



A 3D printed smartphone optosensing platform for point-of-need food safety inspection



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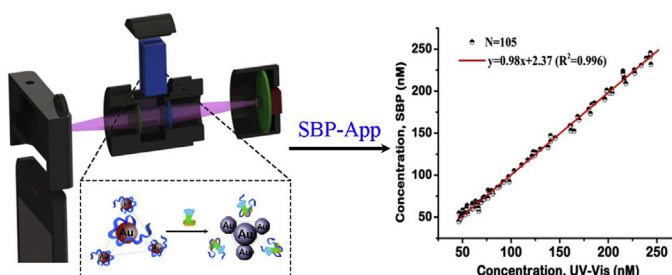
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HIGHLIGHTS

- A rapid and cost-effective smartphone-based method for point-of-need food safety inspection.
- Excellent reliability and applicability of the method for STR quantitation in complicated samples.
- A two-color ratiometric method using aptamer-conjugated AuNPs as colorimetric indicators.
- Fabricated of the smartphone optosensing platform using facile 3D printing technology.
- A user-friendly APP for automatic image processing and result display and sharing.

GRAPHICAL ABSTRACT



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ABSTRACT

The deficiency in rapid and in-field detection methods and portable devices that are reliable, easy-to-use, and low cost, results in the difficulties to uphold the high safety standards in China. In this study, we introduce a rapid and cost-effective smartphone-based method for point-of-need food safety inspection, which employs aptamer-conjugated AuNPs as the colorimetric indicator, and a battery-powered optosensing accessory attached to the camera of a smartphone for transmission images capture. A user-friendly and easy-to-use Android application is developed for automatic digital image processing and result reporting. Streptomycin (STR) is selected as the proof-of-concept target, and its specific quantitation can be realized with a LOD of 12.3 nM ($8.97 \mu\text{g kg}^{-1}$) using the reported smartphone-based method. The quantitation of STR in honey, milk and tap water confirm the reliability and applicability of the reported method. The extremely high acceptance of smartphone in remote and metropolitan areas of China and ease-of-use of the reported method facilitate active food contaminant and toxicant

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screening, thus making the implementation of the whole food supply chain monitoring and surveillance possible and hence significantly improving the current Chinese food safety control system.

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1. Introduction

Food safety is one of the major concerns in every country regardless of the economic and social development. A one-year active surveillance on 10,000,000 people for food safety risk assessment conducted by China National Centre in 2011 revealed that one in every 6.5 people contracts foodborne disease [1,2]. In recent years, China has turned its attention from food supply to food safety, because the frequent occurrence of scandals such as recycled waste cooking oil, illegal additives, and pork disguised as beef, has triggered the growing unrest over food safety. Therefore, it is of vital importance to develop a comprehensive monitoring and surveillance system that not only focuses on the food end products but the whole food supply chain.

Due to the high accuracy and precision, expensive instrumentations such as high performance liquid chromatography (HPLC), gas chromatography (GC) and mass spectrometry (MS) are extensively applied to food safety inspection. However, those methods suffer from complicated sample preparation, professional personnel and long analysis time, and are unsuitable for point-of-need food safety inspection, especially in resource-constrained regions [3–5]. Obviously, the implementation of the whole food supply chain monitoring and surveillance demands rapid and in-field detection methods and portable devices that are reliable, easy-to-use, and low cost.

Currently, the most often used in-field detection method for food safety inspection is the immunoassay, for example, immunochromatographic test strips which always give qualitative results [6], and enzyme-linked immunosorbent assay (ELISA) kits which can give quantitative results but require a second instrumental determination [7]. More importantly, the current immunoassays cannot fulfill the prerequisite of ultrahigh sensitivity for food safety inspection, and the expensive antibodies and cross-reactions with other compounds in food matrix are still the main limitations [6,7].

Aptamers are single-stranded oligonucleotides isolated through a combinatorial biology technique called SELEX, and have attracted tremendous attention in analytical and clinical chemistry due to their high stability, affinity and specificity, ease of modification at arbitrary positions, and small size [8–10]. Especially, aptamers can in principle be selected for any given target, and their chemical synthesis is well developed and hence cost-effective. In the meantime, gold nanoparticles (Au NPs) have become ideal platforms for developing easy-to-use and sensitive sensing systems for quantitation of various target analytes in complicated matrix, owing to its simple and straightforward synthesis, the distance-dependent absorption property, and non-interference with conjugated molecules [11–18]. Particularly, the extremely high extinction coefficient and distance-dependent absorption property make Au NPs appealing colorimetric indicators for chemical and biological sensing. Because of the merit of visualization and hence the independence of large equipment, aptamer-conjugated AuNPs offer great promise for the development of cost-effective colorimetric methods and portable devices for in-field detection, thus implementing the whole food supply chain monitoring and surveillance.

Very recently, thanks to their excellent built-in equipments such as high-resolution digital camera and advanced multi-core processor, smartphone can be extended *via* the addition of accessories

to develop the portable and easy-to-use “all-in-one device” for imaging and sensing, thus eliminating the need for separate detectors and laptop computers for data processing [19]. Taking advantages of smartphone's optical imaging hardware that recently reach to more than 40 million pixels in their digital camera systems, various optical detection technique, for example, microscopy imaging [20], colorimetric [21–30], fluorescent [31–36], surface plasmon resonance (SPR) [37], and bio/chemiluminescent [38,39] detections, have been successfully integrated with smartphone for different biochemical detections. Particularly, colorimetric detection can be performed with smartphone alone by quickly analyzing the color images captured by its digital camera, thus becoming the most promising approach for point-of-need analysis.

To meet the requirement of timely and in-field detection for point-of-need food safety inspection, we herein develop a 3D printed smartphone-based platform (SBP) for the sensitive and specific quantitation of food-safety related markers, which uses aptamer-conjugated AuNPs as the colorimetric indicator and a smartphone for light detection and data processing. A user-friendly Android application (App) is developed to analyze the images captured by a smartphone rear camera, report and share the detection results. In this study, streptomycin (STR) is used as the proof-of-concept target, where its specific recognition is realized by using an anti-STR aptamer, and its concentration is indicated by the ratio of the absorbance at 625 nm (A_{625}) to that at 520 nm (A_{520}) which is consistent with the color change of AuNPs solution. The reliability and applicability of the SBP is assessed with models and real samples such as honey, milk and tap water. The STR quantitation results obtained using the SBP agree well with both those obtained using a laboratory UV-Vis absorption spectrometer and those obtained using a LC-MS, demonstrating the high sensitivity and comparative accuracy of a reliable and cost-effective smartphone spectrometer. Development of affordable, accurate, and easy-to-use methods would facilitate actively screening food contaminants and toxicants, thus significantly improving the current Chinese food safety control system.

2. Experimental

2.1. Chemicals and apparatus

The anti-STR aptamer [40], 5'-TAGGGAATTCGTCGACG-GATCCGGGGTCT GGTGTTCTGCTTGTTCGTCCGGTCGTCTGCAGGTCGACGCAT GCGCCG-3' was purchased from Sangon Biotech Co. Ltd (Shanghai, China). STR, kanamycin (KANA), chloramphenicol (CAP), tetracycline (TC), gentamicin (GM), sodium dihydrogen phosphate, disodium hydrogen phosphate, and sodium chloride were in analytical reagent grade and purchased from J&K (Beijing, China). Ultra-pure water purified by Milli-Q plus system (Millipore Inc, Bedford, MA) was used for preparing all solutions in this study. Absorption spectra were collected with a Beckman DU730 UV-vis spectrometer. The quantitation of STR in honey and milk by national statutory standard method was performed with a LC-MS system (API 4000, Applied Biosystems, USA).

AuNPs with a diameter of 13 nm used in this study were prepared in our lab using the well-established protocol [18], and were characterized by transmission electron microscope (TEM, JEOL JEM

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