Comments on acp-2018-914 "Unraveling the role of silicon in atmospheric aerosol secondary formation: A new conservative tracer for aerosol chemistry"

General comments: In the Earth's crust, silicon (Si) is ubiquitous and relatively inert element. The Si also existed in atmospheric aerosols. In this article, the author reports the role of silicon (Si) in the secondary aerosol formation. They found uncorrelated of the total mass of Si in PM2.5 with the SA formation. Therefore, they suggested Si as a new conservative tracer for estimating the SA contribution to PM2.5. Furthermore, the author also correlated the Si isotopic signatures with SO_4^{2-}, NO^{3-}, NH_4^+, and SOC, then deduced the sources of these secondary components. This is a new method in estimating the secondary aerosol contribution to PM2.5 by using the Si as a tracer. It's meaningful for the atmospheric chemistry. However, there still some comments need to be solved before the manuscript can be published. The author should consider that chemical species of secondary aerosols (e.g., NH_4^+, NO^{3-}, SO_4^{2-}, SOC) as tracers are able to get the detail information of SA compare to use Si as a single tracer, what is the superiority of this method?

Detail comments:

2.2 Sampling of PM2.5 samples

(1) "the PM2.5 samples were collected around Beijing on random haze days (n = 100) in 2013". The author should give more detail information about the samples, such as the concentrations of PM2.5, meteorological parameters, etc. These environmental conditions can help the reader to understand the latter results and discussion more clearly. These are also very important for the conclusion especially the contribution of SA in PM2.5 during this article.

(2) What is the meaning of "random haze days (n=100)"? Can these samples represent the secondary formation of particles?

(3) The sampling sites should be given a brief introduction. For example, what the feature around the sampling sites? What the location of the sites?

2.6 Uncertainty analysis in the estimation of secondary aerosols

(1) Emission Inventory may introduce significant uncertainties because of uncertainties in activity-related data and emission factors. At least, the authors should state clearly the effect of emission inventory so that the readers can judge by themselves. Furthermore, "assuming that the relative emission ratios of primary sources are steady" what's the meaning? The high temporally resolved emission inventory could be used in Table 1, other than the total emissions mass. For example, the monthly emission mass from emission inventory could be used other than yearly emission mass.

From 3.1 to 3.2, the author correlated the Si with the secondary species in PM2.5, the secondary precursors, and relative humidity. There are some comments about this part:

(1) In the part of Methodology, the author stated that random haze days n=100, in this part, n=63. What the meaning of "random haze days"? why these days were used to do the analysis? The author found the PM2.5 concentration showed a clear seasonal trend. How was the representative of the data in the article? Have the author compared their results with references?

(2) All the samples were collected during haze days, why they don't collect some samples during clean days before these haze days? The comparison from clean days to haze days can help to give more information during the formation of SA.

3.4 Secondary aerosol estimate using the Si-dilution method:

(1) The uncertain caused by atmospheric transport should be considered in this study.

(2) Some special episodes should be analysis to support the views and better understand for the readers in this work.

3.5 Comparison with the traditional method

The author stated that the "traditional method bring in huge uncertainties to the result". Then the author compared the Si-dilution method with the traditional method, "the results between the two methods were actually very close". Why?

3.6 Sources of secondary particle precursors revealed by Si isotopic signatures

The discussion in section 3.6 is too simple and less persuasive. The author should give more detail analysis of even one haze case process. Furthermore, there are many works

have been done about the pollution case in the year 2013 in Beijing. The author should add more comparison between their results with the references.

3.7 Implications for air pollution control policies

Huang's results were based on the observation during the high pollution events of 5–25 January 2013 at the urban sites of Beijing. The SA contribution obtained in this article was 88.7% for Jan 2013. This result deduced from just about five days daily data (6/1/2013, 11/1/2013, 18/1/2013, 25/1/2013, 29/1/2013), can these data have the representative for 20 days from 5-25 Jan 2013? The SA contribution obtained was 79.2%, what's the uncertainty of these results?

Besides, there are some grammar and spelling mistakes. The format of some reference should be checked, such as line10 in section 3.3, Line12-13 in section 3.3.