Nature Environment and Pollution Technology
An International Quarterly Scientific Journal

p-ISSN: 0972-6268
(Print copies up to 2016)
e-ISSN: 2395-3454

Vol. 22 | No. 1 | pp. 497-502 | 2023

Original Research Paper

https://doi.org/10.46488/NEPT.2023.v22i01.048

Open Access Journal

Performance of Alum Coagulation and Adsorption on Removing Organic Matter and *E. coli*

E. N. Hidayah*†, O. H. Cahyonugroho* and N. A. Fauziyah**
*Department of Environmental Engineering, Universitas Pembangunan Nasional Veteran Jawa Timur, Indonesia
**Department of Chemical Engineering, Universitas Pembangunan Nasional Veteran Jawa Timur, Indonesia
†Corresponding author: E.N. Hidayah; euisnh.tl@upnjatim.ac.id

Nat. Env. & Poll. Tech.
Website: www.neptjournal.com

Received: 07-08-2022
Revised: 17-09-2022
Accepted: 29-09-2022

Key Words:
Organic matter
E. coli
Coagulation
Adsorption
Chlorination

ABSTRACT

Surface water is the primary resource for raw water in drinking water treatment processes. Therefore, the presence of microorganisms, bacteria, and viruses should be the main focus in drinking water treatment, in addition to natural organic matter, which is composed of organic carbon groups derived from aquatic biota as well as organic material, organic matter from industrial and domestic waste. This study applied coagulation-flocculation followed by adsorption as the advanced treatment with activated carbon for removing organic matter and bacteria simultaneously to know each process's performance. The results indicated that all treatment processes have a good performance for removing dissolved organic matter in water with efficient removal of 28.35%-70.75% of TOC concentration and 26.75%-55.95% of UV_{254} concentration. Further, the selected processes demonstrated a high percentage of removal of *E. coli*, about 65.35%-96.43%. However, the effect of chlorination impacted the increasing THMs concentration up to 36.32%, while the other processes could remove THMs concentration 17.25%-51.08%. Overall, this study conjectures that all treatment processes simultaneously perform well for removing dissolved organic matter, THMs, and *E. coli*. However, chlorination should be managed to control the formation of THMs due to the remaining organic matter in water.

INTRODUCTION

Surface water contains diverse pollutants and biological activities that humans and industries generate. One of the primary pollutants is organic matter, known as natural organic matter (NOM), which is composed of organic carbon groups derived from plants or aquatic biota, as well as organic material and organic matter from industrial and domestic waste. Characterization of the organic matter revealed that organic matter is composed of various molecular weight fractions, aromatic, aliphatic, hydrophobic/hydrophilic, and humus/non-humus (Sillanpää et al. 2015). Surface water is the primary resource for raw water in drinking water treatment processes. Therefore, drinking water treatment should focus on microorganisms, bacteria, and viruses. Numerous microorganisms and bacteria have been identified as the most dangerous pathogens, capable of causing diarrhea, diseases, and death (Edzwald & Tobiason 2011). In developing countries, *Escherichia coli* (*E. coli*) is one of the most common causes of many bacterial infections, including gastroenteritis (Owoseni et al. 2017).

Nevertheless, surface water is used as raw water for the production of clean and drinking water. The most applicable clean and drinking water technologies are coagulation-flocculation, sedimentation, filtration, and disinfection. However, it has limitations in treating all parameters to meet the quality standards optimally, especially to fill the requirement for the lowest concentration of organic matter and bacteria or viruses concentration simultaneously (Chen et al. 2021).

The most commonly used clean water treatment technology to set aside the content of viruses and bacteria is a chlorine disinfection process because cheap and chlorine is relatively stable. It means that chlorine residual remains in the distribution system, preventing the regrowth of viruses and microorganisms (Edzwald & Tobiason 2011). However, chlorine is very reactive and quickly reacts with organic and inorganic materials contained in water. Reaction chlorine on the organic matter can cause the formation of disinfectant by-products (DBPs), namely trihalomethanes (THMs) and haloacetic acids (HAAs), which are carcinogenic and harmful to human health (Bond et al. 2009, Hidayah et al. 2017). Some issue related to the formation of DBPs is increasing chlorine dosage to eliminate viruses and bacteria in production water. In contrast, the treated water still contains dissolved organic matter (DOM). Therefore, it will

trigger an increase in carcinogenic compounds in drinking water (Bond et al. 2009, Hidayah et al. 2016).

Regarding natural organic matter (NOM) removal, generally, the most common and economically feasible standard processes for removing NOM and drinking water parameters are considered coagulation and flocculation, sedimentation, and sedimentation and filtration. The most widely used coagulant is an alum and iron-based coagulant, which has been reported to be consistently more effective in removing NOM (Uyak & Toroz 2007, Matilainen et al. 2010). Coagulation-flocculation has primarily been used in waterworks companies and is typically applied before further treatments. The other treatment process is the application of advanced treatment of adsorption by using activated carbon, charcoal, and diverse biosorbent. Activated carbon is a suitable adsorbent due to its ability to bind with various molecules (Han et al. 2015, Azad et al. 2020). In addition, activated carbon has been found that can be used to remove bacteria and bacteria toxins, in addition to developing the water quality used for consumption purposes.

Furthermore, the active part of adsorbents can remove organic matter in the water (Karanfil et al. 1999, Han et al. 2015) and remove pathogens (Naka et al. 2001). This study applied coagulation-flocculation followed by adsorption as the advanced treatment with activated carbon to remove organic matter and bacteria simultaneously. This study aimed to know the concentration of THMs and *E. coli* under post-treatment of adsorption only and adsorption followed by chlorination.

MATERIALS AND METHODS

The primary source of raw water for water treatment, Kali Surabaya in Surabaya City, provided the source water for this study. The raw water sample was taken in May 2022. This study was conducted under four treatments: alum, adsorption with activated carbon, coagulation-adsorption, and coagulation-adsorption-chlorination. The first treatment, alum ($Al_2(SO_4)_3.18H_2O$, Merck, Germany), was used as a coagulant with various dosages of 10 to 60 $mg.L^{-1}$ with intervals of 10 $mg.L^{-1}$. Jar test equipment was used for flash mixing for 3 min at 100 rpm, slow mixing for 15 min at 35 rpm, and then settling for 15 min. The second treatment, powder-activated carbon (PAC), was used as an adsorbent with various dosages of 10 to 60 $mg.L^{-1}$ with an interval of 10 $mg.L^{-1}$ under slow mixing for 60 min at 35 rpm. In the third treatment, the optimum alum dosage from the first treatment was applied in coagulation, followed by adsorption with the optimum adsorbent dosage at the second treatment. The treated water from the third treatment was injected with 10 $mg.L^{-1}$ $Na(OCl)_2$ (Merck Germany) as a disinfectant solution with 30 min contact time, or it was the fourth treatment. All treated samples from each treatment were collected and filtered through 0.45 m filter paper (cellulose acetate) before being subjected to additional analysis, which included dissolved organic matter in terms of total organic carbon (TOC) and ultraviolet absorbance at 254 nm wavelength (UV_{254}); THMs, and *E. coli*. The TOC and UV_{254} analyses followed Standard Methods and procedures. (APHA AWWA & WEF 2012). TOC was determined using the combustion-infrared method with a total organic carbon analyzer Model TOC Analyzer 5000A Shimadzu, and UV_{254} was determined with a Carry 100 Bio UV-Visible Spectrophotometer with a 1 cm quartz cell. THMs were determined using gas chromatography and headspace-solid-phase microextraction (HS-SPME, 85 m carboxy/polydimethylsiloxane fiber). The DPD ferrous method was used to measure residual chlorine (APHA AWWA & WEF 2012). The Most Probability Number (MPN) test was used to determine the amount of *E. coli* in all treated samples. The MPN is an estimated number of individual bacteria with a 95% confidence interval, with a unit per 100 mL or gram.

RESULTS AND DISCUSSION

Fig. 1 explains the performance of coagulation, adsorption, and its combination processes for removing organic matter. The graph shows the percentage change of each treatment's TOC concentration and UV_{254} concentration. Firstly, the results present the maximum efficiency of each treatment process for TOC concentration, that is, 28.35% removal in alum coagulation, 46.85% removal in adsorption of activated carbon, 63.25% removal in alum coagulation followed by adsorption with activated carbon, and 70.75% removal in alum coagulation combined with adsorption with activated carbon followed by chlorination. This result indicated that all treatment processes effectively remove dissolved organic matter in water. Coagulation is the primary process in all water and wastewater treatment (Sillanpää et al. 2015, Hidayah et al. 2017). Previous studies have revealed the efficiency of coagulation up to 50% in removing dissolved organic matter, which depends on the treated water's characteristics (Aslam et al. 2013, Hidayah et al. 2016). Adsorption is an advanced treatment for removing pollutants such as heavy metals, particulates, organic matter, etc. Several kinds of research have demonstrated adsorption performance with activated carbon as an adsorbent for removing dissolved organic matter up to 30% (Han et al. 2015). The capacity and characteristics of the adsorbent, as well as the capacity and characteristics of activated carbon, determine the efficiency of the adsorption process (Karanfil et al. 1999). Adsorption with activated carbon demonstrated

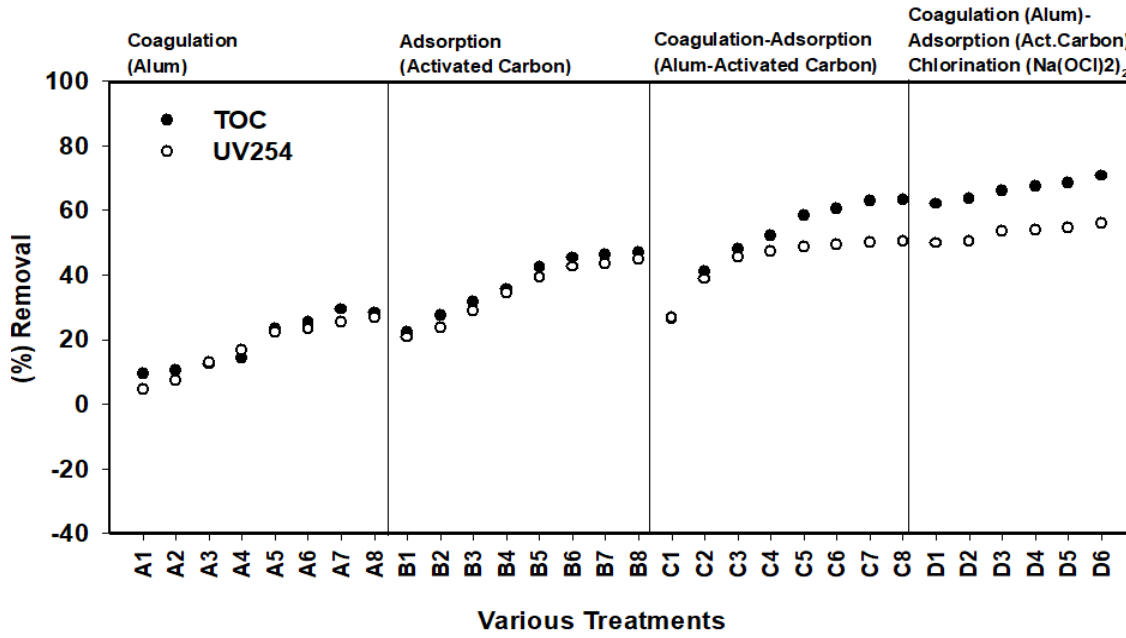

Fig. 1: The efficiency removal of various treatments for removing dissolved organic matter in water.

a high removal of hydrophilic and low molecular weight of dissolved organic matter (Velten et al. 2011).

Secondly, alum coagulation and adsorption with activated carbon have a better efficient removal than alum coagulation. Another combination, coagulation-adsorption-chlorination, performed a similar result to the coagulation-adsorption processes. It indicates that post-chlorination had an insignificant effect on the removal of dissolved organic matter. Chlorination could remove dissolved organic matter through various mechanisms, such as transformation, oxidation, and polymerization. The mechanism of chlorination on microorganisms is to inactivate a microorganism by damaging its cell membrane, and chlorine can be involved in the cell and alter cell respiration and disrupt DNA activities (Bond et al. 2009, Mazhar et al. 2020). Chlorination of organic matter can destroy the organic coating on the particle's surface, causing the zeta potential to change. Oxidation will strengthen the bond between particles and their adsorbed organics, lowering the molecular weight of organic matter.

Furthermore, oxidative polymerization of dissolved organic matter may result in particle aggregation via bridging reactions (Xie et al. 2016). Due to organic being transformed into lower molecular weight, the TOC analyzer and UV_{254} could not detect the lower molecular weight organic compounds. According to previous studies, HPSEC with UVD/OCD revealed the changing molecular weight organic after chlorination (Hidayah et al. 2017) and the changing of fluorophores properties as detected by FEEMs (Han et al. 2015).

Third, UV_{254} concentration decreased to 26.75% removal in alum coagulation, 44.85% removal in adsorption of activated carbon, 50.32% removal in alum coagulation, followed by adsorption with adsorption activated carbon, and 55.95% removal in alum coagulation combined with adsorption with activated carbon followed by chlorination. The changing of UV_{254} concentration demonstrated that these selected processes could remove aromatic dissolved organic matter composed of saturated carbon double bonds. Fourth, comparing the percentage removal of TOC concentration and UV_{254} concentration results in higher removal of TOC concentration than UV_{254} concentration. UV_{254} represents aromatic dissolved organic carbon, while TOC explains all types of dissolved organic carbon. It means a higher removal of TOC indicates a removal of the other kind of dissolved organic carbon instead of an aromatic compound, such as aliphatic non-aromatic. The removal difference between TOC concentration and UV_{254} concentration in alum coagulation was lower than the removal difference in alum coagulation combined with adsorption with activated carbon followed by chlorination. These results follow the performance of each treatment, which mentioned that alum coagulation has a good performance for the removal of aromatic and less for non-aromatic compounds (Aslam et al. 2013, Hidayah et al. 2016). Adsorption has been well known to remove low molecular weight organic compounds,

hydrophilic and non-aromatic compounds (Velten et al. 2011, Azad et al. 2020), and it was related to a higher removal difference between TOC concentration and UV_{254} concentration in adsorption with activated carbon followed by chlorination (Azad et al. 2020).

Fig. 2 presents the changing THMs and *E. coli* concentrations in each treatment process. THMs decreased up to 17.25% in alum coagulation, adsorption with activated carbon removed 35.32% THMs concentration, and a higher removal up to 51.08% in alum coagulation followed by adsorption with activated carbon. However, alum coagulation, adsorption, and chlorination increased THMs concentration by about 36.32%. This result shows compliance with the removal of TOC and UV_{254} concentration, as shown in Fig.1. The selected processes, except chlorination, are potentially carried out to remove dissolved organic matter. It is well known that organic matters are the precursor for the formation of disinfectant by-products, such as THMs and HAAs. There is contradictory information regarding dissolved organic matter fractions are the most important precursors of THMs and HAAs.

Regarding hydrophobicity or hydrophilicity, it has been proposed that the hydrophilic fraction is a more critical precursor of THMs than HAAs. According to the humic compound, a humic substance fraction is a significant precursor for HAAs, while an aliphatic biopolymer fraction is a primary precursor for THMs. The characteristic organic matter with high molecular weight and hydrophobic was the primary source of THM precursors.

In contrast, the low molecular weight components were more significant DHAA (dihaloacetic acids) precursors than THAA (trihaloacetic acids) (Wang et al. 2013, Hidayah et al. 2016). Increasing THMs concentration indicates the formation of by-product compounds due to disinfections reacting with the organic matter in treated water. The higher chlorination dosage caused a higher formation of by-product compounds. In addition, the dominant mechanism of the reaction of chemical biocides with NOMs and other constituents in water is different. Therefore, the type and level of the resultant DBPs are expected to differ (Mazhar et al. 2020, Hidayah et al. 2017). Factors such as oxidant concentration, pH, organic matter characteristics, temperature, bromide existence, and precursor reactivity may affect DBPs formations. Therefore, water work companies have been looking for alternative disinfection processes, especially using chemical compounds, reducing the formation and discharge of DBPs. The most recently used chemical biocide is chlorine dioxide (ClO_2) (Edzwald & Tobiason 2011, Postigo et al. 2021).

Secondly, Fig. 2 shows the *E. coli* concentration gradually reduced up to 65.35% in coagulation, 70.32% in adsorption, 90% in coagulation-adsorption, and 96.43% in coagulation-adsorption-chlorination. The trend of *E. coli* reduction in those treatment processes follows the THMs reduction, except in coagulation-adsorption-chlorination. Treatment processes without chemical chlorination resulted in an inline reduction between THMs concentration and *E. coli* concentration. The reduction of THMs concentration

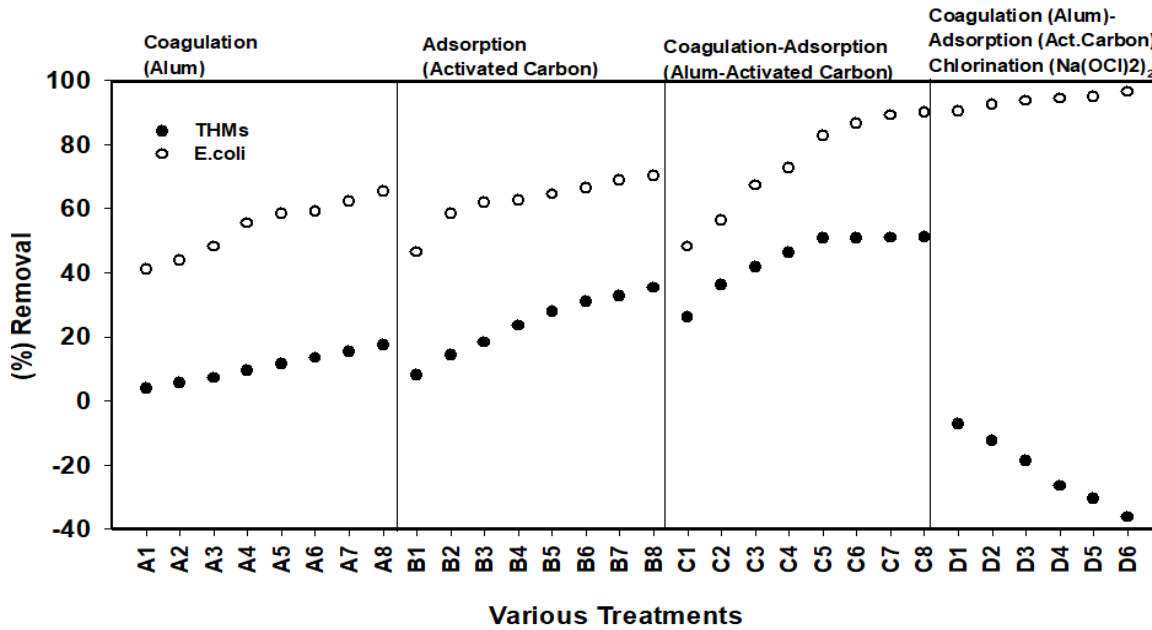

Fig. 2: The efficient removal of various treatments for removing the THMs formation and *E. coli*.

is lower than *E. coli* concentration removal, probably due to THMs indicating the existence of organic matters as precursors of DBPs formations (Mazhar et al. 2020). A higher reduction of *E. coli* through those selected processes is probably due to the power of each mechanism applied. Several mechanisms explain the coagulation of particles and organic substances, including precipitation, charge neutralization, adsorption, bridge aggregation, and sweep-flocculation. The coagulant develops the chemical and physical properties of flocs, especially on the particle bridging, increasing settling velocity.

Further, the cells are more easily taken into flocs, resulting in a greater density, particle size, strength, and settleable flocs. Furthermore, in addition, to charge neutralization, the surface charge of the microorganism is an important factor in the removal of microorganisms during the coagulation process (Aslam et al. 2013). Several studies mentioned the removal of microorganisms and viruses from water by coagulation, flocculation, and sedimentation (Shin & Sobsey 2015, Chen et al. 2013, Swiatlo et al. 2002) because it is most likely due to microorganisms having a negative charge. Microorganisms contain pyruvate, acidic sugars, or phosphate in different serotype capsular polysaccharides, with additional contributions from cell surface structures, which caused the negative charge (Swiatlo et al. 2002). The internal surface area of activated carbon may affect *E. coli* removal, mainly activated carbon with a large area of its pore. The activated carbon has various pore sizes and shapes due to it has complex network inside. Therefore, the high capacity adsorption indicates that the internal pores provide a vast internal surface area for activated carbon. However, in some cases, the activated carbon has low binding capacity because activated carbon does not possess pores of sufficient diameter for those bacteria (Chandy & Sharma 1998, Naka et al. 2001. Li et al. 2013). Combining coagulation followed by adsorption has given a higher efficiency removal of *E. coli* because *E. coli* was continuously removed into two different processes (Chen et al. 2021). Further, the robust chlorination process provides a higher removal of *E. coli* because chlorine is a potential oxidant that destroys the microorganism's nucleic acids and cell membranes, such as the effectiveness of chlorination for *E. coli* inactivation (Edzwald & Tobiason 2011).

CONCLUSIONS

This study presents the performance of alum coagulation, adsorption with activated carbon, alum coagulation followed by adsorption, and the effect of chlorination after the coagulation and adsorption in removing dissolved organic matter as parameters TOC and UV_{254} and in removing *E. coli* and THMs as one of the DBPs potentials. The efficiency performance of those treatment processes shows various percentage removal, such as 28.35%-70.75% of TOC concentration, 26.75%-55.95% of UV_{254} concentration, 65.35%-96.43% of *E. coli* concentration, 17.25%-51.08% of THMs concentration. Regarding dissolved organic carbon, combining alum coagulation and adsorption with activated carbon has a better efficiency removal than alum coagulation only. At the same time, coagulation-adsorption-chlorination performed a similar result to coagulation-adsorption processes. It indicates that post-chlorination had an insignificant effect on the removal of dissolved organic matter. Treatment processes without chemical chlorination resulted in an inline reduction between THMs concentration and *E. coli* concentration. Chlorination affected the increase of THMs formation up to 36.32% due to the reactivity of chlorine with dissolved organic matter. The removal difference between TOC concentration and UV_{254} concentration in alum coagulation was lower than the removal difference in alum coagulation combined with adsorption with activated carbon followed by chlorination.

ACKNOWLEDGMENTS

This study was a part of the Primary University Flagship Research (PDUPT) scheme Basic Research (PD), which the Directorate of Research supports, Technology, and Community Services, Directorate General of Higher Education, Research, and Technology, Ministry of Education, Culture, Research, and Technology in the Year 2022, SP DIPA-023.17.1.690523/2022 2nd revised date April 22nd, 2022

REFERENCES

APHA, AWWA and WEF 2012. Standard Methods for the Examination of Water and Wastewaters. Twenty-First edition. American Public Health Association, Washington, D.C.

Aslam, Z., Chow, C.W.K., Murshed, F., van Leeuwen, J., Drikas, M. and Wang, D. 2013. Variation in character and treatability of organics in river water: an assessment by HPAC and alum coagulation. Sep. Purif. Technol., 120: 162-171.

Azad, M.S., Azhari, S. and Hassan, M.S. 2020. Removal of methylene blue, Escherichia coli, and Pseudomonas aeruginosa by adsorption of activated carbon produced from moringa oleifera bark. Malay. J. Sci. Health Technol., 7: 29-39

Bond, T., Henriet, O., Golan, E.H., Parsons, S.A. and Jefferson, B. 2009. Disinfection by-product formation behavior of natural organic matter surrogates. Environmental Science and Technology, 43:5982-5989.

Chandy, T. and Sharma, C.P. 1998. Activated charcoal microcapsules and their applications. J. Biomater. Appl., 13(2):128-157

Chen, L., Deng, Y., Dong, S., Wang, H., Li, P., Zhang, H. and Chu, W. 2021. The occurrence and control waterborne viruses in drinking water treatment: A review. Chemosphere, 281: 1-11.

Edzwald, J.K. and Tobiason, J.E. 2011. Chemical principles, source water composition, and watershed protection. In Edward, J.K. (ed.), Water

Quality & Treatment: A Handbook on Drinking Water. AWWA McGraw-Hill, New York, pp. 1-76.

Han, Q., Yan, H., Zhang, F., Xue, N., Wang, Y., Chu, Y. and Gao, B. 2015. Trihalomethanes (THMs) precursor fractions removal by coagulation and adsorption for bio-treated municipal wastewater: Molecular weight, hydrophobicity/hydrophily, and fluorescence. J. Hazard. Mater., 297: 119-126.

Hidayah, E.N., Yeh, H.H. and Chou, Y.C. 2016. Using HPSEC to identify NOM fraction removal and the correlation with disinfection by-product precursors. Water Sci. Technol. Water Supply, 16(2): 305-313.

Hidayah, E.N., Yeh, H.H. and Chou, Y.C. 2017. Comparison between HPSEC-OCD and FEEMs for assessing DBPs formation in water. J. Environ. Sci. Health Part A, 52(4): 391-402.

Karanfil, T., Kitis, M., Kilduff, J.E. and Wigton, A. 1999. Role of granular activated carbon surface chemistry on the adsorption of organic compounds natural organic matter. Environ. Sci. Technol., 33(18): 3225-3233.

Li, D., Zeng, S., Gu, A.Z., He, M. and Shi, H. 2013. Inactivation, reactivation, and regrowth of indigenous bacteria in reclaimed water after chlorine disinfection of a municipal wastewater treatment plant. J. Environ. Sci., 25:1319–1325.

Matilainen, A., Vepsäläinen, M., and Silanpää, M. 2010. Natural organic matter removal by coagulation during drinking water treatment: a review. Adv. Coll. Interf. Sci., 159: 189-197.

Mazhar, M.A., Khan, N.A., Ahmed, S., Khan, A.H., Hussain, A., Rahisuddin, Changani, F., Yousefi, M., Ahmadi, S. and Vambol, V. 2021. Chlorination disinfection by-products in municipal drinking water: A review. J. Clean. Prod., 16: 273.

Naka, K., Watarai, S., Tana, Inoue, K., Kodama, Y., Oguma, K., Yasuda, T. and Kodama, H. 2001. Adsorption effect of activated charcoal on enterohemorrhagic Escherichia coli. J. Veter. Med. Sci., 63(3): 281-285

Owoseni M.C., Olaniran, A.O. and Okoh, A.I. 2017. Chlorine tolerance and inactivation of Escherichia coli recovered from wastewater treatment plants in the eastern cape, south Africa. Appl. Sci., 7(8): 810-825.

Postigo, C., Solsona, R.G., Batista, M.F.H., Ferrero, P.G., Alygizakis, N., Ahrens, L. and Wiberg, K. 2021. A step forward in detecting by-products of anthropogenic organic micropollutants in chlorinated water. Trends Environ. Anal. Chem., 32: 116-125.

Shin, G.A. and Sobsey, M.D. 2015. Removal of norovirus from water by coagulation, flocculation, and sedimentation processes. Water Supply, 15(1): 158-163.

Sillanpää, M., Metsämuuronen, S. and Mänttäri, M. 2015. Membranes. In: Silaanpää, M. (eds.), Natural Organic Matter in Water: Characterization and Treatment Method, Butterworth-Heinemann, Oxford, pp. 113-157.

Swiatlo, E., Champlin, F.R. and Holman, S.C. 2002. Contribution of choline-binding proteins to cell surface properties of streptococcus pneumonia. Infect. Immun., 70(1): 412-415.

Uyak, V. and Toroz, I. 2007. Disinfection by-product precursors reduction by various coagulation techniques in Istanbul water supplies. J. Hazard. Mater., 141(1): 320-328.

Velten, S., Detlef, R.U.K., Traber, J., Kaiser, H.P., Guntena, U., Boller, M. and Meylan, S. 2011. Characterization of natural organic matter adsorption in granular activated carbon adsorbers. Water Res., 45: 3951-3959.

Wang, D.S., Zhao, Y.M., Yan, M.Q. and Chow, C.W.K. 2013. Removal of DBP precursors in micro-polluted source waters: A comparative study on the enhanced coagulation behavior. Sep. Purif. Technol., 118: 271-278.

Xie, P., Chen, Y., Ma, J., Zhang, X., Zou, J. and Wang Z. 2016. A mini-review of peroxidation to improve coagulation. Chemosphere, 155: 550-563.