Anonymous Referee #1

General comments

The essential information for major sources as well as evaluation and planning of control measures in Hong Kong, China, was proposed based on 10 years of long-term monitoring data of $PM_{2.5}$. Overall, it is an interesting study. The test and analysis procedures are reliable and the amount of data are sufficient. The manuscript can be accepted with the following revisions:

Response: Thank you for the positive comments. Our response to the comments is given in the following. The response text is marked in blue. References cited in this response document are placed at the end.

Comments

1) *Chlorine loss of sea salt aerosol was always found in the sampling area, however, particulate chloride were not found in the chemical analysis and long term variation analysis. It was also suggested to analyze the trend variation of Criteria gaseous pollutant data.*

Response: As the reviewer has pointed out that chlorine loss of sea salt aerosol was common and was also observed in our sampling location. The chloride concentrations were at very low levels at our study site over the years (e.g., as low as 0.09 $\mu g/m^3$ in 2011). The chloride data were lumped into "salt", which is the sum of Na^+ and Cl^- and shown in the 10-year time series plots of monthly averages and annual averages of $PM_{2.5}$ chemical composition in Figure 2. Shown in Figure 3, "salt" made up a very minor part (<4% in all years) of $PM_{2.5}$. We now also add the individual temporal variation plots of Na^+ and Cl^- in Figure S3.

Due to the low concentration levels, the Cl^- data were associated with large measurement uncertainties. One more complicating factor was that nylon filter substrates were used during 2008-2010 while quartz filter substrates were used during 2011-2017 for IC analysis of the ionic constituents. It is known that nylon filters had higher efficiency in retaining chloride while quartz filters suffer negative sampling artifacts due to volatilization of chloride following acidification to HCl by co-existing sulfate (e.g., HSO_4^-) (Tsai and Perng, 1998; Tsai et al., 2005). The known systematic bias for Cl^- measurements between the two sampling media used during the ten-year period, together with the low concentration levels, does not adequately long-term variation analysis.

The following text is added to comment on chloride data, in association with the "salt" component:

> Line 243 – 244
>
> "Salt", consisting of Na^+ and Cl^-, was a very minor part of $PM_{2.5}$, accounting for less than <4% in all years."

Regarding the trend variation of criteria gaseous pollutants, we'd like to clarify that they are already illustrated in Figure 4 and summarized in Table 2. Extended discussion about the influence effect of gaseous pollutant variation on the secondary formed products (sulfate and nitrate) was provided in section 3.4. Thus, we did not expand the section other than adding a sentence to convey the use of gaseous pollutants in this study.

> Line 152 – 153
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> "The temporal variations of gaseous pollutants serve as additional data valuable for exploring the effects of changes in precursor gases on the secondary formed $PM_{2.5}$ constituents (e.g., sulfate and/or nitrate)."

2) *Figure 1: The resolution of Figure 1 needs to be improved.*

Response: Thank you for careful checking on the resolution of figures. A higher resolution version of Figure 1 is created and incorporated in the manuscript.

3) *Lines 100-113: The authors collected blank samples as a background correction? and how were filter samples stored after collection? More information about sampling and storage should be presented in Ms.*

Response: Field blanks were collected at a frequency of 10% of a sampler's routine operating frequency. They were used to monitor contamination throughout the process from sampling to analysis and for background correction, if necessary. The field blank filters were stored and analyzed following the same procedures as those of $PM_{2.5}$ sample filters for quality assurance (QA) purposes.

The following text is added to provide more details about the sampling and storage procedures in our study:

> Line 112 – 118
>
> "After the collections, the 47-mm Teflon filters were stored in Petri dishes and the 47-mm quartz filters were stored in the dishes lined with aluminum foil. The dishes were sealed with parafilm. While the 20x25 cm quartz filters were folded in half and stored in aluminum foil. All the filters were packed in a thermal bag for transportation to the laboratory and refrigerated under 4°C before chemical analysis. Field blanks were collected at a frequency of 10% of a sampler's routine operating frequency. They were used to monitor contamination throughout the process from sampling to analysis and for background correction. The field blank filters were stored and analyzed following the same procedures as those of $PM_{2.5}$ sample filters for quality assurance (QA) purposes."

4) *Double "in" were found in line 178. Moreover, please recheck the English by a native Editor*

Response: Thank you for careful reading. Line 178 is revised.

5) *Line 136: Table 1 should be revised to a trilinear table.*

Response: We are unsure what is "trilinear table". We now revise the column arrangement of this table to make it more easily comprehended.

6) *Line 230: "For winter PM_5, the relative importance of OM increased (up from 31% in 2008 to 40% in 2017)… as well as EC (down from 7.8% to 5.3%)." I can't find any value of 31% in 2008 to 40% in 2017 in the wintertime figure (Figure 3).*

Response: We double checked the values used for the figures and they were calculated correctly, which means that the number in text are somehow copied wrongly from the results. All the numerical values in the text are now re-examined and any similar issue was now corrected.

The following text is now revised with correct percentage values:

> Line 238 – 239:
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> "For winter $PM_{2.5}$, the relative importance of OM increased (up from 35% in 2008 to 39% in 2017) while the relative abundance of nitrate decreased (down from 9.1% to 6.6%), as well as EC (down from 6.1% to 4.4%)."

7) *Line 231: Valid numbers should be uniform.*

Response: The numbers in text are now rounded to 2 significant figures. (Please refer to the revised sentence in response#6)

8) *Line 233: Do authors have any ideas about the decrease percentage of EC in summer was about 4 times higher than that in winter?*

Response: The changes in percentage (i.e. the species contribution to $PM_{2.5}$) depend on the concentration variations of both the species and the $PM_{2.5}$ mass. As shown in the table below, EC levels had little seasonal variations and the reductions over the 10-year period was profound (by 59.5% in winter and by 62.7% in summer). On the other hand, the $PM_{2.5}$ levels decreased to a much more significant extent in winter (by 43.8%) than in summer (by 26.8%). Therefore, the contribution of EC to $PM_{2.5}$ was more notable in summer (by 49.1%) since the reduction in EC level is more significant than that in the PM mass.

EC, µg/m^3	2008	2017	%Change
Winter	3.06	1.24	-59.5%
Summer	3.06	1.14	-62.7%
$PM_{2.5}$, µg/m^3	2008	2017	%Change
Winter	50.2	28.2	-43.8%
Summer	18.3	13.4	-26.8%
EC/$PM_{2.5}$	2008	2017	%Change
Winter	6.1%	4.4%	-27.9%
Summer	16.7%	8.5%	-49.1%

9) *Figure 3: The percentage values in the chart use an art font that is unclear and needs to be modified.*

Response: Thank you for the comment. The art styles are now removed for easier reading to audiences in the figure. Valid rounding in 2 significant figures is also updated in the figure.

10) *Line 392: "The concentration levels of shipping emission tracers (V and Ni)… respectively", Is this result from Table 2?*

Response: Yes, it is the result from Table 2. We now quote the table number in text for better tracking of the results.

The following text is now revised:

Line 407 – 408:

"The concentration levels of shipping emission tracers (V and Ni) were reduced by 34% and 36% by mass concentration and at a rate of -0.60 and -0.29 ng m^{-3} yr^{-1} (Table 2), respectively."

11) *Line 418: "K+ more representative of burning crop residues high in K+ content vs. levoglucosan representative of burning of cellulose, thus all types of vegetative biomass including hill fires." Grammar errors. The sentence components are unclear and there is no cause-and-effect relationship in this sentence.*

Response: Thanks for the comment. Here we would like to show the examples where K^+ and levoglucosan can represent two different categories of biomass burning. The sentence is revised as below for improved clarity.

Line 433 – 435:

"Specifically, K^+ is a better marker for emissions from burning crop residues, which are typically enriched in K^+, while levoglucosan, a thermal pyrolysis product of cellulose, is commonly found in burning of all types of vegetative biomass including hill fires."

12) *Line 444: "The two elements are highly correlated, reflecting their common material sources and spatial origins", are the correlation values in the literature or your results?*

Response: The correlation values was computed based on our results with the correlation matrix of the starting and ending years as shown in Figure 5. The *R*-value of correlation results is added in this sentence as additional information. As characteristic elements of crustal materials, the high correlation of these two elements (Al and Si) are commonly found in ambient PM samples.

The following text is now revised:

Line 461 – 462:

"The two elements are highly correlated (*R: 0.54 in 205 and* 0.87-0.97 in the other years), reflecting their common material sources and spatial origins."

13) Line 499: Valid numbers in Table 4 should be uniform.

Response: Table 4 summarizes the coefficients resolved from the multiple linear regression. We'd like to clarify that the numbers are uniform in having the same number of significant figures (i.e., 2). Note that the numbers should not round up to the same decimal place, as the concentrations for different species span a few orders of magnitude.

References:

Tsai, C.J. and Perng, S.N., 1998. Artifacts of ionic species for hi-vol PM10 and PM10 dichotomous samplers. *Atmospheric Environment*, *32*(9), pp.1605-1613.

Tsai, C.J., Huang, C.H. and Lu, H.H., 2005. Adsorption capacity of a nylon filter of filter pack system for HCl and HNO3 gases. *Separation Science and Technology*, 39(3), pp.629-643.