

Interactive comment on “Determination of gaseous and particulate carbonyls (glycolaldehyde, hydroxyacetone, glyoxal, methylglyoxal, nonanal and decanal) in the atmosphere at Mt. Tai” by K. Kawamura et al.

Anonymous Referee #1

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This study reports measurements of gaseous and particulate carbonyls (glycolaldehyde, hydroxyacetone, glyoxal, methylglyoxal, nonanal, and decanal) at a mountainous site (Mount Tai) in the North China Plain during June 2006. The measured gaseous concentrations for all six species were among the highest ever reported in the urban and forest atmosphere. The carbonyls were much more abundant in the gas phase than in the particulate phase. Glyoxal, methylglyoxal, and glycolaldehyde were highly correlated with levoglucosan, suggesting that major contributions from open burning of agricultural residue.

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The study is of interest to the community for better understanding the sources of carbonyls in the atmosphere and the contributions of carbonyls to secondary organic aerosols. The concurrent gaseous and particulate carbonyl measurements reported in this study are rare to begin with, and this study was conducted in an area where data are particularly sparse. In that sense, the paper fills a crucial data gap. For that matter, I suggest that the paper be accepted after addressing the major issues raised below.

Major issues: 1. The scientific content of the paper in its current form is fair. But the paper has the potential for much higher scientific relevance. In particular, the paper reports the first (to the best of my knowledge) co-located MAXDOAS and filter-based glyoxal measurements. Glyoxal is of interest due to its potential contribution to the photolytic production of radicals and the formation of secondary organic aerosols. Satellite and ground-based MAXDOAS measurements of glyoxal have emerged during the last decade, but so far validation has been minimal. Here the author compared the glyoxal concentrations from MAXDOAS and filter-sampling but merely stated that the two were positively correlated (and with r of only 0.58). Very little was done to explain the factor of 5 differences between the two measurements. The authors said that there may be stronger photochemical production of glyoxal in the upper atmosphere, but did not mention whether the MAXDOAS measurements were in support of this. Moreover, the analysis with levoglucosan clearly indicates that the gases glyoxal measured by filter-sampling were emitted primarily from biomass burning. I fully understand that a detailed comparison with the MAXDOAS measurement entails a separate study, but some resolution of the issues above would vastly improve the relevance of the current paper.

2. Another important problem with the paper is that the inference for the source of oxalic acid was based purely on the correlation against the gaseous alpha-dicarbonyls and the diurnal variation of product/precursor ratios. The authors jumped to the conclusion that the oxalic acid was produced by the partitioning of alpha-dicarbonyls into

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the aqueous phase followed by oxidation. This was a very premature conclusion, in my opinion. Many studies have shown that biomass burning emits large amounts of both alpha-dicarbonyls and oxalic acid, so the high correlations there may very likely be the sole result of the same primary source. The diurnal variation could merely reflect a larger agricultural burning emission or photochemical production of gaseous dicarbonyls during the day. It is important that the authors fully address these possibilities.

Minor issues: 1. Page 2736, lines 4-5: 'Plain possibly ...': I do not understand this sentence; there seems to be a typo.

2. Page 2737, lines 14, 16: 'Fig 10a' should be 'Fig 11a'. 'Fig 10b' should be 'Fig 11b'.

3. Page 2737, lines 22, 27: 'Fig 11a' and 'Fig 11b' should be 'Fig 12a' and 'Fig 12b'.

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