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Dear Editor-in-Chief,

We would like to thank the referees for their comments that aim at improving the manuscript. Hereafter, we have replied to the different comments. Amendments in the manuscript were done in track changes mode for ease of review. Additionally, a new DOI was generated for the updated database (<https://doi.org/10.5281/zenodo.8022729>).

### **Answers to referee # 1**

#### **General comment**

The paper reports a study of maritime contributions to PM<sub>10</sub> in a rural coastal area of northern France. The topic is of interest and suitable for the Journal. The approach used is quite standard, however, the extensive dataset, the detailed statistical analysis, and the limited availability of data for this area make the paper useful to the scientific community. I would like to see it published after a revision step addressing my minor specific comments.

*We would like to thank the referee for his constructive comments that aim at improving the manuscript. Hereafter, we have replied to the different comments in blue. Amendments in the manuscript has been done accordingly.*

#### **Specific comments**

1. Please remove all instances of etc. in the paper. If something should be added, please do it explicitly.

*The « etc. » term has been removed from lines 42, 44, and 54 of the original manuscript.*

2. Line 67. Please remove the “+”. In addition, this value seems to be very large compared to the measured ones and to the values published in other studies. What it represents Annual average or other short-term contribution?

*The « + » symbol will be removed from line 67 of the original manuscript. This information was retrieved from Ledoux et al. (2018) that studied the influence of shipping emissions on PM concentrations in the port city of Calais, one of the busiest harbors in Europe. The sampling was conducted for three months and the sampling site was situated at a distance of 500 to 1000 meters from boats. PM<sub>10</sub> concentrations were deduced from the real-time particle size distribution measured by an Electrical Low Pressure Impactor. The temporal resolution for PM<sub>10</sub> was 1 min and 15 min for gas concentrations.*

*In order to estimate the impact of in-port ships on PM<sub>10</sub> concentrations, the authors have selected the data registered when the wind was blowing from the direction of the port. Periods showing no concentration peaks of PM, SO<sub>2</sub>, NO, and NO<sub>2</sub> represented the background concentrations whereas periods for which successive peaks were detected represented the “Background + in-port shipping related concentrations”. Results showed that the in-port ship emission increased the background PM<sub>10</sub> concentration by an average of 28.9 µg/m<sup>3</sup> with a maximum increase of 78.9 µg/m<sup>3</sup> for a 1-minute time interval (the value presented in line 68 of the manuscript).*

*The information is modified in the manuscript as follows (Line 67 in the original manuscript):” Punctually, ship emissions led to a marked increase of PM<sub>10</sub> mass of 78.9 µg/m<sup>3</sup> for a one-minute time interval.”*

3. Lines 77-80. It would be worth to mention here that the use of low-sulphur fuels actually reduce also primary PM emissions from ships how it has been demonstrated in several study and, of course, also secondary sulphate. So that the regulation was not done only for SO<sub>2</sub>.

*The authors agree with the referee. The information has been added at the end of the introduction:*

*“Additionally, despite the IMO regulation for global sulfur limit of 0.5% from ship’s fuel oil applied starting January 2020, different countries are still adopting higher sulfur limits. It is worth*

*noting that these limits were set for sulfur content in marine fuels in order to reduce not only SO<sub>2</sub> emissions but also primary PM emissions (Zetterdahl et al., 2016; Shen and Li, 2020). Hence, this study aims at highlighting the contribution of the natural and anthropogenic marine emissions to the PM<sub>10</sub> concentrations in a rural coastal French site.”*

4. Line 101. Are these quartz filters?

*The Pall® QAT-UP filters are indeed no binder quartz filters. The sentence (lines 100-101) has been modified as follows:*

*“Moreover, PM<sub>10</sub> were collected using an automated high-volume sampler (DA80, DIGITEL®, Switzerland) operating at 30 m<sup>3</sup>/h onto 150 mm Pall Tissuquartz™ 2500 QAT-UP filters (no binder) filters.”*

5. Line 172. Are you referring to secondary organic aerosol here?

*The authors were not referring to secondary organic aerosols. The interpretation found in this sentence was based on the comparison between the average concentration of PM<sub>10</sub> at Cape Gris-Nez which is a rural coastal site (with no primary emission sources nearby) and the one at other urban and industrial sites in the Northern France region (during the same period of one year). PM<sub>10</sub> concentrations are mainly governed by the fluctuations of the background levels observed at a regional scale. Additionally, the long-range transport of different organic and inorganic species to Cape Gris-Nez have also an impact on the PM<sub>10</sub> concentration. This was also highlighted in the CWT representations showing that more than 80% of PM<sub>10</sub> concentrations was attributed to regional and/or long-range transport origins.*

6. Section 3.2. Considering that the receptor model used is still not widely applied, it should be useful to mention that it has comparable performances to the PMF model investigated in the recent work of Belis et al. (Atmospheric Environment: X, 5, 2020, 100053).

*The authors thank the referee for the suggestion. However, the “CW-NMF” model developed by the LISIC (Laboratoire d'Informatique Signal et Image de la Côte d'Opale) at the University of Littoral Côte d'Opale and that was used in this study was not mentioned in the recent work of Belis et al., 2020. For this reason, the reference will not be added in this particular context. However, it will be added in the introduction of the manuscript to highlight the importance of the source apportionment models.*

7. Lines 247-256. For the discussion on the V/Ni ratio, I suggest to have a look and mention the work of Gregoris et al (Environ Sci Pollut Res (2016) 23, 6951–6959) that shows ratios lower than expected in coastal areas with relevant impact of shipping as well as a strong spatial variability of this ratio.

*The authors would like to thank the referee for this suggestion. The reference of Gregoris et al. (2016) as well as the information within the paper related to the V/Ni concentration ratio will be added to the final manuscript as follows:*

*“Heavy Fuel Oil (HFO) combustion profile was composed of high loadings of carbonaceous matter (OC and EC) as well as high contributions of V, Ni,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ . The concentration of OC in this profile is 4.7 times higher than EC, which is consistent with other studies (Zhang et al., 2016). The evaluation of the V-to-Ni concentration ratio show a value of 1.3 in this study which is lower than the ones usually found (close to 3) from shipping emissions (Pandolfi et al., 2011; Becagli et al., 2012). Similar observations (V-to-Ni ratio of 1.6) were previously reported for other sites in the Northern region of France (Ledoux et al., 2017). This is mainly due to the position of the sampling sites reported in the latter study as well as the site of this study that are located in a SECA where sulfur content in marine fuels is limited to 1% during the sampling period in 2013, whereas the sites reported by Pandolfi et al. (2011) and Becagli et al. (2012) were not. The quality of the fuel with a low sulfur content used for shipping was shown to contain as well low contents of V and Ni and by that change their ratio (Zhang et al., 2016; Streibel et al., 2017).” **Gregoris et***

*al. (2016) also found V/Ni concentration ratios that were less than 3 in Venice and the species were mainly attributed to HFO combustion from shipping.*

8. Lines 293-295. I would not say that it is underestimated. The point is that it was analysed here only the contribution to primary PM<sub>10</sub> and that there would be also a contribution to secondary aerosol, mainly sulphate, that could be even larger than the primary one.

*This work presented an exhaustive chemical characterization of PM<sub>10</sub> including primary species as well as secondary ones, namely the secondary inorganic aerosols (NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and NH<sub>4</sub><sup>+</sup>). The concentration levels of these secondary species characterized in PM<sub>10</sub> can be found in Table 1 of the manuscript. Additionally, these secondary species were added into the CW-NMF model in order to identify different PM<sub>10</sub> sources and quantify their contribution as well. The HFO combustion profile (Lines 247-256 in the original manuscript) was identified by high loadings of carbonaceous matter, V and Ni, as well as secondary inorganic species (NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>). By that, we cannot say that the HFO combustion source corresponds only to primary PM<sub>10</sub> emissions.*

*However, the authors wanted to highlight the idea that the transformation reaction of SO<sub>2</sub> emitted from the shipping emissions to SO<sub>4</sub><sup>2-</sup> is probably not at the equilibrium yet and maybe more amounts of SO<sub>4</sub><sup>2-</sup> were transformed which may lead to higher contribution of shipping emissions to PM<sub>10</sub>. That is why the authors mentioned the underestimation of the shipping emissions contribution to primary PM<sub>10</sub>.*

*The sentence in the original manuscript was modified as follows:*

*“However, considerable amounts of SO<sub>2</sub> and NO<sub>x</sub> can be emitted from shipping activities and can be transformed into secondary compounds by gas-to-particle conversion that largely contribute to PM mass. Ledoux et al. (2018) have reported that the impact of shipping in the harbor of Calais in Northern France was estimated to 35% of NO, 51% of SO<sub>2</sub>, and 15% of NO<sub>2</sub> average concentrations.*

*Nevertheless, the transformation reaction of SO<sub>2</sub> emitted from the shipping emissions to SO<sub>4</sub><sup>2-</sup> is probably not at the equilibrium yet and maybe SO<sub>2</sub> emissions contribute to the formation of more SO<sub>4</sub><sup>2-</sup> than the quantity sampled close to the sea.*

9. Lines 322-325. Please mention how and where these gases are measured.

*The information regarding the measurement of the gas concentrations will be added in the supplementary material:*

*“SO<sub>2</sub> and NO<sub>2</sub> were continuously measured using an AF21M SO<sub>2</sub> analyzer (Environnement SA, France) and an AC32M NO-NO<sub>2</sub>-NO<sub>x</sub> analyzer (Environnement SA, France), respectively. The calibration of the analyzers was done at the beginning of the campaign and were routinely check by the regional air quality network atmo Hauts-de-France. The temporal resolution was 15 minutes for gas concentrations. More details related to the measurement campaign of the gases at Cape Gris-Nez can be found in Ledoux et al. (2018).”*

10. Figure 4. Please use the same acronym as in the text (i.e. CBPF). To be honest, I do not understand why the CBPF of sea salt and aged sea salt are so different. A better interpretation of this aspect would be useful.

*The terms “CPF” in Figure 4 will be replaced by CBPF.*

*The difference between the CBPF of fresh and aged sea-salts is mainly due to the meteorological conditions as mentioned in the manuscript (Line 317) and the seasonality. It is important to remember that the sampling site at Cape Gris-Nez is a rural coastal site, located at 200 m from the sea and therefore strongly subjected to fresh marine influence from sectors 210° to 50° via the north. This is why, for the fresh sea-salts, the maximum concentrations were observed for winds blowing from the southwest and the northeast sectors corresponding to the English Channel and the North Sea (for medium to high wind speeds > 10m/s).*

*As for the aged sea salts, the highest concentrations were observed for winds blowing from the northeast. In fact, the aged sea-salts are yielded by the reaction between the fresh sea-salts and the SO<sub>2</sub> and NO<sub>2</sub> gases that show the highest concentrations levels on land (Northeast wind sector – Figure S6). This phenomenon will be done according to the trajectory of the air masses. By that, the aged sea-salts may not come from the wind direction open to the sea but from land (Northeast wind sector) especially in a coastal site, which is the case of this study.*

11. Figure S4. The use of three different level of blue is not a good choice because make the figure hardly readable generating confusions among the different sources. Please make a different choice for these colours

*Following the referee's recommendation, various colors will be used for Figure S4 instead of the different levels of blue.*

## **Answers to referee # 2**

### **General comment**

The manuscript reports a standard source apportionment study with a focus on the maritime source. Beyond the original results and the use of a specific receptor site to achieve the goals, the rest of the study presents only the trivial application of well-established source apportionment techniques (PMF-like, CWT, CPF). Another weakness of this study is the "age" of the data, which were collected 10 years ago. However, since the lack of papers focusing on maritime sources, I am in favour of publication. The authors are requested to improve the manuscript by adding more information about the outcomes of the CW-NMF model (in particular about the number of factors extracted).

*We would like to thank the referee for his constructive comments that aim at improving the manuscript. We understand the referee's point of view regarding the outcomes of the CW-NMF model and the "overfitting" problem addressed in the following comments. We have checked again the reconstruction of the species added into the model and came to the conclusion that three elements (La, Mn, and Cr) were not as well reconstructed as the other species. For that purpose,*

*we had run again the CW-NMF by removing these elements. In this case, the optimal number of factors obtained was nine, removing by that the “metal-rich” source. The outputs of the CW-NMF model specifically the reconstruction of PM<sub>10</sub> as well as the different species and the detailed bootstrap analysis that show the robustness of the 9-factor solution will be presented in details in the following comments and will be added in the supplementary information. Amendments in the manuscript especially regarding the outcomes of the CW-NMF model have been done.*

### **Specific comments**

1. Lines 76-79 “Additionally, despite the IMO regulation for global sulfur limit of 0.5% from ship’s fuel oil applied starting January 2020, different countries are still adopting higher sulfur limits. It is worth noting that these limits were only set for sulfur content in marine fuels in order to reduce SO<sub>2</sub> emissions but no regulations for PM components neither in the sea nor at ports were issued”. Not clear why this consideration is reported within the objectives of the study. Is this topic addressed in the current study? Please clarify.

*The authors understand the point of view of the referee. This topic is not addressed in the current study. However, it was relevant to include it in this part of the introduction in order to shed the light on the idea that the data is still relevant nowadays since different countries are still adopting sulfur limits higher than 0.5%, which was the case in 2013. Additionally, it was added following the recommendation of the referee #1.*

2. Lines 66-80. Please comment on the choice of sampling PM<sub>2.5</sub> to quantify the maritime contribution. Combustion emissions are expected to be finer than other sources. On the contrary, other sources in the coastal areas (e.g., sea salt) may emit large particles. Thus, it would be better to analyze PM<sub>2.5</sub>. Please comment to support your choice.

*We agree with the referee regarding the fact that combustion emissions are expected to be in the finer fraction of PM such as PM<sub>2.5</sub>. However, the study was conducted in 2013. Several years before, the EU has placed policies in order to reduce the concentrations of some atmospheric pollutants. The directive 2008/50/EC limits daily PM<sub>10</sub> concentrations to 50 µg/m<sup>3</sup> with a*



*maximum of 35 days of exceedance per year. This directive is binding and forces countries that do not comply with it to seek solutions for improvement. This was particularly the case of France, which was in a dispute on this subject with the European Union. Several regions in France were thus concerned with high number of exceedance days, especially the Northern region. This is why it was important at the time to identify the sources that contribute to the concentrations of PM<sub>10</sub> in order to focus on the reduction of emissions of these sources and to quantify the part of maritime contribution.*

3. Line 90. “The sampling site is far from major continental pollution sources and can be considered as a background site.” Which background? Rural, urban, suburban? If rural, are you sure the site has the characteristics to be classified in this way? Please comment.

*We agree with the comment of the referee regarding the classification of the site as “background”. According to the current Implementing Provisions on Reporting (IPR) 2011/850/EC regarding the type of sampling area and the influence of the immediate surroundings (traffic, industrial or background), the site cannot be considered as a background site due to its coastal typology with marine anthropogenic emissions ([https://www.eionet.europa.eu/aqportal/doc/IPR%20guidance\\_2.0.1\\_final.pdf](https://www.eionet.europa.eu/aqportal/doc/IPR%20guidance_2.0.1_final.pdf)). The term “background” will be removed from the manuscript. As for the definition of a rural site: it corresponds to an area that does not fulfill the criteria of an urban or suburban site with at least 10 km from major sources. The sampling site can be indeed considered as a rural site on the coast.*

4. Section 2.2. The sampling campaign is 1 year long (1st of January to 31st of December 2013) and samples were collected at daily frequency, however only 122 samples were analyzed. Why? How the samples to be analyzed have been selected? Randomly? 1 day over 3? Please explain.

*The sampling campaign was conducted between the 1<sup>st</sup> of January and the 31<sup>st</sup> of December 2013. A total number of 362 samples were collected at Cape Gris-Nez in 2013. 122 samples were chosen*

*for the chemical analysis. They were not chosen randomly and they represent a sampling of 1 day over 3 in order to ensure that these samples are representative of the whole sampling period.*

*The information was added in the manuscript:*

*“Over the sampling period, 362 samples have been collected. 122 samples representing a sampling of one day over three were chosen for the chemical analysis and were presented in this study.”*

5. Line 100. Was the beta monitor tested against the gravimetric measurement? Which calibration audit? Please add info.

*The beta monitor results were not compared to the gravimetric measurement of the filters. However, the LCSQA (Laboratoire central de la surveillance de la qualité de l'air) in France has verified the equivalence of the MPI01M-RST measurements using the beta gauge with the gravimetric reference method in their study conducted in 2011 and published in 2012 (LCSQA, 2012).*

*The information will be added in the final version of the manuscript as follows (Line 100): “During the sampling period, PM<sub>10</sub> concentrations were monitored using a MPI01 beta gauge analyzer (Environment SA®). The analyzer was calibrated at the beginning of the campaign and routinely checked by the regional air quality network atmo Hauts-de-France”.*

6. Line 168 “The yearly-mean PM<sub>10</sub> concentration obtained for the set of sampling days was of 24.3 µg/m<sup>3</sup>”. Is this the real yearly average (365 days in a year possibly measured with the beta monitor) or the average over the 122 analyzed samples? It would be great to see both values to understand if the sample selection is consistent with (indicative of, or similar to) the year average.

The  $PM_{10}$  concentration of  $24.3 \mu\text{g}/\text{m}^3$  reported in the manuscript corresponds to the average over the 122 analyzed samples. For the yearly average considering all the samples, the average concentration was  $22.8 \mu\text{g}/\text{m}^3$ . The closeness of the two values is indicative that the sample selection is representative of the whole year.

7. Figure 2. Please report the error bars relative to the bootstrap results. This would clarify some source profiles.

Following the bootstrap results, the percentiles 25<sup>th</sup> and 75<sup>th</sup> were added to the contribution of the species by factor and were presented in Figure S2.

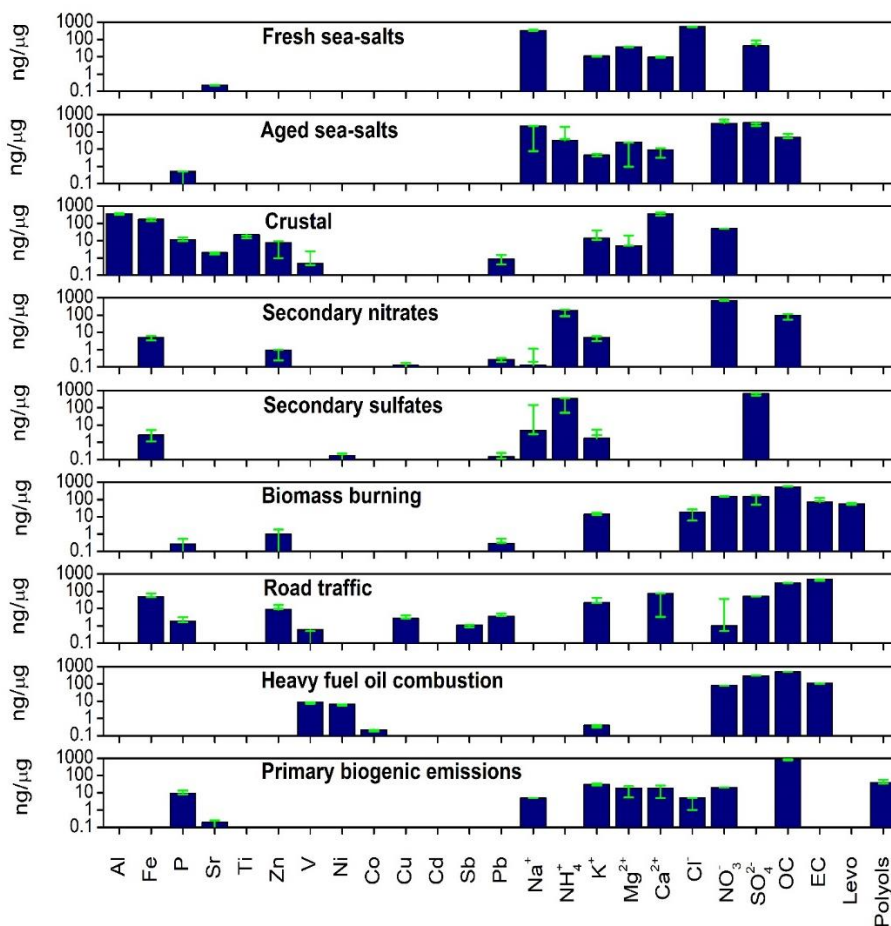


Fig.S2:  $PM_{10}$  source profiles at Cape Gris-Nez (CGN) identified using the CW-NMF model along with the percentiles 25<sup>th</sup> and 75<sup>th</sup> calculated via the bootstrap analysis.

8. Section 3.2. The description of the model setup lacks many details. In addition, it looks like you pushed the model beyond the limits of what it is capable of. It is hard to believe that you can see so many factors with just 122 samples and 28 variables without overfitting the data. For instance, the presence of a “metal-rich” factor may be due to the overfit. Thus, I would ask you to add the scaled residual plots to the SI material file along with all the diagnostics returned by the model. This is to better investigate the goodness of the selected model setup.

*As mentioned before, the authors took into consideration the comment of the referee regarding the “overfitting” of the model.*

*First of all, regarding the number of samples and species, the authors considered a total of 242 samples as input data for the application of CW-NMF model: 122 samples (1 day over three) collected in 2013 + 62 samples collected in 2013 during exceedance days and pollution episodes + 58 samples collected in 2014 at the same site. The contribution of the sources for the 122 samples (1 day over three) in 2013 were extracted from the output data of the model in order to present them in this paper.*

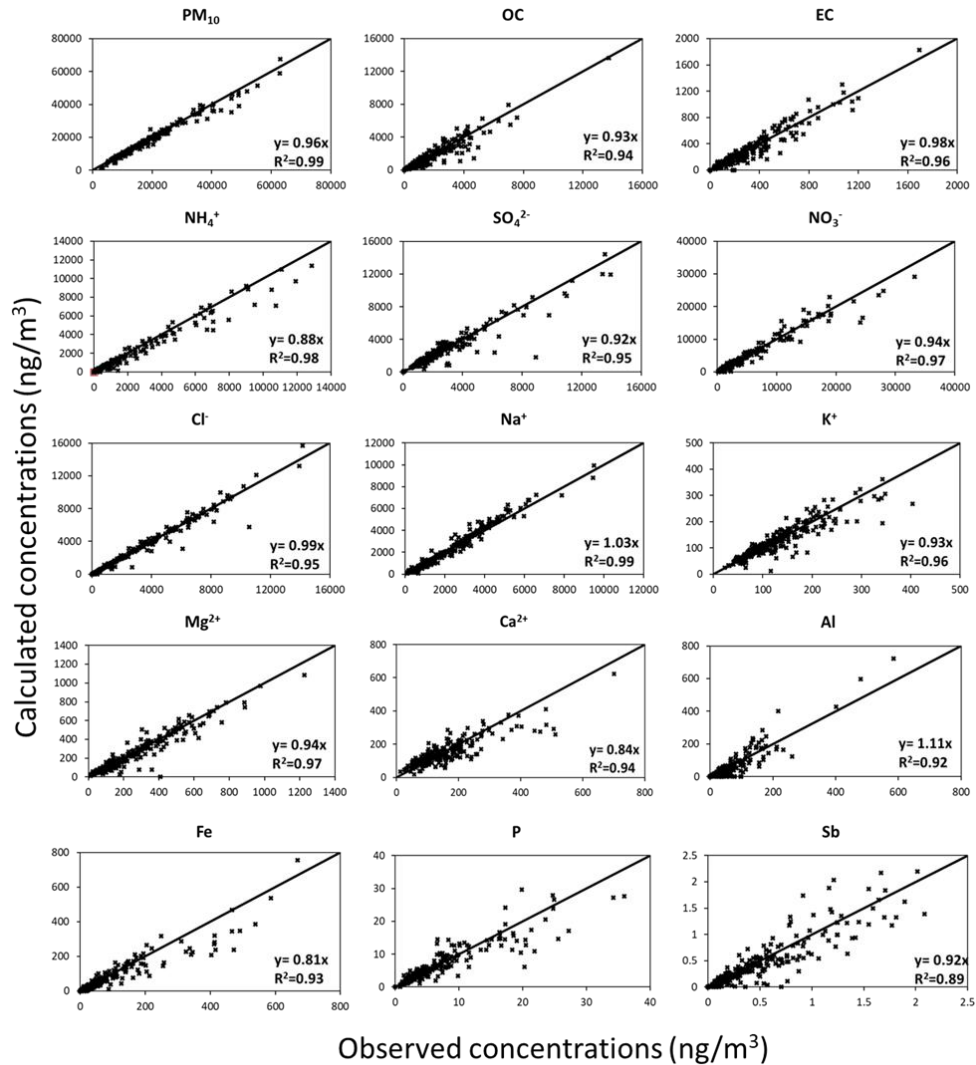
*Second of all, we have checked again the reconstruction of the species added into the model and came to the conclusion that three elements (La, Mn, and Cr) were not as well reconstructed as the other species. For that purpose, we had run again the CW-NMF by removing these elements. In this case, the optimal number of factors obtained was nine, removing by that the “metal-rich” source. The criteria that help us evaluate the goodness and robustness of the solution are the following:*

- *The comparison between the source profiles obtained by the model with the source profiles that can be found in the literature and in databases such as SPECIATE or SPECIEUROPE (Pernigotti et al., 2016; Simon et al., 2010) along with the evaluation of the characteristic ratios of the species in the profiles.*
- *The reconstruction of the concentration of  $PM_{10}$  and all the species added into the model*

- The bootstrap analysis that shows the variability of the solution by studying the effects from random errors and partially including the effect of rotational ambiguity. It is used to find if there is a small set of observations that can largely influence the solution. Mapping over 80% of the factors indicates that the BS uncertainties can be interpreted, and the number of factors may be appropriate.

In the case of the 9-factor solution, the profiles presented in figure 2 were comparable with those of the literature and the details were presented in section 3.2 of the manuscript.

Hereafter, we present the reconstruction of the different species included in the model as well as of  $PM_{10}$  (figures will be included in the supplementary information).



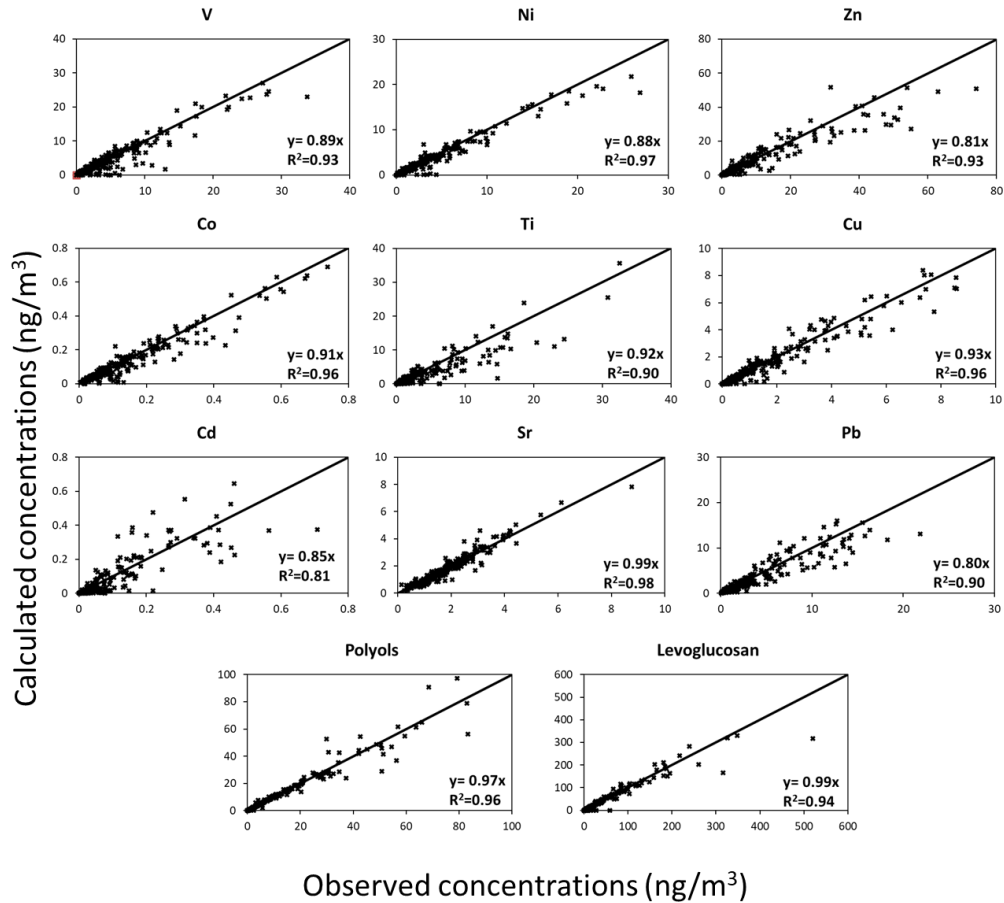


Fig. S3: Calculated species and  $PM_{10}$  concentrations using Constrained Weighted - Non-Negative Matrix Factorization versus observed concentrations for all the species considered in the calculation (with the identity line as reference).

*As we can see,  $PM_{10}$  and all the species were well reconstructed with slopes of the calculated vs observed linear regressions close to 1 (ranging between 0.8 and 1.11) with determination coefficient  $R^2 > 0.9$  with the exception of Cd ( $R^2 = 0.81$ ).*

*Finally, for the bootstrap analysis, the results (presented in the table below) showed values higher than 80%, indicating a stable solution and a reasonable model fit for the 9-factor solution.*

Following all of the above-mentioned arguments, the authors do show that the 9-factor solution is robust and can be presented in the manuscript. For this reason, modifications were done in the manuscript (Section 3.3, 3.4, 3.5 as well as figures 3, 4, and 5 and the supplementary information).

**Table S1:** Bootstrap mapping results for CW-NMF results at CGN site

<b>Bootstrap mapping</b> <b>Min. correlation coefficient <math>r = 0.6</math></b>										
	1	2	3	4	5	6	7	8	9	Unmapped
Boot Factor 1 (Fresh sea-salts)	100	0	0	0	0	0	0	0	0	0
Boot Factor 2 (Aged sea-salts)	0	100	0	0	0	0	0	0	0	0
Boot Factor 3 (Crustal)	0	0	99	0	0	0	0	0	0	1
Boot Factor 4 (Secondary nitrates)	0	0	0	100	0	0	0	0	0	0
Boot Factor 5 (Secondary sulfates)	0	0	0	0	100	0	0	0	0	0
Boot Factor 6 (Biomass burning)	0	0	0	0	0	99	0	0	0	1
Boot Factor 7 (Road traffic)	0	0	0	0	0	0	100	0	0	0
Boot Factor 8 (HFO combustion)	0	0	0	0	0	0	0	100	0	0
Boot Factor 9 (Primary biogenic emissions)	0	0	0	0	0	0	0	0	100	0

9. Line 227. “with an average Cl<sup>-</sup>-to-Na<sup>+</sup> ratio of 1.8 which is commonly observed for fresh sea salts (Seinfeld and Pandis, 2016).” Is the seawater composition also valid for other fresh sea salt ionic species such as K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, and SO<sub>4</sub><sup>2-</sup>?

For the 9-factor solution that is added in the final version of the manuscript, the sea composition is also valid for the other ionic species and are resumed in the following table:

<i>Ratios</i>	<i>Cl<sup>-</sup>/Na<sup>+</sup></i>	<i>K<sup>+</sup>/Na<sup>+</sup></i>	<i>Ca<sup>2+</sup>/Na<sup>+</sup></i>	<i>SO<sub>4</sub><sup>2-</sup>/Na<sup>+</sup></i>	<i>Mg<sup>2+</sup>/Na<sup>+</sup></i>
<i>Sea-water composition</i>	<i>1.8</i>	<i>0.037</i>	<i>0.038</i>	<i>0.251</i>	<i>0.12</i>
<i>Fresh sea-salts profile</i>	<i>1.7</i>	<i>0.033</i>	<i>0.027</i>	<i>0.15</i>	<i>0.11</i>

10. Line 230. Is the cations/anions ratio balanced in this factor?

*The cations/anions ratio in the “aged sea-salts” profile in the 9-factor solution is balanced with a cations/anions ratio of 1.14.*

11. Line 240-255. The authors report some literature data in support of their findings. However, most of these studies refer to finer PM (PM<sub>2.5</sub> or even PM<sub>1</sub>). For example, the paper by Khan et al (2021) reports the EC to levoglucosan ratio in PM<sub>1</sub>. The paper by Salameh et al (2018) refers to the OC-to-EC ratio of PM<sub>2.5</sub>. It is not reliable that the ratios between different variables (chemical species) remain unchanged on PM<sub>10</sub>, PM<sub>2.5</sub> or PM<sub>1</sub>. At least, not all. Please comment or change the references.

*The authors understand the point of view of the referee. The references were changed in order to reflect the concentration ratios between species in the PM<sub>10</sub> fraction. The references of Khan et al., (2021) and Salameh et al., (2018) were replaced by others that present concentration ratios in PM<sub>10</sub> (Amato et al., 2011; Waked et al., 2014; Sonwani et al., 2021).*

12. Figure 3. Please provide the uncertainty associated with the results.

*We agree with the referee on the importance of having uncertainty values for the sources' contributions. However, the output data of the CW-NMF model, just like the PMF model, does not include uncertainties associated with the contribution of the sources. Instead, the average concentration as well as the standard deviations of each source in ng/m<sup>3</sup> were added in the supplementary information.*



Table S2: Average contribution and standard deviation in  $\mu\text{g}/\text{m}^3$  and percentages of the factors to  $\text{PM}_{10}$

<b>Source</b>	<b>Contrib. at CGN (<math>\mu\text{g}\cdot\text{m}^{-3}</math>)</b>	<b>Standard deviation (<math>\mu\text{g}\cdot\text{m}^{-3}</math>)</b>	<b>Contrib. at CGN to <math>\text{PM}_{10}</math> (%)</b>
Fresh sea-salts	4.24	4.57	21.9%
Aged sea-salts	3.0	2.31	15.4%
Crustal	0.18	0.28	0.9%
Secondary nitrates	6.27	9.03	32.3%
Secondary sulfates	1.91	3.24	9.8%
Biomass burning	1.49	3.25	7.7%
Road traffic	0.54	0.61	2.8%
HFO combustion	0.85	1.01	4.5%
Primary biogenic emissions	0.92	1.40	4.7%

13. Section 3.5. It appears that most of the factors come from the same area of origin. This again pinpoints the possible data overfit. However, the results for HFO appear to come from a more "marine" source area than the two secondary factors. It would be better to zoom in on Figure 5 to better visualize the differences. After all, we are not interested in results where the model returns too low a contribution (thus, please cut the more distant areas).

*We agree with the referee regarding his comment to figure 5. The figure was changed by replacing the contribution of the sources with the ones determined for the 9-factor solution. Additionally, the CWT representations were zoomed in order to better visualize the differences.*

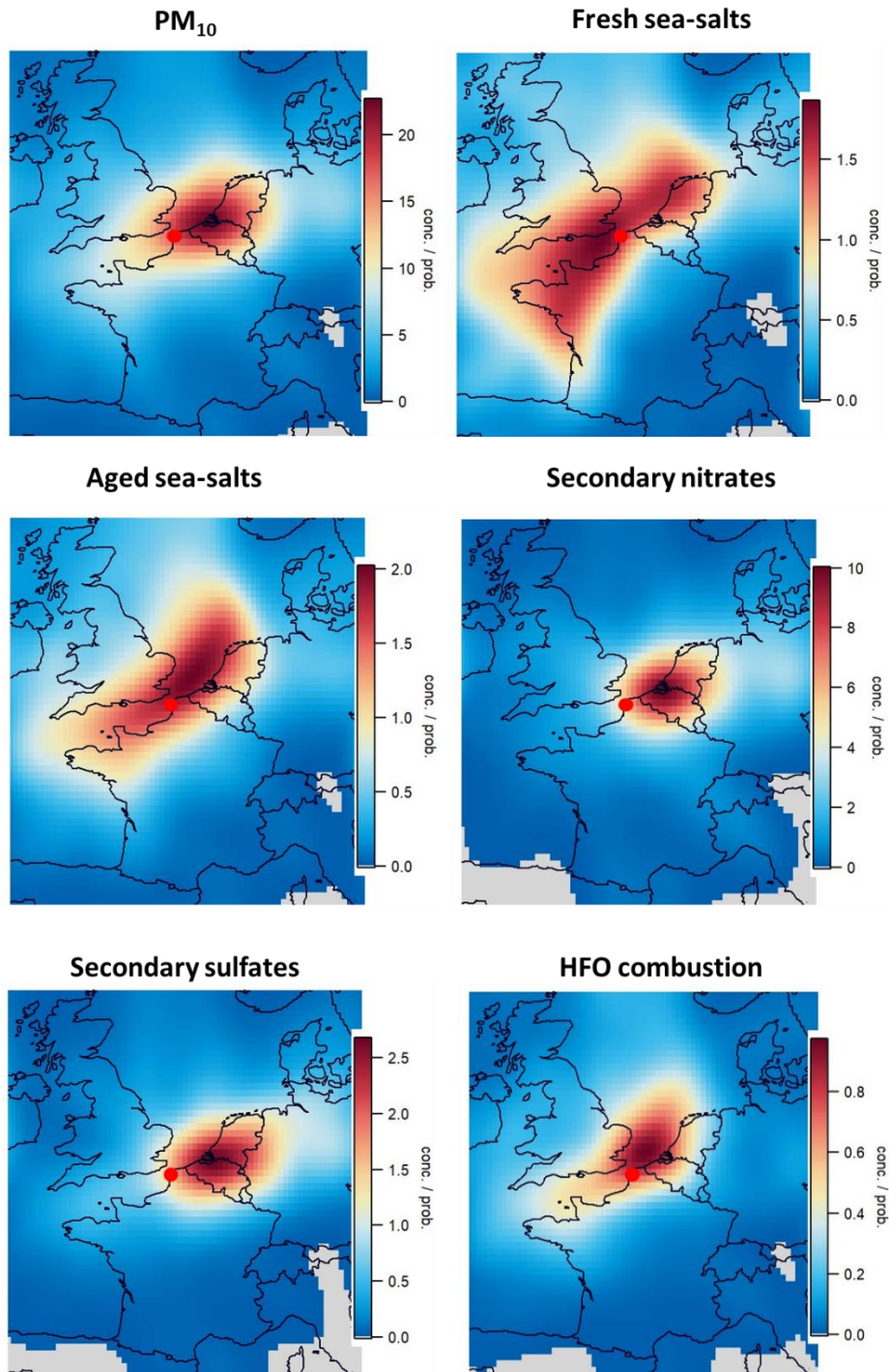


Figure 1: CWT results for PM<sub>10</sub> and some NMF factors (fresh sea-salts, aged sea-salts, secondary nitrate, secondary sulfate, and HFO combustion). Red colors highlight potential emission zones. Contribution scales are in  $\mu\text{g}/\text{m}^3$ .

## References:

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