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Original Paper

The chemistry of rain water in relation to dust fall over Mosul City and Ba'aj Town / Northern Iraq.

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Abstract
Twenty four samples of mineral dust (Nov. 2010 – Oct. 2011) and fourteen samples of rain water (Dec. 2010 --- Apr. 2011) were collected at Mosul City and Ba'aj Town / North Iraq. All the samples were analyzed for (18) elements (Al, As, Ba, Ca, Cr, Cu, K, Li, Mg, Mn, Na, Ni, P, Rb, S, Sb, Sr, Zn) by ICP-MS technique in addition to separate tests and analysis of rain water (pH, EC µs/cm, TDS ppm, SO_4^{2-} ppm). The imprints of the nature of the land use, the soil type and the geological setting noticed in the chemistry of the mineral dust suggest that mineral dust emission is of local source(s). The positive correlation between monthly dust-fall rate with rain content of most elements indicate to the atmospheric dissolution or leaching of element at dust grain/rain droplet boundary. Wash-out ratio (W) of the studied element show great deal of variation but they are held within two extremes i.e. the least mobile Al and most mobile S probably as SO_4^{-2} (Al, Mn, Ni, Cr, Rb, Mg, K, Ba, As, P, Zn, Ca, Sr, Na, S as SO_4^{-2}). Element Cu, Li and Sb are neglected on the basis of poor analysis accuracy and precision. Some elements (Al, Ca, Sr, Na, S as SO_4^{-2}) in the series may comply with their known behavior in the weathering zone but the mobility of others is not common at the supergene environment. Mineral dust is considered to change composition and size once it is in the atmosphere. It is thought that such changes are the result of heterogeneous reactions with trace atmospheric gases. At present this type of reaction is not fully define for all and each element constituents of the mineral dust, especially those of environmental interest.

Keywords: chemistry of dust/droplet boundary, wash-out ratio, heterogeneous reactions.

Introduction:

n general, the chemistry of rain water is derived from natural or /and anthropogenic sources (Jickells and Knap, 1984). The natural source is more effective in undeveloped area of regional arid and semi-arid weather like the case of the present study i.e. Mosul City and Al-Ba'aj Town, where particulate matter of the atmosphere is dominated by mineral dust. Recently the weather conditions start to change in the countries of the dust belt of the Middle east region, with three folds more frequent events and intensities of dust storm occurs (Gerivani *et al.,* 2011) and occasionally accompanied with rain precipitation. This lead some time, to what is known as muddy rain in the study area. The muddy rain is noticed also at several places across the world (Keller, 2011). On the other hand, the colored rain precipitation noticed at several localities across the world was the subject of controversy and debate framed within superstition discussion. It is generally known that color of true solution is generated by the dissolution of highly soluble colored organic or inorganic material and the colored colloid solution, formed by deflocculating of colored colloidal particle forming hydrosol colloidal system. Polarization of water molecules play an important role in the formation of both colored solutions, whereas the concepts of Electrical Double Layers (Berner, 1971) or Surface Complexation (Stumm *et al.,* 1987) theories dominate the discussion of the reactions and interactions across the interface of water (rain) / solid (dust). The discussion

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here is not extended any further because of very limited data and instead, these theories and concepts will be referred properly when element distribution across the water/solid interface is discussed.

The chemistry of rain water is investigated at world regional (Galloway *et al.,* 1982; Church *et al.,* 1982) and local area (Jickells and Knap, 1984) countries. The dissolution of single, pair and group of elements from natural and anthropogenic atmospheric particulates into rain and sea water were thoroughly studied (Statham and Chester, 1988; Thuroczy *et al.,* 2010; Maring and Duce, 1987; Loye-Pilot *et al.,* 1986; Hogde *et al.,* 1978; Jickells *et al.,* 1984). The present study is concerned with the distribution of (18) elements between monthly collected contemporaneous rain water and dust fall samples during rainy season of 2010/2011 at Mosul City / North Iraq and at Ba'aj Town located 30 km. south of Sinjar Town and at about 150 km. west of Mosul City (Fig. 1). The study area belongs to arid and semiarid climate with precipitation rate below 100 mm. per year. In general, the rainy season covers few months and usually start on November/December and extend to January, February , March and April next year. Recently, the study area and Iraq as a whole witnessed great changes in rain precipitation and dust fall rate, since several rain and dust storms strike the Middle East region.

Figure -1: Map of IRAQ showing study area.

Samples and Methods:

Dust samples (24) were collected through a funnel of 23 cm diameter firmly attached with stopper to a conical flask of 1 liter volume has been placed in position in guard frame for a month at a height 0f 3 – 6 m. (authors house roof at both Mosul City and Ba'aj Town). The amount of dust fall (gm/m^2) collected over the period Nov. 2010 - oct. 2011 is shown in Table (1).

Month and Year	Dust fall (gm/m^2) over Mosul City, Ba'aj Town	
Nov. 2010	1.6	2.0
Dec. 2010	1.33	2.25
Jan. 2011	0.19	0.62
Feb. 2011	1.5	3.0
Mar. 2011	6.0	22.0
Apr. 2011	20.5	64.0
May 2011	2.3	21.0
Jun. 2011	2.75	24.3
Jul. 2011	2.23	24.0
Aug. 2011	0.73	12.3
Sep. 2011	1.25	10.5
Oct. 2011	0.75	13.8

Table (1): Amount of dust fall (gm/m^2) over Mosul City and Ba'aj Town for the interval (Nov. 2010 – Oct. 2011).

Rain water samples (14) were collected in a cleaned wide plastic container placed in an open area at authors house in Mosul City and Ba'aj Town. Details of conditions associated with rain sample collection such as commence time of rain, intensity, duration, the occurrence of dust and thunder storms are documented as well as the date and code of rain sample (Table 2). No rain precipitation is recorded in March at both sites.

Area	Sample No.	Date	Start Duration Note Time Hours
Mosul City	RM1	11/12/2010	01 am 6-7 1st. rain Interm. Strong wind
	RM2	31/12/2010	05 am 12 Shower
	RM3	15/01/2011	11 am 12 Interm. very cold
	RM4	29/01/2011	12 noon 7 Interm.
	RM5	15/02/2011	8.30 pm 4 Heavy large quantity
	RM6	21/02/2011	01 am 7 Shower
	RM7	05/04/2011	08 pm 6 Interm. Thunder storm Preceeded by dust
	RM8	21/04/2011	03 pm 48 Last rain Interm. Heavy shower
Ba'aj Town	RB1	11/12/2010	05 pm 13 Interm. Throughout dusty
	RB2	30/12/2010	10 pm 12 preceeded strng wind Dusty
	RB3	04/01/2011	05 am 14 No dust
	RB4	08/01/2011	09 pm 4 Calm conditions
	RB5	14/01/2011	10 pm 10 Intermittent
	RB6	30/01/2011	08 pm 10 Intermittent
	RB7	15/02/2011	- - Sporadic interm. Small quantity
	RB8	21/04/2011	Last rain , heavy shower , dust and thunder storms

Table (2): Details of rain precipitation over Mosul City and Ba'aj Town for the interval Dec. 2010 – Apr. 2011.

The collected samples (dust and rain water) were analysed using ICP – MS instrument type (Perkin Elmer Elan 6000) at Acme Analytical Laboratories Ltd, Vancouver/Canada. Detail of the method of analysis is available at internet site www.acmelab.com. The dust samples were ground by agate mortar to pass 250 mesh (about 63 mμ) sieve. The dust samples were subjected to thorough homogenization and 1 gm of the sample were weight and placed in small well cleaned and labeled polyethylene bag and send by air to the above stated destination. The rain water samples were subjected to filtration using 0.5 mμ porosity filter paper type (Sartorius AG / Germany) attached to Buchner Funnel at the Chemistry Laboratories of the Educational College of Mosul University. Later, 60 ml of the filtered sample is placed in a well cleaned labeled and firmly closed container of the type (HDPE) which is shipped to Acme Ltd./Canada for analysis. The water samples were subjected to various measurements such as EC μs/cm, TDS ppm and pH as well as SO_4^{-2} (Table 3).

The evaluation of the analytical results were made through simple calculation of precision based on the difference between repeated analysis of one sample (T8M Tigris river at Mosul City and B5F Ba'aj fine fraction dust) and comparison between determined and expected analytical result of Acme standards of water (TMDA-70) and Acme standard for solid (OREAS24P). The average precision of the analysis of (18) element in water and dust samples amounts to (2.7%) and (3.5%) respectively (excluding Cu (12.8%), Li (12.05%) and Sb (25%) from precision calculation of dust analysis). The average accuracy of the analysis of (18) elements in water and dust samples reach (4.4%) and (3.2%) respectively (excluding Sb (11.0%) from accuracy calculation of dust sample). Accordingly the element Cu, Li and Sb are excluded from any further discussion. The analytical result is shown in Table (3) for rain water samples and Table (4) for dust samples at Mosul City and Ba'aj Town.

Result and Discussion:

It can be noticed in Table (1) that the highest dust fall (20.5 gm/m^2 for Mosul City and 64.0 gm/m^2 for Ba'aj Town) and the lowest dust fall (0.19 gm/m^2 for Mosul City and 0.62 gm/m^2 for Ba'aj Town) were recorded in April 2011 and Jan. 2011 respectively. The average element constituent show more variation in rain water relative to dust samples (Table 3 and 4).

Ba'aj Town is located near salt deposit (semi-dry Snaisla salt lake) within brown and desert soil rich in carbonate (Caliche) and sulfate (Gypcrete) and lie at the rim of dry and hot weather area known as Palmyra desert along the Iraqi / Syrian international border. Provided that the dust emission is mainly of local source (Gerivani, 2011) and the atmospheric mineral dissolution is carried out within adequate time of more than 20 minutes (Thuroczy *et al.,* 2010), then the higher Ca-, SO_4^{-2} – and Sr contents in rain water (Table 3) of Ba'aj Town (34.90 ppm, 41.25 ppm and 202.8 ppb respectively) relative to Mosul City (28.3 ppm 29.65 ppm and 153.28 ppb respectively) can be explained, whereas , the extensive agricultural activity around Mosul city and the common uses of phosphate fertilizers among farmers may accounts for the three folds higher p-content in rain water at Mosul City (93.0 ppb) relative to Ba'aj Town (30.33 ppb) (Table 3). The lower average pH value (7.24) of Mosul's rain relative to that of Ba'aj (7.97) site may suggests the presence of more soluble species (Mn^{+2}), hence, the enrichment of Mn in rain water at Mosul City (7.04 ppb) relative to (1.52 ppb) of Ba'aj Town. The variation in pH values coincide with the differences in EC µs/Cm (142.87 and 168.45) and TDS ppm (101.39 and 119.63) for Mosul and Ba'aj sites respectively Table (3). Soluble phase of (Al) show an increase in content in Mosul's rain water (19.70 ppb) relative to Ba'aj rain water (6.83ppb) which reflect more hydrolysis of finer size of feldspar grains at Mosul city. The dust's constituent of both sites (Table 4) show closer values (within analytical errors) which reflect similar mineralogy represented by Carbonate, Sulfate, Quartz, Feldspars, Clay and other unidentified fugitive mineral grain in addition to organic material (Al-Bakri *et al.,* 1984; Karydis *et al.,* 2011) and/or reflect more processed or bleached (with nitrate) Asian mineral dust (including those of the present study) relative to African mineral dust (Su and Toon, 2011). The chemical constituents of dust sample (Table 4) are comparable with the exception of Sr which show more variation in content ppm (361.96 at Mosul and 526.25 at Ba'aj sites) relative to Ca content wt.% (12.33 at Mosul and 12.07 at Ba'aj sites). Three minerals namely Feldspars, Gypsum and Calcite may account for the fore-mentioned data, because both feldspars and gypsum minerals contain less Ca- content and high Sr content relative to calcite and this in turn, explain the increase in Ca content and the decrease in Sr content in the dust sample of Mosul City relative to dust sample of Ba'aj town. The higher average sulfur content (0.982 wt.%) and the appreciable increase in Al- , Na- and k- content wt.% (5.656 , 0.597 and 1.096 respectively) in Ba'aj's dust confirm the previous conclusion concerning relative enrichment of Ba'aj's dust with Feldspar and Gypsum , whereas Calcite increase in content in Mosul's dust. Phosphorous content show no appreciable differences in the dust of both sites. This may represent the less soluble phases left over after the dissolution of the highly soluble phosphorous phases. The last remark may also applied to other dust's constituents of both sites.

The studies of (Jickells and Knap, 1984; Merrill and Duce, 1983) investigate into the behavior of chemical element at the dust / rain droplet through calculation of suggested parameters like Wash-out ratio (W) using the following mathematical equation (1).

W = conc. in rain water (ng/l)/conc. in air (ng/cm^2)…(1):

The present study followed the same mathematical equation for the calculation of Wash-out ratio based on the monthly data of the chemical composition of dust and rain water samples (Table 3 and 4) obtained in the present study. This is different from the calculation of (Jickells and Knap, 1984) based on data obtained at different time. Table (5) show the raw monthly data used in the calculation of Wash-out ratio (W) and the average (W) is also shown at the end of the same table. The dust and rain water samples show variation in element content, however, rain water samples show much more variation in element content relative to dust samples. In fact, the rain water samples show prevailing maximum (bold number) and minimum

(underlined number) element content in April and January months respectively (table 5). Such element distribution is shown by (11) elements at Mosul city relative to (8) elements for Ba'aj Town. This is coincide with maximum (20.5 gm/m^2 and 64 gm/m^2) and minimum (0.19 gm/m^2 and 0.62 gm/m^2) dust fall rates at Mosul and Ba'aj sites respectively (Table 1). Among individual element, Phosphorous show much higher content in Mosul's rain relative to Ba'aj's rain which emphasizes the uses of phosphate fertilizers in agriculture activity around Mosul city. The dissolution of sulphates (and feldspar) may explain the increase in calcium content in Ba'aj's rain in comparison with Mosul's rain.

The calculation of wash out-ratio (W) requires recalculation or conversion of rain data expressed by ppm (µg/ml. × 10^6) and ppb (µg/l × 10^3) to ng/l (nano gram/liter) unit and dust data in wt.% (g/100g × 10^9× monthly dust-fall rate g/m^2 ÷ 10^4) ppm (µ/g × 10^3 × monthly dust-fall rate g/m^2 ÷ 10^4) to unit of ng/cm^2 (nano gram/square centmeter). Wash out ratio show great deal of variation among elements, collection site and month, nevertheless, certain variation trend can be noticed for most element where lowest W value is recorded for the month of maximum dust-fall rate i.e. April, where as the highest W values were documented for the month of minimum dust fall rate i.e. January. Grain size variation may explain such relation, where dust grain at Ba'aj town is of larger size and hence element dissolution is at minimum rate. This is backed by lower average W value of all element at Ba'aj site with the exception of Av.W of sulfur which amount to (1182 & 16667) at Mosul and Ba'aj sites respectively (table 5). This explain the increase in (Ec 168.45µs/cm), (TDS 119.63 ppm) and (pH7.97). The studied elements can be arranged in the order of increasing average W value (Table 5) as in the following:

Al-Mn-Ni-Cr-Rb-Mg-K-Ba-As-P-Zn-Ca-Sr-Na-S.

This series of element behavior is partly compared with their mode at the weathering zone. It is generally known that (Al and S as SO_4^{-2}) represent the extremes mobility modes where the former is considered less mobile and the second is highly mobile element probably as soluble sulfate radical. The fore - mentioned element series is not to be mistaken with the actual element concentration in the rain samples. Excluding Al, S, Na, Sr and Ca, the W variation is not correlated with known element mobility series and partly defies element distribution concepts of Goldschmidt, 1954, Ringwood, 1955 and Burns, 1970, and known supergene parameters like activity product and ionic potential and colloid concepts, however, it can be explained by some other uncommon (unknown ?) geochemical parameters prevailed at the dust – cloud system. Interactions between mineral dust and clouds are the subject of considerable research due to the influence of such interaction on climate in general and on cloud droplet formation (Kelly *et al.,* 2007) and may alters the hydrological cycle which affects quality and availability of fresh water. Once start in atmosphere, mineral dust particle change size and composition through heterogeneous chemical reactions (Grassian, 2002). Such reactions involve trace atmospheric gases like nitrogen oxides, sulfur oxides, ozone and organic gases. The impact of such gases on the chemistry of mineral dust is not well defined for each individual element, excluding the attempts made to study solubility changes of Al – and Fe – containing dust as a result of heterogeneous reactions (Ruasinghege and Grassian, 2009). Further detail work is needed to cover heterogeneous reactions of more individual element including those of environmental interests.

Conclusion:

The collected samples of rain precipitate and dust-fall over Mosul and Ba'aj sites were analysed for (18) elements (plus other test and analysis) using ICP-MS technique at Acme Analytical Laboratory Ltd./Canada. The analytical data show that the average composition of rain samples are more variable than those of mineral dust samples. This reflects similar mineralogy and/or processed or bleached mineral dust during transportation in the Troposphere. The rain content of most element is positively correlated with the

amount of mineral dust collected at each month. Such relation is shown by more elements at Mosul relative to Ba'aj sites. Grain size of the collected mineral dust may partly account to the variation of element content of rain samples. Element distribution between rain droplet and mineral dust were followed through calculation of wash-out ratio (W). Excluding Sb, Li and Cu for poor analytical accuracy and precision, the studied element can be arranged in order of increasing average (W) value: Al-Mn-Ni-Cr-Rb-Mg-K-Ba-As-P-Zn-Ca-Na-S. Some of the elements in the series can be compared to the distribution mode at the weathering zone, others is not related to concepts of element distribution in the supergene.

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الملخص العربي

كيميائية مياه الأمطار وعلاقتها بالغبار الساقط فوق مدينة الموصل وقضاء البعاج شمال العراق

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الخلاصة

جرى تجميع (٢٤) أنموذجا من الغبار الساقط (تشرين الثاني ٢٠١٠ – تشرين الأول ٢٠١١) و(١٤) أنموذجا من مياه الأمطار (كانون الأول ٢٠١٠ – نيسان ٢٠١١) في مدينة الموصل وقضاء البعاج /شمال العراق. وخضعت جميع النماذج إلى تحليل ثمانية عشر عنصر وهي:
(Al , As , Ba , Ca , Cr , Cu , K , Li , Mg , Mn , Na , Ni , P , Rb , S , Sb , Sr , Zn) باستخدام جهازتحليل حث البلازما المزدوج – المطياف الكتلي (ICP – MS) فضلا عن فحوصات وتحاليل أخرى (pH , EC µs/cm , TDS ppm , SO_4^{-2} ppm). إن البصمة المسجلة لكل من طبيعة استخدامات التربة والوضع الجيولوجي و نوع التربة والملاحظة في التركيب الكيميائي للغبار الساقط تشير إلى المصادر المحلية لانبعاثات الغبار، أما العلاقة الموجبة بين المعدلات الشهرية للغبار الساقط وبين محتوى الأمطار من معظم العناصر، فتشير إلى إمكانية حدوث إذابة جوية أو خلب عند تماس العالق من حبيبات الغبار وقطرات الأمطار في الغلاف الجوي. تتبان كثيرا نسبة الغسل (wash-out ratio) للعناصر موضوع البحث ولكنها تنحصر بحدين يتمثلان بعنصر الألمنيوم الأقل حركة و عنصر الكبريت بشكل كبريتات ذائبة الأكثر حركة ويمكن ترتيب العناصر حسب نقصان نسبة الغسل وكما في الأتي:
Al-Mn-Ni-Cr-Rb-Mg-K-Ba-As-P-Zn-Ca-Sr-Na-S . وأهملت نتائج تحليل كل من النحاس والليثيوم والانتموني بسبب انخفاض دقتهم وصحتهم التحليلية. وباستثناء تسلسل حركة عناصر الكبريت و الصوديوم و الستروتتيوم و الكالسيوم و الألمنيوم المعروفة في البيئات السطحية, فان بقية العناصر لا يوجد ما يشبه تسلسل حركتها في البيئات السطحية الشائعة وهي بالتالي تبدو بأنها تتقاطع مع المفاهيم الدارج قبولها لتوزيع العناصر في البيئات السطحية. ويعتقد بان معادن الغبار تخضع للتغيير في تركيبها وحجمها حال وجودها في الغلاف الجوي. ويعتقد بان مثل هذه التغييرات ناتجة عن تفاعلات غير متجانسة لغازات الجو الأثرية. إن مثل هذه التفاعلات غير معروفة تماما لكل عنصر من عناصر مكونات معادن الغبار وخصوصا تلك التي لها أهمية بيئية.

الكلمات المفتاحية: كيمياء تماس الغبار بالقطرات، نسبة الغسل، التفاعلات غير المتجانسة.