Atmos. Chem. Phys. Discuss., 8, S7404–S7414, 2008
www.atmos-chem-phys-discuss.net/8/S7404/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Intra-community spatial variability of particulate matter size distributions in southern California/Los Angeles" *by* M. Krudysz et al.

M. Krudysz et al.

Received and published: 23 September 2008

This manuscript presents an analysis of the spatial, diurnal and seasonal dependence of particle number size distributions in the vicinity of the Ports of Los Angeles and Long Beach. Particle size distributions were monitored at 13 sites from April to December, 2007. Spatial variation analysis was made by calculating the coefficient of divergence and correlation coefficients between site pairs. The data are interesting but unfortunately, in some places the manuscript suffers from unsufficient arguments so that many conclusions are not scientifically solid. Thus the manuscript needs major revision before publication in ACP.

The authors agree with the reviewer that the data used for analysis in this manuscript

are very interesting and we would like to emphasize the enormous number of data points and numerous sampling sites. It is outside of the scope of this study to analyze the meteorological data using PMF or to fit log-normal distributions to every sampling site and month. In view of the reviewer8217;s comments, we have revised the manuscript considerably and discussed the effects of ship and industrial emissions on the observed particle number size distributions. In addition, the mono-modality of our data has been discussed in more detail and supported by several additional references, whose results are in agreement with ours.

General comments:

The average particle size distributions for each site and sampling period are presented in Figures 2a-2g. The authors conclude that for most of the sites, a single mode is present at particle sizes 20-60 nm. This is disagreement with the other studies mentioned in the manuscript; for example, Hussein et al., 2005 reported two modes 8211; nucleation mode < 25 nm and Aitken mode 25-90 nm. Log normal modes should be fitted to the observations. After this additional information I believe that the conclusions will be changed. I am also wondering in Table 1 why the geometric mean diameters for the whole size distributions are calculated. What do they describe? I would suggest to calculate the modal geometric mean diameters with std.

The authors acknowledge that, overall, the size distributions averaged to a single mode, but we respectfully disagree with the reviewer8217;s assertion that our conclusions are in complete disagreement with other studies. The Hussein et al. manuscript referenced by the reviewer shows unimodal size distributions (Figure 2 a, b, e, f). In addition, research conducted in close proximity to vehicular exhaust resulted in a unimodal size distributions, and only measurements taken with a thermodenuder showed that the size distribution consisted of the accumulation mode and the more volatile nucleation mode (Ronkko et al., 2007). Also, a recent study has found that for small distances behind a vehicle being investigated, no nucleation modes were observed, only a unimodal particle number size distribution (Morawska et al., 2007). Another study has noted that

ACPD

8, S7404–S7414, 2008

the size of the nucleation mode particles is really small and that at most driving conditions, the GMD of nucleation mode is less than 10 nm. Limiting the lower measured size to >10 nm can result in missing the nucleation mode. The study concluded that the measurement range of the instruments is an important issue in nucleation mode studies (Ronkko et al., 2006). A brief summary of these studies has been included in the introduction and referenced in section 3.2 Particle Number Size Distributions while discussing the modality of our data.

Further, we would like to point out that the size distributions presented in Figures 2a-g are averages of all measurements taken during the indicated month; therefore possible occurrences of bi-modality are masked by the averaging process. For example, bi-modality was found in our data when diurnal patterns were considered. During morning commute hours, a bi-modal distribution was observed at LB5, with a peak at 20-30 nm and a smaller one at 60-70 nm (Figure 4b).

Fitting log-normal modes to the vast amount of data collected over 9 months at 13 sites is outside of the scope of this study. We calculated the geometric mean diameter for the whole size distribution at each site to determine pattern in the distributions. Since high density of vehicular emissions are found in the Long Beach/Wilmington area, these sources are expected to be the primary contributors to the particle number size distributions at the sampling sites. Furthermore, most sites were located very close to these sources, therefore proximity to vehicular emissions can explain the mono-modality of our averaged size distributions. The geometric standard deviations shown in Table 1 are small compared to the geometric mean diameters, implying that the distributions are not, on average, wide and are mono-modal.

The authors state that for the measurement sites LB2, LB8, LB9, W3 (section 3.2.1) higher average particle number concentrations (Ntot) and larger number median diameters in winter than in summer (cf. Figure 2c, e and f and Table 1) could be explained by higher RH in winter. However, the average particle number concentrations given in Table 1 are in contradiction with the above statement because only for W3 the win-

ACPD

8, S7404–S7414, 2008

ter Ntot is higher than in summer. Furthermore, only at LB2 the mean value of RH is clearly larger in winter than in summer. What is the explanation for higher concentrations in winter as shown by Fig. 2e,f,g because the average temperature differences in winter and summer are very small (Table 1). Please, check all values in Table 1. Some statistics for T and RH are needed.

In addition to the changes in RH and T, mixing height influences particle number concentrations. Although no data were available on mixing height during the study period, total particle number concentrations measured simultaneously as the particle number size distributions (Moore et al., 2008) shows distinct seasonal patterns, with relatively higher levels observed in the fall/winter than in the summer. This is consisted with previous observations conducted in this area (Singh et al., 2006).

Although very good agreement was found between total particle number concentrations measured by a separate CPC and the SMPS unit (r>0.9), trends in particle number size distributions from site to site did not always correspond to those for the total particle number concentrations calculated by a CPC. This occurred for several reasons: some data from the concurrently running CPC (Moore et al., 2008) was missing during SMPS sampling periods; and the differences in particle size limits of detection (7 nm and 14 nm for the CPC and the SMPS, respectively) created discrepancies between total particle number concentrations and particle number concentrations detected in the various size intervals. Table 1 has been revised to show total particle number concentrations for each site calculated by summing particle number concentrations across all size intervals measured by the SMPS. Showing total particle number concentrations measured by the SMPS is also a more appropriate comparison to the particle number size distributions. Table 1 shows higher total particle number concentrations at sites LB2, LB8, LB9, W3 during the fall/winter season in comparison to the summer sampling campaigns at those sites. This information has been added to the text in the manuscript to support higher observations in particle number size distributions at sites LB8, LB9, and W3.

ACPD

8, S7404–S7414, 2008

I would expect clear differences in number size distributions during weekdays and weekends. However, the authors have not taken this into account when studying diurnal variability analysis. Could you comment this?

As with other studies (Hussein et al., 2005), our study did not find differences in location of modes between weekdays and weekends, only differences in absolute particle number concentrations. As an example, a figure has been added to the supplemental materials, which compares diurnal patterns between weekdays and weekends at site W2. A discussion on the differences in diurnal particle number size distributions between weekdays and weekends was added to section 3.2.2 Diurnal Variability.

By comparing the background locations SP1 and LB1 (Fig. 6a) very high spatial divergence was observed for particles < 20 nm whereas for larger particles spatial divergence was smaller. The authors state that one reason is that LB1 is influenced by transient emissions from ships entering and leaving the harbour. What is typical size distribution for the ship emissions? I would expect not only high nucleation mode but also high Aitken mode. Its effects should be included in your conclusions.

It should be noted that comparisons between sites LB1 and SP1 discussed in Figure 6a were included only for the winter season (Nov-Dec), when simultaneous data from these sites were available. There was a wind shift during that period and appreciably higher overall particle number concentrations were observed at LB1 compared to SP1 (Table1) and (Moore et al., 2008). This created particle number size distributions influenced by emissions from the Port at LB1, including not only ships but also other diesel vehicles servicing the incoming cargo and auxiliary engines.

The studies available for particle number size distributions from ship emissions show somewhat mixed results. A nucleation mode resulting from ship emissions and a size distribution similar to those observed previously for vehicular emissions has been observed for plumes originating from ships on the Baltic Sea (Hyvarinen et al., 2008). Similarly, other studies have found that ships emit particles with a marked nucleation

ACPD

8, S7404–S7414, 2008

mode (108211;100 nm), but with negligible numbers of particles with radii greater than about 0.1 61549;m. Peaks in nucleation modes were dependent on engine type (Hobbs et al., 2000). Quickly vanishing Aitken mode (61566;20 nm) has been found for ship plums emitted in Asia (Lin et al., 2007). Particle emissions from cruising ships in the English Channel were characterized by a strong mode centered at 15 nm and a second but slightly weaker mode centered around 50 nm, although these modes were shown to change based on the age of the plume (Petzold et al., 2008). Nucleation mode particles, which have formed in the expanding and cooling plume from emitted gaseous precursors were shown to vanish after about 1000s, presumably due to coagulation with Aitken and accumulation mode particles. Similar bi-modal distributions were found for ship plumes passing over an urban area of Sweden with a dominant mode in the diameter range below 40 nm and another, much weaker mode detected in the range 70-100 nm (Isakson et al., 2001). Mono-modal distributions have been observed from low speed marine diesel engine emissions (Kasper et al., 2007). The mean particle diameter was 258211;40 nm, smaller compared to that found for road vehicle engines. In addition, the accumulation mode seemed to be completely missing or was covered.

A brief summary of previous studies on particle number size distributions from ship emissions was included in the manuscript and the influence of ships on the observed size distributions was included in the text. Since it is not possible to distinguish the effects of ship emissions from other port-related emissions on the size distributions observed at SP1, the conclusions were limited to the combined effects of ship emissions and other diesel emissions.

Specific comments:

p. 9643, line 16: Puustinen et al, 2007 does not study LA locations

The sentence referencing Puustinen et at. has been revised to 8220;Studies in various locations throughout the world have shown substantial spatial variability in particle

ACPD

8, S7404–S7414, 2008

number concentrations8221;.

p. 9643, line 29: coagulation might also be important (Kerminen et al., Atmos. Environ., 41, 1759-1767, 2007)

Coagulation has been included in the manuscript as one of the mechanisms by which ambient aerosols undergo atmospheric transformation after emission from a source. The references by Kerminen et al. has also been included.

p. 9645, line 3: the reference Moore et al., 2008 is missing from the reference list

The Moore et al., 2008 reference has been added to the text

p. 9645, line 10: industrial sources are mentioned here but their effects have not been discussed section 3 Results and discussion.

A paragraph has been added in the Results/Discussion section under 3.2 Particle Number Size Distributions, which discussed the possible effect of industrial emissions on the observed particle number size distributions in this study. Specifically, although studies have shown bimodality in particle number size distributions from industrial facilities (Maguhn et al., 2003; Morawska et al., 2006), the large variability in the concentration of particles within those modes can make one or the other mode less visible in the size distribution. The absence of this bimodality in our data set, therefore, can be attributed to either absence or insignificant impact of industrial sources, or the limitation of using particle number size distributions to discern industrial emissions from vehicular emissions.

p. 9645, line 17: explain the abbreviations PoLA and PoLB

PoLA and PoLB were defined as Port of Los Angeles and Port of Long Beach, respectively earlier in the manuscript, at the end of the introduction section, p.9644, lines 26-27.

p. 9647, line 9: might be good to mention the particle size range measured by CPC,

ACPD

8, S7404–S7414, 2008

Interactive Comment

and the number of size sections recorded by SMPS

Descriptions of the particle size range measured by the CPC and the number of size sections recorded by the SMPS were included in section 2.2 Instrumentation.

p. 9649, line 2: where are the meteorological data measured?

As stated in Section 2.2: Instrumentation, A Vantage Pro 2 Weather Station (Davis Instruments, Hayward, California), installed within 10 feet of the SMPS unit, collected meteorological data including temperature, humidity, and wind speed and direction at each site.

p. 9649, line 2: what do you mean here by the word consistent?

The word consistent was used to imply that meteorological conditions did not vary much from site to site. This was explained more clearly by changing the text to 8220;Meteorological data are shown in Tables 1 and 2. Average temperatures and relative humidity did not vary much from site to site, but seasonal variation was observed.8221;

p. 9651, lines 23-26: these qualitative statements need more precise quantification, eg. by positive matrix factorization (see eg. Krecl et al., ACPD, 8, 5725-5760,2008)

Full analysis of meteorological parameters was outside the scope of this study. It is not possible to include a detailed description and analysis of the weather conditions at all sites throughout the course of the study, as such analysis would constitute an entire manuscript. Although we acknowledge that the meteorological data presented in this manuscript are limited, we would like to point out that the goal was to provide a flavor of the meteorological conditions at the various sites to aid in the interpretation of the differences in particle number size distributions among the sampling sites. A sentence was added to the manuscript explaining that quantitative analysis of the meteorological data set was outside of the scope of this study.

p. 9652, line 27: You mention that nucleation mode at around 20 nm from vehicle

ACPD

8, S7404–S7414, 2008

Interactive Comment

emissions arises from condensation of organic species onto solid nuclei (Morawska and Zhang, 2002). The recent studies show that nucleated particles might be formed by binary homogeneous H2SO4-H2O nucleation (volatile nuclei) after the exhaust tube or have non-volatile core formed already in the engine (see eg. Rönkkö et al., Environ. Science and Technology, 41, 6384-6389, 2006.

The authors agree with the reviewer that in addition to condensation of organic species onto solid nuclei, binary homogeneous nucleation can also play a role in particle formation. These additional processes and their references have been included in the manuscript.

p. 9653, line 10: smaller and larger particles - give more specific values

Specific values (< 30 nm and > 30 nm) were given to summarize the spatial variability of PM

p. 9654, line 8: 8220;PM size distribution8221; is misleading word for number size distribution. This is mentioned many times also in Introduction.

The phrase 8220;number size distribution8221; was substituted for 8220;size distribution8221; wherever it occurred in the manuscript.

p. 9657, line 11, W2 to LB9 LB5, is the word AND missing

The word 8216;and8217; has been added to the sentence.

Figure 2 and 6: The quality of the figures is not good enough since it is hard to distinguish the curves.

The symbols in Figures 2 and 6 were enlarged to distinguish the curves.

Figure 7: LB2 should read W2 in the figure caption

The typo in the figure title has been changed.

References

ACPD

8, S7404–S7414, 2008

Interactive Comment

Hobbs, P. V., Garrett, T. J., Ferek, R. J., Strader, S. R., Hegg, D. A., Frick, G. M., Hoppel, W. A., Gasparovic, R. F., Russell, L. M., Johnson, D. W., O'Dowd, C., Durkee, P. A., Nielsen, K. E., and Innis, G.: Emissions from ships with respect to their effects on clouds, J Atmos Sci, 57, 2570-2590, 2000.

Hussein, T., Hameri, K. A., Aalto, P. P., Paatero, P., and Kulmala, M.: Modal structure and spatial-temporal variations of urban and suburban aerosols in helsinki - finland, Atmos Environ, 39, 1655-1668, 2005.

Hyvarinen, A. P., Komppula, M., Engler, C., Kivekas, N., Kerminen, V. M., Dal Maso, M., Viisanen, Y., and Lihavainen, H.: Atmospheric new particle formation at uto, baltic sea 2003-2005, Tellus B, 60, 345-352, 2008.

Isakson, J., Persson, T. A., and Lindgren, E. S.: Identification and assessment of ship emissions and their effects in the harbour of g(o)over-circleteborg, sweden, Atmos Environ, 35, 3659-3666, 2001.

Kasper, A., Aufdenblatten, S., Forss, A., Mohr, M., and Burtscher, H.: Particulate emissions from a low-speed marine diesel engine, Aerosol Science and Technology, 41, 24-32, 2007. Lin, P., Hu, M., Wu, Z., Niu, Y., and Zhu, T.: Marine aerosol size distributions in the springtime over china adjacent seas, Atmos Environ, 41, 6784-6796, 2007.

Maguhn, J., Karg, E., Kettrup, A., and Zimmermann, R.: On-line analysis of the size distribution of fine and ultrafine aerosol particles in flue and stack gas of a municipal waste incineration plant: Effects of dynamic process control measures and emission reduction devices, Environmental Science Technology, 37, 4761-4770, 2003.

Moore, K., Krudysz, M., A., Pakbin, P., Hudda, N., and Sioutas, C.: Intra-community variability in ultrafine particle number concentrations in the san pedro harbor area (los angeles, california), Atmos Environ, Submitted, 2008.

Morawska, L., Johnson, G. R., He, C., Ayoko, G. A., Lim, M. C. H., Swanson, C.,

ACPD

8, S7404–S7414, 2008

Ristovski, Z. D., and Moore, M.: Particle number emissions and source signatures of an industrial facility, Environmental Science Technology, 40, 803-814, 2006.

Morawska, L., Ristovski, Z. D., Johnson, G. R., Jayaratne, E. R., and Mengersen, K.: Novel method for on-road emission factor measurements using a plume capture trailer, Environmental Science Technology, 41, 574-579, 2007.

Petzold, A., Hasselbach, J., Lauer, P., Baumann, R., Franke, K., Gurk, C., Schlager, H., and Weingartner, E.: Experimental studies on particle emissions from cruising ship, their characteristic properties, transformation and atmospheric lifetime in the marine boundary layer, Atmos Chem Phys, 8, 2387-2403, 2008.

Ronkko, T., Virtanen, A., Vaaraslahti, K., Keskinen, J., Pirjola, L., and Lappi, M.: Effect of dilution conditions and driving parameters on nucleation mode particles in diesel exhaust: Laboratory and on-road study, Atmos Environ, 40, 2893-2901, 2006.

Ronkko, T., Virtanen, A., Kannosto, J., Keskinen, J., Lappi, M., and Pirjola, L.: Nucleation mode particles with a nonvolatile core in the exhaust of a heavy duty diesel vehicle, Environmental Science Technology, 41, 6384-6389, 2007.

Singh, M., Bowers, K., and Sioutas, C.: Seasonal and spatial trends in particle number concentrations and size distributions at the children's health study sites in southern california, Journal of Exposure Science and Environmental Epidemiology, 16, 3-18, 2006.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9641, 2008.

ACPD

8, S7404–S7414, 2008