In the following, my comments are provided in red color, below each comment and author's response.

1. Our reviewers have been insisting on trying to delineate the "trends that were caused by internal annual variabilities of weather or climate conditions". However, it was assumed already in the initial version that the search for all the causes of the temporal evolutions of source contributions was not in the scope of our work, and it is still not in the direction that we want for this paper. This would be a totally different work, that would require gathering many other data (on local and large-scale meteorology, on emission inventories, on parameters that are influencing atmospheric chemistry for the formation of secondary chemical species, …) on the long term, and it is not our purpose. One aspect of our paper is focused on showing that the data set allows to identify trends in the evolution of the sources contributions of PM for this rural site, and that these trends are different for one source to another. All of this is already an innovative step compared to most of the (few) similar papers in the literature that discuss of evolutions of the concentrations of some chemical species for similar sites, or evolutions of sources with a set of tracers that is much less elaborated than ours. We believe that this innovative result by itself justify the publication, as a step ahead of other previous work on long-term trends of PM, particularly for rural areas.
However, to address reviewers' comments, we already added a statement emphasizing that the role of meteorology cannot be ruled out. We further included in this version the sentence "In most cases, there is a complex interplay between PM and meteorological conditions that further exacerbate PM mass concentration (Chen et al., 2020).", to strengthen this statement.
Regarding the added sentence, it is not clear what the authors mean by "further exacerbate PM mass concentration" and specifically how this is connected to the revealed (decreasing) trends.

2. As said, the novelty of the paper is the observation of the trends of the PM sources (apportioned using an enhanced PMF methodology) but also that of OP (apportioned using MLR). This allowed the unravelling of the decreasing trend in terms of source contributions by the STL model. The STL deconvolution was applied on all the identified sources, and it clearly shows that the traffic source has the highest tendency (see Table 1 in the manuscript) with a decreasing trend. The other major sources of PM10 (such as biomass burning, mineral dust, nitrate-, and sulphate-rich sources) do not have as much decreasing tendency as the traffic source. This is probably an indication that some of the processes included in "the internal annual variabilities of weather or climate conditions" are not leading factors in the temporal evolutions that can be seen (or not), since they would probably affect PM from different sources in the same way. Again, this aspect is not discussed further, since it is not our purpose to delineate these complex processes.
I would suggest to include part of the response in the revision.

3. However, thanks to the reviewers' comments, we added evidence in the second version that the decreasing traffic contributions is also in good agreement with the decreasing BC emissions from emissions inventory for France. Hence, we stand by one of our key take-aways stating that "While local or regional changes in meteorology may be a factor in the evolution of the concentrations observed, this

is unlikely to be the dominant one in the evolution of the concentrations of chemical species related to traffic emissions, in light of the strong correlation observed with the national emissions inventory in France.".
Ok.

**Anonymous Referee #1 (nominated 18 Mar 2022, accepted 25 Mar 2022, report 02 Apr 2022, Report #2):**

 Lines 34-35: This sentence is not clear: rephrase.

RESPONSE: The sentence in Line 34 to 35 is further improved into:
"Particulate matter (PM) pollution causes various environmental concerns affecting public health and climate."
Still not clear, in particular related with the climate effect of PM (since the dominant effect should be a cooling so compensating for global warming.

 Line 39: and what about aerosol transport apart from its formation?

RESPONSE: The sentence was improved and now reads as:
"Further work has also been carried out in more specific areas to understand particular processes of aerosol formation and transport, as well as specific sources such as in the boreal forest (Yan et al., 2016), polar environments (Barrie and Hoff, 1985; Moroni et al., 2016), high altitude (Rinaldi et al., 2015), or marine sites (Scerri et al., 2016)."
Ok.

 Line 45: well, it is quite restricting to say that the understanding of such processes is only related with the "elaboration" (perhaps also not the most appropriate term) of chemical transport model.

"Studies at such sites enable the understanding of large-scale and mesoscale processes (Anenberg et al., 2010; Mues et al., 2013; Konovalov et al., 2009), which is necessary to elaborate chemical transport models."
RESPONSE: The sentence was improved and now reads as:
"Studies at such sites provide more understanding of large-scale and mesoscale processes (Anenberg et al., 2010; Mues et al., 2013; Konovalov et al., 2009), which can be useful in the development and validation of chemical transport models."
Ok.

 Lines 47-48: This is a general characteristic of the long term time series, and not just of the ones collected at background sites.

RESPONSE: The phrase "in background sites" was removed.

Ok.

 Lines 71-73: This detail is not needed at this point.

“The PMF methodology includes a unique validation with comparison of the chemical profiles of the factors with those obtained in many other studies in France.”
RESPONSE: This improvement (see sentence above) is a response to another referee’s comment (Referee #2) that aims to emphasize the usefulness of this paper. In any case, we believe that it is good to mention that our work indeed included this amount of effort that enabled us to do a comparison of chemical profiles obtained in many different studies in France.
Ok.

This is a feature, allowing check for consistency, that is extremely rare in the literature. In fact, in Figure 5, we have included this discussion through the PD-SID metric.
Ok.

 Lines 69-77: The scopes are very specific and citing already methodologies which happen to be still rather obscure to the reader. Please try to generalize the objectives leaving the details of the methodology for later on in the text.

"The objectives of this work are, first, to achieve for the very first time a study of the main sources of PM in a rural environment in Europe, using a long-term database including several specific organic tracers in the carbonaceous fraction. The PMF methodology includes a unique validation with comparison of the chemical profiles of the factors with those obtained in many other studies in France. The second objective is to quantify the temporal evolution of the contributions of these sources over the period of the study, particularly focusing for the first time on the vehicular emission that have already been shown to decrease in urban environments in Europe during the last decades. Finally, another major objective is to perform the deconvolution of the contribution of the PM sources to the OP measured with AA and DDT assays, and to determine the most important sources for the oxidizing capabilities of PM influencing human health in such an environment."
RESPONSE: This paragraph has been improved and now reads as:
"The understanding of trends of PM sources are essential to evaluate the effects of mitigation policies on air pollution levels. A reference background site offers a good opportunity to gauge the broad effects of certain improvements in the transportation fleet and other regulations aimed at reducing vehicular emissions in large cities. Thus, in this study, an extensive dataset of PM over a 9-year period ($n$ = 434), obtained from a French national background site, was investigated to: (1) provide insights on the long-term trends of PM sources and other emerging health-based metrics of PM exposure, such as OP of PM, (2) quantify the temporal evolution of the contributions of these sources, particularly focusing on vehicular emissions that have already been shown to decrease in urban environments in Europe during the last decades."
Ok.

 Line 140-141: Absolutely not clear how you reconstructed PM10.

RESPONSE: This was discussed in section 3.1, specifically mentioning that the reconstructed mass of PM10 in the OPE site was calculated following Eq. S1. In the SI are the equations used to reconstruct PM mass.
You can then provide a reference to the Supplementary Material for such description.

 Lines 142-144: This detail is not useful here, as the reader does not know anything of this comparison.

“A total of 299 out of 434 (69%) TEOM measurements were paired with reconstructed PM10 data, due to many interruptions in the TEOM functioning, in order to evaluate the semi volatile mass missing in the mass reconstruction with filter chemistry.”
RESPONSE: This sentence (see sentence above) was added as an improvement following the comment of Referee #1, which suggested that the unknown portion of PM be added in Figure 2. In this sentence, the authors wanted to elaborate on how this was done.
Ok.

 Lines 206-207: There are other reasons to increase the uncertainties of some variables (indicating them as weak) or excluding them from the analysis; I assume that this has been considered. also, have other sources of uncertainty (e.g., flow rate) taken into account?

RESPONSE: Section 2.4.2 presented our criteria for a valid solution, which also mentioned that we followed the recommendations of the European guide on air pollution source apportionment with receptor models (Belis, 2019). This was the guideline that was followed, it presents in detail how to perform receptor modelling.
Each sample included was initially checked for its consistent and valid flow rate. We did not include samples that have questionable flow rates during sampling.
The guide does not have a specific value for the added extra uncertainty (to the whole dataset) and in any case it provides general guidelines on how the uncertainty can be determined but many details (e.g., evaluation of the S/N ratio, calculation of the uncertainty for missing data and for data below detection limits, etc.) are absolutely not fixed. I would suggest to include more details on this on the revised version of the manuscript.

 Line 211: How did you take into account the weighted residuals distribution? I mean, could you explain how you analysed it, such as you did with the Q/Qexp ratio?

RESPONSE: This information can be found in the PMF user guide 5.0 and the European guide on air pollution source apportionment with receptor models (Belis, 2019; mentioned in section 2.4.2 in the manuscript). In brief, the residual analysis is based on the uncertainty-scaled residuals. A specie is considered well-modelled, when all residuals are between +3 and -3 and are normally distributed. Species with residuals beyond +3 and -3 was evaluated in terms of their observed vs. predicted scatterplot and time-series analysis.
Ok, but perhaps a detail could be added in the revised version of the manuscript.

 Lines 265-284: Which software or code did you use to apply this analysis?

RESPONSE: It has been mentioned in the manuscript (section 2.6) that it was implemented in Python using the *statsmodels* module.
Ok.

 Lines 266-268: I cannot understand this: meteorology does not have only a seasonal signal, and also how can the interannual variation in the seasonal signal be connected with the effect of meteorology? If you have references, please provide them, because this justification is not convincing.

RESPONSE: Please refer to the general comments.
The response in the general comments does not address my specific comment above.

 Line 298: To be true, I can observe also a reduction in NO3- and NH4, which should be among the main chemical species. So I still cannot understand this.

RESPONSE: $NO_3^-$ and $NH_4^+$ are in fact among the main chemical species in Figure 2. Our point in this sentence was that the changes are not drastically changing through the years. In this paragraph, we were also discussing the yearly average volatile mass (i.e., unaccounted by chemical analysis).
I still see this as confusing, and I would suggest to make the sentence clearer in this regard.

 Lines 299-301: Can you explain at least tentatively the reasons of such differences?

RESPONSE: Our unaccounted portion is well within range generally found in a rural environment and we have added a reference that supports this. As discussed in many papers in the literature, the differences are generally attributed to all semi-volatile chemical species included in the PM (water vapor, organics, ammonium nitrate, …).
Detail on this should be added.

 Line 378: A part of sulphates has a marine origin.

"The **aged sea salt** factor is characterised by high loadings of $Na^+$ and $Mg^{2+}$, with a certain amount of species originating from potentially anthropogenic sources such as nitrates (6% of $NO_3^-$ mass) and sulphates (19% of $SO_4^{2-}$ mass) that can be attributed to mixing and transformation processes in the atmosphere."
RESPONSE: There is no statement in the manuscript that argues against "sulphates has marine origin". Indeed, the reviewer can observe that there is a fraction of sulfate included in the fresh sea -salt chemical profile (Figure S5), that largely increases in aged sea-salt (Figure S6).

 Lines 384-386: And what about the sulphates to Na+ ratio? Did you observe if there is a particular wind direction for this factor? Or reasons to suspect collinearity? Or any other investigation on this factor which could be also a mixed source?

RESPONSE: We made it clear that we did not analyse meteorological data. Please refer to the general comments. Further, it is really difficult with the PMF (and nearly never discussed in papers with PMF results) to distinguish if the chemical profile of a factor includes some species (that are not fully known to be associated in a given source) because of co-linearity of sources or because the species are indeed

internally mixed in the PM because of interactions / modification during transport. In our case, with a multi-year data base, it seems unlikely that the presence of some fraction of OC in the MSA rich factor is present just because of collinearity or mix with another source, that would need to be maintained for the overall period.
The response is incomplete since the absence of meteorological data does not justify the absence of the analysis of the sulphates (or other species) to Na+ ratio.

 Lines 388-390: And what about the ratio of Cl- to Na+? Is there chlorine depletion?

REPONSE: Elaborating about the chlorine depletion does not add any useful information in terms of identifying factors resolved by PMF. However, the reviewer can observe that the chlorine that is present in the fresh sea-salt chemical profile (Figure S5) but is not apparent in the aged sea-salt profile (Figure S6), indicating chlorine depletion with ageing of the sea salt emissions.
Ok.

 Lines 413-416: And what about your study?

RESPONSE: In this paragraph, we talked about the description of primary biogenic factor. We talked about the characteristics of primary biogenic sources as reported in Samake et al. (2019), a paper also published by our group. This is a strong paper supporting the characteristics we have equally found in our study in the OPE site.
Ok.

 Lines 417-424: Any particular temporal pattern for this factor?

RESPONSE: Yes, it has temporal pattern and this is shown in Figure S1. In section 3.3, we have also mentioned that it is a major contributor during winter season.
 Line 432-434: Also here, any particular temporal pattern?

RESPONSE: There is a mild temporal pattern, please refer to Figure S4.
Ok.

 Lines 458-530: Apart from the analysis of trends, could you explain more how to interpret the results of the STL analysis for example in terms of different importance of the three signal components?

RESPONSE: Thank you for the suggestion, we have improved a part of section 2.6 that now reads as: "The STL (Season-trend deconvolution using locally estimated scatterplot smoothing) model is a versatile and robust statistical method allowing the decomposition of a time-series dataset into three components including trend, seasonality, and residual. The trend provides a general direction of the over-all data; the seasonality is a repeating pattern that recur over a fixed period of time; finally, residual is the random fluctuation or unpredictable change in the dataset."

This addition does not completely address my comment, since the information provided are generic for the STL analysis and does not refer explicitly to the results obtained here using this methodology.

 Figure 6: The Figure has poor resolution. Also, there is no unit of measurement on the y-axis.

**Figure 1: The Season-trend (STL) deconvolution of contributions in µg $m^{-3}$ from the traffic factor to $PM_{10}$ from year 2012 to 2020.**

RESPONSE: It is unclear if referee meant poor image resolution (which will be improved in the final paper) or he meant the resolution of the x-axis interval (which is essentially monthly resolution, fit for the purpose of the trend evaluation). The unit is given in the figure caption.

I was referring to the image resolution and to units of the y-axis.

 Line 579: Missing reference to a Figure.

RESPONSE: This has been corrected.

 Lines 660-666: This sentence is still not clear, and has to be revised.

RESPONSE: The paragraph has been improved and now reads as:

“There was a change in sampling duration between the collection performed in year 2012 to 2016 (7-day sampling) and 2016 to 2020 (24-hour sampling). A 7-day filter sample includes both weekdays and weekends, whereas a 24-hour sample will either be a weekday or weekend, depending on the sampling interval. This implies that the weekly collected samples may contain features that are not fully captured in a daily sample. However, since the OPE site is quite distant from direct emissions, the expected difference in the weekday and weekend levels should be relatively small. Further, PMF source apportionment were conducted separately on the two periods (i.e., 7-day samples versus 24-hour samples), leading to very similar results for the chemical profiles and source contributions, justifying the coupled analysis.”

Ok.

 Lines 702-703: Do you mean the improvements in the technology?

RESPONSE: Yes, technology in various aspects.
 Lines 704-705: Not clear what you mean by “persistent changes”. Revise.

RESPONSE: This sentence was improved and now reads as:
“However, persistent changes in meteorological conditions influencing the transport of air masses to OPE or formation of PM during this transport cannot be totally ruled out.”
 Code and data availability: still not in line with the policy of this journal.

RESPONSE: Our declaration is that these “could be made available upon request by contacting the corresponding author” in order to be in line with university and research institution policies and legal terms with our funding groups.
 Table S2 and text: the “nitrate-rich” and “sulfate-rich” factors should be “secondary nitrate” and “secondary sulfates”? also, the “fresh sea salt” and “aged sea salt” share much of the fingerprint, so there is no clear evidence of why they should be separated in two factors, at least from this table.

RESPONSE: The authors deem that the two factors, nitrate-rich and sulphate-rich, are named accurately (as discussed in the description of factors in section 3.3), there is no added value in changing the names as suggested by the referee. Those are terms widely accepted and recognized by all research groups doing PMF studies. Sulfate- and nitrate-rich names were coined because these factors do not only contain sulfate or nitrate secondary components but also aggregate some other chemicals species. As for fresh and aged sea salt, one of the major differences is the depletion of chlorine (as mentioned above), but also the aggregation of other components (like sulfate, also mentioned above).
 Figure S1 and text: is the temporal variation of biomass burning in agreement with your expectations? Does it increase in winter (it seems so) and with wildfires?

RESPONSE: We already mentioned in the main text that the Biomass burning factor is a major contributor during winter. There are not many reported wildfires in proximity to the sampling site, if any it would be relatively more common in South of France (~700 km away). Agricultural fires (which are not so common in France) could be possible to explain some low level contribution out of the winter season, where the emissions are totally dominated by domestic heating with wood burning.
 Figure S4: About the mineral dust factor (possibly better named “resuspension”, as mineral dust would be just the transport of dust from the desert), I would expect an increase in the summer season with less precipitation, and with Saharan dust transports. Is this the case?

RESPONSE: It was also mentioned in the main text that there could be an influence from re-suspended road dust. Yes, there is somehow a seasonal pattern, but not as clear as the pattern seen in the Biomass burning factor. We can assume the influence of precipitation and Saharan

dust transport; this has been extensively published in the past already. The authors deem it is unnecessary to provide meteorological data just so we can support this phenomenon.

 Figure S1-S9: In most cases the BS and DISP bars (in the legend, but are actually lines?) cannot be seen. Also, apart from the temporal pattern, could you investigate the effect of meteorology at least in some of these factors?

RESPONSE: The BS and DISP bars are very close to the contributions estimated in the final constrained PMF solution (100% mapping, discussed in section 3.2). Please refer to the general comments for our response about meteorological data.