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Atmospheric Chemistry and Physics Discussions

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.

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Authors' Responses to Ref. #1

Thank you for the careful review and valuable comments on our manuscript.

Anonymous Referee #1

This study reports measurements of gaseous and particulate carbonyls (glycoaldehyde, hydroxyacetone, glyoxal, methylglyoxal, nonanal, and decanal) at a mountainous site (Mount Tai) in the North China Plain during June 2006. The measured gaseous

concentrtaions for all six species were among the highest ever reported in the urban and forest atmosphere. The carbonyls where 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. 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 comparisons with the MAXDOAS measurement entails a separate study, but some resolution of the issues above would vastly improve the relevance of the current paper.

Response: Thanks for the comments. We added a few sentences in the revised MS as follows: “Although MAX-DOAS measurements did not directly support the photochemical production of glyoxal, it is of interest to note that MAX-DOAS data showed that glyoxal concentrations increased in the afternoon (Fig. 7b), suggesting that glyoxal is photochemically produced in the atmosphere. Alternatively, the significant difference between two methods may suggest that the distributions of glyoxal are not homogeneous between the mountaintop site and ground level. It is also important to note that due to a very thick aerosol layer in this polluted region MAX-DOAS data may have significant uncertainties, potentially causing lower concentrations.” Please see lines 271-278.

2. Another important problem with the paper is that the inferrence for the source of oxalic acid was based purely on the correlation against the gaseious 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 C1197 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.

Response: Thanks for the comments. Based on the comments, we newly discuss several possibilities that control the concentrations of gaseous dicarbonyls. Please see a few sentences added in the revised MS; lines 339-346.

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

Response: This is a typo made during proof preparation. The sentence was rephrased.

Please see lines 268-271 in the revised section 3.3.

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

Response: corrected.

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

Response: corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 2725, 2013.