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Dear Editor-in-Chief,

We would like to thank you for accepting our manuscript for publication in Atmospheric Chemistry and Physics. Hereafter, we have replied to the different minor comments. Amendments in the manuscript were done in track changes mode for ease of review.

 Referee 1 comment 10 "...I do not understand why the CBPF of sea salt and aged sea salt are so different...": Please add clarifying comments to the text along the lines of that supplied in the response document.

We have added in the manuscript additional details that were found in the response to the referee's comment (Lines 340, 341-344, 351-354 in the revised manuscript).

## The paragraph now reads:

"The main reason for these differences might be the meteorological conditions as well as the seasonality. Indeed, the CBPF representation of fresh sea-salts clearly evidenced that the maximum concentrations were observed for winds blowing from the southwest (SW) and northeast (NE) wind sectors and for medium to high wind speeds (> 10 m/s). This is mainly due to the position of the sampling site, strongly subjected to fresh marine influence from sectors 210° to 50° via the North, corresponding to the English Channel and the North Sea, respectively (Error! Reference source not found.). These wind directions were predominant during all months of the year except for the summer season which could explain the higher concentrations of ged sea-salts were obtained when the wind blew from the northeast wind sector with wind speeds higher than 10 m/s (Error! Reference source not found.). This might be explained by the reaction of the fresh sea-salts with SO<sub>2</sub> and NO<sub>2</sub> that also show the highest concentrations in the northeast wind sector (**Figure S7**) to yield aged sea-salts in the Strait of Dover and the North Sea area (Error! Reference

source not found.). This phenomenon occurs 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.

2) Referee 2 comment 2 "Please comment on the choice of sampling PM2.5...": Please add clarifying comments to the text along the lines of that supplied in the response document.

The explanation was clearly added in the introduction of the manuscript (Lines 69-74):

"Several years before 2013, the EU has issued the directive 2008/50/EC that limits daily  $PM_{10}$  concentrations to 50 µg/m<sup>3</sup> with a maximum of 35 days of exceedance authorized per year. The directive is binding and forces countries that do not comply with it to seek solutions for improvement. In this period, several regions in France were concerned by high number of exceedances, especially in and around Paris capital as well as in the North, the East, and the South-East parts of the country (EEA, 2014). This is why it was important to focus on the  $PM_{10}$  fraction in order to understand the reasons behind these exceedances on a regional and national scale. "

 Referee 2 comment 6 "The yearly-mean PM10 concentrations...": Please clarify in the text that the average of the entire year is 22.8 ug/m3 and that this supports the 122 samples is representative of the entire year on average.

The information regarding the yearly average concentration of  $PM_{10}$  and the concentration for the 122 samples was added in the manuscript (Lines 182-185):

"The yearly mean concentration of  $PM_{10}$  for the 362 samples was 22.8  $\mu g/m^3$  in 2013. As for the 122 samples corresponding to a sampling of 1 day over 3, the mean  $PM_{10}$  is 24.3  $\mu g/m^3$ . The closeness of the two values is indicative that the sample selection is representative of the whole year."

4) Referee 2 comment 8 "The description of the model setup...": On line 169 (track changes version) it states that 122 samples were used as the input data, however in the response it suggests additional samples from 2013 and 2014 were also included. Please clarify the input data. If the data from the exceedance days in 2013 was included in the input data, then a discussion on if including exceedance days alters the model outputs is warranted. One can imagine that having a biased sampling (by including extra exceedance days, but not non-exceedance days) would alter the results. The 2014 data would also need explanation if that was included as input.

We understand the editor's comment regarding the input data for the NMF model. We would like to clarify that in the response as well as in the manuscript.

The sampling campaign at CGN covered the year 2013 where 362 samples were collected and a part of the year 2014 (until 17/04/2014) where 107 samples were collected. Not all of these samples were chemically characterized and/or used for the source apportionment.

The choice was done as follows:

- 1 day over 3 samples making a total of 122 samples in 2013 and 36 samples in 2014.
- 51 samples in 2013 and 13 samples in 2014 corresponding to wind directions underrepresented by the selection done by one day over three (non-exceedance days).
- 11 samples with exceedances in PM<sub>10</sub> concentrations for 2013 and 9 for 2014.

This total of 242 samples was considered as the input data of the model. Besides the 1 day over 3 samples (in total 158), samples corresponding to non-exceedance days (64 samples) and to exceedance days (20 samples) were added, removing by that the bias sampling.

Concerning the manuscript, the authors have decided to only present the results of the sampling campaign of 2013 because it covers a whole year and by choosing the 122 samples corresponding to the 1 day over three samples. The contribution of the sources used in Figures 3,4, and 5 as well as in Figure S6 and Table S2 were extracted from the output data of the model and it only represents the 122 samples (1 day over three in 2013). The extra exceedance and non-exceedance days as well as the data of 2014 were not considered.

The information was clearly added in the manuscript (Section 2.2; Lines 110-116):

"Over the sampling period, 362 samples have been collected in 2013 and 107 in 2014. Field blanks (two per month) were also considered by placing a blank filter in sampling conditions but without pumping. Additionally, meteorological data (temperature, wind speed and direction) were recorded on site using the WMT 52 ultrasonic wind sensor (Vaisala Windcap) coupled to the DIGITEL® DA80.

Among these samples, 158 (122 in 2013 and 36 in 2014) corresponding to a one day over three sampling, 64 (51 in 2013 and 13 in 2014) corresponding to wind directions under-represented by the selection done by one day over three (non-exceedance days), and 20 (11 in 2013 and 9 in 2014) representing exceedance days were chosen for the chemical analysis and the source apportionment. In the results of this study, we will only be presenting the results of the 122 samples representing a sampling of one day over three in 2013."

Additionally, the bootstrap analysis would benefit from a few additional sentences of explanation (SI is ok) and the results of Table S1 require a few sentences explanation of the meaning of the table (where is the number 100 coming from?).

A description of the bootstrap analysis method as well as details regarding the number 100 was added in the supplementary information:

"The Bootstrap analysis show the effects from random errors and include partially the effects of rotational ambiguity. It is used to find if there is a small set of observations that can largely influence the solution. This method creates sets of bootstrap data constructed by randomly selecting blocks of observations from the initial dataset. The size of the block was taken as 5 samples for this study. The solution was bootstrapped 100 times to ensure the robustness of the results. Mapping over 80% of the factors indicates that the bootstrap uncertainties can be interpreted, and the number of factors may be appropriate. "

*The interpretation of the results of Table S1 was added in the manuscript as follows (Lines 236-238):* 

"The best results were obtained for the 9 factors solution. The stability of the results was examined via bootstrap analysis and the different source profiles satisfied the validation criterion (**Table** 

*S1*) with a mapping percentage of at least 99% (higher than 80%) showing the robustness of the obtained solution".

5) Referee 2 #9 "Is the seawater composition...": Please add a sentence to the text saying that the ratios were also valid for other ionic species.

The validity of the other concentration ratios was added into the manuscript (Lines 250-251):

"The concentration ratios between the other ionic species were also valid for sea water composition:  $K^+/Na^+=0.03$ ,  $Ca^{2+}/Na^+=0.03$ ,  $SO_4^{2-}/Na^+=0.15$ , and  $Mg^{2+}/Na^+=0.11$ ."

6) Referee 2 #10 "Is the cations/anions ratio balanced in this factor?": Please clarify in the *text that it has a ratio of 1.14.* 

The information was added in the manuscript as follows (Line 255):

The ionic balance is respected in this factor with a cations/anions ratio of 1.14.

Please ensure that the SI material is cited in order. Currently Fig. S3 is cited before Fig. S2.

We have carefully checked the manuscript and changed the order of the figures in the supplementary in order to cite them in order.

8) Fig. S6: Is the 75th percentile of the January value for secondary sulfates off-scale? It is hard to tell with the current axis.

We would like to thank the editor for the comment. The axis for the figure of secondary sulfates (Fig S6) was modified and added in the SI.

## References:

EEA: Air quality in Europe 2014 report, EEA report, Publications Office of the European Union, Luxembourg2014.

Ledoux, F., Roche, C., Cazier, F., Beaugard, C., and Courcot, D.: Influence of ship emissions on NO<sub>x</sub>, SO<sub>2</sub>, O<sub>3</sub> and PM concentrations in a North-Sea harbor in France, J. Environ. Sci., 71, 56-66, <u>https://doi.org/10.1016/j.jes.2018.03.030</u>, 2018.