

### Answer to RC3

This manuscript describes the STRASS equipment, an aerosol sampler for the collection of atmospheric particles. Sampling is performed in short periods of one hour, over a polycarbonate filter surface; that permits the collection of 168 samples, sequentially and automatically, during one week. The exposed filter is sequentially transported to the laboratory, where the direct measurement of elementary aerosol composition by PIXE is performed, without further treatment. This sampling/PIXE analysis methodology permits the evaluation of trace aerosol composition with hourly discrimination at acceptable costs and is therefore potentially interesting for source apportionment purposes.

The STRASS sampler was built to replace and improve the previous STREAKER sampler used for the same objectives, which is not commercially available, anymore.

In the paper the characteristics and performance of the STRASS sampler are evaluated with various experiments to validate the sampler and polycarbonate 0.8  $\mu\text{m}$  pore membrane capability to collect correctly PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> aerosols.

The evaluation of 0.8  $\mu\text{m}$  pore polycarbonate filter was done by parallel sampling with a Teflon membrane filter recognized as having high filter efficiency for submicrometric particles. The results, exposed in Figure 3, show very similar concentration values. However, the presented results are for PM<sub>10</sub> and elements usually associated with coarse particles (with possible exception of S). Therefore, from the experiment, it is not clearly demonstrated the capability of the 0.8  $\mu\text{m}$  pore membrane to collect efficiently fine particles. Results for elements principally associated with fine particles and for PM<sub>1</sub> or PM<sub>2.5</sub> fractions would be more relevant to the evaluation of filter collection efficiency.

For nuclepore with 0.8 micron pores, minimum in collection efficiency is expected below 100 nm (Hinds, 1999). Thus, even sampling fine particles, only elements with a size distribution strongly skewed towards the Aitken or condensation modes can be affected by significant losses. Focusing on the elements detected with PIXE ( $Z > 10$ ), significant contribution to the Aitken and accumulation mode (at least in urban areas) is expected for S and K only, as S is mainly present as secondary aerosol and K may be produced by biomass burning (see e.g. Bernardoni et al., 2017). In the measurements presented in this work and from previous measurements, a fair reproducibility is observed: the loss is always between 15 and 30% for S and between 20 and 30% for K. Conversely, the effect is expected and proved to be minimal for crustal and marine elements, as a result of the measurements we made to evaluate these effects. We did not report PM<sub>2.5</sub> and PM<sub>1</sub> scatterplots as we obtained similar results with PM<sub>10</sub>, however we agree that this information may be of interest and we added it in the paper (a slight effect can be observed for S). We also want to point out that this is a membrane type issue. STRAS can also work with the 0.4 micron pore size polycarbonate membranes, and with these the collection efficiency is close to 100%.

The reliability and accuracy of the STRASS sampler was done in a field validation, by parallel sampling in the external environment with the STRASS and a GEMINI sampler, using both the same polycarbonate 0.8  $\mu\text{m}$  pore membranes. The results show remarkable similarities for both samplers, principally for PM<sub>10</sub>, a clear indication of the reliability of the STRASS sampler which seems not to suffer from flow control or leak problems.

The collection capability of 0.8  $\mu\text{m}$  pore membrane filters is further evaluated in field conditions by using parallel sampling with the GEMINI sampler provided with Teflon membrane filters. The results presented in Figure 7 for PM<sub>1</sub> particles show evidences of inefficient polycarbonate membrane filtration for elements concentrated in submicrometric size ranges, such as S and K.

The results of this campaign confirm the results obtained in the courtyard tests: a loss for S and K, no loss for the other elements. The losses for S and K are slightly different from those found in the tests (about 30% for both elements versus 15% for S and 20% for K). Nevertheless, it should be noted that these data were obtained in a real field campaign, where the sampling difference is not only the type of membrane: in particular, in this case, STRAS was used to sample on an hourly basis while GEMINI

was conventionally used on a daily basis. Further, the campaign was performed in a different sampling site, Bologna, where the S and K size distributions may be different from Milan.

We have added in the conclusions of the text the following recommendations on how to account for these effects:

“Possible PM losses due to a reduced collection efficiency of these membranes were investigated by simultaneous sampling on ring-supported thin Teflon filters: significant effects (15-30% underestimation) were observed only for S and K, which are elements typically related to smaller particles originated by secondary aerosol processes and biomass burning, respectively (and thus with size distributions which extend down to below 100 nm, where the polycarbonate 0.8 membranes have a minimum in collection efficiency). As a consequence, it is important to be aware that S and K concentrations can be underestimated when using these membranes. In our tests these underestimations were found to be quite stable; however, some parallel sampling, even on a daily basis, may be helpful in determining correction coefficients for other sampling sites. It should be noted that the values found in this work can be considered maximum limits of underestimation since the tests were carried on in places characterized by a strong prevalence of ultrafine aerosols, such as secondary and combustion ones.”

Contrary to previous figures, figure 7 shows scatterplots with X and Y in logarithmic scales. From the figure it seems that presented linear regressions and correlation coefficients are also for the logarithm of concentrations. This presentation permits the evidencing of lower concentration values, that, anyway, are already strongly influenced by PIXE detection limits and filter blanks variability. However, visualization of differences at higher concentration ranges are reduced. A parallel figure using linear scales would be useful for a more clear evaluation of polycarbonate filters performance.

Scatterplots of figure 7 are presented in log scales but the linear regression parameters and correlation coefficients are calculated on the original concentrations (not log). We chose log plots due to the larger range of variability of these data; nevertheless, we agree with the referee about the limitations of this visualization. In order to uniform the paper plots, we have decided to change figure 7 to linear plots, as the ones reported in the following:

