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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The contribution of anthropogenic aerosols to aerosol light-scattering and CCN activity in the California coastal zone" *by* D. A. Hegg et al.

Anonymous Referee #1

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This study uses positive matrix factorization (PMF) to identify aerosol sources during three field campaigns. Aerosol filter samples were collected during the summers of 2004, 2005, and 2007 on aircraft in the marine boundary layer off the coast of California. Specifically, the contributions of three or four sources to particle number concentration (here used as a proxy for CCN concentration) and light scattering are presented. While the topic is relevant for *Atmos. Chem. Phys.*, and the methodology and presentation of results is sound, the authors have previously published the results of receptor modeling on the 2007 dataset, as they discuss on page 11488, lines 14-16: "As in Hegg et al. (2009), we therefore use receptor modeling to address the issue of sources of CCN activity and aerosol light scattering. However, while the EPA UNMIX

2.3 model was used in Hegg et al. (2009), in this study we use the EPA PMF model 3.0." In addition to the new receptor model, this manuscript also includes PMF analyses of filter analyses from two previous years. These are the two new contributions of this work, and thus to merit publication in *Atmos. Chem. Phys.*, the manuscript should both demonstrate why the use of PMF 3.0 is an advance over UNMIX 2.3, and use the additional data (years) to broaden the conclusions drawn beyond what could be said based on a single year. I think that the latter has been achieved, but I feel that a major revision is needed to address the former. If this can be done, I would recommend publication after the following issues are addressed.

I. Major points

The authors briefly discuss general advantages of PMF 3.0 over UNMIX 2.3, but should indicate how this particular dataset benefits from PMF 3.0, and also discuss the circumstances under which PMF and UNMIX produce different results. When used on the 2007 dataset, PMF produces four factors, while UNMIX produces only three, but as the authors discuss, this is to be expected, and the authors generally lump the two marine sources together in their analyses of the data. The authors compare the PMF results to the UNMIX results in the first paragraph on page 11492. The most interesting point here is that certain high-CCN samples are less predominantly marine in PMF than in UNMIX. The authors mention that they prefer the PMF results based on the high [CN] for those samples, but do not attempt to explain why the two models disagree. I think such a discussion is necessary, given that the use of PMF3.0 was mentioned as a justification for this study. In general, this section of the paper should be expanded, such that the reader gains a greater understanding of the differences between the models and some idea as to when one would be preferable to the other, and when they yield essentially the same results.

The manuscript also presents PMF analyses of two previously unpublished datasets. In comparing the three summers, the authors note that the pollution contribution was greatest in 2005, and the biomass burning greatest in 2007. Back-trajectory analyses

indicate more offshore flow during the 2005 campaign. In general, I found the discussion of the inter-annual variation satisfactory, although the (appropriate) brevity makes it even more important that the PMF vs. UNMIX issue discussed above be improved.

A weakness of this paper is the use of "cases" instead of more clearly defined samples (e.g., a time series, or a depth profile). For example, the variation in the fraction of the particles attributed to a marine source from year to year could be due in part to variations in the fraction of cases sampled at the lowest altitude, but the authors have not provided any information to test this idea. The authors mention that below cloud base was sampled "when cloud was present" – how consistent was the frequency of cloud formation between the three campaigns? At the very least, add a table that summarizes the three campaigns and includes the fraction of the cases during each campaign taken at various altitudes, under cloudy vs. cloudless conditions, etc.

Why claim CCN for the first two experiments? Why not just say CN? Although this issue is well-discussed in the text, I find the captions from Figs. 4-6, which refer only to "CCN (0.3%) concentration" to be potentially misleading. I think the authors should take the more straightforward approaching of reporting PMF contributions to particle number, as measured by the PCASP. Especially considering the fact that a constant value of soluble fraction (30%) is assumed for all aerosols (from all sources) in the conversion from [CN] to [CCN] (and thus that all particles are equally CCN active), I think the authors are on much firmer ground reporting the source contributions to PCASP particle number. This change would more accurately reflect the data used by the authors, and it would not reduce the significance of this study.

II. Minor points

P11488, L1-6: The PCASP vs. CCN data should be presented to justify use of PCASP as a proxy for CCN. While I agree that 0.69 is a "reasonable" value for r^2, I can't agree with the authors that "the variance structure of the CCN proxy is clearly similar to that of the CCN" without seeing the data. Figures 1-3 could be combined into one if the

authors would like to limit the number of figures.

P11490, L17: Does "secondary" here refer to secondary aerosol, or are the authors using a more general definition? This is potentially confusing.

Fig. 1, and others: Increase font size. Many of the figures would be easier to read if the font size was as large as it is in Fig. 9.

The color scheme is consistent from Fig. 4 to Fig. 8. But earlier source profile figures, as well as Fig. 9, have colors switched, potentially causing confusion. Please standardize the color scheme.

Fig. 4: "Marine 2" label used twice – which one is "Marine 1"?

Fig. 10: Why is this analysis presented for only CARMA III? A three-paneled figure (with much smaller panels) seems warranted, covering all campaigns, unless the other two are essentially identical to this one (and if so, this should be stated).

Figs. 11-12: Are the differences in the contributions from year to year statistically significant, according to the PMF model? One of the benefits of this model is its ability to calculate uncertainty in source contributions given uncertainty in concentration measurements.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 11483, 2010.