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A review of constructive analytical methods for determining the amount of aluminum in environmental and human biological samples

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ABSTRACT

Aluminum is a toxic metal and cause pollution in soil, water, and air. Afterwards, a lot of patients suffer renal failure due to the accumulation of aluminum in the tissues of kidneys. Also, high concentration of aluminum in plants tissues makes agricultural food toxic. Therefore, measuring aluminum in water, soil, air, human organs, tissues of plants and each food (or agricultural product is so necessary for protecting human healthy. In this paper, the analytical methods which have been applied for measuring the amount of aluminum from 1970 to 2019 are focused on. Also, the effect of some parameters such as pH and temperature on decrease or increase in the amount of aluminum in water and other samples are stated. Ultimately, it is worthwhile to mention the analytical methods which are more time-consuming, cost-effective, applicable, and precise for determining the amount of aluminum now. In this review, the analytical methods such as fluorimetric, ICP-MS, colorimetric, graphite furnace/flame atomic absorption spectrometry, etc. which have been applied for measuring the amount of aluminum (especially Al^{+3}) in environmental and human biological samples are assessed.

1. Introduction

Aluminum is a toxic metal. This toxic metal has polluted a lot of water wells, springs, lakes, groundwater aquifers, rivers, and soil in most parts of the world. Unfortunately, aluminum accumulates in plants' tissues. Also, aluminum foils during a little time penetrated into food. Nowadays, a lot of patients who have suffered chronic renal failure in the world were evaluated. People are living next to the mines are exposed to this toxic metal. In addition, aluminum which conveys to our bodies by having agricultural products and drinking water accumulates in the tissues of organs, and after a little time, their functions are suppressed. Also, a lot of analytical methods such as weight loss measurements, environmental scanning electron microscopy, electro-thermal atomic absorption spectrometry, colorimetric, kinetic fluorimetric, chelation, and inductively coupled plasma-mass spectrometry have been presented by a lot of researchers in the world for determining the amount of aluminum especially Al^{3+} in water, soil, and biological samples. World

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health organization (WHO) has studied a lot on the amount of allowed aluminum in every organ of our body [1-5]. Wong et al reported about aluminum and fluoride contents of tea, with emphasis on brick tea and their health implications [1]. Mameri et al showed that defluoridation of water by small plant electrocoagulation using bipolar aluminum electrodes can be separated many pollutions from matrix but can be caused aluminum pollutions in environment [2]. Aluminum and heavy metal contamination of ground water was evaluated and determined by Momodu et al. by ET-AAS [3] and Havas et al. reported analytical method based on aluminum bioaccumulation in soft water at low pH [4]. Shaw et al. showed the effect of aluminum in the central nervous system (CNS) which have caused to toxicity in humans and animals [5] and Brown et al. research in analytical method in aluminum species at mineral surfaces by instrumental analysis [6]. In 1995 -1996, Hodson et al and Neuville et al used different methods based on for measuring Al in plants and glasses which was effect in environment and humans [7,8]. Krewski et al showed Human health risk assessment for aluminium, aluminium oxide, and aluminium hydroxide in humans in 2007 [9] and uptake of fluoride, aluminum and molybdenum by some vegetables from irrigation water was studied by Khandare et al. at 2006[10]. Occasionally, aluminum was used in different industry with variety of method which was hazard effect in humans and environment. For examples, in 2003, Zn/Al hydrotalcite-like compound (HTlc) was used for removal of fluoride from aqueous solution by Das et al [11]. Cosby et al used a modeling the effects of acid deposition for extraction of aluminum and Assessment of a lumped parameter model of soil water and stream water [12]. Human exposure to aluminum was evaluated by Niu and Exley [13-15]. Dunemann et al., showed, simultaneous determination of Hg (II) and alkylated Hg, Pb, Al and Sn species in human body fluids using SPME-GC/MS-MS [16]. Messerschmidt et al used adsorptive voltammetry procedure for the determination of platinum and aluminum baseline levels in human body fluids [16] and release of metal ions from dental implant materials was shown through determination of Al, Co, Cr, Mo, Ni, V, and Ti in organ tissue by Lugowski et al [17]. Also, a review paper about determination of metal-binding proteins by liquid chromatography was reported by journal of Analytical and bioanalytical chemistry in 2002 [18]. The concentration of other metals in human body was evaluated based on different analytical methods by GC-MS, SPME-CGC-ICPMS, GC-MIP-AED, and anodic stripping voltammetry [19-26]. Khanhuathon et al used a spectrophotometric method for determination of aluminum content in water and beverage samples employing flow-batch sequential injection system at 2015 [27]. Liu et al applied Determination of metals in solid samples by complexation/supercritical fluid extraction based on gas chromatography and atomic emission detection for determination metals [28]. Other methods such as, fluorescence detection and film microelectrode based on voltammetry was used for determination metals in body fluids was used [28-31]. The speciation of aluminum in human serum was done by Sanz-Medel et al in coordination chemistry reviews [32]. In 2017, the Nano analysis in biochemistry for separation of aluminum in blood of dialysis patients has been developed with graphene oxide Nanoparticles which have been dispersed in Ionic Liquid [33].

Moshtaghie et al showed a method for aluminum determination in serum of dialysis patients by F-AAS [34]. Halls [35], Bettinelli [36], and Narin [37] have determined the amount of aluminum in dialysate fluids and environmental samples by ET-AAS [34-36]. Aluminum in biological fluids and dialysis patients was determined with 8-hydroxyquinoline/ extraction/fluorimetry by Buratti [38] and Davis et al showed a method for determination of aluminum in human bone [39]. Also, aluminum in biological and water samples based on cloud point extraction /furnace atomic absorption spectrometry was developed by Sang in 2008 [40]. Selvi et al introduce a method analysis for determination of Aluminum in dialysis petients by Atomic Absorption Spectrometry by

Coprecipitation with $LaPO_3$ in 2017 [41]. Many methods for determination of aluminum was done such as lubricating oils emulsified in a sequential injection analysis system, tri-calcium Phosphate (TCP), eriochrome cyanine with CPE, dopamine as an electroactive ligand, by ET-AAS/F-AAS from 2002 to 2016 [41-45]. The risk assessment of aluminum based on determination of aluminum in food/meat was developed by Bassioni and Juhaiman in 2012 and 2015 respectively [46,47]. Novel method for determination aluminum in human brain tissue using lumogallion /fluorescence microscopy was obtained by Mirza in 2016 [48]. Sorenson et al. showed that aluminum in the environment and human samples can be evaluated and Rana Sonia used Schiff base modified screen printed electrode for selective determination of Al^{3+} in different matrix in 2017 [49,50]. Determination of aluminum with deep eutectic solvent/microextraction method was developed in water and food samples in 2018 by Panhwar et al [51] and Lia-yan Liu used by ICP-AES [52]. Zuziak et al., applied a voltammetry for determination of aluminum in 2017 [53]. Chao, Litov and Dórea introduced a analytical method for breast milk samples [54-56]. In addition, many analytical method was used for determination and separation Al from different matrix such as blood and water samples [57-67]. In this research paper, it has been tried that the analytical methods which have been used for quantifying the amount of aluminum in water, soil, and biological samples will be assessed. Also, the assessment helps all scientists and researchers find and use the best analytical approaches which are more precise and accurate for determining the amount of aluminum in the samples. Moreover, this review paper presents the cost-effective and time-consuming methods which have been used since 1970 to 2019.

2. Experimental Procedures

2.1. Methodology

In this section, the analytical methods for measuring aluminum in human body and Environment matrix were studied. The different metals spread on the earth's crust; aluminum (Al) has the third most abundant element as compared to other metals with percentage of 88% gram per kilogram. The free aluminum has never seen in nature and mostly exists in aluminum silicate minerals and rocks [6-8]. Aluminum is also exist in different matrixes such as; air, soil, water, foods and environment. Based on weathering of metals, the metals enter to waters and human. Human activity by industrial processes, waste water effluents and dust as a major constituent of aluminum compounds can be released in air, waters, vegetables and human [9,10].

Many parameters such as coordination chemistry, pH, and characteristics have effects on behavior of aluminum in environment [11,12]. In addition, by biogeochemical cycle of aluminum, geochemical formations and soil particulates, and air particles change to aqueous environments and then enter to soil or sediment. Aluminum is widely was used as applied metal in all world as a building material. The different forms of aluminum compounds have been made by mixing of other elements. In different pH and conditions, Al can be used with other ions with different valence states. Aluminum is used in many fields such as antacids (Al-Mgs), food additives ($Al(OH)_3$), skin ointment, cosmetics products, container, and as a metal contaminants appeared in milk products, juice, fish, and tea [13-21]. Aluminum also enters in drinking water due to the water treatment process, weathering rocks and soils and acid raining. Aluminum is used in many industries due to special physical and chemical property. So, aluminum particulates are seriously exposed by workers of aluminum factories. The absorption of aluminum in human body was achieved by many materials such as citrate, Fe in hem, Ca, F, etc. [21-26]. A precise, sensitive and selective spectrophotometric method for determination of Al^{3+} using (ECR) as a chromogenic reagent in the presence of N, N dodecyl trimetylammonium bromide (DTAB) has been developed by Khanhuathon et al in 2015 [27]. Modern spectrophotometric approach for determining Al^{3+} in waters and soft drinks by using eriochrome cyanine R(ECR) has been

presented by Khanhuathon et al. In addition, in their approach, R(ECR) which is a chromogenic reagent has been used in the presence of N, N dodecyl trimetylammoniym bromide (DTAB). Their study shows that at 584 nm, a maximum absorption is obtained when pH is equal to 5, Al-ECR complex is used. In the mentioned study, the effects of some parameters such as amount and type of surfactant, pH, and concentration of EOR on the rate of absorption of Al have been assessed. Moreover, after optimizing the condition, a linear range of 0-01 to 0.50mgL^{-1} aluminum is received. Also, the range of detection has been 0.0020mgL, and the range of quantification has been 0.0126mgL. Based on their study, the relative standard deviation of their approach has been 13% [28-33]. When the rate of absorption is 0.05 mgL^{-1}, the applicability of their approach for determining Al contents is tested on many water samples and soft drinks. Also, the outcomes of their method are similar to the ICP-AES method. In addition, their methods can recovery up to 80%. In addition, based on their studies, R(ECR) is a suitable reagent for determining the amount of Al in every kind of water. Moreover, in their approach, some cons of using Al-ECR complexes such as time consuming have been there. In addition, in the mentioned approach, pH and temperature of the complexes must be controlled. Also, in the study, the effects of various surfactants on two properties of the complexation and reagent, i.e. spectra and sensitivity, when the pH is equal to 5 have been considered. The consideration demonstrates the highest sensitivity of the absorption spectrum and maximum wavelength which is equal to 584 mm is obtained. Moreover, DTAB (dodecyl trimetylammonium bromide) is the most effective surfactant for improving the sensitivity of AL-ECR [34-42]. Moreover, the study demonstrates that the maximum absorption is obtained when pH is equal to 5, as seen in the following figure (Figure 1).

Yildiz et al have studied on determining Al for tri-calcium phosphate (TCP) anhydrous powder by flame atomic absorption spectrophotometer in 2016. Based on their study, there is about 350 mg/kg (w/w) of aluminum in tri-calcium phosphate anhydrous powder. In the study, the amount of their metal in the powder is determined using atomic

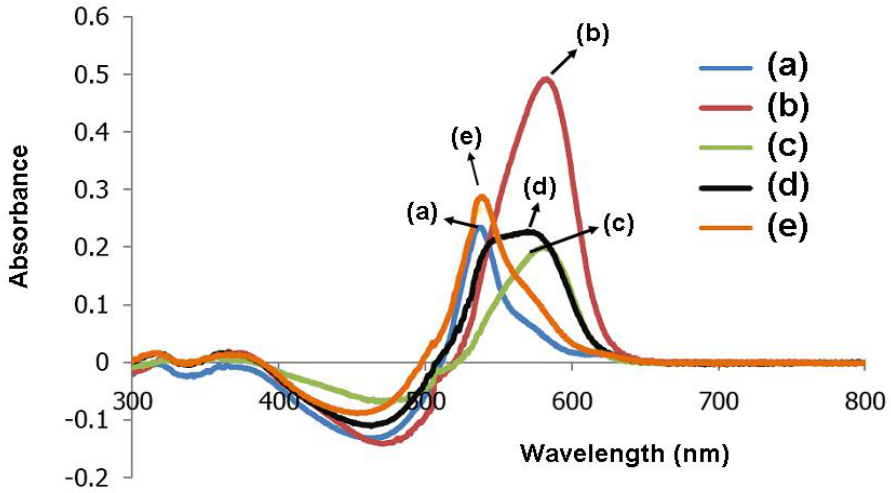

Fig. 1. Spectra of Al-ECR complexes in the absence (a) and in the presence of various surfactants (b) DTAB, (c) CTAB, (d) SDS and (e) Triton X-100. Conditions: 0.2 mg L^{-1} Al, 0.15 mmol L-1 ECR and 3 mmol L^{-1} surfactant, pH 5.

absorption spectrophotometer. In their approach, the outcomes of Al which have been obtained by using the N_2O-C_2A_2 flame are similar to the previous studies. Also, the standard calibration curve has been done automatically. Moreover, the accuracy of their method has been considered using recovery test of aluminum. Finally, their results show that the amount of Al has been 0.5 mg/kg in the detection limit and there is a suitable linearity based on their analysis [43].

Hejri et al has studies on determining trace aluminum with eriochrome cyanine R after cloud point extraction in 2011. In their study, for determining ultra-trace amounts of Al^{3+} in well waters, the approach of cloud point extraction has been used. In addition, during the study, the surfactant of cetyltrimethylammonium bromid has been used. Based on their study, linearity has been ranged from 0.2 ng mgL^{-1} to 20.0 ng mgL^{-1}. In their study, the limits of detection is about 0.05 ng mgL^{-1}; moreover, these limits are governed for determining Al^{3+}, In their method, an interaction is there between surfactants and metal-dye complex. Also, in the mentioned method, a ternary complex which involves surfactant monomers is formed. Moreover, the efficiency of their method increases when pH is equal to 5.5. Also, their results show that by increasing and decreasing pH, sensitivity will be reduced [44]. In addition, determining aluminum in biological fluids using an electroactive ligand, dopamine have been studied by Bi et al in 2002. Based on their study, by increasing Al concentration, decreasing trend of the differential pulse voltammetric anodic peak is linear. Also, when the experimental conditions are optimum, two linear ranges which are about 5.0 $\times 10^{-8}$ to 4×10^{-7} M and 4.0 $\times10^{7}$ to 7.2 $\times 10^{-6}$ M Al^{3+} are gained. They have selected some samples which have been obtained from synthetic renal dialysate, human whole blood, the urine of patients who have suffered diabetes. The amount of Al^{3+} has been measured in the samples using dopamine. Afterwards, they have verified the depression electrochemical activities of DA by making a comparison between electrochemical behaviors and the spectroscopic responses. In their study, an indirect method for determining Al^{3+} with an electroactive ligand has been applies in biological fluids using differential pulse voltammetric. Finally, in the study, a good and suitable agreement between the results of the study and previous studies have been made [45]. In addition, Will et al have considered two methods for determining Al^{3+} concentrations in blood in 1990. In their study, the amount of Al which causes chronic renal failure in patients have been mentioned. In their first method, plasma samples have been diluted with HNO_3/ triton x-100 matric four times. Also, in the second method, samples are diluted with an equal volume of Mg $(NO_3)_2$ matrix, moreover their samples have been atomized from a L'vov platform. In addition, analytical recovery of Al which has been added to is about 98%. Also, they performed and tested the samples in sealed containers to maintain them against contaminations.in the first method, a 10-ml sample which is the representative of whole blood has been selected, then centrifuged. Afterwards, the plasma has been washed by using a disposable polyethylene pasteur pipet at 4°C. In their second method, samples have been diluted in de-ionized water with the solution of $Mg(NO_3)_2.6H_2O$ which is 5-46 mmoL L^{-1}. Also, for analyzing the samples, atomic-absorption spectrometric and electrothermal graphite atomizer with the instruments of model 5100-PE and 5100-PC have been used [46]. Bassini et al have studied on the amount of Al which causes that food would be contaminated in 2012. Unfortunately, transferring aluminum from foil to food is hazardous. In their study, three techniques such as weight loss measurements, environmental scanning electron microscopy, and inductively coupled plasma-mass spectrometry have been used for analyzing the samples which have been selected from the foods that exposure to aluminum foil. The outcomes of their studies show that in acidic food and cooked food, the amount of Al is higher in comparison with the other kinds of foods.in addition, based on their results, the leaching of Al from foil into food solution as a solid phase is the same as liquid and vapor phases.

Moreover, by increasing temperature, leaching of Al is increased. Also, when pH decreases, the rate of leaching rises. Moreover, using aluminum foil causes a lot of diseases in human body [47]. Al Juhaiman has studied the cons of aluminum foil which has been wrapped around baking meat in 2015. Although who has reported the negative effect of Al foils, unfortunately, a lot of companies of producing food use it.in their study, the effect of temperature and cooking time on the amount of Al which leaches into the food has been assessed. Based on their results, the leaching of Al in fish has been the highest, and in chicken, the rate of leaching of Al has been the lowest. Also, cooking foods in aluminum pans or other aluminum dishes increase the rate of leaching. In addition, based on corrosion weight equation, the rate of Al leaching in fish, after 60 minutes cooking, is equal to 38.67 mg Al/kg. Also, when this rate is obtained, CR is about $(7.000.71\pm)\times10^{-3}$ [48]. In addition, Exley et al have studied on the accumulation of aluminum in brain tissue of human in 2016. Based on the study, when aluminum accumulates in brain tissue, the human will suffer neuro-degenerative diseases which include Alzheimer's disease. Also, a few studies have been done on visualization of aluminum. In this study, for measuring aluminum in brain tissue, transversely heated graphite furnace with atomic absorption spectrometry has been used. In their study, fluorescence microscopy and the flour lumogallion have been developed and validated for showing the presence of aluminum in brain tissue. Their research has shown that fluorescence of aluminum in brain tissue is different with other metals that accumulate in brain tissue. Orange fluorescence shows that there is some aluminum in brain tissue. Their method, i.e. fluorescence microscopy helps physicians to get more information about the amount of Al in brain tissue, and thereby the prevention of being suffered Alzheimer's can be followed. Also, Exley et al have used 4-chloro-3-(2,4 dihydroxyphenylazo)-2-hydroxybezane-1-sulphonic acid as a lumogallion for measuring the amount of Al^{3+} in brain tissue. Moreover, the lumogallion has been used for measuring the amount of Al^{3+} in seawater. When it is used in seawater, the limit of detection of CA is equal to 2 Nm. Furthermore, the method of fluorescence has been used for measuring and determining the amount of Al^{3+} in plants. Moreover, during the test of determination of amount of Al^{3+} in the left part of the brain of a patient using fluorescence microscopy and 1Mm lumogallion, ph has been equal to 7.4. based on their results, the concentration of Al^{3+} in the left part of the brain of the patient who has suffered alzimer disease ranges from 0.45µg/g dry wt.(in the hippocampus) to 1.75µg/g dry wt.(in the occipital lobe). In addition, orange fluorescence indicates that there is some aluminum in each tissue. Also, in their study, it is found out that if pure agarose is spread with Ca^{2+}, Cu^{2+}, Mg^{2+}, Fe^{3+} or Zn^{2+}, no fluorescence related to lumogallion is appeared. Moreover, Exley et al have found out that the rate of replication of aluminum concentration in brain tissue ranges from 0.01µg/g dry wt. to 5.58µg/g dry wt. [49]. Moreover, Leng et al have been used chromogenic agent with alizarin reds for determining the amount of trace aluminum in 2015. Moreover, the determination of trace aluminum with the agent has been carried out using ultraviolet spectrophotometer. Also, the effective parameters on the determination such as ph, temperature, and reaction time have been optimized. Their results demonstrates that Fe^{3+} and Cu^{2+} have effects on determining Al^{3+}, while K^{+} and Na^{+} have little influence on the mentioned approach [50-53].

In the procedure, an efficient and new approach based on graphene oxide nanoparticles (GONPs) which have been dispersed in ionic liquid (IL) has been used for in-vitro separation/extraction of trace Al from the blood of dialysis patients by ultrasound assisted-dispersive-micro solid phase extraction (USA-D-µSPE) procedure. Under the conditions which have been optimized, the linear range (LR), limit of detection (LOD), and preconcentration factor (PF) have been obtained 0.1–4.8 µg L^{-1}, 0.02 µg L^{-1}, and 25 for blood samples respectively (RSD<5%). The results of blood samples have demonstrated that the aluminum concentration after dialysis has

been higher than before dialysis (128.6±6.7 vs 31.8±1.6, P<0.05). The mean of blood aluminum has been significantly higher in dialysis patients in comparison with normal control respectively (113 5±7.12 vs 1.2±0.1). The developed approach based on GONPs/IL has been successfully used for extracting critical level aluminum from human blood, and the method is suggested for in-vivo extraction from human body of dialysis patients after being advocated on an appropriate surface with biocompatible materials within the human body. Some other approaches like atomic emission spectrometry preliminary essay to measure the amount of biological materials which have been carried out with existing analytical methods such as spark or flame atomic emission spectrometry 60-63 with sensitivities approximately 300-3000 less than ETAAS29 and before many of the contamination problems associated with sample collection and preparation were fully appreciated. These methods have now been largely abandoned but other sources for atomic emission spectrometry (AES) have proved successful. A constant-temperature graphite furnace and measured aluminum in blood and digested tissues with a detection limit around two- to fourfold better than ETAAS has been developed by Baxter et al. Instrumentation for electrothermal atomization atomic emission spectrometry has to be constructed by the user; however, some commercial inductively coupled plasma atomic emission spectrometry (ICP-AES) systems are available.

Allainss,M-66 has been used ICP-AES for measuring aluminum in serum, water, and dialysis fluids. Although he achieved excellent results, it is the experience of most workers that the sensitivity is insufficient to determine normal concentrations and that time consuming preconcentration steps, with risks of contamination, are necessary. In the other papers, the chemical speciation of aluminum in the low molecular mass (LMM) and high molecular mass (HMM) fractions of human serum has been discussed by Alfredo Sanz-Medel et al [32]**.** The methodologies, the experimental and instrumental requirements and the ability of the reported analytical procedures for identification of HMM and LMM aluminium species in human serum are tested in detail. Nonchromatographic separations coupled to electrothermal atomic absorption spectrometry for aluminum detection are compared with chromatographic techniques (size exclusion chromatography, anion exchange chromatography, and fast protein liquid chromatography) coupled to ETAAS or inductively coupled plasma mass spectrometry (ICP-MS) detection for Al-HMM species assessments. As stated before, the complexity of the human serum samples follows a knowledge and judicious choice of different principle based separations assisted by complementary selective detectors. In this vein, a most advisable first step is the fractionation of the aluminum biocompounds into two broad groups: (a) HMM and (b) LMM type of species. This 'primary' or 'rough information' can provide a constructive preliminary information. Thus, by using non-chromatographic approaches, it seems that about 10% of aluminum in human serum is ultrafiltrable [32-41]; therefore, about 90% of aluminum should be bound to non-ultrafiltrable HMM proteins. The query is now which protein(s) binds aluminum in human serum. In order to reply this query, chromatographic approaches coupled to Al^{3+} specific detectors are the most powerful analytical tools. However, at this stage, a new controversy arose on the type of chromatography to be applied: early workers in this field used size exclusion chromatographic approaches for separating human serum proteins [37,38-41]. The total concentration of Al^{3+} in human serum of healthy subjects which has been reported by Mothes et al [24] ranges from 0.5 to 8 mg dm^3, while a recent report from the Sanz-Medel's group [32] has indicated even lower normal aluminum concentration (in general below 0.35 mg dm^3) [24]. Due to such very low concentrations, the speciation of aluminum in healthy subjects has been possible only in spiked samples. Most of the investigators have used spiked serum in such a way that total serum aluminum, after spiking, ranged between 100 and 200 mg dm^3 matching high concentrations which could

be found in the serum of some dialysis patients. Since reported the concentrations of ultra-filterable aluminum in serum represented only 10/ 20% of total aluminum [41], it was necessary to apply very sensitive analytical procedures in order to identify and quantify the LMM-Al complexes present even in spiked serum and in high aluminum level sera of dialysis patients. Nowadays, there is no doubt that the analytical approaches for Al^{3+} speciation in human serum are needed to appropriately address the biomedical problems still waiting for a solution. The vitality of the research work on the development of new analytical methodologies for Al speciation in human serum is obvious from the number of published papers during the last decade. However, it is obvious that the speciation of Al^{3+} in biological fluids has been full of problems with difficulties in the past as is still in a state of development that has to overcome serious problems for its extensive application. Although earlier work seems to have been plugged with serious contamination problems, some sort of consensus on the chemical speciation of serum aluminum has emerged in recent years based on the results of some work carried out first by ultramicrofiltration, which demonstrates that usually 90% of total serum Al^{3+} is not ultra-filterable (i.e. the metal is bound to HMM bio compounds). Speciation of Al^{3+} in human serum is an extremely difficult task because the basal levels of this element in serum are lower than 2 mg/l and these minute amounts are fractioned in the speciation process. For making matters worse, the risk of significant exogenous Al contamination is very high.

In the other study, a reliable determination of aluminum in serum and aqueous solutions has been described by Moshtagi-Iie et al. In this method, using 10% HNO, for glassware and I mmol EDTA for plastic containers can prevent the problem of contamination since no delectable aluminum has been found by making a comparison between the absorption signals obtained from fresh sera and water samples with those obtained from samples held in the containers (data not shown). The temperature stages used led to the complete atomization of aluminium and produced a sensitivity and detection limit of 15 pg, and 2.1 $mg.L^{-1}$ respectively. Nameless atomic absorption (Perkin-Elmer 603 spectrophotometer) is used by Parkinson et al. Moreover, a sensitivity and detection limit of 35.5 pg and 2.3 $mg.L^{-1}$ have been presented by them. Our findings are in good agreement with their observations. Obviously, the sensitivity produced by our instrument has been much betters due to 10 the atomic absorption model which has been modern. In tile present method, the linearity of our calibration curve has been up 10.60 ng/mL of aluminum. With such a calibration curve, we were able to measure aluminum concentrations in serum, although the serum should be diluted in higher levels of aluminum. Mazzeo Farinaand Cerulli has been reported a linearity of up to 50 ng rnL^{-1} which has been in agreement with our findings. In the other study, it is found out by Halls et al that aluminum toxicity has been shown to be a problem for patients with renal failure on dialysis, leading, in severe cases, to dialysis dementia, bone disease, and anemias [35]. The measurement of aluminum in dialysate fluid can be used to monitor the exposure of patients on dialysis. The change in the concentration of aluminum in the fluid after dialysis can be used to calculate transfer of aluminum to and from a patient, and to follow the removal of aluminum with the chelating agent, desferrioxamine. Moreover, 5 dialysate fluids can be analyzed by electro-thermal atomic absorption spectrometry with electro-thermal atomization (ETAAS-ETA) in the same way as serum. The object of this work has been to develop a sensitive and accurate method based on ETAAS-ETA, which decline the analysis time in accordance with principles which have been described previously.

In the other methods, a modern chelating resin based on poly [4-(1-azo-3-hydroxy-4-(N,N-dicarbo dymethyl) aminophenyl) styrene] for determining traces level of aluminum and titanium have been proposed by Basargin et al. For using it in the solid phase extraction of aluminium, a polystyrene-co-divinylbenzene) commercial resin (Amberlite XAD-4) has been modified by grafting onto salicylic acid

by Bettinelli et al [36]. Also, a new chelating resin by fictionalisation of polystyrene–divinylbenzene with imidazole 4,5-dicarboxylic acid through N=N bonding for the speciation of vanadium (IV) and vanadium (V) have been synthesized. Deionized water is used for preparing all solutions. Otherwise, stated analytical-grade acids and other chemicals used in this study have been achieved from Merck, Darmstadt, Germany. Stock solutions of all metals, containing 1000 mg^{L-1} (Merck) have been used for preparation of the standards for the calibration curve. The calibration standards have never been submitted to the preconcentration procedure.

The XAD-1180-PV column approach has been tested with model solutions prior to the determination of aluminum in the samples. For the metal determinations, 50 ml of solution which contains 0.20 g of Al^{3+} has been added to 10 ml of buffer solution (the desired pH between 2 and 10). The column has been preconditioned by passing buffer solution. The solution has been allowed to flow through the column under gravity at the flow rate of 4 ml min^{-1}. After passing this solution ending, the column has been rinsed with twice 10 ml of water. The adsorbed metals on the column have been eluted with 5–10 ml portion of 2 M HCl. The eluent has been analyzed for determining the concentration of aluminum by graphite furnace atomic absorption spectrometer. The characteristics of XAD-1180-PV chelating resin were prepared. The thermogravimetric analysis curve of the XAD-1180- PV chelating resin is shown in three steps. In the first step, a mass loss of 23% up to 105 °C to be due to adsorbed water on the resin. In the second step, mass loss is 9.0% up to 340.0 °C. In the third step, mass loss is 34.0% up to 458 °C. The mass losses in the second and third steps are similar to pyrocatechol violet. There is an agreement between the situations and the previous studies [24–29]. When the infrared spectra Amberlite XAD-1180 and XAD-1180-PV resins have been compared with each other, there are additional bands at 1720, 1562, 1374, 1195, and 1120 cm^{-1} which seem to originate due to the modification of resin by the ligand. In addition, there are the characteristics of C=O, –N=N–, C–OH, –S–O–, and C–N vibrations respectively.

Moreover, determining trace aluminum in biological and water samples by cloud point extraction preconcentration and graphite furnace atomic absorption spectrometry detection have been studied by Hongbo Sang et al. In the practical application of surfactants in analytical chemistry, separation and preconcentration based on cloud point extraction (CPE) are becoming vital. The approach is based on the property of most non-ionic surfactants in aqueous solutions to form micelles and to separate into a surfactant-rich phase of a small volume and a diluted aqueous phase when heated to a temperature known as the cloud point temperature. The small volume of the surfactant-rich phase obtained with this methodology allows us to design the extraction schemes which are simple, cheap, highly efficient, speedy, and lower toxicity to the environment than those extractions that use organic solvents. Cloud point extraction has been used for separating and preconcentrating organic compounds as a step prior to their determination by liquid chromatography and capillary electrophoresis. The phase separation phenomenon has also been used for the extraction and preconcentration of metal ions after the formation of sparingly water-soluble complexes. By research, a TBS-990 atomic absorption spectrophotometer (Beijing Purkinge General Instrument Co. Ltd., Beijing, PR China) with a deuterium background correction and a GF990 graphite furnace atomizer system has been used for aluminum determination. An aluminum hollow-cathode lamp has been used as radiation source at 309.3 nm. For CPE, aliquots of 10 mL of a solution containing the analyte, Triton X-114 and PMBP buffered at a suitable pH have been kept in the thermostatic bath maintained at 40 °C for 20 min, and the surfactant-rich phase can settle through the aqueous phase. The phase separation could be occur faster by centrifuging for 5 min at 3000 rpm. After cooling in an ice bath, the surfactant-rich phase became viscous and was retained at the bottom of the tube. The aqueous phases can readily

be discarded simply by inverting the tubes. To decrease the viscosity of the extract and allow its pipetting, 200 L of 0.1 mol L^{-1} HNO_3 was added to the surfactant-rich phase. 20 L of the diluted extract was introduced into the GFAAS by manual injection. Calibration has been performed against aqueous standards which have been submitted to the same CPE procedure. Ashing and atomization curves have been established using 10 ng mL^{-1} Al^{3+} solutions which have been sent to CPE procedure and diluted with 10 mL of 0.1 mol L^{-1} HNO_3. In addition, 20 L of the diluted extract has been used for GFAAS analysis. The ashing and atomization curves of Al^{3+} without CPE procedure were also studied with 10 ng mL^{-1} Al^{3+} in 0.1 mol L^{-1} HNO_3. By using CPE procedure, the ashing temperature can be increased by 500 °C over the Al^{3+} solution without using CPE procedure, and the aluminum signal has been enhanced twice. There has been no difference in the shape of the atomization curve for aluminum with and without CPE procedure, only the values of absorbance have been different. In this work, the use of micelle systems as a separation and pre-concentration for aluminum offers some advantages including low cost, safety, preconcentration aluminum with high recoveries and very good extraction efficiency. The surfactant-rich phase can be easily introduced into the graphite furnace after dilution with 0.1 mol L^{-1} HNO_3, and directly determined by GFAAS. The suggested method can be applied to the determination of trace amount of aluminum in various real samples.

As another method, La^{3+} as releasing agent and ion suppressor in flame for determining metal ions has been used by Kılıçkaya Selvia. $LaPO_4$ has been used as co-precipitant for separation and pre-concentration of heavy metals in several water samples. Based on our study, $LaPO_4$ has been firstly used for separation and preconcentration of aluminum in human dialysis samples. This method has several advantages such as low detection limit (LOD), simple, rapid, economic, and precise. The recoveries of aluminum (III) in the presence of the most common matrix elements containing the alkaline and alkaline earth metals were good. A Perkin-Elmer Analyst A800 Model atomic absorption spectrometer (Northwalk, USA) with nitrous oxide/acetylene flame and a D2 lamp with background corrector was used throughout the determination of Al^{3+} in water solutions and human blood samples. For co-precipitation, 2 µg aluminum (III), 150 µg lanthanum (III), and 150 µL phosphoric acid (1:2 diluted water) have been placed in a centrifuge tube. Then the pH of the solution has been adapted to pH=5 with ammonium acetate/acetic acid, and the solution has been diluted to 50 mL with distilled water. After shaking the solution for several seconds, the solution has been allowed to stand for 15 min and centrifuged at 3500 rpm for 15 min. The supernatant has been removed and the precipitate in the tube was dissolved with 0.1 mL of concentrated HNO_3 and the volume was completed to 2 mL with distilled water. The number of five replicates for each analysis was used. The water/serum/blood samples were determined by flame atomic absorption spectrometry.

3. Results and Discussion

Based on some researches which have been carried out, it is demonstrated that the range of concentrations of aluminum next to industrial companies is about 0.4 to 8.0µg/m^3 [28-42, 50-53]. Moreover, aluminum concentration in drinking water ranges from less than < 0.001 to 1.029 mgL^{-1}[54]. Moreover, the amount of aluminum of milk of human breast is about 9.2 to 49µgL^{-1} [55-57]. The concentration of aluminum of soy-based infant formulas is higher in comparison with milk-based infant formulas or breast milk [57]. Moreover, the rate of Aluminum concentration in finished waters is high due to during the treatment of water, Al^{3+} is added to water [58]. In addition, it had better be mentioned that the amount of Al^{3+} in treated water is three times more than the water which has not been treated. Also, the changes of pH and the humic acid content of the water has effects on the rate of Al^{3+} concentrations which have been dissolved. Also, when pH is less than 5, the concentration of Al^{3+} increases. Unfortunately, aluminum particles have been spread in air, water, and foods, so by

inhaling air and having food and water, the rate of Al^{3+} increases in body tissues [59-62]. Moreover, using other consumer items such as antiperspirants, buffered aspirins, antiulereative medications, and antiacids causes an increase in the rate of Al^{3+} in human body. Also, by making a comparison between aluminum which there is in drinking water and food, and medicinal preparations which have Al^{3+} in themselves, the rate of Al^{3+} in medical preparations is much more. The intake or rate of Al^{3+} in food ranges from 3.4 to 9 mg/day [63-65]. The amount of Al^{3+} per tablet/capsule/5 ml dose in many antiacids is about 104 to 208 mg [66]. The vegetables and fruit trees which have been grown using treated water has received more Al^{3+} in themselves. It has been found out by Nayak in 2002 that a decrease or increase in Al^{3+} in human body does not have any effects on mortality (or mental health).

People who are living next to the aluminum companies, plants, and mines, as well as other hazardous waste sites will suffer chronic kidney failure. These people or patients must be treated with phosphate binders and long-term dialysis. The infants which have been fed soya, antiacids, and antidiarrheal can be exposed to high levels of aluminum. Based on TCRI (Toxic chemical release Inventory), the amount of Aluminum which have been released from 329 aluminum facilities to the environment is about 45.6 million pounds [67]. Moreover, total amount of aluminum oxide which has been released from 59 aluminum processing companies to air, water, and soil is about 2.9 million pounds [67].

Table 2-1 list amounts which have been released from these companies or facilities that they are grouped by state.

The data which have been obtained by TRI are

Table 1. Releases to the Environment from Facilities that Produce, Process, or Use Aluminum Oxide (fibrous forms)

[a] **Reported** amounts released in pounds per year[b] Total release									
State[c]	RF[d]	Air[e]	Water[f]	UI[g]	Land[h]	Other[i]	On-site[j]	Off-site[k]	Off-site
AL	2	0	0	0	0	0	0	0	0
AR	1	0	0	0	0	0	0	0	0
CA	1	0	0	0	0	0	0	0	0
CO	1	0	5	0	480	3	485	3	3
CT	1	0	0	0	0	0	0	0	0
GA	2	16	175	0	3	0	191	3	3
IA	2	0	0	0	40	0	0	40	40
IL	5	76	0	0	122	23	76	145	145
IN	3	901	250	0	5	10	1	10	1
KY	3	243	0	0	27	0	243	27	27
LA	2	0	0	0	0	0	0	0	0
MI	2	0	0	0	375	0	0	375	375

[a] The TRI data should be used with caution since only certain types of facilities are required to report. This is not an exhaustive list. Data are rounded to nearest whole number.

[b] Data in TRI are maximum amounts released by each facility.

[c] Post office state abbreviations are used.

[d] Number of reporting facilities.

[e] The sum of fugitive and point source releases are included in releases to air by a given facility.

[f] Surface water discharges, waste water treatment-(metals only), and publicly owned treatment works (POTWs) (metal and metal compounds).

[g] Class I wells, Class II-V wells, and underground injection.

[h] Resource Conservation and Recovery Act (RCRA) subtitle C landfills; other on-site landfills, land treatment, surface impoundments, other land disposal, other landfills.

[i] Storage only, solidification/stabilization (metals only), other off-site management, transfers to waste broker for disposal, unknown

[j] The sum of all releases of the chemical to air, land, water, and underground injection wells.

[k] Total amount of chemical transferred off-site, including to POTWs.

RF = reporting facilities; UI = underground injection

Source: TRI05 2007 (Data are from 2005)

not representative of the amount of Al^{3+} in every region due to TRI has selected a few facilities. In addition, inhaling and digesting Al^{3+} exacerbate renal failure, bone disease, and anemia. Moreover, dialysate fluids are made up (in human body) when aluminium which comes from water supplies is consumed or used. Unfortunately, human-mades have changed for ecosystemand increase the amount of aluminium in the environment. The element of Al^{3+} accumulate in plants and water, and thereby all herbivores are exposed to harmful effect of aluminium. Also, when a place is polluted with Al^{3+}, a decrease in the density of populations is occurred.

Emissions of a lot of Al^{3+} into water and soil decreases the fertility. Al^{3+} as a main factor in acid soil can limit crop productivity. The interaction of Al^{3+} with cell walls can cause the disruption of the membrane of plasma, and the disruption or interaction increases when oxidative damage and mitochondrial dysfunction occur. Also, Al^{3+} can damage DNA. When Al^{3+} accumulates in plants tissue, DNA starts to be ruined, and after a little time, it is observed that the rate of the growth of plants is decreased. In addition, all scientist who are study on the effects of environmental change on the plants are rather hesitant and in a dilemma over whether to adopt the effect of Al^{3+} on the disruption of DNA or not. After carrying out a lot of researches, it has been found out that the accumulation of Al^{3+} in tissue of plants cause that DNA with double strand starts to be broken. In addition aluminum toxicity depends on the acidity of soil and plant resistance. Clinical studies show that the patients who have high concentrations of metals in their brain, bone, and muscle have unexplained syndrome as dialysis dementia. Other researches demonstrated that there are anaemia and ectopic precipitation of calcium in aluminum toxicity syndrome. Here, some effects of aluminum on organ of human body are illustrated, as seen in Figures 2, 3, 4, and 5.

Although by removing dialysis fluid, the rate of Al^{3+} decreases, in the patients who have suffered renal failure, the tissue of their body, especially renal tissues absorbs. More Al^{3+} in contrast with others; therefor, in these patients, measuring the amount of Al^{3+} in the blood of the patient is indispensable.

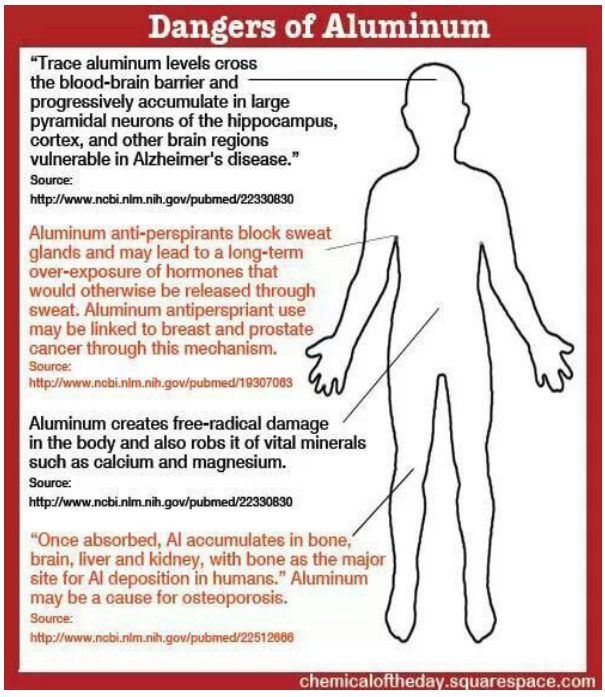

Fig. 2. Some disadvantages of aluminum.

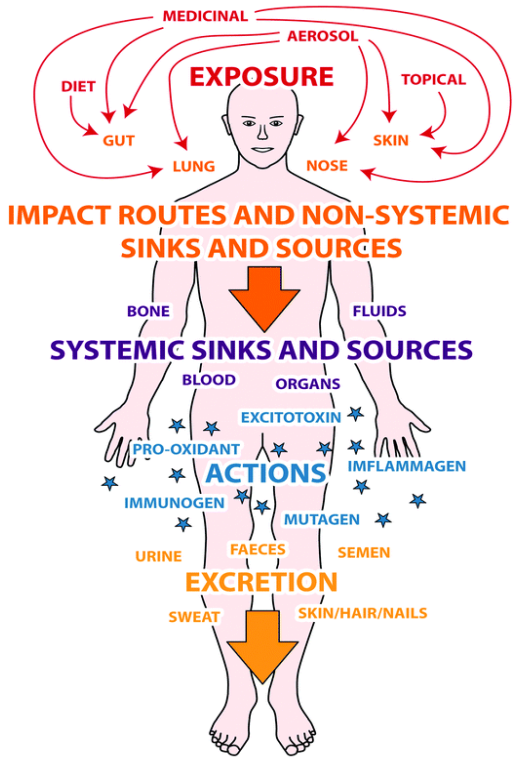

Fig. 3. Aluminum's exposure: A schematic which explores relationships between exposure, immediate targets mediating exposure, sinks and sources of biologically available aluminium with putative mechanisms of action and finally excretion of aluminium.

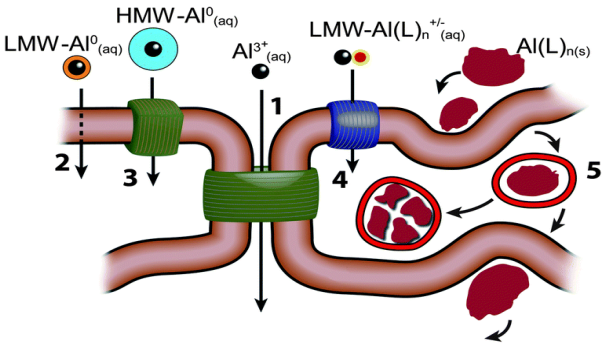

Fig. 4. There are 5 major routes by which aluminium could be transported across cell membranes or cell epi-/endothelia; (1) paracellular; (2) transcellular; (3) active transport; (4) channels; (5) adsorptive or receptor-mediated endocytosis. There are 5 major classes of forms of aluminium which could participate in these transport routes. These are shown in the figure as; the free solvated trivalent cation (Al^{3+}); low molecular weight, soluble complex ($LMW\text{-}Al^{0}_{(aq)}$).

The patients who have suffered chronic renal failure must be in intravenous therapy, and the rate of Al in their blood must be measured after each stage of removing dialysis fluid. Nowadays some researches about the relation of the amount of Al^{3+} and dementic mechanisms of intestinal absorption had better be carried out. Moreover there are a lot of analytical approaches for determining the amount of Al^{3+} in water, human body, and biological samples. Among the methods which have been used for measuring the amount of aluminum in water industry, colorimetric and fluorimetric are common (widespread) methods which have been used. A kinetic fluorimetric approach with a claimed limit of detection of 0.13j.Ig/L^{-1} using 1.0 ml of serum has been described by Iannou and piporaki. The results which have been obtained by flourimetric method is similar to the results which have been obtained by electrothermal atomic absorption spectrometry (ETAAS). For an analysis that more than 1.0 mL of serum is used, the method of conventional fluorophore, lumogallion which has been presented by Suzuki et al is suggested. In the mentioned methods which have been being used for measuring the amount of Al^{3+}, the precipitation of protein, as well making agents occurs. Moreover, in the two mentioned approaches, pH must be controlled carefully. Moreover, in colorimetric and fluorimetric approaches, cationic interferences can be overcome by masking agents. In addition, the two methods may be applied for analyzing serum, but the pros or benefits of the approaches are less than electrothermal atomic absorption spectrometry (ETAAS). In the methods, reagents and equipment which have been required are cheap. The methods

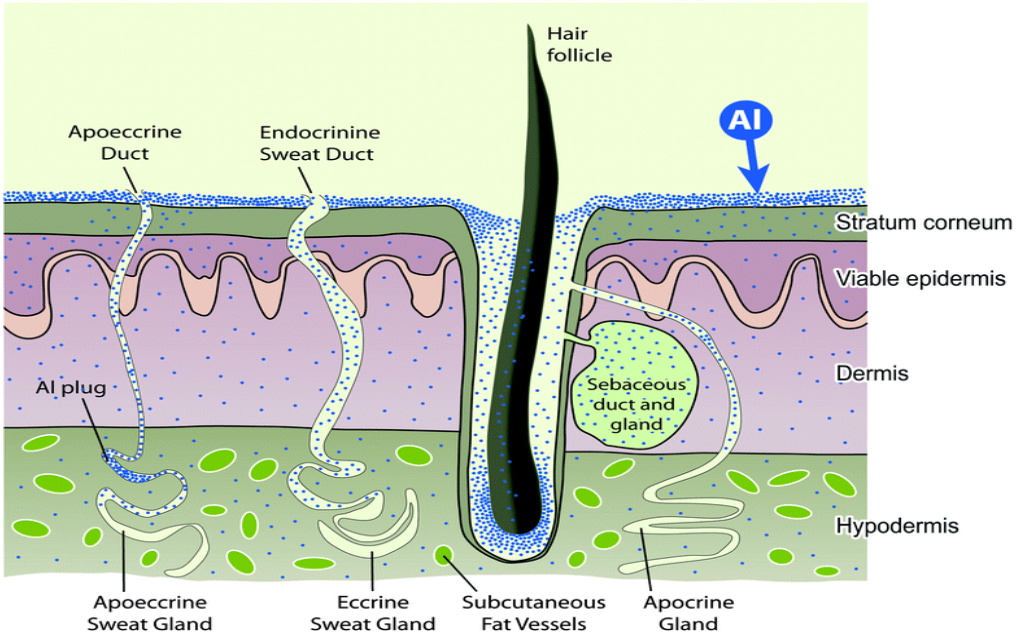

Fig. 5. The skin is a sink for topically applied aluminum and will act as a source of biologically reactive aluminum both to structures within the skin and to the systemic circulation.

of colorimetric and fluorimetric can be used for screening samples which have contamination in themselves. Also, the approaches are constructive for analyzing dialysis concentrates.

The procedure of chelation with eight-hydroxyquinoline when pH is equal to 6 and isobutyl methyl ketones is extracted into 10 ml has been suggested by Mazzeo and Lourenzyi for determining Al^{3+} in 200 ml of dialysis fluid concenter by FAAS. A detection limit of 30µg/l has been obtained. Moreover no interferences from the high salt content of the concentrates have been found. Also, after analyzing the samples which have been dissolved in acid and ashed at 800°C by FAAS, it has been found out that the migration of aluminum occurs at the pH which is equal to 2 while the storage is prolonged and temperature is increasing. Marcin Frankowski et al have used some approaches such as GF-AAS, ICP-AES, and ICP-MS to determine the amount of Al^{3+} in groundwater samples. Moreover, inorganic aluminum complexes have been modeled by them. Their studies have been focused on some ground water samples which have been selected from the Miocene aquifer of the city of Poznan, located in Poland. The amount of Al^{3+} in the aquifer is variable – from 0.0001 to 725µgL^{-1}. Three analytical methods, i.e. graphite furnace atomic absorption spectrometry (GF-AAS), inductively coupled plasma atomic emission spectrometry (ICP-MS), and Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) for measuring the amount of aluminum in the groundwater have been used. The results which have been obtained from analytical methods have been have been used to determine the trend of groundwater from the Mesozoic aquifer to the Miocene aquifer. Distribution of Al^{3+} has been modeled by Frankowski et al in 2011. After modeling, the existence of aluminum hydroxyl complexes in some parts of the groundwater has been confirmed [68]. In addition, based on the study which has been carried out by Frankowski et al in 2011, in spite of the fact that sulphates and organic matter in the most of groundwater samples are dominant, the aluminum complexes have never participated in the reaction with the ligands (based on the modelling) [68]. Also, the change of the amount of aluminum concentration in groundwater aquifers due to aluminum's amphoteric property causes that founa and flora will be ruined. Moreover, the low concentration of aluminum in groundwater aquifer are obtained when the transformations of aluminosilicates occur in the active water exchange zone. Soluble complex bonds with dissolved fluoride (AlF^{2+}, F_2^+, AlF_3^0, AlF_4^-), Sulphate ($AlSO_4^+$, $Al(SO_4)^{2-}$), phosphate ($AlHPO_4^{2+}$, $AlHPO_4^+$) ligands.

With low – molecular organic acids are the major sources of aluminum in groundwater. In most aquifers, based on their studies, Al^{3+} and hydroxide complexes as exchangeable aluminum fractions are the main sources of aluminum [68]. Moreover, the penetration of Al^{3+}, $AlOH^{2+}$, and $Al(OH)_2^+$into the agricultural products causes toxicity to humans. Based on the research which has been carried out by Frankowski et al in 2011, the high concentrations of aluminum in groundwater aquifers demonstrate that hydroxide complexes and organic complexes are dominant in the aquifers [68]. The concentration of trace aluminum in groundwater, surface water, the river have been usually determined by using GF-ASS (graphite furnace Atomic Absorption spectrometry). In addition, for measuring the amount of aluminum in limed lakes, forest soil waters, and springs, using inductively coupled plasma mass spectrometry (ICP-MS) is suggested. Also, for determining the amount of aluminum in drinking water, inductively coupled plasma Optical Emission spectrometry (ISP – OES) has been used. Based on some researches, inductively coupled plasma mass spectrometry method is not constructive for determining the amount of aluminum in water due to the interferences which have been caused by other elements in water samples.

4. Conclusions

In this research paper, the importance of measuring the amount of aluminum complexes in the nature (soil and water) and human bodies has been paid

attention to. Also, some researchers which have been carried out have been selected and assessed. All researches have tried to present the best analytical methods which are more accurate and precise for determining the amount of aluminum in water, soil, and biological samples. From 1985 to 2018, the limit of detection has become lower, and limit of quantification has extended. Nowadays, the approaches which have been used are more precise, time-consuming, cost-effective, and applicable. Also. At the present time, nano-absorbents are used for separation of Al^{3+} from blood of human tissues, water, soil, and plants' tissues. Between 2016 and 2017, flame atomic absorption spectrometry has been used to determine the amount of aluminum in tricalcium phosphate anhydrous powder which contains about 350mgKg^{-1} aluminum in itself. From 2011 up to now, for determining the amount of Al^{3+} in some top and well water samples, in some areas of Iran, surfactant cetyltrimethylammonium bromide and. The method of cloud point extraction have been used with each other. From 2013 to 2019, for quantifying the amount of Al^{3+} in waters and soft drinks of the country of Thailand, spectrophotometric approach using eriochrom cyanine has been used. Also, in this method, the limit of detection is less than 0.0008 and the limit of detection is about 0.0125 mg L^{-1}. From, 1982 up to now, for quantifying the amount of aluminum in human blood, serum, urine, and tissues, in some European hospital, using electro-thermal atomic absorption spectrometry has been suggested. In the decade of 1990, in the hospital of USA, for determining the amount of aluminum in blood, the method of diluting plasma samples with HNO_3/Triton X-100, matrix modifier fourfold was used. Moreover, for measuring the amount of aluminum in the patients who have suffered less renal failure, or their renal functions are normal, diluting samples with an equal volume of Mg $(NO_3)_2$ matrix modifier and atomizing the samples from a L'vov platform were usual methods. Also, based on the studies which have been carried out from 2009 to 2019 about the determination of aluminum in groundwater aquifers, in most parts of Eurasia and USA, the concentration of trace aluminum in groundwater, surface water, and river have been usually quantified by using GF-ASS (graphite furnace atomic absorption spectrometry); moreover, for measuring the amount of aluminum in limed lakes, forest soil waters, and springs, using inductively coupled plasma mass spectrometry (ICP-MS) has rarely been suggested. In addition, for determining the amount of aluminum in drinking water, inductively coupled plasma optical emission spectrometry (ICP-OES) has been used. Also, since 2017 to 2019, in some groundwater aquifers of London, chemometric methods using optimization algorithms have been common among a lot of researchers, scientist, and hydrogeologists for determining the amount of aluminum. Furthermore, based on most researches, when pH is more than 7.0, the solubility of aluminum increases, and then water is polluted. Afterward, lot of people will suffer renal failure or chronic renal failure.

5. Nomenclatures

CNS: Central Nervous System
CPE: Cloud Point Extraction
ETAAS: Electrothermal Atomic Absorption Spectrometry
GONPs: Graphene Oxide Nanoparticles
GF-ASS: Graphite Furnace Atomic Absorption Spectrometry
IL: Ionic liquid
ICP-MS: Inductively Coupled Plasma-Mass Spectrometry
ICP-OES: Inductively Coupled Plasma Optical Emission Spectrometry
LR: linear range
LOD: limit of detection
PF: Preconcentration Factor

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