

## ***Interactive comment on “An automatic collector to monitor insoluble atmospheric deposition: an application for mineral dust deposition” by B. Laurent et al.***

**B. Laurent et al.**

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Response by B. Laurent on Interactive comment on “An automatic collector to monitor insoluble atmospheric deposition: an application for mineral dust deposition”.

Anonymous Referee #1

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=> We would like to thank the three reviewers for their insightful and helpful comments on the manuscript. These comments have helped us to improve the manuscript. Please find hereafter Reviewer's comments and our responses.

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Peer review, and interactive public discussion Does the paper address relevant scientific questions within the scope of AMT? Yes 1. Does the paper present novel concepts, ideas, tools, or data? Yes, the paper presents a practical design and tool for sampling insoluble deposition. 2. Are substantial conclusions reached? Yes. 3. Are the scientific methods and assumptions valid and clearly outlined? Yes, the methods are clearly outlined. Some clarifications on the filter efficiency need to be clarified. 4. Are the results sufficient to support the interpretations and conclusions? Yes 5. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? Yes, as far as I can see. 6. Do the authors give proper credit to related work and clearly indicate their own new/original contribution. Yes. 7. Does the title clearly reflect the contents of the paper? Yes 8. Does the abstract provide a concise and complete summary? The last sentence of abstract is descriptive of what has been done but showed no results. It would good to describe the results of the field work.

=> Results are now briefly presented in the summary: “Insoluble mineral deposition measured on Frioul Island is 2.45 g m<sup>-2</sup> for February to December 2011 and 3.16 g m<sup>-2</sup> for January to October 2012. Nine main mineral deposition events, associated with air masses coming from the southern Mediterranean Basin and North Africa, are measured in periods during which significant MODIS aerosol optical depths are observed.”

9. Is the overall presentation well structured and clear? Yes 10. Is the language fluent and precise? Mostly. It would be good if a native English speaker could have a look before submission.

=> An English colleague read and corrected the paper. We carefully went through the paper.

11. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used. Yes. 12. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? The authors should consider moving

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some of the figures to supplementary materials. For example, Figure 2 and 3 do not really contribute to the science case and only provide supporting evidence and therefore they are not absolutely necessary in the main text.

=> Following the reviewer advice, Figures 2 and 3 are now presented in the supplementary materials (Figures S1 and S2, respectively).

13. Are the number and quality of references appropriate? Yes

Detailed comments

This paper reports an automatic sampler for collecting insoluble deposition. It will prove useful to quantify the aerosol deposition fluxes at various sites, which will be of great value to constrain global aerosol models. The sampler will provide new and exciting opportunities for the aerosol community. The paper is of value to scientific community. My recommendation is to accept the paper after a full consideration of the comments below and those by the other reviewer.

The chosen pore size is 0.8  $\mu\text{m}$ . This is very large. I am sure there are smaller particles passing the filters. It is recommended that a test is carried out to see how much  $<0.8\mu\text{m}$  particles will go through the filter. This can be done by a successive filtering: 0.8  $\mu\text{m}$  pore size filtering of original sample followed by 0.02 $\mu\text{m}$  pore size or 0.1  $\mu\text{m}$  pore size filtering of the filtrates. By comparing the mass of the particles on the first and second filter, it should be possible to estimate the uncertainty associated with the loss of some of the finer particles.

=> There are smaller particles passing the filters with a 0.8  $\mu\text{m}$  porosity. Nevertheless, AA Millipore<sup>®</sup> cellulose esters filters ( $\varnothing$  47 mm, 0.8  $\mu\text{m}$  porosity) are adapted to collecting dust. Regarding the specificities of these filters, Sheldon (Limnology and Oceanography, 1972) pointed out that "all cellulose ester membranes (Millipore) retained particles much smaller than the stated pore size, even from small samples with low particle concentrations". Sheldon (1972) also presented very satisfying retention

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curves for Millipore filters: up to 80% for particles of 0.5  $\mu\text{m}$  in diameter with a 0.45  $\mu\text{m}$  filter porosity, and up to 90% for particles of 1  $\mu\text{m}$  in diameter with a 1.2 filter porosity (estimation based on Figure 2 published in Sheldon R.W., Size separation of marine seston by membrane and glass-fiber filters, Limnology and Oceanography, 17, 3, 494–498, 1972). Sciare et al. (ACP, 2005) presented average concentrations for the particulate mass and chemical species in fine and coarse size fractions in the Eastern Mediterranean Sea during the MINOS campaign in August 2001 (results presented in their Table 1). They distinguished fine ( $< 1.2 \mu\text{m}$ ) and coarse ( $> 1.2 \mu\text{m}$ ) particles. 90% to 95% of the dust concentrations are in the coarse mode, while 84% to 87% of the BC and 82% of the OC are in the fine mode. In order to collect dust particles and to minimize the presence of OC and BC, a filter porosity of 0.8  $\mu\text{m}$  seems relevant.

It may be that this fine fraction is not a major source uncertainty. But if it is, then it would be necessary that the authors consider using smaller pore size filter (slightly longer time does not seem to be a big issue?) or adding a pump although this does add a complexity requiring electricity. Since the pump will only be needed in a short while, it should be able to be powered by car batteries. The study has carefully considered the removal of organic material. However, the black carbon and other anthropogenic particles may still be collected. If the particles are collected onto quartz filter, then it is possible to use a thermal optical carbon analyser to measure both insoluble organic and elemental carbon. The reviewer understand that the purpose of the sampler is for dust deposition evaluation but there is no reason why the sampler could not be used for evaluation of deposition of other types of particles including OC/EC. This may be mentioned in the paper so that a wider impact of the paper could be recognized.

=> If a pump is added, we cannot ensure a passive sampling. => In the present study, the purpose of the sampler is indeed for dust deposition evaluation. The reviewer is right, the sampler could be used for evaluation of other particles, OC/EC, organics etc. after adapting the sampling protocol. => A sentence was added in the paper to recognize the potential wider use of the sampling device: "The CARAGA sampler

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could be used for evaluation of other inorganic or organic particles after adapting the sampling and lab protocols.”

This study assumes that all the insoluble material is from Sahara. This is wrong. When the air masses are coming from the European continent, the deposition can be dust or anthropogenic particles from the Europe. Practically, the method proposed may still be useful if the dust events dominated the total insoluble material deposition. However, the study here and any further studies should discuss in detail the uncertainties associated with the proposed study in estimating Saharan dust deposition. This should include both anthropogenic particles and biomass burning aerosol, both of which will add to uncertainty in estimating dust deposition fluxes.

=> The fact that insoluble fraction is not only Sahara dust is mentioned in the paper, the main other sources being anthropogenic particles, organic and biomass burning aerosol. Bergametti et al. (Tellus, 1989) performed a continuous sampling of particulate matter over a one year period on a Corsican coastal site in the Western Mediterranean. They presented daily atmospheric concentrations measured for elements indicating the major aerosol sources (soil erosion: Al, Si; pollution: S, Pb; sea-salt: Na). The factors influencing the variability of their daily and seasonal concentrations were discussed. The authors conclude that strong daily variations of concentrations are mainly due to the Saharan dusts inputs (for the crustal elements) and to the removal of aerosols by precipitation events. Based on the measurements of the MINOS experiment which took place in summer in the Eastern Mediterranean Basin, Lelieveld et al. (Science, 2002) indicate that “about 80 to 90% of the fine aerosol fraction (< 2  $\mu\text{m}$  in diameter) originates from non-natural sources (BC 6%, etc.). The coarse fraction (> 2  $\mu\text{m}$  in diameter), on the other hand, is probably about 60 to 80% natural” (mainly dust 42%, and sea salts 32%). Nevertheless, the atmospheric concentration of dust in the coarse mode is one order of magnitude higher than the particulate mass concentration of ammonium sulfate and carbonaceous aerosols which composed more than 90% of fine aerosols (Sciare et al., ACP, 2003). These results point out that anthro-

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pogenic aerosols and biomass burning present in the Mediterranean atmosphere could constitute a background deposition flux, but during Saharan dust outbreaks mineral deposition sampled on 0.8  $\mu\text{m}$  porosity filter is mainly due to Saharan dust.

=> This discussion and the references are added in the paper. - Bergametti et al., Seasonal variability of the elemental composition of atmospheric aerosol particles over the northwestern Mediterranean, Tellus, 41B, 353-361, 1989. - Lelieveld et al., Global Air Pollution Crossroads over the Mediterranean, Science, 298, 794-799, 2002.

It is recommended that the authors conduct chemical analyses (including metals, crustal elements, and OC/EC) on the collected samples, which will provide important information on the sources of aerosol as well as the deposition flux of insoluble materials other than dust. The authors should be very careful when deciding where to deploy the samplers. Dust deposition fluxes estimated at sites that are subject to intensive anthropogenic or biomass burning emissions could be largely uncertain.

=> Chemical analyses are far away from the scope of the paper, even if they can be helpful to interpret the sources of aerosols. Moreover, to perform chemical elemental analyses, the sampling protocol needs to be adapted, depending on the studied elements and concentrations encountered. We also have to keep in mind that the sampling is focused on coarse particles (a main part of OC/EC could not be collected) and insoluble deposition (the soluble fraction of is not collected). Therefore the interpretation of chemical analyses could be tricky. These points need to be fully evaluated in another study before conducting chemical analyses. As mentioned by the reviewer, the sampling sites have to be carefully chosen depending on the scientific objectives of the study.

The authors should clarify what the white dots on some of the filters and what is the dark dot on the filter of fourth row and 2nd column.

=> Passive water filtration may lead to accumulation of deposits on certain parts of the filters and other parts without deposited matter (white dots). The filter of fourth row

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and second column in Figure 3 corresponds to the sampling period from October 27 to November 17 and mentioned in the legend of Figure 4. Due to technical problems, no rotation happened during these 3 weeks. During this sampling period, some undefined brown deposits were collected on this filter and may have been accumulated in a part of the filter due to water filtration. When we looked at this filter, we were not able to determine the composition of this brownish deposit but we suspected that it was mainly organic. The deposition mass flux after ignition protocol was 0.26 g m<sup>-2</sup> for this 3 weeks period.

Table 2: clarify what filter; further tests are needed to check how the mass of different particles will change without filter: we know that dust do contain some water and volatile components which may be lost during heating; clarify what is "finest mineral"?

=> Filters refer to cellulose esters filters. This is now mentioned in the legend of Tables 2 and 3. We wanted to perform the tests under the same conditions than for samples collected in-situ. This is why the tests are done for a crushed fraction of soil deposited on the filters. The "finest mineral soil fraction" was replaced by "crushed soil fraction" in the text. It corresponds to soils of Cape Verde and Douz crushed in the lab. It is now mentioned in the text.

Have you checked the blank filter mass change at different temperature? This can be done but putting a blank filter on Al foil. I would like to echo the points by the first reviewer that it is necessary to consider insect contamination. Also new measures should be in place to prevent the bird standing on the funnel causing all sorts of contamination.

=> Yes, we checked the blank filter mass change at different temperature. For instance, there is no noticeable mass change between ambient T° and 40°C. Tests were also done at high T° up to the calcination of the filter. The questions on insect contamination are addressed in the answer to the first reviewer. In order to limit the risk of bird standing, the top of the funnel is made very thin and sharp. Organic contaminations by birds flying over the collector were sometimes observed inside and outside

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the funnel. In this case, the funnel is carefully cleaned every 6 months when the filter series are changed. Organic matters (small insects, pollens, bird's contaminations) collected on the filter during precipitation or automatic funnel rinsing are burned during the calcination protocol.

=> A sentence was added in the section presenting the CARAGA: "In order to limit the risk of bird standing, the top of the funnel is made thin and sharp." => A sentence was added in the section presenting the deposition dataset: "When insects, vegetal debris, pollens or organic matters are collected on the filters they are manually removed only if this manipulation does not affect the sample. If the removal of these elements could damage the sample, we leave them on the filter and the ignition of the samples at 550°C eliminates these organic matters."

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Interactive comment on Atmos. Meas. Tech. Discuss., 8, 2299, 2015.

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