The end user sensor tree: An end-user friendly sensor database

J.L.D. Nelis[a], A.S. Tsagkaris[b], Y. Zhao[a,c], J. Lou-Franco[a], P. Nolan[a], H. Zhou[c,d], C. Cao[a], K. Rafferty[c], J. Hajslova[b], C.T. Elliott[a], K. Campbell[a,*]

[a] *Institute for Global Food Security, School of Biological Sciences, Queen's University, David Keir Building, Stranmillis Road, Belfast BT9 5AG, UK*
[b] *Department of Food Analysis and Nutrition, Faculty of Food and Biochemical Technology, University of Chemistry and Technology Prague, Technická 5, 166 28 Prague 6 – Dejvice, Prague, Czech Republic*
[c] *School of Electronics, Electrical Engineering and Computer Science, Queen's University Belfast, Stranmillis Road, Belfast, UK*
[d] *Department of Informatics, University of Leicester, University Road, Leicester LE1 7RH, UK*

A B S T R A C T

Detailed knowledge regarding sensor based technologies for the detection of food contamination often remains concealed within scientific journals or divided between numerous commercial kits which prevents optimal connectivity between companies and end-users. To overcome this barrier The End user Sensor Tree (TEST) has been developed. TEST is a comprehensive, interactive platform including over 900 sensor based methods, retrieved from the scientific literature and commercial market, for aquatic-toxins, mycotoxins, pesticides and microorganism detection. Key analytical parameters are recorded in excel files while a novel classification system is used which provides, tailor-made, experts' feedback using an online decision tree and database introduced here. Additionally, a critical comparison of reviewed sensors is presented alongside a global perspective on research pioneers and commercially available products. The lack of commercial uptake of the academically popular electrochemical and nanomaterial based sensors, as well as multiplexing platforms became very apparent and reasons for this anomaly are discussed.

1. Introduction

It has been suggested that the scientific community invests little effort in the production of robust, calibrated and reliable tools ready for mass production but often produces a myriad of highly sophisticated 'gadgets' instead (Mohammed et al., 2015; Whitesides, 2013; Chin et al., 2012). Moreover, the cornucopia of different sensors presented both in the scientific literature and commercial resources does not help informed decision making regarding which sensor would best fit the end-users', or researchers' needs. As a result, many different point-of-site sensors of outstanding quality remain hidden from potential end-users. Moreover, a fair comparison of novel developed sensors with similar existing products is inhibited due to the lack of an orderly classification system outlining sensor specifications. A possible remedy is the mapping of sensor capabilities in an open access repository. Such a system could boost commercialisation of recently developed sensors and provide independent expert advice to end-users which can help to promote the use of successful sensors whilst deselecting superfluous systems. Moreover, online repositories can stimulate communication between actors and speed up product design by avoiding the reinvention of the wheel and ignite citizen science developments (Glen Martin

et al., 2017). Some examples are: (i) the microfluidics repository "metafluidics" which allows researchers and science enthusiasts alike to download and upload design schematics for microfluidic related systems (Kong et al., 2017) (ii) the Synthetic Biology Open Language (SBOL) initiative, connecting software developers and wet lab based scientists (Galdzicki et al., 2014) and (iii) the Protein Capture Reagents Program providing a database for validated monoclonal antibodies (Venkataraman et al., 2018). In this work the creation of an end-user-friendly sensor database, mapping sensor capabilities throughout the commercial and scientific sector is described. The platform, named "The End-user Sensor Tree", or TEST, applies a non-linear approach aimed at improving the communication between actors while markedly increasing novel sensor visibility. TEST uses end user focused classification criteria aimed to sort information on available sensors into comprehensive, easy-to-read sections, tailored for both the expert and non-expert. These e-docs are accessible through a decision tree on the website (http://test.foodsmartphone.net/) further facilitating ease of use. The system has currently been established for sensors enabling the detection of four food safety related target groups i.e. aquatic toxins (in marine- fresh- and drinking water), mycotoxins, pesticides, and microorganisms in food. The last of these groupings was split into spoilage

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* Corresponding author.
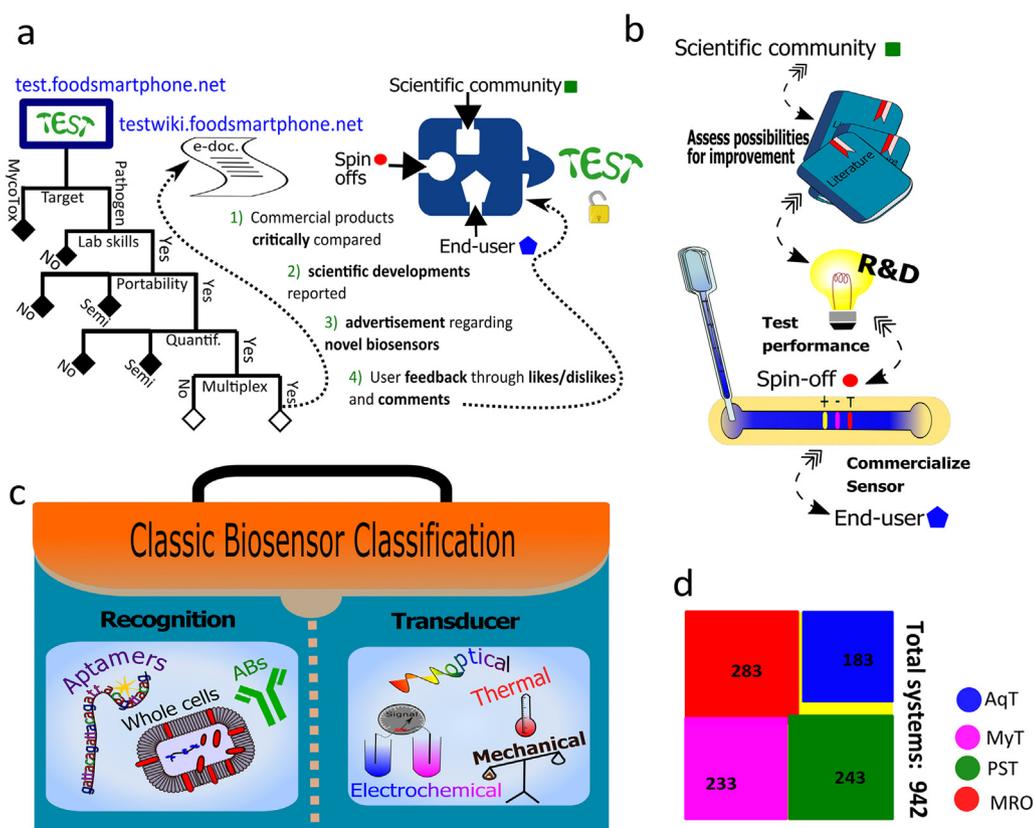   *E-mail address:* katrina.campbell@qub.ac.uk (K. Campbell).

Fig. 1. Overview of the TEST classification system and database. a) The TEST decision tree (left web address in blue) and hierarchal classification system are visualised using pathogens as an example. After running through the nodes the user is directed to an e-doc providing tailored information (right web address in blue) which has a fixed layout (point 1–4 green) and is designed to provide an interactive platform for all users. b) A scheme showing the classic, linear, sensor development pipeline often applied. c) An overview of the classic biosensor classification system showing both the major recognition and transducer elements used. d) A scaled overview of the total amount of detection methods integrated in the TEST repository. Colours are according groups i.e. blue for aquatic toxins (AqT), magenta for mycotoxins (MyT), green for pesticides (PST) and red for microorganisms (MRO). The yellow square represents 100%. The number of systems per square is indicated. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

organisms and pathogens where possible. In the following sections the system was used to analyse and compare the development of novel sensors/methods and commercially available sensors using both classic (transducer based) and novel (TEST) criteria. This study resulted in the generation of a highly informative database providing an easily visualised, in-depth, critical comparison of sensing possibilities encompassing portability, quantification and expertise axes, for each target group, which is introduced here. Moreover, analytical parameters of reported devices are recorded in excel files, generating a dataset that allowed for the global inter-target group comparison reported, which exposes several gaps between commercially available and scientifically reported sensors, and, visualises target group specific trends in sensor use.

2. Material and methods

2.1. Construction of the TEST tree and e-docs

The TEST repository is based on an online decision tree (http://TEST.foodsmartphone.net/) which directs the user to tailored e-docs (http://TESTwiki.foodsmartphone.net/), with a fixed layout (Fig. 1a). The tree uses four classification criteria in the following consecutive order 1) expert training needed 2) sensor portability 3) quantification ability 4) single or multiplex screening ability. This classification system was chosen to enable a more practical, non-linear approach (Fig. 1a), which, unlike the classic linear approach often used for sensor development (Fig. 1b), stimulates interaction of all stakeholders in the sensor development pipeline. Moreover, the system aims to meet the needs of the end-user by using the bespoken classification system rather than the bio-recognition elements and/or transducers based classification system (Fig. 1c) which is classically used (Monošík et al., 2012; Thevenot et al., 2001). The collected information from supplementary tables 1–8 was used to build the decision tree, e-docs and most of the information reported herein. A scaled overview of the division of these

detection methods between target groups is shown in Fig. 1d. The TEST repository aims to provide information both for the expert and the non-expert regarding all detection methods for the mentioned targets. Thus all identified detection methods, were included. This allowed the inclusion of reference method and allowed comparison of novel sensors with the reference. Moreover, some non-biosensor methods are equally commercially available (mass spectrometry (MS) analyses of pesticides and mycotoxins for example) which makes including them valuable for the end-user. Space for the user to leave comments for discussion and for relevant companies to advertise their products are also provided to stimulate stakeholder interaction. In total 942 detection systems (633 research reports and 309 commercial sensors) have been included in the TEST repository.

The following online methods are further described in the Supplementary Material:

2.2. Search criteria to collect literature

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2.3. Definitions used for the TEST criteria to classify sensors

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2.4. The TEST website construction

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3. Results and discussion

3.1. Sensors/methods investigated using TEST classification

A classification system was designed in an attempt to visualise trends and differences between the identified target groups regarding
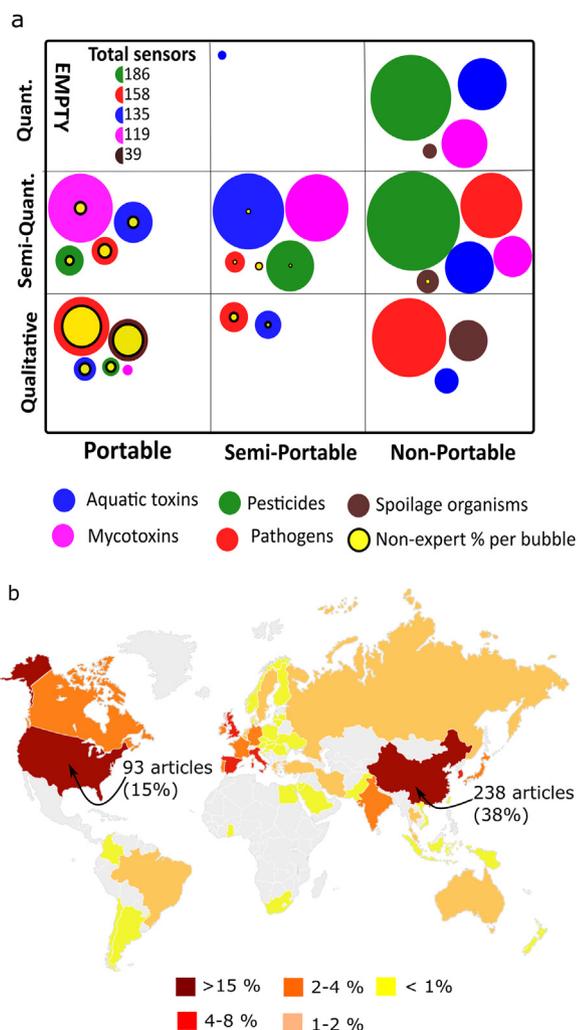
a

b

Fig. 2. TEST sensor classification. a) Each bubble is scaled by area to the total amount of detection methods reported. The 5 target groups (each with a specific colour indicated in legend) are discretely divided between the 9 sections made up of the combinations between portable, semi-portable and non-portable with qualitative, semi-quantitative and quantitative. Non-expert % shown within bubbles is scaled by area to total amount of articles (expert and non-expert) within the individual bubbles. b) Worldwide distribution of detection methods for the listed contaminants reported from 2013 up to 2017. Colours correspond to percentage of total reports as indicated in legend.

sensor development in reported scientific literature. To this end Fig. 2a was analysed over 3 axes i.e. expert/non-expert, portability and quantification. When focussing on sensors developed for the non-expert (Fig. 2a, yellow internal bubbles), the most striking differences were between the microorganism group (20% split into pathogens and spoilage organisms) and the pesticide (4%) target groups. An explanation for this might be the trend towards using quantitative non-portable chromatographic techniques for pesticides (almost half of all reports for pesticides fall in that bubble). This is logical since it is the only method to simultaneously quantify multiple analytes (there are hundreds of pesticides) in a single analytical cycle within a wide linear range. For microorganisms however, Polymerase Chain Reaction (PCR) is often used as a reference method. This technique is more easily made portable by use of isothermal techniques such as Loop-mediated isothermal amplification, for which user friendly portable systems utilizing microfluidics exist although sensitivity is of yet sub-optimal (Sayad et al., 2016; Oh et al., 2016). Equally, electrochemistry (EC) has potential as a non-expert portable technique for microorganisms with even multiplex options at low LODs (Primiceri et al., 2016). EC sensors are equally

reported for other target groups. One notable prototype for pesticide detection, developed by Mishra et al. (2017), is a lab on a glove EC sensor, allowing real time wireless data transmission to a smartphone based device. Even if this pioneering approach did not detect its target pesticides at the required sensitivities, it may pave the way for fast, portable and simple on-site methods using EC. Compared to optical methods however EC is rarely used for non-expert sensors while colorimetric are gaining traction due to easy visual interpretation of the results generated. Cholinesterase inhibition assays in dipstick format (Badawy and El-Aswad, 2014; Apilux et al., 2015) or immunochromatographic assays (Fang et al., 2015) for pesticides are good examples despite the fact they lack quantification. This could be altered if a smartphone camera is used as a detector which has been the case for both pesticides (Comina et al., 2016) and mycotoxins (Machado et al., 2018). In fact when reviewing the portable axes of the TEST system, colorimetric techniques currently prevail with microorganisms (Liu et al., 2015; Wu et al., 2015; Jiang et al., 2016) and mycotoxins (Zhang et al., 2018; Liu et al., 2017; Song et al., 2014) being the most mentioned targets. As for the semi-portable section almost all reports fall in the semi-quantitative group and of these more than half of the reports used a variant of nanotechnology often using EC technology such as Palmsens for detection. The high cost of the potentiostats combined with sensitivity to electrostatic variance, especially for impedance measurements, (which can happen outside of a Faraday cage) might be a reason why these sensors often remain in the semi-portable group. A potential way to overcome this problem is to piggyback on commercially available glucose sensors for other targets. For example, it has been demonstrated that the aquatic toxin brevetoxin (BTX-2) can be detected at very low concentrations (LOD 0.01 μg/L) using a nanocontainer packed with glucose and gated by antibody/hapten complexes without the need for extensive sample handling (Gao et al., 2014). Other semi-portable detection methods reported are portable Raman spectrometers (Yang et al., 2014; S. Huang et al., 2016, Y. Huang et al., 2016) or mass spectrometers (Wang et al., 2016) used for the detection for pesticides. Non-portable sensors, however, still prevail with approximately 80%, 66%, 50% and 43% of the sensors reported for pesticides, microorganisms, mycotoxins and aquatic toxin detection respectively. With liquid chromatography coupled mass spectrometry (LC-MS) being the most described method for pesticides, aquatic toxins and mycotoxin analyses in this section. For microorganisms, quantitative plating techniques are most described for pathogen detection (Lambert et al., 2015) while qualitative metabolomics high resolution MS techniques are mainly used for spoilage organism detection (Xu et al., 2013). An interesting development for non-portable quantitative detection methods is replacing chromatography by direct MS measurements. Indeed, some MS techniques showing sensitive and quantitative analysis without applying chromatography beforehand for aquatic toxin detection using either matrix assisted laser desorption ionisation (MALDI) (Roegner et al., 2014) or ambient source high resolution MS solutions (Roy-Lachapelle et al., 2015) when compared to classic triple quadrupole LC-MS (Pekar et al., 2016). Evidently, such systems can speed up analysis and enable rapid screening in the lab using non-portable devices, an alternative to the point-of-site solution. Finally, when looking at qualitative versus quantitative detection, it is clear that the latter is hardly ever portable. An interesting exception, though lacking in sensitivity, is semi-portable quantitative chromatographic techniques (Abdul Keyon et al., 2014). It combines capillary electrophoresis with a contactless conductivity detector to quantify paralytic shellfish toxins. Qualitative sensors for pesticides and mycotoxin detection are clearly not popular with mycotoxin analysis having only one reported, portable, qualitative multiplex sensors (Beloglazova et al., 2014). This highlights the desired need for quantitative analysis in mycotoxin testing, even for portable devices. For microorganisms and aquatic toxins however, qualitative sensors are being employed. Here most interesting is the use of non-portable eco-toxicity tests for freshwater (Häder and Erzinger, 2017), and seawater (S. Huang et al.,

2016, Y. Huang et al., 2016) screening. Such tests require minimal time while allowing continuous screening which can be useful as an early warning system. Overall, a push towards portable, user friendly quantitative point-of-site instruments was observed throughout the literature. However, as seen in Fig. 2a, these three properties together have not yet been materialized into a sensor. On the contrary, most sensors reported are still non-portable or, in the case of being (semi)portable, show poor quantification (strip tests) or require expertise (EC sensors). Thus further development of these methods is needed to truly realise the desired point-of-site testing so often discussed in the scientific literature. Apart from these observations it is interesting to note the distribution of these (+/- 600) publications on sensors for food worldwide (Fig. 2b), just two countries (China and USA) account for half of them. The EU accounts for another 35%, with the remaining distributed elsewhere in the world with a focus on Korea, Canada, India and Japan.

3.2. Analytical techniques and their sensitivity

In order to accurately represent the numerous analytical methods employed an attempt was made to cluster these into uniform classes based on the applied analytical principle. In this way, 9 different groups were defined including chromatographic, spectroscopic, vibrational, EC, PCR, ambient mass spectrometry and other techniques (cases with less than 3% of the total). The separate cases of enzyme-linked immunosorbent assay (ELISA) and test-strips were separated because they account for 10% of the TEST database and play an important role in

contaminant screening. Focusing on an overall description of the results (Fig. 3a) spectroscopic detection methods were the most widely used (about 23%) while EC techniques accounted for 20%. Importantly, it was noted that both methods used various recognition elements (enzymes, antibodies, aptamersand molecular imprinted polymers) to predominately determine a single analyte demonstrating a significant reason for the predominance of chromatography in multiplex analysis. Surprisingly, chromatographic techniques ranked in the third place of the TEST database (16%) even though it is thought by many to be the gold standard for all target groups except microorganisms (where PCR based techniques are the norm). An explanation may be that only novel use such as nanomaterial implementation for increased sensitivity and non-targeted analysis by high resolution MS, appear in the recent literature while routine LC-MS analysis remains less well reported. Noticeably most other sensors, focussing more on (portable) screening, were strip tests and ELISA for mycotoxins and food pathogens analysis, surface enhanced Raman spectroscopy (SERS) for pesticides screening and EC for aquatic toxins. The reason for this discrepancy in preferred sensors between target groups is unclear but might be the result of intellectual phase locking, with research groups specializing in the detection of a certain target group building on each other's work improving the state of the art of a specific detection method for that target thus increasing reporting on that type of sensor. As for target diversity, Fig. 3b clearly shows that the majority of the sensors/methods were developed against few target analytes. In detail, 70% (aquatic toxins), 67% (microorganisms), 58% (mycotoxins) and 44% (pesticides) of the
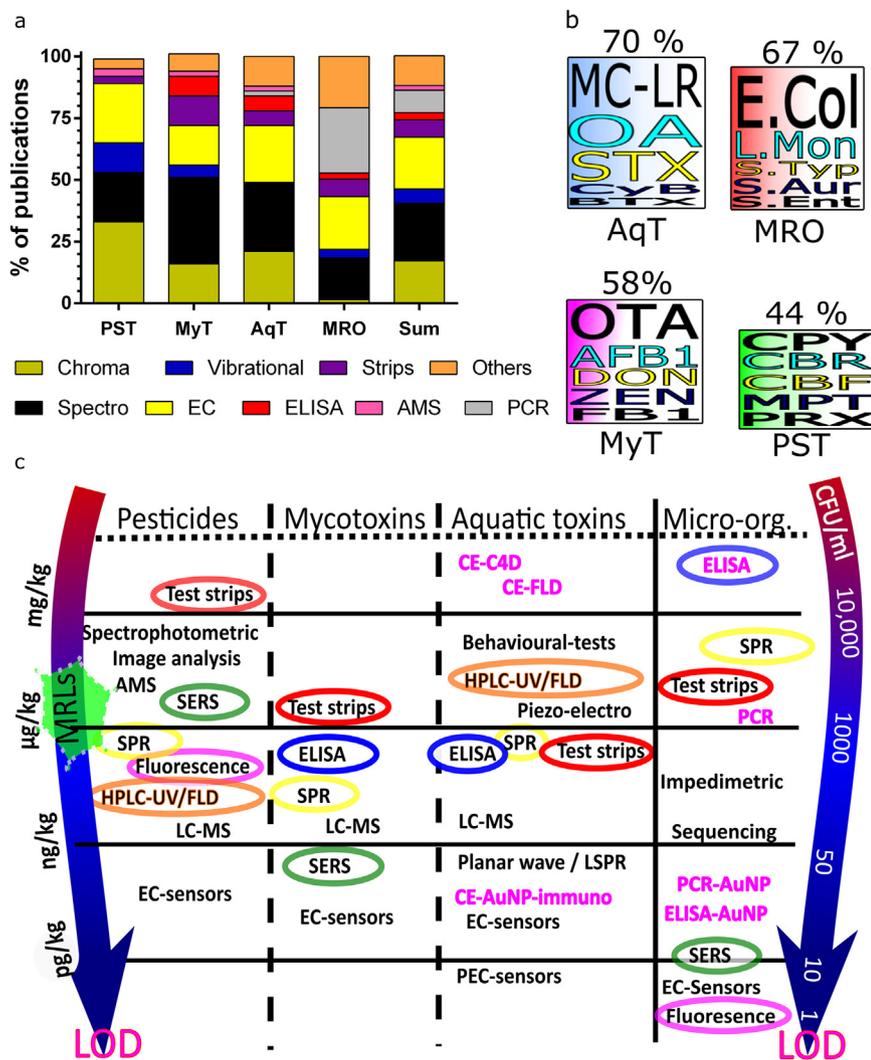

Fig. 3. Analytical methods, main targets and overall sensitivity. a) Analytical method classification per target. Percentages of each analytical technique are shown per target group in colour code (see legend). Sum shows average for total amount of analytical methods reported in the scientific literature. Chroma is chromatography, Spectro is spectroscopy, EC is electrochemistry, AMS is ambient mass spectroscopy, PCR is polymerase chain reaction. PST is pesticides, MyT is mycotoxins, AqT is aquatic toxins, MRO microorganisms and sum is the sum of all targets. b) Top 5 analytes reported per target group (acronyms as indicated above). Percentages (top of the bars) show the fraction (in %) that this top 5 makes up of the total variety in reported analytes within the target groups. The abbreviations for each analyte are scaled to the total analytes within the target group, with the least abundant down and most abundant on top in each bar. For aquatic toxins: MC-LR is microcystin-LR, OA is okadaic acid, STX is saxitoxin, CyB is cyanobacteria, BTX is brevetoxin. For Pathogens: E.col is Escherichia coli, L.Mon is Listeria monocytogenes S.Typ is Salmonella typhimurium, S.Aur is Staphylococcus aureus, S.Ent is Salmonella enterica. For mycotoxins: FB1 is Fumonisin B1, ZEN is Zearalenone, DON is Deoxynivalenol, AFB1 is Aflatoxin B1, OTA is Ochratoxin A. For Pesticides: PRX is paraoxon, MPT is methyl parathion, CBF is carbofuran, CBR is carbaryl and CPY is chlorpyrifos. c) LOD comparison for detection methods reported in literature for all target groups. LODs for the first 3 groups are expressed in g/kg (left arrow) and LODs for microorganism detection in colony forming units (CFU) per ml (right arrow). Similar sensors with different LODs are encircled using the same colour. Sensors having the same transducing element with or without additional enhancement are pink. MRL is maximum residue level, CE is capillary electrophoresis, C4D is contactless conductivity detector, FLD is fluorescence detector, HPLC is high performance liquid chromatography, SERS is surface enhanced Raman spectroscopy, (L)SPR is (local) surface plasmon resonance, Au-NP is gold nanospheres, AMS is ambient mass spectrometry, EC is electrochemical and PEC is photoelectrochemical.

Table 1

Synopsis of commercially available kits for all 4 target groups. This table shows key portable commercial kits extracted from supplementary tables 5, 6, 7 and 8. Devices that enable multiplex detection and/or short analyses time combined with varying levels of quantification are given priority. Moreover, sensors fitting both expert and non-expert needs are listed. LFIA is lateral flow immunoassay. AChE is acetylcholinesterase assay. PP2A is phosphatase inhibition assay. (q)PCR is (quantitative) Polymerase chain reaction. ELISA is enzyme-linked immunosorbent assay. IAC is immuno affinity column. A footnote alphabetically listing the abbreviations used for the analytes in the table is added below.

Analyte	Company	Test type	Sensitivity	Analysis time	Website
Pesticides					
glyphosate	Abraxis	LFIA	2.5 ppb	45 min	https://bit.ly/2oMsyQc
OP & CMs	RenekaBio	AChE strip	0.3–3.5 ppm	13 min + sample prep	https://bit.ly/2CqoBKL
Trifluralin, DDT, carbendazim, ethoxyquin	Bioo Scientific (Perkin Elmer)	ELISA	sub-ppb	< 2 h	https://bit.ly/2MUV68U
OP & CMs	MyDx	AChE strip	0.3 – 100 ppm	10 min	https://bit.ly/2Cp7r08
19 different kits	Creative diagnostics	ELISA	sub-ppb	< 2 h	https://bit.ly/2M3hebV
19 different kits	Abraxis	ELISA	sub-ppb	< 2 h	https://bit.ly/2oMsyQc
atrazine, simazine	Silver Lake Research cooporation	LFIA	< 4 ppb	10 min	https://bit.ly/2NmQurl
Aquatic toxin					
Free Microcystins	Abraxis	LFIA	0.3 µg/l	30 min	https://bit.ly/2PIg7Ic
Domoic acid	Neogen	LFIA	20 mg/kg shellfish	10 min	https://bit.ly/2L1wENa
OA and DTX derivatives	Neogen	LFIA	160 µg/kg shellfish	15 min	https://bit.ly/2L1wENa
STX	Neogen	LFIA	800 µg/kg shellfish	5 min	https://bit.ly/2L1wENa
Microcystins	Zeulab	PP2A	LOQ 0.25 (µg/l)	30 min	https://bit.ly/2OK47xP
Microcystins And Nodularins	Enzolifesciences	ELISA	0.1 (µg/l)	150 min	https://bit.ly/1SP1ubv
STX	Beacon	ELISA	0.02 µg/Kg shellfish	90 min	https://bit.ly/2vR0yPm
OA and DTX derivatives	Beacon	ELISA	0.2 µg/kg shellfish	90 min	https://bit.ly/2vR0yPm
Domoic acid	Zeulab	ELISA	LOQ 2000 µg/kg shellfish	60 min	https://bit.ly/2OK47xP
Mycotoxin					
AFB1/2, AFG1/G2, OTA, FUM, EA, DON, T2, ZEN, DAS, Paxilline	Randox	Chemiluminescent Biosensor	0.25–100 ppb	30–90 min	https://bit.ly/2NNqcvs
ZEN	Vicam	IAC + fluorescence reading	0.1 ppm	15 min	https://bit.ly/2wjzOGP
OTA	Europroxima	ELISA	0.25–1ppb	30 min	https://bit.ly/2ol5brg
DON	Neogen	ELISA	0.1 ppm	10 min	https://bit.ly/2MpWb8g
AFM1	Elabscience	ELISA	0.05ppb	85 min	https://bit.ly/2PL17Iz
T-2 / HT-2	R-biopharm	LFIA	50 ppb	5–10 min	https://bit.ly/2CqI1tH
FUM	Envirologix	LFIA	0.2 ppm	5 min	https://bit.ly/2NlWElt
Microorganisms					
Salmonella	Eurofins	ELISA	1 CFU/25 g	36–46 h	https://bit.ly/2Q8TiqI
Listeria					
E. Coli	DuPont	LFIA	1 CFU/25 g	8–20 h	https://bit.ly/2wO8roD
Salmonella					
Listeria					
Salmonella	DNA Diagnostic	qPCR	1 CFU/25 g	5.5 h	https://bit.ly/2oKpCnm
E. coli	Pall Corporation	PCR	1 CFU/25 g	6–18 h	https://bit.ly/2oKqf0c
Salmonella					
P. aeruginosa					
S. aureus					
C. albicans					
B. cepacia					
E. coli	BIO-RAD	Plating	1 CFU	48 h	https://bit.ly/2NkHRxD

Footnote Table 1: AF- is various Aflatoxins (AF-M1, -B1–2, -G1–2). CM is carbamates. DAS is Diacetoxyscirpenol. DDT is Dichlorodiphenyltrichloroethane. DON is Deoxynivalenol. DTX is dinophysistoxin. EA is ergot alkaloid. FU is Fumonisin. HT-2 is trichothecene toxin HT2. OA is Okadaic acid. OP is Organophosphates. OTA is Ochratoxin A. STX is Saxitoxin. T-2 is trichothecene toxin T2. ZEN is Zearalenone.
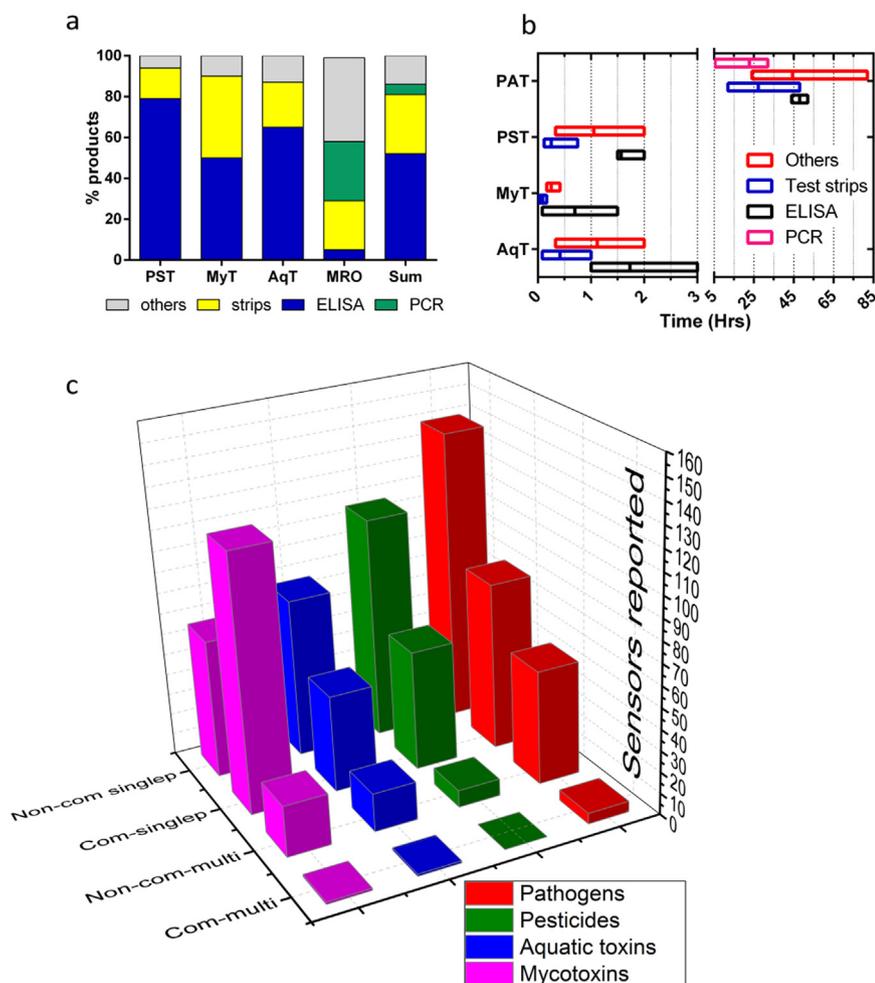
Fig. 4. *Commercially available sensors included in TEST. a)* Analytical method classification of commercially available tests per target. Percentages of each analytical technique are shown per target group. Other (grey) are enzymatic assays, bacterial plating, phage engineering, immune affinity columns, chemiluminescence and fluorescent receptor binding assays. Strips (yellow) are all lateral flow and flow through devices disregarding the recognition element applied. ELISA (blue) are all standard ELISAs. Polymerase chain reaction (PCR) methods are all PCR methods including isothermal PCR, quantitative PCR and real time quantitative PCR. Sum shows average for total amount of commercially available kits for all target groups. b) Timeline showing average (mid-point) and min and max values (bar ends) per analytical technique per target group in hours. Analytical technique classification is the same as in a. In both a and b AqT is aquatic toxins, PAT is pathogens, MRO is microorganisms, PST is pesticides and MyT is mycotoxins. c) Number of commercially available kits is compared with sensors (single and multiplex) mentioned in the literature that potentially could become commercial products (named non-commercial). To avoid unfair comparison all analytical techniques deemed unlikely to become commercialized (chromatographic, infrared, next generation sequencing, and mass spectroscopy based methods) were excluded from the non-commercial sensor groups.

studies focused on 5 specific compounds in each case. The variety is particularly low in the pesticides case, where all the analytes belong to the organophosphorus and carbamate insecticide families sharing a common neurotoxic mode of action using cholinesterase assays. After classification it was attempted to further analyse the quality of the methods reported. Quality characteristics, for instance precision and trueness are indispensable for the analytical evaluation of a method and determine whether the method is fit for purpose. However, sensitivity maybe an equally crucial validation feature regarding food contamination detection as the obtained LODs should be below the regulated maximum residue levels (MRLs) or as low as reasonably possible in the case of an absence of legislation. Thus, the LODs for the various methods included in the TEST database are used as an, albeit somewhat rough, quality indicator here. In Fig. 3c the LODs of various detection methods are shown on a scale of magnitude for each target group. Here detection methods were not classified according the common analytical principle but rather by a combination of transducer elements, recognition elements and use of nanomaterials. This classification was chosen to highlight the effect of adding recognition or nanomaterial elements into a sensor (Fig. 3c, pink sensors) can have on its sensitivity. Perhaps the most striking revelation of Fig. 3c is the sub-ppt sensitivity that can be reached by EC sensors which was only exceeded by fluorescence sensors in one case (pathogens) (Cho et al., 2014b). However, such LODs might be misleading if they were calculated in buffer and not verified in real matrix, which, is sometimes the case (Zhao et al., 2015; Epifania et al., 2018; Lillehoj et al., 2014). Regarding chromatographic methods, the hyphenation of the chromatographic system with various types of detectors strongly affects method sensitivity with more costly MS detectors prevailing over conventional detectors such an ultraviolet-

visible detector (UV–Vis) or a flame ionisation detector. In fact, LC-MS showed LODs rarely exceeding 10 ng/g regardless of the analyte or matrix used. The sensitivity of other techniques however can highly vary between target groups. SERS, in particular, seems to be in this category, showing the sensitive detection of pathogens, with LODs under 100 CFU/ml (Cho et al., 2014a) while sensitive pesticide detection remains a pickle. The latter might be because pesticide detection was performed on the peel of the fruit. However, this seems to be no impediment to reach better LODs in some cases, as it was proved by Yang et al. who were able to detect thiram at 15 pg/kg (Yang et al., 2014). This contrasts with the results found by Tang et al., whose sensor LOD was 150 µg/kg (Tang et al., 2013). The reason for the 7 orders of magnitude difference is unclear but might be related to the nanostructures applied which can strongly affect the enhancement factor observed by SERS which can result in the high sensitivity of SERS sensors (Le Ru and Etchegoin, 2013). Interestingly, across all target groups, nanomaterials seem to be a major factor in effecting sensitivity (pink sensors Fig. 3c). Striking examples were the multiplexed method for the detection of 5 chemical contaminants with LODs of 0.1–5 ppb in drinking water by using monoclonal antibodies conjugated to gold nanospheres (Xing et al., 2015), a plasmonic ELISA for aflatoxin-B1 detection, using gold nanorods with IC_{50} of 22 pg/ml (reported as 32 fold lower than the LOD of conventional ELISA) (Xiong et al., 2018) and a device for pathogen detection which implemented superparamagnetic nanoparticles with a LOD as low as 158 CFU/g in seafood (Liu et al., 2015). However, the targets and matrixes used for these sensors were different. To fairly compare the LODs of such nanomaterial systems between each other and against conventional systems one should, of course, 'compare the comparable'. Thus, all sensors analysing

microcystins were filtered on the term "microcystin" in the analyte column of the supplementary table 2 as an example. This produced 30 records. Eight of the top 10 sensors (classified by LOD) use nanomaterials, with a capacitive immunosensor using gold nanosphere conjugated antibodies which ranked highest with a LOD of 20 pg/L in freshwater (Lebogang et al., 2014) and a photoelectrochemical aptasensor using graphene in third place with an LOD of 30 pg/L in fish (Du et al., 2016). The 10 least performing sensors (in terms of LOD) are either chromatographic or optical sensors with the latter type at the last position with an LOD of 130 μg/L in algae extracts (Brothier and Pichon, 2013). Thus, there are 7 orders of magnitude difference in the LODs for this analyte between sensors while matrixes employed were similar. This comparison clearly showcases the potential high sensitivity using EC sensors combined with nanomaterials which was observed throughout all target groups.

3.3. Commercially available sensors and kits

In total, 309 commercially available detection systems were identified (47 for aquatic toxins, 57 for pesticides, 82 for pathogens and 123 for mycotoxins). These were classified regarding the TEST classification criteria and analytical parameters reported by the test kit providers (supplementary info tables 5–8). A short synopsis, showing key commercial detection systems for each target group, is shown in Table 1. Fig. 4a shows the analytical techniques used per target group. Clearly, ELISA is the predominant test on the market followed by test strips (regardless the recognition element used) for all target groups except microorganisms, where PCR and bacterial plating predominates. For pesticide analysis there are several cholinesterase assays, which, in our opinion, can improve the current status as they can detect both organophosphates and carbamates. However, cholinesterase assays are sometimes not so sensitive. For example some advertised commercial ones had reported LODs that do not meet MRLs set in the EU (Pesticide Detection Test Cards by RenekaBio, Agri-Screen® Tickets by Neogen and OrganaDx by MyDx). Others however (by Envirologix & MyBioSource) did meet EU set MRLs. Lateral flow immunoassays (LFIA) (Abraxis) and ELISA (Bioo Scientific, Creative Diagnostics & Abraxis) tests can also detect at MRLs. Although the library of detectable pesticides using these antibody based techniques is more limited. For mycotoxins, ELISA, LFIAs and fluorometric tests are popular. Fluorometric tests use immunoaffinity columns read by a reader which the company typically offers. For example, Romer labs offers all 3 kit types for a variety of mycotoxins. Moreover, they equally offer LC-MS analyses. Other companies that offer these kits are Vicam, Envirologix and Neogen. Finally, R-biopharm is using smartphone technology and LFIAs for mycotoxin detection thus creating a portable and user-friendly piece of equipment for mycotoxin detection. For aquatic toxins the variety is limited as well with ELISAs and strip tests making up 85% of the total (Fig. 4a). Other kits include non-portable phosphatase inhibition, cholinesterase and fluorescence based tests. Of these, especially the phosphatase inhibition test of Zeulab (MICROCYS) stands out since it claims detection of all toxic microcystin congeners with an LOQ of 0.25 μg/L, 4 fold lower than the limit advised by the WHO. Moreover a similar phosphatase inhibition test developed by Zeulab (OkaTest) is a inter laboratory validated kit for diarrhetic shellfish poison detection (Turner and Goya, 2016). For spoilage organisms, no commercial kits exist currently, except perhaps the Peel Plate Microbial Test from Charm sciences (a non-selective bacterial media to detect potential shelf life issues (i.e. spoilage organisms). Other tests to detect spoilage organisms are electronic noses (Airsense Analytics) which detect volatile metabolites belonging to microbes, and a combination of microbiological techniques (catalase test, Kovac's reagent strips, selective agars) to identify the bacteria. However, none of these methods have developed to a commercially available kit. For pathogens especially non portable quantitative PCR has proven its efficiency going through validation tests, reaching very good LODs. On the other hand several consumer

friendly strip tests have also been developed (Table 1). Importantly, all commercial sensors targeting pathogens require enrichment steps often prolonging analysis times over 24 h (up to 48 h) making this target group by far the slowest in terms of result generation with the biggest time differences between tests (Fig. 4b). The quantitative PCR based *Salmonella* Velox test of DNA diagnostics (Table 1) is the quickest with approximately 6 h test time to detect *Salmonella* at a LOD of 1 CFU/25 g of raw meat or fish. As for aquatic toxins, pesticides and mycotoxins, average analysis times are less with mycotoxins having the fastest tests with some strip tests that can be performed in less than 5 min and some ELISAs marketed with analysis times under 10 min (Fig. 4b). This showcases that, in theory, swift ELISAs could equally be developed for aquatic toxin and pesticide detection. Moreover, the group "other" of mycotoxins, which mainly includes immunoaffinity columns, equally features short analysis times. This might be caused by greater competition in the mycotoxin commercial tests kit sector which was indeed the group with the most commercial kits available, even more then there are novel sensors reported in the identified literature (Fig. 4c). Overall, several interesting gaps came to light when commercial kits are compared to sensors reported in the scientific literature. First off, most of the sensors have been developed by large and established companies. This raises the question if smaller companies simply do not participate much in the development of food screening tests, or small (spinoff) companies remain overlooked. If the last of these causes is the case, increased visibility through orderly classification of such systems might improve upon the current situation. The second gap identified is the lack of availability of commercial multiplex sensors compared to the many reported sensors in the literature (Fig. 4c). Indeed, very few multiplex commercial kits are on offer such as the Myco10 array from Randox Laboratories (Myco10 array) for 10 mycotoxins, and another for pesticides developed by Pall Corporation to detect *Escherichia coli Salmonella* and *Listeria* simultaneously using PCR technology. Most other multiplexed technologies for pathogen detection focus on the detection of closely related pathogens (i.e. different strains from the same species). As for aquatic toxins and pesticide detection, no real multiplex tests were identified at all. Finally, and perhaps most strikingly, no commercial test kits using EC and/or nanomaterial components (including upconversion nanoparticles, graphene, carbon nanotubes, local surface plasmon resonance, SERS or microfluidics) were identified for either of the target groups. Nonetheless, EC and nanomaterial enhanced sensors clearly seemed popular and promising when the scientific literature was analysed. Thus the question could be raised if these sensors will be able to bridge the apparent valley of death between research and commercialisation.

4. Conclusions

The End-user Sensor Tree is the first interactive platform for food contaminant sensors providing a novel classification, based on endusers needs, approach, maximizing the available information for commercial sensors and sensors in development. Rapid, on-site and sensitive quantitative detection is still an almost unreachable dream with lab-based instrumental methods showing a more robust and reliable analytical performance. However, nanomaterial enhanced and EC based sensors show great potential due to their ability to reach high sensitivity and provide semi-quantitative results. Unfortunately, these novel sensors seem still far away from being implemented in real life applications. Test strips however, do seem to successfully bridge the perceived valley of death with many examples of rapid tests requiring little or no expertise. Moreover, rough quantification using a special reader or even a smartphone camera, seems possible. Another step forward to rapid screening is the use of multiplexing. Unfortunately marketing of multiplex tests has equally shown little progress and scientific reports on this are lacking. Overall, it would appear, more effort from the scientific community needs to be invested in the validation of the developed systems. Perhaps capturing the market through the sales

of such systems through spinoffs might further boost development. The lack of commercial novel sensors could also be an effect of the unwillingness of companies to invest into a leap into the unknown. Moreover, it is hoped that TEST will increase sensor promulgation overall and the aim is to further grow TEST into a fully comprehensive sensor platform for numerous analytes in various matrixes providing independent expert advice stretching over both the food and medical sectors. Such development can equally lead to more rapid acceptance of novel systems and provide stakeholders with a means to actively communicate and shape the future of contaminant screening together. Other enthusiasts and stakeholders may wish to contribute to this substantial quest. Ultimately, this might lead to a more time effective and delineated bottom-up biosensing environment replacing the current top-down laboratory-based system.

Acknowledgements

Conflict of interest

The authors declare that there are no financial or commercial conflicts of interest.

Credit author statement

JN was the main author writing the original draft. AT and JL contributed to the writing, reviewing and editing of the original draft. JN was responsible for the data curation. JN, AT, PN, and JL contributed to the investigation in the literature review, preparation of the supplementary tables and in the reviewing and editing of the TEST repository. YZ created the website and decision tree software. CE, KR, CC, HZ and JH provided advice and assisted in reviewing and editing the final document. KC contributed the concept to understand the end user goals, supervised this work in providing guidance and assistance in the investigation and contributed to the review and editing of the draft and final manuscript.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.bios.2019.01.055.

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