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REVIEW ARTICLE

# A review of recent progress in the application of Raman spectroscopy and SERS detection of microplastics and derivatives

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## Abstract

The global environmental concern surrounding microplastic (MP) pollution has raised alarms due to its potential health risks to animals, plants, and humans. Because of the complex structure and composition of microplastics (MPs), the detection methods are limited, resulting in restricted detection accuracy. Surface enhancement of Raman spectroscopy (SERS), a spectral technique, offers several advantages, such as high resolution and low detection limit. It has the potential to be extensively employed for sensitive detection and high-resolution imaging of microplastics. We have summarized the research conducted in recent years on the detection of microplastics using Raman and SERS. Here, we have reviewed qualitative and quantitative analyses of microplastics and their derivatives, as well as the latest progress, challenges, and potential applications.

**Keywords** Micro-/nanoplastics · Raman spectroscopy · SERS detection

## Introduction

Microplastics (MPs) are plastic particles with a diameter of less than 5 mm [1]. Depending on their source, they can be divided into primary and secondary microplastics [2]. Primary microplastics are those that are manufactured, such as microfibers from clothing and abrasive particles from everyday items such as toothpaste. Secondary microplastics are produced as a result of the use, aging, and photodegradation of larger plastic items such as bags, bottles, and other packaging. In 2020, more than 1.56 billion masks were thrown into the ocean [3]. Nanoplastics(NPs) are plastic particles less than 1 μm in diameter [4]. The main polymer types of micro/nanoplastics are polystyrene (PS), poly ethylene (PE), polypropylene (PP), polyamide (PA), polydopamine (PDA), and polyethylene terephthalate (PET) [5–8]. Microplastics are widely distributed in the environment such as water bodies [9], soil [10], and the atmosphere [6]. Recently, the discovery of microplastics in global oceans, lakes, and rivers has greatly increased. Researchers have investigated the distribution of microplastics in different water environments, including the Bohai Sea of China [11], the South China Sea [12], the Mediterranean Sea [13], the North Sea [12], the Pacific [14], the Arctic [15], the Atlantic [16], and the Indian Oceans [12], the Great Lakes [17], and the Yangtze River of China [18]. According to different studies, various degrees of microplastic pollution have been found. Kristina et al. [16] studied the abundance of marine microplastics ≥ 10 μm in the North Atlantic and found that the highest concentration could reach 501 items·m$^{-3}$. Zhao et al. [19] found that the abundance of microplastics in the surface seawater of the Yangtze River Estuary in China could reach $4 \times 10^3$ items·m$^{-3}$. Microplastics are not only found in water but also in soil [10], beaches [20], and even in Antarctica [21].

Due to the slow natural degradation of plastics, the amount of microplastics will accumulate in the environment. According to the statistics [22], the growth rate of

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global microplastics has exceeded that of carbon emissions [23]. Overtime, microplastics will break down into smaller and smaller fragments and eventually become nanoplastics [24]. Microplastics and their derivatives pose hazards to the human body [25] because they can be found in the digestive system, respiratory system, and circulatory system [26] and travel through the blood to other organs or tissues, including the brain [27], placenta [28], and breast milk [29]. Microplastics also can be combined with toxic pollutants, such as bisphenol A [30], benzo(a)pyrene [31], cadmium [32], and pesticides [33], making it easier for microplastics to spread and cause greater harm.

Researchers have explored detection methods to learn more about how to detect MPs [34]. In the early years, researchers mainly used optical microscopes to detect microplastics, limiting the detection of only larger sizes of microplastics [35]. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) improved the minimum detection size of microplastics, but these methods cannot provide chemical information, such as the type of polymer [36]. Thermal analysis is another common method, which decomposes the polymer at high temperatures and then analyzes the product to obtain information about the sample composition [37]. However, thermal analysis cannot obtain physical information such as the size and color of small particles and requires the destruction of the sample, which is not applicable for subsequent analysis [38].

Vibrational spectroscopy technology, including Fourier transform Infrared Spectroscopy (FTIR) and Raman spectroscopy, is the most frequently employed method to detect microplastics (MP). It provides data on the size, variety, quantity, and distribution of plastic particles in the sample. Spectral detection technology's foundation is radiation-molecular vibration interaction. As MP concludes synthetic hydrocarbon polymer, vibrational spectroscopy can utilize the distinct molecular fingerprints of the specimen for the identification of its chemical composition [37]. FTIR is reliant on infrared absorption determined by the change in permanent dipole moment in the chemical bond, allowing it to the detection of polar functional groups in the sample [39]. FTIR's application in MP detection is restricted by the requirement of dry samples [40] and the analyte size exceeding 20 µm [38] in general.

Raman spectroscopy is founded on the principle of photons inelastically scattering while interacting with matter. This technique can provide molecular vibration data for the studied sample [41], which corresponds to the medium's initial and final vibration energies, respectively. Raman spectroscopy is an effective method for analyzing smaller MPs in water and wet samples, as it is not susceptible to water interference. This technique has been utilized to identify common MP polymers, including PE, PP, PET, and PS [20]. By combining microscopy with Raman spectroscopy, MP detection, identification, and quantification can be performed quickly and easily with a certain degree of automation. This enables chemical imaging of microplastics and facilitates high-throughput analysis.

Generally, the Raman signal of a sample is feeble, and it can only be gauged in solid samples or highly concentrated aqueous solutions [42]. However, if the sample being measured is absorbed on a metal surface (typically gold or silver) with nanoscale roughness, the Raman signal of the sample molecules experiences significant amplification, which is known as surface-enhanced Raman scattering. The spectroscopic detection technique developed based on this phenomenon is known as surface-enhanced Raman spectroscopy (SERS). The benefits of SERS include high-resolution spikes that allow for simultaneous multi-component analysis, non-invasive methodology, uncomplicated sample handling, speedy analysis, in situ analysis, and instrument portability [43]. The primary limitations of SERS include the need for close substrate contact with analytes, substrate degradation, selectivity problems, and issues with substrate reusability and uniformity [44]. SERS can enhance sample characterization, but its reliability and reproducibility are closely related to the distribution area of hot spots. As a powerful material analysis technology, SERS has made great achievements in various fields. The advantages and disadvantages of the above detection methods in microplastic detection are shown in Fig. 1.

Additionally, microplastic and nanoplastic particles exhibit complexity in terms of their type, size, shape, density, and other attributes. Nevertheless, due to significant obstructions in sampling, separation, and detection of microplastic particles, information on contamination by such particles is limited. Thus, a crucial research area aimed at closing knowledge and technology gaps is to identify the composition of microplastics and nanoplastics [45].

Therefore, researching both microplastic and nanoplastic contamination is necessary primarily to identify their composition and structure. Analytical technologies based on Raman scattering, such as normal Raman spectroscopy and SERS, have been employed for the detection of MPs. These non-destructive spectral analysis technologies can detect microplastics qualitatively and quantitatively, and when combined with machine learning, the detection limit for microplastics can be significantly lowered. The use of Raman spectroscopy and SERS can lead to a breakthrough in the study of microplastic contamination, as Raman spectroscopy is well suited to determine the specific type and composition of plastic polymers through spectral pattern recognition.

Here, we highlight the latest developments and applications of Raman spectroscopy and SERS methods for the identification of MPs. The application of Raman spectroscopy and SERS in the detection of microplastics will next

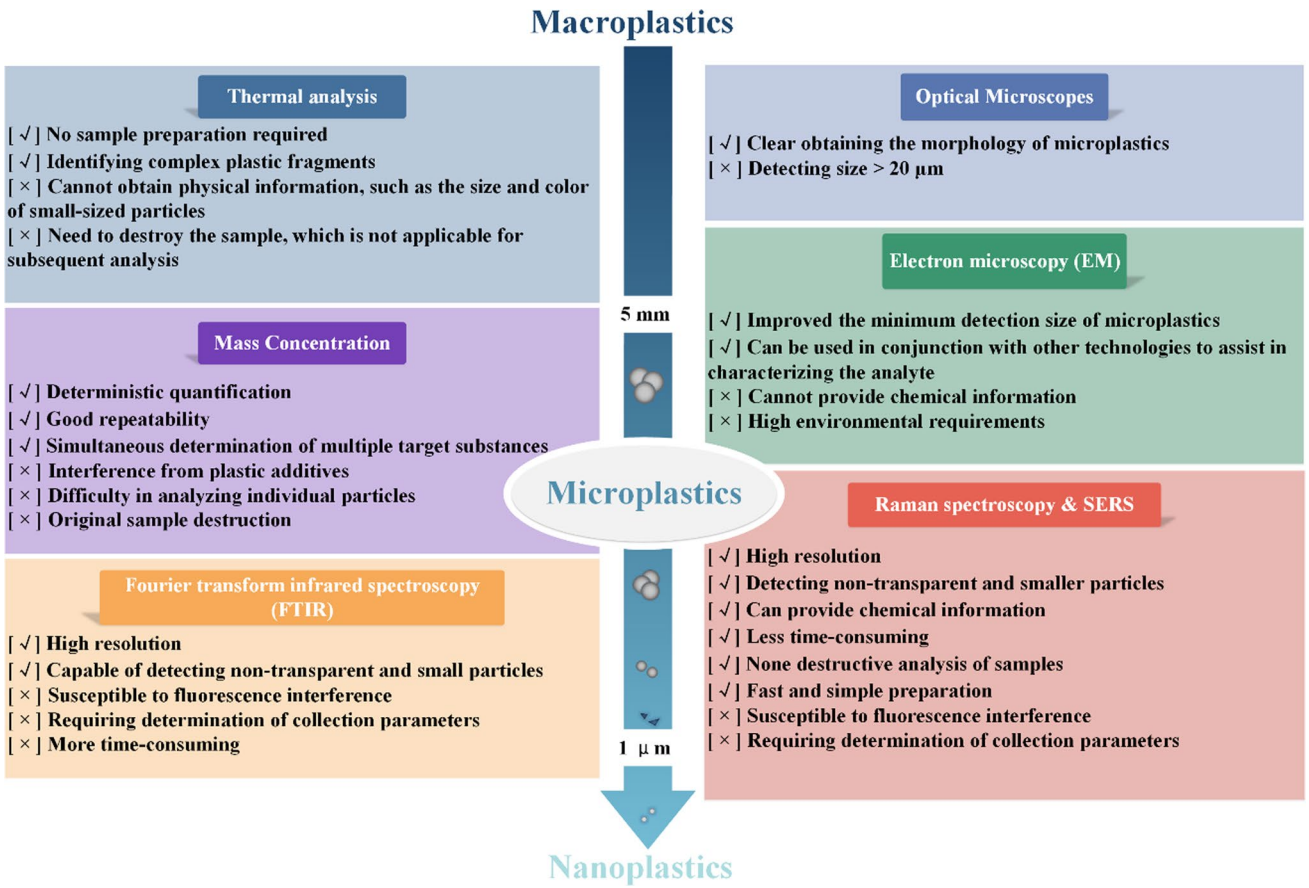


**Fig. 1** Advantages and disadvantages of optical detection technology, electronic detection technology, thermal analysis technology, FTIR, and Raman spectral technology in the detection of microplastics

be discussed after an introduction to the basics of both techniques.

## Research progress in Raman spectroscopy and SERS for the detection of microplastics

### Raman spectroscopy analysis

Raman spectroscopy (named after the Indian physicist Chandrasekhara Venkata Raman) is a vibrational technique based on the inelastic scattering of photons interacting with matter and can provide information about molecular vibration in the sample [41]. It can be divided into two types: Stokes Raman scattering and anti-Stokes Raman scattering, which corresponds to the initial and final vibration energy of the medium respectively. Unlike elastic light scattering, Raman spectroscopy employs inelastic light scattering, which provides material-specific information on its vibrational modes, thereby enabling their identification. As a result, Raman spectroscopy is frequently utilized in chemistry to discern the structural fingerprints of molecules [46].

Hardware for Raman spectroscopy has advanced significantly in recent decades. Samples are illuminated with a laser light source, and the resulting Raman scattering is detected by a spectrometer equipped with a photosensitive detector after passing through the target [47]. In addition, software advancements have emerged in the field of Raman spectroscopy. This technology can now be merged with deep learning and intelligent algorithms to efficiently analyze the sample data collected by Raman spectroscopy devices [48]. Raman spectroscopy has been used for the detection of substances in biological, chemical, and other fields [49].

Raman spectroscopy has been used to identify microplastics and nanoplastics according to recent reports, as shown in Table 1. Unknown particles can be identified by searching for a library of known vibrational fingerprints of microplastics. Raman spectroscopy is not sensitive to water, making it suitable to analyze MPs in water and wet samples. In addition, Raman spectroscopy is an effective technique for determining the type of MPs and has been reported for common polymers such as polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), and polystyrene (PS) [20].

**Table 1** Details of literature available for the detection of MP/NPs using Raman spectroscopy

| Target | Size | Detection environment | Whether came from natural environment | LOD | Excitation wavelength | Power of laser | Reference |
|---|---|---|---|---|---|---|---|
| PE | 303 µm | Tap water and sea water | Yes | 0.09 mg | 785 nm | - | [50] |
| PS, PET, PE, PVC, PP | 1 µm | Soil | Yes | - | 532 nm | 60 mW | [51] |
| PET, PP | 1 µm | Bottled water | Yes | - | 532 nm, 785 nm | 20 mW, 50 mW | [52] |
| PET | 1 µm | Bottled water | Yes | - | 532 nm, 785 nm | 3.2 mW, 5.3 mW | [52] |
| PA, PE, PET, PS, PVC | 4.7 µm | Air | Yes | 2502 particulates·$m^{-3}$ | 785 nm | 19 mW | [53] |
| PP, PE | 10 µm | Marine sediment | Yes | - | 532 nm, 785 nm | - | [54] |
| PE, PET | 108 µm | River sediment | Yes | - | 442 nm | - | [55] |
| PS | 1 µm | Edible oil | Yes | - | 780 nm | 10 mW | [56] |

For example, Sutapa et al. [57] used Raman micro-spectroscopy to analyze the stomach contents of fish from the Pacific Ocean, the laboratory, and the subtropical circulation. Raman micro-spectroscopy is able to detect MPs and provides specific molecular information at the level of a single particle down to 10 µm. The Raman peaks were explained by a combination of fluorescence background removal algorithms to provide size, chemical, and morphological information on microplastic debris.

Raman spectroscopy relies on the change in polarizability of the chemical bond in the sample, so it is sensitive to aromatic bonds, C–H bonds, and C=C bonds. Analysis of microplastics begins with sampling and sample preparation. Different sampling methods can be used depending on the type of sample. Depending upon the nature of the sample, distinct sampling methods should be utilized. Generally, sampling methods include selective sampling, batch sampling, and volume reduction sampling. Selective sampling refers to the direct sorting of MPs after visual recognition. Of course, this method is only practical with larger MPs in the sample. Batch sampling is frequently utilized in practical applications and uses the full volume of the sample, especially for sediments. Volume reduction sampling refers to the method of minimizing the volume of samples, through filtration or sieving and can be used for water and sediment analysis. After large volume and reduced volume sampling, the obtained samples usually require further processing (e.g. density separation, removal of organic matrix) for MP identification.

To avoid interferences in the sample, it is very important to remove organic and inorganic matrices from complex samples [25]. In addition, the selection of appropriate measurement parameters (laser wavelength and power and acquisition time, as well as objective gain and confocal mode) is very important to avoid interference from strong fluorescence and improve spectral quality. The Raman spectral range of MP should cover all the spectral features of the polymer [58] to achieve reliable detection, identification, and quantification of MP.

Additionally, Raman imaging is very time-consuming since it is necessary and usually performed as a point-by-point measurement [26]. To ensure accurate results, the Raman system must be calibrated before collecting the spectrum due to the need for measuring numerous particles. Depending on the instrument, this is usually done using the 520.7 $cm^{-1}$ peak of the silicon wafer and the zero-order correction of the grating used [59].

## Surface-enhanced Raman spectroscopy

In 1974, Fleischmann et al. [60] used Raman spectroscopy to distinguish two types of adsorbed pyridine (a basic cyclic heterodyne compound molecule), which are found on the surface of a silver electrode to mitigate absorption effects. The surface-enhanced Raman spectroscopy (SERS) technique is derived from Raman spectroscopy but has higher sensitivity and unique molecular specificity. In general, the Raman signal of a sample is very weak. It can only be measured in solid samples or highly concentrated aqueous solutions. The Raman signal of the sample molecules is significantly amplified when the sample being measured adheres to the surface of a metal structure (typically gold or silver) with nanoscale roughness. The enhancement mechanism is mainly attributed to two widely accepted mechanisms: electromagnetic enhancement (EM) and chemical enhancement (CM) [61]. The former is based on the excitation of strong electromagnetic fields (localized surface plasmon resonance, LSPR) at the surface of plasma materials (mainly silver and gold nanostructures) [46]. The latter is mainly based on the observation that the intensity of the Raman signal could be increased by the charge transfer between the metal substrate and the molecule which changes the electron cloud density

and polarizability of the molecule. In chemical enhancement, it is mainly considered to be caused primarily by interfacial charge transfer [62], and electromagnetic enhancement is the main cause [43]. Raman scattering is enhanced when plasmonic nanostructures with peaks, narrow gaps, and aggregates act as "hot spots" [63].

SERS and Raman detection can both analyze different sizes of microplastics. However, SERS offers higher sensitivity, lower detection limits, and potentially better quantification capabilities compared to Raman detection. It becomes increasingly challenging to detect smaller particles using Raman detection. SERS utilizes nanostructured substrates to amplify the Raman signal [64], allowing for the detection of smaller microplastic particles. Additionally, SERS may require additional sample preparation steps compared to Raman detection.

As mentioned before, to utilize SERS for microplastic detection, the analysis must commence with the sampling and processing of samples [58]. According to different samples, different sampling methods can be used [65]. To detect analytes via SERS technology, researchers must account for various factors, including the size, shape, composition, orientation, and surface affinity of the nanoparticles, type of substrate, and pH of the solution [66].

In the field of food toxin detection, Neng et al. [67] introduced that SERS technology is commonly used as a substrate for noble metal nanoparticles. SERS offers benefits such as easy preparation, consistent performance, and heightened sensitivity. SERS has the potential to detect toxins, melamine, pesticides, antibiotics, and illegal edible dyes, among others. The development of a sensitive and rapid detection method in clinical samples is crucial for the early diagnosis of infectious diseases. Recently, SERS-based microfluidic devices have captured great attention from numerous researchers due to their tremendous potential to facilitate repeatable point-of-care (POC) diagnosis using SERS. Chen et al. [68] developed a SERS-based aptasensor for highly sensitive and reproducible detection of influenza A or H1N1 virus. This method serves as both a straightforward diagnostic tool for the influenza virus and a dependable detection platform for other viral pathogens.

For more efficient early cancer diagnosis, Rojalin et al. [69] used a simple plasma scaffold composed of a microscale biosilicate matrix embedded with silver nanoparticles to detect circulating nanoscale extracellular vehicles (EVs) for the analysis of SERS analysis of ovarian cancer and endometrial cancer EVs. SERS can also be used to detect contaminants, such as dichlorodiphenyltrichloroethane (DDT), a pesticide classified as a persistent organic pollutant and a possible carcinogen. Marino-Lopez et al. [70]developed a SERS substrate with gold nanoparticles embedded in microporous silica capsules to detect DDT-containing river water and achieved a detection limit (LOD) of 1.77 $\mu g \cdot L^{-1}$. They achieved a detection limit of 1.77 $\mu g \cdot L^{-1}$. Santhoshkumar et al. [71] developed a new SERS substrate by combining silver nanoparticles, graphene oxide, and carbon nitride, and used SERS technology to detect methylene blue and $Hg^{2+}$ in water with high sensitivity.

These examples illustrate the versatility of SERS technology in detecting and analyzing various substances. When employing SERS technology for inspection, the SERS substrate is of utmost importance. Although theoretically, any suitable metal with a rough surface can produce enhancement effects, achieving high, repeatable, and stable enhancement effects is necessary. The two metals commonly used as substrate materials are silver and gold. Solid substrates made of precious metals are suitable for use in microscopes by applying a liquid droplet onto the prepared substrate and observing its surface [72]. Recently, SERS technology has been applied to detect microplastics and nanoplastics in the real environment.

The primary considerations in detecting MPs using Raman and SERS include grasping the practical importance and linking the technology with real-life applications. Figure 2 illustrates the utilization of Raman and SERS technology in detecting MPs.

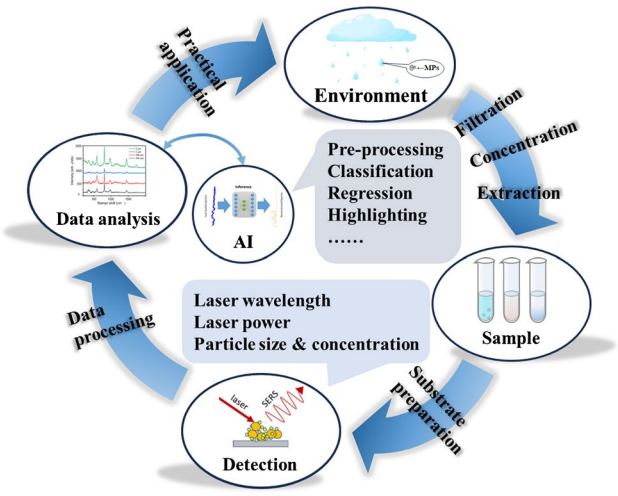


**Fig. 2** Schematic representation of the application of Raman technology in the detection of microplastics. ([73] Copyright (2020) American Chemical Society. [74] (2021) American Chemical Society. [75] (2022) Elsevier)

## Microplastic detection using Raman spectroscopy and SERS

### Detection technology of microplastics based on Raman spectroscopy

Raman spectroscopy, as previously described, is sensitive to variations in the polarizability of the chemical bonds in the sample, making it sensitive to aromatic bonds, C–H bonds, and C=C bonds. Raman spectroscopy is one of the most widely used techniques for the identification and visualization of microplastics. It has been applied to different types of samples, such as deionized water [76], seawater [77], food [78], environmental sand samples [79], and seafood species [80]. As shown in Table 1, Raman spectroscopy-based microplastics detection technology was investigated.

The detection of microplastics based on Raman spectroscopy can be performed in different backgrounds. For example, in the soil, Zahra Sobhani et al. [51]proved that Raman spectroscopy can identify and visualize five different kinds of microplastics, including polystyrene (PS), polyethylene terephthalate (PET), polyethylene (PE), polyvinyl chloride (PVC), and polypropylene (PP). The identified lateral resolution is 1 µm per pixel, which can capture microplastic as small as 1 µm. Their research has demonstrated the advantages of Raman spectroscopy in the detection of microplastics, including less dye addition and sample destruction and the reduced interference of water or organic matter or fluorescence background.

For daily drinking water, microplastics are also ubiquitous. Schymanski et al. [52] used micro-Raman spectroscopy to test and analyze the microplastic content of water in different recyclable and disposable plastic bottles, beverage cartons, and glass bottles purchased, and small (50–500 µm) as well as very small (1–50 µm) microplastic fragments were found in each type of water. Microplastic particles that can be detected by Raman spectroscopy include 30 types of polyester (mainly polyethylene terephthalate PET, 84%) and polypropylene (PP, 7%). According to the water's carbonate content, test samples are divided into different groups. At least 20 small particles (50–500 µm) and very small (1–50 µm) of plastic particles were found in each group of water. This result showed that Raman spectroscopy can be used to detect microplastics under different water conditions. Besides, Ossmann et al. [81] used aluminized polycarbonate filter membranes and micro-Raman spectroscopy to study microplastics in bottled mineral water. Microplastics were analyzed in different packages of mineral water from local grocery stores, and about 90% of the microplastics detected were ≤ 5 µm in diameter, and about 40% were even ≤ 1.5 µm, with a minimum particle size of 1 µm. The range from 2649±2857 per liter in disposable PET bottles to 6292±10521 per liter in glass bottles suggests that other sources of contamination besides packaging need to be considered.

To the best of our knowledge, the e-study by Guo et al. [56] was the first to quantify and identify microplastics in commercially bottled oil. The recovery of PS microplastics from edible oils ranged from 62.73 to 118.00% using a simple hexane filtration method. Four types of oil (olive, rapeseed, sunflower, and coconut) exhibited a substantial particle presence ($1.34\times10^5$ to $5.80\times10^5$ particles per liter). Overall, more than 50% of the particles were identified as microplastics and more than 80% of the particles were smaller than 10 µm. Examples of microscopic images, Raman images, and spectra of PS microplastics are shown in Fig. 3.

In the natural environment, Sutapa Ghosal et al. [57] analyzed the stomach contents of fish living in the Pacific Ocean, laboratory, and subtropical circulation by Raman micro-spectroscopy (RMS). Using Raman micro-spectroscopy for the detection of microplastics, specific molecular information at the level of a single particle can be obtained to identify polymer fragments in the natural environment, including PE, PS, PVC, PET, and PP. Liu et al. [82] used a two-phase ( ethyl acetate-water) system to separate and extract microplastics from beach sand and marine sediment samples, and the qualitative and quantitative analysis of microplastics was conducted by confocal Raman spectroscopy. The samples were analyzed to detect the types and abundance of microplastics.

### Detection technology of microplastics based on SERS

Microplastics are ubiquitous in daily life and their existence has been analyzed in tap water [76], river [76], and sea [83]. Table S1 outlines multiple studies concerning the identification of microplastic particles using SERS.

As previously stated, Raman spectroscopy detects changes in the polarizability of chemical bonds, making it sensitive to aromatic bonds, C–H bonds, and C=C bonds [84]. According to recent research, SERS has already been discussed in many analyses focused on well-known pollutants such as phthalates, plasticizers [84], and xenobiotic contaminants [54]. Compared to Raman spectroscopy, SERS has the following advantages. The advantages of SERS detection technology include a high signal-to-noise ratio and high-resolution peaks.

Simultaneous analysis of multiple components is possible with this technology. Additionally, it allows for non-invasive detection and low-requirement single-particle detection. It also includes the necessary technical terminology abbreviations and maintains a logical flow of information. Moreover,

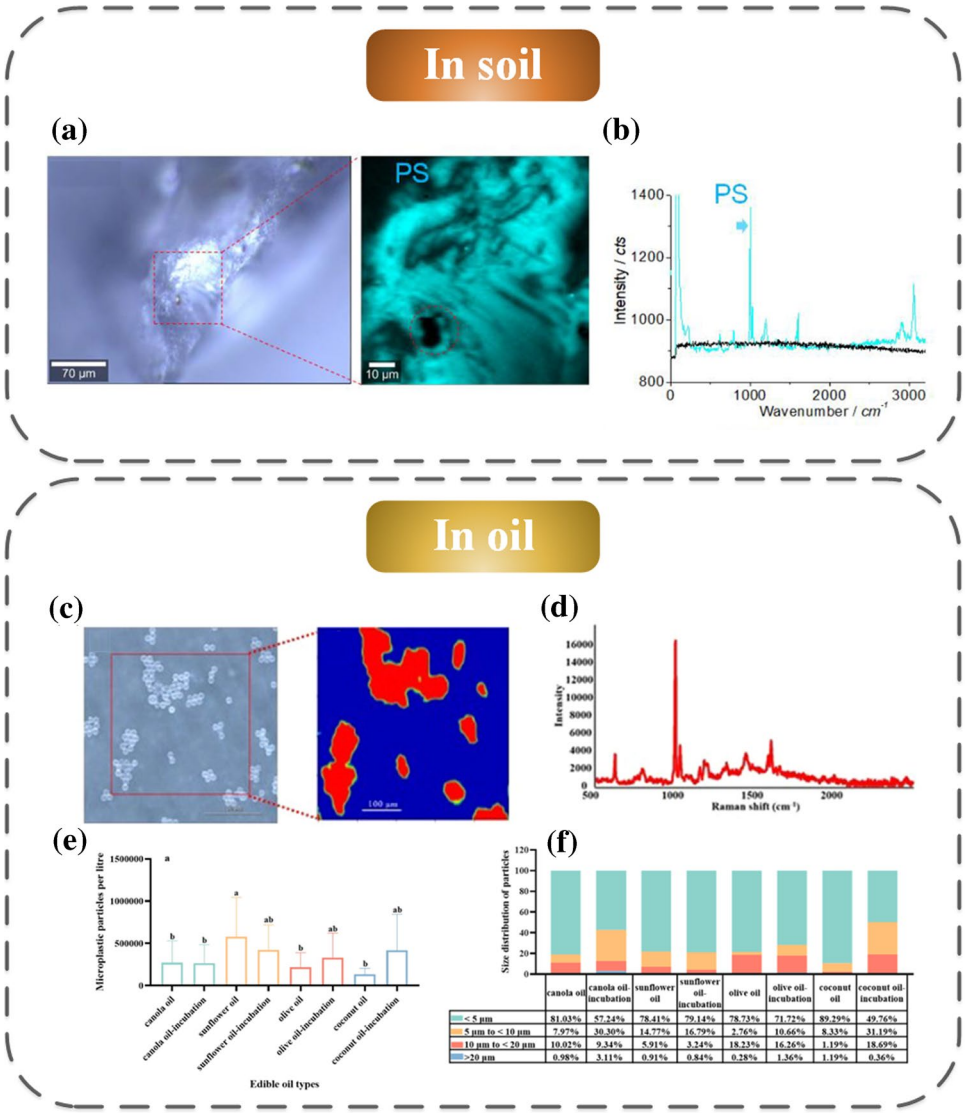


**Fig. 3** Examples of the performance of Raman-based detection of microplastics: **a** Photo and mapping images of PS from the scanning area. [51] Copyright (2019) Elsevier. **b** Typical Raman spectra of PS from the scanning area. [51] Copyright (2019) Elsevier. **c** Microscopic images, Raman images, and spectra of PS images of different sizes and quantities. [56] Copyright (2023) Elsevier. **d** Selected Raman spectra of PS of different sizes and quantities. [56] Copyright (2023) Elsevier. **e** Boxplots of microplastics released from different types of edible oil. [56] Copyright (2023) Elsevier. **f** Size distribution of microplastics released from different types of edible oil. [56] Copyright (2023) Elsevier

in the process of detection, SERS technology has the advantages of easy sample handing, rapid analysis speed instrument portability. Finally, SERS uses molecular fingerprinting to detect and identify MPs or NPs and to realize in situ analysis of analytes [53].

Several factors need to be considered when implementing SERS as a routine sensing technique for analytes. In particular, the intrinsic properties of the metallic nanoparticles, the type of substrate, and the pH of the solution should be controlled to acquire reproducibility criteria [84]. It is crucial to use nanostructured materials for preparation of the SERS substrates, since the metal particles responsible for its operation need to be small compared to the wavelength of the exciting light [43].

The research of new SERS substrate is of great significance to expand the research scope and application field of SERS. The type of nano plasma used and the morphology of the substrate will affect the SPR and molecular adsorption of SERS substrate [8], making them the most important factors affecting SERS. At present, the preparation technology of SERS substrate has become increasingly mature, even enabling the control of the shape and size of nano plasma [85]. SERS hotspots composed of gold and silver nanoparticles are generally hydrophilic, like MPs, so it is important to absorb precious metal nanoparticles uniformly on microplastics to enhance the SERS signal. This can be achieved by changing the substrate structure and using the coffee ring effect [73].

### Metal nanoparticle SERS substrates

In samples of bottled, tap, and river water, Quang et al. [86] used SERS spectroscopy and AuNSs@Ag@AAO substrate to detect micron-grade polystyrene with a minimum diameter of 0.4 μm. A new SERS substrate was created by applying the ultrasound-induced self-assembly technique using

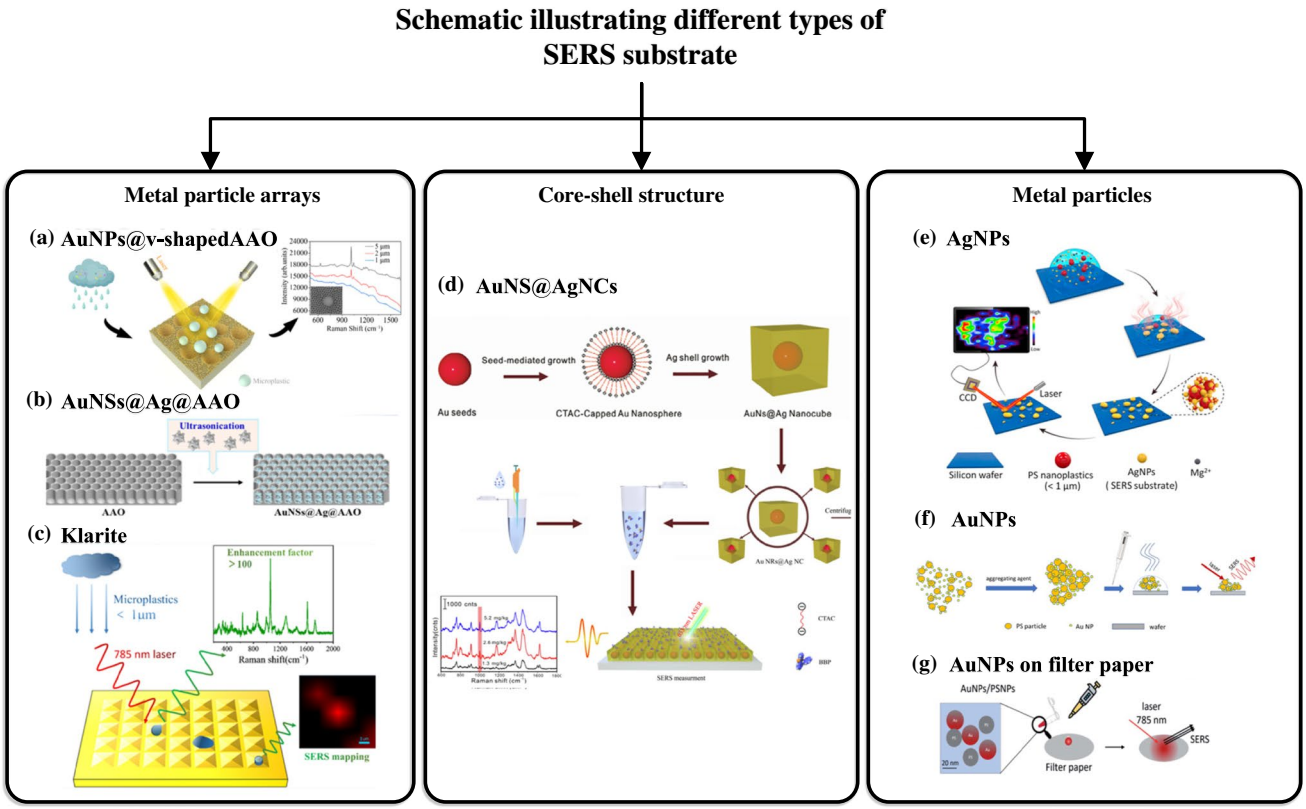


**Fig. 4** Schematic illustrating different types of SERS substrate. **a** Schematic diagram of AuNPs@v-shapedAAO. [87] Copyright (2022) Springer. **b** Synthetic scheme of preparation of the AuNSs@Ag@AAO substrates. The AuNSs@Ag dimers were assembled inside the AAO nanopores [86] Copyright (2020) Elsevier. **c** Schematic diagram of Klarite in SERS detection [73] Copyright (2019) American Chemical Society. **d** Schematic diagram of AuNS@AgNCs in SERS detection. [88] Copyright (2020) Elsevier. **e** Schematic diagram of AgNPs in SERS detection and mapping measurement of PS nanoplastics. [89] Copyright (2020) Elsevier. **f** Schematic diagram of AuNPs in SERS detection. [75] Copyright (2022) Elsevier. **g** Schematic diagram of the system of AuNPs on filter paper in SERS detection. [90] Copyright (2022) Royal Society of Chemistry

the AAO template as a base. The preparation diagram of the substrate is shown in Fig. 4b. The AuNSs@Ag dimer was evenly fixed in the AAO nanopore for sensitive detection of submicron polystyrene MPs. The results showed that 0.4 μm polystyrene MPs can induce plasmon resonance enhancement on the AuNSs@Ag@AAO substrate surface. LOD values of 0.005% were measured in a rapid detection time of a few minutes without sample pretreatment. It is demonstrated that SERS substrate is an effective new sensing platform for trace analysis of MPs at submicron size.

Techniques have been developed to detect river water in real samples. Chang et al. [76] developed a method based on SERS to test the Raman signals of micro-plastics and nano-plastics in bottled, tap, and river water spiked samples, which can detect PS particles as small as 100 nm. For rainwater samples, Liu et al. [87] developed a V-shaped substrate (the schematic diagram is shown in Fig. 4a) for detecting and identifying microplastics, which can detect the PS and PMMA particles smaller than 5 μm. Under different dissolution conditions, Zhou et al. [82] tested real water collected from the Kunyu river. and transformed a dried mixture of AgNPs and PS nanoparticles on a silicon wafer for collecting Raman spectra and was able to detect concentrations as low as 5 $\mu g \cdot mL^{-1}$ (50 nm PS) in river water spiked samples. For biological detection, Li et al. [91] used SERS to achieve a single particle detection of PMMA and conducted testing on carp's spiny muscle tissue.

Xu et al. [73] used Klarite as a substrate for the detection of single microplastic particles in atmospheric or aquatic environments. Klarite is a special SERS substrate (the schematic diagram is shown in Fig. 4c) and is shaped as an inverted pyramid, with an orderly and dense grid structure of cavities (or "pits"). The width of each pit structure is 1.5 μm, making Klarite very suitable for studying nanoparticles. It consists of metal nanoparticles and carbon nanotubes, which enhance Raman spectral signals through the resonance of metal nanoparticles. Using a 785 nm laser excitation, they successfully identified PS and PMMA particles greater than or equal to 360 nm in deionized water in a laboratory environment. Besides, Xu et al. have successfully detected

PMMA and PET microplastic particles with a size as low as 450 nm on Klarite, a commercial substrate made of gold with a dense grid of inverted pyramidal cavities that can produce intense hot spots. This substrate can be used to detect microplastics as small as 360 nm in polystyrene (PS) and polymethyl methacrylate (PMMA) in water, as well as microplastics smaller than 450 nm in ambient air, where the EF of PS reaches 2 orders of magnitude [73].

#### Metal core-shell nanoparticle SERS substrates

Hybrid nanomaterials, consisting of bimetallic nanostructures or nanocomposites, have been used alongside pure noble metals. These materials are effective for Raman-active substrates in biosensors and various environmental applications [92].

For food detection, Wang et al. [88] used SERS combined with plasmonic core-shell Au nanospheres@Ag nanocubes (AuNS@AgNCs) as a substrate to rapidly and sensitively detect butylene phthalate (BBP) in liquor liquid samples. This kind of substrate has a gold nanosphere as the core and a silver nanocube as the shell, whose synthesis schematic diagram is shown in Fig. 4d.

#### Metal nanostructure arrays SERS substrates

In pure water, Caldwell et al. [93] created a substrate in surface-enhanced Raman scattering spectroscopy detection, utilizing spherical gold nanoparticles with 14 nm and 46 nm diameters to improve the scattering signal obtained during Raman spectroscopy measurements. They analyzed polystyrene particles with sizes of 161 nm and 33 nm and poly (ethylene terephthalate) particles with an average size of 62 nm, as a proof-of-concept for the detection of nanoplastic particles by SERS substrates created using colloidal AuNPs. As shown in Fig. 4f, Mikac et al. [75] introduced a technique for detecting polystyrene spheres down to 350 nm in pure water under laboratory conditions. This technique created, characterized, and used gold nanoparticles (AuNPs) of four different types and sizes were created, characterized, and used as a SERS active substrate for the detection of microplastics. Through SEM analysis, the average particle size of spherical samples can reach a minimum of 33.2 nm.

Moreover, Chang et al. [76] used the coffee ring effect to drop the nanoplastic sample on the substrate, and the sample was captured into the pores on the substrate. The prepared nanopore-enhanced Raman substrate was used to detect a single polystyrene nanoplastic with a size of 200 nm and the limit of detecting polystyrene nanoplastic was 5 ppm in the actual water environment. For instance, sea urchin-like gold nanoparticles surrounded by irregular spikes around the core, known as AuNUs, were used as SERS hotspots with polystyrene (PS) particles as probes and it was found that SERS on PS particles can be induced by as few as 1–5 AuNU particles under 785 nm excitation [94].

In the natural environment, Microplastics can also be detected through SERS. In seawater sampled from the actual environment, Lv et al. [77] developed a method based on SERS to test the Raman signals of micro-plastics and nanoplastics in pure water and seawater, using silver colloid as the active substrate, and sodium chloride (NaCl) as the aggregating agent for the qualitative analysis of microplastics and nano-plastics in the water environment. They studied the reinforcement efficiency of plastic particles of different sizes and types by changing the volume ratio between samples and silver colloid, the concentration of NaCl, and the sample with polystyrene (PS), polyethylene (PE), and polypropylene (PP) as the detection target. The Raman signals of micro-plastics and nano-plastics in pure water and seawater showed good enhancement efficiency. The detection method based on SERS overcomes the limitations of micro plastic and nanoplastic in liquid and can detect 100 nm microplastic as low as 40 $\mu g \cdot mL^{-1}$. Under different dissolution conditions, Zhou et al. [89] tested real water collected from the Kunyu river. They transformed a dried mixture of AgNPs and PS nanoparticles on a silicon wafer for collecting Raman spectra and detected concentrations as low as 5 $\mu g \cdot mL^{-1}$ (50 nm PS) in river water spiked samples. The schematic illustration of the SERS mapping measurement of PS nanoplastics is shown in Fig. 4e. A novel filter paper-based SERS method, presented by Shinji Kihara et al. [87], has a detection limit of 10 $g \cdot mL^{-1}$ with a sample volume of 50 mL, and is shown in Fig. 4g.

SERS substrate can also be created by sputtering gold particles on a glass cover [95]. Gold nanoparticles (AuNPs) doped filter paper was used as a flexible SERS substrate to capture MPs in fiber pores. Hot spots generated by AuNPs could enhance the Raman signal of MPs. The flexible SERS substrate has a minimum detection concentration of 0.1 $g \cdot L^{-1}$ for polyethylene terephthalate (PET) in water and a maximum enhancement factor (EF) of 360.5 and has been successful in detecting MPs in tap water and pond water [96]. Analysis was conducted on layer-by-layer assembly of synthetic substrates using positively charged polyelectrolytes and negatively charged AuNPs (14–46 nm), polystyrene particles with sizes of 161 nm and 33 nm, and polyethylene terephthalate particles with an average size of 62 nm were analyzed using substrates. With this technique, plastic particles with a concentration of 10 $\mu g \cdot mL^{-1}$ can be detected [93]. Recent reports indicate that metal-organic backbone (MOF)-cotton fabric (CF) composites infused with silver nanoparticles are effective for detecting simulated microplastics [97]. Additionally, gold-coated vertically aligned carbon nanocones have been identified as suitable SERS substrates for hybrid microplastic detection [98].

To conclude, the qualitative detection of microplastics benefits greatly from the various applications of SERS. SERS is versatile enough to be used in a variety of settings and can detect particles as small as nanometers.

## Quantitative detection technology of microplastics based on SERS and combination of SERS and other technologies in the detection technology of microplastics

Some accurate SERS quantitative analysis methods have been reported [85, 99]. Nowadays, highly sensitive and selective SERS quantitative analysis methods for nano-alcohol have been developed with good accuracy [100].

For example, nanoenzymes and bioenzymes are used to catalyze the reaction of nanoparticles and molecules to amplify the SERS signal, and the aptamer is used to improve selectivity [101]. In Caldwell's study [93], polystyrene particles with sizes of 161 nm and 33 nm and poly (ethylene terephthalate) particles with an average size of 62 nm were analyzed by using two different sizes of gold nanospheres as SERS substrates. Through this technique, plastic particles with a concentration as low as 10 μg·mL$^{-1}$ can be detected, a recovery rate of 81%, and analytical enhancement factors of up to 446 were achieved.

In real lake water samples, Hu et al. [102] provided a SERS method for quantitative analysis of nanoplastics (shown in Fig. 5a). They added KI to Ag nanoparticles as a coagulant to make silver nanoparticles aggregate to form SERS hotspots, and as a cleaning agent to remove impurities on the surface of silver nanoparticles. According to their research, PS nanoplastics in varying concentrations can be found by analyzing particles with different sizes (50, 100, 200, and 500 nm). As shown in Fig. 5b and c, the recovery rate is 87.0–110%, which indicates a strong linear correlation between concentration and the SERS intensity at 1002 cm$^{-1}$.

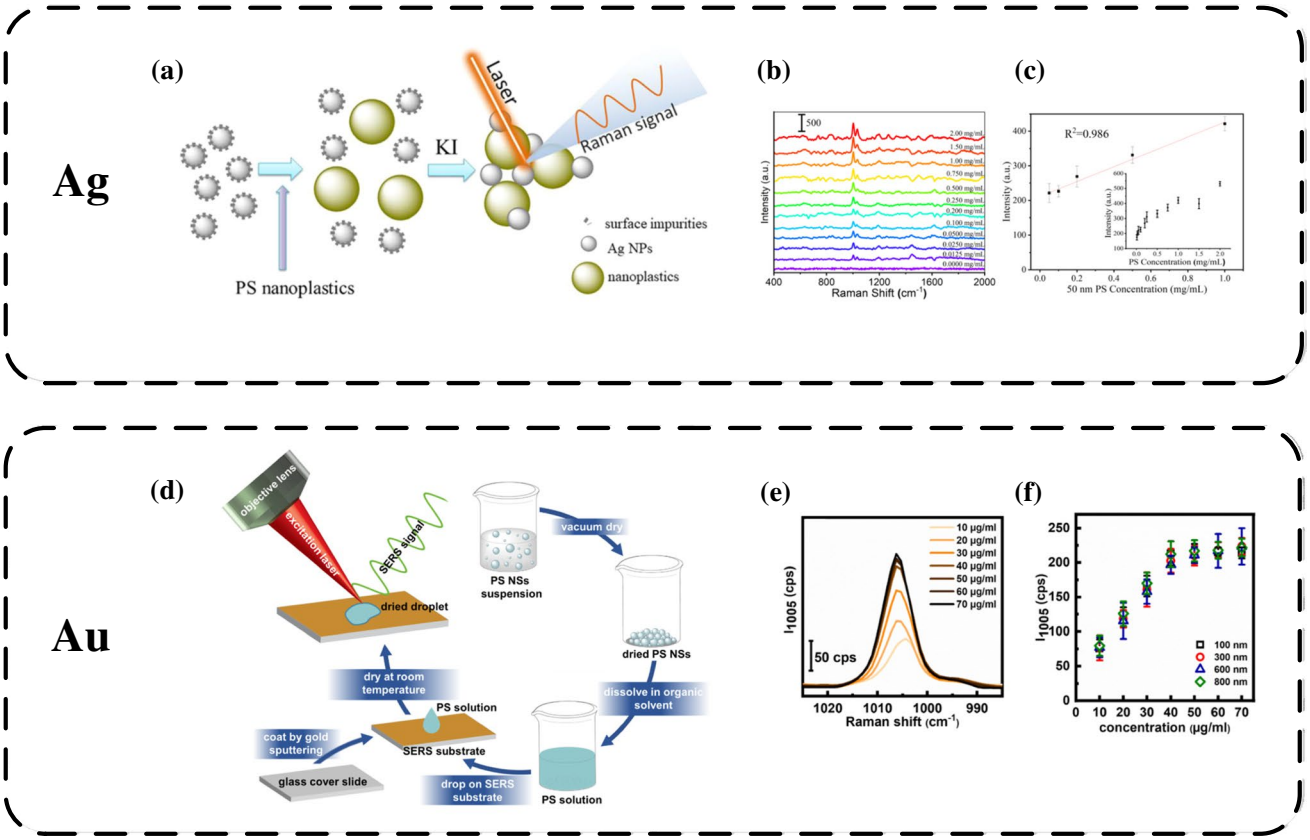


**Fig. 5** Examples of microplastics quantitative detection techniques based on SERS**: a** The schematic diagram of nanoplastic analysis by proposed SERS method. [102] Copyright (2022) Elsevier. **b** Raman spectra of PS nanoplastics at different concentrations, taking 100 nm as an example. [102] Copyright (2022) Elsevier. **c** The relationship between concentration and the SERS intensity at 1002 cm$^{-1}$ of PS nanoplastics, taking 100 nm as an example. [102] Copyright (2022) Elsevier. **d** The procedure for quantitative analysis of NPs in water media using SERS. [95] Copyright (2022) Elsevier. **e** SERS spectra of PS in the region of 990-1020 cm$^{-1}$ at different PS concentrations. [95] Copyright (2022) Elsevier. **f** Standard calibration curves of PS with particle sizes of 100, 300, 600, and 800 nm. [ 95 ] Copyright (2022) Elsevier

In the study by Chaisrikhwun et al. [95], combined with the "coffee ring effect," gold nanoparticles were used as SERS substrates for the quantitative detection of PS particles. The minimum detection PS particle size was 100 nm. The LOD for PS particles was 0.10 $\mu g \cdot mL^{-1}$, whereas the quantitative range of analysis falls between 10 and 40 $\mu g \cdot mL^{-1}$. At the same time, this study also indicated that in the presence of interference, SERS measurements could still perform well, including NaCl, KCl, $MgCl_2$, $CaCl_2$, sucrose, glucose, fructose, arabinose, galactose, SDS, and albumin. Therefore, this developed SERS technique is well suited for detecting and quantifying MPs that are present in the environment. The test procedure is shown in Fig. 5d. The SERS spectra of PS at various PS concentrations and the standard calibration curves of PS with various particle sizes are shown in Fig. 5e and f.

SERS technology can be utilized not only for direct detection of microplastics but also for indirect detection. The concept of indirect detection based on SERS involves establishing a correlation between the spectral changes of metabolites, reaction products, or reporting molecules (RM) and the concentration of target analytes [103]. This method can detect analytes with low or zero levels of Raman vibration signals, and can also realize multi-component detection [104].

The most commonly used method in indirect detection is the utilization of reporting molecules (RM). This involves functionalizing the substrate with one or more molecules, leading to single-wave or multiple-wave detection. This method is effective in detecting changes in the Raman cross-section caused by the interaction with the target analyte. Reporting molecules are typically small in size and possess a high Raman cross-section. Furthermore, they have features such as a narrow Raman spectrum, minimal overlap with bands in the substrates or analyte spectrum, and photochemical stability [105].

In the detection of microplastics using Raman spectroscopy, relying solely on this technique may not provide accurate and precise results, especially in complex environmental samples. Therefore, multiple detection technologies have become increasingly important, which can identify and quantify microplastics and yield more accurate and reliable results [106].

First, various microscopic techniques like scanning electron microscopy (SEM) and transmission electron microscopy (TEM) can be applied to visualize and identify microplastics. These techniques allow for studying the morphology and size of microplastics [37]. In addition, pyrolysis Gas Chromatography Mass Spectrometry (Py-GC-MS) is a technology that can reveal the chemical composition of microplastics. The microplastic sample is heated to a high temperature under anaerobic conditions to decompose the plastic into its constituent chemicals, which are then be analyzed by gas chromatography and mass spectrometry to identify the type of plastic that has been found [38]. Moreover, fluorescence staining provides a fast, convenient, and inexpensive way to quantify microplastics [107]. For example, Nile Red [108] and Rhodamine B [109] are used to stain the tested samples by emitting specific fluorescence after they are adsorbed on the surface of microplastics, microplastics can be identified and roughly counted by image processing and analysis.

Furthermore, SERS can also be combined with various detection technologies. For example, Xia et al. [110] studied the method of using microfluidic in SERS analysis through four combination approaches. First, microfluidic synthetic techniques provide a unified MP fabrication manufacturing method. Second, the integration of microchips and SERS substrate offers advanced equipment for sensitive and efficient detection. Third, sample preparations using microfluidic can rapidly separate and preconcentrate analytes before SERS detection. Finally, highly integrated microfluidic devices can be used in multistep SERS analysis, namely material fabrication, sample preparation, and detection processes.

Chen's research [111] highlights the microfluidic-SERS detection system as a compact, advanced analysis platform that enables high sensitivity, rapid, and high throughput analysis requiring no samples. The effective detection of microfluidic SERS platforms can be grouped into the following categories. The first category is the mixing efficiency between the SERS active matrix and analyte in the microchannel, as optimal mixing contributes to an increase in SERS hot spots [112]. Additionally, the synthesis or modification of the SERS active substrate in microfluidic devices can improve the reproducibility and convenience of SERS detection. Moreover, the capture of target analytes is performed in the microfluidic channel to enhance SERS-specific detection.

The microfluidic-SERS detection system complies with the modern analysis technology development trend and satisfies the current detection demands [113]. Future research should continue to focus on improving detection selectivity and reproducibility, chip versatility, and integration.

In general, the application of multivariate detection technologies to detect microplastics can improve the accuracy and reliability of microplastic detection, leading to a deeper understanding of the prevalence and distribution of microplastics in the environment.

## Application of artificial intelligence algorithm in Raman spectrum detection

Traditional spectral data analysis techniques have several limitations due to the growth of Raman spectroscopy and the expansion of application areas [114]. In practical

applications of microplastics detection, it is not always feasible to optimize all available parameters due to high sample volumes and limited analysis time for each sample [52, 115]. The poor spectral quality requires post-processing steps to increase the confidence of spectral evaluation. As a result, different phenomena can be resolved through various methods. For instance, baseline artifacts can be eliminated using several baseline reduction methods, such as fitting a polynomial or asymmetric least squares smoothing [116]. Nevertheless, customizing the optimal post-processing settings for every single spectrum becomes unfeasible when dealing with a large number of spectra that need to be processed rapidly.

Artificial intelligence (AI) can accelerate experimental analysis and computation considerably by learning efficiently from extensive pre-labeled data and making precise predictions for new data sets. Detection combined with AI algorithm technology can more effectively extract valuable information from spectral data [117].

Figure 6 presents the different applications of AI algorithms in Raman spectroscopy.

In order to implement an AI algorithm for the detection of MP using Raman spectroscopy, the initial step involves collecting Raman spectroscopy data for the microplastics. Then, the collected data should be processed, including denoising and debaselining. In addition, the Raman spectral data can be feature extracted by AI algorithm [122]. Finally, through reasonable algorithm selection, model training, and model evaluation, microplastic detection can be facilitated by integrating AI algorithms.

For example, combining artificial intelligence algorithms allows for the automated completion of the detection analysis during the measurement or processing [123]. According to Brandt et al. [74], a self-coding neural network [124] can be built to effectively remove noise and unwanted artifacts from the vibration spectrum for batch processing of large amounts of spectral data. First, the input data is encoded into a compressed form using the "encoding" phase of the algorithm. The encoded data is then reconstructed into its

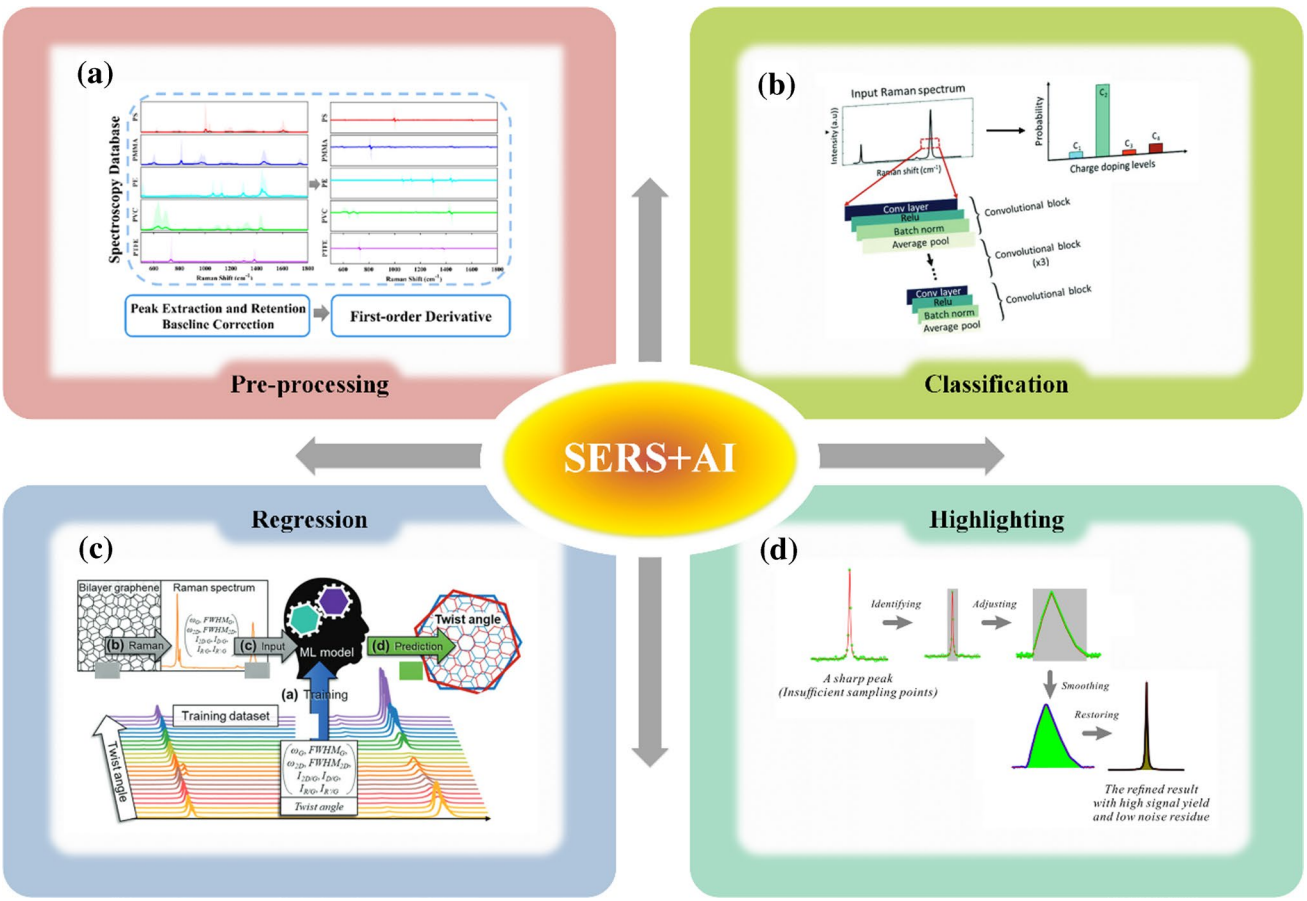


**Fig. 6** Applications of artificial intelligence algorithm to Raman spectroscopy. **a** Schematic of the autoencoder network for spectral reconstruction. [118] Copyright (2023) American Chemical Society. **b** Schematic diagram of an example classification task. [119] Copyright (2022) Royal Society of Chemistry. **c** Schematic diagram of an example regression task. [120] Copyright (2022) American Chemical Society. **d** Schematic diagram of an example highlighting task. [121] Copyright (2021) American Chemical Society

original format using a decoder of the network. In this way, the dimension of the input data is first reduced to capture only the basic information, which is then used to construct the raw data. This method is suitable for a variety of different spectral types.

In traditional deep neural networks, as the depth of the network increases, the gradient gradually becomes smaller and eventually disappears, causing parameter updates to become slow or even stagnant, making the network difficult to train. In 2015, He et al. [125] used residual learning for image recognition, and this problem was solved by introducing ResNet which adds residual connections [126]. These connections allow skipping one or more layers and adding the original input to the output, which enables the network to learn the residual function rather than the original function, alleviating the problem of disappearing gradients [127]. Essentially, there is a shortcut between the input x and the desired output H(x). This network design enables a skip connection, allowing gradient information to pass through the layers and can prevent the vanishing gradient problem. Additionally, an intriguing deep learning architecture known as the generative adversarial network (GAN) was introduced, which consisted of a generator and a discriminator [126].

In terms of noise reduction, Zhao et al. [128] developed a low signal-to-noise ratio Raman signal denoising method based on feature extraction. Raman spectroscopy is a non-destructive technique, with the disadvantages of weak Raman signals, even weaker than noise, due to their short exposure time and low power of the excitation laser. The method's principle is based on the Hilbert Vibration Decomposition (HVD) method, which decomposes the Raman spectrum into two components. The peak is in the first component and compensated by the peak value in the second component. Then, their full width at half peak (FWHM) is calculated based on the position and height of the peaks. Finally, Gaussian signals are used to reconstruct Raman peaks from strong noise and baseline based on the position, height, and FWHM of the peaks.

Artificial intelligence algorithms are being used in the detection of microplastics based on SERS. For example, Henrique et al. [129] applied machine learning classification algorithms to the detection of microplastics by comparing several successful classification models. Four preprocessing methods, namely Baseline correction, Standard Scaler, PCA, and Oversampling, were analyzed and compared. The goal was to evaluate their effectiveness in enhancing the detection process. In addition, a novel technique was proposed to define microplastics using a machine-learning approach. Unlike other methods, this technique measured the multi-class likelihood of a model using logarithmic loss. Ultimately, linear SVC was chosen as the final classifier. The findings revealed that Baseline correction and Oversampling were the most effective methods for the selected models. Additionally, linear SVC is highly effective for scalability. Once trained, the classifier can be directly deployed and used to classify unknown spectra. However, a major drawback of this approach is that the training model cannot predict samples from classes it is not familiar with. This limitation could hinder the overall deployment of the program, especially considering the wide range of polymer types that may exist. Henrique et al. recommend assigning polymer class tags to each spectrum as a solution to this issue, which will reduce unnecessary work and expedite the collective learning process.

For example, Pittroff et al. [44] used Raman spectroscopy to extract the characteristic peak information of MPs with fingerprint features. Combined with machine learning technology, the rapid identification and classification of MP (PET, PVC, PP, PS, PC, PE) particles with six concentrations of 10 ppm, 1 ppm, and 0.1 ppm in five water environments (pure water, rainwater, lake water, tap water, and seawater) were achieved.

In general, the use of artificial intelligence algorithms in Raman spectral detection is effective and can aid researchers in detecting microplastics more quickly and effectively.

## Prospects and conclusions

### The development trends of Raman spectroscopy for microplastic detection

As a powerful materials analysis technique, Raman spectroscopy has attracted attention for its potential in detecting microplastics. The sensitivity and resolution of Raman spectrometers have improved considerably. By using better equipment and materials, researchers can improve better optical performance and achieve more sensitive detection, in detecting smaller plastic particles.

The most important and challenging aspects of using Raman spectroscopy and SERS for microplastic detection are sample extraction, spectral data acquisition, and low-concentration detection.

Firstly, when it comes to extracting samples and detecting low concentrations, it is useful to consider the two aspects together. Raman spectroscopy typically requires a high concentration of sample at the time of detection. Therefore, researchers have developed new sampling methods to collect and concentrate microplastics, such as filtration, centrifugation, and density separation. Developing more sensitive detection equipment and methods to detect low levels of microplastics is the future research direction.

In addition, addressing the issue of spectral data acquisition can be achieved through substrate research and the use of portable Raman instruments. The main limitations of SERS include the requirement of substrate-analyte close

contact, substrate degradation, selectivity issues, and problems with re-usability and homogeneity of substrate.

One particular challenge arises from the uneven distribution of nanoplastics on a silicon wafer, leading to variations in hot-spot densities and resulting in non-uniform enhancement effects for the nanoplastic sample. As a consequence, reproducibility is compromised. To overcome this, further efforts should be dedicated to creating more uniform surfaces by increasing the number of hot spots and ensuring a more homogeneous distribution of nanoplastics. To summarize, while SERS offers great potential for spectral data acquisition, it is crucial to address the limitations associated with substrate-analyte contact, substrate degradation, selectivity issues, re-usability, and substrate homogeneity.

The development of portable Raman spectroscopy equipment has enabled its application in the detection of microplastics in the field. This equipment offers convenience and allows for quick and easy analysis of microplastics on-site. Raman spectroscopy has proven effective in identifying the sources of microplastics in various environments, such as seawater, sediments, soil, and biological tissues. By comparing the Raman spectra of microplastics from different sources, researchers can determine the source and distribution of microplastics in different environments.

In terms of future development, SERS technology is expected to have the following characteristics: it will widely promote the adoption of local environmental standards, harness the minimizing effects of micro/nano plastic aggregation and natural organic matter, exploit the unique hot spot engineering of micro/nano plastic, and continuously optimize SERS parameters like laser power, wavelength, and duration.

## Conclusion

As a result of their versatile applications across different fields, the global production and implementation of plastic materials have experienced a consistent upsurge in recent years. The increasing use of plastic products has resulted in a buildup of microplastic (MP) waste in natural environments, which has become a widespread problem. In this study, our objective is to provide an update on the recent developments in plasmonic nanomaterials-assisted Surface Enhanced Raman Scattering (SERS) platforms, which detect the trace of MP particles in the natural environment.

Therefore, the authors introduce a wide range of novel SERS substrates that have been created from either pure precious metal or unique hybrid nanomaterials. The findings revealed that significant progress can be made in developing new and efficient technologies for the identification of MP particles. These technologies not only offer high selectivity but also enable ultra-sensitive monitoring of MP particles with a low detection limit. Furthermore, to protect human health and the environment from the harmful impact of MP particle pollution, the combination of plasmonic nanomaterials-assisted SERS substrates and Raman spectroscopy holds great promise and attracts researchers interested in portable and rapid analysis of MP particles in environmental nature. According to the previously validated studies, this concise topical review provided an updated overview of recent developments and trends in MP detection using enhanced Raman scattering. Various unique materials, as well as 3D SERS substrates, nanopipettes, and microfluidic chips, are discussed. Additionally, a novel materials-assisted spectral Raman technique and its practical application were introduced for the selective monitoring of trace detection of MPs in both indoor and outdoor environments.

This article introduces the detection principle of Raman spectroscopy and SERS, as well as their application in the detection of microplastics. Additionally, it briefly discusses the use of Raman spectroscopy and SERS in combination with other technologies for efficient and time-saving detection applications. Drawing from validated research, this review provides concise insights into the prospects and future development trends in this field.

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## Declarations

**Conflict of interest** The authors declare no competing interests.

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