

## **Response to Reviewer 1:**

*General comments:*

*Liang and colleagues have written an excellent primer on Raman spectroscopy and its application to the study of atmospherically relevant single-particles. The work is comprehensive and very well written and is an excellent contribution to the literature. My comments below are all quite minor and can be readily corrected during a through readthrough of the manuscript.*

**Author's Response:** Thank you for your comments. The manuscript has been revised accordingly.

*Line 13. Replace 'state' with 'states'.*

**Author's Response:** The word has been corrected.

*Lines 49-51. References should be provided for the different approaches mentioned within this sentence and/or in the paragraph that follows.*

**Author's Response:** The relevant references have been added.

Physical and chemical processes of atmospheric particles have been examined by numerous experimental approaches: a flowing stream of aerosol particles in an open system (flow tube) (Li et al., 2015; Duporté et al., 2017; Heine et al., 2018; Liu and Abbatt, 2020; Liu et al., 2020a; Peng and Jimenez, 2020), a chamber of suspending particles (smog chamber) (Akimoto et al., 1979; Behnke et al., 1988; Liu et al., 2016; Ye et al., 2018; Charan et al., 2019; Liu and Abbatt, 2021; Qin et al., 2021), a collection of deposited particles (Bao et al., 2018; Zhang et al., 2020b), and single particles (Lee and Chan, 2007b; Krieger et al., 2012; Davies and Wilson, 2016; Gen et al., 2019b; Gong et al., 2019; Sullivan et al., 2020).

*Line 60. Is the 'smog chamber' mentioned here the same thing as the 'chamber of suspending particles' mentioned on line 50? The terminology could be tightened.*

**Author's Response:** We do mean smog chambers. The sentences are revised to:

Line 50: Physical and chemical processes of atmospheric particles have been examined by numerous experimental approaches: a flowing stream of aerosol particles in an open system (flow tube), a chamber of suspending particles (smog chamber), a collection of deposited particles, and single particles.

Line 60: Smog chambers enable simulations of various environmental conditions, including gas

concentrations, for studying the formation and chemical transformation of particles.

*Line 84. Replace 'process' with 'processing'.*

**Author's Response:** The word has been corrected.

*Line 101. Define the acronym HULIS*

**Author's Response:** The definition of HULIS has been added.

Light-absorbing chemicals such as [humic-like substances](#) (HULIS) or some brown carbon species may generate fluorescence, interfering with the peak identification in the spectra ([Ivleva et al., 2007a](#)).

*Line 114. It is unclear why the authors mention pharmaceuticals here. There are numerous non-atmospheric applications of Raman that could be mentioned. I'd suggest eliminating the parenthetical expression.*

**Author's Response:**

Line 112-114 is replaced by:

We focus on applications of atmospheric relevance and refer readers to other literature on the principles of Raman spectroscopy and related techniques (e.g., stimulated Raman or coherent Anti-stokes Raman spectroscopy) and applications of less direct atmospheric relevance.

*Line 143. Replaced 'coupling' with 'coupled'*

**Author's Response:** The word has been corrected.

*Line 164. The term 'Bessel beam' has not previously been defined.*

**Author's Response:** The sentence is revised to:

[By configuring a Bessel laser beam, instead of the typical Gaussian beam, in either vertically or horizontally trapping arrangements, optical levitation and tweezer can be applied for trapping submicron particles \(Meresman et al., 2009; Cotterell et al., 2014; Lu et al., 2014\)](#)

*Line 203. References should be included for MDRs and WGMs.*

**Author's Response:** The references related to MDRs and WGMs have been added.

*Lines 194-205. The reference formatting in this section is not consistent with the other parts of the manuscript. Reference numbers are given instead of Author, Date format.*

**Author's Response:** The reference formatting has been corrected.

*Line 280. Replace 'other' with 'differential' Raman characteristics.*

**Author's Response:** The word "other" has been replaced by "different."