Atmos. Chem. Phys. Discuss., 11, C13699–C13704, 2011
www.atmos-chem-phys-discuss.net/11/C13699/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The variability of urban aerosol size distributions and optical properties in São Paulo – Brazil: new particle formation events occur at the site" *by* J. Backman et al.

Anonymous Referee #1

Received and published: 30 December 2011

The manuscript presents an aerosol climatology based on 3 months of observations during the late spring/early summer in the city of Sao Paulo, Brazil. The observations include aerosol number concentrations, aerosol size distributions between 0.8-800 nm, and aerosol light scattering and absorption coefficients. The analysis also includes data from a nearby SO2 monitoring site. The analysis includes comparisons to previous observations, including heavily populated areas such as New Delhi and Beijing, examinations of the diurnal and weekday patterns in the observations, and an investigation into new particle formation events observed at the site.

The methods are described adequately and the analysis is routine but sound. The

manuscript would benefit greatly, however, by clearly tying the observations and accompanying analysis to the motivation for the study described in the introduction: the high use of biofuel ethanol in the study location. The introduction stresses that Sao Paulo is unusual due to the large contribution of biofuel ethanol used in the vehicle fleet and worthy of study to understand the role increased ethanol use could have on air quality and climate. The analysis presents a number of observations, but never links any of them to ethanol use. Comparisons are made to other large cities, such as New Delhi and Beijing, but it is not clear if the observed differences are due to differences in city size, sources or fuels. The analysis of new particle formation focuses on the role of sulfuric acid (from the photo-oxidation of sulfur dioxide), but does not discuss how increased ethanol use in Sao Paulo might affect sulfuric acid concentrations (e.g., via changes in hydroxyl radical chemistry associated with a different mix of VOCs compared to a region with less ethanol use).

Much of the analysis describes well-established concepts regarding aerosol dynamics, their relationship to optical properties and new particle formation. The manuscript points to several previous studies in heavily populated and polluted areas where similar phenomena have been observed, so the novelty of this study lies on the unusually high ethanol use in the study area. The manuscript should be significantly revised to focus on how high levels of ethanol use affect the observations, as well as the comments listed below, before I can recommend it for publication in ACP.

General comments

Since the data indicates a weekday/weekend effect in Sao Paulo the diurnal patterns may be more clear by restricting the analysis to weekdays only to better isolate the traffic effects. It would also be interesting to see the analysis of the weekend/weekday pattern extended to additional observations besides number concentrations, most notably black carbon.

Comparing black carbon observations to those reported for Mexico City (Baumgardner

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et al., ACP 7, 2517-2526, 2007) might be a useful exercise, particularly w/ respect to the higher ethanol use.

The manuscript would also benefit from further copy editing to improve the grammar. Some examples are listed below, but it is not an exhaustive list.

P30420, L22: "This indicates that also other condensable..." P30421, L08: "The aerosol particles ability to affect..." to "The ability of aerosol particles to affect..." P30422, L12: "Ethanol as an additive to fossil fuel increase aldehyde emissions..." P30246, L14: "were calculated" to "were converted" also P30427, L21: "When the CS is low, also small condensable..." P30427, L24: "the vapour abundance...need to be larger" P30431, L7: "behaviour associated of the aerosol dynamics..."

Specific comments

P30422, L12-16: It would be useful to include here any references that discuss differences in particle and/or ozone yields of aldehydes to other VOCs to aid the interpretation of the observations in a region expected to have higher aldehyde emissions relative to other VOCs due to the very high use of ethanol in fuel.

P30423, L15: Sample line lengths, material, internal dimensions and flow rates (or residence time) for the different systems should also be provided here.

P30424, L10-13: The decision to omit the other two SO2 monitoring sites needs to be supported in more detail. How far in km were the other sites and in what directions (could be referred to on the map). Were the stations in different surroundings (e.g., industrial)? What metrics were used in the authors' assessment?

P30425, L2-3: Might be better to move this paragraph to the instrument calibration section. Please also provide method used to calibrate flows.

P30425, L10: Should "ion" also be capitalized?

P30425, L28: Please provide a reference for the inversion method of the NAIS.

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P30426, L3-4: “optical properties tracked. . .black carbon mass” Suggest changing this to “optical properties tracked were light scattering coefficients measured by a nephelometer (Model 3563, TSI. . .) and light absorption coefficients converted from black carbon (BC) mass concentrations reported by a Multi Angle Absorption Photometer (MAAP, Model. . .).”

P30426, L11: ambient RH or RH measured in the instrument?

P30426, L19: replace “nephelometer data” with “temperature and pressure measured in the nephelometer”

P30427, L9: Reporting the diffusional losses in the driers as equivalent lengths of tubing suggests the authors applied a correction for losses, but this isn’t explicitly stated in the manuscript. Was a loss correction applied to the reported size distributions?

P30429, L11-13: Was this for the study period or does it describe typical climate pattern?

P30429, L15+: Number concentrations reported as 20.1 x 103 and 6.28 x 104 (and elsewhere) throughout these two paragraphs. Format should be the same unless significant digits change to reflect uncertainty in the values reported.

P30430, L5: Change “edges” to the particular statistic being referred to (e.g., 5th and 95th percentiles, min/max).

P30430, L23: Please define “median particle number size distribution”. Is this the size distribution corresponding to the median number concentration, the distributions of the medians in each bin, or something else?

P30432, L12: “for carbonaceous aerosol” Does this refer to mass or number?

P30432, L13: Should this be “peak at the same time”? The fact that both scattering and absorption have peaks does not necessarily mean that primary aerosols are driving them. For example, the reference cited describes observations in Mexico City

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that highlight the importance of secondary rather than primary aerosols for scattering properties (see their Figure 10). The statement is also contradicted by P30433, L1 "After the morning rush hour, when convectional mixing increases and photochemistry comes into play, SSA experiences a gradual increase. . ."

P30432, L26-28: The reasoning here is difficult to follow. What exactly is meant by "small in size"? Smaller than the non-absorbing particles? What evidence supports the conclusion that soot aerosols in the primary fleet are dominant in number? Is there a strong correlation between absorption coefficients and aerosol number? Both have peaks in the morning, but by 12 LT the absorption coefficients have already returned to near "background" values (Figure 5a) while number concentrations are still considerably elevated (Figure 2a).

P30433, L8: "The low SSA values presented here indicate that the City of Sao Paulo has a direct net warming effect on a regional scale." This is a strong statement to make on the basis of a measurement at a single location for 3 months of the year. Additional SOA formation downwind of the city (either due to changes in gas-particle portioning with changes in temperature and RH or additional photochemistry) could increase the SSA above the 0.85 critical value, which itself depends on a large number of factors including surface albedo, aerosol distribution and clouds. Also, what is meant by the "City of Sao Paulo"? The city also is responsible for other emissions, such as those needed to generate the electricity it consumes, that are not necessarily represented by the measurement location. Perhaps the statement could be changed to "aerosols emitted sources within the City of Sao Paulo have a direct net warming...".

P30433, L10-16: As written, it is not clear how the discussion of Angstrom exponents adds to the manuscript since size distributions were measured directly. Since the nephelometer was behind a 2.5 um sizecut the reported values cannot fully capture the possible contributions of dust and sea salt regardless.

P30434, L25: Another possibility that should be stated here is that the conversion

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technique used to calculate the H2SO4 proxy from SO2 measurements was off since it was based on a different site, as noted earlier in the manuscript.

P30435, L7: Suggest changing “As both the” to “As the timing of the . . .”

Figures and Tables

An additional table summarizing the study averages (for N, etc.) and comparisons to other locations might be useful.

Fig 1: Suggesting shading roads a different color or grey to distinguish from other labels

Fig 2: Write out abbreviations. Add x-axis label. State what the "statistical edges" of the data are (min/max?). What are the red points?

Fig 3: Not sure why this needs to have a log-scaled y-axis.

Fig 5: panel label locations are too close to the data, move to top left and adjust axis scales if necessary.

Fig 6: Legend is covering some of the data. Suggest adding "black circles, which indicate peaks of the size distribution" to the caption.

Fig 7: Legend covering some of the data. X-axis label is missing.

Fig 9: X-axis label missing. Caption should describe data source for the number time series. Would be helpful to have size ranges for different N concentrations listed in legend or caption (e.g., DMPS (X-800 nm), CPC 3022 (D > X nm), ...).

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 30419, 2011.