



Analytical Study of Affecting Gasoline Quality Properties on Environment and Its Conformity with Specifications

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ABSTRACT

Gasoline is an important fuel in human life, but it is also responsible for deteriorating Ambient Air Quality (AAQ) through fugitive and exhaust emissions. In this study, chemical properties (Sulfur and Lead content) of gasoline production at Al-Daura refinery were verified based on a statistical quality control tool. Gasoline samples were examined in January/2011 and compared with samples examined in January/2019. In this study, it was concluded that the average production process and process variability for Sulfur content and added Lead value are stable. In January/2011 Sulfur content conforms to Iraqi and European standard specifications, but Lead value doesn't conform to Iraqi standards, where internationally the use of lead to enhance engine performance has been banned since the 1970s of a century ago because it's large dangerous on the environment. In January/2019 Sulfur content conforms to Iraqi standards but doesn't conform to European standards specifications. The addition of lead to gasoline is stopped in 2016; imported gasoline was used by blend it with produced gasoline at al Daura refinery to raise its quality. Gasoline quality properties must be improved and tighten up according to international standards to save our environment.

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1. INTRODUCTION

The quality of motor gasoline is specified by measuring its various Physico-chemical properties measured by about 20 various properties. Gasoline consists of organic compounds including Carbon and Hydrogen (HC). Products derived from crude oil have large commercial worth [1]. These products are mixtures, sometimes simple, but more oftentimes very complex. Gasoline is one of these

derived products. It is classified into two categories, premium, and regular based on the octane number. Gasoline with high octane number has many comprehensive benefits of reducing exhaust emissions and engine noise, improved cold starting and engine strength. There is a general claim about the poor engine performance and increased exhaust emission by a large number of consumers from time to time [1]. In the context of increased fuel requirements, including requirements related to their environmental impact, the issue of fuel quality is gaining more importance [2]. Urban air pollution worldwide is categorized to be responsible for 865,000 premature deaths every year and about 60% of these deaths occur in Asia [3]. Continues trends to make gasoline more environmentally and friendly to humans. Many researchers [4,5,6] directed to improving performance and environmental properties for the gasoline by blending it with another material or by using processing techniques in the refinery. So, the research problem is that most petroleum refineries facing the challenges of producing gasoline that has all of the qualifying properties as well as responding to increased environmental regulations and health restrictions on vehicle emissions. In this study, the sulfur and lead content of gasoline produced in the Daura refinery are manipulated to assess the effect of fuel composition on air pollutant emissions and conformity to Iraqi and European standards based on quality control tools.

2.GASOLINE FUEL QUALITY PROPERTIES

Gasoline is a complex mixture of 200 to 300 hydrocarbons, and its properties will vary in the relay on numerous refining and blending processes used to make it. This hydrocarbon is from 5 to 12 carbon. Figure 1 shows the regular carbon chain lengths [7, 8].

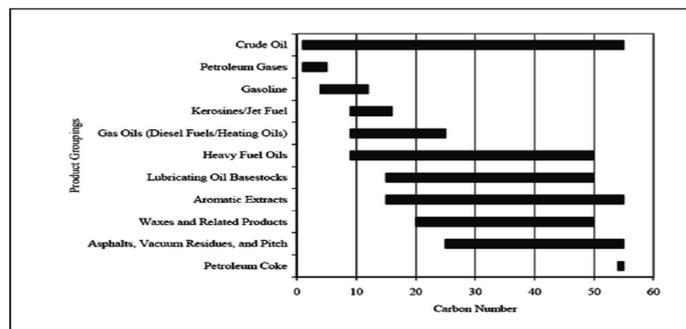


Figure 1: Typical carbon chain lengths [7]

The quality of gasoline is opposed to meeting specific operating and retention requirements, as gasoline must result in clean combustion without being knocked out. The quality specifications of gasoline engines have been greatly increased to reduce the environmental effect of motor vehicles [9, 10]. Figure 2 shows the gasoline requirements in different countries.

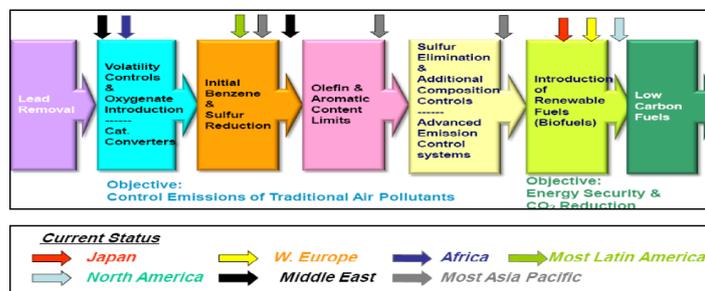


Figure 2: Worldwide gasoline quality development [11]

The main air pollutants in the exhaust gases are insufficient combustion oxides of the hydrocarbon containing carbon dioxide, nitrogen oxide, hydrogen, and particles when gasoline burns in the engine. Changes in the composition of gasoline can reduce vehicle emissions because adjusting the specific gasoline allows engines to perform in the optimum range. Understanding the impact of

fuel on modern gasoline vehicles is important for environmental agencies that develop regulations for modified gasoline [3, 12]. Also, the increase in the number of gasoline vehicles leads to an increase in traffic congestion and problems related to air pollution [13]. Table I summarizes the effects of gasoline quality on emissions from light gasoline vehicles.

TABLE I: Impact of gasoline composition on emissions from light-duty vehicles [12, 14]

Gasoline	No Catalyst	Comment	EU Euro emission standards, g/km				
			EuroI	EuroII	EuroIII	EuroIV	EuroV
lead↑	Pb ↑	CO, HC, NO _x All increases dramatically as catalyst destroyed	CO=2.72	CO=2.20	CO=2.30	CO=1	CO=1
Sulfur↑ (50-450) ppm	SO ₂ ↑	CO, HC, NO _x all increase ~15-20% SO ₂ and SO ₃ increase	HC+NO X= 0.97	HC+NO X =0.50	HC=0.20	HC=0.10	HC=0.1
					NOX=0.15	NOX=0.08	NOX=0.06

Vehicle emission standards from the European Union (EU) are active and require further reduction in carbon exhaust emissions from carbon monoxide (53%), hydrocarbons (67%), and nitrogen oxides (68%) relative to the Euro I standard which became in 1992. This requires adapters (catalytic) on all cars too. The Euro IV standard that came into force in January 2006 requires an additional reduction in exhaust emissions (relative to Euro III) in carbon monoxide (43%), hydrocarbons (33%), and nitrogen oxides (50%). The European Union has also established fuel generation standards that require consecutive cleaner fuels. Consequently, the European Union is looking to reduce transport concerning air pollution, not only by ordering cleaner and more efficient engines and catalytic converters but also by ordering cleaner fuels [15]. The quality of many petroleum products depends on the amount of sulfur. Sulfur is naturally present in crude oil. If sulfur is not removing during the refining process, this will pollute the vehicle's fuel. Since then, many catalytic processes are sensitive to sulfur contaminant [16, 17]. The sulfur content of the gasoline mixture increases emissions of nitrogen oxides, carbon monoxide, and sulfur oxide. The European Commission will require members of the European Union to produce 10 parts per million (ppm), or semi-sulfur gasoline and diesel [16, 18]. Fuel additives are very important since many of these additives can be added to the fuel to enhance eligibility and performance. Using Lead additives is an inexpensive way to refrain from this required property, but the lead is very toxic; its use in gasoline is banned in an increasing number of countries. Removing the lead component from gasoline reduces emissions of HC vehicles. Thus, a lead-free market worldwide is essential, not only for emissions control compatibility but also because of the known adverse health impacts of lead [12, 16, 18]. This is means that leaded gasoline is considered an environmentally unsafe product. [19]. Table II clarifies the changing of gasoline specifications with time.

TABLE II: Specification of gasoline quality [20]

Standards	BStd-II	Euro III	Euro IV	WWFC	Iraqi Std.
Year of Implementation	2000-2001	2005	2010		2000
Sulfur, ppm	500	150	50	<10	500
Research Octane Number (RON), (Min)	88	91	91		85
Motor Octane Number (MON), (Min)	-	81	81		-
Benzene, (Max), Vol %	5/3	1	1		-
Aromatics (Max), Vol %	-	42	35		-
Olefin, (Max), Vol %	-	21	21	10	-
Reid Vapour Pressure (RVP), KPa	35-60	60 (Max)	60 (Max)		44-82.5
Lead g / liter	-	-	-	-	0.1

3. GASOLINE QUALITY PROPERTIES TEST

To measure the quality of motor gasoline, (31) samples were collected randomly from the Research and Quality Control Department at al-Daura Refinery, one of Midland Refineries Company (MRC). These samples were examined in January/2011 and compared with samples examined in January/2019. Al-Basrah crude oil is the main source of production at this refinery. At al- Daura refinery, gasoline blends produced in one grade that is regular RON 82 from various components are:

- 1) Light Straight Run Naphtha (LSRN) RON 63,
- 2) Reformate RON 88.5 from reforming a mixture of 30% Light Straight Run Naphtha (LSRN), and 70% Heavy Straight Run Naphtha (HSRN)],
- 3) Power formate RON 87 is the last component of the reforming process.

The final quality of the terminated products always checked by laboratory tests before market distribution. Sulfur content and lead additive investigated. The sample size is determined according to various component tanks (three tanks) and their value (1 litter). By using Minitab 18 software Statistical quality control tools, “ \bar{X} - R” charts were utilized since these charts help detect the deviation from the process mean and process variability. “Equation (1) and (2) are the central values for the \bar{X} - R charts” [21,22, 23]:

$$\bar{\bar{X}} = \frac{\sum_{j=1}^m \bar{X}_j}{m} \quad (1)$$

$$\bar{R} = \frac{\sum_{i=1}^m R_j}{m} \quad (2)$$

Where $\bar{\bar{X}}$ = average of subgroup means.

\bar{X}_j = mean of the jth subgroup.

m = no. of subgroup.

\bar{R} = average of subgroup ranges.

R_j = range of jth subgroup.

Trial control limits for the charts are established at ± 3 standard deviations from the central value as shown by the formulas:

$$UCL_{\bar{X}} = \bar{\bar{X}} + A_2 \bar{R} \quad (3)$$

$$LCL_{\bar{X}} = \bar{\bar{X}} - A_2 \bar{R} \quad (4)$$

$$UCL_R = D_4 \bar{R} \quad (5)$$

$$LCL_R = D_3 \bar{R} \quad (6)$$

Where UCL = Upper Control Limit. LCL = Lower Control Limit. A2, D3, and D4= are factors that vary with subgroup size.

I. Result of Sulfur Content Test

Tables III and IV show results of the laboratory analysis of the samples examined in the examination laboratories at the Daura refinery in terms of the average, range, and standard deviation of the sulfur content of the gasoline for the two periods (2011 and 2019). These values are used to obtain the quality control charts resulting in the next two Figures 3 and 4 respectively.

TABLE III: Analytical results of sulfur content (ppm) in January/2011

No. of tests	Sample 1 (Tank A)	Sample 2 (Tank B)	Sample 3 (Tank C)	Range	Mean	St.Dev.
1	2	3	5	2-5	3.33333	1.527525
2	3	5	4	3-5	4.00000	1
3	4	2	3	2-4	3.00000	1
4	5	2	2	2-5	3.00000	1.732051
5	3	3	5	3-5	3.66667	1.154701
6	4	2	3	2-4	3.00000	1
7	3	5	4	3-5	4.00000	1
8	5	2	4	2-5	3.66667	1.527525
9	2	3	5	2-5	3.33333	1.527525
10	3	4	2	2-4	3.00000	1
11	5	3	2	2-5	3.33333	1.527525
12	3	4	4	3-4	3.66667	0.57735
13	2	3	5	2-5	3.33333	1.527525
14	6	3	4	3-6	4.33333	1.527525
15	3	2	5	2-5	3.33333	1.527525
16	5	3	4	3-5	4.00000	1
17	2	3	6	2-6	3.66667	2.081666
18	4	3	4	3-4	3.66667	0.57735
19	4	5	2	2-5	3.66667	1.527525
20	5	3	4	3-5	4.00000	1
21	2	3	4	2-4	3.00000	1
22	4	3	5	3-5	4.00000	1
23	5	3	2	2-5	3.33333	1.527525
24	4	4	5	4-5	4.33333	0.57735
25	6	4	3	3-6	4.33333	1.527525
26	3	3	4	3-4	3.33333	0.57735
27	5	3	2	2-5	3.33333	1.527525
28	4	5	3	3-5	4.00000	1
29	2	4	2	2-4	2.66667	1.154701
30	3	3	5	3-5	3.66667	1.154701
31	2	6	5	2-6	4.33333	2.081666

TABLE IV: Analytical results of sulfur content (ppm) in January/2019

No. of tests	Sample 1 (Tank A)	Sample 2 (Tank B)	Sample 3 (Tank C)	Range	Mean	St.Dev.
1	111	124	150	111-150	128.3333	19.85783
2	102	177	108	102-177	129	41.67733
3	167	183	120	120-183	156.6667	32.7465
4	115	110	165	110-165	130	30.41381
5	163	200	123	123-200	162	38.50974
6	172	155	183	155-183	170	14.10674
7	126	107	119	107-126	117.3333	9.609024
8	171	158	144	144-171	157.6667	13.50309
9	176	119	132	119-176	142.3333	29.87195
10	183	126	145	126-183	151.3333	29.02298
11	116	128	114	114-128	119.3333	7.571878
12	172	156	165	156-172	164.3333	8.020806
13	136	145	173	136-173	151.3333	19.29594
14	111	123	153	111-153	129	21.63331
15	138	136	164	136-164	146	15.6205
16	123	146	132	123-146	133.6667	11.59023
17	116	126	135	116-135	125.6667	9.504385
18	162	153	134	134-162	149.6667	14.29452
19	136	123	144	123-144	134.3333	10.59874
20	173	163	133	133-173	156.3333	20.81666

21	126	118	127	118-127	123.6667	4.932883
22	165	158	176	158-176	166.3333	9.073772
23	185	154	147	147-185	162	20.22375
24	148	177	136	136-177	153.6667	21.07922
25	128	136	173	128-173	145.6667	24.00694
26	152	137	149	137-152	146	7.937254
27	182	163	145	145-182	163.3333	18.50225
28	145	127	146	127-146	139.3333	10.69268
29	138	156	144	138-156	146	9.165151
30	135	128	123	123-135	128.6667	6.027714
31	147	128	146	128-147	140.3333	10.69268

While Figure 3 and 4 show that observed samples of Sulfur content for process average and process variability are steady during the tested period of January 2011 and January 2019 where only common causes are present, and no samples out of control limit. Sulfur content in gasoline blends at the period of January/ 2011 was better and conform to Iraqi (500 ppm) and European (50 ppm) standard specifications (previously mentioned in Table II). These values compared with tested periods at January 2019, where was conform to Iraqi standard but doesn't conform to European standard specifications (better to down quality values). This is due to a change in the raw material of crude oil wherein the previous period the refinery was supplied with two types of crude oil (Basrah and Kirkuk crude oil). Currently, Basrah crude oil is the only source for the Daura refinery, since each type of crude oil has different sulfur content due to its nature. An international effort is direct towards reducing this value of sulfur to control or decrease its effects especially on the environment. Sulfur in vehicle fuels leads to unwanted vehicle emissions of sulfur compounds and interferes with vehicle emission control systems that are directed at regulated emissions such as volatile organic compounds, nitrogen oxides, and particulates. Consequently, refineries must have the capability to remove sulfur from crude oil and refinery streams to the extent needed to relieve these unwanted effects. The higher the sulfur content of the crude, the greater the desirable degree of sulfur control, and the higher the related cost.

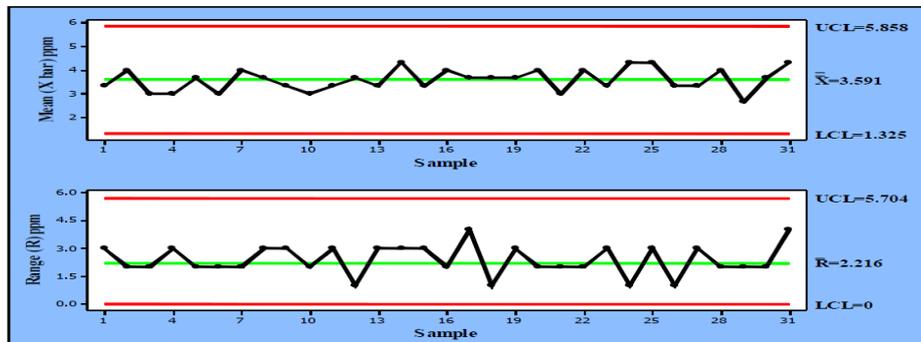


Figure 3: \bar{X} and R charts for gasoline Sulfur content in January/2011

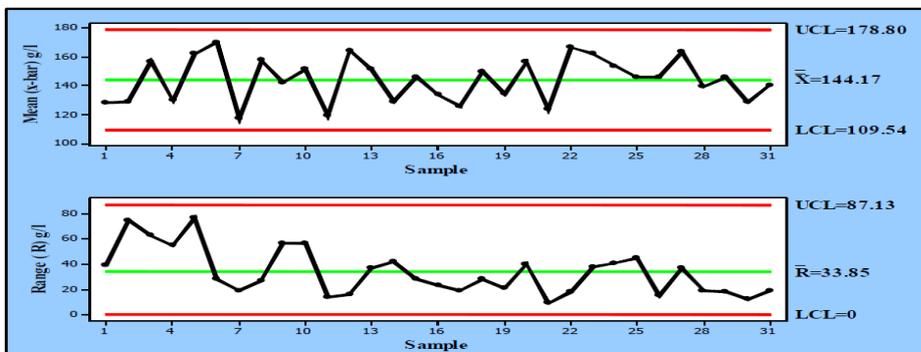


Figure 4: \bar{X} and r charts for gasoline sulfur content in January /2019

The difference between the sulfur content values can be distinguished by the representation of the samples taken for the years 2011 and 2019 by the Radar diagram. We note that there is a large and

clear difference between the samples of the two years as shown in Figure 5. Where the samples close to the center in green color represent samples of the year 2019 and the samples away from the center in blue color represent samples of the year 2011.

Previously, in 2011, the refinery relied on Basrah and Kirkuk crude oil as a raw material to produce gasoline after mixing them. Which attributes to the low sulfur values. Whereas in 2019 Basrah crude oil became the only source for the Daura refinery to produce gasoline, which led to an increasing in the values of its sulfur content. It is also known that Basrah crude oil is distinguished by its higher sulfur content compared to Kirkuk crude oil.

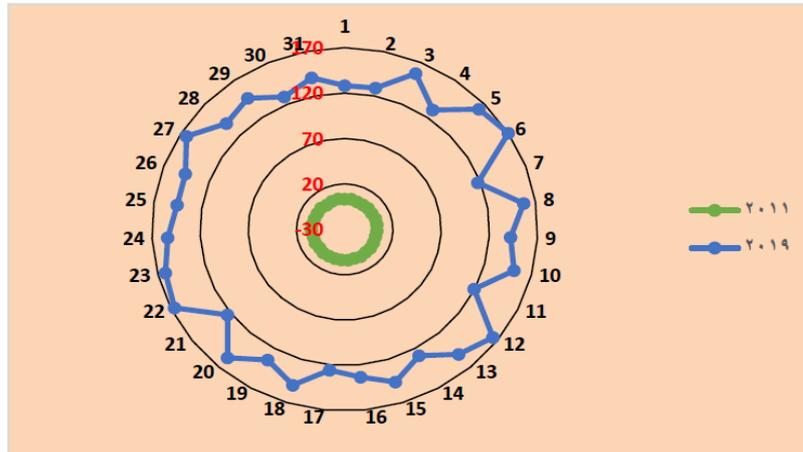


Figure 5: Radar chart for gasoline Sulfur content at years 2011 and 2019

To compare the values of sulfur content in gasoline for the years 2011 and 2019 with the Iraqi (2000) and the European (2010) standards, Figure 6 illustrates this. Whereas the average of the values of sulfur content in gasoline in the year 2011 was (144) ppm and at the year 2019 was (3.6) ppm, which clarifies that these values are within the limits of the Iraqi (500) ppm and European (50) ppm standards for the values of sulfur content in gasoline.

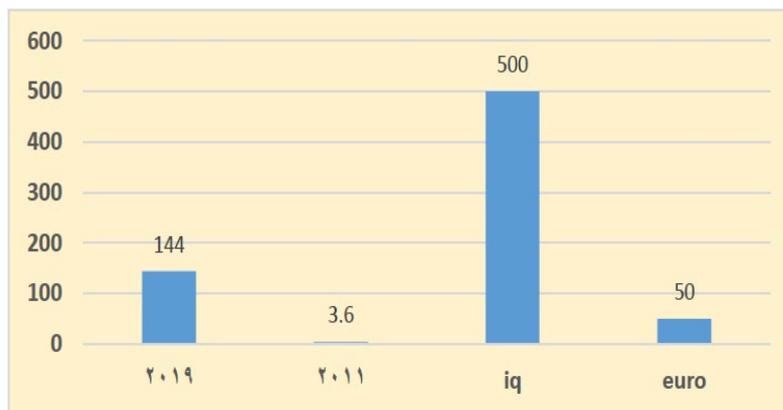


Figure 6: Gasoline Sulfur content compared with IQ and Euro standards

II. Results of Lead Content Test

Table V shows results of the laboratory analysis of the samples examined in the examination laboratories at the Daura refinery in January/2011 in terms of the average, range, and standard deviation of the lead additive in the gasoline. These values are used to obtain the quality control charts resulting in the next Figure 7. It is worth noting that the addition of lead to gasoline was discontinued in 2016.

TABLE V: Analytical results of lead content (g/liter) in January/2011

No. of tests	Sample 1 (Tank A)	Sample 2 (Tank B)	Sample 3 (Tank C)	Range	Mean	St.Dev.
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1	0.15	0.172	0.21	0.15-0.21	0.177333	0.030353
2	0.182	0.165	0.143	0.143-0.182	0.163333	0.019553
3	0.19	0.21	0.175	0.19-0.21	0.191667	0.017559
4	0.165	0.211	0.182	0.165-0.211	0.186	0.023259
5	0.20	0.251	0.114	0.114-0.251	0.188333	0.069241
6	0.152	0.177	0.165	0.152-0.177	0.164667	0.012503
7	0.211	0.154	0.121	0.121-0.211	0.162	0.04553
8	0.132	0.17	0.22	0.132-0.22	0.174	0.044136
9	0.145	0.166	0.21	0.145-0.21	0.173667	0.033171
10	0.172	0.154	0.144	0.144-0.172	0.156667	0.014189
11	0.231	0.197	0.165	0.165-0.231	0.197667	0.033005
12	0.145	0.165	0.176	0.145-0.176	0.162	0.015716
13	0.138	0.21	0.184	0.138-0.21	0.177333	0.03646
14	0.154	0.13	0.17	0.13-0.17	0.151333	0.020133
15	0.176	0.153	0.325	0.153-0.325	0.218	0.093376
16	0.161	0.178	0.117	0.117-0.178	0.152	0.03148
17	0.101	0.171	0.252	0.101-0.252	0.174667	0.075567
18	0.132	0.150	0.210	0.132-0.210	0.164	0.040841
19	0.206	0.095	0.210	0.095-0.210	0.170333	0.065271
20	0.154	0.140	0.187	0.140-0.187	0.160333	0.024132
21	0.220	0.177	0.210	0.177-0.220	0.202333	0.022502
22	0.123	0.161	0.155	0.123-0.161	0.146333	0.020429
23	0.164	0.140	0.175	0.140-0.175	0.159667	0.017898
24	0.132	0.208	0.197	0.132-0.208	0.179	0.041073
25	0.118	0.241	0.121	0.118-0.241	0.16	0.070164
26	0.143	0.162	0.205	0.143-0.205	0.17	0.031765
27	0.161	0.170	0.132	0.132-0.170	0.154333	0.019858
28	0.127	0.156	0.178	0.127-0.178	0.153667	0.02558
29	0.212	0.176	0.160	0.160-0.212	0.182667	0.026633
30	0.153	0.165	0.170	0.153-0.170	0.162667	0.008737
31	0.118	0.210	0.187	0.118-0.210	0.171667	0.047878

In Figure 7 there are no samples out of the control limit, so the process is under control and stable. It is noticed that the lead values don't conform to Iraqi standards since it specified maximum lead content reaches 0.1 g / l. Previously, at al-Daura refinery lead is added to gasoline to enhance the operational performance of automobiles. Lead additive is the cheapest way to raise the quality of gasoline in terms of operational performance. The addition of lead to gasoline in all Iraqi refineries is stopped in 2016 due to the risk to the environment as well as accumulates in catalytic converters and poisons the catalyst. Imported gasoline from outside of the country has relied on octane number (RON 95), it is mixed with the gasoline produced in the Daura refinery with octane number (RON 81) to become gasoline without additives leaded and with octane number (RON 85). Currently, work is underway on the establishment of the isomerization unit process in the refinery to improve gasoline quality properties.

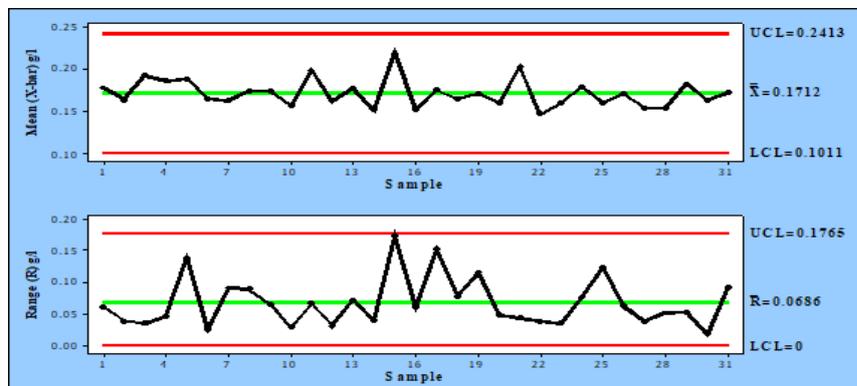


Figure 7: \bar{X} and R charts for gasoline lead additives in January/2011

4. CONCLUSION

The gasoline produced in the Daura refinery in one grade (regular) with octane number 81 through the period January/2011 to January/2019. The sulfur content in gasoline is different depending on the crude oil specifications used, as well as the operational processes at the refinery. In the previous period of 2011, sulfur content was lower (3.6) ppm and conformed to the Iraqi (500) ppm and European (50) ppm standard specifications compared to the samples examined in January/2019. In January/2019 it found that the sulfur content is high (144) ppm, it conformed to the Iraqi standard specification but doesn't conform to the European standard specifications; this is due to the different types of crude oil used at these periods. Also concluded that all the tested samples were within the limits of control. Lead is added as a good improvement to the properties of gasoline but it's not a good effect on the environment. In the Daura refinery and the prior period, lead content as additive was rather high (0.172) g/l and doesn't conform to Iraqi standard specifications (0.1) g/l. The use of lead as an addition to gasoline in al Daura refinery was banned in 2016 while it was banned internationally in the Seventies of the past century. So, we recommended tightening the Iraqi standard and developing it to suit internationally the operational and environmental properties of the gasoline fuels. Intensification of efforts to complete the improvement unit (isomerization) in the Daura refinery to produce gasoline with a high octane number and little impact on the environment. Also, environmental regulations should be developed to protect against high levels of vehicle hazard emissions. Mixing oxygenating materials such as ethanol with gasoline to improve the operating properties of the engines as well as to control environmental pollutants by using harmful substances such as lead.

Abbreviations and Acronyms

AAQ	Ambient Air Quality
EU	European
ppm	part per million
Bstd	British Standard
WWFC	World Wide Fuel Charter
RON	Research Octane Number
MON	Motor Octane Number
RVP	Reid Vapour Pressure
LSRN	Light Straight Run Naphtha
HSRN	Heavy Straight Run Naphtha

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