

We thank the reviewers for the input and resulting improvements to the manuscript.

**Response to Reviewer 2:**

1) Page 11, line 226: “Calibration of the impactor shows a transition from rebound to adhesion between 102 to 1 Pa s in viscosity for sucrose particles (Bateman et al., 2015).” To relate particle bounce to viscosity values, more than one substance should be used in calibrations. The particle bounce properties are not affected only by viscosity, but also other material characteristics which may vary between different substances.

We agree with the reviewer that a more robust understanding of material physical properties as related to their rebound behavior would be beneficial for a more complex analysis of ambient particle rebound behavior. The transition from semisolid to liquid for a variety of inorganic compounds with respect to rebound behavior in this same apparatus has recently been studied (Li et al., 2016). In the absence of a complete study on a variety of material characteristics and their rebound response, the present study assumes that the uptake of water by semi-solid sucrose particles to liquid is a valid model system for the transition of SOM from semi-solid to liquid.

The text is expanded as follows: *“As a point of reference, the transition from rebound to adhesion occurs across a viscosity transition of  $10^2$  to 1 Pa s for sucrose particles for the operating conditions of the impactor (Bateman et al., 2015). The viscosity range corresponding to the rebound transition can depend on particle composition, but this aspect is not investigated herein.”*

2) Page 11-12, lines 245-246: It seems that authors interpret the bounce curves in a way that if the rebound fraction is 5%, approximately 5% of the particles are solid. Even the bounce curves for single components, such as sucrose, are s-shaped curves and rebound value varies between 1 and 0. In the case of sucrose particles all the particles have absorbed same amount of water at certain RH, hence they all have the same physical phase state. The reason why for example 20% of sucrose particles are bouncing off from the impactor is not because 20% of the particles are solid and 80% liquid. 20% of sucrose particles are bouncing off at certain RH and 80% are sticking on the impactor substrate because the velocity of particles in impactor jet depends on their radial distance from the center of the impactor jet. Hence in this example, 20% of sucrose particles have kinetic energy higher than the dissipation and surface adhesion energies. So it is obvious that increasing rebound fraction doesn't necessarily imply external mixture and increasing fraction of solid particles. It can also imply changes in material characteristics of all particles (more solid particles). This should be taken into account in data interpretation and also the text should be changed accordingly throughout the manuscript.

Thank you for this thoughtful comment. It is true that there is variation in the impact velocity and thus rebound fraction as a function of radial distance from the center of the impactor jet. We have modeled the flow-dynamics of our impactor and for the nozzles employed in this study (cf. Figure S3 and S4 of Bateman et al., 2014) and found the kinetic energy of the impacting particles at large diameters should be well above the adhesion energy even at high RH (cf. Figure 7 of the Bateman et al., 2014). This is the main reason for using particle diameters of 190 nm. Under the controlled conditions and thorough understanding of the particle flow dynamics of our impactor we can rule out any change in rebound fraction attributed to kinetic energy and surface adhesion.

We also agree that the reviewer is correct that we assume the particles are a homogeneous mixture during most of the analysis. To this point the caveat is included: “...that increase viscosity when internally mixed with background PM and increased concentrations of non-liquid anthropogenic particles in external mixtures of anthropogenic and biogenic PM.” Overall, to the reviewer’s point, the width of the rebound curve exceeds the width of calibration particles, indicating that there are different particle types undergoing nonliquid/liquid transitions at different RH values.

3) According to main conclusions of the paper the changes in measured rebound correlated with decreasing kappa and decreasing O:C. Still no data on O:C is shown in the whole manuscript. Authors should show, for example, how O:C varies between pollution, biomass burning and background cases and also during day and night (Figure 3). Also O:C panel should be added to figure 4.

The intention was to use PMF-A and PMF-B as the main supporting data for the changes in the measured rebound fractions. To avoid the confusion highlighted by the reviewer’s comment, we removed the statement “...decreasing O:C elemental ratios,” from the conclusions in the manuscript.

We addressed the reviewer’s concern that there is not enough data included for O:C ratios by adding Table S3 to the supporting information. This table includes the average O:C ratios for the various air mass classification categories and day/night time periods used in the manuscript. Text is added as follows: “The average O:C ratios for the various air-mass classifications can be found in Table S3 of the Supplement.” The O:C panel is added to Figure 4.

4) Also the rebound data corresponding different cases (polluted-biomass burning background) should be shown.

These data sets are shown in Figure S1 as whisker-box plots. Text is added as follows, “Figure S1 of the Supplement presents an additional level of detail.”