A survey of uranium levels in urine and hair of people living in a coal mining area in Yili, Xinjiang, China

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ABSTRACT

Recent reports have drawn attention to the uranium contamination arising from coal mining activities in the Yili region of Xinjiang, China due to the mixed distribution of uranium and coal mines, and some of the coal mines being associated with a high uranium content. In this study, we have collected water samples, solid samples such as soil, mud, coal, and coal ash, and hair and urine samples from local populations in order to evaluate the uranium level in this environment and its implications for humans in this high uranium coal mining area. Our results showed that uranium concentrations were 8.71-10.91 μg L^{-1} in underground water, whereas lower levels of uranium occurred in river water. Among the solid samples, coal ash contained fairly high concentrations of uranium (33.1 μg g^{-1}) due to enrichment from coal burning. In addition, uranium levels in the other solid samples were around 2.8 μg g^{-1} (the Earth's average background value). Uranium concentrations in hair and urine samples were 22.2-634.5 ng g^{-1} (mean: 156.2 ng g^{-1}) and 8.44-761.6 ng L^{-1} (mean: 202.6 ng L^{-1}), respectively, which are significantly higher than reference values reported for unexposed subjects in other areas. Therefore, these results indicate that people living in this coal mining area have been subjected to uranium exposure for long periods of time.

Key words: Uranium contamination, Enrichment, unexposed subjects

1. Introduction

Uranium is a radioactive element ubiquitously existing in the Earth's crust with an average concentration of 2.8 μg g^{-1} (UNSCEAR, 2000). Natural uranium consists of a mixture of three radioactive isotopes: ^{238}U (99.2745 % by mass), ^{235}U (0.7200 %), and ^{234}U (0.0054%) (Chu et al., 1999). Due to its existence in soil, surface and ground water, air, plants, and animals, food and water are the primary uranium intake modes for non-exposed people. The average individual uranium intake is estimated to be 1-2 μg d^{-1} from food and 1.5 μg d^{-1} from drinking water (ATSDR, 2013). However, uranium intake is highly variable and depends on a number of factors such as dietary and drinking habits, local geology, fitness, and climate (Harley et al., 1999; Pietrzak-Flis et al., 2001). Drinking water has been identified as the major source of uranium intake in North America (ATSDR, 2013). In contrast, human occupational exposure to uranium occurs at work places such as mining, milling, and ore processing, which may contribute to an increase in uranium intake to potentially harmful levels (Bagatti et al., 2003). Most of the ingested uranium in the body is excreted in faeces and urine over a period of several days (Taylor and Taylor, 1997). The remaining uranium enters the blood stream and deposits mainly in the skeleton and the kidneys (Russell and Kathren, 2004). The kidney is most affected by the toxicity of uranium and very high concentrations of uranium lead to kidney failure. In addition, reproductive and respiratory systems are also affected by uranium exposure (ATSDR, 2013; Durakovic, 1999; ICRP, 1997). A review by Hindin et al., (2005) concluded there was an increased risk of birth defects in the children of persons

exposed to depleted uranium.

Much concern has been raised about the exposure of humans to elevated levels of radionuclides from nuclear-related activities such as uranium mining and processing, nuclear fuel production, nuclear waste storage (Blanchard et al., 1982; Gallop et al., 1998), and depleted uranium (Oeh et al., 2007). However, other uranium-bearing mines, such as coal mines, may also cause pollution in nearby environments. The negative effects of coal mining activities on the environment are mainly due to the disposal of large amounts of wastes that pollute groundwater, surface water, and soil by dusts, leachates, weathering, and self-ignition (Bian et al., 2009). It has been reported that mining, milling, and processing of uranium-bearing minerals has led to elevated levels of uranium not only in the workers but also in the inhabitants of mining and processing sites (Lipsztein et al., 2001; UNSCEAR, 2000). Surface and ground waters were contaminated by uranium and thorium from abandoned dump sites due to leaching (Ragnarsdottir and Charlet, 2000). Soil was found to have elevated level of uranium due to mining activities in Nigeria (Arogunjo et al., 2009). The coal mine sites in the Yili region of Xinjiang, China have been reported to have high levels of uranium in coal, and nearby soil has been contaminated to various degrees during coal mining, transportation, and burning (An et al., 2014). Negative effects to humans may occur by drinking of contaminated waters, consumption of crops grown on polluted land, or by inhalation of dust in these areas.

Urine and hair are two common bioindicators for uranium monitoring in humans. Urine monitoring is the preferred method to determine human exposure to soluble

uranium since the amount excreted per day via urine is related to the total level of uranium in the body (Hoellriegl et al., 2011). In order to make an assessment on an individual's possible exposure, knowledge of background uranium levels is indispensable. However, background levels vary from one person to another and also change greatly over different time scales (Mohagheghi et al., 2005). Levels of natural background uranium in the urine of unexposed subjects have been reported for different regions and population groups and may reflect variable intakes of uranium through food and beverages (Dang et al., 1992; Heitland and Koster, 2006). The average concentration of uranium in urine for major parts of the world is about 10 ng L^{-1} in non-exposed subjects ranging from a few ng L^{-1} to hundreds of ng L^{-1} (WHO, 2001). Measurement of uranium in hair is another useful method for assessing an individual's exposure to uranium. Hair samples have several advantages over urine samples. Firstly, hair is stable and does not need special storage or handling. Besides, hair can reflect the total body intake over an extended period much longer than urine and faeces (Sela et al., 2007). The uranium concentration in hair also greatly varies among unexposed persons. For example, hair samples of 67 Japanese males and 81 females showed ranges of 5-390 ng g^{-1} and 8.2-1280 ng g^{-1}, with means of 38^{-1} ng g and 51 ng g^{-1}, respectively (Imahori et al., 1979). The uranium content in different lengths of a single hair also changes. The concentration in a single hair declined from 212 to 18 ng g^{-1} due to a decrease in uranium concentration in drinking water (Sela et al., 2007).

The purpose of this study was to test and assess uranium levels in hair and urine

of the people living in a coal mining area in Yili, Xinjiang, China, who were likely to have been exposed to uranium above background levels. Uranium concentrations in water samples (groundwater, river water) and solid samples (soil, mud, coal and coal ash) were also monitored in an aid to interpret uranium exposure sources for the study subjects in the target area. Inductively coupled plasma mass spectrometry (ICP-MS) was used for the measurement of uranium due to its high sensitivity, high precision, and high sample throughput (Allain et al., 1991; Haldimann et al., 2001; Oeh et al., 2007).

2. Materials and methods

2.1. Sample collection

A total of 38 people located in and around the coal mining site volunteered for hair sampling. Among them, only 16 volunteered for urine sampling. Most of these 16 volunteers were male adults due to the sensitivity of the region regarding religious and other reasons. Some parameters of the volunteers are listed in Table 1.

The urine and hair samples were collected 4-5th April, 2015. The volunteers were provided with 50 ml polyethylene bottles for urine samples and given instructions on how to collect it without contamination. Urine samples were acidified on-site with 5 ml HCI (20%) per liter to prevent decomposition. The containers were returned to the laboratory after 12h and stored frozen at -20℃ until analysis. Hair samples were collected from the scalp using stainless steel scissors. Then hair samples were stored in 8×10 cm sealed plastic bags to avoid any contamination prior to the analysis in lab. In addition, two groundwater samples ($1^{\#}$ from a well and $2^{\#}$ from a worker's home), and two river water samples were collected using 1.0 liter plastic containers. Two mud samples from a dry river inside the coal mining area were collected using 15×20 cm plastic bags. Two soil samples ($1^{\#}$ from a forest, $2^{\#}$ from a garden orchard) were collected using clean woven bags. One coal ash sample was collected outside a farmer's house with a woven bag. Five coal samples ($1^{\#}$-$4^{\#}$ from different sites of this coal mining area, $5^{\#}$ from a farmer's house) were collected in 0.8 m×1.2 m×0.6 m plastic containers.

2.2. Sample preparation and analysis

Water samples were directly tested by ICP-MS (8800, Agilent, USA) after filtering with a 0.22 μm hydrophilic polyestersulfone membrane. The detection limit DL is 0.0055 μg L^{-1}. Solid samples including surface soil, mud, coal and coal ash were dried at room temperature, pulverized and sieved through a 100 mesh to homogenize them. 0.1g solid samples (except coal) were put into white { HYPERLINK "javascript:void(0);"} tubes for digestion. 4 ml concentrated nitric acid, 4 ml hydrofluoric acid, and 1 ml perchloric acid were added to the tubes which were heated at 180℃ until near dryness. Deionized water was added to the tubes several times. The solutions were transferred to and diluted in 100 ml volumetric flasks before measurement by ICP-MS after filtering with a 0.22 μm membrane. The detection limit (DL) was 0.018 μg L^{-1}. 1.0g of sieved coal samples were heated in a muffle furnace at 500℃ for 4 h in order to decarbonize. Coal samples were prepared for measurement after digestion using the above mentioned procedure. The hair samples were washed in the laboratory with ultra-pure water and acetone to remove only exogenous contaminants on the sample surface (Rodushkin and Axelsson, 2000). 0.05 g of each hair sample was digested in 3.0 ml concentrated nitric acid and 0.3 ml hydrogen peroxide (30%) in white { HYPERLINK "javascript:void(0);"} tubes. The tubes were heated at 100 ℃ until near dryness. Deionized water was added to the tubes several times. The solutions were transferred to and diluted in 10 ml volumetric tubes with deionized water prior to measurement. The detection limit (DL) was 0.032 ng g^{-1}. 1 ml of each urine sample was added to 0.5 ml of 16 M nitric acid, and then diluted to 10 ml with deionized water. Samples were centrifuged at 8000 rpm for 10

140 min before analysis of the supernatant. The detection limit (DL) was 8.91 ng L^{-1}. The
141 DL was calculated using Bessel formulae as following: DL=k •s_1 [DL=detection limit,
142 s_1=standard deviation (9-12 times testing of blank samples), k=2 or 3] (Bowman F,
143 2010). In our study, k and s_1 were 3 and 11 separately.
144 2.3. Statistical methods
145 A non-parametric statistical method (Mann-Whitney-Wilcox) was used to
146 determine whether there were differences between the subgroups (Gibbons, 1997).
147 T-test statistical method was used to determine whether there were significant
148 differences between the values of this study and mean (or average) values of other
149 studies. Spearman rank formula was used to determine the correlation coefficient and
150 its significance between uranium concentrations of urine and hair of the same study
151 subjects.

Results and discussion

2.4. Water and solid samples

Table 2 shows the uranium concentrations in water and solid samples in the coal mining area. The uranium concentrations in all water samples were above the DL of 0.0055 μg L^{-1}. Among them, ground water samples showed higher uranium concentrations, which are about 5-fold higher than the drinking water provisional guideline value (2.0 μg L^{-1}) issued by the WHO in 1998 (WHO, 1998), but lower than the increased provisional guideline values of 15μg L^{-1} (WHO, 2003) and 30μg L^{-1} (given by its chemical toxicity) (WHO, 2012). In contrast, the uranium concentrations in river water samples were measured to be lower than the drinking water guideline values. Among the solid samples, the coal ash displayed a rather high content of uranium (33.1 μg g^{-1} dry wt) due to the enrichment from coal burning. The uranium concentrations in soil and mud samples were significantly higher than the average global soil level of 1.8μg g^{-1} ($p=0.008$) (Eisenbud and Gesell, 1997), but roughly equivalent to the Earth's average background value (2.8 μg g^{-1}, $p=0.59$) (UNSCEAR, 2000). The uranium concentrations in 5 coal samples ranged from 1.85 to 3.77μg g^{-1} (dry wt), are not significantly different from the Earth's average ($p=0.51$).

2.5. Hair samples

Figure 1 and Figure 2 shows uranium concentration and its distribution in the hair of the study subjects in the coal mining area. Uranium levels in the 38 hair samples ranged from 22.2 to 634.5 ng g^{-1} with a mean value of 156.2 ng g^{-1} (Fig. 1). 42.1% of all the participants in this study were shown to have values of 50-100 ng g^{-1}, followed

by 18.4 % ranging between 100-150 ng g^{-1} (Fig. 2). In comparison with other studies where the uranium content in hair was determined, the range and mean concentrations for the whole study subjects were significantly higher than the range and mean values reported for unexposed (occupationally or environmentally) residents of south Israel (10.0-180 and 62 ng g^{-1}, respectively) (Gonnen et al. 2000) and north Sweden (6.0-436 and 57.0 ng g^{-1}, respectively) (Rodushkin and Axelsson 2000b). Our values are comparable to the mean value of Iraqi subjects (160 ng g^{-1}, p=0.86) where people were exposed to depleted uranium (Alaani et al., 2011) (Table 3). This implies that the people in the coal mining area of Yili, China have been subjected to uranium exposure for longer periods of times. Generally, drinking water is the main pathway for uranium intake where uranium levels in drinking water are above a few µg L^{-1}, while food and other beverages may be a major pathway for uranium intake when the concentration in drinking water is below 1 µg L^{-1} (Gonnen et al., 2000, Karpas et al., 2005). A correlation between uranium intake in water and concentrations in hair also was shown by Karpas et al., (2005). An incremental uranium intake of 1 µg d^{-1} was estimated to result in an increase of 37 ng g^{-1} in hair. The uranium concentrations in two ground water samples are 8.71 and 10.91µg L^{-1}, roughly converted to be 13.07 and 16.37µg d^{-1} (1.5 L/d • person). This indicates that the uranium in the ground water might be the main source of uranium intake in this coal mining area. However, uranium concentrations in soil and individual wells may vary greatly, therefore the limited number of water and soil samples in this study are not enough to infer the exact exposure pathways. Further investigation of the local water and soil sources,

196 consumption habits, and food would be necessary in order to indentify additional
197 exposure pathways.

198 In order to evaluate whether significant differences existed between sub-groups
199 such as males and females, age groups, and workers and residents, the non-parametric
200 statistical method (Mann-Whitney-Wilcoxon distribution) was used to determine the
201 differences between sub-groups. As shown in Table 4, only very slight differences,
202 which were certainly statistically non-significant, were found between males and
203 females ($p=0.814$). The dependence of uranium concentration on sex has been
204 reported in other studies. Rodushkin and Axelsson, (2000) observed women had
205 roughly twice the levels of uranium than men in northern Sweden, whereas Gonnen et
206 al., (2000) and Ting et al., (1999) found no significant difference between men and
207 women. Similarly, there were no statistically significant diffences found between
208 different age groups in our study ($p=0.294$, 0.655). These results show that there are
209 no sex or age dependent effects of uranium distribution in hair of the residents in this
210 coal mining area. Although workers have higher mean and median values than the
211 residents, the statistical difference is still non-significant due to the large variation of
212 the test data, implying that coal mining activities may not be the main cause for the
213 high uranium intake in people in this coal mining area.

214 2.6. Urine samples

215 A particular consideration for uranium study in urine would be whether to use
216 spot urine samples or to attempt a 24-h urine collection. Jones' (2007) study showed
217 that there was no systematic difference between the concentrations derived from 24h

or spot sample types. However, 24-h urine samples provided more precision while spot samples could well be sufficient for the purpose of distinguishing elevated concentrations. Due to the difficulty of collecting 24-h urine samples in our study area, we only collected 16 spot samples. Fig. 3 shows the frequency distribution of excreted uranium in urine of the people from the coal mining area. The uranium concentrations in all urine samples were above the DL of 8.91 ng L^{-1}. The uranium levels in the urine samples varied greatly from 8.44 to 761.6 ng L^{-1} with a mean value of 202.6 ng L^{-1}. Of all the volunteers in this study, 37.5% were found to have values lower than 100 ng L^{-1}, followed by 25.0% ranging between 100-200 ng L^{-1}. In comparison with the reference values of spot urine samples, the range of our study was much higher than the ranges reported for unexposed subjects. As shown in Table 5, Uranium concentration in the general public ranged from about 4 to 57 ng L^{-1} (WHO, 2001). Urine spot samples of unexposed subjects showed ranges of 3-40 ng L^{-1} (Dang et al., 1992) and not detectable to 38.1 ng L^{-1} (Jones et al., 2007). Our results are comparable to the mean value of a Jordan study (mean: 320 ng d^{-1}, roughly 228.6 ng L^{-1}, $p=0.62$), where soil is rich with phosphate rock and sand (Al-Jundi et al., 2004). This further confirms that the study subjects in this study area have been subjected to continuous uranium exposure from ground water. However, uranium concentrations in drinking water as well as water consumption rates may vary considerably over time; therefore a spot urine sample is not necessarily a good indicator of long-term uranium exposure. The uranium concentration in hair reflects the natural uranium contamination caused by continuous use of drinking water better than the uranium

concentration in a spot urine sample (Muikku et al., 2009). Therefore, the urine results of this study should be viewed as a supplementary method to the hair results in this survey.

Fig. 4 shows the change of uranium concentration in urine with age, and the correlation between uranium concentrations in both urine and hair of the same study subjects. In general, Uranium concentration in urine fluctuates greater than uranium concentration in hair and did not display a noticeable age-dependent effect with an increase of age (5-75) (Fig.3). This is because hair analysis reflects chronic exposure to toxic elements, while the concentration of trace elements in urine reflects the current or recent metabolic conditions, and fluctuates with daily or weekly changes in physiological and environmental conditions (Agnes and Sidney, 2014). Some earlier studies indicated that uranium excretion rates increased with age (Roth et al., 2001; Werner et al., 1997). The ICRP uranium model (ICRP, 1995a,b) also predicted such an increase under conditions of a continuous level of intake. In contrast, Hoellriegl et al., (2011) and Oeh et al., (2007) concluded that there was no dependency of uranium excretion with age, which is consistent with our conclusion.

Further, we used Spearman rank formula to analyze the correlation between uranium concentrations of urine and hair of the same study subjects. The correlation coefficient and significance were calculated to be 0.299 and 0.261, respectively. The statistical significance was set at $P<0.05$. $0.261>0.05$ means there is lack of significant correlation between urine and hair uranium concentrations. However it is noticeable from Fig. 4 that there are some parallel trends between the uranium

concentrations of hair and urine of the same subjects, suggesting that urine analysis might be useful as a monitor of exposure, being roughly correlated with hair values.

3. Conclusions

This study aimed to gain a better understanding of uranium contamination in a coal mining area in Yining, Xinjiang, China. The uranium concentration in ground water and river water was below the updated WHO drinking water provisional guideline value issued in 2012. The uranium content in soil, mud, and coal samples did not increased significantly compared to the earth's average background level. Coal ash showed fairly high level of uranium concentration due to the enrichment from coal burning. The uranium levels in the hair of 38 study subjects and in the urine of 16 study subjects were significantly higher than reported reference values for unexposed subjects, indicating that the residents in this coal mining area have been subjected to uranium exposure, most possibly from ground water. Uranium concentrations in the hair and urine of these volunteers did not display sex- or age-dependent effects. The low uranium content in coal samples and insignificant difference between workers and residents' hair samples may suggest that coal mining activities may not be the main cause for the high uranium levels in hair and urine of the people in this coal mining area. Due to the limited number of water and soil samples in this study, further investigation of the local water and soil sources, consumption habits, and food would be necessary in order to indentify additional exposure pathways.

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Table 1

Some parameters of the study volunteers for collection of urine and hair samples

Males	Females	Juveniles (5-17)	Adults (18-60)	Elders (61-75)
21	17	9	22	7

Mining workers (urine donors)	Residents (urine donors)	In all (urine donors)
11 (2)	27 (14)	38 (16)

Table 2

Uranium concentrations and standard deviation (SD) of water and soil samples

Samples	Ground water 1#	Groundwater 2#	River water 1#	River water 2#
Concentration (μg L^{-1})	8.71	10.91	1.28	1.23
SD	1.25	1.20	0.21	0.17
Samples	Soil 1#	Soil 2#	Sediment 1#	Sediment 2#
Concentration (μg g^{-1} dry wt)	3.13	2.69	2.46	2.57
SD	0.15	0.15	0.18	0.37
Samples	Coal 1#	Coal 2#	Coal 3#	Coal 4#
Concentration (μg g^{-1} dry wt)	2.70	2.0	3.77	2.44
SD	0.30	0.14	0.45	0.23
Samples	Coal 5#	Coal ash		
Concentration (μg g^{-1} dry wt)	1.85	33.10		
SD	0.30	1.92		

Table 3

Reference data on uranium concentrations in hair (ng g^{-1} dry wt) of occupationally unexposed subjects from different countries: number (N) and age of subjects, mean values and ranges of urinary ^{238}U excretion are presented.

Country	Number	age	Mean value (ng g^{-1})	Ranges (ng g^{-1})	references
Israel	99	3.5-84	62.0	10.0-180.0	Gonnen et al. (2000)
Sweden	114	1-76	57.0	6.0-436.0	Rodushkin and Axelsson (2000)
Slovenia	17	-	13.6	2.7-330.0	Byrne and Benedik (1991)
Brazil	22	17-61	15.4	2.1-498.0	Akamine et al. (2007)
Japan	67 M 81 F	5-60	38.0 51.0	5.0-390.0 8.2-1280	Imahori et al. (1979)
Iraq	25	-	160.0	20.0-400.0	Alaani et al. (2011)
Finland	852	18-66	216.0	0.5-1400	Muikku et al. (2009)
China	38	5-79	156.2	22.2-634.5	This study

Table 4

The statistical analysis, according to a non-parametric distribution, of the uranium content in hair samples of the subgroups characterized according to sex, age and occupation

Subjects	N	Uranium concentration (ng g^{-1})			Non-parametric test	Conclusion
		range	Mean	median	P	
Male	21	35.0-634.5	155.7	103.0	0.814	Not significant
Female	17	22.2-533.1	156.8	97.6		
Juveniles (5-17)	9	52.3-320.6	162.4	120.4	Juveniles/Adults: 0.249	Not significant
Adults (18-60)	22	22.5-533.1	148.6	143.7	Adults/Elders: 0.655	
Elders (61-75)	7	22.2-634.5	157.7	94.0		
Residents	27	22.2-634.5	137.9	91.8	0.260	Not significant
Workers	11	62.3-533.1	201.2	97.6		

Table 5

Reference data on uranium concentrations in urine (ng L^{-}) of occupationally unexposed subjects from different countries: number (N) and age of subjects, mean value, and ranges of urinary ^{238}U excretion are presented. LOQ is limit of quantification.

Country	Number	Age	Mean value (ng L^{-1})	Ranges (ng L^{-1})	Reference
Finland	951	18-66	16.0	10-3700	Muikku et al. (2009)
United Kingdom	25	20-59	-	LOQ-38.1	Jones et al. (2007)
India	20	-	12.8	2.9-40.0	Dang et al. (1992)
Germany	87	18-65	5.0	LOQ-20	Heitland and Koster (2006)
	72	2-17	4.0	LOQ-3	
Japan	168	-	5.6	0.8-35.6	Tolmachev et al. (2006)
Italy	38	20-50	10±7	1-44	Galletti et al. (2003)
Germany	>200	7-84	17.5	2-50	Roth et al. (2001)
USA	499	6-88	11.0	1.42-34.5	Ting et al. (1999)
Slovenia	10	-	12.8	3-49	Byrne and Benedik

					(1991)
Jordan	60	6-95	320 (ng d^{-1})	18-3420 (ng d^{-1})	Al-Jundi et al. (2004)
Finland	205	18-81	485	1-8450	Karpas et al. (2005)
China	16	5-75	202.6	8.44-761.6	This study

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Figures Legends

Fig. 1. Uranium concentrations in hair of the study subjects. a, male and female groups; b, age groups; c, workers and residents. Box plots have the following statistical values: 10th percentile (lower whiskers), 25th percentile (lower boundary of the box), median (solid line within the box), 75th (upper boundary of the box), 90th (upper whiskers), open squares (extreme values), and filled circles (mean values).

Fig. 2. Frequency distribution of uranium concentration in the hair of the study subjects.

Fig. 3. Frequency distribution of uranium concentration in the urine of the study subjects.

Fig. 4. The change in uranium concentrations in urine with age and the correlation of uranium concentrations in both urine and hair of the same study subjects. Data shown are means ± SD (n=3).

Fig. 1

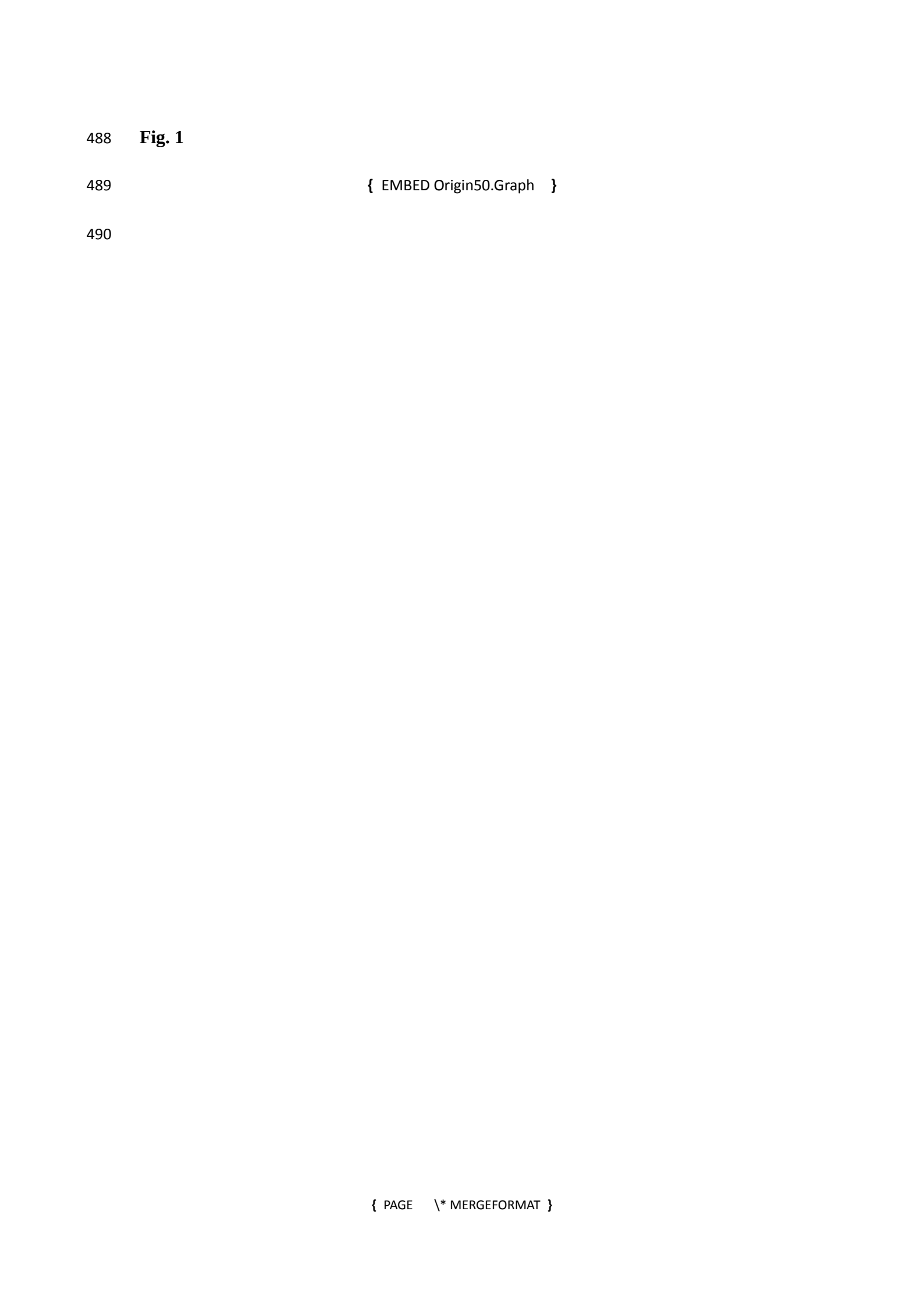

Fig. 2

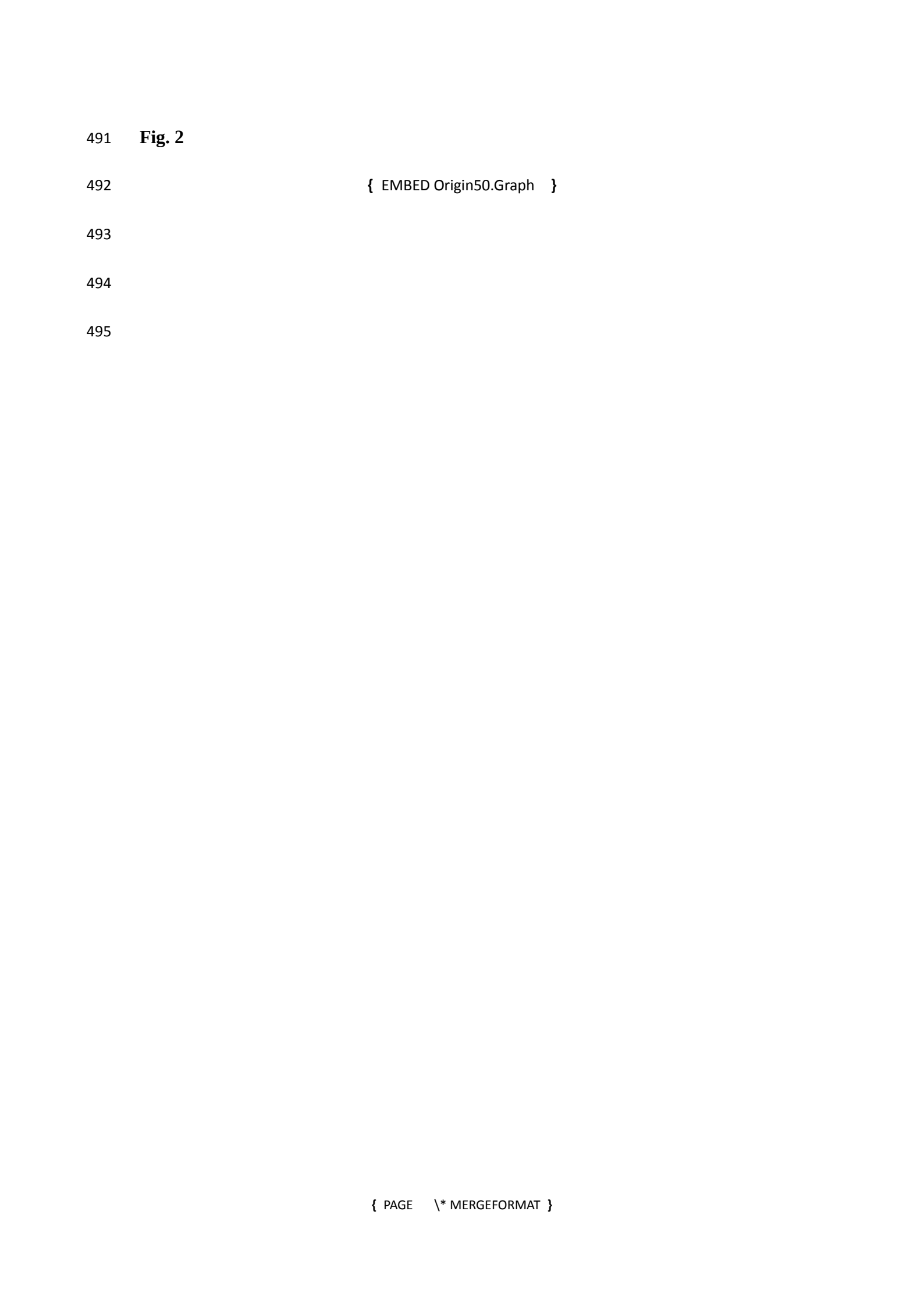

Fig. 3

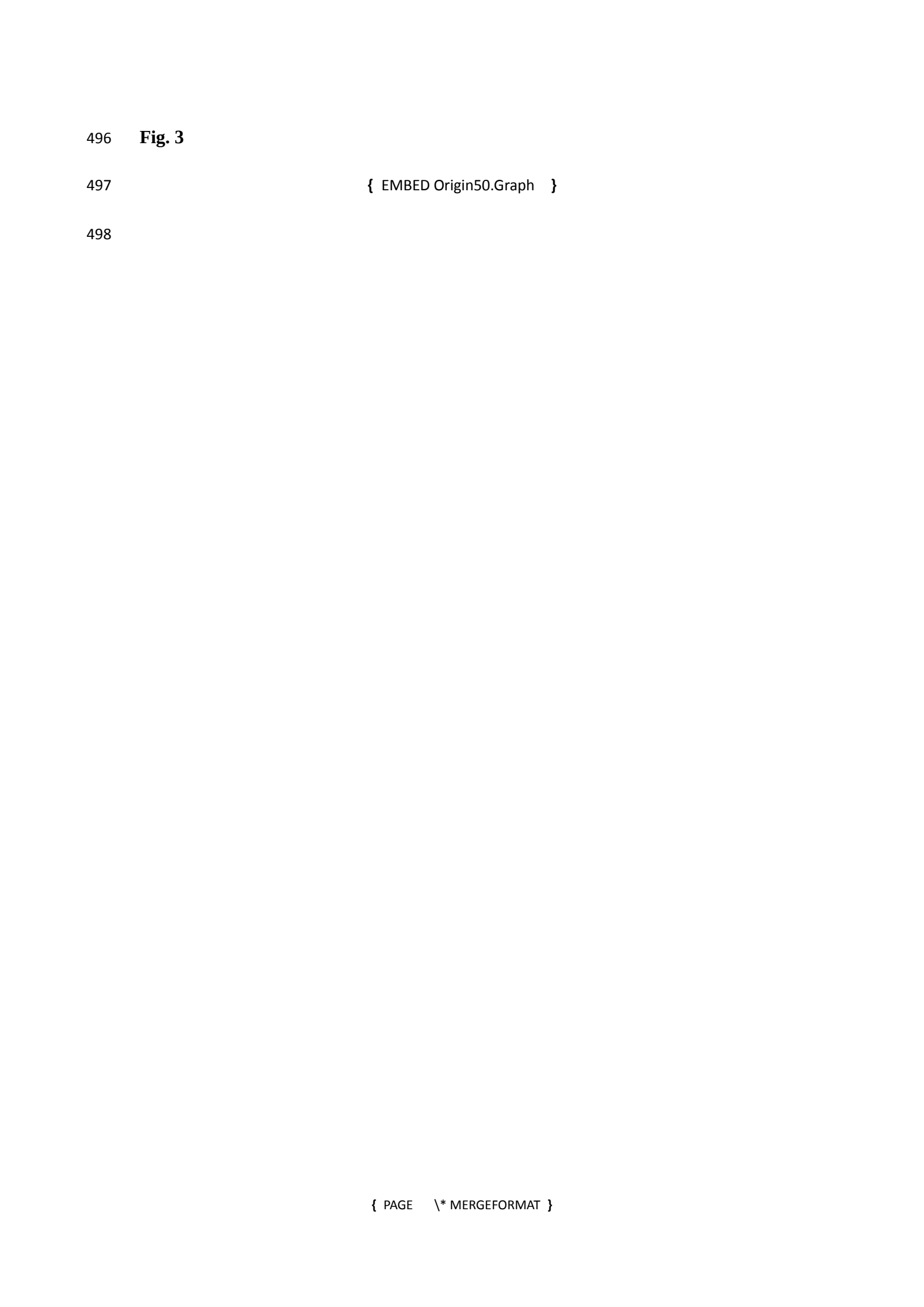

Fig. 4