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Title: Springtime carbon emission episodes at the Gosan background site revealed by total carbon, stable carbon isotopic composition, and thermal characteristics of carbonaceous particles

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Responses to the reviewer's specific comments and questions;

Reviewer #2 (Comments):

This paper shows very interesting use of 13C isotope data and thermal characteristics of the carbonaceous aerosol fraction to explain the origin of carbonaceous aerosol on a small island in Korea. It is one of the most conclusive studies using 13C data on aerosols that I have seen so far and the subject is definitely relevant for ACPD. There are some issues that require clarification and a few points where the interpretation of the data seems questionable, which will be discussed below. Since these points do not affect the main conclusions of the paper I suggest publication, subject to minor revisions.

Major comments

1) The division and subdivision of the different episodes needs to be described more clearly. Reading through the whole paper, this is what I think was done, although I am not completely sure:

(a) First a general subdivision of cases with elevated carbon concentrations (=carbon episodes) using a cutoff of 10ug/m3. Then the carbon episodes were further subdivided in

b) Pollen cases (identified by high loadings of pollen present on the filter)

c) LRT (identified by elevated PM10 concentrations?)

d) LRT + dust (identified by elevated PM 10 concentrations + high Ca + low alpha?)

Response: Mass concentrations of ammonium, nitrate, and sulfate have been added in Table 2. The categorization of the haze episodes for the AD episode has been modified as AD+LTP (Asian dust plus long-range transported pollution).

Following sentences have been added. Please see lines 297-305 in the revised MS.

"Subdivision of the carbon episodes was conducted as follows. Pollen episodes were identified by the elevated concentrations of citric acid and pollen in the TSP samples as described by Jung and Kawamura (2011). Even though the TC value (7.5 μgC m^{-1}) for the KOS612 sample during

the pollen episodes was lower than the threshold value, we included this sample to the carbon episode for comparison of different episodes. LTP episodes were identified by the elevated concentrations of nitrate and sulfate during the carbon episodes. AD+LTP episodes were identified by the elevated concentrations of Ca^{2+} and low aerosol Ångström exponents as well as the elevated concentrations of nitrate and sulfate during the carbon episodes."

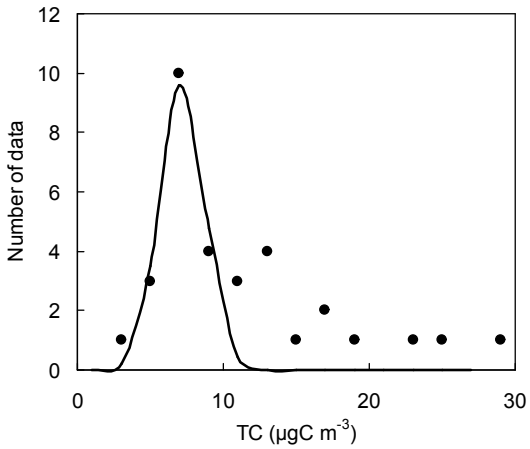
I think it necessary to be more clear on the following issues:

(a) Cutoff of 10ug/m3 for carbon episodes: (i) why was this cutoff chosen?

Response: Following sentences have been added. Please see lines 288-294 in the revised MS.

"The frequency distribution of TC mass concentrations at 2 μgC m^{-1} increments is shown in Fig. 2 with a peak value in the rage of 6−8 μgC m^{-1}. Gaussian fit of the frequency distribution showed a peak center at 7.2 μgC m^{-1} with the width of 3.1 μgC m^{-1}, representing background TC mass distributions during the entire sampling periods. The Gaussian fit was clearly separated from the total TC distribution with a threshold value of 10 μgC m^{-1} (Fig. 2). Thus, this study defined the carbon episode as an average mass concentration of TC > 10 μgC m^{-1}."

Following figure has been added in Fig. 2.



(ii) If this was the main criterion, why is there a "pollen episode" in figure 5 with a TC concentration < 10ug. Does this mean the presence of pollen took precedence over the criterion of 10ug/m3.

Response: Following sentences have been added. Please see lines 299-301 in the revised MS.

"Even though the TC value (7.5 μgC m^{-1}) for the KOS612 sample during the pollen episodes was lower than the threshold value, we included this sample to the carbon episode for comparison of different episodes."

Where the other non-episode cases pollen free?

Response: Following sentence has been added. Please see lines 371-374 in the revised MS.
"Mass concentrations of citric acid during the non-episodes (range: 0.17 − 18 μg m^{-3}) were several dozen times lower than the pollen episodes (range: 20 − 320 μg m^{-3}) and almost no pollen grains were observed from the microscopic image of the TSP samples."

(iii) Did the non-event cases also sometimes have back trajectories from China or did the back trajectories come from elsewhere?
Response: Air mass backward trajectories during the non-episodes have been added in Fig. 4d. Following sentence has been added. Please see lines 376-378 in the revised MS.
"Air mass backward trajectories during the non-episodes mainly originated either from the northern part of China or the western North Pacific Ocean (Fig. 4d)."

(b) Pollen cases: (i) What was the exact criterion for a sample to be classified as "pollen enriched" e.g. how many pollen on the filter?
Response: Pollen episode was identified as follows and then discussed chemical and carbon isotopic properties in the TSP samples. Please see lines 348-350 in the revised MS.
"Identification of pollen episodes was based on daily human observation of pollen blowing and the microscopic image of pollens collected in the TSP samples (Jung and Kawamura, 2011)."
However, quantitative estimation of pollen count in the TSP samples was not conducted.

Was there a relatively gradual transition from non-episodes to pollen episodes, or was there a very clear cutoff?
Response: Following sentence has been added. Please see lines 353-355 in the revised MS.
"Temporal variations of mass concentrations of citric acid and stable carbon isotopic composition during the pollen episodes showed gradual transition from the non-episodes to the pollen episodes (Fig. 3)."

(ii) Did the pollen cases also sometimes have back trajectories from China? I.e. what was done with the cases that showed both pollen on the filter as well as LRT characteristics?
Response: Air mass backward trajectories during the pollen episodes have been added in Fig. 4c. Following sentence has been added. Please see lines 362-364 in the revised MS.
"Air mass backward trajectories during the pollen episodes mainly originated from the northern part of China and the western North Pacific Ocean (Fig. 4c)."
Following sentence has been added. Please see lines 355-358 in the revised MS.
"Average mass concentrations of NO_3^- and SO_4^{2-} during the pollen episodes were ~2−3 times lower than those during the LTP and AD+LTP episodes (Table 2), implying that a relatively

low impact of anthropogenic emissions from the Asian continent during the pollen episodes."

(c) How exactly were the LRT cases classified? (i)Were they just the cases of carbon episodes that were not pollen enriched? Or were they selected based on PM10 or back trajectories? (ii) Were there any carbon episodes that showed neither LRT nor pollen characteristics?

Response: Following sentences have been added. Please see lines 301-305 in the revised MS.

"LTP episodes were identified by the elevated concentrations of nitrate and sulfate during the carbon episodes. AD+LTP episodes were identified by the elevated concentrations of Ca^{2+} and low aerosol Ångström exponents as well as the elevated concentrations of nitrate and sulfate during the carbon episodes."

Following sentence has been added. Please see lines 306-308 in the revised MS.

"Mass concentrations of nitrate and sulfate during the LTP episodes were more than 2 times higher than those during the pollen and non-episodes (Table 2), indicating strong influences of anthropogenic pollutants."

(d) Dust episodes: Are the MODIS pictures that are presented just an illustration for one day, or was this analysis done for all cases and used to identify the dust episodes? The division of cases has to be made very clear (maybe even with the help of a drawing, but at least with a clearly divided list) in the beginning of the paper.

Response: AD episode has been modified as AD+LTP episode.

The sentences beginning "A high AOT >1.0 and low α <0.4 …" have been modified as follows. Please see lines 327-331 in the revised MS.

"A high AOT >1.0 and low α <0.4 during the selected days (31 March 2007 and 25 May 2007) of the AD+LTP episodes clearly showed the presence of dust plumes over the Yellow Sea (Fig. 5e-h). The α obtained from the AERONET AOT also showed low values (0.37 ± 0.06) during the KOS627 AD+LTP episode (Table 1), indicating large size particles in the dust plumes."

The sentence beginning "The elevated concentration of nss-Ca^{2+} …" has been modified as follows. Please see lines 334-337 in the revised MS.

"The elevated concentrations of nss-Ca^{2+} (avg. 7.5 ± 0.2 μg m^{-3}), nitrate (avg. 16.0 ± 7.6 μg m^{-3}), and sulfate (avg. 32.5 ± 15.1 μg m^{-3}) in the KOS603 and KOS627 filter samples supported the presence of dust particles and anthropogenic pollutants (Tables 1-2)."

All MODIS images during the AD+LTP episodes were investigated and clearly showed dust layer over the Yellow sea and Korean Peninsular. MODIS data during the second AD+LTP episode (25 May 2007) has been added in Fig. 5g,h.

2) I think the subdivision between LTP_EC and LTP_NEC cases is not too well justified (the back trajectories are not so extremely different, since most of them start in NEC). The divison does not add much to the scientific content of the paper. There are few LTP data points anyway and subdividing then into even smaller subcases just raises questions about the statistical validity of the conclusions. The few differences that are observed (for just 2 data points in the LTP_EC case) could also be due to different meteorological conditions during transport or other influences. Unless it is a priori known that aerosols in EC and NEC differ very strongly and this difference is also seen in the data here, I would strongly advise to just omit this subdivision. It will only make the main conclusions of this paper stronger.

Response: The LTP_EC and LTP_NEC episodes in the original MS have been merged to the LTP episode.

3) The non-event cases should be included in Table 2 and Table 3 for the sake of comparison (one line with average non-event values would be sufficient.) This is especially important since the "carbon episodes" are defined against the non-event cases and the non-event cases are also included in some of the figures. This would probably also clarify some of the questions raised in point 1.

Response: The non-episodic cases have been added in Table 2. Please see Table 2 in the revised MS. Following sentences have been added. Please lines 368-376 in the revised MS.

"Average mass concentrations of TC during the non-episodes (avg. 7.1 ± 1.7 μgC m^{-3}) were ~2 times lower than those during the carbon episodes (Table 2). Average mass concentrations of nss-Ca^{2+} during the non-pollen episodes (avg. 1.4 ± 0.76 μg m^{-3}) were ~5 times lower than those during the AD+LTP episodes (avg. 7.5 ± 0.2 μg m^{-3}). Mass concentrations of citric acid during the non-episodes (range: 0.17 − 18 μg m^{-3}) were several dozen times lower than the pollen episodes (range: 20 − 320 μg m^{-3}) and almost no pollen grains were observed from the microscopic image of the TSP samples. These results indicated that airborne pollens and dust particles rarely had an impact on the TSP samples during the non-episodes."

4) The weaker Asian dust episode seems a bit questionable: Apart from low alpha and high Ca, all other characteristics (PM10, isoptopes, carbonates, thermal evolution, etc: : : blend in well with normal LRT data). This can also be seen in the figures where the strong dust episode is often an outlier, whereas the weak dust episode usually lies among the LRT data points. This might mean that a weak dust episode does not strongly affect the other aerosol characteristics (or maybe that it was a normal LRT episode?) This should be discussed in the paper a bit more clearly.

Response: AD episode has been modified as AD+LTP episode. MODIS data during the second AD+LTP episode (25 May 2007) has been added in Fig. 5g,h. The sentences beginning "A high AOT >1.0 and low α <0.4 …" have been modified as follows. Please see lines 327-331 in the revised MS.

"A high AOT >1.0 and low α <0.4 during the selected days (31 March 2007 and 25 May 2007) of the AD+LTP episodes clearly showed the presence of dust plumes over the Yellow Sea (Fig. 5e-h). The α obtained from the AERONET AOT also showed low values (0.37 ± 0.06) during the KOS627 AD+LTP episode (Table 1), indicating large size particles in the dust plumes."

The sentence beginning "The elevated concentration of nss-Ca^{2+} …" has been modified as follows. Please see lines 334-337 in the revised MS.

"The elevated concentrations of nss-Ca^{2+} (avg. 7.5 ± 0.2 μg m^{-3}), nitrate (avg. 16.0 ± 7.6 μg m^{-3}), and sulfate (avg. 32.5 ± 15.1 μg m^{-3}) in the KOS603 and KOS627 filter samples supported the presence of dust particles and anthropogenic pollutants (Tables 1-2)."

5) Figure 5a. The strong correlations observed in these Figure are mainly due to two outlier data points at very high concentrations. The interpretation of this figure should be used with great caution and the conclusions are not entirely reliable.

Response: This study roughly estimated the contribution of airborne pollens to total TC using TN and TC regression approach as shown in Fig. 6a,b and Eq. (1). In the abstract and conclusion sections, we have decided to delete the TN and TC regression approach.

The phrase "and the TN and TC regression approach" in the abstract and conclusion sections was deleted.

6) p 12883, line 26: In my opinion a likely reason for the diverging delta13C TC values are is the variability of the delta 13C of non pollen carbon (see e.g. Figure 6a), which is seen more strongly at low citric acid concentrations.

Response: Following sentence has been added. Please see lines 564-565 in the revised MS.

"The divergence of the $\delta^{13}C_{TC}$ values also can be explained by the variability of the $\delta^{13}C_{TC}$ of non-pollen carbon."

7) Figure 11: If the LRT cases are not subdivided then it can be said that the LRT cases have a relatively constant OC2, and a strongly variable OC1 fraction. This could also be an effect of aging during transport and might not necessarily reflect the different sources. In any case a regression slope derived from of 2 or 3 data points is largely meaningless (page 13886, line 11).

Response: Following sentence has been added. Please see lines 628-630 in the revised MS.

"Thermal evolution patterns of OC during the LTP episodes showed relatively constant OC2 mass concentrations but strongly variable OC1 mass concentrations (Fig. 12)."
Following sentences have been added. Please see lines 644-648 in the revised MS.
"Thus, different evolution patterns of OC obtained for the LTP and AD+LTP episodes can be explained by different formation mechanisms of secondary organic aerosols and the effect of aging of organic aerosols during long-range atmospheric transport. Different sources of organic aerosols from the Asian continent may also contribute to the different thermal evolution patterns of OC."

8) In the pollen cases, OC1 and OC2 are highly correlated and I am surprised this is not discussed more detail, considering how much discussion is spent on the statistically weaker data points.
Response: Following sentence has been added. Please see lines 665-669 in the revised MS.
"A positive correlation between mass concentrations of OC1 and OC2 (Fig. 12) and a negative correlation between the normalized OC1 and OC2 fractions (Fig. 13) during the pollen episodes imply that a small fraction of pollen also evolved in the OC1 temperature step but dominant fractions of them evolved in the OC2 temperature step."

9) Page 13887: I think this paragraph is too speculative, especially regarding the role of dust in SOA formation. It is impossible to conclude this from two data points, where one of them is in my point of view not even very clearly a dust episode.
Response: The sentences beginning "The increase of OC2 fraction … during the long-range atmospheric transport" were deleted.
Following sentences have been added. Please see lines 644-648 in the revised MS.
"Thus, different evolution patterns of OC obtained for the LTP and AD+LTP episodes can be explained by different formation mechanisms of secondary organic aerosols and the effect of aging of organic aerosols during long-range atmospheric transport. Different sources of organic aerosols from the Asian continent may also contribute to the different thermal evolution patterns of OC."

10) Figure 12: If the pollen mostly evolves at the OC2 temperature step, do you have any explanation for the strong correlation between OC1 and OC2 in the pollen cases?
Response: Following sentence has been added. Please see lines 665-669 in the revised MS.
"A positive correlation between mass concentrations of OC1 and OC2 (Fig. 12) and a negative correlation between the normalized OC1 and OC2 fractions (Fig. 13) during the pollen episodes imply that a small fraction of pollen also evolved in the OC1 temperature step but dominant

fractions of them evolved in the OC2 temperature step."

11) This paper needs to be corrected by a native speaker, before it can be published

Response: A native speaker has proofread the manuscript before submitting the revised one.