Answers to referee # 1

General comment

The paper reports a study of maritime contributions to PM_{10} 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 will be done once the final version uploaded.

Specific comments

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

The « etc. » term will be removed from lines 42, 44, and 54.

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. 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_{10} concentrations were deduced from the real-time particle size distribution measured by a Electrical Low Pressure Impactor. The temporal resolution for PM_{10} was 1 min and 15 min for gas concentrations.

In order to estimate the impact of in-port ships on PM_{10} 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_2, NO, and NO_2 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_{10} concentration by an average of 28.9 $\mu g/m^3$ with a maximum increase of 78.9 $\mu g/m^3$ for a 1-minute time interval (the value presented in line 67 of the manuscript).

The information will be modified in the manuscript as follows (Line 67 in the original manuscript):" Punctually, ship emissions led to a marked increase of PM_{10} mass of +78.9 $\mu g/m^3$ 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 SO2.

The authors agree with the referee. The information was 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_2 emissions but also primary PM emissions (Shen and Li, 2020; Zetterdahl et al., 2016). However, no regulations for PM components neither in the sea nor at ports were issued. Hence, this study aims at highlighting the contribution of the natural and anthropogenic marine emissions in the degradation of the air quality in a coastal background 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) will be modified as follows:

"Moreover, PM_{10} were collected using an automated high-volume sampler (DA80, DIGITEL®, Switzerland) operating at 30 m^3/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_{10} at Cape Gris-Nez which is a rural background site with no primary emission sources nearby and other urban and industrial sites in the Northern France region and during the same period (one year). PM_{10} 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_{10} concentration. This was also highlighted in the CWT representations showing that more than 80% of PM_{10} 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, NO_3^-, and 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 (Becagli et al., 2012; Pandolfi et al., 2011). 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 (Streibel et al., 2017; Zhang et al., 2016)." ***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 pint is that it was analysed here only the contribution to primary PM10 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_{10} including primary species as well as secondary ones, namely the secondary inorganic aerosols (NO_3^-, SO_4^{2-}, and NH_4^+). The concentration levels of these secondary species characterized in PM_{10} 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_{10} 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_3^- and SO_4^{2-}). By that, we cannot say that the HFO combustion source corresponds only to primary PM_{10} emissions.

However, the authors wanted to highlight the idea that the transformation reaction of SO_2 emitted from the shipping emissions to SO_4^{2-} is probably not at the equilibrium yet and maybe more amounts of SO_4^{2-} were transformed which may lead to higher contribution of shipping emissions to PM_{10}. That is why the authors mentioned the underestimation of the shipping emissions contribution to primary PM_{10}.

The sentence in the original manuscript was modified as follows:

"However, considerable amounts of SO_2 and NO_x 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_2, and 15% of NO_2 average concentrations.

However, the transformation reaction of SO_2 emitted from the shipping emissions to SO_4^{2-} is probably not at the equilibrium yet and maybe a higher amount of SO_4^{2-} could be formed from SO_2 which may lead to a probable underestimation of shipping emissions to primary PM_{10}."

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_2 and NO_2 were continuously measured using an AF21M SO_2 analyzer (Environnement SA, France) and an AC32M NO-NO_2-NO_X 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). 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_2 and NO_2 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.

References:

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