Internet of Things as a Tool for Sustainable Analytical Chemistry: A Review

Alegre N. S. Cadeado,^{a,b} Caio C. S. Machado,^a Geandre C. Oliveira,^{®a} Daniela A. S e Silva,^a Rodrigo A. A. Muñoz^{®a} and Sidnei G. Silva[®]*^a

^aInstituto de Química, Universidade Federal de Uberândia, Avenida João Naves de Ávila, 2121, Santa Mônica, 38400-902 Uberlândia-MG, Brazil

^bDepartamento de Ciências e Tecnologia, Universidade Licungo, Quelimane, Moçambique



Sustainable analytical methods are highly demanded in the modern society. Within the green principles, novel procedures that attend the sustainable development goals have been proposed and the internet of things (IoT) can play key role to achieve this goal. The association of IoT with analytical chemistry enables the real-time obtaining of analytical data to control industrial processes as well as for monitoring different environmental scenarios and human health by accessing remotely analytical information. In this context, this review presents the main IoT technologies used for analytical chemistry as well as a tutorial for beginners in the field.

Keywords: do it yourself, sensors, Arduino, Raspberry Pi, automatization

1. Introduction

In September 2015, the United Nations adopted a resolution for sustainable development entitled "transforming our world: the 2030 Agenda for Sustainable Development". In this resolution, an enthusiastic world plan was shown for taking action by 2030 to tackle the global challenges of environmental degradation and poverty.¹ There are 17 universal goals for 2030, each providing guidelines and targets for all countries, industries and organizations to adopt in line. Behind the 17 goals are a set of 169 specific targets which describe, in detail, the ways in which the goals may be measured and achieved.² Analogous to the Sustainable Development Goals (SDGs), the Sustainable Chemistry concept may serve as a way to accomplish these objectives for a considerable number of targets for chemicals and waste management.^{3,4} From this perspective, analytical chemistry may contribute to the target SDGs, facilitating data collection and monitoring.⁵ This typically requires the development and application of chemical analytical devices which integrate industry, manufacturing, chemicals, transportation and laboratories according to the internet of the things (IoT) principles.^{6,7} The internet of things remain an influential concept referring to a range of technology and concerns the interconnection of

*e-mail: sidgon@ufu.br Editor handled this article: Teodoro S. Kaufman machines and computing devices through the infrastructure of internet. Modern technologies such as IoT offer a wide number of applications such as water and energy management, environmental monitoring, healthcare and chemistry. The IoT demonstrates the potential to address some of the most acute human, economic and environmental needs. It can also directly contribute to achieve the targets in the Sustainable Development Goals.

Therefore, analytical chemistry working together with IoT devices can provide data for the monitoring of different analytes. Furthermore, the design of portable analysis devices facilitates for *in situ* analysis, which generally require low volume of the sample and reagents that contributes to "zero waste" generation. A new generation of small, lowpower computers and microcontrollers boards systems are positioned to play a key role in the development of wireless and remote-controlled system for analytical chemistry.

Frequently applied in do it yourself (DIY) projects, the IoT consists of connected board embedded with sensors, software, step motors among other technologies. IoT devices provide easy affordable alternatives to sense and measure chemical and physical properties. The most typical applications involve environmental and healthcare monitoring. In this context, open-source contributions to the production of communications resources (software and hardware) increase the opportunities for democratization of production, governance and knowledge exchange. The most popular, open-source, platforms are Arduino and Raspberry Pi boards, designed for building electronics devices projects for analytical chemistry purposes. Arduino was developed in Italy in 2005 and it became popular due to its low cost, for running on different platforms (Windows, Mac, Linux), and simplicity for programming it. Raspberry Pi was created specifically for promoting computer science teaching to elementary school students.

A revision about internet of things for environmental analytical chemistry has been reported by Capella et al.⁸ A review about the use of biosensors and chemosensors within the analytical chemistry field, exploiting IoT integration, has also been showed.9 The authors suggested the integration between IoT technologies can generate an internet of analytical things (IoAT). The focus of this review is to feature sensor-based devices for IoAT applications, highlighting the use of wearable sensors. All the review papers about internet of things and analytical chemistry in the literature do not focus on the interface, platform board, and other necessary peripherals for implementing IoT devices. In this context, this review aims to fill this gap by providing a comprehensive review of the field, highlighting the use of microcontrollers and microcomputer available to date with their respective advantages and drawbacks, device designs and modifications to accommodate different assay needs, detection strategies and the growing applications of internet of things. Finally, we discuss how the field needs to continue moving forward to achieve its maximum potential. We want to emphasize the groundbreaking work done previously before the term of IoT devices was even invented and has inspired later research groups to find their attractive analytical devices. This includes publications with concepts relevant to all of the IoT's devices, such as microcontrollers, small PC boards and some modules and sensors.

2. Scientometrics

The publications of scientific papers in the field of internet of things have increased significantly in recent years (Figure 1). More than 41,000 works have been published in the last 10 years. Analytical chemistry is one of the most established fields in internet of things and accounts for 5.79% of the papers and received more than 24,000 citations. Most of these papers have been published in journals with high impact factors; they are mostly published in the area of sensors and biosensors. However, many of these papers involving IoT's devices do not necessarily present analytical applications to our knowledge. Many works involve the development and applications of smart sensors for industrial and domestic use, such as humidity and temperature monitoring, among others.

The term "internet of things" was cited by the first time¹⁰ in 2013. The authors developed a wireless sensor

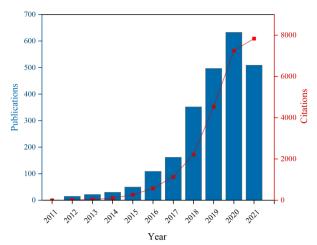


Figure 1. Evolution of publications and citations of analytical chemistry in the field of internet of things.

tag with potentiometric input for use with pH and ionselective electrode. The sensor is able to autonomously measure, store electrode potential, then transfer logged data wirelessly, via radio frequency identification (RFID) to a nearby reader or by near field communication (NFC) to a smart phone. The wireless interface of the smart tag was built employing an instrumentation amplifier as the electrode interface (Texas Instruments Inc., Dallas, USA) and the electronic circuits were designed using schematic capture and printed circuit board design tools. The main contributions of internet of things devices for analytical chemistry, highlighting wireless apparatus (Bluetooth and Wi-Fi) and the use of Arduino, including applications, are shown in Figure 2 as a timeline.

3. Devices

To develop an IoT device for chemical analysis requires to choose the development platform. Singleboard microcontrollers and computers are frequently used platforms for IoT projects. A single-board microcontroller is a single integrated chip on-board that contains all of the circuitry necessary for a useful control task: a microprocessor, I/O (input/output) circuits, a clock generator, RAM (random access memory) or ROM (readonly memory) memory, any necessary support integrated circuit (ICs). The intention is that the board is immediately useful to an application developer, without requiring them to spend time and effort to develop controller hardware. On the other hand, a single-board computer is a complete computer containing a microprocessor, memory, I/O circuit and other structures. Some of them include Raspberry Pi,¹¹ The Beagles¹² (BeagleBoard, BeagleBoard-xM, BeagleBone, BeagleBone Black), MK802 and MK808,13 Cubieboard,¹⁴ MarsBoard,¹⁵ Udoo¹⁶ and others. In this

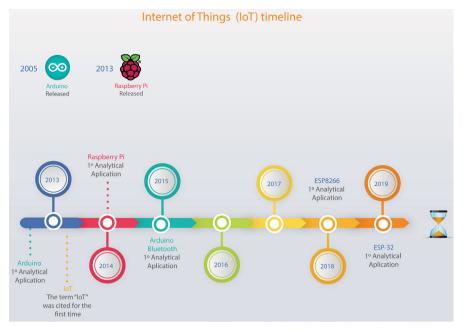


Figure 2. Timeline showing the main internet of things (IoT) contributions.

way, with everything embedded on a single board, without double, a microcomputer has higher performance capabilities than a single-board microcontroller.

The two most popular microcontroller and microcomputer single-board are Arduino¹⁷ and Raspberry Pi,¹¹ respectively. Arduino is based on the ATmega family microcontroller, and has an easier time interfacing with different hardware's, such as sensors, motors, or other devices, while the Raspberry Pi has a more complicated path requirements to interfacing sensor readings, it is often necessary a middle-level programming languages knowledge. However, both have their advantages and disadvantages. If you have little knowledge of programming IoT devices, Arduino is the best choice, because the setup is really easy to manipulation, you will need to plug a USB (universal serial bus) cable, connect the board to a computer, download the software from the Arduino website, and you are ready to go. Raspberry Pi requires before starting it, an SD (secure digital) card for the operational system, some computer peripherals such as a keyboard and a mouse, an HDMI (high-definition multimedia interface) screen and cable. You will also need to install the right operating system on the SD card. In terms of connectivity, Raspberry Pi has on-board connectivity (Bluetooth and Wi-Fi), while Arduino needs additional modules to obtain this type of wireless communication. Finally, to summarize that brief comparison, Raspberry Pi has inputs and outputs ports, general purpose input/output (GPIO), and supports the I²C (inter-integrated circuit) backpack and serial peripheral interface (SPI), however, these are all digital ports. Analog inputs can easily get on the Raspberry Pi by

using analog-to-digital converters. Some Arduino boards, such as Arduino Uno, has digital inputs and outputs and also, analog inputs.

The publications of scientific papers and citations of devices based on Arduino and Raspberry boards, for analytical chemistry applications, follow the trend of increasing publications and citations related to the internet of things (Figure 3), which demonstrates the popularity and ease of implementation of these devices. Likewise, publications involving wireless communications via Bluetooth and Wi-Fi have been growing (Figure 4).

In order to help beginners to easily get started on building a prototype based on Raspberry Pi and Arduino, from scratch, we here outline some guidance. Table 1 summarizes websites, online lessons, e-books and places for buying electronic components, including some sensors. We encourage you to read some recommended papers aimed at implementing IoT devices for analytical chemistry.³³⁻³⁸

4. Arduino Based Devices

Arduino board designs are an open-source equipped with sets of digital and analog input/output (I/O) pins that may be interfaced to various expansion boards and can be programmed using the C and C++ programming languages. Arduino based devices have become increasingly useful in the field of analytical chemistry due to their low cost and integrated development interfaces. The first analytical device use of Arduino was proposed in 2013³⁹ for a design and construction of a three-dimensional (3D) printable,

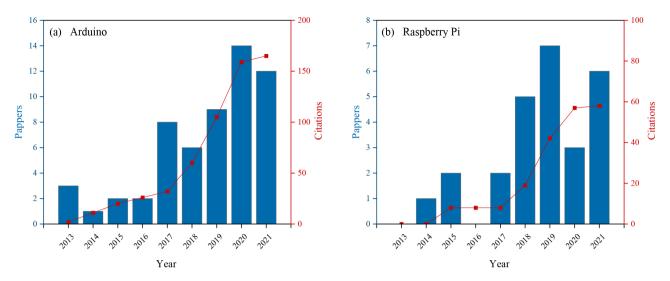


Figure 3. Evolution of publications and citations of analytical chemistry in the field of (a) Arduino-based boards and (b) Raspberry Pi-based.

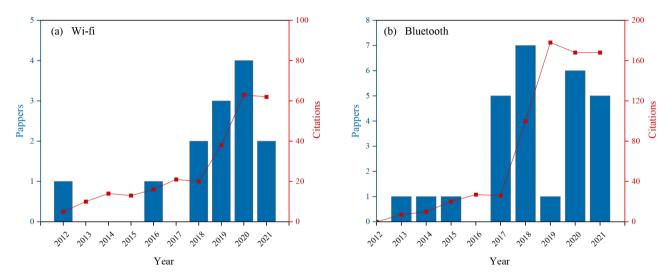


Figure 4. Evolution of publications and citations of analytical chemistry in the field of (a) Wi-Fi and (b) Bluetooth communications.

Type of resource	Remarks	Reference
	trial account in order to download books and video	18
Books and video lessons	a free book for Arduino	19
	inspired by the teachings of prototyping laboratory	20
	a list of free Raspberry Pi e-books	21
	with some free courses	22
Tutorial	with some free courses	23
Where to buy		24
		25
		26
		27
		28
		29
Forum and code		30
library		31
		32

open-source colorimeter exploiting open-source hardware and software. A light-emitting diode (LED) with an emission peak near 606 nm was used as a light source and a TSL230R light sensor was used as light detector. Device performance was evaluated for chemical oxygen demand determination.

A total of 228 publications on Arduino, published between 2012 and 2021 (with the number of publications increasing annually), were retrieved from the database. Original research articles were the most common type of publication. After an analysis of the 228 articles, it was found that only 57 were devoted for analytical chemistry purposes, with a total of 586 citations. Different detections strategies were exploited in these papers.

Also, it is quite true that these analytical applications are not necessarily IoT devices, although some of these have potential for implementation. In this case, it would be important to consider the incorporation of a wireless communications protocols. Several different wireless communication technologies and protocols can be used to connect the smart device such as Internet Protocol version 6 (IPv6), ZigBee, Bluetooth Low Energy (BLE), NFC, among others. To improve wireless data communication controller, a wireless module could be attached. Usually, wireless based modules are cheap and easy to interface with microcontroller or other components and shows a low power consumption. Here are some Arduino compatible modules examples: ESP8266⁴⁰ (Wi-Fi), ESP32⁴¹ (Wi-Fi and Bluetooth), HC-05⁴¹ (Bluetooth), RFID NFC PN532⁴¹ (NFC) and Xbee (ZigBee).⁴¹

We selected some Arduino papers to help DIY enthusiasts and encourage new laboratory improvement for chemical analysis. We recommend some articles presented in the literature, all of them exploring IoT devices for analytical measurements. A device for data acquisition to collect voltage signals³⁴ and data acquisition systems based on analog-to-digital converter modules for microscale separation systems (e.g., electrophoresis) are some of opensource Arduino based examples for analytical chemistry. Selected contributions of IoT devices employing Arduino boards for analytical chemistry applications are presented in Table 2.

Optical based techniques are the most prevalent Arduino-based detection utilized, because of its easy-to-use hardware and software, portability and robustness, with a range of official commercially available sensors, modules and including boards, modules and optical transducers.

Photometry Arduino-based measurements exploited the use of a LED as a light source.^{42,44-47} Usually, the Arduino

Table 2. Selected contributions of IoT devices employing Arduino, Bluetooth and Wi-Fi boards for analytical chemistry applications

Wireless connection	Technique	Analyte	Board	Reference
_	photometry	Zn ^{II}	Arduino Due	42
_	photometry	SO ₄ ²⁻	Arduino Due	43
-	photometry	NO_3^-	Arduino Mega	44
	photometry/colorimetry	antioxidant (tea infusion)	Arduino	45
HC-05	photometry	oxalate	Arduino	46
-	photometry/fluorimetry	orthophosphate	Arduino Mega 2560	47
	fluorimetry	rhodamine dyes	Arduino	48
-	fluorimetry	Hg ²⁺	Arduino	49
-	fluorimetry	Cu ²⁺	Arduino Uno R3	50
-	fluorimetry	quercetin (QC-Y ³⁺)	Arduino	51
-	fluorimetry	HCN	Arduino Mega	52
	fluorimetry	2,4-DNT	Arduino	53
-	fluorimetry and colorimetry	Fe ^{III} , Co ^{II} , Hg ^{II} , Sn ^{II}	Arduino Uno	54
-	colorimetry	Ni ^{II}	Arduino Uno R3	55
-	colorimetry	cyanide	Arduino Uno	56
-	colorimetry	Pb ²⁺	Arduino Uno	57
-	chemiluminescence	H_2O_2	Arduino Due	58
-	laser-scattering	proteins	Arduino Leonardo	59
-	photothermal	Fe ^{II}	Arduino A Mega 2560	60
HC-05	amperometry	uric acid, hydrochlorothiazide, ascorbic acid	Arduino Nano	61
Bluetooth module	voltammetry	dopamine	Arduino Mega 2560	62
-	voltammetry / chronoamperometry	H_2O_2	Arduino Uno	63
ESP32	voltammetry/amperometry	glucose	ESP32	64
Wi-Fi module	voltammetry	ferricyanide	Arduino Uno	65
HC-06	impedance	volatile organic compounds	Arduino board	66
HC-05	impedance	prostate specific antigen	ATmega328P	67
HC-06	impedance	2,4,6-trinitrotoluene	Arduino uno	68
	electroanalytical	ferricyanide	RFduino	69
_	electrophoresis	K+, Ca2+, Na+, Mg2+, Li+, NH4+	Arduino Nano	70

board are used to control the power and activation of the light source. The use of optoelectronic devices can provide a natural filter because they operate within a specific range of wavelength and easy integration with Arduino boards. Optical sensors TSL230R light³⁹ and TSL2591,⁴⁶ photodiodes,^{42,44} photoresistors⁴⁵ and a LED⁴⁷ were used as a photosensor with satisfactory results.

Arduino-based devices also was developed for fluorescence measurements. They require a light source. and LEDs were more extensively used due to their easy integration with the Arduino, stable monochromatic illumination, low power and low voltages requirements, develop little heat, and are small in size. These papers describe integrated fluorescence detection systems that uses LED,⁴⁷ digital camera,⁴⁸ photodiode,^{51,53} light sensor^{49,50} and RGB sensors^{52,54} as detectors. RGB sensors have a white LED, responsible for ambient lighting. The detector consists of a photodiode color array that comprises a 4×3 array of red, green, and blue-filtered photodiodes. The RGB sensor, in principle, operates as an emission filter for fluorescence measurements, eliminating light scatter and background emission of the sample. Also, the sensor has in on single board the excitation source and the detection system, simplifying the integration with the Arduino board. Colorimetric Arduino-based devices have been successfully developed based on the use of RGB color sensors,55-57 interfaced with Arduino board. These sensors respond linearly to color variations, providing signals in the RGB scale.

Chemiluminescence detection was performed in a flow-based system for hydrogen peroxide determination.58 An Arduino Due board was used to control the propulsion system (solenoid pumps) and signal acquisition, by the use of a photodiode. Turbidimetric and nephelometric measurements for total protein determination in body fluids was proposed by Strzelak et al.,59 who showed a 3D printed silicone microfluidic with simultaneous turbidimetric and nephelometric measurements for the determination of total protein in body fluids. The microfluidic system consists of a two-channel syringe pumps, responsible for the injection of the sample, reagent, and carrier solution. The detection system consists of a laser (650 nm) and two RGB sensors, which one sensor was placed at the angle of 90° and the other one at 180° for nephelometric and turbidimetric measurements, respectively.

Other optical-based Arduino devices included a tunable multireflection thermal lens spectrometer.⁶⁰

Arduino-based potentiostats for electroanalytical measurements have been reported in the literature.⁶¹⁻⁶⁹ A potentiostat is an analytical instrument in which the electronic hardware is based on the control of the potential

difference between two electrodes, working and reference electrodes. A third electrode, a counter electrode, is usually added to the device in order to isolate the electrode used as a potential reference (so current flows from counter to working electrode or vice versa). A basic potentiostat consists of electronic components, such as capacitors, operational amplifiers and resistors, used to assemble an electronic circuit capable of generating and measuring potentials and currents. Arduino-based potentiostats use an Arduino microcontroller board for two purposes, to control electrochemical parameters (e.g., potential) and for data acquisition (current, amperometric signals, etc.). Digital-toanalog converters (DACs) and analog-to-digital converters (ADC) are important building blocks which interface electrochemical devices used for the measurements to digital systems, such as Arduino boards. An ADC takes an analog signal and converts it into a binary one, while a DAC converts a binary signal into an analog value. Both converters, DAC and ADC, are commercially available as modules converts. An Arduino module integrates all the necessary components and connections that are required for a device to work and presents you with a ready-to-use product, facilitating integration and its use in electronic devices.65,69

Agustini et al.61 developed an Arduino-based miniaturized electronic, allocated in a 3D printed platform, to support three different electrodes: graphite electrodes (for the determination of uric acid); carbon-printed electrode (for the determination of hydrochlorothiazide) and gold electrode (for the chromatographic separation and electrochemical determination of ascorbic acid and epinephrine). The devices were based on the use of operational amplifiers and an Arduino board, allowing the application of potentials between 0.0 and 5.0 V and electric current readings up to 4.5 µA. As power source two 9 V batteries were used. The signals were transmitted by USB connection to computer or via Bluetooth to a smartphone. Shen et al.⁶² reported the use of screen-printed electrodes as a sensor for dopamine detection and the electrochemical determination using a smartphone-based potentiostat. The circuit is mainly composed of a microcontroller module, Arduino Mega, used to control an external DAC to apply a specific analog potential signal to external electronic analysis electrodes. The resultant digital signal was acquired and stored via an ADC. An Arduino-based potentiostat for hydrogen peroxide determination was proposed by Gao et al.63 In this work an Arduino-based potentiostat performed chronoamperometry and cyclic voltammetry measurements. Using an external DAC module, a step voltage from -1 to 1 V was generated. A glassy-carbon electrode was used as working electrode,

an Ag/AgCl/saturated KCl electrode as a reference electrode, and a platinum electrode as a counter electrode. An integrated biosensor platform for glucose detection was related in the literature.⁶⁴ The device was based on a wireless potentiostat, capable of cyclic voltammetry and amperometry measurements and an ESP-32 microcontroller was used to control the potentiostatic and for data acquisition.

ESP32 is a low power Wi-Fi enabled microcontroller, and can be programmed using many different programming languages, for example, Arduino IDE.

Also, Arduino-based potentiostats for impedance measurements were reported in the literature.⁶⁶⁻⁶⁸ These works related the use of an impedance converter (such as an AD5933 module) with a high precise system that combines an on-board frequency generator and an ADC converter. The frequency generator allows an external impedance to be excited with a known frequency. The AD5933 also contains an internal temperature sensor, a 3.0 V reference to act as a stable supply voltage (to separate analog and digital sections of the device) and an ultrahigh precision regulator. These works reported the detection of volatile organic compounds,⁶⁶ prostate specific antigen⁶⁷ and 2,4,6-trinitrotoluene.⁶⁸

5. Raspberry Pi Based Devices

The Raspberry Pi is a low-cost, single-board microcomputer developed by the Raspberry-Pi Foundation. The use of open-source microcontrollers and microcomputers quickly became popular in the electronics and robotics community, and has been used in various scientific applications. In teaching labs, these devices allow students to gain hands-on experience in building and programming instrumentation used in class projects. In analytical chemistry it has become increasingly common to use these devices for various purposes, such as for construction of a photometer, acquisition of analytical signals and automation. Due to its low cost as well as its small size and diversity of resources, Raspberry is used in the development of portable analysis instruments and with the possibility of integrating data acquisition and device control in a single system.

We analyzed individual publication profiles of over 136 papers whose first record paper appeared in the Web of Science database between the years 2014 and 2021. These works provide the means for the development of wireless, remote controlled, simple and fast analysis, with low-cost chemical procedures. Also, these developments are already assisting in improved point-of-care, clinical diagnosis and environmental monitoring, and in future will facilitate low-cost, more frequent monitoring of occupational health and the environment. Given the uptake of IoT devices worldwide, these developments should not be restricted to developed nations, but will also be beneficial to developing countries.

The earliest record of literature about the use of Raspberry Pi in analytical chemistry indexed in the Web of Science appeared in 2014.⁷¹ After an analysis of the 136 articles on Raspberry Pi, it was found that only 22 were devoted for analytical chemistry purposes, with a total of 202 citations. Different detections strategies were exploited in these papers. Selected contributions of IoT devices employing Raspberry for analytical chemistry applications are presented in Table 3.

Owing to the wide range of accessories available, smallsize properties and low-cost, Raspberry Pi devices based on optical measurements have been used as a splendid alternative for improvement of IoT analytical platforms. Photometric measurements are based on the use of LED as light sources for absorption photometric detection and a camera module, connected to the Raspberry Pi board, used as detector. In this case, both setups are directly connected to the board and could be controlled by the Raspberry Pi by the use of libraries.^{78,79} Tok et al.⁷⁷ developed a device employing fiber optics to transport the light directly to the detector (Raspberry Pi Camera), for the detection of E. coli and total coliform based on absorption and fluorescence measurements. Capable of performing automated detection, the device shows more sensitive than manual count procedure, with a total cost of approximately \$600. In fact, this is an embracing IoT analytical applications, involving automation, detection and environmental monitoring. Fluorescence measurements were related in the literature employing Raspberry Pi microcomputer. Chen et al.⁸¹ developed a portable ultraviolet light emitting diode (UV-LED) induced fluorescence, composed of a commercially available 370 nm LED and a spectrometer as light source and the optical detector, respectively. The developed fluorimeter was employed for routine analysis of amiloride in pharmaceutical tablets and human serum. Fluorescence fingerprints of various petroleum oil samples were obtained for sample classification.80 In this work, the authors proposed a device that uses the computational power and affordability of a Raspberry Pi, coupled with three UV-LEDs and a Pi camera as a detector in order to collect a large data set of UV fluorescence signals. Finally, phosphorescence signals were measured using a sensor system for the monitoring of the uptake of oxygen by mammalian cells as a direct indicator for the metabolism. The device includes a microfluidic chip with an integrated oxygen-sensitive phosphorescent film, heater and temperature sensor. An excitation LED and a Raspberry Pi camera were used to measure the phosphorescent signal.

Principle	Detection principle	Analyte	Raspberry Pi model	Reference
		Optical technique		
Enzyme kinetic constants calculated directly from paper	µPad color intensity detection (Raspberry Pi camera)	-	Raspberry Pi 2 model B	72
Nanohole array-based biosensing platform	surface plasmon resonance imaging (SPRi)	Escherichia coli	Raspberry Pi 3 model B	73
Electronic eye (EE) for qualitatively discriminating of tequila	image sensors	-	Raspberry Pi 3 model B	74
Point-of-care testing using a smartphone and a Raspberry Pi	image sensors	progesterone	Raspberry Pi 3 model B	75
Microfluidic biosensor	image sensor	Salmonella typhimurium	Raspberry Pi 4B	76
Enzymatic method based on Colilert reagent	colorimetry and fluorometry	<i>Escherichia coli</i> and total coliform	Raspberry Pi 3 model B	77
Antioxidant capacity measurements by DPPH assay	photometry	antioxidant activity	Raspberry Pi B+	78
Different approaches for data acquisition using the Raspberry Pi camera	photometry	reactive oxygen metabolites	Raspberry Pi 3 model B	79
Predict some physiochemical content for petroleum classification	fluorescence	-	Raspberry Pi 3B+	80
Amiloride fluorescence emission after UV-LED excitation	fluorescence	amiloride	Raspberry Pi 3 model B	81
Sensor system to monitor the uptake of oxygen by mammalian cells	phosphorescence	oxygen	camera Raspberry Pi	82
		Electroanalytical		
Nickel modified screen- printed electrode	amperometry	cortisol	_	83
Pt-decorated WO ₃ nanoparticles onto an electrode pattern	metal oxide semiconductor sensors	H_2	Raspberry Pi 2	84
Detection of volatile compounds	electrochemical gas sensors	ammonia, amines, alcohol, CO_2 and acetone	Raspberry Pi	85
		Separation technique		
Monitor the spectral separation of food dyes of different colors	Office Chromatography (OC) - OCLab2	steviol glycoside	Raspberry Pi 4B	86
Monitoring of dynamic chemical processes	GC-MS	volatile organic molecules	_	71
Miniaturized capillary flow- through deep-UV absorbance detector	briefcase-sized portable capillary HPLC system	13 organic compounds	Raspberry Pi 3 model B	87
A low-cost, open-source digital strip chart recorder for chromatographic detectors		-	Raspberry Pi 3 model B	88

Table 3. Selected contributions of IoT devices employing Raspberry Pi boards for analytical chemistry applications

DPPH: 2,2-diphenyl-1-picrylhydrazyl; UV-LED: ultraviolet light emitting diode; GC-MS: gas chromatography-mass spectrometry; HPLC: high performance liquid chromatography.

Microfluidics with colorimetric detection have played an increasing role in the analytical chemistry due to its significant strengths, such as less reagent and sample requirements, automatic operation and portability. Those devices require images to be captured with a digital camera. After the images are collected, they are subjected to an image processing, by the use of a software, improving their quality, extracting some sample characteristics required for specific computational algorithms for their interpretation. In this sense, image sensors based on Raspberry Pi are presented in the literature.⁷²⁻⁷⁷ LEDs were used as light source and a camera module (Pi camera) were used to capture the imagens. These papers showed relevant for point-of-care applications, especially for regions around the world where accessibility to medical facilities is heavily dependent. Those IoT devices enables onboarding, configuration, overall management of IoT devices, data manipulation and also, be done using cloud platforms.

Connecting an Arduino board to Raspberry Pi, some advantages are achieved with unique capabilities, especially if there are sensors, motors, and actuators. In this case the Arduino can send values to and from the Raspberry Pi, sorting out the computing intensive tasks (done by the Raspberry Pi) and controlling tasks (done by the Arduino). This strategy was exploited to control a stepper motor, in which an automatic nanometric positioning system was developed and controlled by an Arduino nanoboard.⁷³

The electrochemical techniques are very compatible to IoT devices due to the miniaturized systems and integrated electronic circuits. In this way, amperometric measurements were performed by a potentiostat, controlled by a Raspberry Pi board.⁸³ This lab-made point-of-care device was used for electrochemical determination of cortisol levels in salivary samples. A portable system was proposed by Järvinen et al.⁸⁴ for gas monitoring. A gas sensor was developed with an H₂ sensor and an ink was prepared using WO₃-Pt nanoparticles for sensor fabrication. Other commercial sensors were incorporated in the gas sensor board, for temperature and humidity monitoring. Also, the sensor board included four socketed sensors enabling to evaluate distinct gases simultaneously. The device utilizes an Arduino microcontroller to control the sensor routine, connected on a Raspberry Pi computer.

Open science as an instrument for the democratization of knowledge was demonstrated by the use of Raspberry Pi for chromatographic techniques. Some improvements were demonstrated to increase the access and affordability of chromatographic data acquisition,^{71,85,86} especially for systems in which software control has become obsolete.⁷¹

A miniaturized Office Chromatograph (OC), constructed with an open-source hardware, 3D printing of operational parts and open-source software, was proposed. Exploiting inkjet printing of sample and mobile phase solutions, imaging of the miniaturized chromatogram via inexpensive light emitting diodes and Raspberry Pi camera as a detector was proposed by Schade *et al.*⁸⁶ The open-sourced device consists of a miniaturized, low-cost and an open-sourced system for planar chromatography, ideal for point-ofcare and on-site applications. This system allowed the quantification of steviol glycosides and bioanalytical screening. Foster *et al.*⁸⁸ developed a low-cost, opensource digital strip chart for chromatographic detectors. A Raspberry Pi single-board was used for data acquisition. In this way, an AD7705 analog-to-digital converter IC was used to read the analog pins from a detector. Any detector that provides a standard analog voltage output may be used with this device.

The detection of volatile compounds emitted by bacteria was carried out by the use of four commercial gas sensors, TGS-826 used for ammonia and amines, MQ-3 used for alcohol, MQ-135 for CO₂ and MQ-138 for acetone.⁸⁵ These gas sensors were connected to an Arduino board which read their values, and these values were then sent to a Raspberry Pi board via USB port.

As alternative for endpoint determination in neutralization, redox and complexation titrations were proposed by Alessio *et al.*⁸⁹ A simple automatic device was developed using a contactless thermometric sensor for temperature monitoring and a lab-made syringe pump to titration control. Open-source software developed by Python was built to plot the results in a real-time chart and suggested the endpoint of the titration automatically. The proposed device was applied for the determination of the acidity of sauces and the ascorbic acid and calcium content of pharmaceutical samples.

6. Future Directions

The development of sustainable analytical methods demands the correlation of low impact to the environment (green analytical chemistry that requires reduction of waste generation approaching to "zero waste"), economy (affordable devices that can be executed in developing countries) and benefits to the society in a wide range of interests. The IoT analytical devices described in this review attend to such statements that enable us to conclude that they contribute to a new generation of sustainable analytical methods.

Scientific papers and citations of devices based on IoT increased exponentially in recent years and analytical chemistry applications follow the trend of increasing publications and citations. The papers discussed in this article provide a small overview of IoT-based devices for analytical chemistry, focusing on the use of Arduino and Raspberry Pi boards. Most of these works provide a simple way, based on plug and play and DIY concepts. However, the challenges for the future of IoT-based devices will depend on some improvement on computer hardware, embedded system, networking, display, control devices, especially for some analytical techniques such as chromatography and mass spectrometry. In these cases, an intermediate/advanced knowledge of programming such as Python language will be necessary, especially for software enhancements and artificial intelligence (AI). Big data and artificial intelligence have a synergistic relationship to control and manipulate chemical analysis. Through such Arduino-based and Raspberry-based apparatus, humans and AI, it is possible to enhance each other's complementary strengths, presenting a pilar to the advance of IoAT. Although the evolution for AI-based analytical instrumentation is moving forward, the implementation is in the early stages and outcomes not yet materialized, and analytical goals were not well defined. The development of novel algorithms for interpretation of data is one of the challenges for analytical IA-based development. If you are interested in artificial intelligence applied in chemistry, we recommend reading some articles.^{90,91}

Finally, the 5G connection is expected to happen in the near future. According to reports, 5G will enhance the speed and integration of other technologies. Due to 5G boosts network capacity, the cloud computing, artificial intelligence and edge computing will all help to handle the data volumes generated by the IoT devices, allowing the devices to function with intelligence levels similar to that of humans.

Acknowledgments

The authors would like to thank the financial support from National Council for Scientific and Technological Development (CNPq, process numbers: 425494/2018-7 and 307271/2017-0) and Coordination for the Improvement of Higher Education Personnel (CAPES, financial code 001).

Author Contributions

Alegre N. S. Cadeado, Caio C. S. Machado, Geandre C. Oliveira and Daniela A. S e Silva were responsible for the conceptualization, investigation, methodology, writing original draft; Rodrigo A. A. Muñoz for the conceptualization, data curation, funding acquisition, investigation, methodology, resources, writing original draft, writing review and editing; Sidnei G. Silva for the conceptualization, data curation, funding acquisition, investigation, methodology, project administration, resources, writing original draft, writing review and editing.



Alegre N. S. Cadeado is Graduated in Chemistry and Biology, MSc in Chemistry Education / Teaching from the Pedagogical University of Mozambique. He is Professor of Chemistry in the Department of Science

and Technology at Licungo University, Quelimane, Mozambique. He is a PhD student in Chemistry at UFU, where he is a member of Prof Silva's research group. *He works with the development of portable devices for colorimetric analysis.*



Caio C. S. Machado is graduated in Industrial Chemistry at Universidade Federal de Uberlândia (UFU) where he also concluded his MSc and actually is a PhD student at UFU, where he is

member of Prof Silva's research group, developing remoted controlled analytical devices, mainly for point-of-care health monitoring and food control.



Geandre C. Oliveira has a degree in Chemistry from the University of Uberaba and is a professor at the Instituto Federal do Triângulo Mineiro (IFTM). He is currently finishing his doctorate at the

Universidade Federal de Uberlândia (UFU), where he is a member of Prof Silva's research group. He works with the development of portable devices for colorimetric analysis.



Daniela A. S. e Silva is graduated in Chemistry at the Instituto Luterano de Ensino Superior de Itumbiara-GO. She is specialized in Environmental Management and Quality at the Universidade Federal

de Goiás. Actually, she is an MSc student at Universidade Federal de Uberlândia, where she is member of Prof Silva's research group, developing remoted controlled analytical devices for environmental monitoring.



Rodrigo A. A. Muñoz is graduated in Chemistry from the University of São Paulo, Brazil (2002), and received his PhD in Analytical Chemistry from the same university in 2006, an internship

period at the Oxford University, UK (2005). He completed a postdoctoral research at the Arizona State University (USA) during 2006-2007 and a postdoctoral research at the University of São Paulo during 2007-2008. He is currently Associate Professor of Chemistry at the Federal University of Uberlândia, Brazil, Associate Editor of Journal of the Brazilian Chemical Society and an affiliate member of the Brazilian Academy of Science. His current research interests focus on electrochemical (bio)sensors and devices, 3D-printing technology and novel materials.



Sidnei G. Silva is graduated in Chemistry and received his PhD in Analytical Chemistry at Institute of Chemistry of Universidade de São Paulo, an internship period at the Universitat de València, Spain, working with automation in analytical chemistry and optical methods of analysis. He completed a postdoctoral research at the Universidade Federal de São Carlos and at the Wake Forest University (USA). He is Assistant Professor at Universidade Federal de Uberlândia. His research is focused on instrumentation in analytical chemistry, optical methods of analysis, sensors for internet of things and related applications.

References

- 1. United Nations, https://www.unep.org/pt-br/node/1495, accessed in March 2022.
- Axon, S.; James, D.; Curr. Opin. Green Sustainable Chem. 2018, 13, 140. [Crossref]
- Fatimah, Y. A.; Govindan, K.; Murniningsih, R.; Setiawan, A.; J. Cleaner Prod. 2020, 269, 122263. [Crossref]
- 4. Friege, H.; Sustainable Chem. Pharm. 2017, 6, 57. [Crossref]
- Chen, T. L.; Kim, H.; Pan, S. Y.; Tseng, P. C.; Lin, Y. P.; Chiang, P. C.; *Sci. Total Environ.* 2020, *716*, 136998. [Crossref]
- Capella, J. V.; Bonastre, A.; Campelo, J. C.; Ors, R.; Peris, M.; *Trends Environ. Anal. Chem.* 2020, 27, e00095. [Crossref]
- Grudpan, K.; Kolev, S. D.; Lapanantnopakhun, S.; McKelvie, I. D.; Wongwilai, W.; *Talanta* 2015, *136*, 84. [Crossref]
- Capella, J. V.; Bonastre, A.; Ors, R.; Peris, M.; Sensors 2019, 19, 5528. [Crossref]
- Mayer, M.; Baeumner, A. J.; Chem. Rev. 2019, 119, 7996. [Crossref]
- Kassal, P.; Steinberg, I. M.; Steinberg, M. D.; Sens. Actuators, B 2013, 184, 254. [Crossref]
- 11. Raspberry Pi, https://www.raspberrypi.com, accessed in March 2022.
- 12. Beagle Board, https://beagleboard.org, accessed in March 2022.
- 13. Rikomagic, http://www.rikomagic.co.uk, accessed in March 2022.
- 14. Cubieboard, http://cubieboard.org, accessed in March 2022.
- Mars Board, https://www.marsboard.com, accessed in March 2022.
- 16. Udoo, https://www.udoo.org, accessed in March 2022.
- 17. Arduino, https://www.arduino.cc, accessed in November 2021.
- 18. O'Reilly, https://www.oreilly.com, accessed in March 2022.
- Introduction to Arduino; http://www.introtoarduino.com/ downloads/IntroArduinoBook.pdf, accessed in March 2022.
- Open Softwear, http://softwear.cc/book/files/Open_Softwearbeta090712.pdf, accessed in March 2022.
- Magpi Raspberry Pi, https://magpi.raspberrypi.com/books, accessed in March 2022.
- 22. Udemy, https://www.udemy.com, accessed in March 2022.
- 23. Programming Electronics Academy, https://www. programmingelectronics.com/, accessed in March 2022.
- 24. RS, www.rs-online.com, accessed in March 2022.

- 25. Sparkfun, https://www.sparkfun.com/, accessed in March 2022.
- 26. Adafruit, https://www.adafruit.com, accessed in March 2022.
- 27. Conrad, https://www.conrad.com, accessed in March 2022.
- Filipe Flop, https://www.filipeflop.com/, accessed in March 2022.
- 29. Raspberry Pi Forums, https://forums.raspberrypi.com/, accessed in March 2022.
- Arduino Forum, https://forum.arduino.cc/, accessed in March 2022.
- 31. Git Hub, https://github.com/, accessed in March 2022.
- Instructables, https://www.instructables.com/, accessed in March 2022.
- Fitzpatrick, D. E.; O'Brien, M.; Ley, S. V.; *React. Chem. Eng.* 2020, 5, 201. [Crossref]
- Grinias, J. P.; Whitfield, J. T.; Guetschow, E. D.; Kennedy, R. T.; *J. Chem. Educ.* 2016, *93*, 1316. [Crossref]
- Bougot-Robin, K.; Paget, J.; Atkins, S. C.; Edel, J. B.; *J. Chem. Educ.* 2016, 93, 1232. [Crossref]
- Ayres, L. B.; Lopes, F. S.; Garcia, C. D.; Gutz, I. G. R.; *Anal. Methods* 2020, *12*, 4109. [Crossref]
- Francisco, K. J. M.; do Lago, C. L.; *Electrophoresis* 2009, *30*, 3458. [Crossref]
- da Costa, E. T.; Mora, M. F.; Willis, P. A.; do Lago, C. L.; Jiao, H.; Garcia, C. D.; *Electrophoresis* 2014, *35*, 2370. [Crossref]
- Anzalone, G. C.; Glover, A. G.; Pearce, J. M.; Sensors 2013, 13, 5338. [Crossref]
- 40. Adafruit HUZZAH ESP8266 Breakout, https://www.adafruit. com/product/2471, accessed in March 2022.
- Adafruit ESP32S3-DevKitC-1 ESP32-S3-WROOM-2 32MB Flash 8MB PSRAM, https://www.adafruit.com/product/5364, accessed in March 2022.
- Santos, G. F.; Reis, F. B.; *Microchem. J.* 2021, *163*, 105918. [Crossref]
- Lima, M. J. A.; Kamogawa, M. Y.; Reis, B. F.; *Microchem. J.* 2019, 145, 921. [Crossref]
- Khanfar, M. F.; Al-Faqheri, W.; Al-Halhouli, A.; Sensors 2017, 17, 2345. [Crossref]
- Grazioli, C.; Faura, G.; Dossi, N.; Toniolo, R.; Abate, M.; Terzi, F.; Bontempelli, G.; *Microchem. J.* **2020**, *159*, 105584. [Crossref]
- Machado, C. C. S.; Petruci, J. F. S.; Silva, S. G.; *Microchem. J.* 2021, *168*, 106466. [Crossref]
- Bzura, J.; Fiedoruk-Pogrebniak, M.; Koncki, R.; *Talanta* 2018, 190, 193. [Crossref]
- Hossain, M. A.; Canning, J.; Yu, Z.; Ast, S.; Rutledge, P. J.; Wong, J. K. H.; Jamalipour, A.; Crossley, M. J.; *Analyst* 2017, *142*, 1953. [Crossref]
- Daniel, S. C. G. K.; Kumar, A.; Sivasakthi, K.; Thakur, C. S.; Sens. Actuators, B 2019, 290, 73. [Crossref]
- Ng, S. M.; Wong, D. S. N.; Phung, J. H. C.; Chua, H. S.; *Talanta* 2013, *116*, 514. [Crossref]

- Nghia, N. N.; Huy, B. T.; Lee, Y. I.; Analyst 2020, 145, 3376. [Crossref]
- Greenawald, L. A.; Boss, G. R.; Snyder, J. L.; Reeder, A.; Bell, S.; ACS Sens. 2017, 2, 1458. [Crossref]
- 53. Gillanders, R. N.; Samuel, I. D. W.; Turnbull, G. A.; Sens. Actuators, B 2017, 245, 334. [Crossref]
- 54. Heo, G.; Manivannan, R.; Kim, H.; Kim, M. J.; Min, K. S.; Son, Y.-A.; *Sens. Actuators, B* **2019**, *297*, 126723. [Crossref]
- 55. Maejima, K.; Hiruta, Y.; Citterio, D.; *Anal. Chem.* **2020**, *92*, 4749. [Crossref]
- 56. Singh, H.; Singh, G.; Mahajan, D. K.; Kaur, N.; Singh, N.; Sens. Actuators, B **2020**, 322, 128622. [Crossref]
- de Morais, C. L. M.; Carvalho, J. C.; Sant'Anna, C.; Eugênio, M.; Gasparotto, L. H. S.; Lima, K. M. G.; *Anal. Methods* 2015, 7, 7917. [Crossref]
- Brandão, E. G.; Perdigão, S. R. W.; Reis, B. F.; *Microchem. J.* 2021, 171, 106789. [Crossref]
- Strzelak, K.; Malasuk, C.; Oki, Y.; Morita, K.; Ishimatsu, R.; Microchem. J. 2020, 157, 104936. [Crossref]
- Cabrera, H.; Akbar, J.; Korte, D.; Ramírez-Miquet, E. E.; Marín, E.; Niemela, J.; Ebrahimpour, Z.; Mannatunga, K.; Franko, M.; *Talanta* 2018, 183, 158. [Crossref]
- Agustini, D.; Fedalto, L.; Agustini, D.; dos Santos, L. G. M.; Banks, C. E.; Bergamini, M. F.; Marcolino-Junior, L. H.; *Sens. Actuators, B* 2020, *304*, 127117. [Crossref]
- Shen, X.; Ju, F.; Li, G.; Ma, L.; Sensors 2020, 20, 2781. [Crossref]
- Gao, W.; Luo, X.; Liu, Y.; Zhao, Y.; Cui, Y.; Sens. Actuators Rep. 2021, 3, 100045. [Crossref]
- Mercer, C.; Bennett, R.; Conghaile, P.; Rusling, J. F.; Leech, D.; Sens. Actuators, B 2019, 290, 616. [Crossref]
- 65. Meloni, G. N.; J. Chem. Educ. 2016, 93, 1320. [Crossref]
- Liu, L.; Zhang, D.; Zhang, Q.; Chen, X.; Xu, G.; Lu, Y.; Liu, Q.; *Biosens. Bioelectron.* 2017, *93*, 94. [Crossref]
- Khan, M. S.; Dighe, K.; Wang, Z.; Srivastava, I.; Daza, E.; Schwartz-Dual, A. S.; Ghannam, J.; Misra, S. K.; Pan, D.; *Analyst* 2018, *143*, 1094. [Crossref]
- Zhang, D.; Jiang, J.; Chen, J.; Zhang, Q.; Lu, Y.; Yao, Y.; Li, S.; Liu, G. L.; Liu, Q.; *Biosens. Bioelectron.* 2015, 70, 81. [Crossref]
- Ainla, A.; Mousavi, M. P. S.; Tsaloglou, M. N.; Redston, J.; Bell, J. G.; Fernández-Abedul, M. T.; Whitesides, G. M.; *Anal. Chem.* 2018, *90*, 6240. [Crossref]
- Itterheimová, P.; Foret, F.; Kubáň, P.; Anal. Chim. Acta 2021, 1153, 338294. [Crossref]
- Ting, H.; Hu, J. B.; Hsieh, K. T.; Urban, P. L.; *Anal. Methods* 2014, 6, 4652. [Crossref]
- Boehle, K. E.; Doan, E.; Henry, S.; Beveridge, J. R.; Pallickara, S. L.; Henry, C. S.; *Anal. Methods* 2018, *10*, 5282. [Crossref]

- Gomez-Cruz, J.; Nair, S.; Manjarrez-Hernandez, A.; Gavilanes-Parra, S.; Ascanio, G.; Escobedo, C.; *Biosens. Bioelectron.* 2018, 106, 105. [Crossref]
- Gómez, A.; Bueno, D.; Gutiérrez, J. M.; *Biosensors* 2021, *11*, 68. [Crossref]
- Jang, H.; Ahmed, S. R.; Neethirajan, S.; Sensors 2017, 17, 1079. [Crossref]
- 76. Qi, W.; Zheng, L.; Wang, S.; Huang, F.; Liu, Y.; Jiang, H.; Lin, J.; *Biosens. Bioelectron.* **2021**, *178*, 113020. [Crossref]
- Tok, S.; de Haan, K.; Tseng, D.; Usanmaz, C. F.; Koydemir, H. C.; Ozcan, A.; *Lab Chip* 2019, *19*, 2925. [Crossref]
- Tonelli, A.; Candiani, A.; Sozzi, M.; Zucchelli, A.; Foresti, R.; Dall'Asta, C.; Selleri, S.; Cucinotta, A.; *Sens. Actuators, B* 2019, 282, 559. [Crossref]
- Tonelli, A.; Mangia, V.; Candiani, A.; Pasquali, F.; Mangiaracina, T. J.; Grazioli, A.; Sozzi, M.; Gorni, D.; Bussolati, S.; Cucinotta, A.; Basini, G.; Selleri, S.; *Sensors* 2021, *21*, 3552. [Crossref]
- Bills, M. V.; Loh, A.; Sosnowski, K.; Nguyen, B. T.; Ha, S. Y.; Yim, U. H.; Yoon, J.-Y.; *Biosens. Bioelectron.* 2020, 159, 112193. [Crossref]
- Chen, W.; Xiong, Y.; Wang, W.; Wu, T.; Li, L.; Kang, Q.; Du, Y.; *Talanta* **2019**, *203*, 77. [Crossref]
- Bunge, F.; van den Driesche, S.; Waespy, M.; Radtke, A.; Belge, G.; Kelm, S.; Waite, A. M.; Mirastschijski, U.; Vellekoop, M. J.; Sens. Actuators, B 2019, 289, 24. [Crossref]
- Gevaerd, A.; Watanabe, E. Y.; Belli, C.; Marcolino-Junior, L. H.; Bergamini, M. F.; *Sens. Actuators, B* 2021, *332*, 129532. [Crossref]
- Järvinen, T.; Lorite, G. S.; Rautio, A. R.; Juhász, K. L.; Kukovecz, Á.; Kónya, Z.; Kordas, K.; Toth, G.; *Sens. Actuators, B* 2017, 252, 983. [Crossref]
- Alvarez, C. S.; Sierra-Sosa, D.; Garcia-Zapirain, B.; Yoder-Himes, D.; Elmaghraby, A.; Sensors 2019, 19, 1523. [Crossref]
- Schade, F.; Schwack, W.; Demirbas, Y.; Morlock, G. E.; *Anal. Chim. Acta* 2021, *1174*, 338702. [Crossref]
- Lam, S. C.; Coates, L. J.; Gupta, V.; Wirth, H. J.; Gooley, A. A.; Haddad, P. R.; Paull, B.; *J. Chromatogr. A* 2020, *1631*, 461540. [Crossref]
- Foster, S. W.; Alirangues, M. J.; Naese, J. A.; Constans, E.; Grinias, J. P.; *J. Chromatogr. A* 2019, *1603*, 396. [Crossref]
- Alessio, K. O.; Tischer, B.; Voss, M.; Teixeira, I. D.; Brendler,
 B. M.; Duarte, F. A.; Helfer, G. A.; Costa, A. B.; Barin, J. S.;
 Talanta 2020, 216, 120975. [Crossref]
- 90. Gasteiger, J.; ChemPhysChem 2020, 21, 2233. [Crossref]
- Ayres, L. B.; Gomez, F. J. V.; Linton, J. R.; Silva, M. F.; Garcia, C. D.; *Anal. Chim. Acta* 2021, *1161*, 338403. [Crossref]

Submitted: December 2, 2021 Published online: March 17, 2022