Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-721-AC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diagnosis of dust- and haze pollution-impacted PM_{10}, $PM_{2.5}$, and PM_1 aerosols observed at Gosan Climate Observatory" *by* Xiaona Shang et al.

Xiaona Shang et al.

meehye@korea.ac.kr

Received and published: 25 December 2018

Correspondence to Referee #2

Thank you very much for your thorough and constructive comments on our manuscript acp-2018-721, entitled "Diagnosis of dust- and haze pollution-impacted PM10, PM2.5, and PM1 aerosols observed at Gosan Climate Observatory". We added all available information to provide more solid evidences and revised the manuscript according to your comments. The response is given to each comment. In the revised manuscript, changes are colored in blue and page and line numbers are given for the revised manuscript.

General Comments

Comment 1: As for principle component analysis, three factors explained 71% of the total variance. It means 29% of the variance was not explained. The residue was quite large. What's the possible contribution for this unexplained part? Response 1: As you pointed out, the residual is about 30%. In general, the two factors of the largest distance in eigenvalue are chosen for PCA, which account for more than 60% of the total variance in this study (Figure given below). The reason for we selected three principle components is not only that the 3rd factor (PC3) is in charge of 9 % of variance, but also that PC3 exhibits a clear increasing trend with time (for details of increasing trend see Response 2). In addition, PC3 loadings were discernible during warm season, in contrast to other two components that were dominated in cold season. The contribution of the rest components was much less than 10 %. For instance, the fourth component (PC4) accounts for 6 % and indicates salts influence by high loadings of Na+ and Cl–. The rest components contributed no larger than 5 %. This information is added to Supplementary Information (SI 3).

SI 3. Eigenvalues according to the number of principal components.

Comment 2: Because the number of the tracer was limited and the tracers were lack of uniqueness, the results about source apportionment were not so robust. For example, PC3 was dominated with NH4+ and Ca2+, while secondary formation of NH4+ might also lead to the correlation between NH4+ and Ca2+. Response 2: There is no fancy item in the measurement list. Nevertheless, you agreed to review this manuscript and gave constructive comment. We really appreciate it. In order to understand the factors determining the variation and magnitude of particulate matters in the study region, we had measured major aerosol constituents in three size-cuts for a long period. This is we believe the uniqueness of this study that led us to main findings.

NH4+ concentration used to be in good correlation with PM mass, SO42– or NO3– . NH4+ is a neutralizing compound of aerosol and is so Ca2+, which is an indicator for the impact of Asian dust in the study region. NH_3 concentration is the highest due to fertilizer application in spring and summer, and enhanced due to anthropogenic emissions in winter. On the other hand, the source strength of Ca^{2+} is the greatest in spring because of the sandstorm transported by northerly and northwesterly wind. NH_4^+ is possibly correlated with Ca^{2+} due to secondary formation but in association with SO_4^{2-} or NO_3^-.

In previous studies, Sulfur Oxidation Rate (SOR) = [$nSO_4^{2-}/(nSO_4^{2-}+nSO_2)$] and Nitrogen Oxidation Rate (NOR) = [$nNO_3^-/(nNO_3^-+nNO_2)$] was found to be high during summer (n represents molar concentration), which indicates the efficient conversion of precursor gases to secondary ions in particles. In this study, the SOR and NOR were not increased with time like PC3 loadings presented in the following figure (R^2 = 0.53), implying that there was no significant relation between PC3 and secondary formation. To avoid the confusion, therefore, the relevant discussion was added in the revised manuscript.

Time series of SOR, NOR, PC3 scores, and linear fitting of PC3.

Page 9 line 236-237: "These results indicated the agricultural influence on PC3 loading due to fertilizer usage."

Comment 3: The time series of mass concentration for each PC should be given after the PCA data. This is important for understanding the sources of PM. Response 3: According to your suggestion, we provide the time series plot of PC loadings to the Supplementary Information.

SI 4. Time series of scores for each principal component.

High scores of PC1 along with the high SO_4^{2-} and NO_3^- concentrations were observed particularly in winter, confirming that PC1 represented the influence of anthropogenic pollution sources (Zhang et al., 2013b). In comparison, PC2 loading was high for Ca^{2+}, Mg^{2+}, Na^+, and Cl^- in spring and well correlated with OC4 concentration in

$PM_{2.5}$ and PM_{10}, which used to be elevated upon dust events (Lim et al., 2012). It is noteworthy that PC3 loading was high in spring and summer and increased with time, reaching to the highest in 2010. In addition, NH_4^+ concentration was moderately related to PC3 loadings in $PM_{2.5}$ and PM_{10} with low concentrations of particulate matter, indicating the agricultural influence on PC3 loading due to fertilizer usage.

Comment 4: 4. It was unclear about the diagnosis of dust and haze based on frequency distribution. There was no robust criterion to differentiate haze from dust events. As shown in Fig.4, the mass loading of PM on dust or haze events varied greatly. Response 4: First of all, it is not the purpose of this study to differentiate between haze and dust. In Korea, the reference concentrations of PM_{10} and $PM_{2.5}$ have been established for warning or air quality forecast. However, there are no criteria for mass concentrations of haze or dust occurrence, which was the motivation of this study. The result of this study reveals that the GCO PMs are under consistent influence of dust, haze, or the two being mixed. The samples of PM_{10}, $PM_{2.5}$ and PM_1 were concurrently collected approximately every 6–8 days during the five years. As a result, our samples do not cover all officially issued haze and dust events, thereby being suitable for diagnosing the effect of haze and dust on particulate matters.

We also tested if these criteria were valid for the recent measurements at Gosan from January 2016 to October 2017. $PM_{2.5}$ measurement officially began in 2015 and the hourly measurements of PM_{10} and $PM_{2.5}$ are available through http://www.airkorea.or.kr/. During this period, dust and haze events were observed for eight and twelve days, respectively, for which daily average concentrations are presented in Figure 5. As shown in Figure 5, all dust and haze events are found above the mean+SD of PM_{10} (52 $\mu g/m^3$) and $PM_{2.5}$ (32 $\mu g/m^3$), respectively, even for the recent two years. It demonstrates that the criteria suggested in this study are robust and useful to diagnose the effect of dust and haze impacted particles.

Figure 5 is replaced with the one shown below in the revised manuscript.

Figure 5. Frequency distributions of PM10, PM2.5 and PM1 mass concentrations for all measurements. Mass concentrations are given as ln values in x-axis. The green lines stand for mean+SD. The individual samples collected during dust or haze events are marked as different symbols along the x-axis. For comparison, added right below the x-axis are the daily average concentrations of haze and dust days during January 2007 ∼ October 2012 (http://www.airkorea.or.kr/).

All figures are in the file uploaded in Supplement.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-721/acp-2018-721-AC1-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-721, 2018.

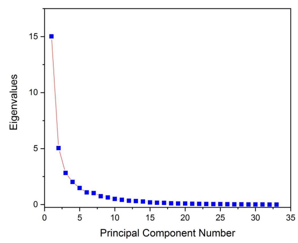

Fig. 1.

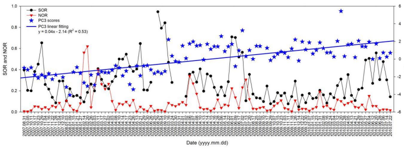
SOR
NOR
PC3 scores
PC3 linear fitting
y = 0.04x - 2.14 (R^2 = 0.53)
SOR and NOR
PC3 scores
Date (yyyy.mm.dd)

Fig. 2.

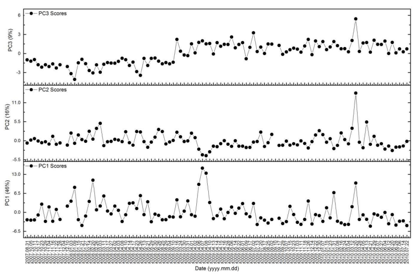
PC3 Scores
PC3 (9%)
PC2 Scores
PC2 (16%)
PC1 Scores
PC1 (46%)
Date (yyyy.mm.dd)

Fig. 3.

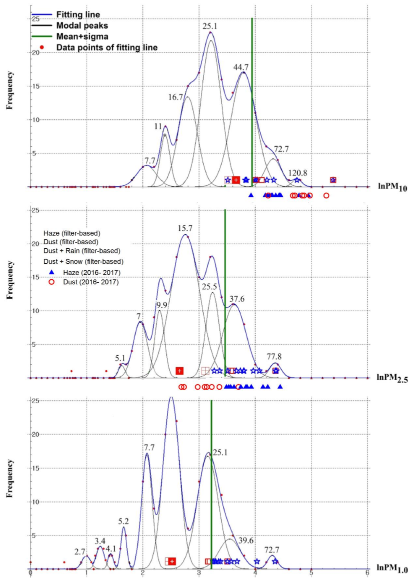
Fitting line
Modal peaks
Mean+sigma
Data points of fitting line
Frequency
lnPM10
Haze (filter-based)
Dust (filter-based)
Dust + Rain (filter-based)
Dust + Snow (filter-based)
Haze (2016- 2017)
Dust (2016- 2017)
lnPM2.5
lnPM1.0

Fig. 4.