

## Answer to RC2

General comments:

This paper introduces a new aerosol sampler for observing particulate matter composition capable of taking many samples with high temporal resolution, offering the potential for more efficient atmospheric monitoring—an increasingly critical need. However, the manuscript lacks several key elements, including a quantitative evaluation and thorough discussion of the sampling method, as well as a comparison of its advantages and disadvantages relative to conventional methods. These points require further elaboration.

We thank the referee for raising these issues, discussed in more detail in the following. We have accordingly changed the paper.

A major concern is the observed loss of fine particles for certain elements compared to Teflon filters. This raises questions about the suitability of the method for fine particle sampling ( $PM_{2.5}$ ,  $PM_{1}$ ). If the sampler is intended for fine particle applications, the authors should clearly discuss its validity and propose solutions to address these issues.

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. This allows a reasonable evaluation of these effects due to the collection efficiency and the correction of the results. 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%.

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.”

Additionally, it was difficult to understand the superiority of STRAS over the conventional streaker sampler and the Gemini sampler used for this comparison. I expect that STRAS allows the collection of a larger number of samples and a larger amounts of particles in one spot, but there is insufficient data regarding the Gemini sampler's capacity, making it difficult for readers to draw meaningful comparisons. I would recommend to make a table to explain merits and limitations of each sampling method together with other methods mentioned in the introduction.

We have explained more clearly in the paper the differences and advantages of STRAS with respect to the STREAKER and the conventional sequential samplers like Gemini (lines 97-102).

#### Advantages of STRAS against STREAKER:

- higher areal load, and so higher sensitivity for PIXE
- flexibility in time-resolution
- possibility to work directly with commercial US-EPA inlets
- STREAKER sampler is not produced anymore

-data logging for sampling parameters (air flow, pressure drop, elapsed time etc..) and for sample position (the position of every single spot is registered and can be reproduced also in case of black-outs or stepper motor malfunctioning). STREAKER registered the sampling parameters (in a far less user-friendly mode with respect to STRAS) but not the sample positioning data. [This information was missing: we added it at lines 165-167 in the sampler description].

- better separation of the sample deposits (i.e. sampling slots) avoiding the moving average due to the continuous frame rotation present in the STREAKER sampler

#### Advantages of STRAS against the GEMINI:

- higher areal load, and so higher sensitivity for PIXE
- flexibility in time-resolution (down to 1h)
- the number of samples that can be collected with no need of filter change or operator intervention: GEMINI can perform up to 21 samples without any operator intervention (less than a day on hourly basis); STRAS can collect up to 168 samples without any need of changing filters, allowing 1 full week of samplings on a hourly basis with the instrument unattended. While unattended, both GEMINI and STRAS can be controlled remotely.

#### Specific comments

##### 1. Introduction:

· As noted in the general comments, please consider adding a table to summarize the merits and limitations of different sampling and analytical methods. In addition, include additional details, such as the differences in detection limits, applicable elements, costs, etc., between EDXRF and PIXE, and the advantages of STRAS sampling system over traditional streaker samplers. This would enhance the clarity of the introduction.

We agree with the referee that a direct comparison of the pros and cons of the different sampling techniques may help and guide the reader. Therefore, we added more information in the paper, at the end of the introduction section, in a discursive way, which we felt was clearer than a table (lines 97-102). The paper focuses on a new sampling device, therefore a thorough discussion of the analytical techniques for the determination of aerosol composition is beyond the scope of this work. Nevertheless, we added an explanation for having designed a sampler “dedicated” to PIXE analysis and added some references for the comparison of the different analytical techniques (lines 103-109).

##### 1. Sampler design:

· Please include information about the size and weight of STRAS sampler, since you mentioned that the sampler was designed to be “compact and easily transportable.”

We have added this information to the paper, lines 171-172.

· Please provide details on how particle size resolution is achieved. There was no information on size separation. In addition, it was not clear for me whether the sampler can collect different size fractions at a same time or not.

The STRAS version described in this paper is a single stage sampler. It collects PM by filtration on polycarbonate membranes. It can be equipped with commercial single cut-off inlets designed to select

PM10, PM2.5 or PM1 at 1 m<sup>3</sup>/h. So, depending on the mounted inlet it can collect one of these fractions (one at a time).  
An implementation of STRAS is currently in progress to simultaneously collect both the fine and coarse PM fractions as previously done by the streaker samplers, but it is not described in this paper.

#### 1. PIXE analysis of STRAS samples

• P6. L174: How did you determine the errors (especially for deposit area)? If they were based on analytical data or references, include this information.

We added the following information (lines 201-203):

Measurement quality assurance checks were routinely performed (by analysing the NIST SRM 2783). Measurements and uncertainty estimates were performed according to “IBA measurements guidelines” (<https://www.actris-ecac.eu/pmc-elements.html>).

As for the deposit area, its size is determined by the dimensions of the sucking orifice, which is obtained by 3D print. The used printer has an accuracy of less than 1 tenth of millimetre. A slightly more conservative estimate of the uncertainty was taken into account due to a possible broadening of the deposit on the sides (not evidenced by the careful measurements performed on the deposit spots).

<https://www.actris-ecac.eu/pmc-elements.html>

• P6. L177: If filters were washed with acid, would this reduce the blank values? Trace metal concentrations can be as low as several ng/m<sup>3</sup> in remote areas like marine atmosphere.

For PIXE analysis, with clean filters as Polycarbonate, or Kapton/Teflon/Polypropylene..., the main limitation to detection limits (DLs) comes from the thickness of the filters as this increases the continuum background in the spectra due to secondary bremsstrahlung and Compton scattering. For this reason, in the past, we have deeply characterized the DLs with the different collecting substrata, as reported in Calzolari et al., 2015.

Polycarbonate membranes are very clean: rarely we have observed small contaminations of Si and Br in Polycarbonate filters, but these batches have been discarded.

#### 1. STRAS validation

• P7. L184: Please specify the sampling position (longitude, latitude) and height.

The sampling campaign was performed inside the Dept. of Physics courtyard, an urban background site located in the eastern side of Milan (45,47665 9,23082; 122 m a.s.l). We have added this information to the paper.

• 4.1: You mentioned potential underestimation of S and K due to their enrichment in fine particles. However, you don't address any solutions or recommendations for it. In addition, I don't understand why other elements have good agreement even in PM<sub>1</sub> (Fig. 7) despite that they are all fine particles.

As aforementioned, for nuclepore with 0.8µm pores, minimum in collection efficiency is expected below 100 nm. 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. Therefore, most of the elements have good agreement whatever the sampled PM fraction (PM10 or PM2.5 or PM1). We added some recommendations in the paper on how to handle this effect.

- 4.2: Please explain why Gemini data has larger errors (for example, Figure 5, 6). Is this due to the smaller sample quantities on the filter spots?

Yes, it is due to the lower areal densities on Gemini samples. We added this information in the paper (lines 263-265).

- Figure 7: Why Ca concentrations at low levels are higher in STRAS sampler compared to the Gemini sampler?

It may be due to an unperfect spectra fitting at low concentrations, due to the subtraction of the continuum background due to Bremsstrahlung in the spectra. Nevertheless, the effect is small (please note the very high uncertainties and the y axis range)

#### 1. Conclusion

- P12 L284: This should be mentioned in section 4. Also please refer some papers here.

Yes, we added this information in section 4 (with references).

#### Technical comments

- P2. L46: optimal solution  $\Rightarrow$  optimal analytical solution

Yes, we did it.

- P3. L93-94: I don't think the sentence "Furthermore...samplers." is necessary in the introduction, this should be moved to conclusion or discussion.

Yes, we moved it in the conclusion section.

- P8. L224: How about adding "(e.g. S and K)" at the end of the sentence?

Yes, we did it.

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