Atmos. Chem. Phys. Discuss., 7, S1091–S1095, 2007
www.atmos-chem-phys-discuss.net/7/S1091/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "A study on the relationship between mass concentrations, chemistry and number size distribution of urban fine aerosols in Milan, Barcelona and London" *by* S. Rodríguez et al.

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Received and published: 10 April 2007

Authors Comments - Final Response including the answers to the questions raised by referee#2.

Before starting with any detail, we would like to thank the comments of all anonymous referees. We feel that their comments have made us to reflect on some specific aspects of the results of this study. On one hand the comments have leaded to synthesise and highlight the most important results of the study, and on the other hand, these comments have leaded us to enrich the some parts of the discussion of other part of the manuscript. These comments and suggestions have really contributed to improve

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the manuscript and make it more useful and interesting for future readers.

In this FINAL RESPONSE phase, we include the answers to the questions raised by the referee#2 (which were not replied separately).

The main motivation of this study was to investigate what the relationship between the "chemical composition" and "physical properties derived from the number size distribution" of the fine PM2.5 particles is. Although these two topics have been subject of research in a number of previous studies, this had been done separately, in such a way that there was a gap between the knowledge on the urban aerosols obtained by two very important branches of the aerosol research field: "PM2.5 chemistry and source apportionment" and "physical properties and number size distributions". In order to provide a general view of the aerosol features in urban areas, in this study we have focused on the general relationships between the different aerosol parameters observed at the three study cities (MILAN, BARCELONA & LONDON). In the final version of the manuscript, a part of the Introduction has been reworded, as suggested the referee#2, in order to highlight the "motivation of this research" and to highlight "what was missing on the aerosol studies and what is new in this article".

The study has been performed in three cities of Western Europe: MILAN, BARCELONA & LONDON. We fully agree with the referee#2 when he says that "the three sites are not completely representative for whole Europe". In fact along the article we always refer to the three study regions as representative of different regions of "Western Europe" (even not all the type of regions of Western Europe are covered). We know that data from Central Europe, Eastern Europe and Scandinavian countries are available for this period; however, these were not included in the article because data on "PM2.5 chemistry" obtained by using the same methodology were not available. In the European Aerosol Phenomenology performed by Putaud et al. (2004; A European aerosol phenomenology, report EUR 20411), it was observed that comparisons between the aerosol composition across Europe was not "obvious or easy" because a number of methods are in used in across Europe for the aerosol sampling and chemi-

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cal analysis. This is because this campaign in MILAN, BARCELONA & LONDON was designed by using the same methodology at the three sites.

We agree with the opinion of several referees: the amount of information included in the manuscript (and specifically in some Figures, such as 3 and 6) is really large. With regard to this, the suggestions of the referee#2 have really been very useful for us. Thus, in the final version of the manuscript, the number of subfigures in the Figures 3 and 6 have been reduced, and the section "4.4 Seasonal evolution and PM2.5 events" has been divided in several subsections (4.4 PM2.5 events, 4.4.1 Urban PM pollution events, 4.4.2 Saharan dust events and 4.5 Seasonal evolution). With this modification the discussion is organized more clearly. The management of this large amount of information on aerosol chemistry and physics is really not easy, however once this is organized in a proper way it provides a comprehensive picture on the involvement of the microphysical processes on the urban aerosols evolution in daily, day-to-day and seasonal basis.

Finally, the comments of the referee#2 on the "summary and conclusions section" encouraged us to slightly reorganised this section for highlighting the major findings of this study, which in our opinion include: 1) the identification of the processes leading the different behaviour of the mass and number concentrations in a daily, day-to-day and seasonal basis, 2) the different association of some aerosol chemical components with different particle number concentration in different size range (e.g. OM and BC with ultrafine particles, and the other compounds with particles $>$100nm), 3) the microphysical processes involved in the day-to-day changes in the aerosol composition and size distribution, and 4) the nature of the PM2.5 winter pollution events (showing how the increase in the concentrations of the semi-volative species observed by the filter sampling fit very well with the increase in the size distribution modes).

In two parts of its report, the referee#2 asked if the results of this study lead to conclusion about if it is better to monitor the number or mass concentrations, or both. The reply to this question has been included in the "summary and conclusions section" of

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the final version of the manuscript (where it was added a new short paragraph in order to highlight how the results of this study may contribute to improve the urban air quality monitoring). These results shows how, in urban areas, the number concentration is mostly representative of “fresh ultrafine particles” (with a short life time) mostly linked to vehicle exhaust emissions, whereas PM2.5 is mostly representative of the “accumulation mode particles linked to the aged urban background aerosol”. These are two different forms of aerosol pollution. These results are important since the number and PM2.5 mass concentrations can be used as metrics for air quality monitoring exhibiting complementary properties.

Answers to other specific questions of the referee#2

-The Introduction has been slightly modified. Now references to other studies performed in central Europe and Scandinavian countries have been added (as suggested by this referee). We have performed a quick summary of previous studies; however the cited studies are not “revised” one by one, but all them in general, by stating “what have been done up to now”, “what is missing” and “what is new in this article”. The referee suggests talking about if mass concentration or number concentration should be monitored; this has been done in the summary and conclusions section.

-More details about the measurements sites in each city. In the previous version of the article, it was already stated that the study was performed in “central urban background” sites (i.e., not kerbside or street canyon). In the final version of the manuscript more details on the environment and surroundings of the sites are provided.

-About including other sites of central and northern Europe. As stated above, other sites of Europe were not included because data on chemistry collected with the same methodology were not available. In our measurements campaign we planned only Western Europe. We agree with the referee that it would be interesting to perform other study by including more sites, but it is necessary to rethink how to work with chemical data obtained with different sampling and techniques.

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-Details on the DMA and DMPS are provided in the final version of the manuscript. DMA of TSI (3071 model) were used by custom built softwares.

-the size distribution modes DpN were calculated simply as the diameter where dN/dlogD reached the maximum value. No fitting performed.

-> Page 619, line 10ff: Why is the correlation coefficient much higher for Milan? This question was also performed by the referee#1. With our data base we can not provide a definitive explanation for this. In Figure 9, it can be observed how the much higher correlation at Milan (with respect to the other sites) is enhanced in the particles size >100nm. We speculate that this may be the result of the confluence of several facts: 1) the number concentration of particles >100nm is much higher in Milan than in the other site, 2) mineral dust concentrations are much higher in Barcelona than in the other sites, and this mineral dust significantly contributes to the mass but not to the number concentration (because of the much high density of mineral dust when compared to that of other aerosol compounds).

-> I miss a comparison of the major results, e.g. numbers, correlations with previous studies. In the final version of the manuscript, comparison with other studies performed in Leipzig, Copenhagen Helsinki and Stockholm have been included.

-About the conclusions. As stated above, the “summary and conclussions” section have been significantly improved in such a way that now the most relevant findings, including those suggested by the referee, are highlighted.

-the manuscript has been reviewed again by English native speakers.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 605, 2007.