Supporting Information

Klarite as Label-Free SERS-Based Assay: A Promising Approach for

Atmospheric Bioaerosol Detection

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Experimental

Reagents and Materials.

All the chemicals were purchased either from Sigma Aldrich or mentioned otherwise. *Escherichia coli* (DH5 alpha) was purchased from Tiangen Biotech co. Ltd, Beijing China. HyClone phosphate buffered saline (PBS) solution was purchased from GE Healthcare Life Sciences. Klarite[™] was purchased from Reinshaw Diagnostics Ltd, Glasgow, U.K. All the solutions were prepared with Milli-Q water.

Microorganisms Preparation.

Shock-frozen *E.coli* (DH5 alpha) cells were separately cultivated in sterile LB medium at 200 rpm and 37 °C for 14 h. 40 mL of each of the bacterial solutions were harvested and then centrifuged at 3750 rpm for 10 min at 4 °C (Beckman Coulter Allegra X-12, Beckman Coulter, Inc., American). Afterwards, the supernatants were discarded, and pellets were washed twice with 20 mL PBS by centrifugation at 3750 rpm for 10 min at 4 °C. Finally, the resulting pellets were resuspended in PBS after vigorous shaking.

Characterization and Instruments.

SEM images were taken with an accelerating voltage of 3000 V with TM-1000 (Hitachi). SERS measurements were performed with XploRA PLUS confocal spectrometer (HORIBA Scientific, France).



Fig. S1. SERS spectra of *E.coli* on (a) Klarite, (b) Cu/Ag film-glass surface, (c) Au nanoparticlessilicon wafer, (d) Au film-glass surface and (e) Au film-Cu foil. The concentration of *E.coli* used was 1.8×10^9 cells mL⁻¹.



Fig. S2. Different growth rate was observed when *E.coli* grown to OD_{600} 0.4, 1.5 and 2.5 and then harvested.



Fig. S3. SERS mapping of ambient bioaerosol samples. SERS mapping of (a) positive sample collected from real environment and (b) negative sample collected from air flow chamber exposed under UV light. SERS mapping performed at 735 cm⁻¹. 50x objective lens was used.



Fig. S4. SERS spectra of different concentrations of *E.coli* from 1.8×10^9 to 1.8×10^1 cells mL⁻¹.

| Raman Shift (cm ⁻¹) | Assignment |
|--------------------------------------------------------------|-------------------------------------------|
| 625 | phe skeletal |
| 652 | δ(COO-) |
| 735 | adenine, glycosidic ring mode |
| 805 | υ(CN) |
| 828 | ring breathing Tyr |
| 852 | ring breathing Tyr |
| 955 | υ(CN) |
| 1002 | phenylalanine |
| 1030 | phenylalanine C-H in plane |
| 1128 | amide III, adenine, polyadenine and DNA |
| 1158 | proteins |
| 1210-1310 | amide III |
| 1227 | amide III, polyadenine and DNA |
| 1330 | $v(NH_2)$ adenine, polyadenine, DNA |
| 1320-1340 | DNA/RNA, Proteins |
| 1360-1440 | υ(COO-) symmetric |
| 1376 | v _s (COO-) |
| 1440-1460 | $\delta(CH_2)$ saturated lipids, proteins |
| δ: Bend υ: Stretch Tyr: Tyrosine Phe: Phenylalanine | |

Table S1. Peaks assignment of the SERS spectra of the *E.coli*.¹⁻⁷

Phe: Phenylalanine v_s : Symmetric Stretch

References

- 1. A. Sengupta, N. Brar and E. J. Davis, Bioaerosol detection and characterization by surfaceenhanced Raman spectroscopy, *J. Colloid Interf. Sci.*, 2007, **309**, 36-43.
- 2. R. M. Jarvis, N. Law, I. T. Shadi, P. O'Brien, J. R. Lloyd and R. Goodacre, Surfaceenhanced Raman scattering from intracellular and extracellular bacterial locations, *Anal. Chem.*, 2008, **80**, 6741-6746.
- 3. Y. Liu, Y.-R. Chen, X. Nou and K. Chao, Potential of surface-enhanced Raman spectroscopy for the rapid identification of Escherichia coli and Listeria monocytogenes cultures on silver colloidal nanoparticles, *Appl. Spectrosc.*, 2007, **61**, 824-831.
- 4. K. Maquelin, C. Kirschner, L.-P. Choo-Smith, N. van den Braak, H. P. Endtz, D. Naumann and G. Puppels, Identification of medically relevant microorganisms by vibrational spectroscopy, *J. Microbiol. Methods*, 2002, **51**, 255-271.
- 5. H. Zhou, D. Yang, N. P. Ivleva, N. E. Mircescu, S. r. Schubert, R. Niessner, A. Wieser and C. Haisch, Label-free in situ discrimination of live and dead bacteria by surface-enhanced Raman scattering, *Anal. Chem.*, 2015, **87**, 6553-6561.
- 6. M. Knauer, N. P. Ivleva, X. Liu, R. Niessner and C. Haisch, Surface-enhanced Raman scattering-based label-free microarray readout for the detection of microorganisms, *Anal. Chem.*, 2010, **82**, 2766-2772.
- 7. O. Samek, H. Telle, L. Harris, M. Bloomfield and D. Mack, Raman spectroscopy for rapid discrimination of Staphylococcus epidermidis clones related to medical device-associated infections, *Laser Phys. Lett.*, 2008, **5**, 465-470.