEN). BEN is a kidney disease and it is geographically confined to Balkan countries where extensive lignite deposits are located.
Organic Contaminants	The most accepted cause of BEN is the use of untreated waters contaminated by lignite-derived organic contaminants. Afsin-Elbistan basin contains approximately 50 % of the lignite reserves of Turkey which
Inorganic Contaminants	makes it an ideal location for the current study. In this work, water samples were taken from 10 different
Groundwater	locations in the Afşin-Elbistan basin and they were analyzed for organic and inorganic contaminants. Results showed that none of the water samples were contaminated with inorganic contaminants and indeed met the Turkish drinking water standards. GC-MS analyses of the water samples revealed similar chromatograms. Briefly, all the water samples have similar organic compound types such as n-alkanes, chlorophyll-derived phytols, and plant- and animal-derived acids (i.e. palmitic acid). On the other hand, none of the samples were contaminated with carcinogenic and/or nephrotoxic organic compounds such
	as polyaromatic hydrocarbons and aromatic amines, which is contrary to many, but not all of the previous works conducted in Balkan countries. All these results may indicate that the influence of coal deposits on the groundwaters is minimal.

Cite

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1. INTRODUCTION

Coal is still extensively used all over the world for electricity generation, industrial purposes, and heating. As it is well known, there are serious health problems and environmental issues associated with coal mining and use (Ding et al., 2001; Tiwary, 2001). Low-rank coals, such as lignites, contain various toxic organic and inorganic compounds such as polyaromatic hydrocarbons (PAHs), aromatic amines, lead, and cadmium. Groundwater and surface water leaches some of these organic compounds from the lignite beds and the use of these waters may lead to health problems like kidney diseases and cancer.

One of the health problems (possibly) related to the low-rank coals is a kidney disease named Balkan Endemic Nephropathy (BEN; Feder et al., 1991; Finkelman et al., 2002; Orem et al., 1999; 2002). BEN is geographically restricted to Balkan countries where Pliocene lignites are extensively found. On the other hand, studies are showing BEN-like diseases outside of Balkan countries (i.e. U.S.A) which is called Pan-endemic Nephrophaty (PEN; Orem et al., 2007). There are numerous hypotheses for BEN aethiology such as exposure to toxins (Grollman & Jelaković, 2007; Lukinich-Gruia et al., 2022; Pfohl-Leszkowicz, et al., 2002; Stiborová et al., 2016, Stoev, 2017), to organic compounds leached from low-rank coal deposits (mainly lignites; Feder et al., 1991; Ojeda et al., 2019) and deficiency of some metals. However there are still some debates, the Pliocene lignite hypothesis is a plausible hypothesis mainly because of the high correlation between lignite deposit

locations and the BEN-endemic villages (Voice et al., 2006). The lignite hypothesis briefly suggests that organic contaminants leach into groundwater from lignite beds and the use of these untreated waters leads to acute exposure to low concentrations of nephrotoxic compounds which may lead to BEN (Orem et al., 1999).

Previous studies showed that untreated well waters taken from Balkan countries (mainly used for drinking) have numerous toxic and potentially toxic organic compounds such as PAHs (i.e. naphthalene, phenanthrene, anthracene, fluoranthene), phenolic compounds, heterocyclic compounds (N-, S-, O- containing cyclic compounds) and aliphatic compounds (Chakraborty et al., 2017; Orem et al., 1999; 2002). Leach experiments, indeed, indicated that water can leach many aromatic organic compounds from coal (Maharaj, 2014) therefore same compounds can be found both in the waters and in the coal samples (Maharaj et al., 2013; Maharaj, 2014; Orem et al., 2002). Concentrations of organic compounds in endemic area waters were found to be very low in concentration (usually <0,1 μ g/L) however both concentrations and the number of compound types are higher compared to control group samples (Finkelman, 2007; Orem et al., 1999). On the other hand, the inorganic compounds such as lead and cadmium and anions like nitrate, sulfate are almost identical in both endemic and control group samples which leads to exclusion of metal deficiency hypothesis.

Although numerous studies were done in the Balkan countries and the lignite hypothesis is one of the accepted cause of BEN (Maharaj, 2014), new studies conducted on other lignite bearing areas from different parts of the world will surely help to understand (a) if untreated waters which are being in use show the similar contamination patterns and (b) if BEN-like diseases are common in these regions as well.

According to Ministry of Energy and Natural Sources (2017) data, approximately 50 % of the lignite reserves of Turkey, with a measured reserve amount of approximately 4 billion tonnes, are found in the Afşin – Elbistan basin. Considering the massive lignite reserves and water pollution being the secondary priority problem in Kahramanmaraş (Directorate General of Environmental Impact Assessment, 2018), this region is geographically ideal for the present study. The overarching aim of this study was to understand if untreated well waters taken from the Afşin-Elbistan basin are contaminated with (lignite-derived) organic and inorganic compounds and to see if the results are in line with the previous studies. In addition, although a BEN-like disease has not been reported in the study area yet, the results of this work may be a reference study for similar works and for the research on the kidney health of the residents of the area.

2. MATERIAL AND METHODS

2.1. Study Area and Sampling

Water samples were collected from Berçenek, Alemdar, Çoğulhan, Kışlaköy and Çomudüz villages of Afşin-Kahramanmaraş (Table 1; Figure 1). All of the villages are near the Afşin-Elbistan thermal power plant and they are located on coal beds. 9 untreated well water samples and 1 surface water sample (sample number 7, as a control sample) were collected in the field. On every sampling location, in-situ measurements (i.e. pH and temperature) were conducted.

Different sampling methodologies were followed for different analyses types. For organic compounds sampling, all glasswares were solvent-cleaned (acetone) before use to avoid any organic contamination. On each location, on-site filtration was conducted by using a glass vacuum filtration apparatus with a manual pump and 0,45 μ m glass fiber filters attached. 2 liters of water sample was filtered and collected in pre-cleaned glass bottles and 60 ml of methylene chloride (DCM) was added to each bottle to retard bacterial activity during storage. When sampling is done at one location, glass fiber is removed and all glass parts of the filtering apparatus was cleaned with acetone and DCM. Upon arrival to the laboratory, water samples were stored in the fridge until extraction.

For inorganic analyses, 100 ml water samples were filtered through cellulose syringe filters $(0,45\mu m)$ and collected in polyethylene sampling bottles. Upon arrival to the laboratory, water samples were stored in the fridge. These samples were directly sent to KSU-USKİM (Kahramanmaraş Sütçü İmam University) and ACME Laboratories (Canada) for anion and cation analyses respectively.

Sample No	Latitude Longitude		Location	pН	Temperature
1	38°18'02.2''N	37°04'30.9"E	Kuşkayası	6,6	13.3
2	38°20'32.7"N	37°01'02.4"E	Çoğulhan	7.1	15
3	38°21'51.3"N	37°01'36.0"E	Alemdar	7.2	17.3
4	38°22'02.4"N	37°01'35.4"E	Alemdar	7.1	17.3
5	38°23'25.3"N	37°00'24.6"E	Çomudüz	7.3	13.6
6	38°23'30.5"N	37°00'18.5"E	Çomudüz	6.7	15.4
7	38°25'11.6"N	36°54'55.6"E	Tanır	7.3	11.1
8	38°23'11.4"N	36°58'49.1"E	Berçenek	7.4	14.8
9	38°22'58.9"N	36°58'41.6"E	Berçenek	7.2	14.1
10	38°25'22.9"N	36°59'08.3"E	Büget	7.1	14.0

 Table 1. Coordinates of Sampling Locations



Figure 1. Map of the Sampling Locations

2.2. Extractions and Instrumental Analyses for Organic Analyses

Extractions of the water samples were done in Geochemistry Laboratory, Kahramanmaraş Sütçü İmam University. The extraction methodology was adapted from Orem et al. (2007). Briefly, water samples were extracted by liquid/liquid extraction with four successive volumes of GC-MS grade DCM (60ml). The combined total lipid extract (240ml, TLE) was reduced to a volume of a few milliliters by rotory evaporation which was concentrated to 200 μ l by evaporation under a nitrogen blow-down unit. A fraction of TLE was then used during gas chromatography/mass spectrometry (GC-MS) analyses.

GC-MS analyses were done on the TLE extracts (ULUTEM research facilities, Gaziantep University). Assignment of organic compounds was performed using a Shimadzu QP2020 GC-MS. A RESTEK Rxi-5ms column (30 m x 0.25 mm x 0.25 µm, 95 % dimethyl polysiloxane, 5 % diphenyl) was used for GC-MS analyses. The GC/MS operation settings were as described in Chakraborty et al. (2017).

Sample No	Fluoride (ppm)	Chloride (ppm)	Sulfite (ppm)	Nitrite (ppm)	Nitrate (ppm)	Phosphate (ppm)
1	0,38	14,77	54,31	BDL	32,8	BDL
2	1,8	6,05	99,54	BDL	7	BDL
3	0,36	37,98	98,69	BDL	84,42	BDL
4	0,44	16,28	38,7	BDL	58,91	BDL
5	0,16	2,47	30,19	BDL	7,43	BDL
6	0,22	36,31	40,7	BDL	127,61	BDL
7	0,08	0,77	4,26	BDL	4,43	BDL
8	0,37	9,84	25,68	BDL	29,1	BDL
9	0,31	14,67	38,05	BDL	40,69	BDL
10	0,3	48,46	24,34	BDL	168,16	BDL
Mean	0,44	18,76	45,45	_	56,06	_

 Table 2. Concentrations of Anions

3. RESULTS AND DISCUSSION

3.1. Inorganic Analyses

Anion analyses showed that in all samples, nitrite and phosphate concentrations were below detection limits which are 0.054 ppm and 0.114 ppm respectively (Table 2; Figure 2). Fluoride concentrations ranged between 0.08-1.8 ppm (mean value of 0.44 ppm); chloride concentrations were between 0.77 - 48.5 ppm (mean value of 18.45 ppm); sulfate and nitrate concentrations ranged between 4.3-99.5 ppm and 7-168.2 ppm (mean values of 45.45 ppm and 56.06 ppm), respectively. Sample 2 showed the highest fluorite and sulfate concentrations with being 1.8 ppm and 99.54 ppm respectively. Sample 10 had the maximum chloride concentration (48.46 ppm) and nitrate concentration (168.16 ppm) while the control sample (sample 7) had the lowest concentrations of all anions.

These anion results showed that all well water samples, although they are mainly used for irrigation purposes, meet the Turkish regulations for drinking water criteria (Doğrul Selver & Uras, 2022). Considering the TS266 and WHO drinking water standards (Table 3), four of the samples exceed the acceptable limit for nitrate (sample number 3, 4, 6, and 10; 50 mg/L) on the other hand, all samples were below the acceptable limit for sulfate (250 mg/L), fluoride (1.5 mg/L) as well as chloride (250 mg/L; Table 2 and 3).

Orem et al. (2002) reported high nitrate concentrations (225 mg/l) in untreated well water samples taken from both BEN endemic and non-endemic sites which suggests that high nitrate concentrations can not be associated to BEN etiology. In this study, maximum nitrate concentration was measured in the water sample taken from the Büget region (sample number 10; 168.8 ppm) which is a large farmyard. On the other hand, in all the samples sulfate concentrations are much higher (4.2 - 99.5 mg/L) which is similar to what Orem et al. (2002) reported.

Cation results showed that none of the samples appear to be contaminated with heavy metals or with the metals suggested to have nephrotoxic effects. For instance, among toxic heavy metals, Hg, Cd, and Fe concentrations were below the detection limits in all water samples (Figure 3, Supplementary Table 1). All the other heavy metals (i.e. Cr, Co, Cu, Mn, Ni, and Zn) had concentrations below the minimum allowed limit for drinking water (Table 3). In addition, there was no major difference between the control group sample (sample 7) and

the other samples. Similarly, previous studies indicated that metal concentrations are similar in endemic and non-endemic sites (Batuman, 2006; Orem et al., 2002; 2004). Cd and Pb were suggested to have nephrotoxic effects and therefore it was suggested that these metals should be analyzed in well waters as well (Fowler et al., 2004; Orem et al., 1999; Weeden, 1991). In all the water samples analyzed in the present study, Pb and Cd concentrations are below detection limits (except for sample 8 and sample 10 for Pb concentration). In addition, As, B, Br, Cl, Cr, F, Li, Na, P, Rb, Se, Sr, and W are found to be statistically related to renal cancer rates in northwest Lousiana (Bunnell et al., 2006) however in the current study none of these elements are higher than allowed limits as well. Consequently, inorganic contaminants results are in line with the previous studies showing insignificant concentration values to be evaluated for environmental and health aspects.

Element	WHO Standard (1993)	TS266 (2005)	Element	WHO Standard (1993)	TS266 (2005)
Al (µg/L)	200	200	Mn (μg/L)	100	50
As (µg/L)	10	10	Ni (µg/L)	70	20
B (μg/L)	2400	1000	Pb (µg/L)	10	10
Ba (µg/L)	700	_	Zn (µg/L)	3000	_
Cd (µg/L)	3	5	Flouride (mg/L)	1.5	1.5
Cr (µg/L)	50	50	Chloride (mg/L)	250	250
Cu (µg/L)	2000	2000	Nitrate (mg/L)	50	50
Fe (µg/L)	300	200	Sulfate (mg/L)	250	250
Hg (µg/L)	1	1	Nitrite (mg/L)	3	0,5

Table 3. WHO and Turkish Drinking Water Standards

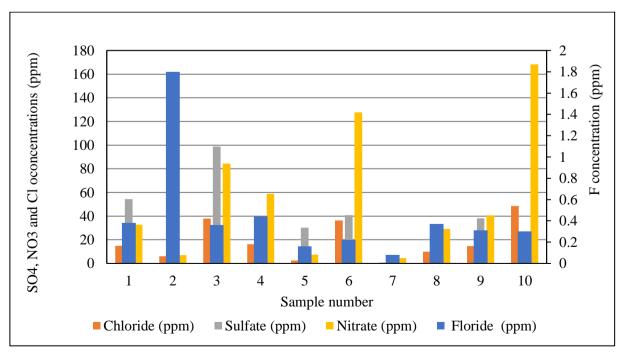


Figure 2. Histograms Showing Concentrations of Anions

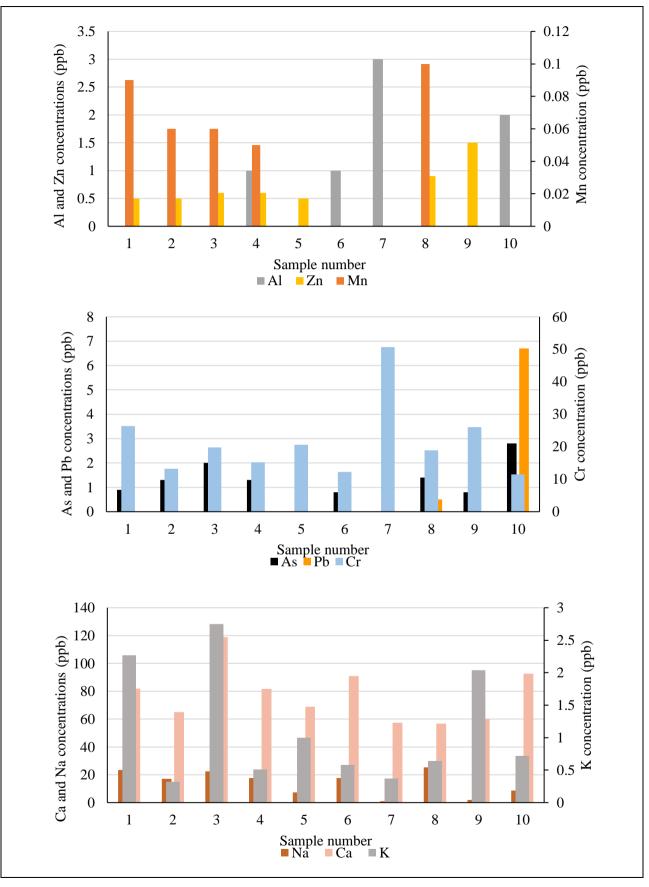


Figure 3. Histograms Showing Concentrations of Different Metals

3.2. Organic Analyses

A non-target screening methodology has been applied therefore the concentrations of compounds were not calculated. Similar chromatograms were obtained for all samples and major peaks in the chromatograms were identified. As a representative of all samples, labelled gas chromatogram of sample 9 was shown in Figure 4. Briefly, results showed that all water samples mainly contain n-alkanes, some animal and plant derived compounds (i.e. phytol and palmitic acid) and phthalates. None of the target compounds as being carcinogenic and nephrotoxic natures, such as PAHs and aromatic amines, were observed.

n-alkanes (C12-C34) are abundantly found in the samples which could indicate the addition of plant-derived compounds into the water sources. Palmitic acid which is a common compound found in plants and animals, and chlorophyll indicators such as phytol were also found in almost all the water samples.

Earlier studies indicated that, in well water samples taken from BEN endemic areas, the concentration and the number of organic compounds are higher than those of the non-endemic regions (Feder et al., 1991; Maharaj, 2014; Orem et al., 2002; 2007). On the other hand, the GC-MS results of the present study revealed that the organic compound composition of all the water samples is similar to each other. In almost all the water samples, phthalates, the most widespread man-made contaminants, were found. This finding is in line with the earlier studies where they found abundant and numerous phthalates (Bunnell et al., 2006; Maharaj, 2014). Bunnell et al. (2006) suggested that these phthalates are mainly originated from PVC well casings and plastic parts of the pump systems. Contamination during sample handling and extraction can be another explanation for phthalates in the water samples.

Most of the previous studies (Feder et al., 1991; Maharaj, 2014; Orem et al., 2002; 2007) showed that water samples taken from BEN-endemic areas contain coal- originated organic compounds such as PAHs, aliphatic& aromatic compounds, aromatic amines, some of which are known as nephrotoxic and/or carcinogenic compounds. In contrast, water samples taken from Afşin-Elbistan area are not contaminated with these abovementioned nephrotoxic and carcinogenic organic compounds, which may suggest a minimal influence of coal deposits on the groundwater quality. This outcome is parallel to the results of Iordanidis et al. (2016) and Kosateva et al. (2017a) reported. PAHs were not found in Amyneto lignites, Northern Greece and similarly coal derived PAHs were also not found in groundwater samples taken from Amyneto lignite basin which in turn indicates a minor influence of lignite deposits on groundwaters (Iordanidis et al., 2016). In other studies, Kosateva et al. (2017a) did leach experiments on Bulgarian lignites for 10 weeks by using different solvents and analytical equipments however did not find any harmful/ toxic organic compounds in the leachates (Kosateva et al., 2017a; 2017b). Considering all these, the absence of PAHs and other toxic and/or carcinogenic compounds in groundwaters related with lignites, as in the present study, and in leachets of lignites is not confounding.

4. CONCLUSIONS

In the current study, 10 water samples taken from the Afşin-Elbistan Basin were analyzed for organic and inorganic contaminants to understand whether these sources are contaminated and to evaluate the possible effects of coal beds on these water sources. Inorganic analyses showed that none of the toxic heavy metals including the ones that suggested to have nephrotoxic and/or carcinogenic effects (such as Cd, Pb, Fe and Al) are above the drinking water standards. This finding is in-line with most of the previous studies. In addition, organic analyses indicated that all water samples have similar organic matter compositions and have limited organic compound variation. To examplify; while n-alkanes, phthalates, and some other common plant- & animal- derived organic compounds are present in almost all water samples, coal-derived compounds (such as PAHs and aromatic amines) are not found in any of the water samples. The lack of coal-derived organic molecules in well water samples may suggest that the effect of coal deposits on the groundwater quality is trivial. Surely, this work can be a reference study for similar prospective studies (on the research of the kidney health of the local people of the area as well) however, triplicate GC-MS runs for each sample and concentration measurements of the organic compounds would allow for more robust interpretations.

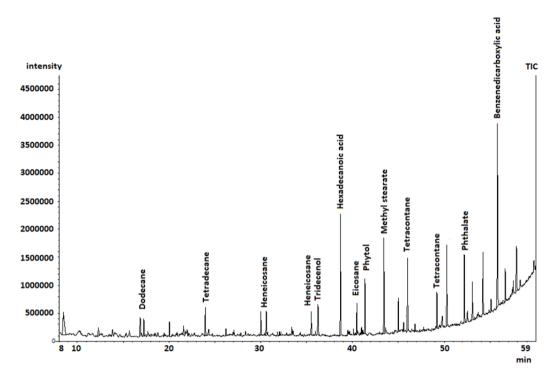


Figure 4. GC-MS chromatogram of sample 9

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

REFERENCES

Batuman, V. (2006). Fifty years of Balkan endemic nephropathy: Daunting questions, elusive answers. *Kidney International*, 69(4), 644–646. doi:10.1038/sj.ki.5000231

Bunnell, J. E., Tatu, C. A., Bushon, R. N., Stoeckel, D. M., Brady, A. M. G., Beck, M., Lerch, H. E., McGee, B., Hanson, B. C., Shi, R., & Orem, W. H. (2006). Possible linkages between lignite aquifers, pathogenic microbes, and renal pelvic cancer in northwestern Louisiana, USA. *Environmental Geochemistry and Health*, 28(6), 577–587. doi:10.1007/s10653-006-9056-y

Chakraborty, J., Varonka, M., Orem, W., Finkelman, R. B., & Manton, W. (2017). Geogenic organic contaminants in the low-rank coal-bearing Carrizo-Wilcox aquifer of East Texas, USA. *Hydrogeology Journal*, 25(4), 1219–1228. doi:10.1007/s10040-016-1508-6

Ding, Z., Zheng, B., Long, J., Belkin, H. E., Finkelman, R. B., Chen, C., Zhou, D., & Zhou, Y. (2001). Geological and geochemical characteristics of high arsenic coals from endemic arsenosis areas in southwestern Guizhou Province, China. Applied Geochemistry, 16(11–12), 1353–1360. doi:10.1016/S0883-2927(01)00049-X

Directorate General of Environmental Impact Assessment, Permit and Inspection (2018). Türkiye Çevre Sorunları ve Öncelikleri Değerlendirme Raporu (2016 yılı verileriyle). <u>PDF</u>

Doğrul Selver, A., & Uras, Y. (2022, June 16-18). Organic and Inorganic Contaminants In Water Sources Around Elbistan Basin. In: International Symposium on Advanced Engineering Technologies. Kahramanmaraş.

Feder, G., Radovanovic, Z., & Finkelman, R. (1991). Relationship between weathered coal deposits and the etiology of Balkan endemic nephropathy. *Kidney International*, 40(34), S9–S11.

Finkelman, R. B. (2007). Health impacts of coal: facts and fallacies. *Ambio*, 36(1), 103–106. doi:10.1579/0044-7447(2007)36[103:HIOCFA]2.0.CO;2

Finkelman, R. B., Orem, W., Castranova, V., Tatu, C. A., Belkin, H. E., Zheng, B., Lerch, H. E., Maharaj, S. V., & Bates, A. L. (2002). Health impacts of coal and coal use: Possible solutions. *International Journal of Coal Geology*, *50*(1–4), 425–443. doi:10.1016/S0166-5162(02)00125-8

Fowler, B. A., Whittaker, M. H., Lipsky, M., Wang, G., & Chen, X.-Q. (2004). Oxidative stress induced by lead, cadmium and arsenic mixtures: 30-Day, 90-day, and 180-day drinking water studies in rats: An overview. *BioMetals*, *17*(5), 567–568. doi:10.1023/B:BIOM.0000045740.52182.9d

Grollman, A. P., & Jelaković, B. (2007). Role of Environmental Toxins in Endemic (Balkan) Nephropathy. *Journal of the American Society of Nephrology*, *18*(11), 2817–2823. doi:<u>10.1681/ASN.2007050537</u>

Iordanidis, A., Schwarzbauer, J., & Gudulas, K. (2016). Organic Pollutants in the Groundwaters Used for Irrigation Purpose Within a Coal-Bearing Basin of Northern Greece. *Bulletin of the Geological Society of Greece*, *50*(4), 2155–2162. doi:10.12681/bgsg.14268

Kosateva, A. D., Stefanova, M., Marinov, S. P., & Gonsalvesh, L. (2017a). Organic components in leachates from some Bulgarian lignites. *Bulgarian Chemical Communications*, 49(D), 25–29.

Kosateva, A. D., Stefanova, M., Marinov, S., Czech, J., Carleer, R., & Yperman, J. (2017b). Characterization of organic components in leachables from Bulgarian lignites by spectroscopy, chromatography and reductive pyrolysis. *International Journal of Coal Geology*, *183*(October), 100–109. doi:10.1016/j.coal.2017.10.005

Lukinich-Gruia, A. T., Nortier, J., Pavlović, N. M., Milovanović, D., Popović, M., Drăghia, L. P., Păunescu, V., & Tatu, C. A. (2022). Aristolochic acid I as an emerging biogenic contaminant involved in chronic kidney diseases: A comprehensive review on exposure pathways, environmental health issues and future challenges. *Chemosphere*, 297(February). doi:10.1016/j.chemosphere.2022.134111

Maharaj, S. V. M. (2014). Limitations and plausibility of the Pliocene lignite hypothesis in explaining the etiology of Balkan endemic nephropathy. *International Journal of Occupational and Environmental Health*, 20(1), 77–91. doi:10.1179/2049396713Y.000000046

Maharaj, S. V. M., Orem, W. H., Tatu, C. A., Lerch, H. E., 3rd, & Szilagyi, D. N. (2013). Organic compounds in water extracts of coal: Links to Balkan endemic nephropathy. Environmental Geochemistry and Health, 36(1), 1–17. doi:10.1007/s10653-013-9515-1

Ministry of Energy and Natural Resources. (2017). Kömür. (Accessed:01/01/2017) <u>enerji.gov.tr/bilgimerkezi-tabiikaynaklar-komur</u>

Ojeda, A. S., Ford, S. D., Gallucci, R. M., Ihnat, M. A., & Philp, R. P. (2019). Geochemical characterization and renal cell toxicity of water-soluble extracts from U.S. Gulf Coast lignite. *Environmental Geochemistry and Health*, 41(2), 1037–1053. doi:10.1007/s10653-018-0196-7

Orem, W. H., Feder, G. L., & Finkelman, R. B. (1999). A possible link between Balkan endemic nephropathy and the leaching of toxic organic compounds from Pliocene lignite by groundwater: Preliminary investigation. *International Journal of Coal Geology*, 40(2–3), 237–252. doi:10.1016/S0166-5162(98)00071-8

Orem, W. H., Tatu, C. A., Feder, G. L., Finkelman, R. B., Lerch, H. E., Maharaj, S. V. M., Szilagyi, D., Dumitrascu, V., Paunescu, V., & Margineanu, F. (2002). Environment, geochemistry and the etiology of balkan endemic nephropathy: lessons from Romania. *Facta Universitatis, Medicine and Biology Series*, 9(1), 39–48.

Orem, W. H., Tatu, C. A., Lerch, H. E., Susan V. M. Maharaj, N. P., Paunescu, V., & Dumitrascu, V. (2004). Identification and environmental significance of the organic compounds in water supplies associated with a Balkan endemic nephropathy region in Romania. *Journal of Environmental Health Research*, *3*(2), 53–61.

Orem, W., Tatu, C., Pavlovic, N., Bunnell, J., Lerch, H., Paunescu, V., Ordodi, V., Flores, D., Corum, M., & Bates, A. (2007). Health effects of toxic organic substances from coal: Toward "panendemic" nephropathy. *Ambio*, *36*(1), 98–102. doi:10.1579/0044-7447(2007)36[98:heotos]2.0.co;2

Pfohl-Leszkowicz, A., Petkova-Bocharova, T., Chernozemsky, I. N., & Castegnaro, M. (2002). Balkan endemic nephropathy and associated urinary tract tumours: A review on aetiological causes and the potential role of mycotoxins. *Food Additives and Contaminants*, *19*(3), 282–302. doi:<u>10.1080/02652030110079815</u>

Stiborová, M., Arlt, V. M., & Schmeiser, H. H. (2016). Balkan endemic nephropathy: an update on its aetiology. *Archives of Toxicology*, 90(11), 2595–2615. doi:10.1007/s00204-016-1819-3

Stoev, S. D. (2017). Balkan Endemic Nephropathy – Still continuing enigma, risk assessment and underestimated hazard of joint mycotoxin exposure of animals or humans. *Chemico-Biological Interactions*, 261, 63–79. doi:10.1016/j.cbi.2016.11.018

Tiwary, R. K. (2001). Environmental Impact of Coal Mining Onwater Regime and Its Management. *Water, Air, & Soil Pollution, 132*(1-2), 185-199. doi:10.1023/A:1012083519667

Voice, T. C., McElmurry, S. P., Long, D. T., Dimitrov, P., Ganev, V. S., & Peptropoulos, E. A. (2006). Evaluation of the hypothesis that Balkan endemic nephropathy is caused by drinking water exposure to contaminants leaching from Pliocene coal deposits. *Journal of Exposure Science & Environmental Epidemiology*, *16*, 515–524. doi:10.1038/sj.jes.7500489

Weeden, R. P. (1991). Environmental renal disease: lead, cadmium and Balkan endemic nephropathy. *Kidney International Supplement*, *34*.

Supplementary Table 1. Cation Concentrations of the Water Samples (in ppb (μ g/L))

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Sample No	Ag	Al	As	Au	В	Ba	Be	Bi	Br	Ca	Cd	Ce	Cl	Со
1	BDL	BDL	0.9	BDL	206	88.55	BDL	BDL	231	81.94	BDL	BDL	13	0.03
2	BDL	BDL	1.3	BDL	222	105.65	BDL	BDL	96	64.94	BDL	BDL	9	0.06
3	BDL	BDL	2	BDL	125	42.44	BDL	BDL	176	118.8	BDL	BDL	37	0.02
4	BDL	1	1.3	BDL	132	73.01	BDL	BDL	143	81.75	BDL	BDL	17	BDL
5	BDL	BDL	BDL	BDL	38	98.35	BDL	BDL	43	68.82	BDL	BDL	4	BDL
6	BDL	1	0.8	BDL	67	73.89	0.06	BDL	142	90.81	BDL	BDL	30	BDL
7	BDL	3	BDL	BDL	8	6.23	BDL	BDL	7	57.28	BDL	BDL	1	BDL
8	BDL	BDL	1.4	BDL	81	74.96	BDL	BDL	65	56.7	BDL	BDL	10	BDL
9	BDL	BDL	0.8	BDL	58	95.62	BDL	BDL	65	60.04	BDL	BDL	14	BDL
10	BDL	2	2.8	BDL	32	20.2	BDL	BDL	89	92.55	BDL	BDL	42	BDL
Sample No	Cr	Cs	Cu	Dy	Er	Eu	Fe	Ga	Gd	Ge	Hf	Hg	Но	In
1	26.4	BDL	0.4	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
2	13.2	BDL	0.7	BDL	0.02	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
3	19.8	BDL	1.2	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
4	15.2	BDL	0.5	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
5	20.6	0.01	0.5	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
6	12.2	0.03	0.6	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
7	50.7	BDL	0.2	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
8	18.9	BDL	0.4	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
9	26	0.01	0.6	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
10	11.5	BDL	0.5	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Sample														
No	K	La	Li	Lu	Mg	Mn	Мо	Na	Nb	Nd	Ni	Р	Pb	Pd
1	2.27	BDL	9.5	BDL	12.4	0.09	1.6	23.34	BDL	BDL	BDL	13	BDL	BDL
2	0.32	BDL	15	BDL	24.63	0.06	5.4	17.11	BDL	BDL	2.3	BDL	BDL	BDL
3	2.75	BDL	11.2	BDL	25.01	0.06	1.1	22.39	BDL	BDL	0.7	BDL	BDL	BDL
4	0.51	BDL	11.4	BDL	21.52	0.05	0.8	17.69	BDL	BDL	0.5	BDL	BDL	0.03
5	1	BDL	11.9	BDL	15.34	BDL	0.6	7.33	BDL	BDL	BDL	14	BDL	0.02
6	0.58	BDL	8.6	BDL	13.17	BDL	0.5	17.76	BDL	BDL	0.6	BDL	BDL	0.02
7	0.37	BDL	0.6	BDL	4.18	BDL	0.2	0.97	BDL	BDL	BDL	BDL	BDL	BDL
8	0.64	BDL	9.6	BDL	12.59	0.1	1.3	25.4	BDL	BDL	BDL	BDL	0.5	0.02
9	2.04	BDL	15.5	BDL	18.11	BDL	0.8	2.033	BDL	BDL	0.3	23	BDL	BDL
10	0.72	BDL	2.2	BDL	14.8	BDL	0.6	8.57	BDL	BDL	BDL	BDL	6.7	BDL

Supplementary Table 1. (continued)

Supplementary Tuble 1. (commune)													
Pr	Pt	Rb	Re	Rh	Ru	S	Sb	Sc	Se	Si	Sm	Sn	Sr
BDL	BDL	0.53	0.18	0.01	BDL	15	BDL	BDL	2.7	11404	BDL	BDL	517.11
BDL	BDL	0.31	0.03	0.03	BDL	25	0.1	3	1.6	13558	BDL	BDL	1147.04
BDL	0.02	0.49	0.27	0.03	BDL	24	BDL	4	0.9	16080	BDL	BDL	916.47
BDL	BDL	0.32	0.27	0.02	BDL	12	BDL	3	1.5	12841	BDL	0.18	878.35
BDL	BDL	0.94	0.01	0.01	BDL	9	BDL	2	BDL	10971	BDL	BDL	458.68
BDL	BDL	0.43	0.07	0.02	BDL	10	BDL	2	1.1	10524	BDL	BDL	556.98
BDL	BDL	0.18	0.01	0.01	BDL	2	BDL	BDL	BDL	3332	BDL	BDL	85.84
BDL	BDL	0.54	0.01	0.01	BDL	9	BDL	3	0.9	12297	BDL	BDL	486.56
BDL	0.02	0.87	0.02	0.02	BDL	10	BDL	3	0.5	11538	BDL	BDL	569.81
BDL	0.03	0.43	0.03	0.03	BDL	9	BDL	3	BDL	10705	BDL	BDL	628.74
Та	Tb	Te	Th	Ti	Tl	Tm	U	V	W	Y	Yb	Zn	Zr
BDL	BDL	BDL	BDL	BDL	BDL	BDL	1.99	4.3	BDL	BDL	BDL	0.5	BDL
BDL	BDL	BDL	BDL	BDL	BDL	BDL	11.32	8.1	BDL	BDL	BDL	0.5	BDL
BDL	BDL	0.13	BDL	BDL	BDL	BDL	1.04	9.7	BDL	BDL	BDL	0.6	BDL
BDL	BDL	BDL	BDL	BDL	BDL	BDL	3.93	5.7	BDL	BDL	BDL	0.6	BDL
BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.36	2.6	BDL	BDL	BDL	0.5	BDL
חח	BDL	BDL	BDL	BDL	BDL	BDL	0.99	2.2	BDL	BDL	BDL	BDL	BDL
BDL	DDL												
BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.14	BDL	BDL	BDL	BDL	BDL	BDL
			BDL BDL	BDL BDL	BDL BDL	BDL BDL	0.14	BDL 6.6	BDL BDL	BDL BDL	BDL BDL	BDL 0.9	BDL BDL
BDL	BDL	BDL											
	BDL BDL BDL BDL BDL BDL BDL BDL BDL BDL	BDL BDL BDL BDL BDL BDL BDL BDL BDL BDL BDL BDL BDL 0.02 BDL 0.03 BDL 0.03 BDL 0.03 BDL 0.03 BDL BDL BDL BDL BDL BDL BDL BDL	BDL BDL 0.31 BDL 0.02 0.49 BDL BDL 0.32 BDL BDL 0.94 BDL BDL 0.94 BDL BDL 0.94 BDL BDL 0.43 BDL BDL 0.43 BDL BDL 0.43 BDL BDL 0.18 BDL 0.02 0.87 BDL 0.03 0.43 BDL 0.03 0.43 BDL 0.03 0.43 BDL BDL B.7 BDL BDL B.1 BDL BDL BDL BDL BDL BDL	BDL BDL 0.31 0.03 BDL 0.02 0.49 0.27 BDL BDL 0.32 0.27 BDL BDL 0.32 0.27 BDL BDL 0.32 0.27 BDL BDL 0.94 0.01 BDL BDL 0.43 0.07 BDL BDL 0.43 0.01 BDL BDL 0.54 0.01 BDL 0.02 0.87 0.02 BDL 0.03 0.43 0.03 BDL 0.03 0.43 0.03 BDL BDL BDL BDL BDL BDL BDL BDL	BDL BDL 0.31 0.03 0.03 BDL 0.02 0.49 0.27 0.03 BDL BDL 0.32 0.27 0.02 BDL BDL 0.32 0.27 0.02 BDL BDL 0.94 0.01 0.01 BDL BDL 0.43 0.07 0.02 BDL BDL 0.43 0.07 0.02 BDL BDL 0.43 0.01 0.01 BDL BDL 0.54 0.01 0.01 BDL 0.02 0.87 0.02 0.02 BDL 0.03 0.43 0.03 0.03 BDL 0.03 0.43 0.03 0.03 BDL BDL BDL BDL BDL BDL BDL <td< td=""><td>BDL BDL 0.31 0.03 0.03 BDL BDL 0.02 0.49 0.27 0.03 BDL BDL BDL 0.32 0.27 0.02 BDL BDL BDL 0.94 0.01 0.01 BDL BDL BDL 0.43 0.07 0.02 BDL BDL BDL 0.18 0.01 0.01 BDL BDL BDL 0.54 0.01 0.01 BDL BDL 0.02 0.87 0.02 0.02 BDL BDL 0.03 0.43 0.03 0.03 BDL BDL BDL BDL BDL BDL</td><td>BDL BDL 0.31 0.03 0.03 BDL 25 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL 0.94 0.01 0.01 BDL 9 BDL BDL 0.43 0.07 0.02 BDL 10 BDL BDL 0.43 0.01 0.01 BDL 2 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 0.02 0.87 0.02 0.02 BDL 10 BDL 0.03 0.43 0.03 0.03 BDL 9 BDL 0.03 0.43 0.03 0.03 BDL 9 BDL</td><td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL BDL BDL 0.43 0.01 0.01 BDL 2 BDL BDL BDL 0.54 0.01 0.01 BDL 9 BDL BDL 0.02 0.87 0.02 0.02 BDL 10 BDL BDL 0.03 0.43 0.03 0.03 BDL 9</td></td<> <td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 BDL BDL 0.18 0.01 0.01 BDL 9 BDL 3 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 3 BDL 0.02 0.87 0.02 0.02 BDL 10 BDL 3 BDL 0.03 0.43 0.03 0.03 BDL 9 BDL<</td> <td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 BDL BDL 0.18 0.01 0.01 BDL 2 BDL <t< td=""><td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3322 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL 0.03<td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3332 BDL BDL BDL 0.18 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL BDL 0.02 0.87 0.02 0.02 BDL 10<!--</td--><td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL 0.18 BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL BDL 0.97 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL</td></td></td></t<></td>	BDL BDL 0.31 0.03 0.03 BDL BDL 0.02 0.49 0.27 0.03 BDL BDL BDL 0.32 0.27 0.02 BDL BDL BDL 0.94 0.01 0.01 BDL BDL BDL 0.43 0.07 0.02 BDL BDL BDL 0.18 0.01 0.01 BDL BDL BDL 0.54 0.01 0.01 BDL BDL 0.02 0.87 0.02 0.02 BDL BDL 0.03 0.43 0.03 0.03 BDL BDL BDL BDL BDL BDL	BDL BDL 0.31 0.03 0.03 BDL 25 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL 0.94 0.01 0.01 BDL 9 BDL BDL 0.43 0.07 0.02 BDL 10 BDL BDL 0.43 0.01 0.01 BDL 2 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 0.02 0.87 0.02 0.02 BDL 10 BDL 0.03 0.43 0.03 0.03 BDL 9 BDL 0.03 0.43 0.03 0.03 BDL 9 BDL	BDL BDL 0.31 0.03 0.03 BDL 25 0.1 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL BDL BDL 0.43 0.01 0.01 BDL 2 BDL BDL BDL 0.54 0.01 0.01 BDL 9 BDL BDL 0.02 0.87 0.02 0.02 BDL 10 BDL BDL 0.03 0.43 0.03 0.03 BDL 9	BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 BDL BDL 0.18 0.01 0.01 BDL 9 BDL 3 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 3 BDL 0.02 0.87 0.02 0.02 BDL 10 BDL 3 BDL 0.03 0.43 0.03 0.03 BDL 9 BDL<	BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 BDL BDL 0.18 0.01 0.01 BDL 2 BDL BDL <t< td=""><td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3322 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL 0.03<td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3332 BDL BDL BDL 0.18 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL BDL 0.02 0.87 0.02 0.02 BDL 10<!--</td--><td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL 0.18 BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL BDL 0.97 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL</td></td></td></t<>	BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3322 BDL BDL 0.54 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL 0.03 <td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3332 BDL BDL BDL 0.18 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL BDL 0.02 0.87 0.02 0.02 BDL 10<!--</td--><td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL 0.18 BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL BDL 0.97 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL</td></td>	BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL BDL BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL 0.43 0.07 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL BDL 0.43 0.07 0.02 BDL 2 BDL BDL 3332 BDL BDL BDL 0.18 0.01 0.01 BDL 9 BDL 3 0.5 11538 BDL BDL 0.02 0.87 0.02 0.02 BDL 10 </td <td>BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL 0.18 BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL BDL 0.97 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL</td>	BDL BDL 0.31 0.03 0.03 BDL 25 0.1 3 1.6 13558 BDL BDL BDL 0.02 0.49 0.27 0.03 BDL 24 BDL 4 0.9 16080 BDL BDL BDL BDL 0.32 0.27 0.02 BDL 12 BDL 3 1.5 12841 BDL 0.18 BDL 0.94 0.01 0.01 BDL 9 BDL 2 BDL 10971 BDL BDL BDL BDL 0.97 0.02 BDL 10 BDL 2 1.1 10524 BDL BDL