Interactive comment on "In-situ physical and chemical characterization of the Eyjafjallajökull aerosol plume in the free troposphere over Italy" by S. Sandrini et al.

Anonymous Referee #2

Received and published: 18 September 2013

REFEREE: The manuscripts gives a quite complete description of the characterization of two Eyjafjallajökull
aerosol plumes detected at the climatological station of Monte Cimone, in the Italian Appennines. This is one more study on the same subject but has some peculiarities (site latitude and height, PM speciation) which, in my opinion, make it interesting and suitable for publication in ACP. The overall quality of the manuscript is good however I recommend to consider the following points/issues:

1) Pag. 5, line 16: actually the MAAP measures the aerosol absorption coefficient which is usually indicated with babs while sigmaabs is fixed in the MAAP at the value of 6.5 m2/g

ANSWER: There are several papers using σ_{abs} and σ_{sca} for identifying the main optical aerosol properties (absorption and scattering). Anyway, in order to avoid confusion, we changed the indication b_{abs} in the paper.

REFEREE: 2) pag 7, line 1-5: this is my major concern. The list of elements measured by PIXE/PIGE includes those for Na to K which determination by X ray fluorescence techniques is very complex when the PM is collected on quartz fiber filters (reasons are the tails of the huge Si peak and the X-ray self-attenuation in the filter thickness). As a matter of fact is nearly impossible a reliable quantification of these elements in the experimental conditions quoted in the manuscript (further problems could also come for Al and Mg due to self-attenuation in the PM grains in the coarse fraction). In the text the elemental concentration values are quoted several times and it is not clear when they have been measured by PIXE and when by ICP. Information on the particular quartz fibre filters used in the experiment should be also provided. A clear and complete discussion on this issue in this paragraph is needed while, along the text, values obtained by PIXE and ICP should be indicated. Furthermore: did PIGE play any role? If so, please discuss it otherwise it should not be mentioned.

ANSWER: The referee correctly points out the main critical issue of PIXE measurements on particulate matter collected on quartz fibre filters, i.e. the detection of the light elements with X-rays peaks rising above the low-energy tail of the Si X-ray peak and the attenuation problems arising by the absorption of the less energetic X-rays both inside coarse particles and in the quartz fibre filter matrix, that may lead to an underestimation of the light elements.
As concerns the first issue, the authors agree that Na, Mg and Al detection by PIXE is not a trivial exercise when particulate is collected on quartz fibre filters due to the high Si signal from the filter; nonetheless, the set-up of the LABEC laboratory where measurements were performed includes a

Silicon Drift Detector (SDD) for the detection of the low-Z elements: this detector sustains high counting rates, thus allowing the quantification of also Na, Mg and Al, even if, obviously, with higher detection limits than with other filter media. Moreover the use of pile-up rejection circuitry in the signal processing electronic chain, allows to reduce the background and not to worsen the detection limit in the energy region above the Si X-ray peak up to 3.5 keV (close to K X-ray lines). More information on the set-up and an example of the acquired spectra during PIXE measurements on particulate matter collected on quartz fibre filters may be found in the cited paper "Calzolai et al., 2006".
Coming to the second issue, the authors acknowledge that PIXE may underestimate light elements concentrations due to attenuation problems; therefore, they used the PIGE technique, which is not affected by such problems as it exploits the much more energetic gamma radiation, in order to estimate suitable correction factors for the attenuation effects.
As pointed out by the referee, the paper lacks of information on PIGE measurements; we thank him/her for detecting this omission. Comments about the possible underestimation by PIXE and the role of PIGE measurements in order to estimate correction factors allowing an accurate quantification of light elements have been added in the text, as suggested by the referee.

The following paragraph has been added after the PIXE description at par. 2.3 and the references to Calzolai et al., (2010) and to Formenti et al., (2010) added in the References.

"Nevertheless, PIXE may underestimate the concentrations of the light elements due to the absorption of the low-energy X-rays both inside the particles in the coarse fraction and in the quartz fibre filter matrix itself. PIGE exploits the much more energetic γ-ray radiation and therefore it does not suffer from the afore-mentioned absorption problems [Calzolai et al. 2010]. However, a quantitative PIGE analysis may be performed only for selected elements; in the present case, PIGE measurements of Al were performed simultaneously to PIXE in order to estimate attenuation correction factors for an accurate quantification of the lighter elements (Na to Ti) (Formenti et al. 2010)."

Information on the particular quartz fibre filters used in the experiment have been added in the text in par. 2.3 "Chemical Analyses" as suggested by the referee.

REFEREE: 3) pag 15 and in general ash and PM composition, Fig. 6, etc: all this parts are connected to my comments at point 2....

ANSWER: Elemental concentrations obtained by PIXE-PIGE or ICP-OES have been specified in the text

REFEREE: 4) table 1: do the uncertainties quoted in the table represent the SD of the measured values?
ANSWER: Yes, the uncertainties represent the standard deviation of the measured values. This information has been added in the table caption.

REFEREE: 5) pag 7, line 28: XRF should be ED-XRF
ANSWER: Corrected in the text.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 20195, 2013.