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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Springtime carbon episodes at Gosan background site revealed by total carbon, stable carbon isotopic composition, and thermal characteristics of carbonaceous particles" *by* J. Jung and K. Kawamura

Anonymous Referee #1

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This paper reports on the analysis of total suspended matter from filter samples taken in April/May 2007 and April 2008 at the Gosan site on Jeju Island. The samples were analysed with respect to total carbon, organic and elemental carbon, total nitrogen and citric and oxalic acid concentration. The thermal characteristics of organic and elemental carbon are reported for individual filter samples. Furthermore the carbon isotopic composition of total carbon and oxalic and citric acid were measured and used for the interpretation of the aerosol sources. Aerosol particles influence climate and have adverse health effects. Elucidating the source of particles and identifying potential strate-

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Interactive Discussion
Discussion Paper

gies for reducing atmospheric burdens of particles is an up to date topic. A particular challenge in this respect is the organic fraction of aerosols which has a vast variety of sources and is known to change properties and composition upon atmospheric aging. Therefore the paper in principle merits publication in ACP. Unfortunately the paper has several weaknesses which should be corrected before publication.

Major comment:

The main issue with the interpretation of these data comes from the classification of individual episodes. It seems from the manuscript that the classification into long range transport, pollen and Asian dust, is based on HYSPLIT backward trajectory analysis. While the sampling was performed 3m above ground level, the backward trajectories were calculated for 500m above ground level. No explanation is given why a trajectory at 500m would be representative of the 3m sampling height in the observation period. This is a crucial point and a proper justification for using backward trajectories at 500m must be given. This is particularly important when taking into account that the filter samples were taken over long averaging times (2 to 6 days). These long averaging times certainly increase the risk that samples can not be unambiguously assign to one air mass origin or episode. In particular the data in table 2 shows, that hardly any difference can be seen between the different classes (see also attached figure). Within the reported standard deviations no difference can be seen between episodes. If at all the main difference between the episodes is seen in d13C. On the other hand e.g. the data for the two pollen episodes shown in figure 2 seem to differ substantially in terms of d13C and TC/TN ratio, further raising the question how representative the classification of episodes is.

The authors should furthermore provide information on how they accounted for the different number of samples available for the individual episodes. Were the reported values weighted with number of samples or is the difference in data coverage not considered when comparing different episodes?

ACPD

11, C5754–C5759, 2011

The sampling and methods part of the manuscript lacks detailed information in a number of places, partly making it difficult to assess the validity of data interpretation. For instance the authors need to describe the procedure used to separate pollen from an ambient sample in much more detail and show that this sample contains pollen only. Also the standard pollen samples used need to be further specified, in particular with respect to the author's statement on page 13882, line 4 that d13C of pollen samples may depend on geographical location. Table3 shows a much lower (-28‰ d13C for the pollen in the ambient sample than measured for the standard pollen (-25.4 and -23.3‰ for cedar and cypress pollen, respectively). Also the huge differences in the d13C of the water soluble fraction, especially of oxalic acid is not well explained, probably due to the small number of samples (here only one ambient pollen sample is discussed).

In several parts of the manuscript the discussion remains very speculative and is in this reviewers' opinion not sufficiently underpinned by the data. For instance on Page 13883 line 23 the authors interpret a "divergence of the d13C values at a certain level of citric acid-C/TC ratio" as due to "different adsorption efficiency of citric acid on pollens and different emission strength of citric acid from tangerine fruit". This is certainly not the only possible explanation, e.g. mixed air mass influence on some of the samples, other sources of citric acid, fractions of carbon with low d13C other than pollen etc. could all lead to the same observation. Similarly the authors interpret the difference of d13C of citric acid in the ambient sample and the d13C of citric acid in tangerine peel as due to the kinetic isotope effect. This is highly speculative. To this reviewers knowledge no data on KIE of citric acid in atmospheric reactions is available in the literature. In any case the presence of a KIE and its influence on observed d13C in citric acid would imply a chemical loss process of citric acid. This would at the same time mean that no robust correlation between citric acid content of a sample and e.g. d13C of total carbon can be expected.

Similarly the discussion of thermally resolved OC components and their changing abundance during long range transport (page 13887 line 10 and following) remains

ACPD

11, C5754–C5759, 2011

very vague and speculative. In the discussion of the dependence of d13C on thermally evolved OC fractions, it seems that the main fractions OC1 and OC2 have to show the observed behaviour. Since both fractions are normalized by total OC, if one increases, the other will necessarily decrease. An anti-correlated dependence of d13C on OC1 and OC2 therefore seems to provide no information on the source or characteristics of the aerosol.

Minor and technical points: Figure 1 is not referred to in the text.

"Carbon episodes" as used in the title and throughout the manuscript is not a well defined term.

Page 13869 line 8: sentence not clear

Page 13879 line 4: sentence not clear

More detailed information on aerosol sampling should be provided. For instance what was the cut-off size of the sampling set-up, which sampling flows were used?

Page 13871 line 24: what were the tangerine fruit prepared for?

Filter areas used for individual analysis are given surprisingly exact. How well determined is the filter area use?

Page 13874 line 15: were blank measurements for this procedure performed?

Page 13876 line 7: starting from here results are presented.

Page 13880 line 21: removed_C is not the abbreviation used in figure 6.

Page 13881 top paragraph: some explanation could be provided why carbonate C is expected to have high d13C.

Discussion of citric acid in pollen samples (page 13883): the cited reference (Jung and Kawamura 2011) is not available and the conclusions are therefore difficult to assess.

Table3: sum formulas of oxalic acid and citric acid could be provided

ACPD

11, C5754–C5759, 2011

Interactive Comment

Figure 10: Why is it that for most of the data the signal at times 0 and 1000 is >0? If this is an issue of offsetting the data for visualization, a better way of presentation should be chosen.

Figure 11: this again shows how little difference between episodes is seen in some of the aerosol features: LTP_NEC is equal to pollen events in terms of OC1 and OC2 characteristics.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13867, 2011.

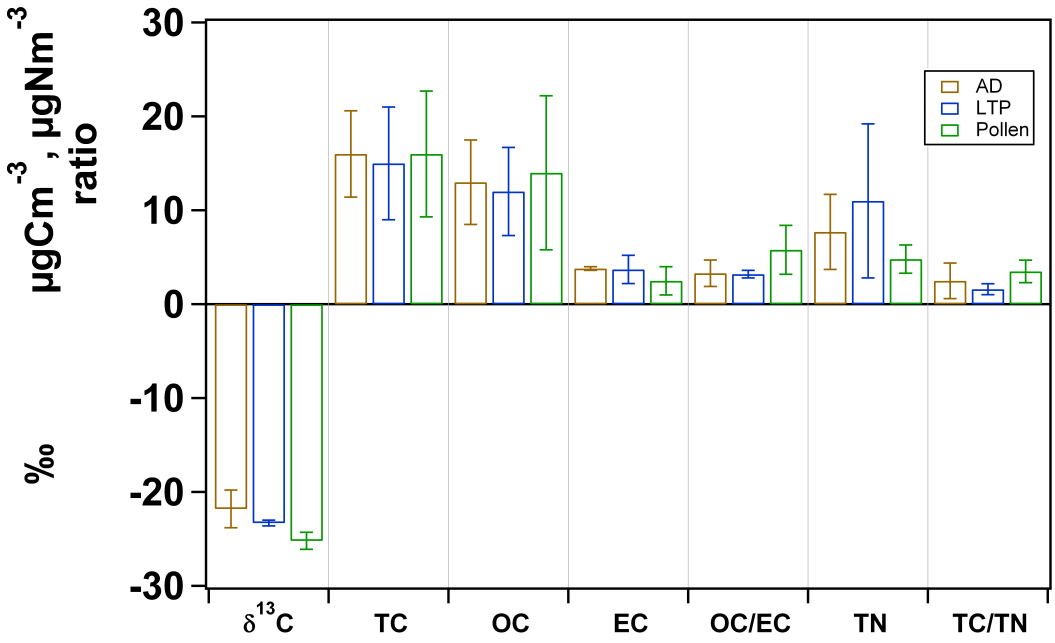

Fig. 1.

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11, C5754–C5759, 2011