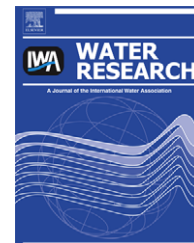


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Dispersion of C₆₀ in natural water and removal by conventional drinking water treatment processes

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ABSTRACT

The first objective of this study is to examine the fate of C₆₀ under two disposal scenarios through which pristine C₆₀ is introduced to water containing natural organic matter (NOM). A method based on liquid–liquid extraction and HPLC to quantify nC₆₀ in water containing NOM was also developed. When pristine C₆₀ was added to water either in the form of dry C₆₀ or in organic solvent, it formed water stable aggregates with characteristics similar to nC₆₀ prepared by other methods reported in the literature. The second objective of this study is to examine the fate of the nC₆₀ in water treatment processes, which are the first line of defense against ingestion from potable water – a potential route for direct human consumption. Results obtained from jar tests suggested that these colloidal aggregates of C₆₀ were efficiently removed by a series of alum coagulation, flocculation, sedimentation and filtration processes, while the efficiency of removal dependent on various parameters such as pH, alkalinity, NOM contents and coagulant dosage. Colloidal aggregates of functionalized C₆₀ could be well removed by the conventional water treatment processes but with lesser efficiency compared to those made of pristine C₆₀.

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1. Introduction

Carbon fullerenes such as C₆₀ have been at the center of the recent prosperity in nanoscale science and engineering. C₆₀ consists of 60 carbon atoms arranged in 20 hexagons and 12 pentagons that form a perfectly symmetrical cage structure, approximately 1 nm in size. With increasing commercial interest in its unique chemical and physical properties, the manufacture and use of fullerenes are expected to grow rapidly over the next decade (Ball, 2001; Colvin, 2003; Wiesner et al., 2006). However, as with many other engineered nanomaterials, information required to accurately assess the influence of fullerenes on the natural environment and

human health is rather scarce. Such assessment requires major research efforts on multi-disciplinary, multi-scale subjects, including the physical and chemical status of C₆₀ in the environmental media to which it is released, transport behaviors in both small and large scales, potential physical, biological and chemical transformations in both natural and engineered systems, and the ultimate interaction of pristine or transformed fullerene with various biological receptors.

Generally, C₆₀ has not been considered a potential contaminant in aquatic systems since it is extremely hydrophobic and virtually non-wettable. However, recent findings have suggested that, upon release to water, C₆₀ forms stable, nanoscale colloidal aggregates (commonly referred to as

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nano- C_{60} or nC_{60}) of which the dispersion status is affected by natural water constituents (Chen and Elimelech, 2007; Deguchi et al., 2001; Fortner et al., 2005). Consequently, numerous studies have been conducted or are currently underway to assess the fate and transport characteristics of nC_{60} in natural and engineered environments, including chemical transformation in engineered oxidation processes (Fortner et al., 2007) and transport behavior in natural subsurface environments (Chen and Elimelech, 2007). Several recent studies (Lyon et al., 2006; Sayes et al., 2004) have also suggested that nC_{60} interacts with numerous types of cells (including human cells), potentially causing toxic effects.

These aggregate forms of C_{60} can be prepared using various laboratory procedures. In the solvent exchange method, C_{60} is first dissolved in a water miscible, polar organic solvent, such as tetrahydrofuran, and mixed with water. The transfer organic solvent is subsequently removed via distillation (Deguchi et al., 2001). This method has been widely applied to test the toxicity and reactivity of nC_{60} due to the high yield and control of aggregate size. Alternatively, nC_{60} can be formed by dissolving C_{60} in a water immiscible organic solvent such as toluene and applying ultrasound to gradually transfer C_{60} into water as the organic solvent is evaporated (Lee et al., 2007). Although the above widely used methods are relatively convenient and useful for the laboratory studies, they do not represent the most likely scenarios of C_{60} release to natural aqueous environments.

The first objective of this study is to examine the fate of C_{60} under two disposal and spillage scenarios through which pristine C_{60} is introduced to water containing natural organic matter (NOM): (1) introduction of dry phase C_{60} to the aqueous phase, and (2) contact of water immiscible organic solvent containing C_{60} (i.e., used for storage purposes) with the aqueous phase and subsequent inter-phase transfer of C_{60} from organic phase to the aqueous phase, both under simple and prolonged mixing (without application of unrealistic external factors, such as widely used sonication). Additionally, a method to quantify nC_{60} in water containing NOM was developed.

The second objective of this study is to examine the fate of these nC_{60} particles in water treatment processes, as these are the first line of defense against ingestion from potable water consumption – a potential route for direct human consumption. Jar tests were performed to evaluate the removal of nC_{60} by a conventional water treatment process, which consists of a suite of alum coagulation, flocculation, sedimentation and granular media filtration steps to remove particulate contaminants. The effects of NOM and water quality parameters such as pH and alkalinity on the removal of nC_{60} were evaluated.

2. Material and methods

2.1. Materials

C_{60} of over 99% purity was obtained from the MER Corporation (Tucson, AZ). The ACS grade (>99% purity) sodium dodecylsulfate (SDS, $CH_3(CH_2)_{11}OSO_3Na$) was purchased from Aldrich Chemical Company (Milwaukee, WI). The Suwannee

River NOM (SRNOM) stock solution was prepared by mixing a known amount of SRNOM (International Humic Substances Society, St. Paul, MN) with ultrapure water for 24 h. Dissolution of SRNOM was facilitated by adding 1 N potassium hydroxide (KOH) to increase the solution pH to 7.0. The solution pH was then lowered to the original value by adding 1 N nitric acid (HNO_3) and the concentration was measured using a total organic carbon (TOC) analyzer (TOC-Vw, Shimadzu, Columbia, MD). This stock solution was diluted to prepare solutions of target carbon concentrations (5 mg-C/L and 50 mg-C/L). Ultrapure water produced by a Milli-Q water filtration system (Millipore, Billerica, MA) was used for the preparation of all solutions.

2.2. nC_{60} preparation methods

Two different methods were used to prepare nC_{60} : single phase mixing and two phase mixing. In the single phase mixing procedure, 5 mg of solid C_{60} was added to 100 mL of either ultrapure water, 5 mg-C/L SRNOM solution, 50 mg-C/L SRNOM solution, or 1% SDS solution (50 mg C_{60} /L in each solution), respectively, and the suspensions were then vigorously mixed for 4 weeks using a magnetic stirrer. After 4 days of quiescent settling, the supernatant (around 60% of total volume) was filtered with a 10 μ m nominal pore size filter (Model 453, VWR, Suwannee, GA) and further experiments were performed using the filtrate. For the two phase mixing procedure, a solution of 500 mg/L C_{60} in toluene (Aldrich Chemical Company, Milwaukee, WI) was prepared by mixing dry C_{60} in toluene for 6 h. A 20 mL aliquot of each fullerene solution in the organic solvent was added to a 125 mL Erlenmeyer flask containing 100 mL of various aqueous solutions (ultrapure water, 5 mg-C/L SRNOM solution, 50 mg-C/L SRNOM solution, and 1% SDS solution) and agitated for 1 week. After 3 days of quiescent settling and phase separation, the aqueous phase was carefully taken and purged with 99.999% purity N_2 (NI UHP300, Airgas Inc., Radnor, PA) for 3 h to remove residual toluene.

2.3. Jar test procedure

Jar tests were performed with 150 mL of solution transferred to 250 mL beakers using a Phipps and Bird Model 7790-400 Jar Tester (Richmond, VA). The total alkalinity of each solution was adjusted before the test using 1 M $NaHCO_3$ solution. The solution pH was adjusted by adding an adequate amount of 1 M NaOH and 1 M H_2SO_4 . After adding a predetermined amount of alum ($Al_2(SO_4)_3 \cdot 18H_2O$) (Aldrich, Milwaukee, WI) from 10 g/L stock, the solution in the jar was mixed at a paddle speed of 200 rpm for 2 min, followed by 30 min slow mixing at 25 rpm. After 1 h of quiescent settling, the supernatant was removed for further analyses. Filterability of the flocs formed from the jar test by a typical granular media filtration was estimated by filtering the supernatant with a Whatman Grade No. 40 Quantitative Filter (Florham, NJ) (Crittenden, 2005).

2.4. Analytical methods

The size of nC_{60} was analyzed by dynamic light scattering (DLS) using a Zetasizer ZS90 (Malvern Instruments,

Worcestershire, UK). Zeta potential was analyzed by a Zeta Plus Zeta Potential Analyzer (Brookhaven Instruments Co., Holtsville, NY). Electron micrographs were obtained by a Philips 120 transmission electron microscope (TEM) (New York, NY). TEM specimens were prepared by placing a droplet of fullerene suspension on a copper carbon grid (Electron Microscopy Science, Hatfield, PA) and drying overnight at room temperature. The concentration of nC_{60} in organic free water was determined by UV absorption at 347 nm (Fortner et al., 2005) using an Agilent 8453 UV-vis Spectroscopy System (Palo Alto, CA). In the presence of NOM, nC_{60} in the aqueous phase was first extracted to toluene (liquid-liquid extraction, LLE), following the procedure from the previous studies (Isaacson et al., 2007; Santa et al., 1995; Xia et al., 2006), with a minor modification discussed below. The concentration of C_{60} in the toluene phase was analyzed using an Agilent 1100 series High Performance Liquid Chromatography (HPLC) system equipped with an Agilent Zorbax 4.6 \times 150 mm XDB-C8 column and a diode-array detector at 333 nm wavelength. The HPLC was operated using a mixture of 40% acetonitrile and 60% toluene as an eluent, at a flow rate of 1 mL/min and sample injection volume of 0.1 mL. The method detection limit of HPLC analysis was 0.25 mg/L and all the extractions and HPLC analyses were duplicated.

3. Results and discussion

3.1. Dispersion of C_{60} in aqueous phase

Solid C_{60} mixed with all the aqueous phases (i.e., ultrapure water, 5 mg-C/L SRNOM solution, 50 mg-C/L SRNOM solution, and 1% SDS solution) formed stable suspensions (Fig. 1a).

Characteristics of the C_{60} suspension were consistent with those of the nC_{60} reported in the literature (Deguchi et al., 2001; Fortner et al., 2005; Lyon et al., 2006). They showed characteristic orange–yellow color, while the intensity of the color varied depending on the solution composition. TEM analysis suggests that most C_{60} aggregates existed as oval or circular shapes in cross section and were surrounded by SRNOM (Fig. 2a), while the agglomerates of SRNOM and the nC_{60} could have formed during the sample drying process. Average diameters of nC_{60} measured by DLS ranged from approximately 380 nm (in 50 mg-C/L SRNOM solution) to 580 nm (in 5 mg-C/L SRNOM solution) and matched well with the observations by the TEM analysis.

Fig. 3a shows UV-vis spectra of nC_{60} suspensions prepared by the single phase mixing method after subtracting the spectra of a solution containing only an equal amount of SRNOM. The UV-vis absorbance of all the nC_{60} suspensions shows two characteristic absorption peaks of solvated C_{60} , which are approximately located between 330 and 360 nm (Fortner et al., 2005; Lyon et al., 2006) (Fig. 3a). Peaks were red-shifted compared to C_{60} molecularly solubilized in the toluene phase, since C_{60} was present in solid forms (Fortner et al., 2005). Broad band absorption at 400–550 nm also indicates the presence of aggregated forms of C_{60} . However, these spectra were considered inappropriate for the quantification of nC_{60} in these aqueous solutions, since a portion of SRNOM which adsorbed onto nC_{60} might contribute differently to the overall absorbance than SRNOM freely solubilized in the aqueous phase. In addition, baseline increase over the entire wavelength, which was not related to the light absorption by nC_{60} , was apparent in these spectra. During nC_{60} preparation by the single phase mixing protocol, portions of black C_{60} powders might not have rearranged into crystalline nC_{60} and remained

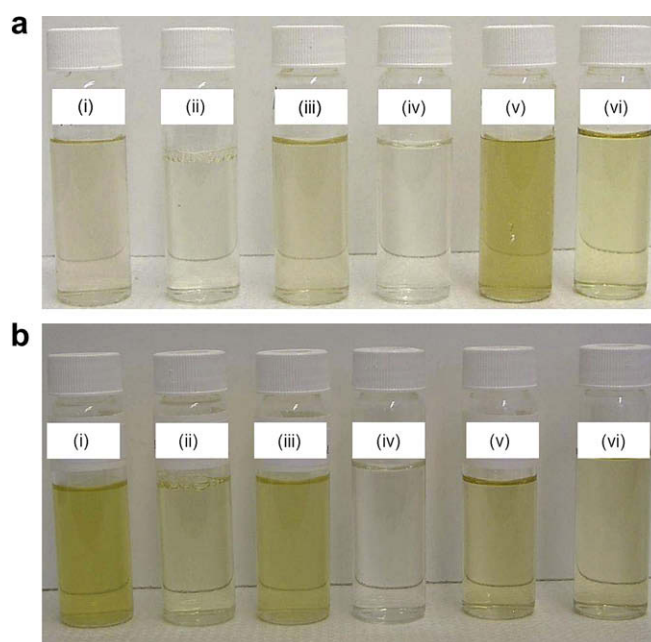


Fig. 1 – Visual examination of C_{60} in (i) Milli-Q water, (ii) 1% SDS solution, (iii) 5 mg-C/L SRNOM solution, and (v) 50 mg-C/L SRNOM solution prepared by (a) single phase mixing and (b) two phase mixing. 5 mg-C/L and 50-mg-C/L SRNOM solutions without C_{60} are also shown in (iv) and (vi).

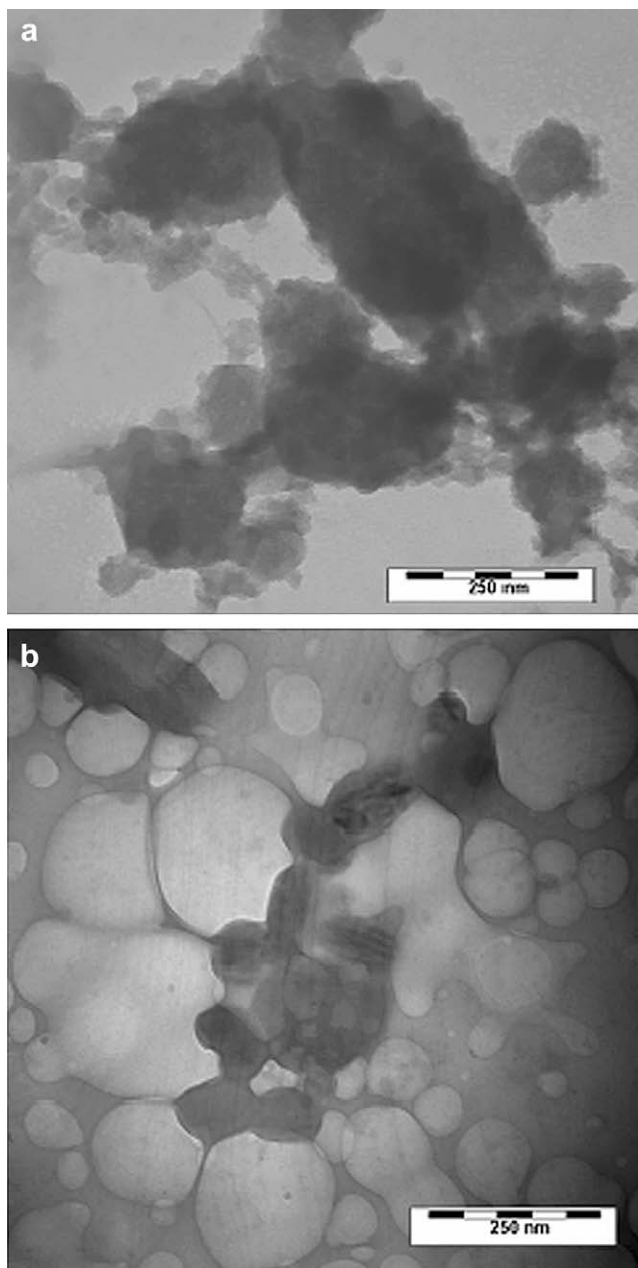


Fig. 2 – TEM images of fullerenes stabilized in SRNOM solutions produced by (a) single phase mixing and (b) two phase mixing.

suspended as black carbon in the solution. They were presumably further stabilized through the interaction with SRNOM and passed through the 10 μm filters. It is likely that these black carbon powders (*e.g.*, C_{60} that did not form crystalline aggregates) scattered incident light and resulted in transmittance loss over the entire wavelength, even though the existence of this form of C_{60} could not be confirmed using the current techniques. Rough estimation of absorption intensity at 360 nm by subtracting the baseline absorbance at 800 nm from the original spectrum, suggests that the absorption at 360 nm of the C_{60} suspensions in the 50 mg-C/L SRNOM solution (≈ 0.156) was higher than that in the

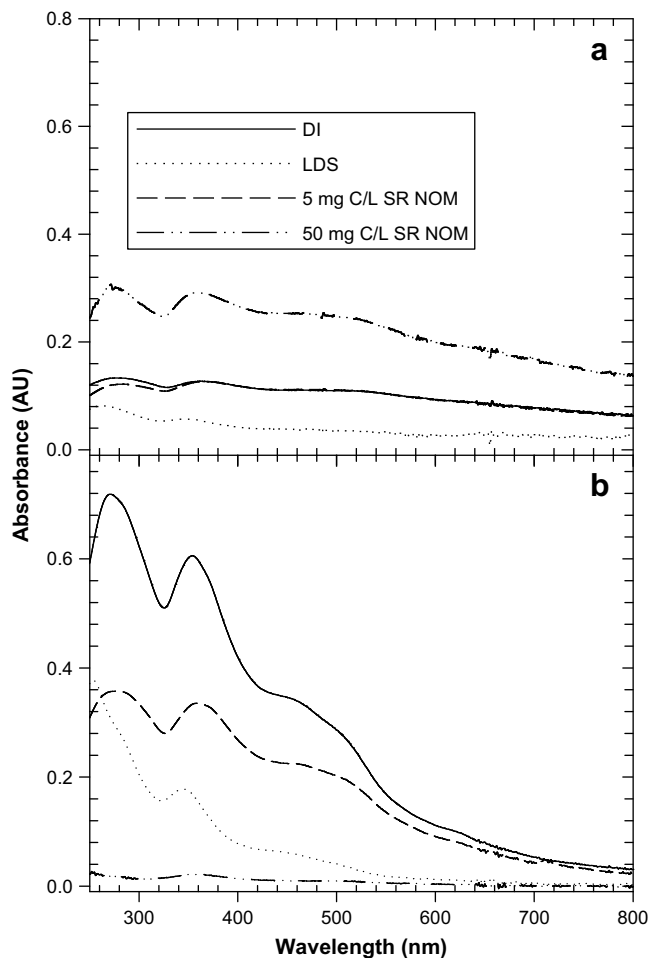


Fig. 3 – UV-vis spectra of C_{60} suspensions prepared by (a) single phase mixing and (b) two phase mixing. Spectra of C_{60} in NOM solutions were obtained after subtracting the spectra of SRNOM only solution from the original spectra.

ultrapure water (≈ 0.064). This suggests that SRNOM might improve the dispersion of C_{60} in the aqueous phase, probably due to the enhanced steric stabilization and charge repulsion provided by the adsorbed SRNOM.

A suspension of $n\text{C}_{60}$ which was stable for over several months also formed when the organic phase containing C_{60} was placed in contact with the aqueous phase. As C_{60} transferred from the organic phase into the aqueous phase, the aqueous phases gradually acquired characteristic orange-yellow color of the $n\text{C}_{60}$ suspension (Fig. 1). TEM images (Fig. 2b) showed circular or oval shaped C_{60} aggregates surrounded or connected by NOM. Consistent with the TEM observation, the diameter of $n\text{C}_{60}$ prepared by two phase mixing was smaller than that produced by single phase mixing according to the DLS analysis. The average diameters of the $n\text{C}_{60}$ in these samples ranged from around 120 to 250 nm.

All the C_{60} suspensions prepared by two phase mixing showed red-shifted UV-vis absorption peaks at 360 nm as well as broad band absorption between 400 and 550 nm (Fig. 3b), confirming the existence of $n\text{C}_{60}$. It was noted that the intensity of UV-vis absorbance at 360 nm (after subtracting the absorbance of the background SRNOM solution) decreased

significantly as the SRNOM concentration increased. This result appeared contrary to single phase mixing in which SRNOM enhanced the dispersion of C_{60} in the aqueous phase. It is postulated that when C_{60} transfers from the organic phase to the aqueous phase, the SRNOM accumulating at the interface of two solvents might hinder inter-phase transport of C_{60} . Likewise, SDS also appeared to retard the transport of C_{60} from the organic phase to the aqueous phase, compared to pure water, but to a much lesser extent than SRNOM when the same carbon mass basis was compared (50 mg-C/L SRNOM versus approximately 5000 mg-C/L SDS).

3.2. Analysis of nC_{60} in water containing NOM

The characteristic UV absorption of nC_{60} at around 340–360 nm has been widely used to quantify the concentration of nC_{60} in the aqueous phase. When the NOM in the aqueous phase interferes in this wavelength, however, such a direct spectral analysis is not possible. Therefore, an LLE-HPLC method was developed to quantitatively analyze nC_{60} prepared from the two phase mixing protocol as a representative nC_{60} . The method was modified from past studies (Isaacson et al., 2007; Xia et al., 2006) with the addition of salts, acids, and/or oxidants to facilitate the extraction process. The extraction kinetics of 5 mg/L C_{60} from 2.5 mL of water containing 5 mg-C/L SRNOM and 10 mM extraction agent, to 2.5 mL of toluene are shown in Fig. 4. Three different representative extraction agents including a salt (KCl), a mild oxidant ($Mg(ClO_4)_2$), and an acid (glacial acetic acid (GAA), CH_3COOH) were used. After shaking a capped test tube with a rotary shaker at 200 rpm, two phases were gravimetrically separated for 1 h. Then, 0.5 mL of toluene in the top layer was carefully taken for the HPLC analysis. Among these three chemicals, $Mg(ClO_4)_2$ showed the fastest extraction kinetics with recovery reaching nearly 100% in less than 2 h. With KCl, the recovery quickly reached a plateau value of approximately 10%. The GAA showed relatively slow

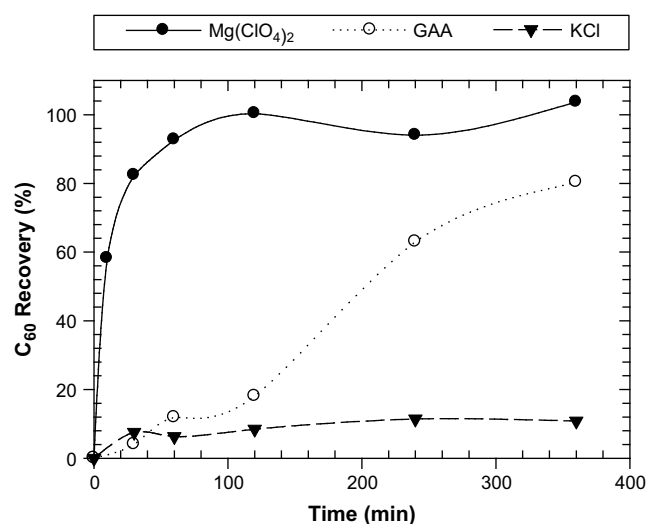


Fig. 4 – Extraction kinetics of C_{60} obtained using different extraction agents (extraction agent concentration = 10 mM, initial C_{60} concentration = 5 mg/L, NOM concentration = 5 mg-C/L).

extraction kinetics but the recovery gradually increased up to around 80% after 6 h of extraction.

The effect of the extraction agent concentration on C_{60} recovery after 6 h of agitation is shown in Fig. 5a. The $Mg(ClO_4)_2$ showed ~100% C_{60} recovery even at low concentrations (5–10 mM), but the recovery gradually decreased to less than 30% as the concentration was increased to 100 mM. At high $Mg(ClO_4)_2$ concentration, the formation of an emulsion at the interface was visibly observed. This is known to occur in the presence of large amounts of proteins, lipids, and surfactants found in natural samples (Xia et al., 2006). Such emulsions might hinder the inter-phase transport of C_{60} from the aqueous phase to the toluene phase. In contrast, C_{60} recovery increased as concentrations of KCl and GAA increased and reached almost 100% when KCl and GAA concentrations were greater than 50 mM. Moreover, for KCl and GAA, no emulsion formation was observed in the range of concentrations investigated.

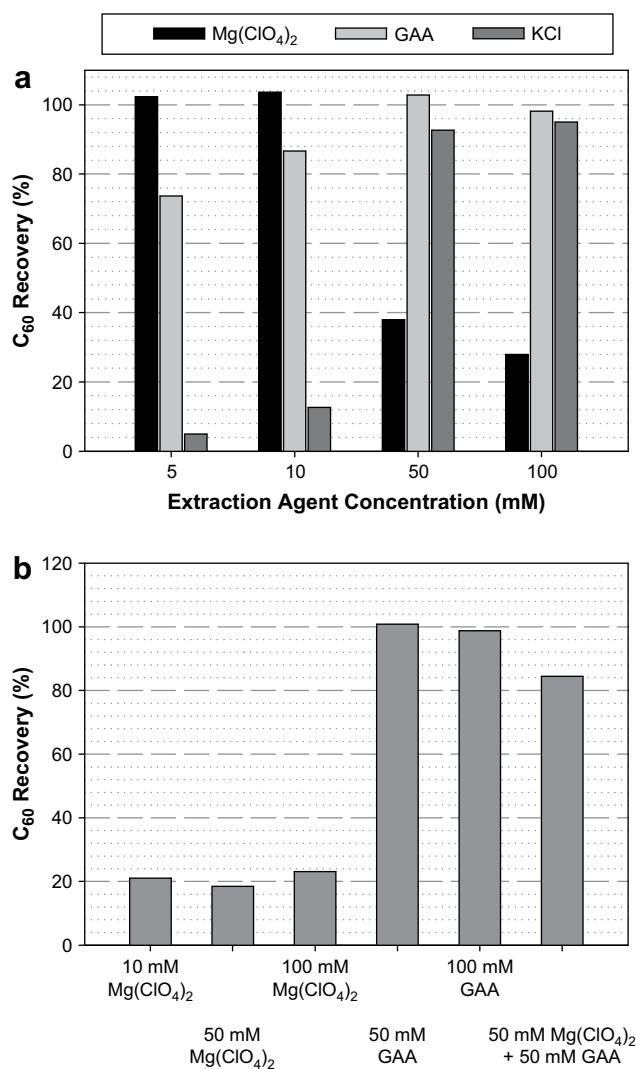


Fig. 5 – Effect of the extraction agent concentration on the C_{60} recovery: (a) without alum and (b) with 25 mg/L alum (extraction time = 6 h, initial C_{60} concentration = 5 mg/L, NOM concentration = 5 mg-C/L).

As the later part of the study involves the use of alum as a coagulant, the effect of alum on the extraction was also examined. The presence of alum coagulant further aggravated the emulsion formation in the case of $\text{Mg}(\text{ClO}_4)_2$. When 25 mg/L alum was added to the solution containing 5 mg-C/L SRNOM, the C_{60} recovery decreased to 20% due to emulsion formation and presumable mass transfer hindrance at the interface, even with a low concentration of $\text{Mg}(\text{ClO}_4)_2$ at 10 mM (Fig. 7). A past study reported that GAA could suppress emulsion formation if added at the same time with $\text{Mg}(\text{ClO}_4)_2$ during C_{60} extraction (Xia et al., 2006). However, when a mixture of 50 mM GAA and 50 mM $\text{Mg}(\text{ClO}_4)_2$ was added to the solution containing alum and SRNOM, the recovery still remained relatively low at 85%. Therefore, even though $\text{Mg}(\text{ClO}_4)_2$ has been frequently used to destabilize nC_{60} in organic free water (Santa et al., 1995), it appears to be a poor choice when the water contains both SRNOM and alum coagulant. In contrast, the recovery by GAA was not influenced by the presence of alum, although a slight decrease was observed with the GAA dose of 100 mM. Based on the above experimental results, 100 mM of GAA was used to extract C_{60} from the aqueous phase, containing both SRNOM and alum coagulant, into toluene in the subsequent work.

3.3. nC_{60} removal by conventional water treatment processes

Jar tests were performed using nC_{60} prepared from the two phase mixing protocol as a representative nC_{60} . Experimental results shown in Fig. 6 suggest that nC_{60} can be effectively removed by a suite of coagulation, flocculation and sedimentation processes, although the removal was largely dependent on water quality parameters. When the total alkalinity of the feed water was 100 mg/L as CaCO_3 , more than 60% of C_{60} removal was achieved for pH range of 6–8 with all coagulant dosages, except the lowest dose of 10 mg/L (Fig. 6a). Removal reached its peak at a pH between 7 and 8 in all cases. In particular, when the alum dosage was 100 mg/L, more than 60% of the C_{60} removal was achieved throughout the entire pH range investigated; and the removal reached up to 97% at a pH between 7 and 8. However, at lower alkalinity (50 mg/L as CaCO_3), nC_{60} was removed in a limited pH range (7–8) when alum dose was less than 50 mg/L (Fig. 6b). When the dose was 100 mg/L, C_{60} removal reached up to 93% only at a pH near 7, and drastically decreased as the pH deviated from this value. The removal of nC_{60} assessed in our study matched well with sweep coagulation zone of the coagulation diagram developed by Amirtharajah and Mills (1982), suggesting that the sweep coagulation might be the dominant mechanism of nC_{60} removal. Considering these experimental results, and the fact that a typical dosage of alum in practice ranges from 10 to 150 mg/L (Crittenden, 2005), nC_{60} is expected to be effectively removed by most of the conventional particle destabilization water treatment processes.

The filterability of nC_{60} flocs in the supernatant was estimated by filtering the supernatant with a Whatman Grade 40 Quantitative Filter. These filters are widely used for the simulation of a rapid granular media filtration in conventional water treatment processes (Crittenden, 2005). The filterability of nC_{60} flocs suspended in the supernatant after

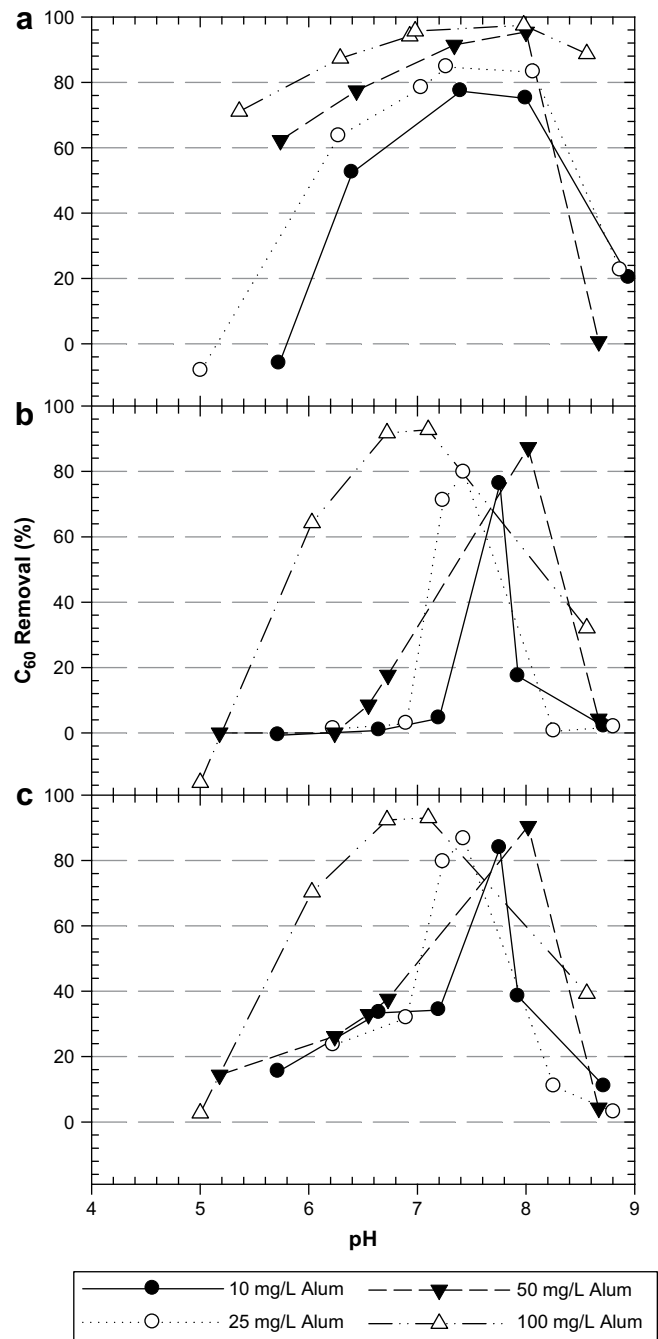


Fig. 6 – Removal of 1 mg/L nC_{60} during jar test results at: (a) alkalinity of 100 mg/L as CaCO_3 , and (b) alkalinity of 50 mg/L as CaCO_3 . (c) The removal after filtration at alkalinity of 50 mg/L as CaCO_3 .

sedimentation is presented in Fig. 6c. An approximate 20% increase of C_{60} removal was generally observed after filtration, when nC_{60} removal by the previous coagulation step was less than 20%. However, when nC_{60} removal by coagulation was already high (more than 60%), additional removal by filtration was minimal.

The presence of SRNOM dramatically changed the removal of nC_{60} during the coagulation/flocculation/sedimentation processes. Experiments were performed using the same alum

dose of 25 mg/L but with different SRNOM concentrations. Compared to the results obtained in the absence of SRNOM (Fig. 6), the range of pH that exhibited maximum removal shifted toward lower values as SRNOM concentration increased (Fig. 7). In addition, the range with greater than 80% removal became narrower as well. The observed decrease in the removal of nC₆₀ in the presence of NOM might be due to the preferential interaction between NOM and the coagulant. For instance, O'Melia et al. (1999) suggested that metal coagulants preferentially bind with NOM and the coagulant dosage is mostly determined by the NOM-metal ion interactions rather than particle-metal ion interactions. Hence, SRNOM that is not bound to nC₆₀ could consume metal ions upon the addition of coagulants, limiting the opportunity to destabilize nC₆₀-SRNOM complexes. Furthermore, a previous study (Hyung and Kim, 2008) suggests that the adsorption of NOM provides carbon fullerene with thermodynamically more favorable surfaces, as well as steric and electrostatic stabilization. Thus, the adsorption of NOM to nC₆₀ would be enhanced as the concentration of NOM in the aqueous phase increases. The enhanced stability of nC₆₀ in the presence of SRNOM was further investigated by measuring the zeta potential of the nC₆₀ suspension under different NOM concentrations at a pH range of 5-9. Results shown in Fig. 8 suggest that as the SRNOM concentration increased, the zeta potential of C₆₀ shifted toward more negative values, which would be accompanied by enhanced colloidal stability.

Another set of jar tests was performed using a representative fullerene derivative, fullerol (C₆₀(OH)₂₄), which is also known to form polydispersed colloidal aggregates in the aqueous phase (Brant et al., 2007). Compared to nC₆₀, the removal of derivatized C₆₀ by coagulation/flocculation and sedimentation was less efficient under the same water quality conditions (Fig. 9). When the alum dosage was less than 25 mg-Al/L, more than 80% of fullerol was removed, but only in a very narrow pH range of 7-8. When the alum dosage was as high as 100 mg-Al/L, although the maximum removal

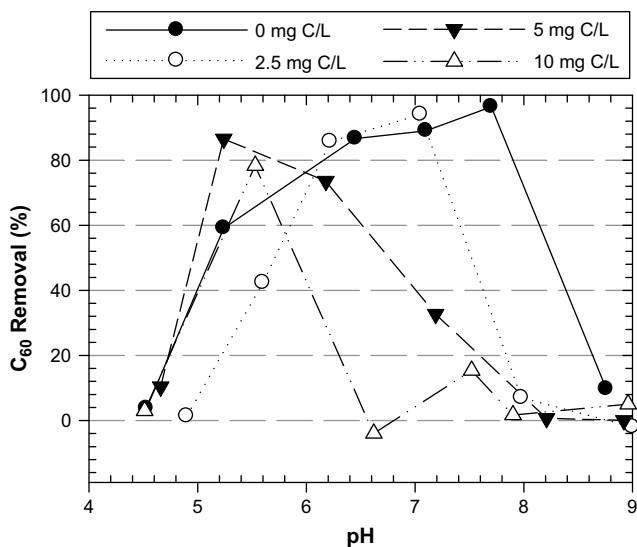


Fig. 7 - Effect of NOM concentration on nC₆₀ removal (initial C₆₀ concentration = 5 mg/L, alkalinity = 100 mg/L as CaCO₃, alum dose = 25 mg/L).

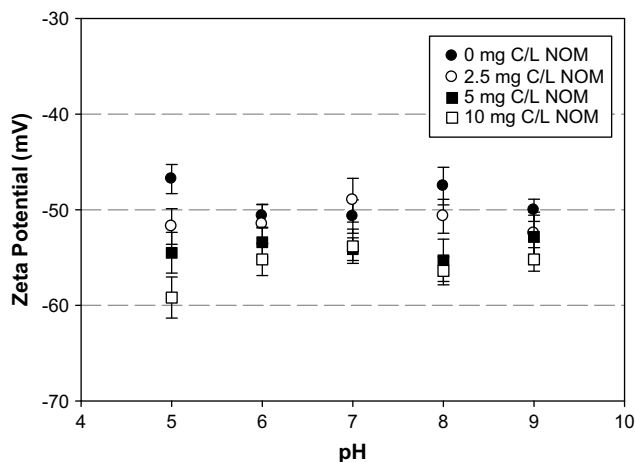


Fig. 8 - Zeta potential of nC₆₀ at different background SRNOM concentrations.

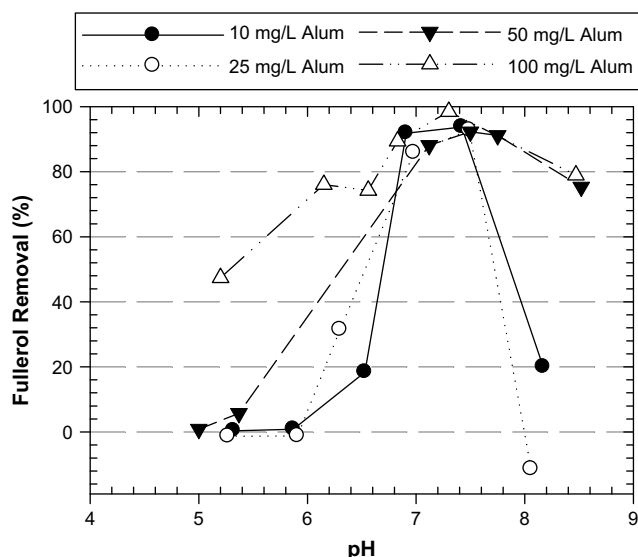


Fig. 9 - Removal of fullerol colloids by coagulation (initial fullerol concentration = 1 mg/L, alkalinity = 100 mg/L as CaCO₃).

increased up to 98% at pH 7.5, the removal sharply decreased when the pH deviated from this value. Although relatively high removal could be achieved, the coagulation process should be carefully operated at optimal sweep coagulation condition in order to effectively remove fullerol. This result implies that C₆₀ that has been derivatized to enhance dispersion in the aqueous phase, or which may occur during pretreatment steps (for example, preozonation during enhanced coagulation processes (Fortner et al., 2007)) might reduce the removal efficiency of fullerenes during water treatment.

4. Conclusions

The results of this study illustrate that in the event of a large scale release of pristine C₆₀ into a natural water body - either

in the form of dry C_{60} or in organic solvent – it will form water stable aggregates in the presence of NOM, with characteristics similar to nC_{60} prepared by other methods reported in the literature. NOM is expected to play critical role on the transfer of C_{60} in aqueous phase either hindering or facilitating dissolution of C_{60} depending on disposal scenario. These colloidal aggregates were adequately removed by a series of particle destabilization and removal unit operations widely used in water treatment, including alum coagulation, flocculation, sedimentation and filtration. Yet, the efficiency of removal greatly depended on water quality parameters, existence of NOM, and coagulant dosage. More focused research is required to evaluate the exact removal mechanisms, though it is likely that these particles are removed by sweep coagulation – a commonly utilized coagulation method in surface water treatment. It is noteworthy that the removal became less efficient as C_{60} was chemically derivatized to hydroxylated C_{60} . As various C_{60} derivatives with enhanced water solubility become more commercially available and nC_{60} can undergo chemical derivatization during ozonation, future research should focus on evaluating the removal of various forms of fullerene during water treatment processes.

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