

Interactive comment on “The time evolution of aerosol size distribution over the Mexico City plateau” by L. I. Kleinman et al.

Anonymous Referee #3

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The paper presents aerosol number and volume size distributions from measurements made on an aircraft during the MILAGRO experiment in Mexico City. The paper is to some extent a sister paper to the Kleinman et al., 2008 paper which used the same measurement suite to discuss chemical transformations of particulate in the plume of Mexico City and related these to a measure of the photochemical ageing of oxidised nitrogen. The present paper uses the same relationships to investigate how the aerosol number and volume distributions are transformed downwind of the city. The paper discusses the extreme cases of condensational and volume limited growth of the particulate and is careful to argue the caveats to the approach whilst highlighting the advantages of the simple analysis. The conclusions and behavioural analysis of how an aerosol size distribution changes with age in such a plume and the implications

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that has on the organic fraction of the aerosol mass are well explained and the paper certainly contributes significant new material. I certainly recommend it for publication in ACP. I do have some comments that I believe would add to the paper.

Page 1624, line 25 and following: Whilst there is a detailed discussion of how D_p was obtained from D_{va} for the AMS data there is no discussion of how scattering diameter as measured by the PCASP was transformed into D_p . This is important and ought to be discussed and/or referenced as line 16 on page 1629 and the discussion at the foot of page 1630 demonstrate. The independence of the PCASP, AMS and/or DMA regressions will to some extent depend on how this was performed.

Page 1625; line 1 The c ToF AMS acronym has not been defined.

Page 1628; line 16-22: AM and PM data are separated to investigate time of day effects. I can imagine that temperature and mixing effects are important diurnally and hence there are differences in time of day no matter where in the plume the measurement takes place. However, one can also envisage that the emission rates into the urban plume also vary diurnally. By segregating the measurements as a function of time no matter how far the measurement is made from source analysing morning and afternoon separately an implicit assumption is made that the diurnal influences on aerosol once formed are much larger than the diurnal variability of the sources. No evidence is given that this is indeed the case as far as I can see. It would be good to see such evidence or if it is not possible a discussion of the assumptions made should be included.

Page 1632; line 9: I can how the intercept of the regressions of figure 5 are used to derive background concentrations but I cannot see how the slopes are involved in this analysis.

Page 1632: line 24-25: The variability in the Aitken mode between day and night appears to suggest that source variability has a major influence, the authors need to comment on this.

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Page 1633; lines 17-23: The authors don't discuss the how the inorganic changes with photochemical age at all. Given that the authors note differences between the AM and PM behaviour of the organic fraction compared to the total volume it would be useful to see the contribution inorganic aerosol are making to the change in behaviour.

Page 1634: The near source CO data in table 4 certainly suggest that there is a significant diurnal variability in source strength and hence most likely a significant variation in primary aerosol number than explains some of the Aitken mode number concentration in the plume. The PM data in the far field are likely to be similar air masses to the AM near source data if indeed the paper assumptions are correct and the plume is from the city source only. Can the authors speculate the impact this would have on their AM/PM variations in some of the earlier figures, it may help to interpret the seemingly high ratios in the far field PM data compared to those in the AM. This could be folded into the discussion at this point.

Page 1638; end of page: Whilst extreme cases are discussed, it ought to be pointed out that between these extremes a continuum of vapour pressures exist and therefore characteristic timescales that cover several orders of magnitude. In reality it is unlikely that a parcel in such an environment is ever in full equilibrium across the whole aerosol population. Page 1639; lines 17-18: Coagulation of small particles to the accumulation mode is unlikely to be significant, however the authors should demonstrate that this can be ruled out.

Page 1640; lines 27-29: The phrase condensation growth reproduces the main feature of the aging process; namely that increased volume is caused by more particles not larger particles should be qualified. The authors mean that there is an increase in accumulation mode particles. Table 5, I am sure, should also refer to accumulation mode particles but does not. It would be helpful if the authors defined what was meant by accumulation mode when making these statements.

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Page 1641; lines 11-14: Entrainment of residual air containing aerosol aged during the previous day from aloft into the boundary in the morning may also give rise to the presence of near field and far field aged aerosol in the morning and should be mentioned.

Page 1642; line 2: It is true that the VMD doesn't change. However, that is not because the distribution is stationary it is because the increase in diameter of the particles initially in the accumulation mode is offset by the influx of smaller particles into the accumulation mode. The VMD averages these two modes and hence remains close to constant. This should be reflected in the discussion though I see that it is raised in the conclusions.

Page 1642; line 20: This is the first discussion of inorganic species. It would be good to relate these changes to the organic changes earlier as the difference in ageing profile between AMS total volume and the AMS organic mass are significant and must be due to the inorganic fraction.

Page 1643; line 4: The key point here is that the authors can only measure the net change in organic mass on the particulate. It is highly likely that as the aerosol ages, partitioning to the gas phase as a result of dilution does occur but in this case is smaller than the net condensation presumably resulting from either the formation of lower volatility products in the gas phase or from particulate organic reactions as the air ages. This should be stressed in this discussion.

Page 1656; table 5: The number concentrations should be referred to explicitly as accumulation mode number concentrations.

Page 1659; figure 3 caption vs CO should be per CO as defined in the text as it refers to a specific methodology defined by the authors

Page 1660; figure 4: It is conventional that the axis labels appear at the bottom of the lower panel.

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