

Development of a versatile source apportionment analysis based on positive matrix factorization: a case study of the seasonal variation of organic aerosol sources in Estonia

Athanasia Vlachou¹, Anna Tobler¹, Houssni Lamkaddam¹, Francesco Canonaco¹, Kaspar R. Daellenbach^{1,a}, Jean-Luc Jaffrezo², María Cruz Minguillón³, Marek Maasikmets⁴, Erik Teinemaa⁴, Urs Baltensperger¹, Imad El Haddad¹, and André S. H. Prévôt¹

¹Department of General Energy Research, Paul Scherrer Institute, Villigen PSI, CH-5232, Switzerland

²Université Grenoble Alpes, CNRS, IRD, G-INP, IGE, 38000 Grenoble, France

³Institute of Environmental Assessment and Water Research (IDAEA), CSIC, 08034 Barcelona, Spain

⁴Estonian Environmental Research Centre, 10617, Tallinn, Estonia

^anow at: Institute for Atmospheric and Earth System Research/Physics, Faculty of Science, University of Helsinki, P.O. Box 64, 00014, Helsinki, Finland

Author's response:

We thank Referee #1 for the careful revision and comments which helped improving the overall quality of the manuscript. A point-by-point answer (in regular typeset) to the referees' remarks (in the *italic typeset*) follows. Changes to the manuscript are indicated in [blue font](#).

In the following page and lines references refer to the manuscript version reviewed by anonymous Referee #1.

- 1) *This article presents source apportionment of OA contained in PM10 in Estonia using PMF technique on off-line AMS data sets. It also estimate the uncertainty of the PMF solution by a bootstrap analysis. Overall, this manuscript is well written, and the findings of the study are in agreement with previous studies. I have minor concerns for the manuscript as listed in the following:*

Number of samples: The author collected only 39 samples from KJ. I am afraid that only 10 samples/ season is not enough to present for the seasonal variation of organic aerosols. It would be more useful if the author could describe in more details of sampling time for 150 samples. Do those samples collected from three sites in same sampling days/ time?

We agree with anonymous referee#1 that 39 samples are in general not enough to perform a robust PMF analysis. However, these 39 samples refer only to those from KJ and the PMF analysis was performed on 150 samples collected from three cities during the same period, where aerosol sources are very similar (except for the enhancement in OilOA in KJ). In addition, the errors on the PMF time-series estimated by the bootstrap analysis are similar at the three different locations (Table S3).

Aerosols were sampled for 24h (Page 3, line 32) at the three sites. The samples were not collected on the exact same dates at the 3 sites. Following the reviewer comment, the dates are now displayed in the following

Fig. S1 and Table S1, added in the supplementary. According to the suggestions of anonymous referee#1 we also changed the text in the Methods (Page 3, Line 33) as follows:

The measurements were performed with 24 h integrated PM₁₀ quartz fibre filter samples from KJ (31/08/2013 to 25/08/2014), Tallinn (05/09/2013 to 01/09/2014) and Tartu (05/09/2013 to 31/08/2014) (see Table S1, Fig. S1 for details).

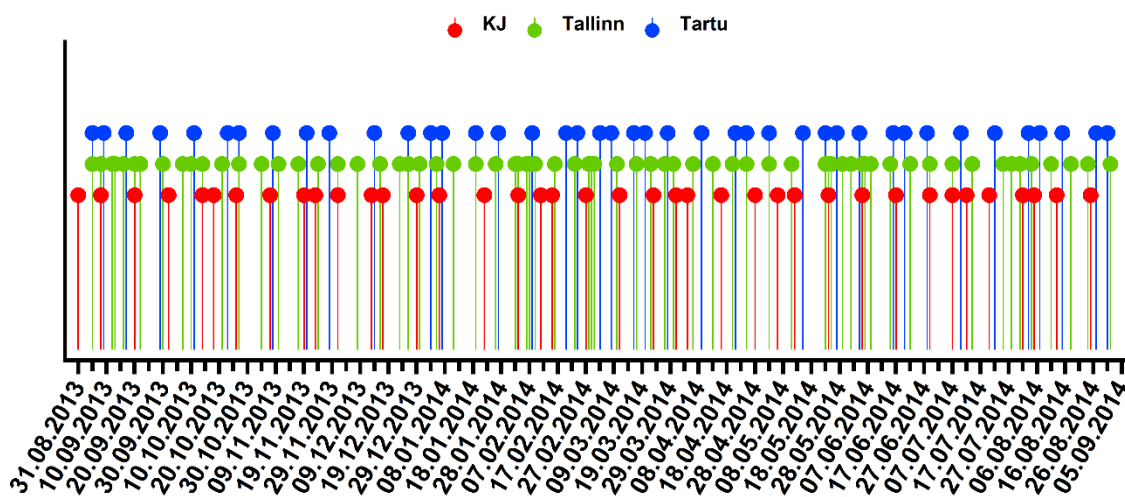


Figure S1. Dates per site. KJ represented in red, Tallinn in green and Tartu in blue.

Table S1. Dates per site.

KJ	Tallinn	Tartu	KJ	Tallinn	Tartu
2013			2014		
31.08.2013					03.01.2014
	05.09.2013	05.09.2013		05.01.2014	
08.09.2013	08.09.2013	09.09.2013	06.01.2014		07.01.2014
	12.09.2013			11.01.2014	
	13.09.2013			19.01.2014	19.01.2014
	16.09.2013		22.01.2014		
		17.09.2013		26.01.2014	
20.09.2013	20.09.2013			02.02.2014	
	22.09.2013				27.01.2014
		29.09.2013	03.02.2014	03.02.2014	
	30.09.2013			06.02.2014	
02.10.2013				07.02.2014	
	07.10.2013				08.02.2014
	10.10.2013			09.02.2014	
		11.10.2013	11.02.2014		
14.10.2013	14.10.2013		15.02.2014		
18.10.2013				16.02.2014	
	21.10.2013				20.02.2014
		23.10.2013		23.02.2014	
26.10.2013					24.02.2014
	27.10.2013	27.10.2013	27.02.2014		
	04.11.2013			28.02.2014	

07.11.2013		08.11.2013		01.03.2014	
	10.11.2013			02.03.2014	04.03.2014
	17.11.2013				08.03.2014
19.11.2013		20.11.2013	11.03.2014	10.03.2014	
23.11.2013	24.11.2013			17.03.2014	16.03.2014
		28.11.2013		22.03.2014	20.03.2014
01.12.2013	01.12.2013		23.03.2014	27.03.2014	
	08.12.2013			30.03.2014	28.03.2014
13.12.2013		14.12.2013	31.03.2014		
	16.12.2013		04.04.2014	06.04.2014	
17.12.2013	23.12.2013			13.04.2014	09.04.2014
	26.12.2013	26.12.2013		20.04.2014	
29.12.2013	30.12.2013		16.04.2014	25.04.2014	21.04.2014
				03.05.2014	25.04.2014
			28.04.2014	11.05.2014	03.05.2014
			06.05.2014		
			12.05.2014	23.05.2014	15.05.2014
				24.05.2014	23.05.2014
			24.05.2014	25.05.2014	
				29.05.2014	27.05.2014
				01.06.2014	
			05.06.2014	05.06.2014	04.06.2014
				06.06.2014	
				08.06.2014	
				15.06.2014	16.06.2014
			17.06.2014		
				22.06.2014	20.06.2014
					28.06.2014
			29.06.2014	29.06.2014	
			07.07.2014	07.07.2014	
					10.07.2014
			12.07.2014		
				14.07.2014	
			20.07.2014		

			22.07.2014
		25.07.2014	
		28.07.2014	
		31.07.2014	
	01.08.2014		
			03.08.2014
		04.08.2014	
	05.08.2014		
			07.08.2014
		11.08.2014	
	13.08.2014		
			15.08.2014
		18.08.2014	
		24.08.2014	
	25.08.2014		
			27.08.2014
			31.08.2014
		01.09.2014	

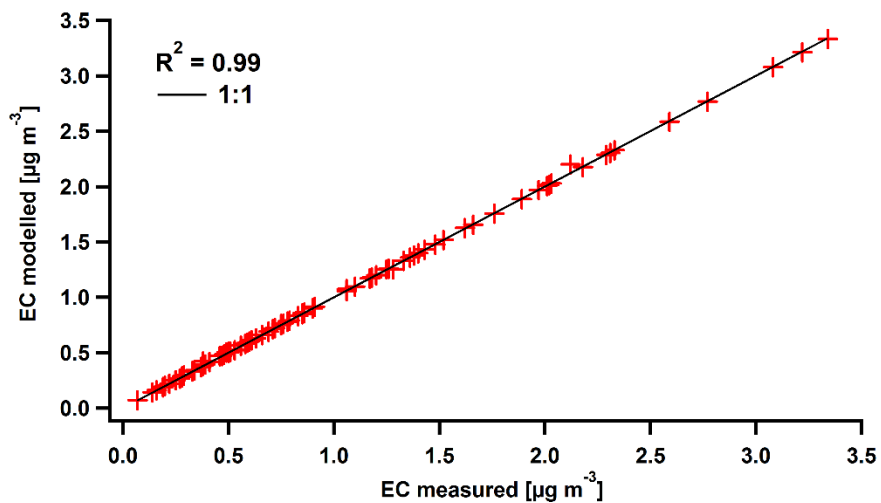
- 2) *PMF techniques: The bootstrap techniques were commonly used to check uncertainty of PMF solutions for decades. I do not think it is "novel technique" as the author state in Page 3, Line 10. Could the author explain more the novelty of their bootstrap techniques?*

We did not intend to claim that the bootstrap technique used here for the uncertainty exploration of PMF is novel. We apologize for this misunderstanding. The novelty corresponds to the selection technique applied after the bootstrap analysis, which enables evaluating the quality of a large set of solutions. We changed the text accordingly (Page 3, Line 10):

In this study, we propose a novel technique to evaluate the selection of the PMF solutions generated through a large number of bootstrap iterations.

- 3) *In equation 3, do the authors assume the EC/WSOA ratio for each source is constant? If yes, please provide any references for that. In addition, the author should provide the correlation between measured EC and modeled EC concentration based on equation (3).*

According to the definition of PMF (Paatero, 1999), the sources are represented by constant profiles and varying intensities; therefore, as noted by the reviewer the EC/WSOA ratio for each source is constant. We note that this ratio is not assumed a priori, but determined through the PMF analysis. We also note that the PMF analysis performed to assess the EC sources is fully constrained. Therefore, EC modelled corresponds to the measured EC as shown by the figure below:



Correlation between EC measured by the thermal/optical method and the modelled EC calculated by PMF (EC = EC_{tr} + EC_{bb} + EC_{oil} + EC_{rrd}).