

Interactive comment on “Electrospray-Surface Enhanced Raman Spectroscopy (ES-SERS) for probing surface chemical compositions of atmospherically relevant particles” by Masao Gen and Chak K. Chan

Anonymous Referee #2

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Authors present ES-SERS approach to characterize atmospheric aerosol particles. This is not a new approach, but new in terms of application as this is not been applied to atmospheric particles in the past. It seems such approach can be applied to atmospheric aerosols, and results are encouraging. Such approach was also recently published (El-Khoury, Johnson et al. 2015), therefore this is not a new approach and manuscript should be modified to reflect this. I have few major comments, and after these are addressed, I recommend the manuscript for publication.

Comments:

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To simplify the reading, I suggest authors revise the particle definition terminology. It is confusing as aerosols are typically defined in the sense that these are the particles that are present in the atmosphere and affect Earth's radiative properties (optical and cloud). In this paper aerosol is defined as silver nanoparticles, and conventionally defined aerosol as 'analyte particles.' Please revise such that these analyte particles are now called aerosols and Ag nanoparticles as Ag nanoparticle only. Defining Ag nanoparticles as aerosol is confusing.

It is not clear why sucrose compound was used? Does this represent atmospheric organic aerosol composition? Please provide some references. Please clarify the necessary to coat the silicon wafer with gold. Are these wafers home grown or commercially available (part number? and vendor?).

In section 2.2 it is mentioned that SEM images were obtained. Can they be inserted into the main paper? Why UV-vis was used? Describe typical CPC concentration values. What was the size of Ag nanoparticles (53 nm?) that was size-selected.

How spraying time of 1 hour was determined.

Model name and software version of the Renishaw Raman microscope should be mentioned. Please specify wavenumbers corresponding to compounds specified on line 198 (page 8).

Section 2: Please mention here how submicron ambient aerosol were collected in this section. It is already mentioned in section 3.5. How particles between 0.05 and 0.15 μm were collected. Using impactor? What stage and what was the 50% cut size. How samples were dried? And why it was necessary to dry the particles. Can drying process could alter the surface composition? Do you have Raman maps for ambient particles?

Section 3.2: It is bit confusing to understand the comparison between Ag-coated SERS substrate and current SERS method. Why this comparison is carried out? In both

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cases the test aerosol are similar, so I expect the spectra from both methods should be similar. In terms of intensity, it can be just coincidence. It also raises important question, if both methods yield similar results, why one should use ES-SERS technique. One can use commercially available SERS substrates, which can save a lot of time in regards to sample preparation. Any thoughts? This result also contradicts the advantages of ES-SERS described in Introduction section.

Section 3 in general. One has to go back and forth between the main paper and supplementary information to understand the results. I suggest if some figures can be inserted into the main paper to improve the readability.

Section 3.3: Line 314-316. Is this observed in this study? and how such distance is determined? Are there any SEM images that shows such information.

Line 332: To improve readability I suggest to add the values of wavelength and NA.
Line 334: ..'gravity had affected..' is not clear. How gravity affects the shape. Shape (morphology) can be also altered during impaction. Line 347-348: Please mention what components were observed in minor and major category. Line 427:429: It is bit confusing. Does this imply that ES-SERS can only give surface composition but not bulk. Line 428: 'comma' after word composition is missingcompositions, respectively, . . .

Section 4: Please revise this section. Remove any new material that is not discussed in the main text. Remove any speculations or theories. Discuss them in the main text. Some editorial work is needed to understand the main message of the paper. Main conclusions are not clear after reading this section. Please see below some comments to improve this section further.

Line 433: This is not a new technique. Rephrase the sentence. Line 436-439: Sentence is not clear. Cannot understand the phrase, please rewrite. Line 447-449: To improve readability, I would move this sentence to the main text. These past studies are not discussed either. Please discuss these. DMA is already defined for differential

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mobility analyzer (line 159). Line 465-466: This is new and speculative. Please move to the main text and discuss. Do you have any maps that support this premise? Check (Baustian, Cziczo et al. 2012) for further information regarding maps and morphology. Line 467-472: Paragraph text is confusing. How this is relevant to the present study. I suggest delete or modify and move to the main text.

Figure 8: In caption mention the wavenumber corresponding to sulfate and organics. How Organics peaks were identified?

References:

Baustian, K. J., D. J. Cziczo, M. E. Wise, K. A. Pratt, G. Kulkarni, A. G. Hallar and M. A. Tolbert (2012). "Importance of aerosol composition, mixing state, and morphology for heterogeneous ice nucleation: A combined field and laboratory approach." *Journal of Geophysical Research-Atmospheres* 117.

El-Khoury, P. Z., G. E. Johnson, I. V. Novikova, Y. Gong, A. G. Joly, J. E. Evans, M. Zamkov, J. Laskin and W. P. Hess (2015). "Enhanced Raman scattering from aromatic dithiols electrospayed into plasmonic nanojunctions." *Faraday Discussions* 184(0): 339-357.

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