

"Measurement report: Spatiotemporal and policy-related variations of PM2.5 compositions and sources during 2015-2019 at multisite of a Chinese megacity"

Feng et al. present a measurement report on a PM2.5 filter data set that covers the years 2015-2019 at ~19 stations in the urban area of Chengdu, China. This is a valuable dataset, which displays the transformation of PM2.5 mass and composition in a rapidly developing urban area. Therefore, it is certainly within the scope of ACP. The general message is that regulations on coal combustion take effect within these five years, while PM2.5 due to traffic emissions and secondary particles increases. This is an important finding and worth publishing. However, in the current state, I cannot recommend the manuscript being published for several reasons: (1) the manuscript requires heavy language editing. Under "technical notes" I listed a few mistakes, I could have listed more, (2) the major message of the paper is that PM2.5 decreased between 2015 and 2019, but this is only based on the average of a large number of filter samples – no test for statistical significance (see major comment on chapter 3.1), (3) the methodology part is far from being complete – reproduction by fellow scientists would be impossible, (4) a few labels of clusters between figures and text are mixed up, (5) some of the data are interpreted in a highly speculative manner, often not convincingly supported by the data (see comment on chapter 3.2.1).

Overall, I conclude that the manuscript would need a major revision in order to meet the standards of ACP.

Major comments:

I. 22: A mass ratio is not necessarily an indicator for emission reduction. What is with the absolute values of sulfate and nitrate?

I. 25: A large fraction of secondary particles → this cannot be attributed to individual sources, such as traffic, coal combustion, industrial emissions? E.g. secondary sulfate can partly originate from coal combustion.

I. 28: What do you mean by "stronger distribution patterns."?

Chapter 2.2: there is very little information provided how the chemical analysis has been done in detail, especially for the ion chromatography and for the ICP-AES part. E.g. I miss the information what mobile phase (e.g. which buffer) was used, which column? Was a suppressor involved? How did you calibrate?, etc. For the ICP-AES analysis, there is no explanation on how the acid digestion was conducted and how the system was calibrated. Although mentioned in the header, I find no information on quality assurance and quality control.

Therefore, the experimental information provided is not sufficiently complete and precise to allow the reproduction by fellow scientists (ACP review criterion no.6).

Chapter 3.1.: The evolution of the average PM2.5 concentration is presented for the years 2015-2019, however, I miss the presentation of the spread of the PM2.5 concentration of the individual filter samples. As a total of more than 800 PM2.5 samples were analysed for this study, it would be interesting to see a histogram of the PM2.5 distribution for each year, showing all PM2.5 masses of every single filter. Based on such solid statistics that is provided with >800 samples, one could also make a statement on the significance of the PM2.5 reduction from 2015 to 2019. Without the presentation of the spread of the individual sample data, I have a problem seeing a trend in the data from the years 2016-2019, and **I cannot say that the results are sufficient to support the interpretations and conclusions (ACP review criterion no.5).**

I.243-246: the information given in the text do not agree with the labelling in Figure 4. "cluster 1 (C1) included all the sites in 2015", while in Figure 4, C1 shows the years 2018 and 2019.

I. 245: How many samples are averaged behind 2016DJY3? What is the distinct pollution feature at this station? Based on Figure 4B, I cannot see a distinct chemical profile of C4, compared to the other clusters (may be a bit more ammonium, less chloride, more heavy metals?).

Chapter 3.2.1: The statements in this chapter appear to me as highly speculative. E.g. a high OC fraction does not necessarily mean that this OC only stems from fuel combustion and biomass burning. Do industrial processes not emit VOCs at layer 3? What is the role of biogenic emissions in layer 3?

Furthermore, it is stated "It's interesting to find that the spatial clustering in each year was generally consistent with the classification of three layers." – I don't see that in Fig. 5: for example, for year 2018, cluster 2 appears in layer 3 and in layer 1; year 2017, cluster 4 appears in layer 2 and layer 3.

I. 282-284: The relative contribution of sulfate decreased, okay, but what about the evolution of the absolute concentration of sulfate, nitrate and chloride in these years? I cannot find these numbers. Maybe the relative fraction only decreases, because other compounds (e.g. secondary organics) increased from 2015-2019?

Minor comments:

I. 75: "A total of 836 samples were collected in 19 sites [...]" → this information rather belongs to chapter 2.1.

I. 113: unit of sampling is likely $L \text{ min}^{-1}$, not min L^{-1} . Otherwise 100 min L^{-1} would be really slow.

Chapter 2.5.: A comment on the specific choice of the distance metric (cosine) and linkage function (average) would be helpful. Is the result with this particular choice robust, such that other distance and linkage functions lead to the same/ a similar result?

I. 335: "The effectiveness of the SWPSCF method was well-evaluated during the investigation." How?

I. 386: Hysplit and meteorological data are not all presented data. Please add a data repository with all presented data, or explain how to get these data.

Technical notes:

General: The term "layer" to describe a specific area appears to me as a bit odd. With layers I associate rather vertical stacked "layers".

I. 13 + 19 (and throughout the manuscript): better use singular of "compositions"

I. 13: "at multisite." ?? → at multiple sites.

I. 22 + 23: but dropped to less than 1.

I. 29: were occurred → occurred.

I. 45: small number of literatures → better: small number of publications

I. 61: consider rewriting the sentence “For PSCF method, due to the sources showed...”. Eventually make two or three sentences out of it, for a better reception of the message.

I. 64: would be developed in this work → has been / was developed in this work, which combines [...].

I. 91: “time interval of highly polluted vehicles”? You likely mean highly polluting vehicles, but what do you mean by time interval? Time interval of vehicle registration? Please specify.

I. 108: may not be fully consistent...

I. 132: The sentence is missing a verb.

I. 137: extracting → extraction.

I. 165: chemical compositions → better write “chemical species”.

I. 245: DJY3 is only one station → the only site far from the other sites.

I. 337: are shown in Fig. 8