Interactive comment on "Meteorological-gaseous influences on seasonal PM 2.5 variability in the Klang Valley urban-industrial environment" by N.Amil et al.

Anonymous Referee #1

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General comments:

Amil et al. investigate the variability of PM2.5 with season, meteorology, and gas-phase species in an urban-industrial location in the Klang Valley, Malaysia. The authors present results from data collected over a period of approximately one year, detailing the PM2.5 chemical composition and its temporal variation, as well as applying factor analysis to identify potential sources of PM2.5 in the area. To put the findings from this study into context, the authors compare the results to those from previous studies from the same location and similar locations. However, the uniqueness and importance of the study are not highlighted thus it is not obvious how the findings from this work contribute to scientific knowledge. The paper is unnecessarily long, which is in part due to the results detailed in tables being described in full and partly due to the extensive comparison of the findings with those from other studies. Although the results are interpreted in line with the title of the manuscript i.e. how meteorological and gaseous parameters influence PM2.5 variability, little is offered in the way of the significance or importance, which would strengthen the paper if included. For example, the factors identified from PMF analysis are correlated with various parameters with the results described e.g. strong positive or negative correlations, but it is often not suggested as to why such correlations are observed. Similarly, the motivation for some of the analyses is missing. The figures could also be greatly improved, particularly in the time series plots where the text is difficult to see. However, once these major revisions have been addressed along with the comments below, I recommend this manuscript be published in ACP.

Response:

The authors would like to thank the reviewer #1 for all detail suggestion and very useful comments. Each general and specific comment has been addressed in detail below, and appropriate additions and changes to the manuscript have been made where necessary. The uniqueness and importance of the study has been highlighted in the last part of our introduction.

This study was conducted in a period of one-year assessment on PM$_{2.5}$ covering all four seasons (including haze events) to investigate it's variability in the Klang Valley (urban industrial) tropical environment. We have revised our manuscript and deleted unnecessary information. Based on the suggestion of reviewer #2, we have revised our title to 'Seasonal Variability of PM$_{2.5}$ Composition and Sources in the Klang Valley Urban-Industrial Environment'. At the same time we have tried our best to improve our discussion on the influence of meteorological-gaseous parameters to the variability of PM$_{2.5}$ compositions and also the motivation of each analysis in our manuscript. Further, we have improved the quality of our figures.

Specific comments:

Abstract, page 26424, line 5: It may be useful to briefly mention that the seasons are characterized by monsoons/wind flow rather than the more typical summer/fall/winter/spring classification.

Response:

We accept the suggestion and revised the sentence to:

'In total, 94 daily PM$_{2.5}$ samples were collected during a one-year campaign from August 2011 to July 2012. These covered all four seasons, distinguished by the wind flow patterns.'

Abstract, page 26424, line 6: Please clarify what is meant by the 'aerosol pattern' e.g. temporal, spatial.

Response:

We understand the missing keyword and therefore clarify with the revised sentence to:

'The chemical compositions were statistically analysed and the temporal aerosol pattern (seasonal) was characterised using descriptive analysis, correlation matrices, enrichment factors (EF), stoichiometric analysis and chemical mass closure (CMC).'

Abstract, page 26424, line 22: It is not clear how information on the coarse particles was obtained as a high volume PM2.5 sampler was used in this study.

Response:

We understand the concern. Thus, added new sentence regarding this matter:

'In addition, secondary data of total suspended particulate (TSP) and coarse particulate matter (PM10) were used for PM ratio assessment.'

Abstract, page 265424, line 25: Please state in which order the CMC components are listed e.g. in decreasing order of mass or mass contribution.

Response:

We agree with the observation and revised the sentence to:

'The CMC components identified were in the decreasing order of (mass contribution): black carbon (BC) > secondary inorganic aerosols (SIA) > dust > trace elements (TE) > sea salt > K^+.'

<u>Page 26426, line 1:</u> Please clarify what is meant by 'background of an area'.

Response:

Here, 'background of an area' referred to different sites (~1000) at different states of USA used in the study (Tai et al. 2010; Tai et al. 2012) whereby each sites was characterised discretely by the surroundings i.e. topography, local activities i.e. anthropogenic and/or natural air pollution emission etc. over a period of 11 years. Furthermore, the study uses spatial interpolation of 2.5^0 X 2.5^0 grid square of 24-h averages $PM_{2.5}$ fields to produce time series of $PM_{2.5}$ concentrations for each grid square which eventually independently represent an area. To make it short, we use 'background of an area' as summary to this explanation as below:

'Aside from meteorological and gaseous pollutants, seasonal changes and background of an area (topography and local activities affecting anthropogenic and/or natural air pollution emissions) also influenced the $PM_{2.5}$ chemical variability (Tai et al., 2010, 2012).'

<u>Page 26426, lines 8-15:</u> It is not clear why the findings from these studies in particular are referred to; are the locations close by or the conditions/locations similar to that of this study?

Response:

As per mentioned in (Page 26426, lines 2-3), the reason for these studies (Page 26426, lines 8-15) highlighted is the similarity in the sense of seasonal variation of $PM_{2.5}$ mass and its chemical composition thus sources of $PM_{2.5}$ reported in Asia cities. However, we see the unclear heads up on this matter. Therefore, we revised the sentence accordingly:

'Seasonal variation of $PM_{2.5}$ mass and its chemical composition <u>for the Asian region</u> has been widely reported. For example, Balasubramanian et al. (2003) reported that Singapore $PM_{2.5}$ mass temporal…'

<u>Page 26426, line 27:</u> To describe receptor modeling as measuring PM <u>and working backwards to determine sources</u> seems a bit too simplistic. Please expand on the latter half of this sentence. For example, receptor modeling uses temporal and chemical variations to separate total PM into different factors, where marker species are used to identify the sources.

Response:

We agree and appreciate the suggestion from referee #1. Thus, added the following sentence as per suggestion:

'Receptor modelling uses temporal and chemical variations to separate total PM into different factors, where marker species are used to identify the sources.'

Page 26427, lines 4 and 7: What is the difference in these versions of PMF and is it noteworthy?

Response:

We appreciate the feedback. The difference between PMF versions is the computational components of a model. For example, the initial version of the EPA PMF model (version 1.1) did not provide any rotational functions (such as FPEAK, Fkey, and Gkey) as implemented in PMF2. The latest version of the PMF (PMF5.0) has an additional two key components to previous PMF3.0; an addition of two error estimations methods and source contribution and profile constraints. In addition, PMF5.0 has the ability to read data from multiple sites and reduced model run times. However, looking back at context of the manuscript particularly for this section of *Introduction,* we understand the concern. To avoid confusing the readers, we believed that it is not noteworthy to mention the version of the PMF used since the intention here is just to show that PMF is widely used for source apportionment purpose regardless of different type of chemical composition dataset. Thus, we have revised the paragraph by deleting the PMF versions.

Page 26427, line 14: Why is one of the current trends to apply more than one receptor model for SA? Please expand on the purpose, such as is it to increase the confidence in the results or to better characterize the full PM2.5 due to limitations of each of the model or to enable comparison of the results with those from other studies that have used different techniques?

Response:

We understand the requirement to expand on the purpose of applying more than one receptor model. As per suggestion, an additional line has been added to the paragraph:

'One of the current trends of SA is to apply more than one receptor model, a trend set by a number of countries i.e. Belgium, Germany, Portugal and Spain (Viana et al., 2008). Due to limitations of a single model, applying more than one receptor model will enhance the SA analysis, leading to enhanced characterisation of an element and/or source and thus increasing the confidence in interpretations from the results. The study also reports that the most frequent combinations used for SA are principal component analysis (PCA)-cluster analysis (CA), PCA-Lenschow, PCA-chemical

mass balance (CMB), PCA-back-trajectory analysis, PMF-UNMIX-multilinear engine (ME), and CMB-mass balance.'

Page 26428, line 20: Please clarify what is meant by wind flow. Is it a combination speed and direction? Is it a quantitative definition as it is stated in Section 3.1.4 that wind flow correlates with PM2.5 mass? How exactly is it used to distinguish the seasons e.g. a significant change in the wind flow for a period of time?

Response:

We see the missing point. According to Malaysia Meteorological Department (MET Malaysia), though the wind over the country is generally light and variable, there are, however, some uniform periodic changes in the wind flow patterns. Based on these changes, four seasons can be distinguished. Thus, we clarify the 'wind flow' term with the revised sentences:

'Wind flow pattern distinguishes the seasons for Peninsular Malaysia, namely the South-West monsoon (SW), the North-East (NE) monsoon and two shorter periods of inter-monsoons (INT.2 and INT.1) (METMalaysia,2013). During the SW monsoon (usually established during the middle of May until the middle of September), the prevailing wind flow is generally south-westerly and light (below 7.72 m s^{-1}). Known as the dry season, haze is expected to occur during this period. On the other hand, during the NE monsoon (established early November until the middle of March), steady easterly or north-easterly winds of 5.14 to 10.3 m s^{-1} prevail. During periods of strong surges of cold air from the north (cold surges), the winds over the east coast states of Peninsular Malaysia may reach 15.4 m s^{-1} or more. With the highest rainfall intensity and the possibility of flooding, NE monsoon is known as the wet season.'

Page 26430, line 13: Why were the samples stored overnight in the refrigerator? Is it because the analysis needed to be performed all in one day or is refrigeration a necessary part of the chemical analyses method?

Response:

The samples were stored overnight in the refrigerator to allow for equilibrium of the solution. Also yes, since the analysis using ion chromatography is not done immediately after extraction, the proper storage of the extracted solution is to store at 4°C (i.e.in refrigerator).

We understand the requirement and thus add the explanation in the text as follows:

'The extracted solutions were stored overnight in a refrigerator at 4∘C to allow for equilibrium of the solution before analysis using ion chromatography (IC).'

Page 26430, line 20: Why are only 6 method blanks used to calculate the MDLs as there were 12 (one from every month)? Are the other 6 used for the analysis as detailed on page 26431, line 17?

Response:

In any laboratory analysis, the aim for method detection limit (MDL) is to get the lowest yet consistent possible (to minimise the standard deviation) value so as to accurately quantify an element in a solution/etc since this value will be used to be subtracted from the samples filter paper in which state higher confidence level of an analysis. Therefore, we have limit the number of field blank to be used in an analysis. So, yes, for major ions, only six field blanks were used while the other six is used for trace elements analysis.

Page 26433, line 23: has this approach been used in other studies? Please provide a citation if so.

Response:

Yes, this method is referring to the one mention early of the section from Bressi et al. (2013), but we understand the confusing statement/section. We have revised the section as the following:

'Due to our low Al element recovery (36%), and lack of Si and S elements which are the dominant elements in soil from PM$_{2.5}$ (Rahman et al. 2011) the dust fraction is therefore calculated using a straightforward approach used by Bressi et al. (2013). The dust fraction was calculated as the contribution of nss-Ca^{2+} in mineral dust. The 8.3% mineral dust mass contribution for the Klang Valley area estimated by Rahman et al. (2011) was employed for the calculation.'

*We also corrected typo error on the mineral dust mass; changed from '11%' to '8.3%'; also corrected in the CMC equation.

Pages 26433-26434, Sections 2.5.2. and 2.5.3: Due to the low recovery of Al, nss-Ca2+ was used for CMC and yet Fe was used for the EF analysis. How confident are you that the use of these elements are giving similar results as to if Al had been used? Why was Fe chosen as the reference element for the EF analysis?

Response:

As per listed in Table S2, the recovery for Al is just 36% while Ca^{2+} and Fe recoveries were 93% and 69%, respectively. So, the reason why both CMC and EF were calculated as such using such element was that it is the only reasonable thing to do since we have limitations in terms of low recoveries of Al and limit of element determined (to use other method/equation. In addition, Fe were also listed by Lawson and Winchester (1979) as reference for elemental enrichment factors

calculation besides Al, Si, and Ti. Studies by Ho et al. (2006); Kuo et al. (2007); Han et al. (2009) have successfully used Fe in their studies. Both referred source for CMC and EF calculation were a good source. The CMC referring to Bressi et al. (2013) while the EF value are based on the abundance of elements in the Earths's crust (Taylor 1964). Thus, we are confident with the results and the interpretations made from it.

Page 26434, line 20-21: As stated, Cesari et al. (2012) defined EF between 2 and 4 as being of mixed origin. Is there a range in EF values for mixed origins in this study?

Response:

We thank you and really appreciate this feedback. Looking back at our results, it is in fact reasonable to consider having "mixed origin" category for this study since our results reveal some elements of mixed origin during different seasons. Therefore, we have revised the text as follows:

'Due to the low recovery of Al, in this study we opted to use Fe as our reference element for the enrichment factor (EF) analysis. For the cut-off point, we follow Cesari et al. (2012). The study derived a two-threshold system of EF in which, for re-suspended soils, elements with an EF of smaller than two (2) were considered to be from crustal sources, EF of larger than four (4) were considered from an anthropogenic origin while those in between were considered of mixed origin.'

Page 26435, Section 2.5.4: Please provide a few lines of overview on the source apportionment analysis.

Response:

We accept the requirement. Thus, revised the section on the source apportionment analysis as per following:

'A combination of PMF version 5.0 (PMF 5.0) and MLR analysis was employed to determine source apportionment where results of the MLR were used to apportion the $PM_{2.5}$ chemical compositions in order to quantify sources. Details of the PMF procedure used in this study are similar to our previous work as discussed in Khan et al. (2015). In brief, two data files were used as an input, i.e. 1) concentration; and 2) uncertainty. For the concentration data file, the chemical composition dataset were first pre-treated and validated. To ensure a strong signal from the data was evident, species having more than 50% of the data below MDL were discarded. For the rest, the missing values were replaced by half of the MDL while data with values, but below MDL, were left as they were. The final dataset used for the PMF analysis contained 80 samples with 31 elements (including $PM_{2.5}$ mass). Based on the signal to noise ratio (S/N), NO_3^- and Na^+ were set as 'weak'

species while the rest were categorised as 'strong' species. The PM$_{2.5}$ mass was also categorised as "weak" so as not to affect the PMF solution. The second data file is the uncertainty value of each variable in each sample estimated from an empirical equation. An additional 5% uncertainty was added to account for methodological errors during preparation of filter papers, gravimetric mass measurements and preparing the calibration curves. Upon running the PMF analysis, different numbers of factors and Fpeak values have been explored to obtain the most meaningful results with 100 bootstrap runs and a minimum R^2 of 0.6 to test the uncertainty of the resolved profiles. It was observed that a 5 factor solution provided the most meaningful results, based on the lowest Q (Robust) and Q (True) value of 1581.27 with the Q (true)/Q exp value of 0.94 after 390 computational steps and the convergence of the results. PMF factors were resolved on 20 runs and seed value of 9, with Fpeak = 0.5 found to be the most reasonable. The model output of source contribution is provided as normalised or dimensionless (average of each factor contribution is one). To express the output of PMF, the mass concentrations of the identified sources were scaled by using the MLR analysis.'

Page 26436, line 7: Why does low wind speed result in low concentrations? Low wind speeds can result in stagnant conditions thus leading to a build-up of pollution. Is the pollution in this region mostly transported?

Response:

We understood the reviewer concern. Low wind speed will limit the dispersion of air pollutants especially for PM. This will increase the concentration of local and transported PM within the region. Even though the wind speed is low, the rainfall factor is expected to influence more on the PM concentration during this wet season. Therefore, we exclude the wind speed factor to avoid conflict. Thus, revised the sentence with the following:

'The small number of exceedances during the NE monsoon was due to high rainfall (precipitation) during this time.'

Page 26439, lines 2-6: Please can you suggest why these correlations may be the case? For example, the significant positive correlation of PM2.5 and O3 possibly indicates a secondary source of PM2.5 as well as the already identified combustion-related traffic source, which is primary.

Response:

We agree and thankful for the suggestion. Thus, we added the explanation as per suggestion:

'The significant positive correlation of PM$_{2.5}$ and O$_3$ possibly indicates a secondary source of PM$_{2.5}$ as well as the already identified combustion-related traffic source, which is primary.'

Page 26439, lines 26-27: Again, please suggest some reasons for these negative/positive correlations.

Response:

We accept the requirement and thus added the following sentence to justify the statement:

'*This condition is perhaps because during haze episode, the small particle envelope the atmosphere and reduce the UV radiation that can reduce the temperature of earth surface.*'

Pages 26439-26440, lines 29-2: According to the table this statement is incorrect. The SW monsoon has a negative relationship with RH whereas the HAZE has a positive relationship with RH.

Response:

We admit our mistake of the word 'agree' which make the confusing statement where the correct term should have been 'disagree'. We revised the word as per following:

'*However, HAZE events which occur during the SW monsoon, disagree with the generic pattern of the SW monsoon $PM_{2.5}$-RH relationship.*'

Page 26440, lines 17-18: What is the importance/significance of the cation:anion ratio? Is it to indicate neutralization or dominance of a source type, for example?

Response:

Yes, the importance of the cation:anion ratio is to indicate neutralization or to express the acidity of an aerosol at the site which in this case, ratio value > 1 indicate that the aerosol is acidic. We understand the requirement and thus add the explanation to the text as follows:

'*The total anion mass measured was 1.67 μgm^{-3} (6.0 % of $PM_{2.5}$ mass) while the total cation mass was 1.75 μgm^{-3} (6.3 % of $PM_{2.5}$ mass). As shown in Fig. S2, the equivalent charge ratio of total cation to total anion ratio was 0.46 indicates that the aerosol at the site is acidic due to the excess of anions, also experienced by other study (He et al. 2012)*'

Page 26441, line 11 and Figures 3a and 3b: Where is the value of 19% from in the text? The annual values in Fig. 3a differ to the annual values in Fig. 3b.

Response:

The value of 19% (Fig 3b; the actual value is 19.45%) is the combination of all inorganic mass:

19% = 1% of sea salt + 7% of dust + 1% of NO_3^- + 4% of NH_4^+ + 5% of nss-SO_4^{2-} + 1% of TE + 1% of K^+

The reason why the values in Fig. 3a is different from Fig. 3b is that Fig 3a is actual mass value of an element determined while Fig. 3b is the constructed mass using chemical mass closure calculation.

Page 26442, lines 14-15: According to the table legend, what is stated in the text is not how the NR was calculated. NR = [NH4+]/[SO42-] + [NO3-] in the table legend. Should these concentrations be divided by the respective molecular weights?

Response:

We really appreciate this comment in particular. Indeed, we are so sorry for the confusing statement in the text. The formula written in the table is correct but we did not include the word 'equivalent value'. Yes, the concentration was divided by the molecular weight and multiplies with the number of charge. We thank you for pointing out about the NR value. We double-check and admit that we did make mistake in the NR calculation. Thus, we recalculate the NR value, and revised both text and table value, pertaining to this matter:

'Also known as the acidity ratio, the neutralisation ratio (NR) was calculated to further investigate the acidity of the atmospheric aerosols, as reported in Table S1. The NR was calculated based on the ratio of the NH_4^+ (eq m^{-3}) to the sum of SO_4^{2-} and NO_3^- (eq m^{-3}) (Squizzato et al. 2013). The overall NR obtained for this site was 0.26, indicating an excess of SO_4^{2-} and NO_3^-. The NR ratio varied with season. The highest recorded NR was during the HAZE episodes with 0.35. The rest of the values showed the following trend: SW (0.31) > NE (0.22) > INT.2 (0.21) > INT.1 (0.17).'

Table S1. Descriptive statistics of experimental data (meteorological, gaseous and ion parameters); unit: mean ± std (min - max). Remarks:

ud = undetected (below detection limit); SIA = secondary inorganic aerosol; NR = neutralisation ratio = $[NH_4^+]$ / ($[SO_4^{2-}]$ + $[NO_3^-]$), (eq m^{-3}) (Squizzato et al. 2013); SO_2 gas was converted from ppm to µg m^{-3} assuming 1 ppm = 2619 µg m^{-3} (25 °C,1 atm).

Elements	Unit	ANNUAL 5 Aug 2011 - 18 July 2012 n = 81	SW 15 May - 14 Sept n = 29	INT.2 15 Sept - 30 Oct n = 7
API	-	50 ± 16 (29 - 127)	60 ± 21 (36 - 127)	49 ± 6 (40 - 59)
T	°C	28.5 ± 1.19 (26.1 - 31.6)	28.9 ± 1.36 (26.4 - 31.6)	28.5 ± 1.20 (27.1 - 30.4)
RH	%	71.2 ± 7.91 (50.4 - 86.7)	68.2 ± 9.22 (50.4 - 86. 7)	72.9 ± 8.50 (59.7 - 82.7)
WS	ms^{-1}	1.29 ± 0.194 (0.873 - 1.77)	1.39 ± 0.187 (0.966 - 1.77)	1.25 ± 0.198 (1.01 - 1.53)
WD	Degree	129 ± 31.6 (23.1 - 208)	123 ± 38.0 (23.1 - 205)	128 ± 22.0 (100 - 167)
Rainfall	mm	10.4 ± 17.5 (0.000 - 85.4)	6.27 ± 10.6 (0.000 - 34.2)	8.46 ± 16.9 (0.000 - 45.4)
CO	ppm	1.29 ± 0.31 (0.61 - 2.16)	1.26 ± 0.32 (0.61 - 1.99)	1.43 ± 0.32 (1.10 - 1.93)
O_3	ppm	0.012 ± 0.006 (0.000 - 0.029)	0.010 ± 0.007 (0.000 - 0.025)	0.017 ± 0.008 (0.010 - 0.029)
SO_2	ppm	0.003 ± 0.001 (0.001 - 0.008)	0.004 ± 0.002 (0.001 - 0.008)	0.004 ± 0.001 (0.002 - 0.005)
NO_X	ppm	0.062 ± 0.013 (0.028 - 0.109)	0.057 ± 0.012 (0.028 - 0.076)	0.072 ± 0.013 (0.059 - 0.091)
NO	ppm	0.030 ± 0.010 (0.008 - 0.067)	0.025 ± 0.008 (0.008 - 0.041)	0.033 ± 0.008 (0.025 - 0.047)
NO_2	ppm	0.032 ± 0.007 (0.016 - 0.049)	0.032 ± 0.007 (0.016 - 0.048)	0.038 ± 0.006 (0.034 - 0.049)
SO_4^{2-}	µg m^{-3}	1.3 ± 0.88	1.8 ± 1.2	1.6 ± 0.78
ss-SO_4^{2-}	µg m^{-3}	0.076 ± 0.090	0.060 ± 0.023	0.022 ± 0.0079
nss-SO_4^{2-}	µg m^{-3}	1.3 ± 0.90	1.8 ± 1.2	1.61 ± 0.79
NO_3^-	µg m^{-3}	0.21 ± 0.13	0.19 ± 0.077	0.29 ± 0.22
NH_4^+	µg m^{-3}	0.99 ± 0.85	1.5 ± 1.2	1.00 ± 0.64
SIA	µg m^{-3}	2.4 ± 1.7	3.3 ± 2.3	2.8 ± 1.6
SIA/$PM_{2.5}$	%	8.5 ± 3.0	8.7 ± 3.4	9.6 ± 3.0
NR	-	0.26	0.31	0.21
SO_4^2-SO_2	µg m^{-3}	1.3 - 8.2	1.8 - 9.5	1.6 - 10

Continuation of Table S1 (2)

Elements	Unit	NE 1 Nov - 14 Mar n = 35	INT.1 15 Mar - 14 May n = 10	HAZE n = 11
API	-	44 ± 8 (29 - 58)	45 ± 9 (33 - 58)	78 ± 22 (49 - 127)
T	^{o}C	28.1 ± 1.02 (26.1 - 30.4)	28.8 ± 0.78 (27.5 - 30.2)	29.5 ± 1.33 (26.7 - 31.6)
RH	%	73.6 ± 6.79 (56.5 - 85.5)	70.5 ± 4.01 (65.1 – 77.0)	63.0 ± 9.91 (50.4 - 81.6)
WS	ms^{-1}	1.20 ± 0.167 (0.873 - 1.46)	1.32 ± 0.18 (1.08 - 1.71)	1.49 ± 0.138 (1.27 - 1.70)
WD	Degree	132 ± 31.2 (83.2- 208)	128 ± 25.1 (103. - 178)	103 ± 33.2 (23.1 - 137)
Rainfall	mm	15.1 ± 22.7 (0.000 - 85.4)	7.04 ± 9.69 (0.000 – 24.0)	2.28 ± 5.18 (0.000 - 15.8)
CO	ppm	1.29 ± 0.30 (0.92 - 2.16)	1.32 ± 0.28 (0.84 - 1.75)	1.45 ± 0.31 (0.89 - 1.99)
O_3	ppm	0.013 ± 0.005 (0.004 - 0.025)	0.014 ± 0.004 (0.003 - 0.018)	0.016 ± 0.004 (0.011 - 0.025)
SO_2	ppm	0.003 ± 0.001 (0.001 – 0.005)	0.003 ± 0.001 (0.001 – 0.005)	0.003 ± 0.001 (0.001 - 0.005)
NO_X	ppm	0.065 ± 0.014 (0.044 - 0.109)	0.059 ± 0.011 (0.039 - 0.072)	0.057 ± 0.013 (0.028 - 0.074)
NO	ppm	0.034 ± 0.010 (0.021 - 0.067)	0.029 ± 0.008 (0.013 - 0.039)	0.022 ± 0.008 (0.008 - 0.0038)
NO_2	ppm	0.031 ± 0.006 (0.021 - 0.049)	0.030 ± 0.008 (0.018 - 0.044)	0.035 ± 0.008 (0.020 - 0.048)
SO_4^{2-}	µg m^{-3}	0.98 ± 0.41	1.1 ± 0.70	2.4 ± 1.2
ss-SO_4^{2-}	µg m^{-3}	0.048 ± 0.029	0.16 ± 0.15	0.059 ± 0.00
nss-SO_4^{2-}	µg m^{-3}	0.95 ± 0.42	0.90 ± 0.60	2.4 ± 1.2
NO_3^-	µg m^{-3}	0.19 ± 0.13	0.27 ± 0.13	0.22 ± 0.14
NH_4^+	µg m^{-3}	0.65 ± 0.34	0.82 ± 0.49	2.2 ± 1.5
SIA	µg m^{-3}	1.7 ± 0.81	2.0 ± 0.88	4.7 ± 2.6
SIA/$PM_{2.5}$	%	8.2 ± 2.8	8.4 ± 2.3	7.5 ± 2.7
NR	-	0.22	0.17	0.35
SO_4^2-SO_2	µg m^{-3}	0.98 - 6.6	1.1 – 8.0	2.4 - 9.1

Page 26442, lines 15-23: What is the importance of the NR value? If only just more than half of the aerosol acidity has been neutralized by ammonium what implications does this have?

Response:

We have revised our calculation based on the suggestion by the reviewer where the new NR value was calculated based on the concentration divided by the molecular weight and multiply with the number of charge. The new NR value is 0.26 indicate an excess of SO_4^{2-} and NO_3^- at the site. Therefore, we feel it is reasonable to remain having the NR analysis results in the manuscript.

Page 26443, lines 7-8: What does this sentence mean? How was the slight difference in these elements identified? What are they different to?

Response:

We see the missing point and thus the revised sentences:

'*However, the seasonal results revealed a slight difference in several elements (Cu, Rb, V and Ni); as shown in Fig. S3. For example, Cu during SW monsoon follows the annual grouping of anthropogenic source while during other seasons, it is drawn from crustal source. Meanwhile, Rb, V and Ni during the SW monsoon to originate from the anthropogenic source which is contrary to the annual and other seasonal pattern. Ni and V are known as heavy oil combustion indicators (Jiang et al., 2014), Cu is known to be associated with the traffic (Contini et al. 2014) while Rb known drawn from crustal source (Khan et al. 2010). A study in Taiwan also argued that these four element (Cu, Rb, V and Ni) are likely to be affected by both soil and non-soil emission (Balakrishnaiah et al., 2012).*'

Page 26444, lines 23-25: Why might the results from this study be low compared to most other SEA cities? Please offer some possible reasons.

Response:

We accept the requirement and thus try to reason the situation with the following sentence:

'*On a regional scale, our results here are comparatively low compared to most other SEA cities as reported by Reid et al. (2013). One possible reason is because this study was carried out on a long-term basis while the others mostly concentrated on a particular season and/or event, especially haze episodes.*'

Page 26447, lines 1-9: If this factor (combustion of engine oil) predominantly primary, why might the reason be that a positive correlation is observed between O3 and this factor during INT.2?

Response:

We understand the requirement and thus added the following explanation:

'The positive correlation during INT.2 was perhaps due to higher measurements of NOx and NO_2 during this time period compared to other seasons. NO_2 provides an O-radical which contributes to the formation of O_3 with the assistance of sunlight and volatile organic compounds (VOCs). High concentrations of O_3 and other organic pollutants can lead to the formation of secondary organic aerosol; this may explain the observation results.'

Page 26447-26448, lines 28-1: This sentence (and similar sentences in other sections describing the factors) needs to be rephrased. Currently it sounds like the correlation is between HAZE and the gases, when it is the correlation of the factor with the gases during the HAZE period.

Response:

We accept the feedback and revised them accordingly as follows:

1) Page 26447-26448, lines 28-1:

'This factor during HAZE seems to be affected by a few gaseous parameters i.e. NOx and NO with r = 0.650 (p = 0.042) and r = 0.698 (p = 0.025), respectively.'

2) Page 26449, lines 26-28:

'With highest mass contribution during SW, this factor showed significant (p < 0.05) positive correlations with CO, O_3, and NO_2 at r = 0.612, r = 0.597 and r = 0.422, respectively.'

3) Page 26451, lines 19-22:

'Season-wise, following high mass contribution, this factor during INT.2 showed significant correlation with NOx and NO_2 with r = 0.774 (p < 0.05) and r = 0.766 (p < 0.05), respectively. On the other hand, during the NE dry season this factor showed a negative correlation with O_3 (r = −0.351; p < 0.05) and WS (r = −0.507; p < 0.05).'

Page 26448, lines 20-22: What is the relevance of the discussion on different uses of potassium to estimate biomass burning? For example, is the Kb indicator used in this study?

Response:

The reason being is to backed up our choice of using only K^+ instead of total K (not determined) as an indicator for biomass burning. However, we see the irrelevance of too many studies for this purpose. Therefore, we have limited the reference list and shorten the section (by deleting unnecessary discussion) as follows:

'The combined sum of ammonium sulfate and ammonium nitrate represents the secondary inorganic contribution to the $PM_{2.5}$ mass. This study is clearly dominated by ammonium sulfate. The potassium ion (K^+) on the other hand is an indication of major soil elements, usually from biomass burning. Echalar et al. (1995) has indicated that potassium (K) may be considered a good tracer for the flaming phase of forest fires. Watson and Chow (2001) reported that 85% of the K is in the soluble form K^+, which is consistent with most vegetative burning profiles. Due to this established relationship, K^+ in PM was seen in many studies as a marker of biomass origin, either in the European region (Reisen et al. 2013) or the SEA region (Tahir et al. 2013; Wahid et al. 2013; Mustaffa et al. 2014; Ee-Ling et al. 2015). Reche et al. (2012) reported that K^+ from biomass burning was mostly emitted in the fine fraction of PM rather than coarse particles. Characterised by high levels of NH_4^+ (59% of NH_4^+ mass), SO_4^{2-} (46% of SO_4^{2-} mass) and K^+ (49% of K^+ mass), the third and biggest factor for this site was identified as a mix of SIA and biomass burning and makes up 42% of the $PM_{2.5}$ mass on annual basis. Studies by Mooibroek et al. (2011), Zhang et al. (2013), Almeida et al. (2005), Yin et al. (2010) and Song et al. (2006) also identified a major contribution by the secondary aerosol fraction to $PM_{2.5}$.'

<u>Page 26449, lines 25-26:</u> Please consider rephrasing this sentence to something like

'The strongest correlations between this factor and gaseous-meteorological parameters were observed during the SW monsoon season'.

Response:

We accept the suggestion and revised the sentence to:

'The strongest correlations between this factor and gaseous-meteorological parameters were observed during the SW monsoon season.'

<u>Page 26450, lines 17-18:</u> What about the EF analysis of Se and Rb? These two elements are not discussed in this section.

Response:

We admit our mistake. Actually, we accidently include Se and Rb in the *section title*. Therefore, we have revised the *section title* accordingly as follows:

'3.3.4 Factor 4: mixed traffic and industrial (NO_3^-, Pb, NO_2^-, Zn, As, Bi, Cd, BC)'

<u>Page 26451, lines 11-13:</u> Please list some of the possible anthropogenic activities in the surrounding area that could produce ammonium nitrate.

Response:

We accept the suggestion and thus added some possible anthropogenic activities to the text:

'NO_3^- and NO_2^- could also possibly come from the secondary aerosol of ammonium nitrate from anthropogenic activities in the surrounding area such as motor vehicle exhaust, industries (petrochemical industry, iron/steel plant etc), and stationery combustion sources (coal plant etc).'

Page 26452, lines 21-23: This result is not obvious. If this was the case then it would be expected that INT.2 had the lowest ratios.

Response:

We admit our mistake for this sentence. Supposedly, it should be highlighting the INT.1. Therefore, we corrected the season from 'INT.2' to 'INT.1' as follows:

'From these results, it is obvious that INT.1 contributed more Ca^{2+} and Na^+ with higher occurrences of chloride loss or the "chlorine deficiency" phenomenon compared to other seasons.'

Page 26453, lines 18-23: These few sentences are not necessarily needed as it is all detailed in the table that is referred to in the text. Possibly shorten these sentences or use the values to compare the composition of the two HAZE periods rather than listing the contributions of the sources.

Response:

We accept the comments and thus revised the sentence:

'For a total of 19%, four other factors were identified: combustion of engine oil, sea salt, mineral dust, and mixed traffic and industrial. These factors do not seem to have a strong influence on HAZE 2011. However, HAZE 2012 was strongly influenced by those four factors, with a combined contribution of 44%.'

Page 26454, lines 15-18: Please clarify that during HAZE 2011, strong correlations are observed but they are not significant.

Response:

We accept the suggestion and thus the following revised sentence:

'Further analysis showed that HAZE 2012 was more influenced by the meteorological and gaseous parameters whereas during HAZE 2011, strong correlations were observed but they are not significant; as shown in Table S6.'
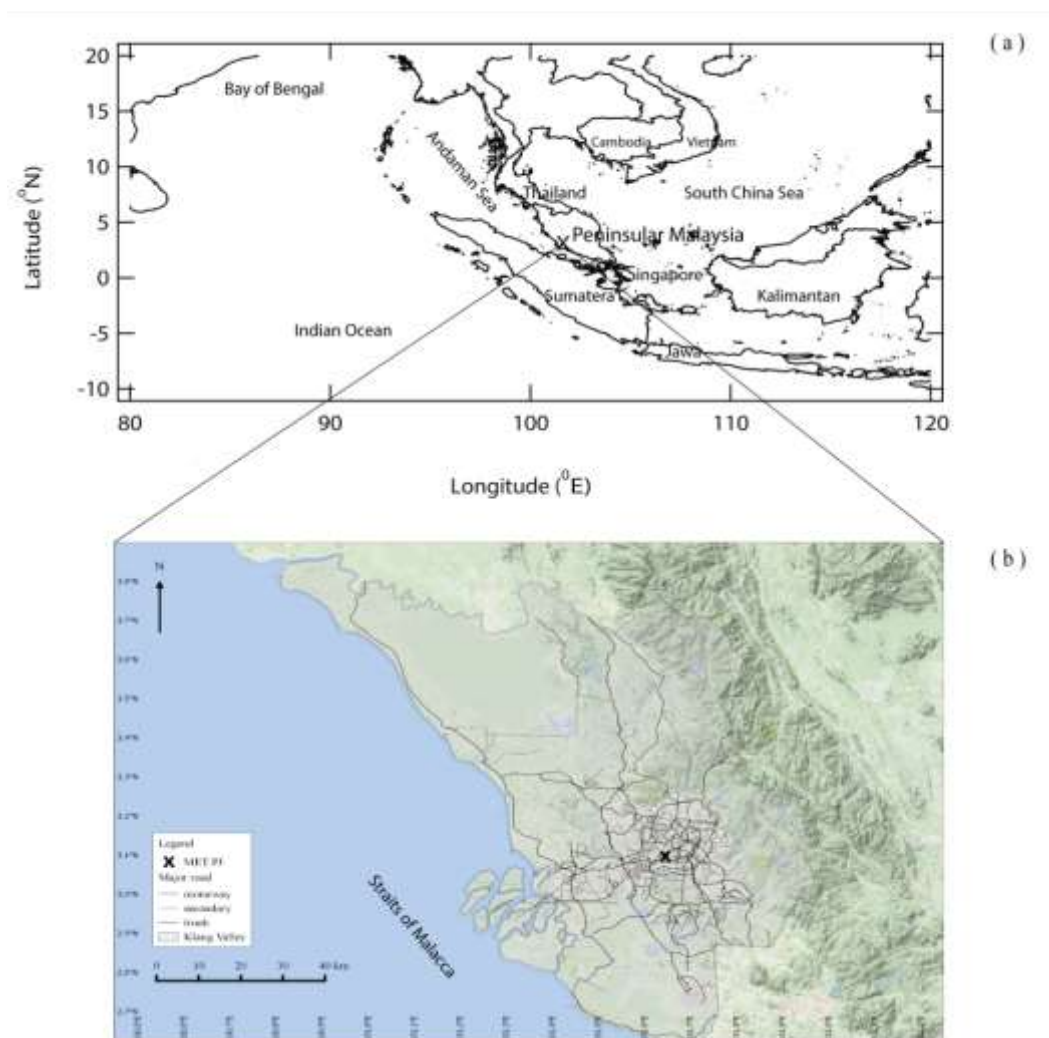
<u>Page 26475, Figure 1:</u> Please consider adding a few place names to the map in figure (a). What does 'trunk' refer to in the legend of figure (b)?

Response:

We accept the requirement and thus revised the map and also added remarks for the legend used in Fig 1b (in the caption) as follows:
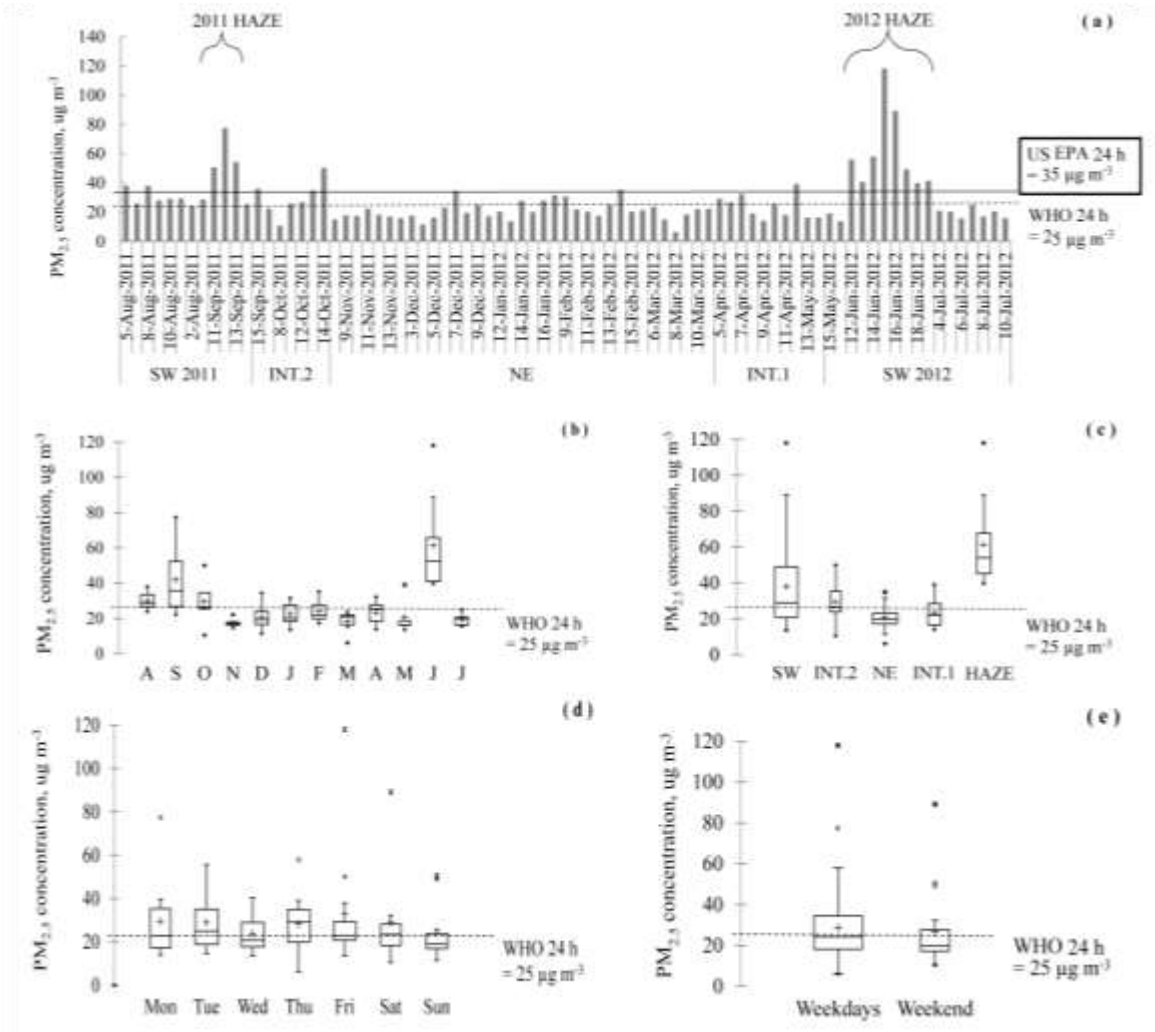

'Figure 1. Location of the sampling site mark as "X" in: a) the Southeast Asia region where the area is the boundaries of MODIS fire hotspot data used; and b) the Klang Valley area in the Peninsular Malaysia. Remarks: motorway = toll highway; secondary = main road; trunk = highway (main road) with traffic intersection'

Please state what the box and whiskers represent e.g. 5th and 95th percentiles. The size of the text in figure (a) in particular is too small and difficult to read.

Response:

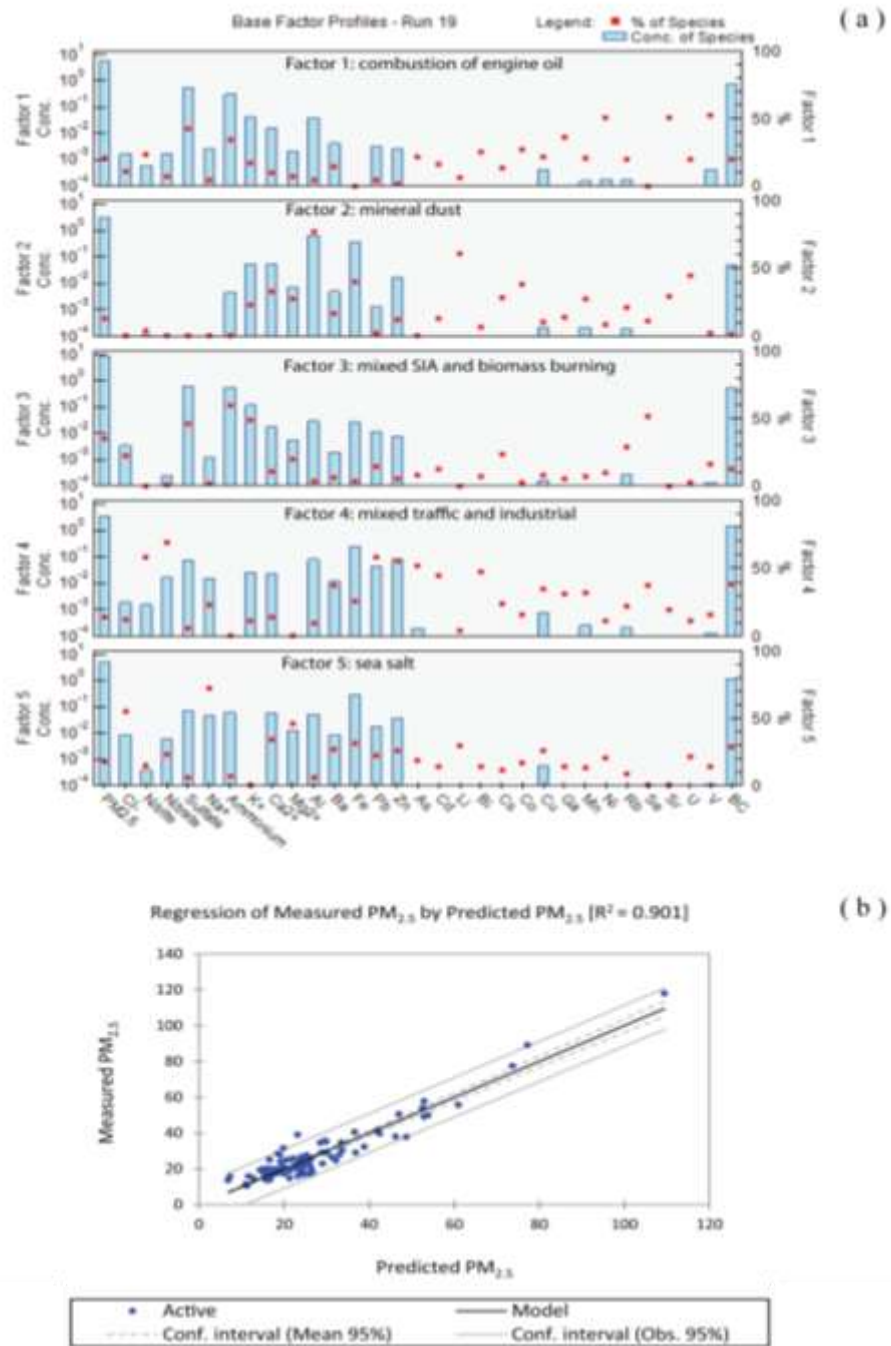We accept the requirement. Thus, revised the caption and also Fig 2a:

Figure 2. The PM$_{2.5}$ mass concentration on the: (a) daily basis; with box and whisker plots (of the: (b) monthly; (c) seasonal; (d) days; and (e) weekdays/weekend. For the box and whisker plots, the horizontal line within the box indicates the median, boundaries of the box indicate the 25th and 75th percentile, and the whiskers indicate the highest and lowest values of the results. The "+" marked in the box indicates the mean. All figures were also subject to World Health Organisation (WHO) daily PM$_{2.5}$ guideline and United States Environmental Protection Agency (US EPA) daily PM$_{2.5}$ standard, accordingly'

<u>Page 26478, Figure 4:</u> What do all the different lines in figure (b) represent?

Response:

We accept the suggestion. Thus, the following revised figures and caption:

'*Figure 4. Source apportionment results from positive matrix factorisation (PMF) analysis:*

(a) source profile; and (b) regression plot between measured and predicted PM$_{2.5}$ mass. Remark:

SIA = secondary inorganic aerosol'

<u>Page 26479, Figure 5:</u> Should figure (c) be a fraction or percentage if it is relative contribution or mass concentration rather than contribution?

Response:

We admit our mistake. Thus revised the caption:

'*Figure 5. Time series of daily and monthly variations (left to right) of: (a) gaseous; (b) meteorological parameters; and (c) mass concentration of PM$_{2.5}$ sources.*'

<u>Supplement: Table S1:</u> It is not necessary to have as many decimal places as are used here. Consider using 3 significant figures throughout. What is SO_4^{2-}-SO_2?

Response:

We accept the suggestion to use 3 significant figures throughout. Thus, we revised all tables and text manuscript accordingly.

'SO_4^{2-}-SO_2' value is given to show the conversion of gas-to-particle conversion of SO_2. Sulphates in the lower atmosphere are generally assumed to be a secondary airborne particulate formed from gas-to-particle conversion of SO_2.

<u>Figure S1:</u> This figure is very hard to read. Possibly making the figure landscape rather than portrait might help. The synoptic wind fields in particular need to be addressed.

Response:

We agree that the figure need revision. Hence, the revised figure; which will be made in landscape in the supplementary information (SI).
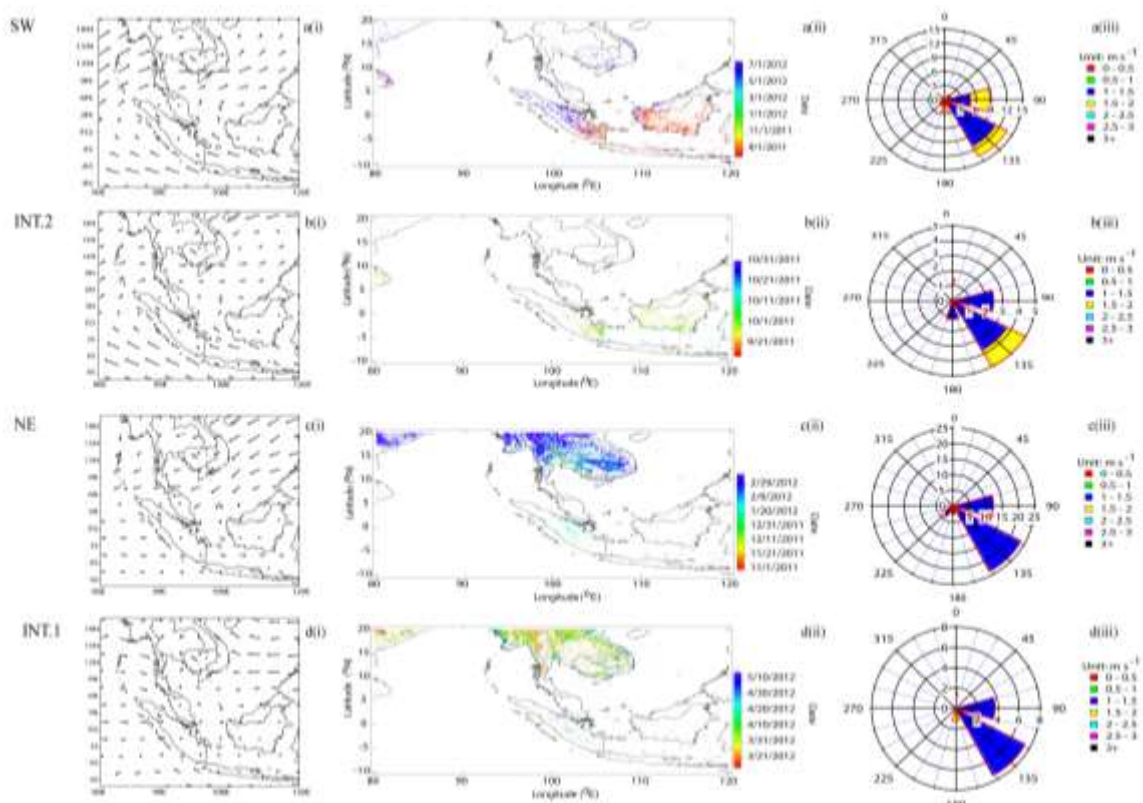
<u>Figure S2:</u> Please consider using color in this plot as well as the different symbols as it's very difficult to tell the fit lines from the data points apart. The cluster of points towards to the lower left of the plot all merge too even though different symbols are used. Color would probably really help this figure.

Response:

We agree that the figure need revision. Hence, the revised figure as per suggestion:

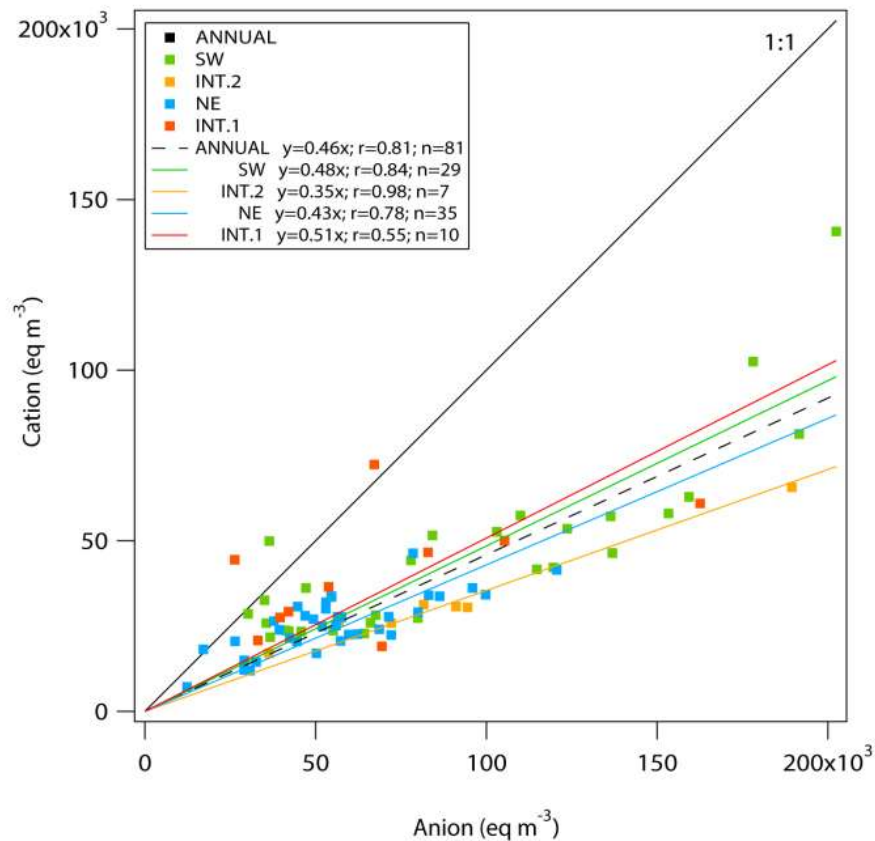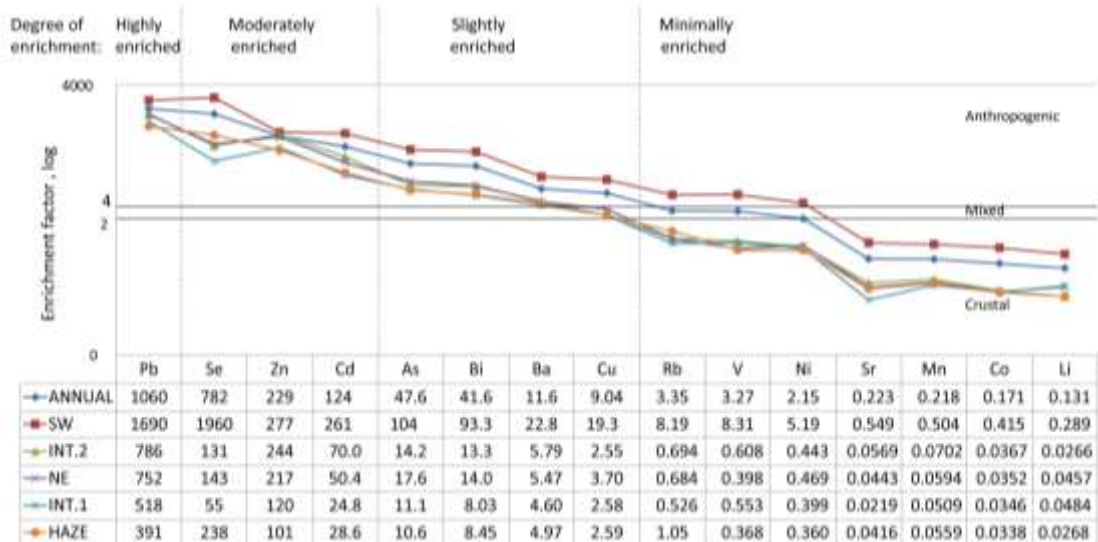<u>Figure S3:</u> This figure is not particularly simple to interpret. Please consider including additional horizontal lines and tick marks to indicate the different levels of EF (the different source types cut offs as well as the highly enriched, moderately enriched values etc.).

Response:

We agree that the figure need revision. Thus, we revised the figure as well as the text as per suggestion:

Degree of enrichment: Highly enriched | Moderately enriched | Slightly enriched | Minimally enriched

Enrichment factor, log

4000

4
2

0

Anthropogenic

Mixed

Crustal

	Pb	Se	Zn	Cd	As	Bi	Ba	Cu	Rb	V	Ni	Sr	Mn	Co	Li
ANNUAL	1060	782	229	124	47.6	41.6	11.6	9.04	3.35	3.27	2.15	0.223	0.218	0.171	0.131
SW	1690	1960	277	261	104	93.3	22.8	19.3	8.19	8.31	5.19	0.549	0.504	0.415	0.289
INT.2	786	131	244	70.0	14.2	13.3	5.79	2.55	0.694	0.608	0.443	0.0569	0.0702	0.0367	0.0266
NE	752	143	217	50.4	17.6	14.0	5.47	3.70	0.684	0.398	0.469	0.0443	0.0594	0.0352	0.0457
INT.1	518	55	120	24.8	11.1	8.03	4.60	2.58	0.526	0.553	0.399	0.0219	0.0509	0.0346	0.0484
HAZE	391	238	101	28.6	10.6	8.45	4.97	2.59	1.05	0.368	0.360	0.0416	0.0559	0.0338	0.0268

'*Figure S3. Seasonal variation of enrichment factor (EF) in the trace element of $PM_{2.5}$.*
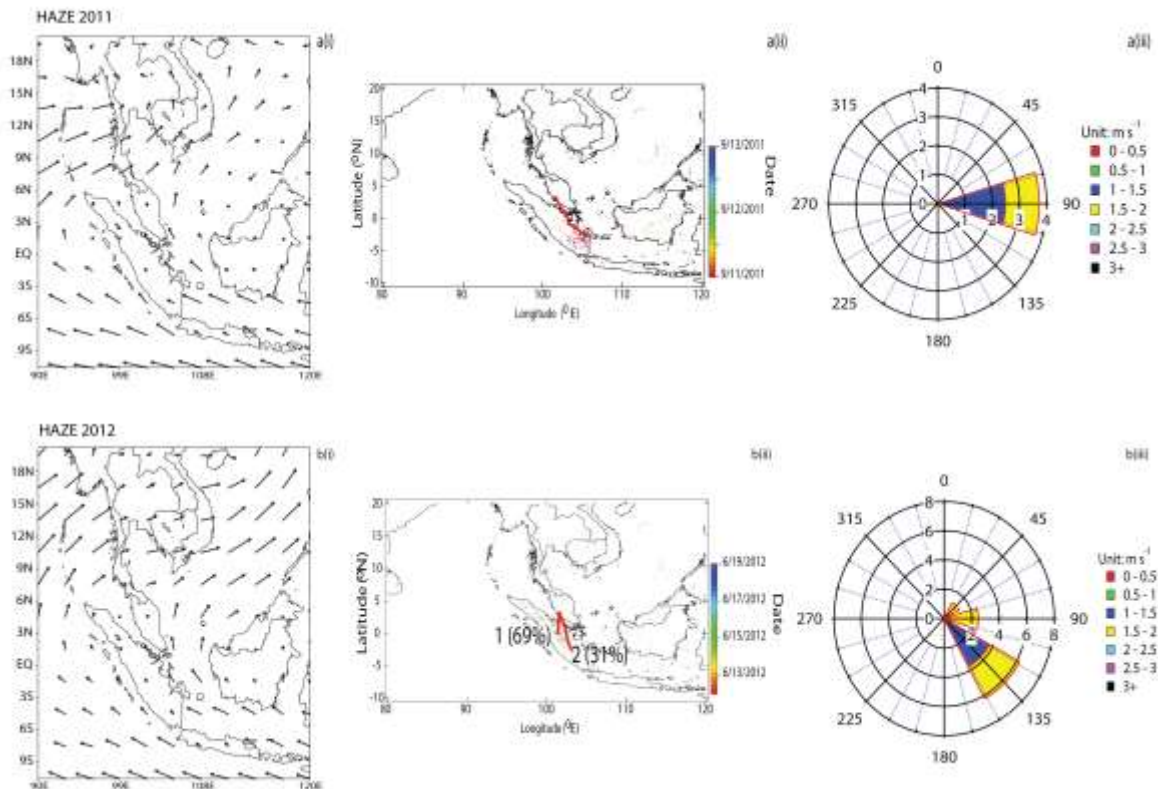
Remarks:

EF category = Anthropogenic: EF > 4; Mixed: 2 < EF < 4; Crustal: EF < 2.

Degree of enrichment = Highly enriched: EF ≥ 1000; Moderately enriched: 100 < EF < 1000; Slightly enriched:10 < EF < 100; and Minimally enriched: EF < 10.'

Figure S4: Similar comments as for Fig. S1. The figure needs to be made clearer.

Response:

We accept the requirement as thus the following revised figure; which will be made in landscape mode later on in the Supplementary Information (SI):

Minor and technical corrections:

Abstract, line 14: Please consider rephrasing this sentence to something like 'On a daily basis, the PM2.5 mass ranged between 6 and 118 µg m−3 with the daily WHO guideline exceeded 43 % of the time.'

Response:

We agree and appreciate the suggestion. Thus, revised the sentence to:

'On a daily basis, the $PM_{2.5}$ mass ranged between 6 and 118 µg m^{-3} with the daily WHO guideline exceeded 43 % of the time.'

Abstract, line 21: Please define TSP.

Response:

We appreciate the referee highlighted the missing definition of TSP in the section. In conjunction with other requirement, the following sentence was added to the abstract and hopefully cover this matter.

'In addition, secondary data of total suspended particulate (TSP) and coarse particulate matter (PM10) were used for PM ratio assessment.'

<u>Abstract, page 26425, line 2:</u> Please define SIA.

Response:

We already define SIA in previous page (page 26424, line25) which is still in the same section of the *abstract* section.

<u>Page 26425, line 14:</u> Please consider using a different term than 'pristine' such as rural or less polluted.

Response:

We accept the suggestion and thus changed 'pristine' to 'less polluted'.

<u>Page 26425, line 19:</u> Please consider using a different term than 'stable' as this is usually used more for radioactive species rather than volatilities. Perhaps just say that the fine particles are composed of compounds of a range of volatilities.

Response:

We agree and accept the suggestion. Thus revised the sentenced to:

'The fine particles, which are composed of compounds of a range of volatilities, appear to do more harm to human health than coarse particles (Dockery et al. 1993; Schwartz et al. 1996; Laden et al. 2000; Lanki et al. 2006; Pope III & Dockery 2006; Krewski et al. 2009; Tagaris et al. 2009; WHO 2013).'

<u>Page 26427, lines 8 and 10:</u> Remove the word 'has' after the reference.

Response:

Removed the word 'has' after the reference.

<u>Page 26428, line 19-20:</u> Please consider including some values for the uniform temperature, high humidity etc.

Response:

We understand the suggestion and therefore revised the sentence to:

'Overall, Peninsular Malaysia experiences relatively uniform temperature (~28.5^{o}C), high humidity (more than 70%) and copious rainfall (6.27-15.1mm) throughout the year.'

<u>Page 26429, line 3:</u> Change 'were' to 'are'.

Response:

Changed 'were' to 'are'.

<u>Page 26429, lines 4 and 5:</u> it is not necessary to provide all of these values to 2 decimal places. Please consider using 3 significant figures here and <u>throughout the manuscript.</u>

Response:

We accept the suggestion and thus corrected the sentence to '*The average temperature (T) at the site during the sampling campaign was 28.5 ± 1.19◦C and the average relative humidity (RH) was 71.2 ± 7.91 %.*'

In addition, we also corrected the value for Page 26429, lines 8 from 'Rainfall was lowest during SW (6.27±10.63 mm) and highest during NE (15.13±22.69 mm).' to '*Rainfall was lowest during the SW monsoon (6.27±10.6 mm) and highest during the NE monsoon (15.1±22.7 mm).*'

<u>Page 26430, line 22:</u> Change 'use' to 'used'.

Response:

Changed 'use' to 'used'.

<u>Page 26431, line 3:</u> Is power of 0 W correct for setting number 2 or is this a typographical error?

Response:

The value for power of 0 W for setting number 2 is correct as stated but there is a typographical error for time of setting number 4. The sentence has been corrected to:

'*For the digestion process, one strip (2.54 cm × 20.32 cm) of loaded filter was used with the following setting of time (m) and power (W) was used: 1) 1, 250; 2) 1, 0; 3) 8, 250; 4) 5, 400 and 5) 5, 650.*

<u>Page 26433, line 23:</u> Please rephrase this sentence as 'nss-Ca2+ over the mineral contribution' does not make sense.

Response:

We accept the comment and therefore rephrase the sentence to:

'*Due to our low Al element recovery (36%), and lack of Si and S elements which are the dominant elements in soil from $PM_{2.5}$ (Rahman et al. 2011) the dust fraction is therefore calculated using a straightforward approach used by Bressi et al. (2013). The dust fraction was calculated as the contribution of nss-Ca^{2+} in mineral dust. The 8.3% mineral dust mass contribution for the Klang Valley area estimated by Rahman et al. (2011) was employed for the calculation.*'

Additional info: we made correction the typo error for the mineral dust contribution from '11%' to '8.3%'

Page 26435, line 5: Please change 'remain as it is' to 'remained as it was'.

Response:

Changes were made accordingly with latest version of the section.

Page 26435, line 19: Please change 'days' to 'daily'.

Response:

Changed 'days' to 'daily'.

Page 26436, lines 6-7: Please rephrase this sentence to something like 'The small number of exceedances during the NE monsoon was due to. . .'.

Response:

We accept the suggestion and rephrased to:

'The small number of exceedances during the NE monsoon was due to high rainfall (precipitation) during this time.'

Page 26436, line 23: Please change 'ompared' to 'compared'.

Response:

Changed 'ompared' to 'compared'.

Page 26439, line 9: Insert 'is' between 'which' and 'also'.

Response:

Inserted 'is' between 'which' and 'also'.

Page 26441, line 12: Please consider removing 'of the 34% chemical composition determined' as this makes the sentence a little confusing.

Response:

We accept the suggestion and deleted them. Thus, the revised the sentence:

'*In this study, IM accounted for 19 % of PM$_{2.5}$ mass while BC accounted for 15 %.*'

Page 26451, line 20: Insert 'other' between 'the' and 'hand'.

Response:

Inserted 'other' between 'the' and 'hand'.

Page 26453, line 16: Change 'characters' to 'characteristics'.

Response:

Changed 'characters' to 'characteristics'.

Page 26472, Table 2: A reference for the Gombak study is missing.

Response:

We are sorry for the confusing table. We mistakenly draw a line after the Petaling Jaya, Klang Valley, Malaysia study. The Gombak study is actually within the Keywood et.al (2003). We corrected the table as per following:

Table 2. Comparison of $PM_{2.5}$ mass recorded in this study with other previous studies.

Location	PM$_{2.5}$ mass (ug m^{-3})	Site description	Sampling period (24 h)	Reference
Petaling Jaya, Klang Valley, Malaysia	28 ± 17	Urban - industrial	5 Aug 2011 - 10 July 2012	This study
Kuala Lumpur, Klang Valley, Malaysia		Urban Metropolitan	Jan - Mar 2013	Ee-Ling et al. (2015)
	30 ± 7	Urban Metropolitan		
	18 ± 3	Semi-urban		
	10 ± 4	Rural		
Kuala Lumpur, Klang Valley, Malaysia	27 + 10	Urban	Jan 2004 - Dec 2008	Rahman et al. (2011)
Kuala Terengganu, Malaysia	14 ± 7	Coastal, Sub-urban	Aug 2006 - Dec 2007	Tahir et al. (2013)
Petaling Jaya, Klang Valley, Malaysia	33	Urban - industrial	Dec 1998 - Dec 2000	Keywood et al. (2003)
Gombak, Klang Valley, Malaysia	28	Urban - residential	Dec 1998 - Dec 2000	
New Taipei City, Taiwan	22 ± 8	Urban Industrial	May 2011 - Nov 2011	Gugamsetty et al. (2012)
Agra, India	140 ± 22	Urban Industrial	Nov 2010 - Feb 2011	Pachauri et al. (2013)
	308 ± 52	Traffic		
	91 ± 17	Rural		

Paris, France	15 + 10	Urban	11 Sept 2009 - 10 Sept 2010	Bressi et al. (2013)
	15 + 11	Semi-urban		
Qincheng, China	51 ± 18	Industrial complex	5 - 16 Aug 2009; 24 Jan - 4 Feb 2010	Huang et al. (2013)
Beijing, China	135 + 63	Urban	Apr 2009 - Jan 2010	Zhang et al. (2013)
Venice, Italy	33	Urban	Mar 2009 - Jan 2010	Squizzato et al. (2013)
	33	Industrial		
	26	Semi-urban		
Birmingham, United Kingdom	12	Urban	May 2007 - Apr 2008	Yin et al. (2010)
	10	Rural		
Palermo, Sicily, Italy	34	Metropolitan; Urban 1	Nov 2006 - Feb 2008	Dongarrà et al. (2010)
	24	Urban 2		
Singapore	27 ± 10	Urban	Jan – Dec 2000	Karthikeyan and Balasubramanian (2006)

Anonymous Referee #2

The manuscript "Meteorological-gaseous influences on seasonal PM2.5 variability in the Klang Valley urban-industrial environment" by Amil et al. analyzes the chemical composition of PM2.5 aerosol samples collected in an urban environment characterized by emissions of several industrial activities and a nearby harbor. PM2.5 aerosol sources are then identified based on chemical characterization using chemical mass closure approach and positive matrix factorization (PMF5.0) analysis. The manuscript discusses data collected during an entire year and representative of four different seasons, together with two haze episodes. In the referee's opinion, the set of data presented and discussed by the authors is interesting and worth of publication, nevertheless major corrections are needed before the manuscript could be accepted for publication in ACP.

Major comments

The manuscript title is misleading. The text describes in several sections the correlation between meteorological parameters and PM composition and/or PM sources, although often the correlation, or the lack of correlation, is not discussed or justified. Similarly, the correlation between gas phase species and PM sources is presented, but not discussed. For example the correlation between dust and NO2 is observed, but no explanation or hypothesis are offered to the reader. I would suggest re-phrasing the title. An example could be: "Seasonal variability of PM2.5 composition and sources in the Klang Valley urban-industrial environment".

Response:

The authors would like to thank the reviewer #2 for all detail suggestion and very useful comments. We understand the concern of the reviewer. We have tried our best to amend our manuscript based on the suggestion and improved our discussion on the correlation between meteorological-gaseous and $PM_{2.5}$ mass/composition/sources. However, we agree with the suggestion of the reviewer due to the limitation of our discussion. Therefore, we changed the title to *'Seasonal Variability of PM$_{2.5}$ Composition and Sources in the Klang Valley Urban-Industrial Environment'.*

<u>Results and discussion</u> section presents monthly and annual averages of PM values. Formally, it is not correct to talk about monthly averages when only 1 week of each month was monitored. It would be more accurate to talk about weekly average representative of the month. This is important especially when comparing the mean values with mean values collected at other sites for the entire month, or when comparing with limits set by the WHO or by the legislations.

Response:

We understood the concern of the reviewer and accepted the suggestion. As a reminder to the reader on this matter, we therefore emphasize the 'weekly average representative of the month' in the text as follows:

'The annual $PM_{2.5}$ mass (weekly average representative of the month) for this study averaged at 28 ± 18 μg m^{-3}.

<u>The discussion of PM2.5 to PM10 ratios needs revision and further discussion.</u> The similar PM2.5 to PM10 ratio during the wet and the dry season indicates that meteorological parameters, specifically rainfall, are affecting in the same way fine and coarse particles. This is also confirmed by the good correlation of PM2.5 and PM10. Since it is likely that coarse particles are dominated by dust, and source apportionment and CMC show that dust is not a dominant component of PM2.5, it is unlikely that the correlation of fine and coarse particle mass was due to common sources at this site. Finally during INT1 period the PM2.5 to PM10 ratio is the highest. Is it possible to discuss this ratio at the light of the source apportionment results?

Response:

We agree with the reviewer comment. Based on the results, it is suggested that both coarse and fine mode aerosols are not from the same sources. Nevertheless both coarse and fine mode aerosols can be reduced by rainfall. We agree that the reviewer has suggested a very interesting point for the discussion of ratio at the light of the source apportionment results. Unfortunately, we only determine the source apportionment based on our $PM_{2.5}$ results (fine mode aerosols). We take note this important point and have revised our sentence in the our main text. We will consider this important point in our future studies. We have included this point in our conclusion as well revised the section pertaining to this matter:

'The mean $PM_{2.5}/PM_{10}$ ratio for the site was 0.72 ± 0.18 and the ratio for $PM_{2.5}/TSP$ was 0.46 ± 0.13; Table 1. PM_{10}/TSP ratio was 0.63 ± 0.12. The $PM_{2.5}/PM_{10}$ ratio at this site is comparatively higher than other studies in Asia as reported by Hopke et al. (2008) where

most of the sites studied showed ratios of lower than 0.50. From the aforementioned study, however, an urban site in China and suburban site in Lembang, Indonesia recorded similar $PM_{2.5}/PM_{10}$ ratio to our result of more than 0.70. Our $PM_{2.5}/PM_{10}$ ratio was also in agreement with other cities in Europe (Gehrig & Buchmann 2003; Gomišček et al. 2004; Contini et al. 2014). Despite having different characteristics, the SW and NE monsoons still came out with similar values to the annual ratio at 0.72 ± 0.10 and 0.71 ± 0.13, respectively. The similar $PM_{2.5}$ to PM_{10} ratio during the wet and the dry season indicates that meteorological parameters, specifically rainfall, are affecting in the same way fine (particle with an aerodynamic diameter of less than 2.5µm) and coarse (particle with an aerodynamic diameter of greater than 2.5µm) particles. This is also confirmed by the good correlation of $PM_{2.5}$ and PM_{10} (r = 0.963; p < 0.0001). Both inter-monsoon seasons recorded the opposite mass concentration trend. INT.2 (average mass of 29 ± 12 µg m^{-3}) showed a higher mass concentration than INT.1 (average mass of 23 ± 8 µg m^{-3}) but a lower $PM_{2.5}/PM_{10}$ ratio (0.62 ± 0.17) than INT.1 (0.85 ± 0.40). This ratio of INT.1 is the highest $PM_{2.5}/PM_{10}$ ratio among all seasons, even higher than during HAZE episodes. HAZE-episode-only ratios were 0.74 ± 0.07. To further examine the particle at the site, the seasonal $PM_{2.5}/TSP$ ratio was calculated. During the dry season (the SW monsoon), ambient air at the site had particles in the ratio of approximately 50/50 coarse to fine particles ($PM_{2.5}/TSP$ = 0.50 ± 0.081). During INT.2 and the NE monsoon (wet season), the air was filled with more coarse particles, resulting in $PM_{2.5}/TSP$ ratios of 0.44 ± 0.12 and 0.40 ± 0.087, respectively. INT.1 and HAZE episodes on other hand both had a $PM_{2.5}/TSP$ ratio of 0.54, implying the ambient air contained almost the same portion of fine and coarse particles. With these ratios, we can conclude that fine particles are very significant in the ambient air of the Petaling Jaya urban-industrial area in Klang Valley. Similar observation on the significance of the fine particle were also reported for SEA cities (Kim Oanh et al. 2006).'

We also have added sentence to the Conclusion section:

'However, our study is limited to only fine particle. Parallel sampling of both fine and coarse particle will give better insight on the actual condition of the aerosol at a site. With the use of meteorological-gaseous parameters, concrete conclusion can be achieved, as to whether meteorological-gaseous parameters are affecting in the same way fine and coarse particles, and whether both fine and coarse particle share common sources.'

Results of the chemical mass closure analysis are presented reporting the absolute concentration of each PM component and the percentage mass contribution. The results are then discussed based on the percentage mass contribution. Since the goal of the discussion is to investigate the effect of meteorological parameters on PM sources, the mass fraction is not a useful measure since the normalization to the total mass removes some meteorological effects, like dilution or accumulation in the boundary layer, or removal by rainfall. It is necessary to verify the consistency of PMF and CMC results, or discuss the differences. For example, during the HAZE period CMC indicates that dust accounts for 6% of PM2.5, while PMF assigns to dust 19% of PM2.5. Sea salts accounts for 1% of PM2.5 according to CMC, and 17% according to PMF.

Response:

We accept the suggestion and verified the consistency of the PMF and CMC results through multilinear regression between the two methods. Thus, add new section (Section 3.4) to the text manuscript pertaining to this matter:

'3.4 Comparison between CMC and PMF Source

As shown in Fig. 4b and S5, predicted mass modelled by PMF and reconstructed mass by CMC were compared to those measured $PM_{2.5}$ mass. Both approaches resulted with good regression at R^2 = 0.901 and R^2 = 0.784, respectively. Further, seasonal regressions and time series between these two approaches were shown in Fig. S6. The analysis were run on selected components that have similarity to compare: 1) CMC dust vs. PMF factor 2 mineral dust; 2) CMC SIA & K^+ vs. PMF factor 3 SIA & biomass burning; and 3) CMC sea salt vs. PMF factor 5 sea salt.

Overall, 'dust' and 'SIA & biomass' component shows similar trending, as shown in Fig. S5b. Both component have good correlations between CMC and PMF approach (R^2 more than 0.70) except during INT.1 and NE monsoon (R^2 less than 0.50). Rainfall, which was higher during these two seasons compared to other season, could be the reason. The ANNUAL CMC/PMF ratio for seasonal 'dust' component is 0.29 (r = 0.89) while seasonal regression (including HAZE) range between 0.24 and 0.53. The seasonal CMC/PMF ratio for 'SIA & biomass' component ranging between 0.13 and 0.24 with an annual ratio of 0.15(r = 0.87). The inconsistency ratio for both components maybe due to the fact that PMF contribution includes absorbed elements other than selected elements considered by the CMC approach. The 'sea salt' component however, did not show good agreement between two approaches.

However, for HAZE dataset, the two approaches seem to have a very good agreement (r = 0.94) on 'sea salt' component. As shown in Fig. S6c, the seasonal regression of CMC and PMF for sea salt showed that this pair is moderately correlated. Similar observation of large difference on 'sea salt' component (also known as marine aerosol) between two approaches with one of them being the mass closure calculation was seen and discussed by Almeida et al. (2006) and Farao et al. (2014). The observation on this matter was that perhaps CMC calculation did not include all the components associated sea spray and due to reaction of NaCl with inorganic acids (HNO_3 and H_2SO_4) which resulted with the loss of Cl^- ion.

The different estimation derived from the two approaches was expected. According to Harrison et al. (2003), CMC is a hybrid between comprehensive chemical analysis method and simper statistical procedure. It is a simple approach yet effective model to assort the measured PM compounds into different source categories. One of the highlights of this method is that CMC treats sulphate and nitrate separately. This is crucial since different ambient condition can leads to different response of the aerosol which will further affect organic carbon thus secondary organic carbon trend. Moreover, CMC also separate the sea salt and crustal components which have different respond to changes in traffic volume. PMF on the other hand, is an advanced computational tool to identify sources and eventually the mass contribution based on the work by Paatero and Tapper (1994). So, it is likely to have two different results and thus almost impossible to verify results from the two different methods (Hellén et al. 2003; Hopke et al. 2006; Vallius et al. 2008; Vecchi et al. 2008; Favez et al. 2010; Hellebust et al. 2010). This issues have been highlights by Viana et al. (2008) on Europe source apportionment studies. The study stated that it is difficult to obtain coinciding results with different receptor models for the same data. This statement is supported by Vallius et al. (2008) which stated that different methods yield different results when they are applied to air pollution data.'
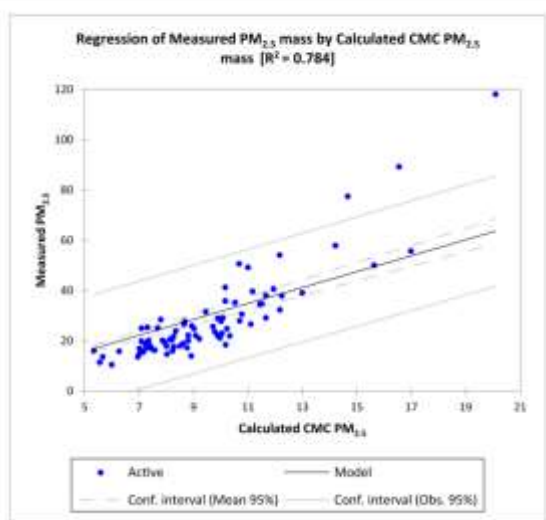
Figure S5. Regression plot between calculated chemical mass closure (CMC) and measured PM$_{2.5}$ mass
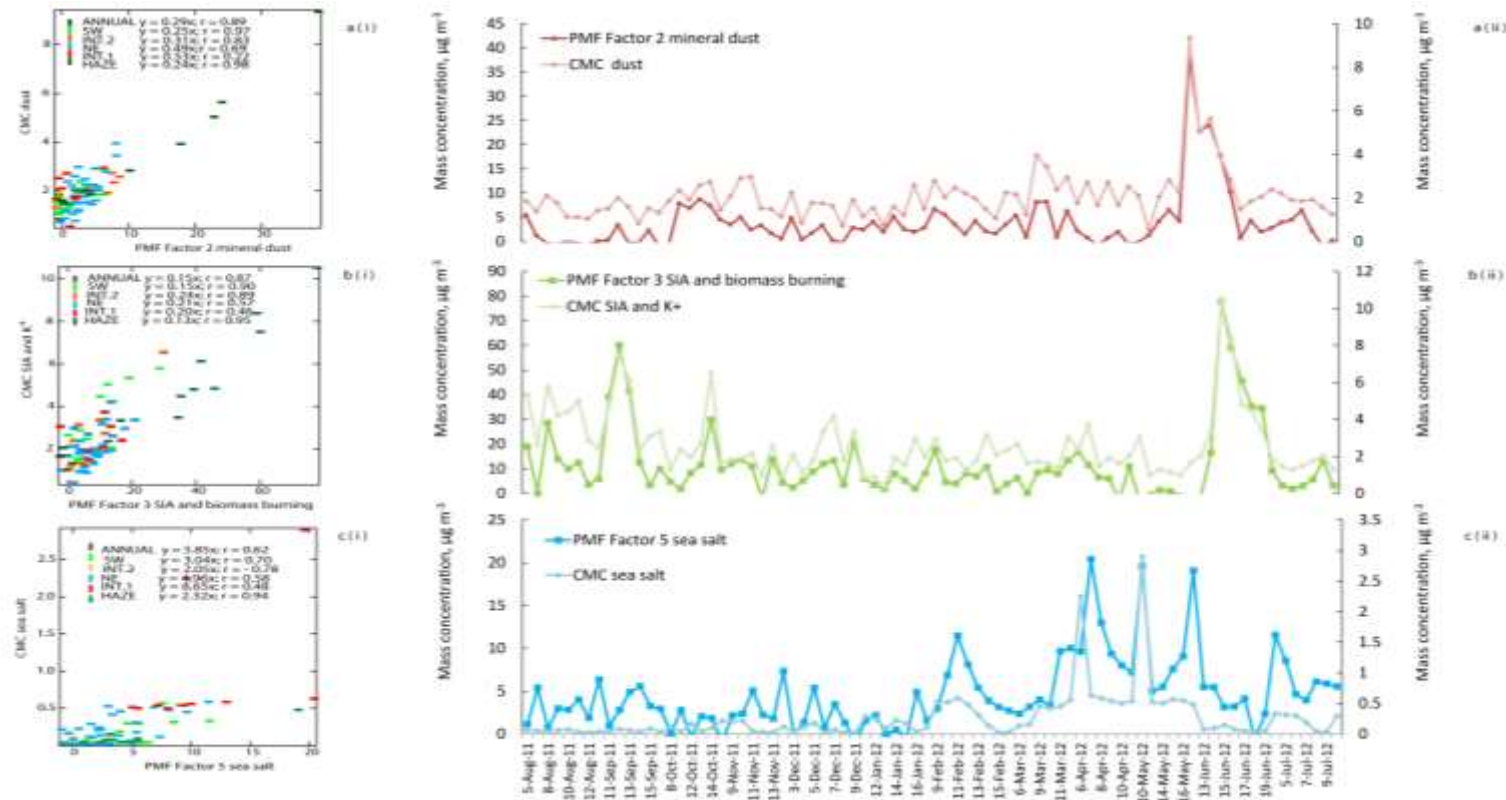
Figure S6. Comparison results between chemical mass closure (CMC) and positive matrix factorisation (PMF) results on three selected factors: a) dust; b) secondary inorganic aerosol (SIA) and biomass burning; and c) sea salt; where i) Correlations between CMC and PMF of $PM_{2.5}$ are on a seasonal and annual basis, each with respective linear regression equations; and ii) time series of the mass concentration predicted by both CMC and PMF approach.

<u>The conclusions state:</u> " The results of our study clearly suggest that chemical constituents and sources of PM2.5 were greatly influenced and characterized by meteorological and gaseous parameters". Although the conclusion is sound, it is in contrast with the discussion at pages 26439-26440 and the phrase: "on a seasonal scale daily PM2.5 mass during all seasons appeared to be affected by the gaseous parameters but not meteorological conditions". The correlation between PM concentration and meteorological parameters, discussed at pages 26439-26440, is actually not the correct approach to investigate the effect of the different meteorological variables. For example rainfall affects PM components removing particles from the air, but also leaves the soil and road surface wet, preventing or reducing the contribution of road dust during the following days, as well. Wind direction is the average wind direction or the prevailing wind direction? The effect of WD on PM could be better investigated looking at the prevailing wind direction (or polar plots) associated to the different PMF factors. The discussion of how meteorological parameters affect PM and PM components should be revised through the text.

Response:

We understand the confusion between the two statements in conclusion and discussion; since some of the factors were greatly influenced by both meteorological and gaseous parameters while some others were only influenced by either meteorological or gaseous parameters. We have modified the text accordingly to show the influence of meteorological-gaseous factor to the $PM_{2.5}$ mass and its composition. However, since we have changed the title of our manuscript to 'Seasonal variability of $PM_{2.5}$ composition and sources in the Klang Valley urban-industrial environment', we now consider the argument on the influence of meteorological-gaseous towards $PM_{2.5}$ correlation suit with the manuscript. Based on new discussion on our main text especially regarding the influence of meteorological factors to PM concentration, we have revised the related information in our conclusion as below:

'These results are connected to the urban-industrial background of the area, where gaseous parameters affect $PM_{2.5}$ mass both annually and seasonally. However, correlation between the chemical constituents and sources of $PM_{2.5}$ towards meteorological and/or gaseous parameters largely varied with different season. Overall, this study suggests that $PM_{2.5}$ and its constituents here in Klang Valley urban–industrial environment, were characterised by the local and regional activities as well as the seasonal tropical change.'

In this study, the average wind direction is used since our PM$_{2.5}$ is a daily measurement. We understand that polar plots can show association of PMF factors to different wind direction. At this stage, we regret to inform that we cannot incorporate the suggestion. However, we will definitely consider this polar plot in our future work and suggested it in our conclusion part.

'The potential source contribution function (PSCF) could also enhance the analysis of local and regional long-range transport. Alternatively, a simple yet effective approach, looking at the prevailing wind direction (or polar plots) associated to the different PMF factors could answer the effect of wind direction on PM.'

Minor comments

Introduction: It would be useful to add some more recent references to the first part of the introduction. In addition, please add some details about the sources identified by previous studies in the area, or similar regions. For example, the author mentioned that previous source apportionment studies have been performed in SEA, but the results are not reported. The introduction could mention which are the most important PM sources we should expect to find.

Response:

We agree with the reviewer. We have included recent references in the manuscript. We have included some details about the sources identified by previous studies in this particular areas and similar region. We also have included important information on the possible sources of PM in this area.

'1 Introduction

Airborne particulate matter (PM) significantly impacts global climate (Jacobson 2002; Vieno et al. 2014; Mallet et al. 2016), causing visibility degradation in both urban and less polluted environments (Diederen et al. 1985; Doyle & Dorling 2002; Watson 2002; Chang et al. 2009; Hyslop 2009) and accelerates material decay (Grossi & Brimblecombe 2002). (Fuzzi et al. 2015) revealed that climate-aerosol interaction, as well as effects of PM on human health and the environment, were underpinned by many new processes and development in the science. Different sizes of PM have been found to have varying toxicities impacting human health (Schwartz et al. 1996; Katsouyanni et al. 1997; Pope III 2000; Ruuskanen et al. 2001;

Eatough et al. 2003; Halonen 2009; Ross et al. 2013; Khan et al. 2016). The fine particles, which are composed of compounds of a range of volatilities, appear to do more harm to human health than coarse particles (Dockery et al. 1993; Schwartz et al. 1996; Laden et al. 2000; Lanki et al. 2006; Pope III & Dockery 2006; Krewski et al. 2009; Tagaris et al. 2009; WHO 2013).

The fraction and composition variability of fine particles (PM$_{2.5}$; particles with an aerodynamic diameter of less than 2.5 μm) are strongly influenced by seasonal meteorological factors, gaseous parameters and location. Megaritis et al. (2014) showed that PM$_{2.5}$ in Europe appears to be more sensitive to temperature changes compared to other meteorological and gaseous parameters in all seasons. Aside from meteorological and gaseous pollutants, seasonal changes and the background of an area (topography and local activities affecting anthropogenic and/or natural air pollution emissions) also influenced the PM$_{2.5}$ chemical variability (Tai et al. 2010; Tai et al. 2012). Seasonal variation of PM$_{2.5}$ mass and its chemical composition for the Asian region has been widely reported. For example, Balasubramanian et al. (2003) reported that Singapore PM$_{2.5}$ mass temporal variability was influenced by a number of factors including changes in emission strength, wind direction (WD) and other meteorological parameters. Also, their chemical mass closure (CMC) components (i.e. soil dust, metallurgical industry, biomass burning and automobiles, sea salt, and fuel oil combustion) at times were significantly attributed to Indonesian forest fires compared to local traffic and industrial emissions. Ye et al. (2003) reported varied CMC elements (ammonium sulfate and nitrate, carbonaceous material, crustal components, potassium) for Shanghai seasons where significant changes in the PM$_{2.5}$ mass were observed with changing season. Meanwhile, sources of PM$_{2.5}$ in Beijing (dust, secondary sulfate, secondary nitrate, coal combustion, diesel and gasoline exhaust, secondary ammonium, biomass aerosol, cigarette smoke, vegetative detritus) showed distinct seasonal trends (Zheng et al. 2005). India PM$_{2.5}$ sources (i.e. motor vehicles, biomass burning, marine aerosol, tyre and brake wear, soil, secondary PM, and others) were observed to have considerable seasonal and weekday/weekend variations (Srimuruganandam & Shiva Nagendra 2012). A study by Louie et al. (2005) on PM$_{2.5}$ chemical compositions showed variations between different locations in Hong Kong where elevated concentrations of a source marker species at a site explained a higher influence of that source. The study identified carbonaceous aerosol as the largest contributor, followed by ammonium sulfate, crustal material, sea salt, and ammonium nitrate. Similar observations were also evident for Indonesia where source

apportionment analysis on the elemental composition of PM revealed different numbers of factors for urban and suburban areas (Santoso et al. 2008).

$PM_{2.5}$ in the atmosphere consists of primary and secondary pollutants including volatile, non-volatile and semi-volatile components which originate from various sources (Eatough et al. 2006). Source apportionment (SA) is an approach that aims to identify and quantify the various sources of air pollutants (Hopke & Song 1997; Watson et al. 2002; Wagstrom & Pandis 2011). The most common method is receptor modelling. Receptor modelling measures atmospheric concentrations of chemically-speciated particles to infer the sources responsible for their emission, or the pathways of formation of secondary particles (Viana et al. 2008). The method starts by collecting and measuring ambient PM at a receptor (location), and works backwards to determine the sources. Receptor modelling uses temporal and chemical variations to separate total PM into different factors, where marker species are used to identify the sources. The goal of receptor models is to solve the chemical mass balance between measured species concentrations and source profiles. One of the models used to solve the chemical mixture is positive matrix factorisation (PMF), first developed by Paatero and Tapper (1993). Subsequently, numerous other studies have employed this method into their $PM_{2.5}$ receptor modelling including many undertaken in the Asian region. For example, Begum et al. (2004) have successfully applied PMF on inorganic and BC datasets to lead to source identification for $PM_{2.5}$ in Bangladesh. Srimuruganandam and Shiva Nagendra (2012) made an evaluation of $PM_{2.5}$ sources for Chennai city, India using only inorganic (elemental) compositions. A study by Zhang et al. (2013) has successfully discussed the seasonal perspective of $PM_{2.5}$ sources (soil dust, coal combustion, biomass burning, traffic and waste incineration emissions, industrial pollution, secondary inorganic aerosol) in Beijing, China using PMF on inorganic and organic datasets. Similar applications of PMF to apportion the sources of $PM_{2.5}$ have also been successfully carried out here in Southeast Asia (SEA). For example, Santoso et al. (2008) used inorganic and BC datasets to identify five major sources of $PM_{2.5}$ as biomass burning, soil, two stroke engine emissions, sea salt, secondary sulfate, motor vehicle emissions, and road dust. A study by Rahman et al. (2011) also used similar chemical compositions for the SA analysis of $PM_{2.5}$ samples from the Klang Valley, which resulted in five sources: two stroke engine emissions, motor vehicle emissions, smoke/biomass burning, soil and industry. PMF was also effectively applied by Khan et al. (2015) to their polycyclic aromatic hydrocarbons (PAHs) dataset to characterise the $PM_{2.5}$ for

the semi-urban area of Bangi, Malaysia. This study revealed three main sources: gasoline combustion, diesel and heavy oil combustion, and natural gas and coal burning. One of the current trends of SA is to apply more than one receptor model, a trend set by a number of countries i.e. Belgium, Germany, Portugal and Spain (Viana et al. 2008). Due to limitations of a single model, applying more than one receptor model will enhance the SA analysis, leading to enhanced characterisation of an element and/or source and thus increasing the confidence in interpretations from the results. The study also reports that the most frequent combinations used for SA are principle component analysis (PCA)-cluster analysis (CA), PCA-Lenschow, PCA-chemical mass balance (CMB), PCA-back-trajectory analysis, PMF-UNMIX-multilinear engine (ME), and CMB-mass balance.

Reid et al. (2013) discussed in detail how the SEA region holds a complex relationship between geographic, socio-economic, meteorological, and aerosol microphysical factors. The review emphasised timing and location of sampling when trying to achieve a representation of the actual condition of the aerosol system, as the urban and industrial aerosol environments differ between urban centres. For example, in Jakarta of Indonesia, two stroke engine vehicles, high emitters of particles and incomplete combustion products, were the major factor. Meanwhile, mobile sources are significant in Bangkok, Thailand, whereas Manila was significantly affected by diesel truck and bus emissions. Having said that, most urban centres in the region share the major sources of meat cooking and oil-gas-petrochemical industry activity as well as shipping influences. In addition, the region is also affected by haze episodes caused by biomass burning. Taking this into consideration, we conducted a one-year assessment of $PM_{2.5}$ covering all four seasons (including haze events) to investigate its variability in the Klang Valley (urban-industrial) tropical environment. The samples were subjected to chemical measurements of inorganic matter (IM) compositions and black carbon (BC). We identified and apportioned the sources to $PM_{2.5}$ mass by employing CMC construction and the PMF-MLR model in conjunction with the cluster analysis of the back trajectory. All variables of $PM_{2.5}$ mass, their chemical compositions identified, as well as the sources predicted, were further analysed using correlation matrices with the meteorological-gaseous pollutants for comprehensive assessment.'

Trace elements: details about the preparation of ICP-MS standard solution can be moved to the supplementary section.

Response:

We accept the suggestion and removed the parts into supplements under the section of:

"Experimental quality assurance and quality control (QA/QC)".

Trace element analysis: Two sets of solutions were prepared for two modes of inductively coupled plasma mass spectrometry (ICPMS) analysis as follows: solution (1) 50 mL stock of concentrated solution for elements with lower weight; and solution (2) further diluted 1 : 4 (50 mL concentrated: UPW) for heavier weight elements. Four point calibration curves were performed for each mode of analysis as follows: mode (1) 10, 20, 30 and 50 ppb for Ag, As, Cd, Cr, Li, Be, Bi, Cs, Co, Cu,Ga, Mn, Ni, Rb, Se, Sr, U and V; and mode (2) 125, 250, 500 and 1000 ppb for Al, Ba, Fe, Pb and Zn. It was ensured that all element regression coefficients were better than 0.999 before the sample analysis was begun and that every 15th sample, the QA/QC were samples run, i.e. the UPW, 1 ppm multi-element standard and standard reference material (SRM1648a). The reference material was 10 mg of SRM1648a Urban Particulate Matter obtained from NIST (National Institute of Standards and Technology, MD, USA) while 1 ppm Multi-Element Calibration Standard 3 (Perkin Elmer Pure Plus, Perkin-Elmer; USA) both prepared the same manner as the samples to test the recovery and validation of the method. All lab ware used for trace element analyses was Teflon material, except for the syringes.

Black carbon. Since the instrument used in the present study is not common to most of the readers, please add some details about BC measurements. For example BC is measured based on optical or thermo-optical properties? Aerosol particles are collected on a substrate, like an aethalometer, or are suspended in the air, like a photo-acoustic instrument? What are the assumptions made for the quantification of BC (for example, which is mass absorption cross sections?)

Response:

We accept the suggestion and thus added some details to the section as per following:

2.3.3 Black carbon

Black carbon (BC) concentration was determined using Smokestain Reflectometer with calibration (Diffusion Systems Ltd.; Model EEL 43M; United Kingdom). In brief, this method

involves measurement of darkness of the stain (on the filter paper) through its reflectance of white light (using a reflectometer). The reflectance is relative to the light reflected by a clean filter of the same material with an assumption of 100% reflection. The absorbed light then converted (through calculation) for BC mass. In this study, five points throughout the filters were taken where the average was then used as the final measured percentage of reflectance for mass calculation. Additional explanations pertaining to this instrument and the calculation involved have been discussed elsewhere (Wiwolwattanapun et al. 2011; Moldanová et al. 2013).

Quality assurance. Avoid to add a paragraph just to mention that QA details are reported in the supplementary.

We accept the suggestion and thus added a sentence on this matter at the end of section 2.3.2, as follows:

'Percentage recoveries are based on SRM1648a Urban PM (National Institute of Standards and Technology, MD, USA) and these varied between 29 and 101 % (Table S2). Details of experimental quality assurance and quality control (QA/QC) for both trace elements and major ions are provided in the Supplement.'

Meteorological and gas measurements. Please specify which analytical techniques were used to measure the gas species. No further details are needed.

Response:

We accept the suggestion and thus inserted the analytical techniques used for the gaseous measurement:

'The instrument and measurement principle used for the gaseous were as follows: O_3 = Analyzer 400A (chemiluminescene); NO, NO_2, NO_X = Teledyne Advanced Pollution Instrumentation 200A (chemiluminescene); SO_2 = Teledyne Advanced Pollution Instrumentation M100A (fluoroscene); and CO = Teledyne Advanced Pollution Instrumentation M300 (non-dispersive infrared absorption).'

The neutralization ratio (page 26442 line 13) takes into account only ammonium and sulfate. Why the other anions and cations are not considered?

Response:

We understand the reviewer concern. In this section, we only calculate the neutralization ratio covering three elemensts i.e. ammonium, sulfate and nitrate. The reason being, both ammonium and sulfate are two major cation and anion which have significant concentration in this study. Other parameters do not show significant results. Having said that, we did utilise other elements in other section accordingly i.e. ratio of Mg^{2+}/Ca^{2+} and Cl^-/Na^+ in other section i.e. section 3.3.5 pg 26452 line 18-21.

Technical corrections.

Please remove "comparatively" from sentences like comparatively higher than To refer to tables and figures, the author can write table x reports or figure x shows. Do not write just the table or number figure out of a sentence.

Response:

We accept the suggestion and thus revised all the necessary (deleted comparatively; add Table shows at front of sentence compared to previous just write up no of table and figures at the end of sentence) as follows:

Page 26436 line 29 – Page 26437 line 4: Deleted 'comparatively'; and revised sentence

'Table 2 reports that $PM_{2.5}$ mass average for this study was very low compared to other big cities of Asia i.e. in India and China (Balakrishnaiah et al. 2012; Huang et al. 2013; Pachauri et al. 2013; Zhang et al. 2013) but variable when compared to other parts of the world (Dongarrà et al. 2010; Yin et al. 2010; Bressi et al. 2013; Squizzato et al. 2013).'

Page 26437 line 8-10: Deleted 'comparatively'

'Furthermore, our result for Petaling Jaya is higher than other parts of Peninsular Malaysia (Tahir et al. 2013; Ee-Ling et al. 2015).'

Page 26437 line 11-13: Deleted 'comparatively'; and change 'is' to 'was'

'The $PM_{2.5}/PM_{10}$ ratio at this site was higher than other studies in Asia as reported by Hopke et al. (2008) where most of the sites studied showed ratios of lower than 0.5.'

Page 26437 line 19-21: Deleted 'comparatively'; in accordance with other comments

'Our $PM_{2.5}/PM_{10}$ ratio was also in agreement with other cities in Europe (Gehrig & Buchmann 2003; Gomišček et al. 2004; Contini et al. 2014).

<u>Page 26444 line 23-25:</u> Deleted 'comparatively'

'On a regional scale, our results here are low compared to most other SEA cities as reported by (Reid et al. 2013).'

<u>Page 26444 line 23-25:</u> Deleted 'comparatively'

'The portion of our IM and BC were also low compared to the previous study of the site by Keywood et al. (2003) with 28 and 30% (normal days), respectively.'

<u>Page 26430 line 23:</u> Revised sentence

'The percentage recovery for all elements was between 86 and 131%, as reported in Table S2.'

<u>Page 26431 line 18-20:</u> Revised sentence

'Percentage recoveries are based on SRM1648a Urban PM (National Institute of Standards and Technology, MD, USA) and these varied between 29 and 101%, as reported in Table S2.'

<u>Page 26437 line 10-11:</u> Revised sentence

'The mean $PM_{2.5}/PM_{10}$ ratio for the site was 0.72 ± 0.18 and the ratio for $PM_{2.5}/TSP$ was 0.46 ± 0.13, as reported in Table 1.'

<u>Page 26438 line 19-20:</u> Revised sentence

'Referring to Table 3, the Pearson correlation revealed that $PM_{2.5}$ mass on an annual basis was significantly influenced by meteorological and gaseous parameters.'

<u>Page 26440 line 3-5:</u> Revised sentence

'As mentioned earlier, Table 1 reported that the $PM_{2.5}/PM_{10}$ ratio for both major seasons (SW and NE) were almost the same at ~0.70.'

<u>Page 26442 line 12-14:</u> Revised sentence

'Also known as the acidity ratio, the neutralisation ratio (NR) was calculated to further investigate the acidity of the atmospheric aerosols, as reported in Table S1.'

<u>Page 26445 line 19-22:</u> Revised sentence

'Referring to Table 4 and Fig. 4a, the PMF 5.0 model resolved five factors, identified as: 1) combustion of engine oil; 2) mineral dust; 3) mixed SIA and biomass burning; 4) mixed traffic and industrial; and 5) sea salt;'

Page 26446 line 25-26: Revised sentence
'Factor 1 seems to not be particularly affected by gaseous parameters or meteorological conditions, as reported in Table S3.'

Page 26452 line 12-13: Revised sentence
'As shown in Table 4 and the time series illustration of Fig. 6c, the sea salt factor is seasonally high during INT.1 (45 %), April until early May.'

Page 26453 line 15-16: Revised sentence
'However our investigation as reported in Table S4, revealed that the two episodes to have quite different characters.'

Page 26453 line 23-25: Revised sentence
'As reported in Table S5, the PMF factor 3 of mixed SIA and biomass burning was further investigated through a correlation matrix between CMC and the source for a better understanding of the composition/characteristics.'

Page 26428 line 5-7: Revised sentence
'As shown in Fig. 1, the sampling took place on the rooftop of the Malaysian Meteorological Department (MET) located in the city of Petaling Jaya (MET PJ; 3°06'09.2"N 101°38'41.0"E), about 100 m above the sea level.'

Page 26435 line 18-20: Revised sentence
'As shown in Fig. 2b, 2d and 2e, strong variability can be observed from the monthly and days averages of $PM_{2.5}$ mass.'

Page 26436 line 4-6: Revised sentence
'Fig. 2c showed that during the NE monsoon, only 17% exceedance of the daily WHO guideline was recorded while for three other seasons, more than 50% exceedance of the daily WHO guideline was recorded.

Page 26436 line 17-19: Revised sentence

'As shown in Fig. 2a, it is important to note that haze events always occur during the SW monsoon, thus it is anticipated that they will directly affect the SW overall mass concentration (PM$_{2.5}$).'

Please replace > with "less than" through the manuscript. For example at line 15: season with less than 50% sample exceedance.

Response:

Page 26424 line 13-15: Changed '<' to 'less than'

'The North-East monsoon (NE) was the only season with less than 50 % sample exceedance of the daily WHO guideline.'

Page 26424 line 20-23: Changed '>' to 'particle bigger than'

'Further analysis on the PM$_{2.5}$/PM$_{10}$, PM$_{2.5}$/TSP and PM$_{10}$/TSP ratios reveal that meteorological parameters only greatly influenced the coarse particles (particles with an aerodynamic diameter of greater than 2.5μm) and less so the fine particles at the site.'

Page 26428 line 26 to Page 26429 line 1: Changed '> ' to ' more than'

'In this study, air pollution episodes are defined considering PM2.5 mass (more than 40 μgm−3) and the Air Pollution Index (API) (more than 50), hereafter defined as HAZE samples.'

Page 26432 line 13-15: Changed 'dp < 10 μm' with 'particle with aerodynamic diameter less than 10 μm'

'Daily rainfall readings, daily PM10 (particle with aerodynamic diameter less than 10 μm) and TSP (total suspended particulate) mass (high volume sampler) were obtained from MET of Petaling Jaya recorded at the sampling site.'

Page 26438 line 2-4: Changed 'dp < 10 μm' with 'particle with aerodynamic diameter less than 10 μm'

We make changes accordingly pertaining to other comments:

'The similar PM$_{2.5}$ to PM$_{10}$ ratio during the wet and the dry season indicates that meteorological parameters, specifically rainfall, are affecting the fine (particle with an

aerodynamic diameter of less than 2.5μm) and coarse (particle with an aerodynamic diameter of greater than 2.5μm) particles in the same way.

Page 26425 line 23 "particle with aerodynamic diameter less than 2.5 μm" instead of "dp<2.5 μm"

Response:

Changed from 'dp<2.5 μm' to 'particle with aerodynamic diameter less than 2.5 μm'

Page 26427 line 20. Reid et al. instead of a study by Reid et al.

Response:

Revised the reference from 'a study by Reid et al.' to 'Reid et al….'

Page 26427 line 28. "Cluster analysis of back trajectory" instead of "Trajectory cluster"

Response:

Changed from 'trajectory cluster' to 'cluster analysis of back trajectory'

Page 26429 line 3-7 please verify the number of decimal digits.

Response:

We have made all changes accordingly; using until 3 significant figures throughout the text.

Page 26429 line 8 during SW monsoon

Response:

Changed from 'during SW' to 'during the SW monsoon' and also 'during NE' to 'during the NE monsoon'

Page 26429 line 13 The aerosol sampling

Response:

Corrected the term from 'The sampling' to 'The aerosol sampling'

Page 26430 line 4 "loaded quartz filter" instead of "loaded filter paper"

Response:

Changed from 'loaded filter paper' to loaded quartz filter'

Page 26433 line 20 PM mass instead of filter mass.

Response:

Corrected the term from 'filter mass' to 'PM mass'

Page 26435 line 3 did you mean missing values?

Response:

Yes, we meant 'missing values'. We understand it can be confusing. Therefore, revised the term from 'undetected data point' to 'the missing values'; in conjunction with latest version of the section.

Page 26436 line 23 higher compared instead of higher ompared

Response:

Corrected the typo from 'higher ompared to' to 'higher compared to'

Page 26439 line 6 A correlation with r=0.29, corresponding to r2=0.08 cannot be considered significant. Revise the adjectives used to define the correlations through the text.

Response:

Even though r and r^2 value are low, statically the correlation is still significant, referring to *p*-value ($p < 0.01$).

Page 26443 line 14 dust was instead of dust as

Response:

Changed from 'dust as' to 'dust was'

Page 26447 line 2 correlate with gaseous parameters instead of influenced by with gaseous parameters

Response:

We accept the suggestion and revised the sentence:

'For gaseous parameters, factor 1 seemed to correlate with gaseous parameters mostly during the NE monsoon, with significant positive correlations with CO (r = 0.498; p = 0.005), SO2 (r = 0.436; p = 0.016), NOx (r = 0.471; p = 0.009) and NO2 (r = 0.529; p = 0.003).'

Page 26449 line 9-12 revise this section. It is not clear.

Response:

We agree with the reviewer and thus revised the sentences to the following:

'Except for INT.1, the other seasons show very significant correlations between this factor and secondary aerosol components, i.e. SO_4^{2-}, NH_4^+ and K^+. During HAZE, this factor contributed 63% of the $PM_{2.5}$ mass.'

Page 26454 line 26 page 26455 line 1. The weekly trend does not look significant enough to discriminate between days with higher concentration (Fridays) and days with lower concentrations (Wednesdays).

Response:

We agree with the reviewer and deleted the sentence in the conclusion.

Figure 4a. Add factor labels to the panels.

Response:

We accept the suggestion to include the factor names in the figure caption. The revised figure are as below:
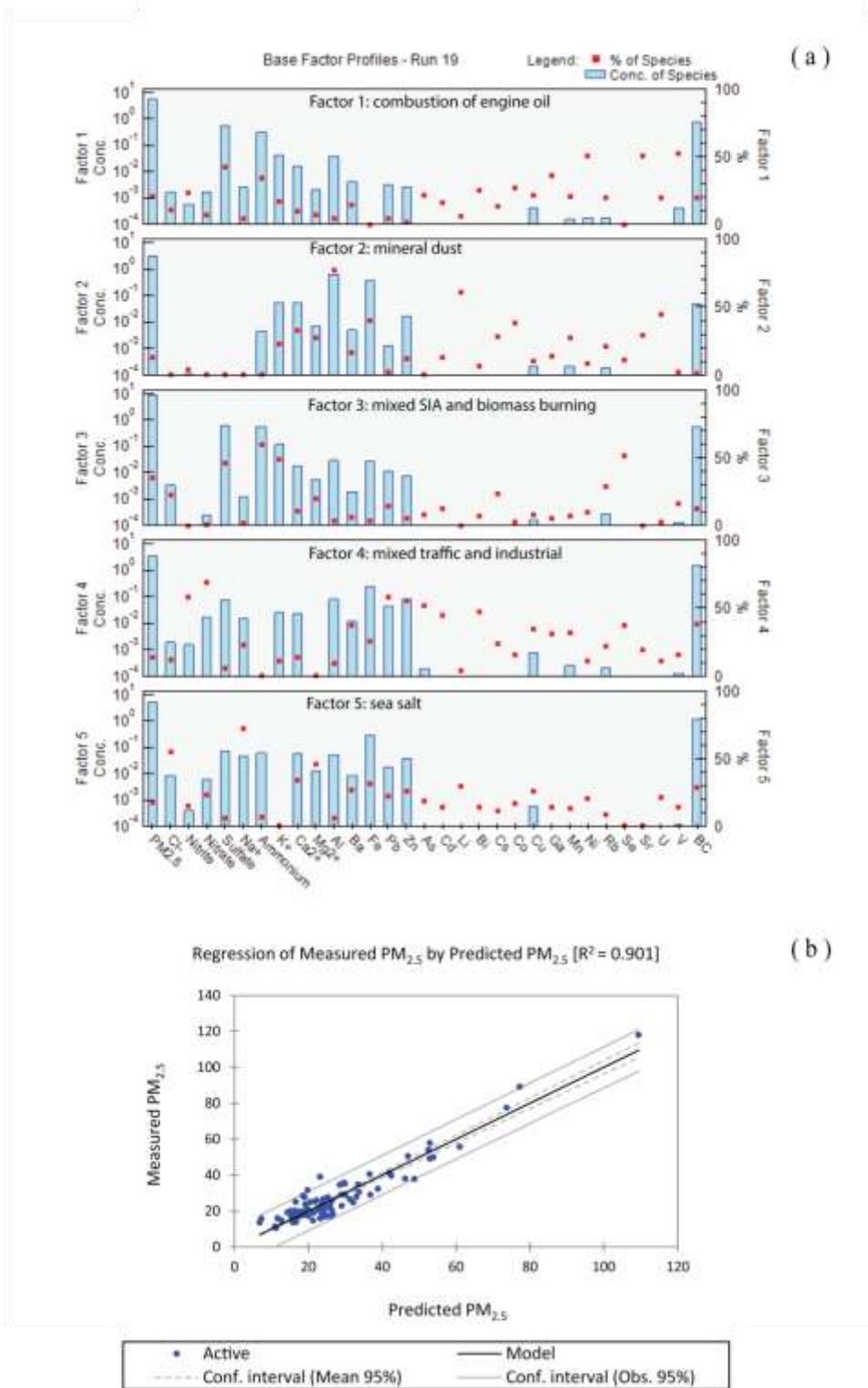
Figure 5. Use different scale to report meteorological parameters because it is impossible to read variation in temperature and difficult to appreciate the variability of RH.

Response:

We accept the suggestion and thus revised the figure as per following: