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Interrogation on Optical Properties of Electrolyte Minerals, TSS and Turbidity in Water - Spectrophotometer and IOP Profiler Based Hyperspectral Analysis Approach

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Abstract

The optical properties of total suspended solids, turbidity, and electrolyte minerals in water such as sodium, potassium, and calcium were studied under laboratory and field-oriented conditions. Spectrophotometer and Wet Lab Instrument Inherent Optical Properties profiler have been used to measure the optical properties. The polynomial fit between the field measured absorption spectra and laboratory measured absorption spectra of each mineral explains in detail the chemical water quality parameters and optical properties. The spectrophotometrically derived absorption coefficient of electrolyte minerals such as sodium (1,000 ppm), potassium (100 ppm), and calcium (1,000 ppm), TSS (500 ppm) and Turbidity (100 NTU) makes virtuous correlation with the IOP profiler measured absorption coefficient donates the best polynomial fit R^2 values of 0.9076, 0.9019, 0.9645, 0.9823 and 0.9354 respectively. Results in this study exhibits that, the absorption coefficient of electrolyte minerals, TSS, and Turbidity in water increases with increasing their concentrations at the wavelengths range from 200 to 300 nm.

Keywords: Total suspended solids, Turbidity, Electrolyte minerals, Spectrophotometer, IOP absorption coefficient, Spectra

Introduction

Water is an essential component for human life which can be explained in terms of optical properties, (i.e.) translucent to the electromagnetic radiation that emerging in the region of visible wavelength, and it is thick-headed towards the up and down of this wavelength [1]. Though, in water body, visible light can be affected by both refraction and attenuation [2].

The studying of optical properties of the water quality parameters is a part of physics and remote sensing which deals with the light field that interacts with the water environment can be called as aquatic optics [3]. The dealings of light with the water column affected by the particles that changes the magnitude of the spectra from its normal form. The aquatic optics can be subdivided into limnetic and oceanographic optics. Recent days there was an attempt made to study the optical properties of inland lake water using the optical profilers. Surface water optics is a new term that has been used now a days to elaborately and continuously monitor the inland water bodies.

In general, some radiant energy from sun is reflected at the water surface and not intruded by the water body [4]. These energy at the water surface is attenuated by inherent optical properties and transformed to other measures of energy, (i.e.) heat produced from it can evaporates the water either it is utilized by aquatic plants for photosynthesis or for some other process [5]. The light source which is not absorbed by the water, scattered by suspended particles present in the water. This scattered energy is bounded to various directional paths and it is directed upward to get recorded in the remote sensing sensor. Scattered light and reflected light which is returned from the suspended particles determines the quality of surface water [6]. The intensity of light radiation affected by water molecules, dissolved salts, organic substances, and suspended particles [7]. **Figure 1** shows, how the light is interacting with matter and it's separated into several forms such as diffusion, reflection, absorption and transmission [8-10]. This is how the spectroscopy measure optical constants of the liquid column provided in the cuvette.

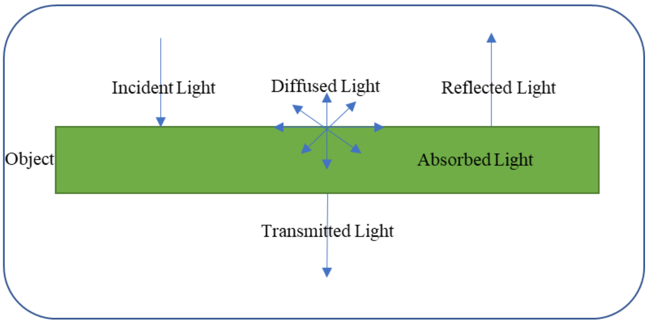


Figure 1 Light Interaction with object.

Li *et al.* [11] developed the optical properties of sodium chloride solution between the wavelength of 300 to 2,500 nm at room temperature using double optical pathlength transmission method. It is concluded from this study is, the refractive index of Nacl solution was increases with Nacl concentrations and also the linear fitted methods of the absorption and refractive index of NaCl solution of wavelength and various NaCl concentration are delivered.

Warren, 2019 [12], conducted an experiment to study the optical properties (Absorption coefficient and refractive index) of ice and snow. The absorption, transmittance and reflectance are disturbed by the size of the grain, bubbles and inclusions of brine. Darren. S. Baldwin and William Valo, 2015 [4], studied the optical properties of water with respect to the effect of dissolved organic carbon. Absorbance and fluorescence at the wavelengths of 240 and 350 nm correlated with DOC and posses' correlation with r^2 value of 0.8.

The optical properties of chemical water quality parameters is lacking in previous studies. In this study, an experiment has been processed to analyze the optical properties of electrolyte minerals and physical parameters present in the water at different level of concentrations. The objective of this study is to obtain the optical constant (Absorption coefficient) of Electrolyte minerals such as calcium, potassium, sodium chloride, and TSS, turbidity at various concentrations using spectroscopic method under laboratory and field-oriented conditions.

Materials and methods

Figure 2 shows the methodology involved in this study. Spectroscopy optical pathlength transmission method was used here in this study to develop the optical constants of electrolyte minerals, TSS and turbidity. It is used to measure and interpret the EMR (Electromagnetic Radiation) absorption or reflection of the molecules over the energy state when it is moving from 1 state to another. As a light source Tungsten filament lamps and Hydrogen-Deuterium lamps are widely used one in the field. The wavelengths from 200 to 800 nm was recorded for the various concentrations of the solution. A distilled water was used as base solution to develop the standard curve. In this work OPTM was implemented to obtain the optical characteristics of above said constituents (**Figure 3**).

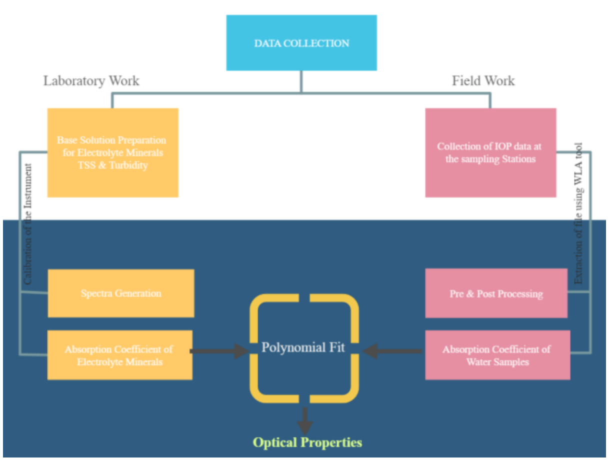


Figure 2 Methodology flow chart.

Figure 3 Standard solutions preparation for development of Spectra for physical and chemical water quality parameters.

Transmittance of the solutions can be expressed as Eq. (1);

$$T_\lambda = f(n, k, L) \tag{1}$$

Where n & k complex refractive index, L is the thickness of the cuvette. The absorption coefficient values are calculated from absorbance measured from UV spectroscopy. The absorption coefficient can be calculated by using the following formulae (Eq. (2));

$$Absorption\ coefficient\ (m^{-1}) = 2.303 * Absorbance \tag{2}$$

Experimental procedure

By using the standard normal procedure, solutions made for various concentrations for various materials. For 1,000 ppm Nacl solution 2.542 g of sodium chloride dissolved in 1 L of distilled water [5]. Calcium chloride with 1,000 ppm solution prepared by, mixing of 21.9072 g of cacl2 with 1 L of distilled water. Preparation of 1,000 ppm of Potassium chloride done by, mixing of 7.4549 g of kcl with 1 L of distilled water. 1,000 ppm magnesium chloride solution prepared by, dilution of 20.3299 g of $mgcl_2$ with 1 L of distilled water. However, these solutions can be used to prepare 10 ppm, 50 ppm, 200 pm, and 500 pm stock solutions. The 400 NTU turbidity solution prepared by mixing of 1 % solution of anhydrous barium chloride ($BaCl_2$) and 1 % solution of sulfuric acid (H2SO4) Combine with the barium chloride and sulfuric acid solutions to form a turbid suspension and it is kept in the room temperature for 24 h to remain it is suspended. As per the standard protocols 10NTU, 50 NTU and 100 NTU stock solutions were made.

Distilled water used as base solution to calibrate the instrument. The instrument was calibrated for 200 to 900 nm and dark correction was done. For different various concentrations, the absorbance spectra were developed for the wavelengths. The protocol for the development of absorbance spectra is shown in **Figure 4**. It is proposed in this study, an optical constants inversion model occurred depends on absorbing layers containing several reflections which is opt for liquid measurements. The energy produced from the sun at this wavelength ranges from 200 - 800 nm will be a 90 % of the total energy produced from solar radiation. Here transmittance was measured at the wavelengths ranges from 200 to 800 nm with the spectral resolution of 10 nm using UV spectroscopy. This UV-Spectroscopy consists of monochromator with tungsten lamp to produce monochromatic light from UV to near infrared. The measurements were carried out at under normal room temperature and atmospheric pressure.

Experimental uncertainty: In order to access the experimental uncertainty of this method, each sample solutions were involved for 5 iterations. The normal spectral absorbance measurements have good repeatability taken into account for inference with the SD (Standard Deviation) value less than 0.5 %. The absorbance accuracy measurement was directly taken from the instrument at every wavelength. The standard uncertainty of the measured absorbance (absλ) was calculated by Eq. (3);

$$abs_\lambda = \sqrt{(1.11\mu_a)^2 + \triangle_\lambda^2} \tag{3}$$

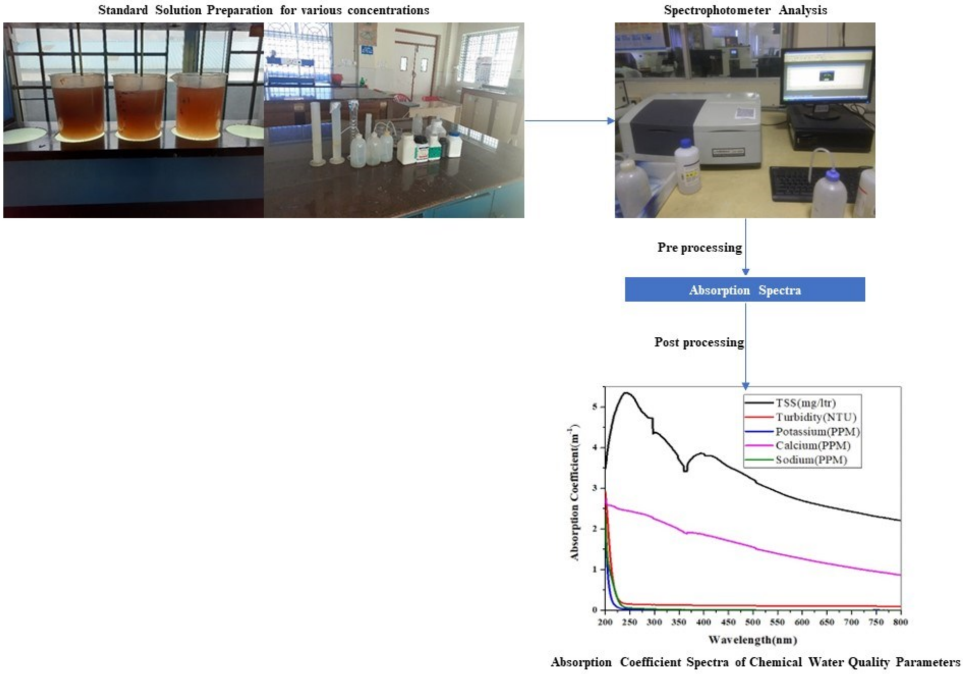


Figure 4 Work flow of spectra generation of electrolyte Minerals, TSS and turbidity.

The IOP data were collected using AC-S sensor wet lab instrument. The AC-S sensor fixed along with the Conductivity, Temperature and Depth (CTD) inside IOP gauge. The instrument calibration done using the drift tracking method. The milli-Q water forced continuously inside the flow tubes at the rate of 1.5 L/min. The calibration file recorded in the data logger through wet view software connected over the instrument for further processing. The calibration values of absorption and backscattering should read 0 m^{-1} unless otherwise, the process may be repeated until getting the values of +/−0.003 m^{-1}.

The sensor was immersed just 3 meters below the water surface at every sampling location. The data recorded for 86 wavelengths ranges from 400 to 750 nm at 1 nm interval.

The raw dataset recorded and stored in the data logger extracted using the WET Lab Archive Processing tool. There are 2 processing methods involved, one is Correction of Temperature and Salinity and another one is Tube scattering correction.

The temperature and salinity absorption coefficient correction on water can be done by using Eq. (4) [13];

$$a_{mts} = a_m - [\psi_{ta} * (t - t_r) + \psi_{sa} * S] \tag{4}$$

Where a_{mts} – corrected absorption coefficient (m^{-1}), a_m – Extracted values from the sensor (ACS (m^{-1}), t, tr – In situ and reference temperatures, S – Salinity, ψ_{ta} – correction coefficient of temperature, ψ_{sa} – correction coefficient of salinity

The following Eq. (5) used to do the tube scattering correction [13];

$$a_t(\lambda) - a_w(\lambda) = a_{mts}(\lambda) - \frac{a_{mts}(\lambda_{ref})}{[c_{mts}(\lambda_{ref}) - a_{mts}(\lambda_{ref})]} * [c_{mts}(\lambda) - a_{mts}(\lambda)] \tag{5}$$

(λ_{ref}) - reference wavelength

Savitzky-Golay filter in origin pro software used to smoothen the data. **Figure 5** shows the photographs taken during the field survey.

Figure 5 Field measurement photograph IOP measurement.

Results and discussion

The absorption peak at 665 nm inhered due to the TSS dominance. In Vembanad lake, (**Figure 6**) In the reference wavelength 710 nm, the coefficient of absorption recorded as 0.154706 to 1.207210 m^{-1} with a mean and median value of 0.467386 and 0.421155 m^{-1}. There is a change observed in the absorption curve around 410 and 430 nm, indicates the presence of highest content electrolyte minerals. Absorption trough observed near 700 nm reflects its low optical constituents value. In this lake potassium ranges between 70.36 to 104.13 ppm, sodium ranges from 141 to 225.67 ppm, calcium ranges from 111.67 to 192.74 ppm, TSS ranges from 0.306 to 20.72 ppm and Turbidity ranges from 1 to 4.9 NTU. The developed spectra for each concentration of electrolyte minerals, TSS and turbidity were correlated with the field measured absorption coefficient and thus characteristics were discussed.

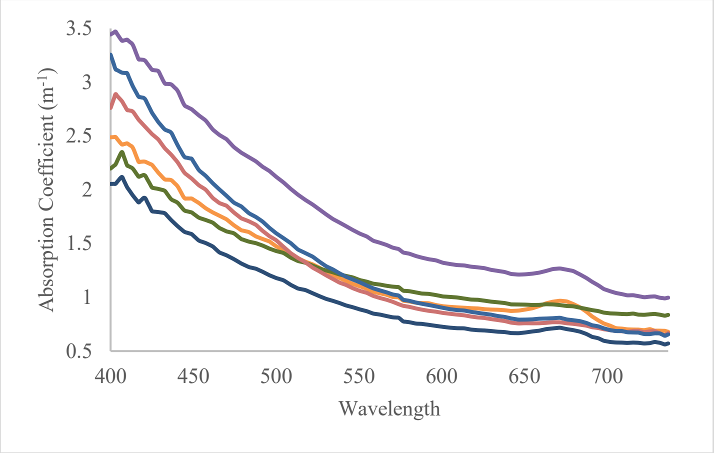


Figure 6 IOP profiler measured absorption coefficient.

Sodium

Anions and temperature present in the solution are the deciding factors for taste threshold of sodium. The threshold values change at room temperature for sodium carbonate it is 20 mg/L, sodium chloride it is 150 mg/L, sodium nitrate it is 190 mg/L, for sulfate it is 220 mg/L, for sodium nitrate 190 mg/L and for sodium bicarbonate it is 420 mg/L. In general sodium salts are highly soluble in water and it can be leached from the atmospheric to surface water and groundwater [11]. Since they are non-volatile in nature it is found with particulate matter in the atmosphere. **Figure 7** shows the absorption coefficient curve of sodium chloride developed using UV-spectroscopy with close view of the spectra with between the wavelengths of 200 to 400 nm. The spectra of absorption coefficient for sodium developed using UV-spectroscopy for different concentrations. The absorption coefficient values for sodium with concentration of 100 ppm ranges from 0 to 0.48 m^{-1}, 300 ppm it ranges from 0 to 0.84 m^{-1}, 500 ppm it ranges from 0 to 1.48 m^{-1} and for 1,000 ppm it ranges from 0 to 2.365 m^{-1}. The refractive index values for sodium with concentration of

100 ppm ranges from 0.015 to 0.024, 300 ppm it ranges from 0.015 to 0.034 and for 500 pm it ranges from 0.015 to 0.067. Sodium in water fully absorbed (NIL) in the visible and NIR regions. Though it starting the absorption peak from the 230 nm (Opposite direction) (i.e) ULa Violet region. There was a certain peak found in 200 nm. The absorption spectra of sodium are exponentially increasing with decreasing wavelength. There was a certain trough found in 203 nm. The different concentrations of spectrophotometrically derived absorption coefficient were correlated with the field derived absorption coefficient values. The absorption coefficient of sodium at 300 ppm, possess less correlation with the absorption coefficient derived in the field with R^2 value of 0.5. On the other hand, Absorption coefficient of 1,000 ppm sodium concentrations having highest correlation with the absorption coefficient derived in the field gives R^2 value of 0.9076. **Figure 8** and **Table 1** shows the polynomial regression analysis incorporated between the absorption coefficient at the field versus absorption coefficient in the laboratory. The absorption coefficient values are increasing with the increasing concentration of sodium.

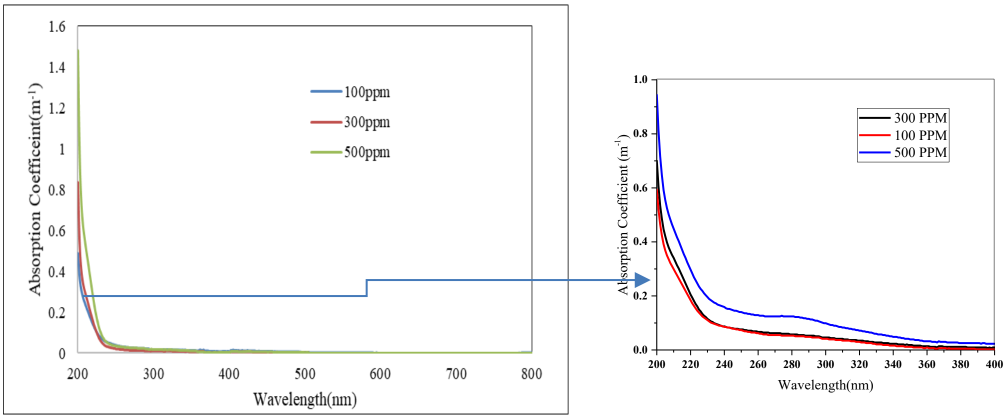


Figure 7 Absorption coefficient spectra of sodium.

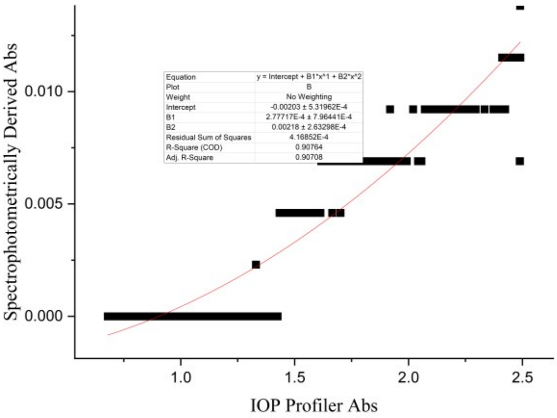


Figure 8 Regression analysis fit between field measured absorption coefficient and laboratory measured sodium absorption coefficient.

Potassium

The absorption spectra developed and it is shown in **Figure 9**. for various concentration of 100, 300 and 500 ppm. The absorption coefficient for 100 ppm of potassium ranges from 0.014 to 0.026 m^{-1}, 300 ppm it ranges from 0.014 to 0.041 m^{-1}, 500 ppm it ranges from 0.014 to 0.058 m^{-1}, and 1,000 ppm it ranges from 0 to 2.388 m^{-1}. Potassium in water fully absorbed in the visible and NIR regions There was a certain peak found at 230 nm. The absorption spectra of potassium are exponentially increasing with decreasing wavelength. There was an extreme peak in absorption found in 200 nm. It is inferred from the study that an unintended optical band gap energy broadly weakened with increasing potassium concentration. The results of this work represent that, optical constants are affected by the presence of potassium in the water. The absorption coefficient of potassium at 500 ppm, possess less correlation with the absorption coefficient derived in the field with R^2 value of 0.7. On the other hand, Absorption coefficient of 100 ppm of calcium concentrations having highest correlation with the absorption coefficient derived in the field gives R^2 value of 0.90186. **Figure 10** and **Table 1** shows the polynomial regression analysis incorporated between the absorption coefficient at the field versus absorption coefficient of potassium in the laboratory.

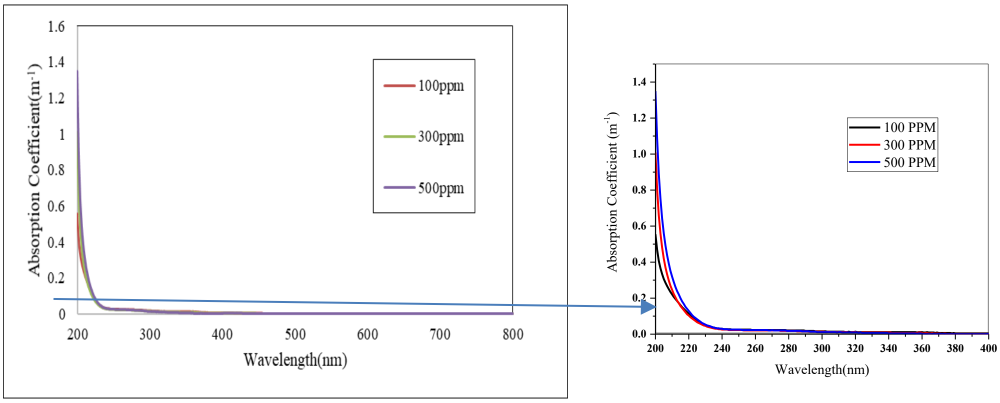
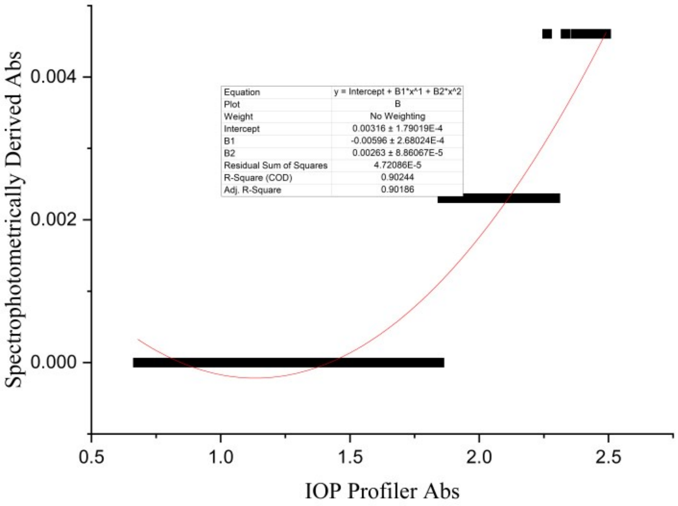


Figure 9 Absorption coefficient spectra of potassium.



Equation	y = Intercept + B1*x^1 + B2*x^2
Plot	B
Weight	No Weighting
Intercept	0.00316 ± 1.79019E-4
B1	-0.00596 ± 2.68024E-4
B2	0.00263 ± 8.86067E-5
Residual Sum of Squares	4.72086E-5
R-Square (COD)	0.90244
Adj. R-Square	0.90186

Figure 10 Regression analysis fit between field measured absorption coefficient and laboratory measured potassium absorption coefficient.

Calcium

It is highly soluble in water with molecular weight of 110.98 g/mol with density 2.15 g/cm^3; it is used to develop the solutions having higher densities. Calcium is one of the most important cations linked with hardness in the surface water. The concentration less than 75 mg/L caco3 is soft water and more than 300 mg/L referred as hard water. Cacl2 densities at different values and temperature have been identified. **Figure 11**. shows the absorption coefficient spectra of calcium developed using UV spectroscopy. The absorption coefficient for 100 ppm values ranges from 0 to 0.7 m^{-1}, for 300 ppm it ranges from 0 to 0.59 m^{-1}, and for 500 ppm 0.016 to 0.946 m^{-1}. Refractive index of 100 ppm ranges from 0.015 to 0.030 m^{-1}, for 300 ppm ranges from 0.015 to 0.027 m^{-1}, 500 ppm it ranges from 0.015 to 0.038 m^{-1} and for 1,000 ppm it ranges from 0.87 to 2.74 m^{-1}. Calcium in water absorption peak starting from 400 nm unlike sodium. There was a certain peak starting in 203 nm. The absorption spectra of calcium are exponentially increasing with decreasing wavelength as similar to sodium. Highest concentration of calcium having 0.95 m^{-1} absorption coefficient value. The absorption coefficient of calcium at 300 ppm, possess less correlation with the absorption coefficient derived in the field with R^2 value of 0.5290. On the other hand, Absorption coefficient of 1,000 ppm calcium concentrations having highest correlation with the absorption coefficient derived in the field gives R^2 value of 0.9645. **Figure 12** and **Table 1** shows the polynomial regression analysis incorporated between the absorption coefficient at the field versus absorption coefficient of calcium in the laboratory. The absorption coefficient values are increasing with the increasing concentration of calcium.

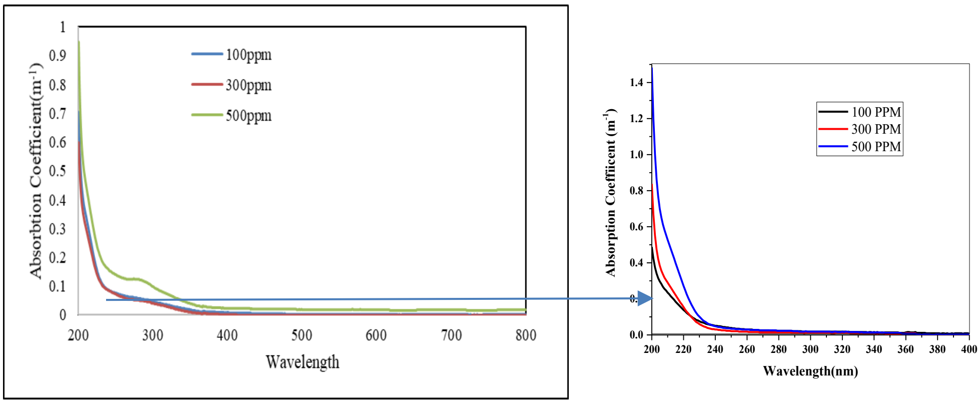
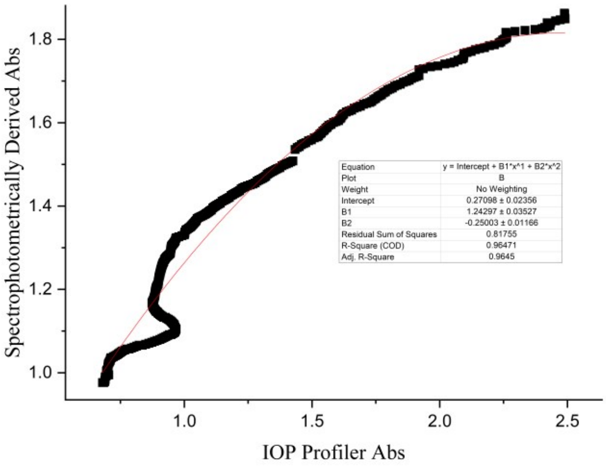


Figure 11 Absorption coefficient spectra of calcium.



Equation	y = Intercept + B1*x^1 + B2*x^2
Plot	B
Weight	No Weighting
Intercept	0.27098 ± 0.02356
B1	1.24297 ± 0.03527
B2	-0.25003 ± 0.01166
Residual Sum of Squares	0.81755
R-Square (COD)	0.96471
Adj. R-Square	0.9645

Figure 12 Regression analysis fit between field measured absorption coefficient and laboratory measured calcium absorption coefficient.

Total suspended solids

Figure 13 shows the absorption spectra of total suspended solids for various concentrations such as 100, 300 and 500 mg/L. The intensity level of light at absorption spectra is increasing with increasing concentrations of suspended particles. TSS in water having more absorption than any other chemical properties. Absorption of TSS is taking place in all region having very good reaction with the light properties There was a certain trough found in 350 nm for highest concentrations of TSS. There was an extreme peak in absorption found in 230 nm. Absorption shoulder noticed in 400 nm. The strong absorption found between 550 and 750 nm, and another between 750 and 850 nm, and it does indicate with small concentration of TSS, found to strong absorption of water. The absorption coefficient of TSS with concentration of 100 ppm ranges from 0.33 to 0.88 m^{-1}, 300 ppm ranges from 1.20 to 2.49 m^{-1}, 500 ppm ranges from 1.39 to 4.37 m^{-1} and for 1,000 ppm it ranges from 2.20 to 5.35 m^{-1}, and the refractive index values for 100 ppm ranges from 0.020 to 0.036, 300 ppm ranges from 0.05 to 0.19 and 500 ppm ranges from 0.06 to 2.72. By comparing with all the elements, total suspended sediments having higher density and make changes in the optical constant's values. Based on the spectra developed, an algorithm may be developed to design a sensor to predict the concentration value of TSS present in the water. The absorption coefficient of TSS at 100 ppm, possess less correlation with the absorption coefficient derived in the field with R^2 value of 0.96681. On the other hand, Absorption coefficient of 300 ppm TSS concentrations having highest correlation with the absorption coefficient derived in the field gives R^2 value of 0.98233. **Figure**

14 and **Table 1** shows the polynomial regression analysis incorporated between the absorption coefficient at the field versus absorption coefficient of TSS in the laboratory.

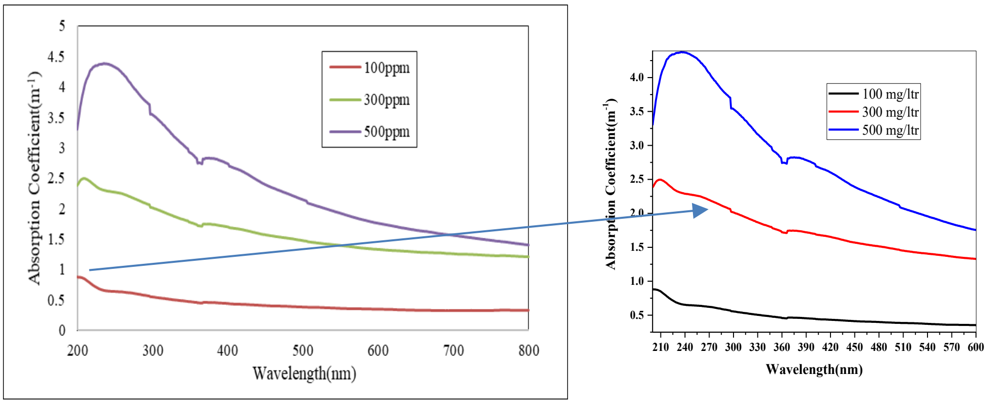
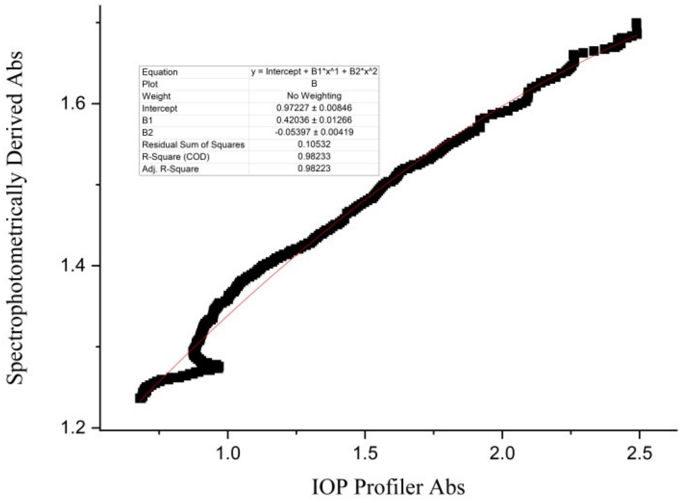


Figure 13 Absorption coefficient spectra of total auspended solids.



Equation	y = Intercept + B1*x^1 + B2*x^2
Plot	B
Weight	No Weighting
Intercept	0.97227 ± 0.00846
B1	0.42036 ± 0.01266
B2	-0.05397 ± 0.00419
Residual Sum of Squares	0.10532
R-Square (COD)	0.98233
Adj. R-Square	0.98223

Figure 14 Regression analysis fit between field measured absorption coefficient and laboratory measured TSS absorption coefficient.

Turbidity

Turbidity is the blurred vision of water caused by the suspended particles. The concentration level of turbidity may vary depend upon the size and distribution of suspended particle present in the water. The absorption curves show an evolution from a solitary, well defined absorption trough at 550 nm at lower turbidity values to a broader, more flat-topped absorption encompassing from 550 to 650 nm at higher turbidity levels. Turbidity in water having absorption properties in all regions but less compare with TSS. There was a extreme peak in absorption found in 200 nm. The absorption spectra of Turbidity are exponentially increasing with decreasing wavelength as similar to sodium, Calcium, and Potassium. The absorbance at 750 nm can be used to measure the turbidity level in water. The absorption coefficient of turbidity at 40 NTU ranges from 0.02 to 1.04, 100 NTU ranges from 0.04 to 1.8 and 400 NTU ranges from 0.092 to 2.92. On the other hand Refractive index of turbidity at 40 NTU ranges from 0.015 to 0.042, 100 NTU ranges from 0.015 to 0.092 and 400 NTU ranges from 0.016 to 0.3 which is higher than the low turbidity refractive index value. **Figure 15** shows the absorption coefficient spectra of different concentration level of turbidity produced using UV spectroscopy used to study the turbidity level. The absorption coefficient of TSS at 100 ppm, possess less correlation with the absorption coefficient derived in the field with R^2 value of 0.83389. On the other hand, Absorption coefficient of 500 ppm TSS concentrations having highest correlation with the absorption coefficient derived in the field gives R^2 value of 0.93535. **Figure 16** and **Table 1** shows the polynomial regression analysis incorporated between the absorption coefficient at the field versus absorption coefficient of turbidity in the laboratory.

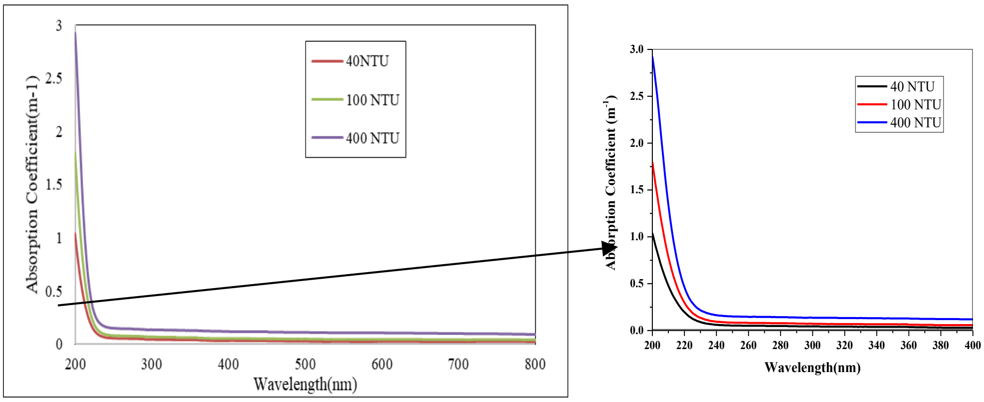
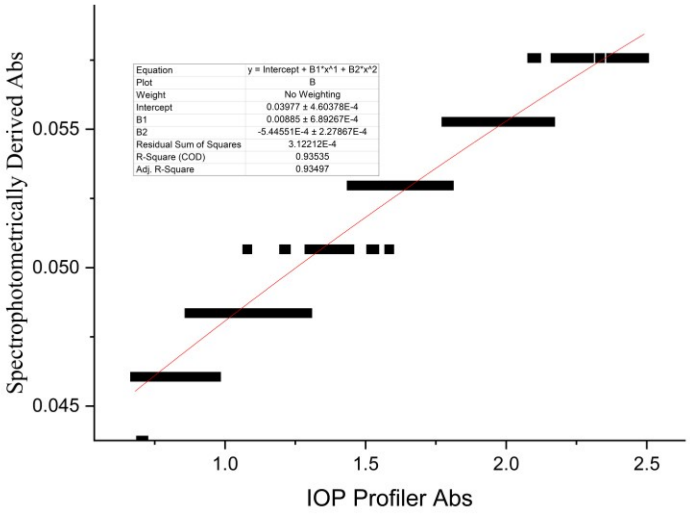


Figure 15 Absorption coefficient spectra of turbidity.



Equation	y = Intercept + B1*x^1 + B2*x^2
Plot	B
Weight	No Weighting
Intercept	0.03977 ± 4.60378E-4
B1	0.00885 ± 6.89267E-4
B2	-5.44551E-4 ± 2.27867E-4
Residual Sum of Squares	3.12212E-4
R-Square (COD)	0.93535
Adj. R-Square	0.93497

Figure 16 Regression analysis fit between field measured absorption coefficient and laboratory measured TSS absorption coefficient.

Figure 17 shows the consolidated absorption coefficient spectra of Total suspended solids, Turbidity, potassium, calcium, and sodium. The absorbance intensity level of potassium is lower than other minerals present in the water column. Alternatively, TSS produces and having higher impact on the optical absorption coefficient value. The absorption trough at 350 nm and peak at 256 nm for TSS shows the clear view of the TSS effect over optical constant and continuous absorption trough taken place after 400 nm to 800nm for the minerals present in the water. Followed by TSS, calcium provides a higher impact over optical constant as hardness having higher absorption values.

Table 1 Polynomial fit between field measured absorption coefficient and laboratory measured water quality parameters absorption coefficient.

Sl. No	Water quality parameters	Linear equation	R^2	Residual sum of squares
1	Sodium (1,000 ppm)	$\mathrm{Abs}_{\mathrm{sodium}(1,000\ \mathrm{ppm})} = -0.00203 + 0.000277*\mathrm{Abs} - (0.00218 * \mathrm{Abs})^2$	0.90764	0.0004168
2	Potassium (100 ppm)	$\mathrm{Abs}_{\mathrm{Potassium}(100\ \mathrm{ppm})} = 0.00316 - 0.00596* \mathrm{Abs} + (0.00263 * \mathrm{Abs})^2$	0.90186	0.0000472
3	Calcium (1,000 ppm)	$\mathrm{Abs}_{\mathrm{calcium}(1,000\ \mathrm{ppm})} = 0.27098 + 1.24297* \mathrm{Abs} - (0.25003 * \mathrm{Abs})^2$	0.9645	0.81755
4	TSS (300 ppm)	$\mathrm{Abs}_{\mathrm{TSS}(300\ \mathrm{ppm})} = 0.97227 - 0.42036* \mathrm{Abs} - (0.05397 * \mathrm{Abs})^2$	0.98233	0.10532
5	Turbidity (100 NTU)	$\mathrm{Abs}_{\mathrm{Turbidity}(100\ \mathrm{NTU})} = 0.03977 + 0.00885* \mathrm{Abs} - (0.0003122 * \mathrm{Abs})^2$	0.93535	0.0003122

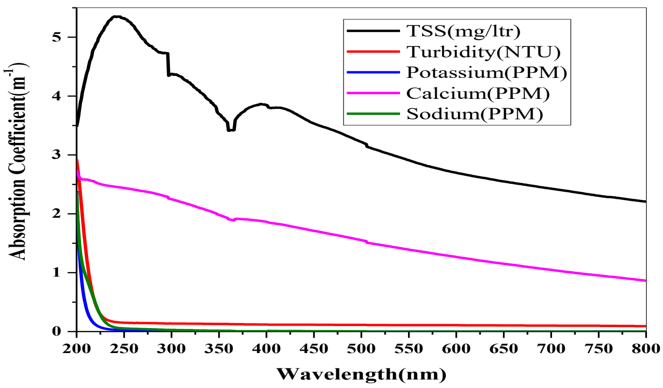


Figure 17 Comparisons of absorption coefficient spectra of different water quality parameters.

Conclusions

The optical properties of electrolyte minerals, TSS and Turbidity were studied spectrophotometrically and correlated with the field measured absorption coefficient. There were intensifying spectra visualized for TSS at all the concentrations level unlike other components which inferred that it endues strong correlation ($R^2 = 0.9823$) with the IOP profiler measured absorption coefficient followed by calcium ($R^2 = 0.9645$). The electrolyte minerals absorption coefficient spectra almost follow the similar patterns of each other and throws in resemblant optical characteristics. The absorption spectra of electrolyte minerals increase with increasing concentrations. The magnitude and shape of the standardized spectra of chemical and physical water quality parameters of each concentration compared and correlated with the atmospherically corrected satellite image derived spectra, could delineate the chemical water quality characteristics. It plays a vital role in the spatial and temporal distribution analysis through remote sensing method using empirical and analytical band models. In future, heavy metals standardized spectra also may be derived and developed.

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