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The Fitting Distribution Model of Chromium in Adhesive Building Materials Used in Saudi Arabia

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Abstract: **Many of building materials or geochemical data are not homogeneous nor normally distributed. Thus, most of scientists have used parametric tests, e.g., multiple regression, ANOVA, to robust their inhomogeneous or non-parametric distributed results produced through analytical work. Some researchers are not familiar with traditional transformations (e.g., square root, inverse, log) that may help to improve the normality tests (e.g Shewhart confidence limit). In the present work, more emphasis was on data transformations models (e.g Box-Cox) were carried out using Cr data in adhesive building materials analysis. Therefore, this paper deals with traditional normalizing transformations and their models where the purpose of is to find an appropriate probability distribution function to apply into the obtained data.**

Keywords: **Model of Chromium, Adhesive Building Materials.**

1. INTRODUCTION

Researches of heavy metals in the environment are an essential part to fully understand their behaviors and accumulates into our environments. Most of the work were greatly focused on heavy metals in water, sediments and soils owing to their high potential hazards (Luo et al,2012). In fact, heavy metals levels in water environments are well organized. Thus, significant work have been expended to evaluate their presence in ecosystem (Howard, Ibigoni Clinton and Olulu, Briggs Amonia et al, 2013) (Zhang, Weiguo et al, 2009)(Hong-gui,, et al, 2012) whereas almost no investigation has been considered the heavy metals in building materials particularly marbles. Therefore, e orts are needed to assess presence of heavy metals in building materials.

The high accumulation of Cr heavy metal in adhesive where building materials come from, can act as a source of Cr because sediments provide essential information of heavy metal accumulation and reffect the environmental impacts of them (Sedak et al, 2014) (Nordberg, Gunnar F el at. 2014).

Cr is belonged to VIB group in the periodic table, with an atomic number 24 and atomic weight of 51.99 and density of 7.1 g/ml. The two important forms are hexa and trivalent Cr where both forms can take place in water. The solubility of hexavalent is very soluble whereas the trivalent is relatively insoluble (Zhu, Yinian et, al, 2016).

Most of hexavalent Cr in the environment produces through human activities. The sources of hexavalent Cr mainly arise from mining, metal casting, pigment industries, and fossil fuels combustion. Whereas trivalent Cr sources are glass production, photographic synthesis and building material production (Chen, Tan, et, al, 2015).

Cr can enter our body through adsorption by respiratory system and gastrointestinal. When Cr is absorbed onto cells, it is rapidly excreted into the urine. The Cr can be determined in urine via routine analysis (Sedak, M, 2014).

The risk effects of Cr are linked with Cr hexavalent. It is well recognized that biological hazards of hexavalent of Cr might be reduced to Cr(III) that reacts with intracellular macromolecules. Exposure to Cr production can result in lung problems (Prasad, Ananda S, 2013).

The data transformations (DTs) are commonly used in quantitative chemical analysis as well as other sciences, particularly medical fields. DT can be used as tools to prove the normality of data distribution. Thus, DT plays important role to minimize the statistical errors and boost the data normality. There have been many applications of data transformations as such square roots, adding factors, reflecting, and wave trigonometric (Mueller, Ralph O and Hancock, Gregory R, 2008).

The important issue in modeling of chemical data is the identification of proper statistical distribution to fit with produced set of chemical data (Hosking, Jonathan RM, 1986).

The utilize of data transformation makes it more precise, enhances the improve of normality data of the reported results, and identifies the exact fitting distribution for the reported data (Osborne, Jason W, 2008).

In this work, it is wise step to highlight the importance of fitting distribution and its applications to this work. In fact, the fitting distribution of the data, if the data follow normal distribution, then they must substantially follow any type of fitting distribution or fitting model. Thus, the performance of fitting distribution can be used as check for quality control of the assumed normal distribution of the data sets.

It is suggested here to carry out fitting distribution to identify the proposed model of normality. When applying different fitting distribution models into the obtained chemical data, the come out model should follow normal, Box-Cox transformation, and loglogistic (2P) and (3P) distribution.

2. EXPERIMENTAL WORK

The marble materials were collected from different houseware stores in Riyadh. The proposed materials weathering-effect were removed at the spot and later transported to the lab. The materials were then crushed using crushing machine. A polyamide screen sieve (mesh size 1mm) was used and then the crushed materials were spread on the sieve by using plastic spatula and soft-shaking. Later, crushed materials were placed in an oven at 110-5 C for overnight in order to ensure no moisture is present in the crushed materials.

Approx. 5gm of each sample was milled to reduce the particle size, and to homogenize the powder sample. After drying, roughly 0.2 gm of the homogenized sample weight was very carefully measured out into vessel, and weight was recorded with an accuracy of 0.0001gm. A solution of HCl,HF, and HNO_3 was added to the vessel.

The performance was done by microwave assisted digestion using 0.2 g dried sample. After digestion H_3BO_3 was added for complexation of fluorides. Adding boric acid to the digested solution not only complexes the free fluoride ions in the solution, but also facilitates the dissolution of the precipitated fluorides. The solution in the bottle was the sample diluted to 50 ml in 3.5%

HNO_3.

Microwave conditions were: 60 bar in PTFE (polytetra uoroethylene) vessels; 35 minutes at 1400 W using a Multiwave 3000 (Anton Paar; Graz, Austria) microwave digestion system. All acids were Merck Suprapur. Determination of heavy metals was carried out by ICP-MS (Inductively Coupled Plasma-Mass

Spectrometer): NexION 300D (Perkin Elmer, USA) at the chemistry department, king Saud University. The selected parameters of operational system used in this analysis are listed in Table.1.

Table 1: Instrumentation operating System for ICP-MS

RF power	1600 W
Nebulizer gas ow	0.92 L/min
Lens Voltage	9.25 V
Analog Stage Voltage	-1762.5 V

Pulse Stage Voltage	1050 V
Number of Replicates	3
Reading / Replicates	20
Scan Mode	Peak Hopping
Dwell Time	40 ms
Integration	1200ms

ICP standard solution was created for the analysis with eight varying levels for each element. High purity certified elemental standard (6 CertiPUP, Merck Plasma Standards) was used in this analysis. To ensure that acids used in this work did not affect the ICP-MS reported data, blank was carried out and acids were used in the standard to the same levels as the sample digestion. It was found out that the acids did not have any affect. Every 8th sample run by ICP-MS was standard, to monitor the quality of instrument. Moreover, an internal standard was used to ensure that the instrument did not go out of calibration.

For quality assurance, certified reference materials were used. The used reference materials were purchased from USGS and they were 69 b bauxite, 1646 a Estaurine Sediment, 1 d Limestone, GBW 07106 Rock, and GBW 07108 Rock. The reported results of the certified reference materials by ICP-MS lab were in the target relative standard deviation of less than 10%.

The target precision of the certified materials had to be above 90% to produce very healthy and comparable results. Fortunately, the obtained result precisions were above 90% by ICP-MS lab.

Table 2: Reported results of reference materials using ICP-MS

	V	Cr	Mn	Fe 57	Co 59	Ni 60	Cu 63	Zn 66	Ga 69	As 75	Sr 88	Mo 98	Cd 111	Te 130	Ba 138	Tl 205	Pb 208	Bi 209	U 238
lab results	ppm	ppm	ppm	%	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
69 b bauxite	147.6	75.7	685.8	6.94	1.3	12.9	11.4	27	96	27.7	9.2	26.5	2.1	0.3	76	0.5	10.3	7.1	9.3
1646 a Estaurine Sediment	49	36.2	219.4	1.73	4.9	21.4	10.8	46.6	4.5	5.2	71	1.8	0.2	0.3	192	0.2	8.8	0.3	1.3
1 d Limestone	9.3	9.1	227.3	0.17	24.4	4.7	3.4	15.2	0.9	10.6	236.7	0.9	0.6	0.1	22.3	0	0.4	0.1	0.9
GBW 07106 Rock	36	18.3	157.7	2.32	6.7	15.3	17.4	15.7	0.49	6.2	63.7	4.7	27	200	129.3	0.3	6.1	0.2	1.9
GBW 07108 Rock	35.3	24.9	414.4	1.68	8.8	18.5	22.9	47.8	0.23	7.2	876.2	6.3	29	57	110	0.2	10.4	3.3	1.4
edCerti material																			
	V	**Cr**	**Mn**	**Fe 57**	**Co 59**	**Ni 60**	**Cu 63**	**Zn 66**	**Ga 69**	**As 75**	**Sr 88**	**Mo 98**	**Cd 111**	**Te 130**	**Ba 138**	**Tl 205**	**Pb 208**	**Bi 209**	**U 238**
	ppm	ppm	ppm	%	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
69 b bauxite	156	75	850	7.14	1.1			28						0.214	80	.....	11.8		
1646 a Estaurine Sediment	45	40.9	234.5	2.01	5	23	10	48.9	5	6.23	68	1.8	0.148		210	<0.5	11.7		2
1 d Limestone	10		233	0.22		4		18	1		260				30				1
GBW 07106 Rock				2.50	6.4	16.6	19	20	0.76	5.9	58	5.3	29	214	143		7.6		2.1
GBW 07108 Rock		32		1.70	9	17.8	23.4	52	0.38	6.6	913	7.1	32	62	120			3.16	

3. ANALYTICAL RESULTS

The studied adhesive materials chemical data for Cr are listed in Table.3. The statistical calculations are also reported in Table.3. Cr levels associated with adhesive are discussed herein with more emphasized on statistical models.

Cr levels in adhesive materials were slightly elevated with an average of 13ppm compared to the upper earth crust of 10ppm. The highest reported value was 66 ppm whereas the lowest reported value was 0.5 ppm. The results distribution of 25% was 1.2 ppm, 50% was 2.5 ppm, and 75% was 22.4 ppm. The normality test using A-D test proved most of the results followed relatively the normal distribution as shown in Fig.1 as the z-score located between 2 to -2. Most of the obtained results for Cr in adhesive material passed the Shewhart confidence limit as shown in Fig.2. except one sample which may be regarded as outlier. The data were analysed using 95% confidence interval (5% significant level).

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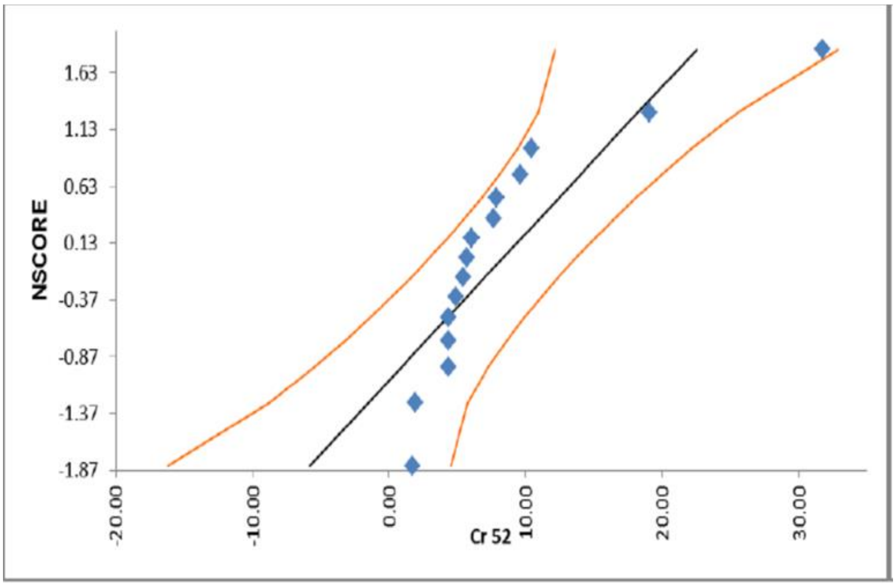

Figure 1: Normal distribution of Cr level in adhesive material

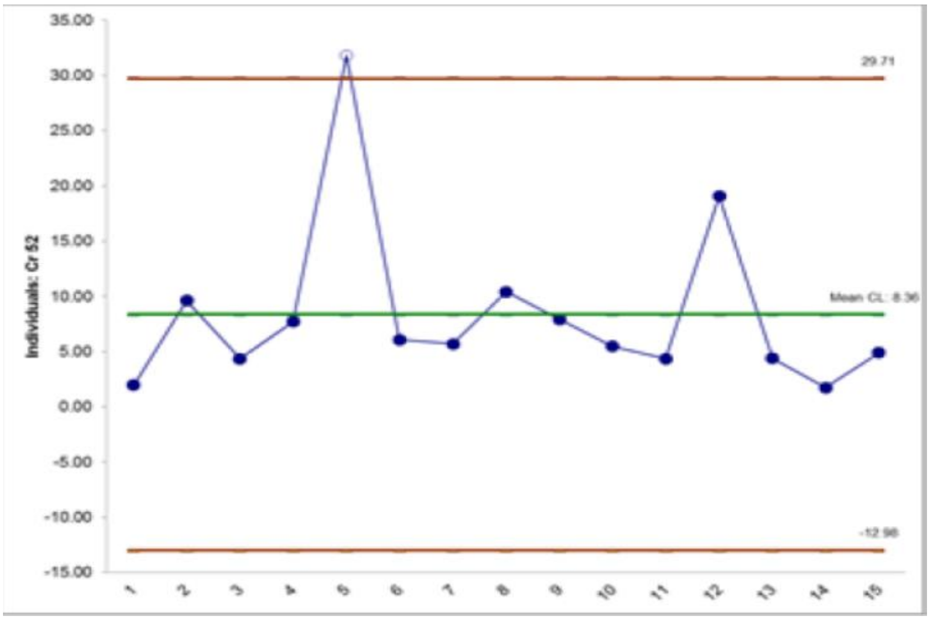

Figure 2: Shewhart confidence limit for Cr level in adhesive material

4. FITTING MODEL DISTRIBUTION DISCUSSION

The used models shown in Fig.3 up to 8, illustrated very good agreement between each model and normality test. The used log scale (2P) in Fig. 3 looks fit well of the used chromium data. Fig.3 shows the data were very normally distributed. The obtained data transformation of : Loglogistic (3P) in Fig.4 demonstrated that chromium results were in good level of confidence with log (3P) as well as normality test using A-D test. The Box-Cox Transformation showed also good normal distribution of reported data as illustrated in Fig.4. The calculated of Log-Likelihood of Box-Cox was similar with other models.

Inversely, the Smallest Extreme values did not match well with other models. By considering Fig.6, the Smallest Extreme model was not suitable model of the used data. Also, the half-normal distribution did not match well as merit of data transformation

Although the adhesive materials were not homogenous, they showed clearly normal distribution Therefore, most of the applied models worked well with the used data.

DISTRIBUTION: LOGLOGISTIC (2P):

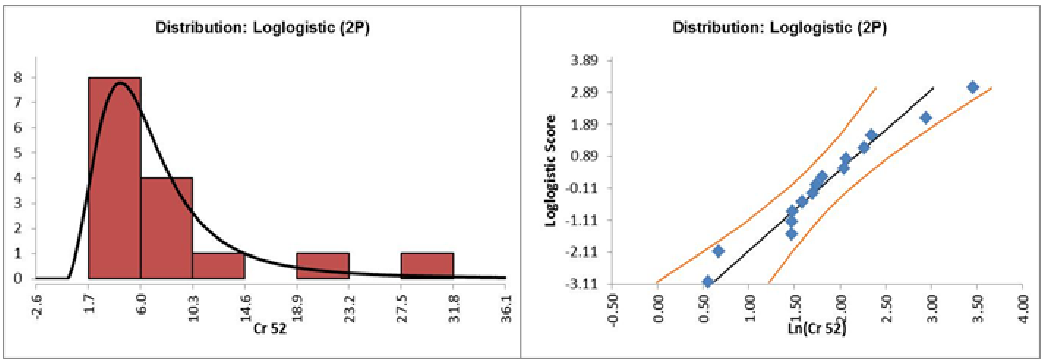

Figure 3: Distribution: Loglogistic (2P)

Distribution: Loglogistic (3P):

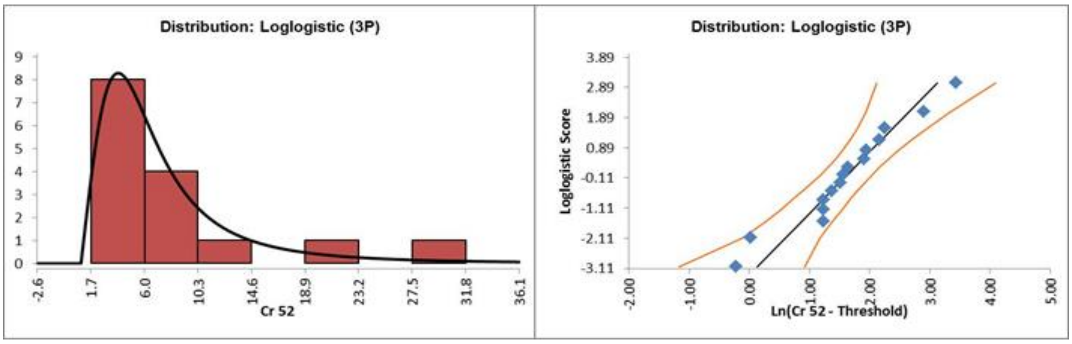

Figure 4: Distribution: Loglogistic (3P)

Distribution: Box-Cox Transformation:

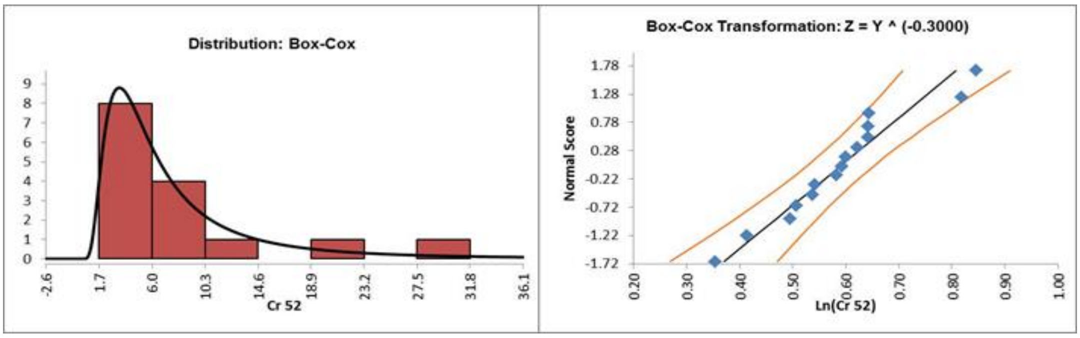

Figure 5: Distribution:Box-Cox Transformation

Distribution: Smallest Extreme Value (2P):

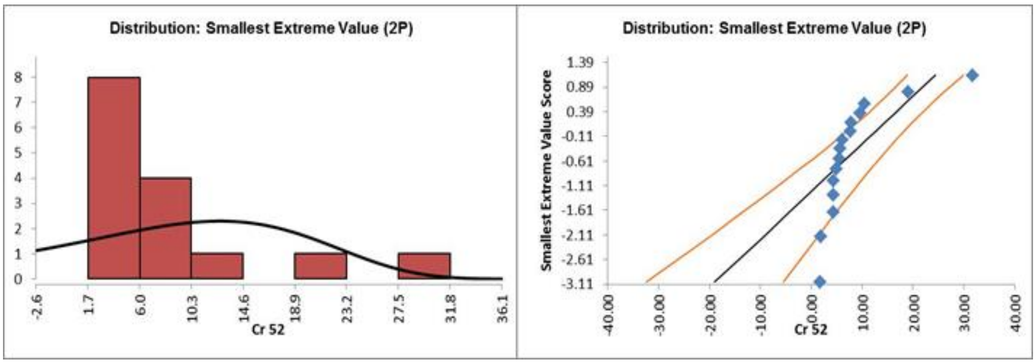

Figure 7: Distribution: Normal

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Distribution: Half-Normal:

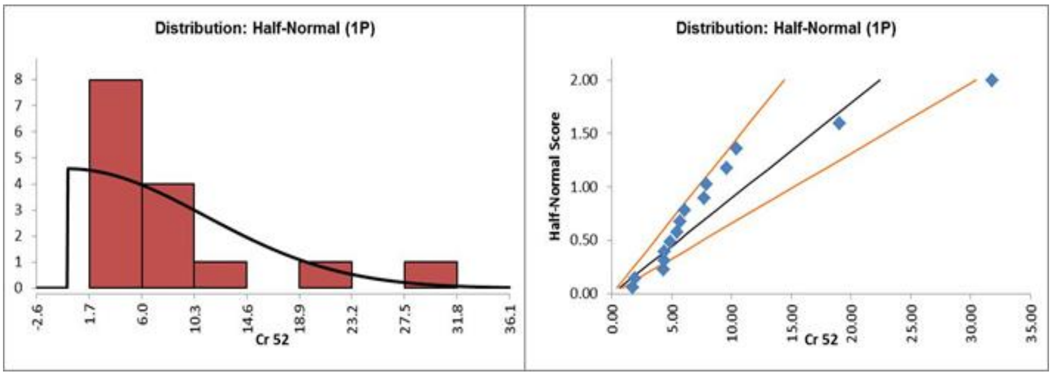

Figure 8: Distribution: Half Normal

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Table 3: Cr levels in adhesive materials

Sample Id		Cr 52
Adhesive S78		1.957113
Adhesive S79		9.611449
Adhesive S80		4.351221
Adhesive S81		7.702654
Adhesive S82		31.78752
Adhesive S83		6.073011
Adhesive S84		5.698459
Adhesive S85		10.42565
Adhesive S86		7.91672
Adhesive S87		5.481078
Adhesive S88		4.37149
Adhesive S89		19.05974
Adhesive S90		4.384569
Adhesive S91		1.745554
Adhesive S92		4.88921
Count		15
Mean		8.364
Stdev		7.723
Range		30.042
Minimum		1.746
25th Percentile (Q1)		4.371
50th Percentile (Median)		5.698
75th Percentile (Q3)		9.611
Maximum		31.788
95.0% CI Mean		4.087 to 12.64
95.0% CI Sigma		5.654 to 12.18
Anderson-Darling Normality Test		1.640
P-Value (A-D Test)		0.0002
Skewness		2.370
P-Value (Skewness)		0.0005
Kurtosis		6.080
P-Value (Kurtosis)		0.0031

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