Comparison of Enrichment Factors for Heavy Metals in Urban Street Dust and Air Aerosols

Pavel KANTOR[1)], Barbora ŠVÉDOVÁ[2)], Jarmila DROZDOVÁ[3)], Helena RACLAVSKÁ[4)], Marek KUCBEL[5)], Konstantin RACLAVSKY[6)]

[1)] Ing.; VSB – Technical University Ostrava, Institute of Geological Engineering and ENET Centre, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic; pavel.kantor@vsb.cz
[2)] Ing., Ph.D.; VSB – Technical University Ostrava, Department of Power Engineering and ENET Centre, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic; barbora.svedova@vsb.cz
[3)] Ing., Ph.D.; VSB – Technical University Ostrava, Institute of Geological Engineering and ENET Centre, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic; jarmila.drozdova@vsb.cz
[4)] Prof., Ing.; VSB – Technical University Ostrava, Institute of Geological Engineering and ENET Centre, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic; helena.raclavska@vsb.cz
[5)] Ing., Ph.D.; VSB – Technical University Ostrava, Institute of Environmental Engineering and ENET Centre, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic; marek.kucbel@vsb.cz
6) Prof., Ing.; VSB – Technical University Ostrava, ENET Centre, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic; konstantin.raclavsky@vsb.cz

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Abstract

The article deals with the possibility of comparison of enrichment factors in air aerosols (PM_{10}) and in the street dust, which is influenced by air pollution. For Pb and Sb, the same value of enrichment factor for both matrices was determined. The highest enrichment index (> 100) in air aerosols was found for Cd, Sb, and As, and also for Pb, and Zn (approximately 100). Enrichment factor shows significant differences in the summer and winter period, for Cd, Sb, As, Pb, and Zn it is > 2x higher in winter. Enrichment factor in street dust was also evaluated using geochemical background of soils from Olomouc. It was found that the highest value of enrichment factor is achieved by Cu, which comes from transport (probably bus operation).

Keywords: heavy metals, street dust, enrichment factor, Clarke value, geochemical background, soils

Introduction

Street dust often contains both industrial and vehicle-generated pollutants (Wang et al., 2012). It is an important environmental indicator of heavy metal contamination from atmospheric deposition (Yildirim and Tokalioglu, 2015). Street dust can be used for monitoring and identification of air pollution sources (Bucko et al., 2010).

Road dust consists of heterogeneous particles that are affected by the soil environment, anthropogenic activity and climatic conditions. Soil minerals form the main component (60%) of street dust. From this amount, quartz represents 40-50%, other main mineral phases are minerals from the feldspar group and phyllosilicates (chlorite and muscovite). Organic matter of plant origin form approximately 2%. The remaining particles come from transport (exhaust and non-exhaust emission) (Gunawardana et al., 2012). Metals in urban road dust have geogenic (soil erosion with subsequent resuspension of particles) or anthropogenic origin: long-distance transport, road traffic, industrial activity and power industry, local heating (Adamiec et al., 2016). During abrasion of tyres, Zn, Cd, Co, Cr, Cu, Hg, Mo, and Ni are released into the environment. The braking systems release mainly Fe, and also Cu, Sb, Ba, Al, Si, S, Ti, Zn, Ni, Cr, and Pb as well as a small amount of Cd (Adachia and Tainoshob 2004, Hjortenkrans et al., 2007). Corrosion and wear of metal parts of cars can be a source of Cu, Ni, and Cr (Al-Khashman and Shawabkeh, 2006). An important aspect for determining the environmental load by risk elements is the calculation of enrichment factor, which allows the identification of anthropogenic sources and geogenic resources. If EF is referenced to the same reference element, it is applicable to both environments – total suspended particles (TSP) and street dust. The anthropic activity results in a consequence of the increase of EF > 10 in air aerosol (Gelado-Caballero et al., 2012). Values higher than 10 in TSP appear for V, Zn, Cd, Sb, and Pb, which are generally enriched in aerosol derived from oil and other combustion and industrial processes (e.g., metallurgy). Primarily lithogenic elements, e.g. Fe and Mn, have also been observed as enriched in anthropogenically impacted aerosols (Shelley et al., 2015).

The aim of the article is to demonstrate the possibility of using enrichment index in air aerosol and street dust to identify the anthropogenic origin of the elements and to demonstrate the effect of incineration on the area load based on the change in their concentrations in air aerosols in the heating and non-heating seasons. To verify this method, the city of Olomouc,

the sixth largest city in the Czech Republic was used. The dominant sectors of industrial production are food processing and machinery. The chemical industry, plastics processing, electronics production, and construction industry are located in this place too.

Materials and methods

Sampling sites and analytical methods

Samples of street dust were obtained by brushing from an area of 1 m² (asphalt surface) at 21 localities in the city of Olomouc (autumn 2017). The locations of the sampling sites within the studied area are shown in Fig. 1. Sampling of the street dust was performed at roads with relatively low traffic. Samples were screened on sieves with hole size > 2 mm (gravel), 2 – 0.063 mm (sand) and < 0.063 mm (silt). Particle size class < 0.063 mm was used for chemical analysis in order to eliminate the particle size distribution on concentrations of elements (Sierra et al, 2015). The analyses were performed according to the U.S. EPA method 6200:2005 using portable X-ray fluorescence spectrometry (Innov-X DELTA PROFESSIONAL). The X-ray powder diffraction analysis was used for identification of mineralogical composition of dust particle samples (samples 10 and 20). Concentrations of risk elements in the street dust can be significantly influenced by pollution sources located in the near vicinity. Localities were divided into the three groups according to the prevailing source of pollution: localities influenced by industrial activity: 1, 2, 4, 9, 10, and 19; localities influenced by traffic (in the vicinity of roads with high traffic load): 6, 8, 18, and 20; localities without an important pollution source: 3, 5, 7, 11, 12, 13, 14, 15, 16, 17, and 21. In the year 2016 during the heating and non-heating seasons, four-day samplings for determination of EF in TSP were performed using high-volume sampler ThermoScientific in the area of Olomouc-Řepčín – locality 4 (Moravian Ironwork, a.s.) and Olomouc-Slavonín, locality 5 (built-up area). In the year 2017, samplings were performed during transitional and winter seasons in the street Velkomoravská, locality 6 (the main transport line through the city) and in the street Luční, locality 2 (without influence of traffic). Chemical analysis of risk elements in TSP was performed in accredited laboratories of ZUOVA (Health Institute in Ostrava) using the method of X-ray fluorescence (SPECTRO).

Statistical and assessment methods

Exploratory data analysis (EDA), statistical analysis for descriptive statistics, and correlation analysis were performed using the statistical software Statgraphics Plus 5.0 and OriginPro 8.5. The Kolmogorov-Smirnov test and Shapiro-Wilk test were performed for normality testing of the datasets (the significance level of $p < 0.05$). The map of anomalous concentrations was created using Surfer 9 (Golden Software, Inc.). Because most of the data were not normally distributed, the logarithmic transformation was performed to ensure the assumption of normality. The map was constructed using the geostatistical gridding method - kriging (the logarithmic variogram model, without drift). The best model was selected based on the cross-validation results. Anomalous concentrations of heavy metals correspond to values greater than the third quartile (75% percentile) of transformed data. Finally, the transformed data were converted using back-transformation into the normal (original) values that appear on the map.

Enrichment factors (EFs) can be utilized to distinguish between the metals originating from a natural procedure and those from human activities, and they were calculated from Eq. 1.

$$EF_{crust} = (X/R)_{air}/(X/R)_{crust} \quad (1)$$

in which X_{air} is content of the examined element in the PM_{10} or street dust, X_{crust} is Clarke value of examined element (Taylor, 1964), Rcrust is Clarke value of Fe.

If EF is < 1, the element is depleted in the environment and crustal sources are predominant. If enrichment factor is > 1, the element is relatively enriched in the environment. In general, EF > 5 indicates that a large fraction of the element can be attributed to non-crustal or anthropogenic sources (Wu et al., 2007). Different authors in literature have various opinions on the enrichment degree expressed for air aerosols by means of EF. Shelley et al. (2015) states that only samples with EF value > 10 can be considered as showing significant enrichment.

Results and discussion

The minimum and maximum content of risk factors in PM_{10} for individual periods is shown in Table 1. Enrichment index was calculated from the median value. The highest enrichment index in the range of 100–1000 was found for Cd, Mo, and Sb, which is consistent with EF reported by Enamorado-Baez et al. (2015) for the Huelva region of Spain. A higher enrichment index value (PM_{10} in Olomouc) than reported by the literature was found for As, Mo, and Zn in the winter season. Comparing the EF values in winter and in summer, it is clear that for most elements enrichment factor is higher in the winter season than in the summer one. The highest difference of up to 10 times was found in Mo, Zn (4x), As (3x), Cd, Ni, Pb, Sb (2x), Cu (1.2x). On the contrary, Mn enrichment index decreased, it was higher in summer than in winter (3x). It is clear from the above results that combustion processes have more than 50% share in increasing the concentrations of As, Cd, Ni, Mo, Pb, Sb, and Zn in the winter period.

Tab. 1. Concentrations of risks elements (minimum and maximum values for the monitored period) and enrichment factors EF

	Winter (min-max) ng/m^3	Transition (min-max) ng/m^3	Summer (min-max) ng/m^3	EF winter	EF trasition	EF summer	Enamorado Baez et al. (2015)
As	3-4	0.8-2.26	1.01-1.29	244	116	87	10-100
Cd	15-18	1.61	7.97-10.85	1,029	754	642	100-1,000
Cr	3-4	1.22-2.47	2.07-2.79	4.2	3.2	3.2	1-10
Cu	10-12	3.47-13.7	7.04-8.85	24	17	19	10-100
Fe	396-542	274-554	380-466				
Mn	10-12	3.37-10.8	23.29-49.46	1.4	1.2	5.3	1-10
Mo	81-85	12.6-22.5	7.22-9.19	683		74	10-100
Ni	18-19	8.86-14	1.4-8.5	31	15	10	1-10
Pb	13-16	5.65-7.89	4.7-7.4	141	81	64	100
Sb	22-24	4.27-4.89	7.4-10.6	1,424	541	617	100-1,000
Se	2		2	11	12	14	100-1,000
V	2-2.5		0.7-0.87	1.8		0.8	1-10
Zn	60-80	52.7-69.2	16.7-17.5	120	63	32	100

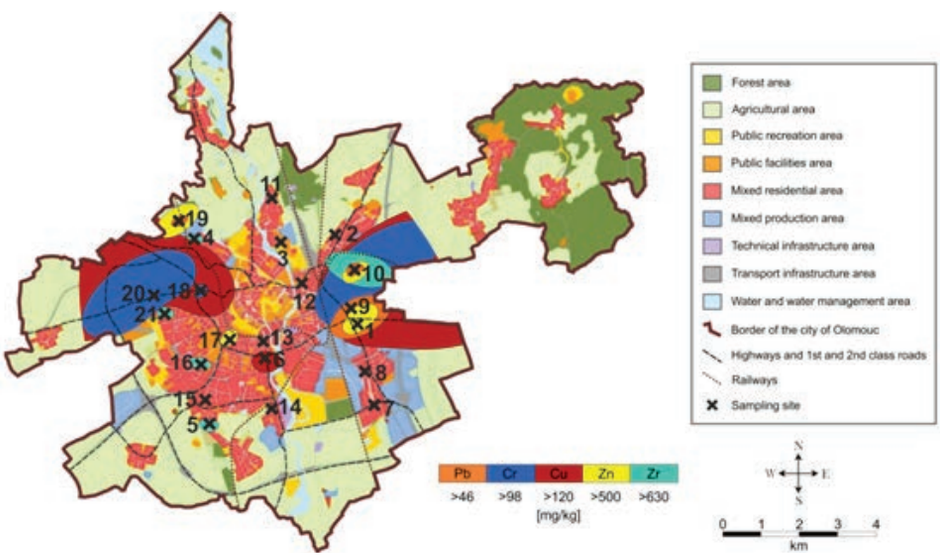

Fig. 1. Anomalous concentrations of Pb, Cr, Cu, Zn, and Zr in the city of Olomouc

In order to assess the impact of transport on the pollution load in the city of Olomouc, street dust sampling was performed. On average, up to 73% of particles in street dust are sand particles (particle size 0.063 – 2 mm), only 18% of the particles are larger than 2 mm and only 9% of the particles represent a grain size < 0.063 mm. From mineralogical analysis it follows that street dust is formed mostly by quartz (48 – 62%) and feldspars (albite 18 – 25% and orthoclase 5 – 6.2%). Feldspars come from resuspension and erosion of soils and quartz is released from soil or building materials (Sýkorová et al., 2017). Other crystalline phase identified in street dust samples include phyllosilicates: muscovite (8 – 10%) and chlorite (4 – 5%). Also calcite (2 – 6%) was identified at locality 20. In the sample from locality 10 (U panelárny), also minor concentration of akermanite (Ca2MgSi2O7) was found (2%). Akermanite is a common component of the high-furnace slag. It can be present in concrete in which Portland cement was partly replaced by high-furnace slag (Jackson et al., 2014).

The anomalous concentrations of the elements occurred mainly in the western and eastern part of the city (Fig. 1). The eastern part, in which the anomalous concentrations of Cr, Zn, Cu, and partly Pb and Zr were identified, is mainly affected by industrial sources (localities 1, 9, 10). Near highly frequented roads in the western part of Olomouc (locations 18, 20), the occurrence of anomalous concentrations of Cu and Cr was identified.

Nine elements (As, Cr, Cu, Mn, Ni, Pb, Sb, Zr, and Fe) were used to calculate street dust enrichment index, which also corresponded to the PM10 analysis. The identification of anthropogenic stress using street dust risk elements was performed for Cu, Cr, Zn, Pb, Fe, and Zr.

The main component method shows that higher Fe, Cu, and Cr content is typical for the localities affected by transport, the localities affected by industry are characterized by high levels of Zn and Pb (Figure 2). The association of elements involved in the identification of pollution sources follows from the results

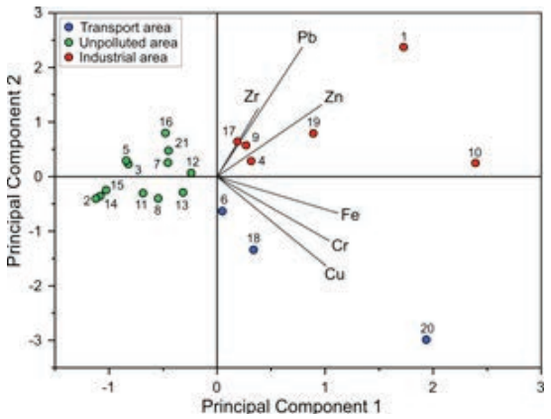

Fig. 2. Principal component analysis

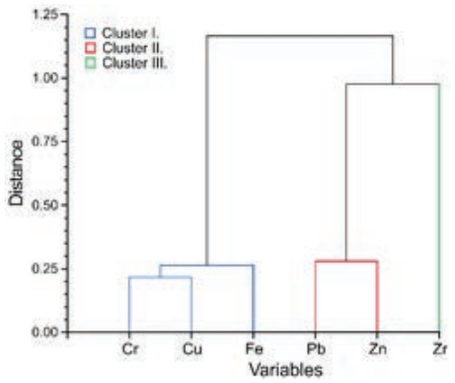

Fig. 3. Cluster analysis

of the cluster analysis (Figure 3). Zirconium, which is of geogenic origin in this area (occurrence of zircon) has a separate cluster status. Anomalous Zr values were found in unpolluted areas (locality numbers 5, 16, and 21). The occurrence of Zr is related to geogenic factors, it is present in particles released from the soil environment rather than from anthropogenic activity (De Vos et al., 2006).

The highest concentrations of heavy metals in street dust were found for localities Nos. 10 and 20. Locality No. 10 with concentrations of Cr (412 mg/kg), Zn (868 mg/kg), Cu (211 mg/kg), and Zr 811 mg/kg) is located in the industrial area (engineering, metal machining, concrete production, etc.). Locality No. 20, which is heavily loaded with Cr (349 mg/kg) and Cu (312 mg/kg), is located close to major traffic arteries (car and tram transport). Cu and Cr can be an important part of asphalt surfaces Adamiec et al., (2016). The highest concentration of Pb (91 mg/kg) was found at location No. 1. An important source of Pb is apparently the Olomouc heating plant combusting hard coal, situated to the west of this locality. For this locality, the highest concentrations of sulphur (2938 mg/kg) and As (13 mg/kg) were found. The elements As, Pb, and S in road dust are considered to be trace elements from coal combustion (Borbély-Kiss et al. 1998). Anomalous Pb values were also found for industrial localities Nos. 9 (50 mg/kg), 10 (55 mg/kg), and 19 (48 mg/kg).

The concentrations of Fe and Cr are higher than the results of Pal et al. (2011) in Scotland and Khoder et al. (2012) in Saudi Arabia. On other hand, the concentrations of Cu and Zn are comparable with Pal et al., (2011) and Khoder et al., (2012). The concentrations of Pb are lower than in Scotland and Saudi Arabia, see Table 2.

Correlation analysis of the concentrations of Cr, Fe, Cu, Zn, Zr, and Pb in street dust samples in the city of Olomouc (n = 21) was performed. Since most variables were not normally distributed and had many outliers, Spearman's correlation coefficient (rs) was used, at the significance levels of $p < 0.05$ and 0.01. The results are shown in Table 3. Significant linear relationship (rs = 0.69 to 0.84 at the significance level of 0.01) was found among Cr, Fe, Cu, and Zn. A slightly weaker linear dependence (at the significance level of 0.05) was proven between Cr and Pb (rs = 0.44), Fe and Pb (rs = 0.54), and between Zn and Pb (rs = 0.70).

The results of the correlation analysis suggest that most metals in road dust are mutually affected by a common source of pollution. The amount of metals released when the road surface dilapidates is also significant. At present, only very small amounts of Pb are emitted from transport. Significant dependence between Pb and Zn documents the predominance of other pollution effects (combustion processes and industry). Iron concentrations evaluated by traffic intensity showed high correlations with Pb, Cu, and Mn (Spearman r >0.50) in less frequented roads. Similarly, it was observed that Fe correlated with Pb, Cu, and Zn tobe greater than 0.50 for samples collected on minor roads. Interestingly, Fe correlation with Zn was as high as 93 % (r = 0.93) on major roads (Apeagyei et al., 2011).

Table 4 shows the results of enrichment index calculated according to Equation 1 and the results calculated from the value of the geochemical background for soils in Olomouc. The values > 5 that represent the enrichment of elements due to anthropogenic activity are marked grey and brown (Wu et al., 2007). The comparison of enrichment index for the particles PM10 and street dust shows that Pb and Sb have approximately the same value in both matrices, but in calculating enrichment index from the background value for soils in Olomouc, enrichment index for Pb is insignificant and it is comparable for Sb. The difference in the EF value for Pb is due to a higher concentration of Pb in soils (defined as geochemical background) – 43 mg/kg of dry matter and lower Pb content in street dust, ranging from 24 to 56 mg/kg with an average value of 35.13 ± 8.35 mg/kg. A similar difference in the EF value from the soil geochemical background was also found for

Tab. 2. Comparison of metal concentrations in the street dust (< 0.063 mm) from the territory of the city of Olomouc and towns in Scotland and Saudi Arabia (SA) v mg/kg

	Transport area[1]	Industrial area[1]	Unpolluted area[1]	Urban area (Scotland)[2]	Urban area (SA)[3]	Industrial area (SA)[3]
	Range (minimum – maximum) (median)					
Cu	84-312 (181)	82-211 (106)	44-70 (63)	52-212 (116)	71-158 (131)	61-176 (81)
Zn	384-825 (455)	146-1,032 (506)	202-626 (282)	89-720 (415)	267-477 (323)	100-420 (246)
Pb	27-91 (30)	24-55 (48)	29-45 (33)	4-338 (153)	41-115 (59)	67-179 (97)
Cr	89-349 (111)	45-412 (90)	44-83 (60)	7-135 (22)	56-99 (73)	16-78 (45)
Fe	22,191-40,584 (29,745)	19,419-32,966 (30,647)	20,575-28,298 (23,254)	-	7,796-11,802 (10,439)	4,914-14,038 (7,074)
Zr	422-635 (530)	433-811 (567)	436-739 (548)	-	-	-
	[1] Olomouc [2] Pal et al., 2011 [3] Khoder et al., 2012					

Tab. 3. Correlation between selected metals

	Cr	Fe	Cu	Zn	Zr	Pb
Cr	1.00					
Fe	0.74**	1.00				
Cu	0.81**	0.69**	1.00			
Zn	0.84**	0.73**	0.74**	1.00		
Zr	0.09	0.00	0.14	-0.02	1.00	
Pb	0.44*	0.54*	0.34	0.70*	0.27	1.00

As. Even in this case, for As, the geochemical background value was higher for soils (13.5 ± 1.04 mg/kg) than concentrations in street dust (9.01 ± 1.89 mg/kg). Since the soil sample was taken from the Ao horizon, the content of the risk metals may be higher because they are influenced by the long-term environmental load.

Enrichment index for Zn in PM_{10} ranges from 32 to 100, while in street dust only from 2.3 to 6.2. If we calculate enrichment index from the geochemical background value for soils in Olomouc, it shows the highest value of Cu enrichment index (29–7), with the EF value for the area affected by transport being the highest. These results correspond to the findings of Moreno et al. (2015), who dealt with the identification of elements released into the environment during bus, tram, and metro operation, and walking. The highest concentration of elements is released into the environment during bus operation, the lowest during tram operation. Copper is released into the air during bus operation at a concentration of up to 170 ng/m³, while only 24.98 ng/m³ in tram operation. The amount of Sb released is significant, when during bus operation it is 24.05 ng/m³ and only 0.66 ng/m³ during tram operation. During operation of buses, Zn is also released into the atmosphere, at a concentration of 100 – 150 ng/m³. Another significant element is Zn, only 53 ng/m³ is released during tram operation. The finding that during bus operation, up to 32.37 ng/m³ of Zr with expected geogenic origin is released, is significant as well. Pb is released during walking, and tram, bus, and metro operation at a comparable concentration of 6.61 – 7.45 ng/m³. Fe is released in the highest concentration, when 2.25 µg/m³ is released during operation of buses, and only 0.74 µg/m³ during tram operation.

Conclusions

The results of the study of enrichment factor (EF-crust) showed that for Pb and Sb, approximately the same values (100 for Pb) and >100 for Sb are reached in both matrices, PM10 and street dust. Enrichment factor values for PM10 are significantly higher in the winter season than in the summer season. The highest difference of up to 10 times was found for Mo, Zn (4x), As (3x), Cd, Ni, Pb, Sb (2x), Cu (1.2x). The different behaviour had Mn, its enrichment factor decreased, it was higher in summer than in winter (3x). Higher values of enrichment index in the winter season confirm the

Tab. 4. Enrichment factors (EF) calculated according to Equation 1 and enrichment factors calculated from the geochemical background of soil samples from the area with minimal load in the city of Olomouc

	Industrial (Fe_{crust})	Transport (Fe_{crust})	Unpolluted (Fe_{crust})	Industrial (Soil Olomouc)	Transport (Soil Olomouc)	Unpolluted (Soil Olomouc)
EF Cr	1.88	2.24	1.47	1.63	1.94	1.27
EF Cu	3.96	7.93	2.96	38.38	76.88	28.69
EF Zn	3.52	6.2	2.31	2.27	4	1.49
EF Pb	96.3	77.86	106.71	1.66	1.34	1.84
EF As	11.32	10.06	11.24	0.67	0.55	0.67
EF Mn	1.42	1.17	1.48	0.78	0.67	0.84
EF Ni	9.33	10.73	10.55	7.84	9.24	9.28
EF Sb	107	153	156	199	218	199

impact of combustion processes on the chemical composition of PM10. The study of the chemical composition of street dust in Olomouc shows that enrichment index calculated from the geochemical background of soils from Olomouc shows an increased value for Cu in the area affected by transport of up to 77, while in the area without pollution, the value is 29. Increased values of Cu in street dust may be affected by operation of buses in urban transport. Copper is, in addition to Fe, the most important element that is released during bus operation. The calculation of enrichment index (EF-soil) cannot be used for Pb and As, which have higher geochemical background values than those determined in street dust. The calculation of enrichment index using the reference average concentration of the element in the Earth´s crust (Clarke value) can be considered optimal.

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