

Interactive comment on “Morphology, Chemical Composition and Mixing State of Atmospheric Aerosols from Two Contrasting Environments in Southern India” by C. R. Hariram et al.

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We thank the reviewer for the comments and overall summary evaluation.

General comment:

1. Firstly, the authors used quartz filters to collect aerosol particles and study individual particles. The information could not describe the right single particles in the air. The quartz filter looks like fibrous. Many fine particles are hiding in the filter and hence the SEM could not see all the particles in the filter. I would like to say that you need to use the flat substrate such as TEM grids, silicon substrate, and polycarbonate filter. I think

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that the authors should look at these references cited in the paper.

- This is an important point, and we agree with the reviewer that many fine particles could hide in the filter (in depths) and thus one may not see all the particles. The reviewer suggests for the use of a filter substrate with smooth surface for optimal analysis of individual particles. While we agree with this suggestion, we feel that these arguments are correct in case of a general aerosol-focussed study especially concerned with fine mode particles. We, on the other hand, are interested to study the morphology and state of mixing of dust-BC composite aerosol system (dust particles with BC depositions), which mainly falls under coarse mode aerosols. A recent study (Beatriz et al. 2016) has reported that quartz fiber filters efficiently collect particles of different size ranges in the top layer ranging from diameter of $0.199 \mu\text{m}$ to $10.272 \mu\text{m}$. It is also reported that the largest volume fraction (34.216 %) of particles captured is in the outer layer. Moreover, it is also estimated that about 65 % of the volume fraction of particles is found in the top two layers of the filter (Beatriz et al. 2016). Additionally, it is reported that, the inner layers are found to trap only the fine particles of mean particle diameter $0.533 \mu\text{m}$. In this study, since our interest was centered around the coarse mode aerosols, we could easily identify them from the top layer of the quartz filter. Moreover, use of aerosol loaded quartz filter for automated SEM/EDX studies, has also been supported in the literature (e.g., Willis et al., 2003) Our study was completed with a semi-automated SEM/EDX system and a careful post processing with the help of an image processing software which solved the tribulations of the noisy fibrous background. The software helped in enhancing the image and cropping the independent particles easily from background. Thus, with the help of sophisticated instrumentation and post-processing, we examine the state of mixing of dust-BC aerosol system over two stations with contrasting environments in southern India.

2. Secondly, the author totally made a mistake about the core-shell structure shown in Figure 2. Core should be in center of particles and shell look like coating on the shell. In the Figure 2, the red part only overlapped on other part. I am pretty sure that the

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particle is not core-shell structure. Based on these two points, the rest figures could not be right data analysis. Even in Figure 4, I could not find any core-shell structure.

- We agree with the reviewer that core should be in the center of the particle and the shell should form a covering on it, in a totally core-shell structure. However, in this study we find particles to be only partially coated and some regions of the core part of the particles appear to be exposed without any shell cover on it and this is one of the important finding. Such partially coated particles when viewed from top, look similar to the ones shown in figure 2. Thus, on the account of the projection of the image, the partially coated particles reported in this study, do not look strictly similar to the theoretical shell-core structured particles. Some of the particles reported in figure 4 also depict such partially coated shell-core structure. Small particles that seem to adhere on the coarse particle like BC chain found on a portion of the surface of a bigger particle as in figure-8 is also considered as coating.

3. Thirdly, because the authors selected one wrong sample filter, they could not get any good images to secondary particles, soot (BC), metal, and other particles. EDX did show quit high Si from quartz filter. The EDS data could not be quantitatively analyzed to show C, O, and Si.

- We would like to undeniably state that all the chemistry data reported in the paper is qualitative. With the present-day available techniques, it is impossible to probe into the individual particle level chemistry on a quantitative basis. As such, we have used the energy dispersive spectroscopy (EDX) which is a widely used qualitative approach to examine the elemental composition of atmospheric aerosols. In this study, we have performed multiple EDX analysis on different portions of the same particle to arrive at the average elemental composition to minimize the error. Also, the concern of background artefact was studied by performing an experiment comparing with the aerosol samples collected on a carbon tape. We computed the background error of 2.05 ± 0.8 % together contributed by Si & O₂. About 50 independent coarse mode particles were used for this study. Furthermore, the EDX was performed carefully on clearly exposed

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surface of the particles with minimum possibility of background interference. This increases our confidence in the reported results. These are included in the revised ms.

Other comments:

1) P5Line 4-5 how do you know the BC?

- We have used an Aethalometer (model AE42; Magee Scientific, USA) for simultaneous measurements of ambient black carbon (BC) mass concentration. Those measurements have been utilised in this study to examine their association with carbon coating on dust particles. Details are provided in the revised ms

2) P5Line 33 why are Si and O dominant in the particles? You might analyse coarse particles because the fine particles penetrate in the filter. Or the quartz filter influence the EDS.

- As already mentioned in reply to 1st general comment from the reviewer, the focus of our work is coarse mode aerosol particles. These particles could be easily identified with sufficiently high magnification (50,000-1,00,000) under SEM as they mostly get trapped in the outer most layer of the quartz filter, unlike the fine mode particles which penetrate the filter. Since the coarse mode particles are mainly composed of Si and O (silica i.e. dust origin), we get dominance of these elements in EDS. The concern of background artefact (quartz filter) in EDS results has been analysed by performing an experiment comparing with the aerosol samples collected on a carbon tape. We have computed the background error of 2.05 ± 0.8 % together contributed by Si & O. About 50 independent coarse mode particles were used for this study. Also EDX was performed carefully on clearly exposed surface of the particles with minimum possibility of background interference. This increases our confidence in the reported results.

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