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Analytical Methods

Digital imaging devices as sensors for iron determination

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ABSTRACT

Portable, sensitive and cost-effective sensors represent an unmet need, especially in resource-limited settings and locally deprived communities. Digital imaging devices can fill the gap. Thus, we have tested a desktop scanner, a digital camera and a smartphone to determine iron using three standard colour reactions. Images of reacting solutions were analysed to obtain the RGB (red, green and blue) non-uniform colour space parameters. To improve the calibration linearity, sensitivity, and detection limit, we converted the RGB intensities into six uniform colour space values and two colour differences attributes. The converted signals surpassed the RGB signals and compared well with reference spectrophotometric signals. The simplicity and sensitivity of this approach make digital imaging devices as excellent competitors to field-monitoring instruments and sophisticated spectrophotometers. Our approach was successfully applied to the assessment of iron in Nile river water, soils, plant materials and meat and liver samples.

1. Introduction

In locally deprived communities and/or resource-limited environments, digital-imaging devices can be used as simple, sensitive and cost-effective sensors for quantitative analysis in chemical, biochemical, and environmental monitoring. These imaging devices include desktop scanners, smartphone cameras, digital cameras, and webcams.

The non-uniform RGB colour space is generally used for digital image-based chemical analysis (DIBA). In DIBA, an analyte reacts with a chromogenic reagent to produce a coloured species, whose digital images are analysed to give standard RGB (red, green, and blue) intensity values (I_R, I_G, I_B, and I_{RGB}). The RGB intensities (Kudo, Yamada, Watanabe, Suzuki, & Citterio, 2017; Martinez, Phillips, Butte, & Whitesides, 2007; Rakow, Sen, Janzen, Ponder, & Suslick, 2005; Scheeline, 2010; Souza, Duarte Junior, Garcia, & Coltro, 2014) and the RGB absorbance values (log (I_{balnk}/I_{sample})) (Andrade et al., 2013; Bang-iam, Udnan, & Masawat, 2013; Barros, Oliveira, Santos, Wisniewski, & Luccas, 2017) have been described previously as sensory signals for analytical assessment. However, the RGB model is a non-uniform device-dependent model; i.e., different devices detect or reproduce a given RGB value differently; this is because the colour elements and their response to the individual RGB levels vary in the same device over time and also from one manufacturer to another (Hunt & Pointer, 2011). Thus, an RGB value does not define the same colour across devices without some kind of colour management. On the other hand, a uniform colour space is generally formed from two-chromaticity plane parameters in addition to a lightness/luminance parameter, with the major advantage of uniformity of the chromaticity plane such that at constant luminance, the colour differences represented by equal distances are perceived as being equal. (Hunt & Pointer, 2011) Therefore, conversion of RGB values into the corresponding uniform colour space parameters can be advantageous. The mathematical conversion can be performed manually or at a button click, using a variety of freeware programs (Capitan-Vallvey, Lopez-Ruiz, Martinez-Olmos, Erenas, & Palma, 2015; Hunt & Pointer, 2011). Among the well-known uniform colour space parameters, it is worthy to mention those of the CIE ($L^*a^*b^*$, Luv, LCh, $\Delta E_{L^*a^*b^*}$, ΔE_{Luv}) and the cylindrical (HSV and HSL) transformations, respectively. The previous notations stand for: CIE (Commission Internationale de l'Eclairage), $L^*a^*b^*$ (lightness/luminance/intensity, red(+)/green(−), yellow(+)/blue(−)), Luv (lightness, chromaticity values), LCh (lightness, chroma, hue angle), HSV (hue, saturation, brightness value) and HSL (hue, saturation, lightness), respectively.

During the past decade, the RGB signals were commonly used in DIBA (Andrade et al., 2013; Bang-iam et al., 2013; Barros et al., 2017; Kudo et al., 2017; Souza et al., 2014). Other less common signals including the CIE-$L^*a^*b^*$ (Baş, 2017; Bueno, Valdez, Gutierrez Salgado, Marty, & Munoz, 2016; Khimchenko & Eksperiandova, 2012; Komatsu et al., 2016), Hunter Lab (Condés, Añón, Dufresne, & Mauri, 2018; Cubero, Albert, Prats-Moltalbán, Fernández-Pacheco, & Blasco, 2018), CIE-Luv (Cheng et al., 2014; Schwaebel, Trapp, & Bunz, 2013), CIE-LCh (Giusti, Caprioli, Ricciutelli, Vittori, & Sagratini, 2017; Mendez-Cid,

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Lorenzo, Martinez, & Carballo, 2017; Tuberoso et al., 2014), HSV (Ahirwar, Tanwar, Parween, Kumar, & Nahar, 2014; Erenas, de Orbe-Paya, & Capitan-Vallvey, 2016), HSL (Cantrell, Erenas, de Orbe-Paya, & Capitan-Vallvey, 2010; Ivanov, Samarina, & Figurovskaya, 2010; Paciornik, Yallouz, Campos, & Gannerman, 2006), $\Delta E_{L^*a^*b^*}$ (Khimchenko & Eksperiandova, 2012; Suzuki et al., 2002), and ΔE_{Luv} (Cheng et al., 2014), have been also reported. In these reports, images have been captured using computer scanners (Ahirwar et al., 2014; Khimchenko & Eksperiandova, 2012; Kudo et al., 2017; Paciornik et al., 2006), mobile phone cameras (Baş, 2017; Scheeline, 2010) digital cameras, (Cheng et al., 2014; Erenas et al., 2016; Komatsu et al., 2016), web cams (Andrade et al., 2013; Bang-iam et al., 2013; Barros et al., 2017), CMOS (complementary metal-oxide semiconductor) modules (Bueno et al., 2016), and colorimeters (Giusti et al., 2017; Mendez-Cid et al., 2017; Tuberoso et al., 2014). However, some other excellent papers on the topic have been undoubtedly missed and those might have be found in featured reviews of the topic (Apyari, Gorbunova, Isachenko, Dmitrienko, & Zolotov, 2017; Capitan-Vallvey et al., 2015; Morbioli, Mazzu-Nascimento, Stockton, & Carrilho, 2017; Yamada, Henares, Suzuki, & Citterio, 2015).

Despite the above-mentioned progress, the direct application of RGB signals in quantitative determinations is characterized by the poor linearity of its calibration graphs, especially for trace analyte concentrations encountered in most environmental samples (Andrade et al., 2013; Bang-iam et al., 2013; Barros et al., 2017; Kudo et al., 2017; Souza et al., 2014). Other limitations may include, the use of optical elements; e.g., wavelength selectors, signal amplifiers, and/or the relatively high-cost of commercially available imaging-based colorimeters/spectrophotometers (Giusti et al., 2017; Mendez-Cid et al., 2017; Scheeline, 2010; Tuberoso et al., 2014). Moreover, the individual use of the hue (Erenas et al., 2016; Giusti et al., 2017; Mendez-Cid et al., 2017; Tuberoso et al., 2014), chromaticity (Giusti et al., 2017; Mendez-Cid et al., 2017), or luminance (Giusti et al., 2017; Mendez-Cid et al., 2017) parameters of LCh, HSV and HSL could not be generalized for quantitative analysis of various coloured reaction systems, using simple, low-cost platforms. Therefore, the adoption of a more simple, sensitive, cost-effective, and generalized signalling tool that is linearly responsive to various coloured systems without any need of optical elements/signal amplifiers is highly desirable.

Herein, we describe the design, validation, and application of a simple homemade platform for the assessment of iron, as a model pollutant, in complex environmental samples, including Nile river water, soils, plant materials, meat and liver samples. The iron content was determined based on its reaction with 1,10-phenanthroline, 2,4,6-tris(2-pyridyl)-s-triazine and salicylate, respectively. Our simple and cost-effective design is free from any lens, slit, mirror, wavelength selector, diode, photomultiplier, and signal amplifier. The platform consisted of (1) two cells or test tubes, (2) a digital imaging device as a sensor, and (3) a white paper as a diffuser. For simplicity, we adopted a digital camera as a model-sensing device. However, a smartphone camera and a desktop scanner have been also used. The current work compares for the first time the analytical performance of seven colour spaces RGB, Hunter-LAB, CIE-$L^*a^*b^*$, CIE-Luv, CIE-LCh, HSV, and HSL in addition to two colour-difference parameters, $\Delta E_{L^*a^*b^*}$ and ΔE_{Luv}, as quantitative generalized signalling tools. The obtained $\Delta E_{L^*a^*b^*}$ and ΔE_{Luv} data surpassed the commonly used RGB parameters regarding the calibration graph linearity, limits of detection (LOD), limits of quantification (LOQ), and the more generalized response to various coloured reactions and also compared well with the corresponding data of a sophisticated Shimadzu 1601 PC spectrophotometer. The platform design, optimization, application, and validation have been thoroughly investigated and incorporated into the recommended procedure.

2. Experimental

2.1. Reagents

All reagents were of ACS grade or equivalent and were used as received from Sigma-Aldrich (St. Louis, MO, USA), or Merck (Darmstadt, Germany). Ultrapure water and aqueous solutions were used throughout. 1,10-Phenanthroline (Phen), 2,4,6-tris(2-pyridyl)-s-triazine (TPTZ), sodium salicylate, ferrous ammonium sulfate hexahydrate, ferric ammonium sulfate dodecahydrate, and L-ascorbic acid were also used. Standard solutions for the determination of iron using phenanthroline ("EPA Method 315B," 1996; Marczenko & Balcerzak, 2000; "Method 3500-Fe B in Standard Methods for The Examination of Water and WasteWater," 2011), TPTZ ("AOAC Methods 937.03, 970.13," 2000; Collins, Diehl, & Smith, 1959), and salicylate (Mitchell-Koch, Reid, & Meyerhoff, 2008) were prepared according to the established standard/reference methods.

2.2. Instruments and software

Spectrophotometric measurements were performed on a Shimadzu 1601PC UV/VIS Spectrophotometer controlled by the UVProbe-2.5 software (Kyoto, Japan). Eppendorf 10 to 100 and 100 to 1000-µL variable-volume pipettes (Westbury, NY, USA) and a calibrated pH meter (EDT, Dover Kent, UK) were also used. Decomposition of food and plant samples was performed using a microwave ETHOS-1600 closed digestion system (Milestone, Italy).

DIBA measurements were performed on a homemade, simple platform consisting of a Canon PowerShot A810 camera with a 16.0 MP sensor as an image-capturing device, two cuvettes mounted on a 3D-printed cell holder and a white cartoon paper as a background diffuser. These three components were fixed on the same line on a 20 × 20 cm wood plate; where each of the camera and the diffuser were set at a 5 cm distance from the cell holder, as shown in Fig. 1. We have also tested a 3.15 MP low resolution camera of a Sony Xperia E mobile phone, and the HP scanner of the M1536dnf all-in-one printer as alternative imaging devices. Images were captured on the bench of our laboratory with the conventional fluorescent daylight lamp fixed to the ceiling serving as the light source. For DIBA measurements using the desktop scanner, the cuvettes were replaced by a flat-bottomed 96-microwell plate.

2.3. Recommended procedure

2.3.1. River water and soil samples treatment

Water samples were collected and filtered through a 0.45 µm membrane filter and acidified to pH < 2.0 using concentrated HCl

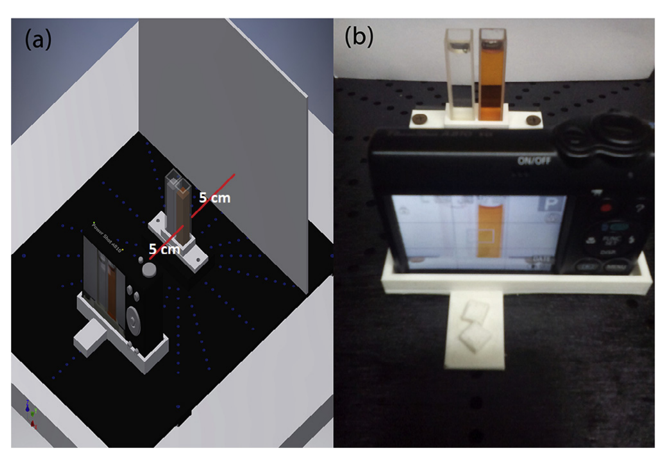

Fig. 1. The homemade platform design.