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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical Note: In-situ quantification of aerosol sources and sinks over regional geographical scales" *by* G. Buzorius

G. Buzorius

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Anonymous Referee #2

Received and published: 12 February 2009

General comments

Manuscript by Buzorius is a continuation of the paper by Buzorius et al. (2006) where the aircraft eddy correlation flux measurement system was presented and verified. This paper enlarges on the aerosol particle flux measurement results. To me it seems that the results presented here could have been squeezed in to Buzorius et al. (2006), but the author have chosen to make two separate papers. Also, the Buzorius et al. (2006) is more technical paper than this which is written as a technical note. However, the results presented here are promising and interesting. Hence, I would recommend ac-

ceptance of the manuscript and have only a few minor comments. **I would like to comment that when 2003 study was conducted and published in 2006, at that time it seemed that there will no opportunity to conduct more similar measurements. During the review of 2006 paper, referee asked to remove pollution episodes into separate manuscript (that become continental example in this manuscript). When measurements were repeated in 2006 we found out that (1) conditions that were qualified as "clean marine environment in 2003" were actually polluted marine environment; (2) really clean episodes were hardly observed in 2003 and much more abundant in 2006 study. Thus, a better example of the clean marine environment was presented here, along with the several plumes in marine environment (typical observation for 2003 study) and the continental example.**

Technical corrections

In general, the writing should be improved throughout.

Introduction: Page 1303, lines 13-17: few references are needed after both of these sentences. **Added. Fountakis et al. (JGR 2007) See referee #1 comment.**

Experimental setup: The purpose for the CPC modification should be mentioned here, although it could be guessed by experienced reader**. It was mentioned in the previous paper**.

Methodology Page 1306, line 9: It seems that during the plumes aerosol concentration could change almost like a step function. Is the linear de-trending applicable and reasonable to use also in those situations? **Generally, it is better to use the filter. If decision was to use the filter, it is better to stay consistent. Of course, there is always possibility to go back to raw data and analyze it in more detail. I checked using the raw data from the entire flight to see how de-trending affected concentration time series and the flux values. (1) The de-trending created a slope in**

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the de-trended series unless the spike is in the middle of the data time series for which covariance is calculated. (2) Skipping de-trending from flux calculation created erroneous flux values (rapid fluctuations in both directions with unrealistic values) on other segments (with no plumes) from the same flight. (3) If de-trending was not used, the plume flux values would be smaller by a factor of 2 to 3 than presented in figure 2. Cancelling the filter created more problems: fluxes were so fluctuating outside the plume that the plume flux could not identified from flux time series alone. Ideally, for a step change in concentration, data stream should be split in two segments: prior and post-change. Both segments can be de-trended separately, and conjunct for covariance calculation.

Page 1306, line16: Repeating the measurements in the same area reduces the random errors only if the source/sink remains the same. **Yes, for example by flying over the same area within an hour gives a reasonable expectation of the same meteorological conditions for the certain cases.**

Page 1308, line 23: Abbreviation SST is not explained. **Corrected in the text. It is Sea Surface Temperature.**

Results and discussion: Chapter 4.3 could be shortened. Now it mainly concentrates to present the results by others providing only average aerosol number concentration and flux range measured by the author. **This chapter became chapter 4.1 as suggested by referee #1. This chapter discusses background conditions and provides a "frame" in which the study results are presented in relation to the prior published datasets.**

Page 1316, line 7: Reformulate the sentence starting with "It is typical to observe..." It is fuzzy. **Corrected**.

Conclusions and summary I wouldn't say that "This study increases the range of appli-

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cations

of EC..." and "The presented method is...", because the method was already

presented in Buzorius et al. (2006). **The range in this paper is increased not in terms of the flight distance but in terms variety of scientific applications. In this paper fluxes are resolved at distances over few kilometers that is comparable distance to EC measurement from tower when sampled at the same altitude. In addition to typical (for tower sampling) planetary surface-atmosphere exchange fluxes, here flux at the relatively higher elevation was observed, where the flux was controlled by the mixing of two different air masses. Such measurements were not reported in previous paper. Changed to "variety of scientific applications"**

References: Buzorius, G. (2001) is in volume 35, not 25. **corrected**

Anonymous Referee #3

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The manuscript presents interesting study on airborne eddy covariance flux measurements over the ocean and land. The measured particle concentrations vary greatly occasionally due to single (ship plumes) or more extended (highway or cities) sources. This is the challenge for the technique which assumes stationarity in time or homogeneity in space. Filtering could reduce to some extent the problem but on the other hand this would result in underestimation of fluxes since transport by large eddies would be excluded in calculated co-variance. The author recognizes sufficiently the uncertainty arising from non-stationarity in interpretation of results. After improving spatial resolution the author verifies that the calculated fluxes are in good agreement with larger scale averages. The results are interesting qualitatively as well as the technique could be very useful for exchange studies of atmospheric aerosols. The manuscript is suitable for publication in ACP after addressing the comments below. Page 1304 lines

7-9 would be useful to indicate directly that the study aims to present fluxes with 3 km resolution compared to 10 km resolution in previous study. **Corrected by inserting in Line 8 "compared to 10-km resolution in the previous study"**

Page 1306 line 8, it is stated that over bar means an average over the 200 or 60 s time period. Indicate also that these two time periods correspond to 10 km and 3 km resolution with the average airplane speed. **A sentence added "200 s and 60 s time periods correspond to 10-km and 3-km resolution with the averaged airplane speed."**

Page 1306 lines 11-18 about errors. Two first errors are described as random. Mention also that the third error is systematic in nature and give typical value with reference. **In this study attenuation of fast fluctuations in aerosol concentration time series were corrected by "speeded-up" method. The method was presented in the previous paper. The typical error estimates are usually given based on Horst formulae, but those estimates are not applicable for "speeded-up" method.**

The same page lines 21. Explanation of footprint should be better formulated although exact definition is not necessary. 'The sea surface fetch' is not definitely the footprint. **Corrected. This was notified by other referees as well.**

Page 1308 lines 4-6. Is the Webb correction due to temperature fluctuations applicable here since 1 m long conductive tubing was used? 1 m long metal tube could rather well damp temperature fluctuations and in that case the respective corrections should no be done. This seems not to be of big practical importance since the corrections are relatively small as reported in results section (page 1309 lines 24-27). **Used tubing is electrically conductive but not metallic (it is TSI Inc. manufactured soft, non-elastic-rubber type tube with part number 3001788, this information was given**

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on page 1305 line 15). Electrical conductivity reduces/eliminates static chargers on tubing walls and subsequently reducing losses of electrically charged particles. Poor heat conductivity is expected for that material. Yes, in this study corrections were small.

Page 1313 lines 5-6 the downward fluxes inside the bay, western side of the pollution plume. What would be the deposition velocities of aerosols to sea surface and could the observed downward fluxes be rather the result of downward mixing of plume in strong vertical concentration gradient conditions? **In that area aerosol concentration is dropping from several thousands to below thousand of particles per cc. To answer the comment I picked a data point at -122.09 longitude and 36.853 latitude, the running (60 s) mean concentration is 1330 particles/cc. that point is located on -10 flux-line in figure 6. the ratio of both gives about 7.6 mm/s deposition velocity. From 2003 experiment it is known that Aitken mode dominates aerosol size distribution (more specifically diameters $<$ 50 nm) in this sampling area. Additionally, the vertical spiral during the same flight several minutes later performed at -122.16 longitude and 36.94 latitude showed steady (289.2$\pm$0.3 K) virtual potential temperature from 33 m to 120 m elevation, while aerosol number concentration was about 700$\pm$120 particles/cc from 33 m to 80 m where it started increasing and reached 1300 particles/cc at 120 m elevation. Due to vertical gradient above 80 m elevation it can be argued in favor of the downward mixing, but there was no significant gradient below 80 m elevation. Also, the atmosphere was well mixed, and measured deposition velocity had similar value to the expected dry deposition velocity.**

Line 1314 line 9 'measured fluxes' in parenthesis is confusing. **Its measured flux in the text, flux in singular. Measured flux represents the sum of dry deposition flux and emission flux. If both are present, the measured flux is a sum of both.**

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Page 1315 line 16, what does it mean decoupling of virtual potential temperature? Was it strong gradient as an indicator of inversion layer, i.e. a layer between two decoupled layers? **Yes. Text was corrected by moving sentence from line 13 page 1316, where it was explained that the temperature jumped from 292 to 294 K aloft.**

Page 1316 line 16-17, an uptake of particles is a confusing expression because uptake did not occur but the upward transport. **The sentence was rephrased. It was upward flux due to dilution aloft.**

Page 1318 lines 3-6 the sentence is not clear. **To assure that the largest atmospheric eddies were sampled, it is important to limit applications to environment where the largest eddies are several times smaller than the spatial resolution. The chosen criterion was the limit on mixed boundary layer height being several times smaller than the spatial resolution. Beginning of the sentence was changed to "Measurements were conducted under ambient conditions. . . "**

Figures 3, 5 and 6, was there any interpolation technique used in figure consruction in addition to averaging of fluxes to 0.035x0.035 degrees cells? If yes, then mention.**yes, the color of each cell resulted from bilinear interpolation of the color of its four vortices. Specificially Matlab function PCOLOR was followed by function shading('interp'). A statement was added to the text on page 1311 line 28 explaining that. Also found a mistake: figure 3 used 0.035*0.035 cells, while figures 5 and 6 used smaller 0.025*0.025 cells. Corrected correspondingly.**

Anonymous Referee #4

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The manuscript reports airborne eddy covariance flux measurements over the ocean and land in the atmospheric mixed layer. One of the innovative aspects of this study raised by the author is an improvement of the spatial resolution to 3 km segments in

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measuring airborne aerosol flux. The choice of the spatial resolution is very challenging and it should be done considering the ability to capture almost all the turbulent flux co-spectral modes and the spatial homogeneity which is a basic requirement of the eddy covariance technique. Somehow the author acknowledges the systematic and random uncertainty due to the shorter spatial resolution and non-stationarity of the turbulent time-series. However the author needs to address the detailed comments below, before the manuscript is suitable for publication in ACP.

Page 1304 line 23. remove "and" after the comma, e.g. "below the flown altitude, correlating the aerosol fluxes...". **removed**

Page 1305. The experiment setup section should include a better description of the experimental sites. I would change the title of this section into "The site description and the experiment setup". I would start with the description of the sites and then proceed with the experiment setup. The lines 6-13 in the section 4.4 give a description of the continental site and then should be put here and not in the results part. **The section was modified by adding a descriptions for vertical profile samplings and continental site. More detailed site description is in previous paper.**

Page 1305 line 9 add "(CPC)" after "condensation particle counter". **Corrected.**

Page 1306 line 5 remove "shown in Eq.(1) below". **removed**

Page 1306 lines 7-9 are fragmented, please reformulate this sentence. Add also the corresponding spatial averaging (10 km and 3 km) to the mentioned averaging time periods. **Changed and added as suggested also by another referee.**

Page 1306 line 12 replace "condensation particle counter (CPC)" with "CPC". **replaced**

Page 1306 lines 11-18. About the random flux errors. Those errors are due to vertical

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wind speed error and limited counting statistics. The author reports an estimation of the flux error equal to 0.1 x 1e6 # m-2 s-1 referring to Buzorius et al., 2006. The author should clarify if in this study such flux error estimation is for a time window of 60 or 200 sec. Assuming that the std of w and c would be the same for 60 and 200 sec (which probably is not true), the error flux estimated according to Buzorius et at, 2006 equations would increase by a factor of 2 reducing the time window from 200 to 60 sec. The difference is small, but please comment on that. **Added: " Improvement in spatial resolution to 3-km increased random error for each single flux data point by a factor of sqrt(200/60)≈1.8". spatial averaging across latitude-longitude cells in figures 5 and 6 reduced the error.**

Page 1306 line 11-18. About the systematic error. The author should give typical value for this error, which is due to limited response time of CPC. Since few lines below the author refers to Buzorius et al., 2006 for a detailed discussion of the measurement error, I assumed that he used the same approach to correct the fluxes in this study, e.g. the flux correction for high frequency loss amounts to about 30%, as derived by the Horst (1997) formula. Buzorius etal. (2006) shows that the wT co-spectrum (Fig. 7g) and the wc co-spectrum (Fig. 7h) follow the expected surface layer scaling at high and low frequencies, then the use of Horst(1997) formula could be justified. However the normalized frequency nm at which the co-spectra peak is about 0.2, then larger than the theoretical surface layer value (0.085) for neutral and unstable stratification. Using the actual nm value (0.2) in the Horst(1997) formula, the flux correction for high

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frequency loss increases to about 50%. Since this systematic error is quite large, the author needs to check this issue and eventually recalculate the fluxes accounting for the actual correction factors. **The sentence was added to 3.1 chapter to specify that "speed-up" method was applied. The method is independent of atmospheric turbulence spectra; therefore, it is superior to Horst model (for comparison between both methods see the previous paper). Therefore the comment is irrelevant.**

Pages 1306-1307. Footprint. The first two sentences of this section could be removed, since a general definition of footprint function is not necessary. The rest of the section should be better reformulated, and the author should give footprint estimation for both the marine and continental sites. **Beginning of the chapter was corrected. It was noted by other referees as well. Footprint estimates were given in page 7 lines 2 to 9 for marine site, and line 14 page 1315 for continental site.**

Pages 1307-1308. Webb correction. In my opinion also this section (included the Eq.2) should be removed, since an explanation of the Webb correction is not necessary. The author could briefly discuss it at the end of section 3.1, giving the correction percentages

for the marine and continental sites. **Eq 2. was used in presented results. It is preferred to demonstrate equations that were used in work.**

Page 1308 Line 11. Why "two atmospheric scalars"? **Atmospheric scalar is a general term used in eddy-correlation literature for gasses and particle concentrations, wind speed components, temperature. The presented statement in that paragraph applies to various flux measurements not just aerosol flux measurements.**

Page 1308 Line 19-25. You should mention in the methodology section (3.1) that the

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flux are calculated using a "kind of moving average" as is explained in Buzorius etal (2006), otherwise the reader cannot understand the Fig. 1, where over a distance of 40 km (800 sec) you have so many points representing fluxes averaged over 5 km (100 sec) and 3 km (60 sec). **yes. Page 1306 line 10 was added with sentence explaining that.**

Page 1309 Lines 24-27. can be moved to the methodology section as I suggested above. **Those lines present results of Web correction calculations. Since they part of results, they were left in results chapter**.

Page 1310 Lines 15-20 How the linear detrending works in presence of such sharp rise in aerosol concentration?**referee #2 had the same question. Answered there.**

Page 1311 Lines 9-11. What are the estimated values of the effective source strength? **To estimate the effective strength in units of particles emitted per second, additional transect across the plume in perpendicular direction compared to the first one, is required. Assuming the identical flux distribution across the plume in both directions, the averaged flux was 54e6 and 14e6 particles per mˆ2 per s for 3-km and 10-km resolution respectively. Plume width was about 200 m (4 seconds at the base in concentration time series, whereas at half height about 3 seconds or 150 m). 10-km resolution flux data point contains concentration time series that are about 196 seconds outside the plume. For comparison purposes 10-km resolution averaged flux 14e6 is compensated for the difference in footprint size by multiplying by (200-4) and dividing by (60-4), the result is 50.69e6 particles per mˆ2 per s, less than 10% difference from 54e6 value that was obtained from 3-km resolution. Continuing with later number: assuming that plume occupies horizontal area of 4e4 mˆ2, the estimated effective strength is about 2.15e12 particles per second. this estimate involves several assumptions, therefore is left-out from the manuscript, since future analysis is required.**

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Are those values reasonable for what you expect from ship emission rate? **More research is required to provide reliable comparisons with other studies. It is important to remember that this number was obtained measuring flux with 3-km resolution while source cross-section distance was about 200 m. it is not known how well turbulence field (vertical wind speed) in 200 m distance represents the entire spectrum average. On the other hand, the provided number represents flux in vertical direction for the time moment when the airplane crossed the plume, while plume is spreading horizontally as well.**

Page 1315 Lines 7-16 I would move these lines to the site description and footprint sections, as I earlier mentioned. **Moved.**

Page 1315 Lines 16-18 I would remove the sentence about the large eddies from here. **removed**

Page 1316 Line 3. Remove the sentence "The aerosol number concentration was sampled at 10 Hz". **Removed.**

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1301, 2009.

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