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Interactive comment on "One-year record of organic and elemental carbon in fine particles in downtown Beijing and Shanghai" *by* F. Yang et al.

Anonymous Referee #2

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This manuscript presents data for organic carbon (OC) and elemental carbon (EC) for a one-year period (1999-2000) for the cities of Beijing and Shanghai. The data were obtained from weekly samplings (in practice, 49 samples were taken in Beijing, 51 in Shanghai). The data seem to be of some value because they were obtained with the well-known and well-documented thermal-optical reflectance (TOR) method, presumably the method described by Chow et al. (1993) or a variant thereof. (Incidentally, if so, reference should be made to Chow et al., 1993). The same OC and EC data have essentially already been given in the papers of He et al. (2001) and Ye et al. (2003). What is new in the current manuscript is (1) that the OC and EC data are deeper discussed, (2) that some data from 14C analyses are presented, to resolve modern

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(mainly biomass burning) from fossil fuel carbon, (3) that the EC data are related to data of non-crustal K, a well-known tracer for biomass burning, and (4) that an attempt is made to derive what fraction of the OC is secondary organic carbon (SOC).

Overall, the manuscript gives a rather sensible interpretation of the data. However, on a number of occasions, the analysis and/or interpretation are unclear, flawed or simply wrong. The references are also not really up to date. As indicated below, some key references are missing. Therefore, major revision is definitely needed before this manuscript may eventually become acceptable.

I have extreme difficulties with the authors' SOC data. In my opinion, their SOC are worthless and the data and their discussion should be removed from the manuscript. The approach to derive SOC, as used by Turpin and Huntzicker (1991), Turpin et al. (1991), Turpin and Huntzicker (1995) and several others, should be employed with great care. It is well known that the (primary OC)/EC ratios vary considerably from source to source and that for ambient samplings at a certain location, the (primary OC)/EC ratio will be influenced by meteorology, diurnal and seasonal fluctuations in emissions, and the influence of local sources (e.g., Turpin et al., 1991). Therefore, the approach can really only be employed when the ratio of (OC/EC)pri during the sampling campaign can be assumed to remain reasonably constant. This means that it can only be used for rather short campaigns (within a single season). Also, the approach is best used with short sampling times, such as the 2-hour samplings of Turpin and Huntzicker (1995). The approach may still give rather reasonable results with 12-hour or 24-hour samples, provided the samples are taken within a short campaign, as for example, done by Cao et al. (2003) in four cities of the Pearl River Delta Region (PRDR) during a winter 2001 period. Using it with weekly samples taken over a full year, as done in the current manuscript, is stretching it really too far. For sure, the (OC/EC)pri ratio will not remain constant throughout the year.

Page 221, lines 10-13: The authors give a sensible argument why they selected for Shanghai the data of Tongji instead of Hainan Road. They should also explain why, for

Beijing, they took the data from Chegongzhuang instead of those from Tsinghua.

Page 221, lines 27-28: The authors write here “It is recommended to analyze only the front quartz filter (Chow et al., 1994; US EPA/NARSTO, 1998).”

Here the authors incorrectly quote Chow et al. (1994) and draw wrong conclusions. Chow et al. (1994) write on page 2063 “The organic carbon on the backup quartz-fiber filters was generally 10-30% of the concentrations on the front quartz-fiber filters. These biases do not exhibit a consistent pattern, and were not used to adjust carbon values on the front quartz-fiber filters. More study of carbon sampling and analysis methods is needed to fully resolve this issue”. Also the report US EPA/NARSTO (1998) does not provide any recommendation. On page 49 of this report there is written: “Now, I know some of you use two filters, two quartz filters to measure organic carbon, but the panel felt that we really don’t know what that second filter means. Should we add it or subtract it or multiply it? We have no idea, so we felt, in the meantime, until we solve this, we better just measure only the front filter. It’s cheaper and probably more reasonable.”

Since 1994 and 1998 several other studies with front and back filters have been made and also the key review paper by Turpin et al. (2000) comments on this topic. Another key paper that deals with this issue is that by Mader et al. (2003).

Page 222, lines 11-12: The authors write here “The slight weekly variations and low levels of OC and EC concentrations in the summer are reasonable since the local wind is mild in this season.”

I presume that the authors mean by “mild” wind that the wind is weak (i.e., that the wind-speed is low). If so, I do not see why it is reasonable to have low levels of OC and EC at low wind speed. Rather, since the sources of the OC and EC are local (and regional) and the low wind speed does not favor dispersion of the local pollution, I would rather expect a build-up of OC and EC levels then. A more logical explanation for the observed lower OC and EC levels in the summer could be that there is more removal by rain in summer and/or that the sources of these species are weaker in this season. Incidentally, on page 224, lines 14-16, the authors invoke precipitation in summer to explain the low OC and EC levels then.

Page 226, lines 7-10: It is well-know that the EC/TC ratio (with TC = OC + EC) is method-dependent. It is dangerous to compare data from different locations if different OC/EC analysis methods were used. Were the data at the various cities listed here all obtained with the TOR method?

Page 226, lines 12-13: Light absorption indeed contributes to visibility impairment. But also light scattering does so. The OC particles are efficient light scatterers and could be responsible for a large fraction of the visibility impairment.

Page 226, lines 18-19: It is unclear where the K and Fe data come from. Presumably, from parallel samples analyzed by ion chromatography (IC) and/or X-ray fluorescence (XRF), as reported in He et al. (2001) and Ye et al. (2003). Some more explanation and/or reference to these two papers is needed here. Also, K should not only be corrected for the crustal contribution, but also for the sea-salt contribution. Thus, non-crustal/non-sea-salt K should be calculated and used as indicator for biomass burning.

Page 227, lines 10-15: The samples used for 14C were collected in 2001, whereas the main samples for this manuscript were taken in 1999-2000. To use the similarity in OC and EC levels of these two years as motivation that the sources were the same makes no sense.

Page 228, lines 19-21: What the authors write here is in contradiction with what they write on page 229, lines 8-14. There they write that the OC/EC ratios are quite different for different sources. Since the relative contribution from the various sources is certainly not the same throughout the year, how can they state that the "OC/EC ratios were not sensitive to ... changing source emissons"?

Page 238, Figure 1: The dates in the abscissa should be arranged in such a way that the same months are vertically aligned above each other. This would make an investigation of the seasonality in the data and of the similarities/differences therein between the two cities a lot easier.

Minor comments:

Page 219, line 16: “Pear river” should be replaced by “Pearl river”.

Page 219, line 22: I suggest to replace “Guangzhou, Beijing, and Shanghai,” by “Guangzhou, Shanghai, and Beijing,” to have the cities in the same order as the regions mentioned in lines 16-18.

Page 226, line 11: “Hller” should be replaced by “Höller”.

Page 230, line 10: “exhibted simialar” should be replaced by “exhibited similar”.

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 217, 2005.