**Response to referee #1**

We are grateful for referee #1's comments. Those comments are all valuable and helpful for improving our paper. We answered the comments carefully and have made corrections in the submitted manuscript. The corrections and the responses are as following:

In the revised manuscript, the red color was marked as the revised places.

General comments: The authors' replies did not answer my comments. They showed only relevant sentences in the revised manuscript. For example, I had three questions in my major comment #2, but none were answered. The same miscommunication can be applied to many of the specific comments. The most important comment, major comment #1, is misinterpreted.

**Reply: We seriously considered your comments and did more experiments to answer your question. Indeed, the revised manuscript was improved a lot.**

Major comment #1. The major comment #1 in the previous report was about three-dimensional shape (configuration) in the atmosphere. The authors' reply was about the representativeness of 3D morphology measured by TEM and did not answer my comment. My concern is that 3D shapes can be changed on the substrate from that in the atmosphere no matter how they are analyzed (e.g., AFM and TEM). Thus, a careful discussion will be necessary to interpret the 3D shape of aerosol particles in the atmosphere. For example, consider the particle configuration of the upper right image in Fig 9 (the particle with three soot particles with sulfate core and organic coating). When rotating the particle 90 degrees, one of the soot particles should be middle of the sulfate core in the 2D image. That means the particle shapes in Fig 6 c-d could be different from those in the atmosphere, but they had changed their shape on the substrate. If they had been the same organic coatings, the organic should have coated the entire surface, including over and behind the particles. Although I do not say that the particle shapes on the substrate are not useful, the shapes also have some information, including those when they were in the atmosphere. However, it cannot be directly compared to the particle shape on the substrate and those in the atmosphere.

So careful discussion should be provided here.

**Reply:** We appreciate the reviewer’s comments. We undertake a carefully examination of this issue through a comprehensive and in-depth discussion.

Firstly, large amounts of studies have proved that microscopy imaging of the samples on substrate can be used to investigate the liquid-liquid phase separation (LLPS) process of the atmospheric particles. You et al. (2012) presented images of real-world samples collected on quartz fiber filters to reveal that atmospheric particles can undergo LLPS. O'Brien et al. (2015) investigated the LLPS in individual aerosol particles by collecting the samples on grid-supported carbon-filmed grids which is similar to our study. Another experiment shows that coated soot particles on the flat grid experienced minimal structural changes during the sampling and storage (Chen et al., 2017). In addition, the aerosol optical tweezer measurement provides the evidence that the core-shell particles are formed after LLPS with the variation of RH (Gorkowski et al., 2020). Therefore, the LLPS indeed occurred in aerosol particles although the aerosol particles become flat on the substrate as the below Figure.

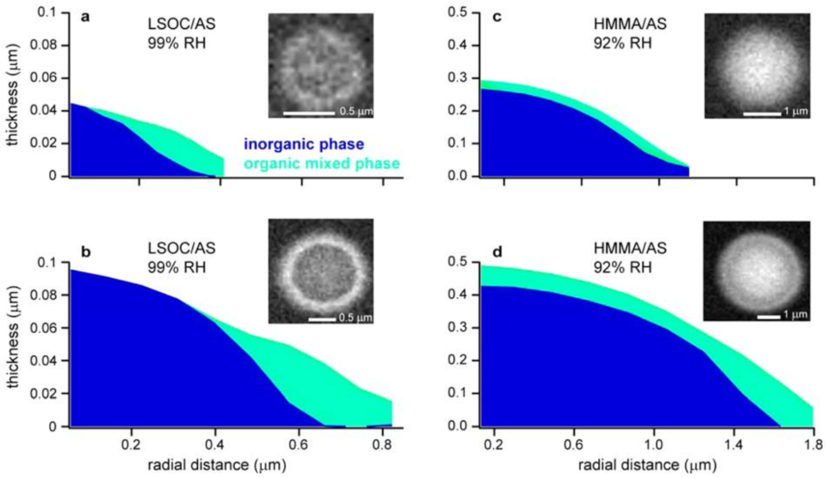


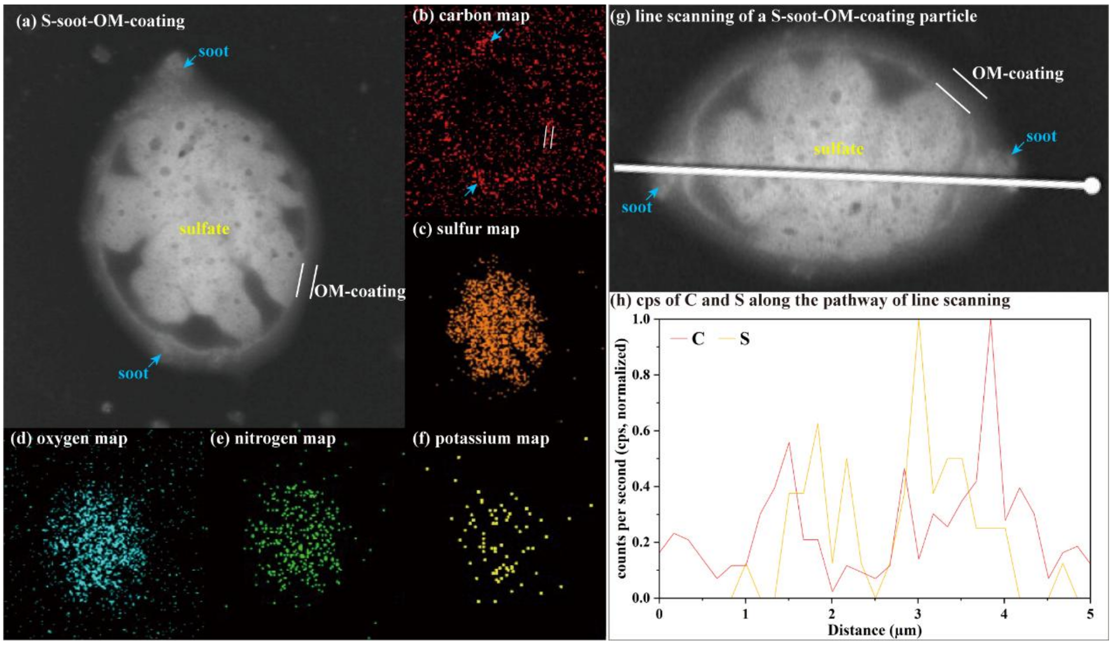
**Figure 3.** Thickness maps for a mixed organic/inorganic phase (cyan) and a pure AS inorganic phase (blue) from radial scans for two different sized LSOC/AS 8:1 OIR particles at 99% RH (a, b) and two different sized HMMA/AS 1:1 OIR particles at 92% RH (c, d). The organic mixed phase is overlaid on top of the inorganic phase. Insets show STXM images for each particle taken at 288.5 eV.

Thickness maps for a mixed organic/inorganic phase (cyan) and a pure AS inorganic phase (blue) from radial scans for two different sized LSOC/AS 8:1 OIR particles at 99% RH (a, b) and two different sized HMMA/AS 1:1 OIR particles at 92% RH (c, d). This figure is cited from O'Brien et al. (2015).

Secondly, as your comments, we did additional measurements by using the same sample. Elemental maps and line scanning of C, S, O, N, and K of the typical S-soot-OM-coating particles were conducted from the TEM-EDS. The results show the clear soot cores distributing in the OM-coating. We added the analysis method and results in the revised manuscript as follows:

In context, line 118-119: “To analyse elemental distribution in individual aerosol particles, EDS mapping and line scanning experiments were conducted using scanning TEM (STEM) mode in the JEM-2100F TEM.”

Line 167-169: “Elemental mapping and line scanning analysis of the core-sell particles all reveal that the coating predominantly contains abundant C, while the inorganic core contains abundant S, N, O, and minor K (Fig. 3).”



**“Figure 3. Elemental distribution in individual S-soot-OM-coating particles. (a) A dark-field TEM image of a S-soot-OM-coating particle. (b-f) TEM-EDS elemental maps of C, S, O, N, and K of the S-soot-OM-coating particle. (g) A TEM image of a S-soot-OM-coating particle for line scanning detection. (h) Counts per second (cps) of C and S along the pathway of line scanning over the S-soot-OM-coating particle.”**

Major comment #2: I am not convinced by the idea that "liquid-liquid phase separation" and "soot redistribution" processes caused the mixing states. I think that the mixing states can also be explained by coagulation between soot and sulfate (Fig. 9 left two processes) and then condensation of organic matter without LLPS and soot redistribution. I am not convinced by the process between the second left and the third left particle images (LLPS) in Fig 9. Why is the soot embedded within the sulfate core instead of simply attaching to the surface? The processes that the authors claimed may be possible, but evidence and discussion are not sufficient. In the specific comments #2, #3, #4, and #7 in my previous referee report, the authors added some more references and showed previous studies but did not discuss the results obtained in this study, including line 164-182. Although previous studies have shown LLPS processes, they do not prove that the results in the current study are the same processes. The authors did not discuss other possibilities. It is acceptable to discuss any possibility, but this manuscript has too strong a conclusion for a possibility. E.g., line 24-27 "One-third of soot-containing particles showed the LLPS phenomenon between organic matter and inorganic aerosols in individual particles, which further induced soot redistribution. The results show that a larger LLPS particle size and a higher ratio of organic coating thickness to soot size tended to drag soot from the sulfate core into the organic coating" and line 232-234 "The TEM images clearly demonstrate the transferred position of soot from the inner sulfate core to the outer organic coatings following the increasing OM/soot (Fig. 6c-e)."

**Reply:** We appreciate the reviewer’s comments.

1. The atmospheric processes like condensation and coagulation of soot with secondary aerosols can indeed affect the morphologies of the internally mixed particles (Corbin et al., 2023). However, the coagulation process between soot and secondary particles is generally negligible except very near the source (Sedlacek et al., 2022) and is unlikely to cause the compaction of soot (Corbin et al., 2023). Condensation and other processes may participate in the aging process during long-range transport. However, here we do not concern the detail about the mixing process of soot and secondary aerosols during the long-range transport because the mixing process is very complicated and it is not the focus of this study. We focused on the structural changes of soot particles after long-range transport and influenced by

the liquid-liquid phase separation. In order to weaken the description of the related conclusions, we revised the contents as follows:

In context, line 24-27: “One-third of soot-containing particles showed a core-shell structure that probably formed the LLPS phenomenon after long-range transport. Particle size and ratio of organic coating thickness to soot size are two of the major possible factors that likely induce soot redistribution between organic matter and inorganic aerosols in individual particles.”

Line 178-179: “Therefore, S-soot-OM-coating particles as shown in Figure 2 were likely considered as soot particles mixed with the LLPS particles after long-range transport.”

Line 196-197: “The results indicate that a substantial amount of soot-containing particles may undergo the LLPS process during the sampling period.”

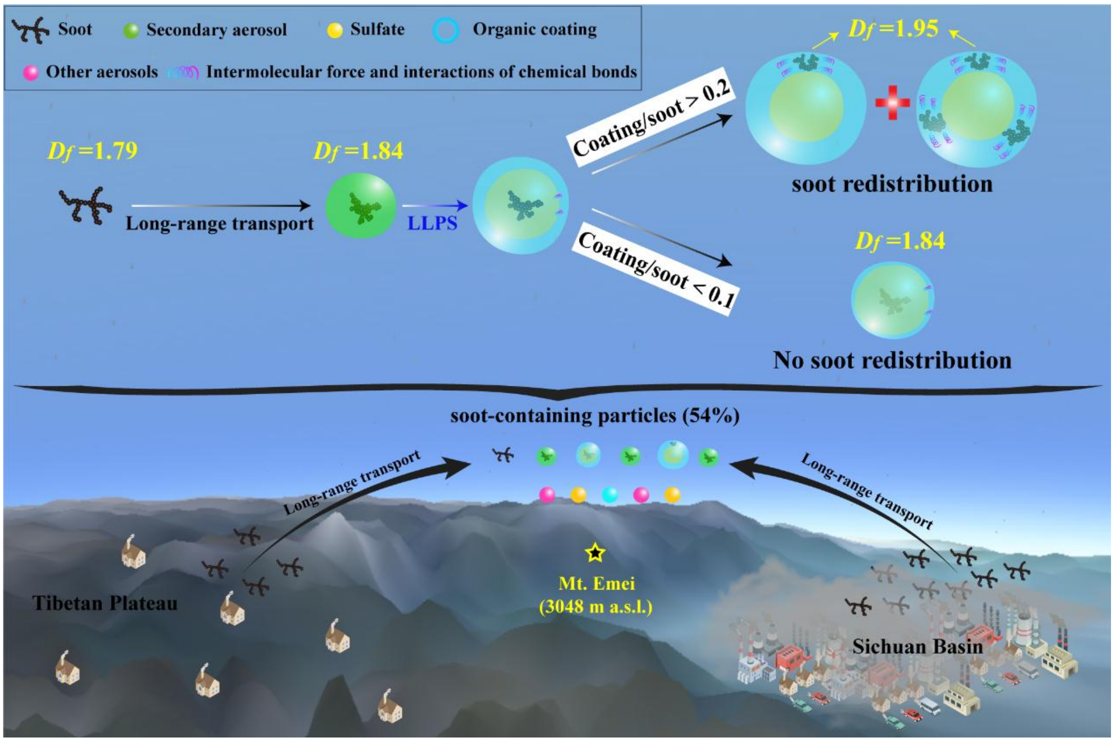
Line 229-230: “These results suggest that soot exhibits a high likelihood of distributing into the organic coating instead of the inorganic core following an increasing ratio of OM/soot (Fig. 8b).”

Line 248-249: “All of these observations provided evidence supporting the potential phenomenon of soot redistribution within LLPS particles in the atmosphere over the eastern TP.”

Line 277-279: “Our morphological analysis suggests that the entire particle size and ratio of organic coating thickness divided by the size of soot (OM/soot) are two of the major possible factors affecting the redistribution of soot.”

Line 281-282: “Consequently, larger particles with thicker organic coatings are more inclined to drive and capture more smaller soot particles from the inorganic phase to the organic phase.”

2. To avoid misleading in Fig.11. We have revised it to facilitate a better understanding of the content of this paper.



"**Figure 11.** A conceptual model illustrating the atmospheric processes of soot on the eastern rim of the Tibetan Plateau."

Major Comment #3: (Response to Comment #2 in the previous report): If soot particles were not included in the plot, how were the soot EVDs obtained? For example, soot EVD values are shown in Figure 4. Due to the fractal shape of soot, if soot EVD values were obtained from the plot without measuring soot particles, the soot EVD values would be inappropriate.

**Reply:** We appreciate the reviewer's comments. We did not clarify this issue in previous version. Although secondary particles and soot particles both appear to be two-dimensional in transmission electron microscopy observations (as shown in Fig. 7a, 7b), they exhibit different three-dimensional features when analyzed from other perspectives. To make it clearly, we did the surface morphology measurement by using scanning electron microscope (SEM). Sulfate and organics mostly showed a thin and flat morphology while soot was more three-dimensional (Fig. 7c, 7d). AFM analysis further show that soot exhibits a higher height than that of secondary particles (Fig. 7e, 7f). Therefore, we used ECD of soot and EVD of secondary particles to represent the individual particle size. We used the difference between the EVD of the entire core-sell particle and the EVD of inorganic core to calculate the thickness of organic coating.

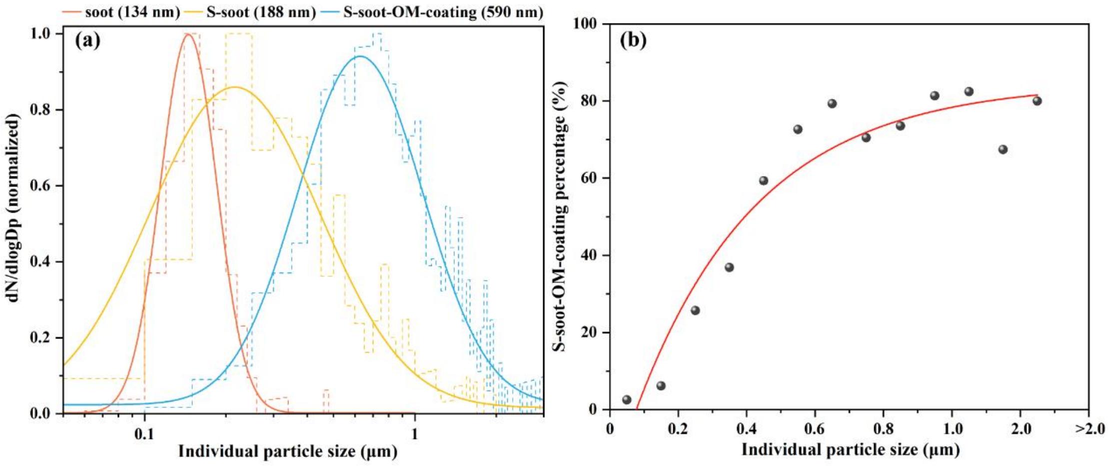
In general, we modified a figure (Fig. 5), added a new figure (Fig. 7), and revised the content as follow:

In context, line 75-77: “In this study, individual particle collection, transmission electron microscopy (TEM), scanning electron microscope (SEM), and atomic force microscopy (AFM) were comprehensively employed to investigate the mixing structures of soot particles at a mountain site on the eastern fringe of the TP.”

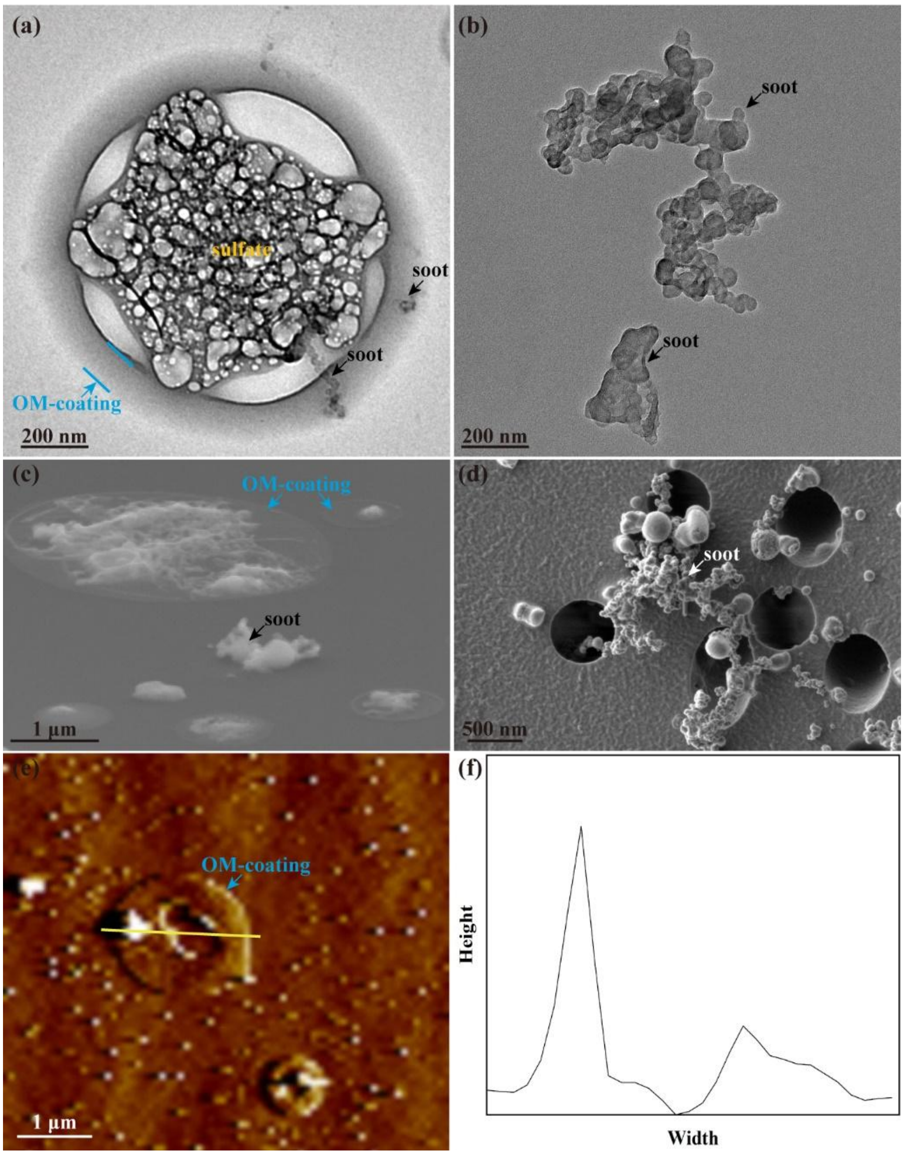
Line 121-124: “The SEM (Zeiss Ultra 55, Germany) was used to obtain detailed information on the surfaces of individual aerosol particles. To obtain three-dimensional morphological information of individual particles on the substrate, we tilted the sample stage to 75° with accelerating voltage of 10 kV and work distance of 6.6 mm, and then captured the particle images at a magnification of 7000×.”

Line 130-131: “The sizes and coating thicknesses of secondary particles are calculated based on the EVD and the sizes of soot particles are calculated based on the ECD. The detailed calculation method can be found in supplement.”

Line 213-214: “Similar to the method employed by Zhang et al. (2022), we calculated the OM-coating thicknesses and the entire particle sizes based on TEM and AFM (Figs. 7, S2, S3).”



**“Figure 5. (a) Size distributions of soot, S-soot and S-soot-OM-coating particles. The numbers in parentheses represent the log-normal peaks. (b) Variations in the percentage of S-soot-OM-coating particles in all soot-containing particles with sizes.”**

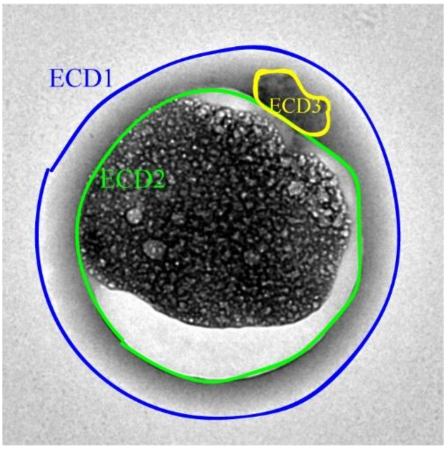


"Figure 7. Different views from the different microscopic techniques. (a) A TEM image of S-soot-OM-coating particle, (b) A TEM image of soot particles, (c) A SEM image at 75° tilt angle of individual particles, (d) A SEM image of soot particles, (e) An AFM image of S-soot-OM-coating particle, (f) The cross-sectional analysis from the left AFM image."

Specific Comment #1: OM/Soot

Figure 6. How were the OM/soot values obtained for each soot particle within particles in Fig. 6 c-e?

**Reply:** We appreciate the reviewer's comments. Firstly, we obtained the ECD of the entire S-soot-OM-coating particle (ECD1, blue line, Fig. S3) and ECD of the inorganic core (ECD2, green line, Fig. S3) through the Radius software. Secondly, we transform the ECD1 and ECD2 to EVD1 and EVD2 according to the AFM analysis, respectively. Thirdly, we calculate the OM-coating thickness (OM for short, (EVD1-EVD2)/2). Finally, we calculate the OM/soot ratio (OM/ECD3).



“**Figure S3**. The calculation diagram of the ratio of OM coating thickness to soot size (OM/soot) of a typical S-soot-OM-coating particle. OM/soot = [0.4144(ECD1-ECD2)/2]/ECD3”

Lines 268-270: "Once the OM/soot ratio exceeded 0.2, more than 80% of the soot tended to be distributed in the organic coating due to the possible intermolecular forces and interactions with increasing coating thickness (Figs. 6b, 9)". The OM/soot ratio >0.2 means that there is more OM compared to the soot, indicating that the soot tends to distribute in the organic coating regardless of "the possible intermolecular forces and interactions with increasing coating thickness".

**Reply:** We appreciate the reviewer’s comments. We revised this sentence as follow:

In context, line 279-280: “Once the OM/soot ratio exceeded 0.2, more than 80% of the soot tended to distribute in the organic coating (Figs. 8b, 11).”

## References

Chen, C., Enekwizu, O. Y., Ma, Y., Zakharov, D., and Khalizov, A. F.: The Impact of Sampling Medium and Environment on Particle Morphology, Atmosphere, 8, 162, 2017.

Corbin, J. C., Modini, R. L., and Gysel-Beer, M.: Mechanisms of soot-aggregate restructuring and compaction, Aerosol Science and Technology, 57, 89-111, 2023.

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You, Y., Renbaum-Wolff, L., Carreras-Sospedra, M., Hanna, S. J., Hiranuma, N., Kamal, S., Smith, M. L., Zhang, X., Weber, R. J., Shilling, J. E., Dabdub, D., Martin, S. T., and Bertram, A. K.: Images reveal that atmospheric particles can undergo liquid&#x2013;liquid phase separations, Proceedings of the National Academy of Sciences, 109, 13188-13193, 2012.