

We would like to thank to the anonymous referee #2 for his/her constructive comments, which definitively help to improve the manuscript. The replies to the referee's questions are listed below. Most of the suggestions have been introduced in the manuscript and will appear on its final version.

Comments are addressed following the order of the referee's questions.

Introduction:

Comment-1. Referee asked "Page 17754, Line 4: What do you mean by 'ultrafine' particles?. This word is used for very different size ranges, thus it needs to be defined in the beginning of each paper".

Reply (C1): The ultrafine particle definition has been added in the introduction of the paper. We define ultrafine particles as those with a diameter < 100 nm.

Comment-2. Referee said "Page 17756, line17: I think there are instruments available measuring below 2.5 nm (NAIS and so on, there is also one new CPC , measuring down to 2 nm, developed by Mikko Sipilä). This should be mentioned here".

Reply (C2): Thanks for this comment and for providing this information. This information will definitively appear in the final version of manuscript.

Measurements:

Comment-3. Referee said "Page 17759, Line 20ff: The measurement period is given here, but usually not all data are available all the time. Please give data coverage for the different parameters".

Reply (C3): The coverage of data for each parameter is shown below, indicating the number of valid cases and the availability of data related to sampling planned:

- N: 7310 cases. N~70%.
- BC: 10414 cases. BC~95%
- PM10: 8665 cases. PM10~80%
- NO<sub>x</sub>: 9111 cases. NO<sub>x</sub>~85%
- SO<sub>2</sub>: 9315 cases. SO<sub>2</sub>~85%
- O<sub>3</sub>: 9526 cases. O<sub>3</sub>~87%
- RAD, WD, WS, T<sup>a</sup>, HR: 10944 cases, with 100% data.
- Precipitation: 9541 cases. Precipitation~87%

Comment-4. Referee said “Page 17760: I do not believe that your CPC is measuring down to 2.6 nm. Did you measure the calibration curve? If yes, please show it. Otherwise you may refer to published calibration measurements, e.g. Hermann et al., 2007, J. Aerosol Sci., there the DP50 was usually above 3 nm.

Reply (C4): After reading this comment, we checked again the documentation (manual, brochure, etc...) provided by the CPC manufacturer (TSI™) and the papers published on this matter (including Hermann et al., 2007). It is really true that the part of the manuscript where the limit detection of our CPC is described should be clarified. In the manuscript we said that the detection limit was 2.5 nm because this is the information provided by the manufacturer. Unfortunately, we have never determined the calibration curve of the CPC used in this study, model 3776, SN: 70530189. However, at the beginning of the measurement campaign presented in this paper (April 2008) we performed a CPC inter-comparison. The inter-comparison of two CPC 3776 is shown in the Figure 1 (below). It can clearly be observed that the difference between the records of the two instruments is extremely low, ~0.2%. It allows to conclude that the CPC used for this study (SN: 70530189) has recorded the data according to the manufacturer technical setting. Now the question is: the manufacturer says that the lower detection limit of the 3776 model is 2.5nm, however, calibration curve determined by Hermann et al., 2007 have shown that the 50% efficiency for these instruments is reached at diameters ( $d_{50nm}$ ) between 3 and 4 nm (for NaCl and Ag particles). In order to clarify this, a text similar to this will be introduced in the final version of the manuscript (section: measurements):

Number concentration of particles was monitored by using an Ultrafine Condensation Particle Counter (UCPC, TSI™), model 3776. According to the manufacture, this instrument detects particles coarser than 2.5 nm. Experimental measurements performed Hermann et al. (2007) have shown that the 50% efficiency for these instruments is reached at diameters ( $d_{50nm}$ ) between 3 and 4 nm. The CPC unit used for this study (SN: 70530189) was intercompared with other unit (SN 70601252) of the same model at the beginning of the campaign, results shows that the differences were of about 0.2% for 10 minutes averaged data (Figure 1).

CPC3776: SN 70601252 vs SN 70530189

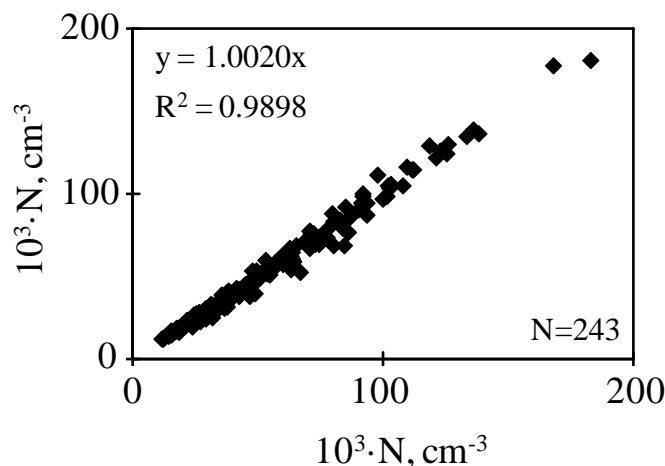


Figure 1. Number concentration of particles measured with two CPC-3776 units operated simultaneously in Huelva city during an inter-comparison. Data: 10 minutes averaged.  $N=243$ , number of 10-min data used in the plot.

Results:

Comment-5. Referee said “Page 17763, line: 11: 0 – 23 h should be 0 – 5 h.

Reply (C5): done

Comment-6. Referee said “Page 17764, line 15 ff: The method to segregate between directly emitted particles and those formed later from the gas phase is very interesting and may help to interpret other datasets. But, it is difficult (or impossible?) to distinguish between new particle formation from anthropogenic sources and others?! This has to be discussed in the paper. If the authors think this is possible a clear explanation needs to be given.

Reply (C6): The method used in this study is based on studying the degree of correlation between black carbon and particle number. It is then useful for distinguish between particles emitted by the vehicle exhaust (N1) from other none emitted by the vehicle exhaust (N2). The origin of the particle non linked to vehicle exhaust emissions (N2) may change site to site, and it should be studied specifically for each location. The methods is relatively new (described by Rodriguez and Cuevas, 2007, Aerosol Science 38 (2007) 1207 – 1219) and has only be applied in Santa Cruz de Tenerife and Huelva (urban sites, the latter affected by industrial emissions). In principle, we think this is useful for distinguish vehicle exhaust particles from other anthropogenic particles. We think that it would allow to identify biogenic sources. In the case of Huelva (this study), we try to show that “non-vehicle emission” sources may also contribute

significantly to ultrafine particles in urban ambient air and that in the case of this city, those other sources are related with industrial emissions.

Comment-7 a). Referee said “Interpretation of Figure 4: If N and BC are correlated new particle formation seems to play a minor role. This should be mentioned and discussed. In fact, the parallel BC measurements are a good way to distinguish between anthropogenic and other emissions.

Reply (C7a): Yes, the referee is right and these BC-N correlations or not-correlations are the principle of the method we used. In Figure 4 it can be observed periods in which BC and N are correlated, e.g., 18-23h in Fig 4D, in this period new particle formation is very low (Fig 5A). In contrast, during the 10-15h period correlation between BC and N is low (Fig. 4C) and new particle formation is very high (Fig 5A).

Comment-7b). About PC-Analysis, referee asked “Why did the authors not include meteorological parameters?”

Reply (C7b): We introduced several meteorological parameters, except wind. In Temperature and Relative Humidity can be observed in the PCA shown in Table 2. We only found an involvement of Temperature and Relative Humidity. We are now re-processing the data for introducing the two horizontal wind components (U, V) in the PCA. The results obtained will be introduced in the final version of the MS.

Comment-8. Referee said “Page 17767, line 28: PC1 should be PC2, please check also the rest of the MS for this!”

Reply (C8): In the PCA performed during the 11-17h period, nucleation of sulphate is observed in summer in PC1.

Comment-9. Referee said “Page 17768, line 1, ..the analysis: : : Which analysis do you mean here?”

Reply (C9): An analysis with data averaged in intervals of  $25 \mu\text{g NO}_x/\text{m}^3$  and  $1 \mu\text{gSO}_2/\text{m}^3$  was performed in order to reduce the other influencing parameters. We will check the text in order it can be clearly understood.

Comment-10. Referee said “Page 17768, line 7: N1 should be N2, please check also the rest of the MS for this!”

Reply (C10): In the manuscript N2 is written.

Conclusions:

Comment-11. Referee said “More interpretation concerning the different sources would be nice. E.g. is it possible to distinguish between traffic and industrial emissions? What is the major and new outcome of the study? It should be clearly stated what the advantage of this methods is and also where the limitations are!

Reply(C11). Thanks for this comment. We are now preparing the final version of the manuscript and this suggestion will definitively be added.