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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The role of mixing layer on changes of particle properties in lower troposphere" *by* L. Ferrero et al.

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To Referee 2:

REFEREE 2 GENERAL COMMENT: This article presents a set of size distribution and chemical composition measurements performed in Milan during 3 years. In a first step, a description on how the particle size distribution and chemical composition change with altitude over Milan city is presented. In a second step, the observed behaviour is described by a statistical model. To study how the size distribution changes with height is an issue that has rarely been investigated by airborne in-situ measurements. Although this is a topic of high interest, the results provided are not significant. I think

that a more elaborated data treatment could be performed in order to obtain significant results.

ANSWER:

Thank you for your useful comments.

In this respect the aim of this work is to describe, by a modelling approach, the typical particle size changes along height, within and above the mixing layer. These are shown under different meteorological situations (tab. 2) by a general law (eq. 6). This kind of knowledge is fundamental in remote sensing applications (Satellite, lidar, photometer. . .) to retrieve the main physical parameters of atmospheric particles (PM ground concentrations, optical properties and size distribution along height). Along the whole paper, the innovative points, not yet clearly resolved in literature as far as we know, are the following: 1. we find new relationships between Hmix and changes in particle size both for fine and coarse particles (fig. 5.b and 7) under stable conditions. This is a topic of interest because, as reported in the introduction (from pag. 16485, line 29, to page 16486, lines 1-6) the connection between the columnar aerosol size distribution and details of the vertical profiles is still under discussion. In this respect, as reported by Campanelli et al. (2003) and by Corrigan et al. (2008), the remote sensing (satellite and AERONET) approach need assumptions about aerosol physical, optical and chemical parameters, as well as homogeneity, which may not lead to valid results (Dubovik et al., 2000; Levy et al., 2004). A common basic assumption of all the above-mentioned algorithms is that the atmosphere is vertically homogeneous and that the aerosol optical characteristics are constant over the whole air column. Our results, collected in Milan, in the stagnant conditions of the Po Valley are totally new as regards the description of the relationship between aerosol size distribution and details of the vertical profiles under these meteorological conditions. This is also reported in the result and discussion section at page 16494 (lines 17-27), and in the conclusions. 2. coarse particles cannot be dispersed into the ML in the same way as finest ones especially during winter (fig. 5a; low wind speed conditions). This is a point of interest

because Hmix is usually used in literature (Levy et al. 2007; Di Nicolantonio et al., 2007; Liu et al., 2005; Sarigiannis et al., 2004) to retrive PM concentrations at ground from satellites (i.e. MODIS on Terra and Aqua platforms) as a proxi of the aerosol effective scale height (Heff) introduced by Kaufman and Fraser in 1983. The use of Hmix instead of Heff requires, for example, that an homogeneous mixing is present into the mixing layer. Figure 5a in the paper gives a first answer to the accuracy of a such approximation. High stability conditions can make the two fractions differently dispersed; in these cases the use of Hmix in PM retrieval algorithms can induce an error. 3. the present study proposes a statistical model for assessing the size distribution changes of the fine fraction along height (section 3.2), which is a basic trait of the work done, that proposes a general tool for handling similar data sets. All vertical profiles together enter in a probabilistic model, after standardising vertical profiles with respect to the ML height (proposed in section 3.2). In this model parameters are estimated, in a statistical sense, in order to permit to find a general law, valid for similar meteorological situations (tab. 2). Moreover the results can be expressed via credibility bands and not only via point values: this is allowed by managing model and data uncertainty via probability tools. The prototypal model we propose can moreover be used in any previsive situation. The basic ideas are the following: a) If we assume that, at any heights, the basic information is the OPC size class relative contribution to the total count, it has to be treated with appropriate statistical tools (i.e. compositional data analysis), this approach is proposed for the first time. b) A statistical model has been estimated separately on each of four homogeneous groups of launches, characterized by different external conditions. In this way typical vertical profile behaviours have been identified (fig. 9): this is a novel result. c) The statistical model at the same time takes into account compositions, heights and launches of the same group and arrives at modelling counts within the OPC size class. 4. the model developed is able to predict particle properties along height starting from ground measurements (aerosol and meterological) and ML height. This is reported in the section 3.2 and in the conclusions. In this respect, we have been able to compute the model counterpart of several experimen-

tal evidences and to confirm experimental conjectures. In particular, Tables 6 and 7 report the model counterpart of the average relative contribution of each size class in homogeneous sub-groups. The 95% credibility intervals of Table 7 contain the empirical average relative size distribution. This shows how statistical modelling permits one to compute measures of uncertainty of estimated values that render results more thorough than descriptive syntheses of data. Among the outputs of the hierarchical statistical model, also appear the estimated MPD values corresponding to the empirical values. The MPD growth estimated via the model is $1.6 \pm 0.4\%$ for group A; $2.2 \pm 0.5\%$ for group B; $-2.0 \pm 0.5\%$ for group C and $6.1 \pm 1.2\%$ for group D, in accordance with experimental evidences.

Finally we would remark that these results came from the collection of long-term data series (3 years, more than 300 profiles), using a thetered balloon approach, and thus avoiding the common limit of direct sampling along vertical profiles: the temporal significance of these measurements and their interpretations. Infact, in literature balloon size-distribution measurements where done mainly during short-peculiar sampling campaigns (Laakso et al., 2007; McKendry et al., 2004; Maletto et al., 2003; Stratmann et al., 2003).

REFEREE 2 SPECIFIC COMMENTS:

QUESTION 1) In several sections of the manuscript, it is provided the percentage of growth of the particles across the mixing layer. In many parts it sound some ambiguous. For example: - Abstract: “An increase in the mean particle diameter....in the summer”. Such growth is observed between two altitudes. I guess that between the ground and the top of the mixing layer. In my opinion, this should be described in the text, and the typical value for the mixing layer depth should be provided. - Section 3.1.1, pag 16492 – 16493: “mean reduction in MPD was 14.9% and 10.7% .....”. Between what altitudes?. Between the ground and top of the mixing layer?. The same applies when describing the particle growth of fine particles in section 3.1.2.

ANSWER 1: The fine fraction growth (FFG), and the coarse fraction reduction (CFR), in terms of MPD changes, were obtained comparing data over the mixing layer, with those collected into it near ground-level. In the abstract we refer to a such comparison at page 16484 (line 11 and 14) describing that the MPD changes occurred "passing through the mixing layer" and "across the ML". In the text (result and discussion section 3.1, page 16490, lines 19-20), we elicitated: "the number size distributions measured at ground level and over the ML". In addition, in the result and discussion section, continuous mentions about a comparison of such data, into and over the ML, are present. Regarding the mixing layer height values, they are reported in table 2 (page 16510) and table 4 (pag 16512). However, probably these points turn out to be not clear, and so we are going to clearly elicitate them at the result and discussion section opening, to avoid any doubt.

QUESTION 2) Section 2.1, page 16488, lines 23-25: "At ground level, a continuous monitoring activity of PM1 and PM2.5 was also present using CEN equivalent (according to EN12341) samplers (FAI-Hydra dual channel low volume sampler; PTFE filters, Ø=47 mm; EU sampling inlet, 2.3m3/h)". This text needs corrections. The standard EN-12341 applies for PM10 particles (not measured in this study). The standard for PM2.5 is EN-14907, which is not cited in the text. As far as I know there is not, at the moment, European reference method/standard for PM1.

ANSWER 2: Thank you for remarking these point which was due to a simple lapse error. We'll mend the manuscript.

QUESTION 3) Section 3.1.1, page 16492, line 25: "On average the sedimentation process was observed in 94% of cases in the winter and in only 49% of cases in the summer. In the other cases no clear evidence in MPD decrease was found. Higher atmospheric instability weakened particle sedimentation in the summer; however less windy conditions and higher stability, in the winter, favoured particle sedimentation. The mean reduction in MPD was 14.9$\pm$0.6% and 10.7$\pm$1.0% in the winter and summer respectively, with maximum reductions of 27.4% and 31.6%". I agree with this

interpretation. However, I think that more processes are involved. Several studies have shown that mineral dust is the most important contributor to coarse particles, and that mineral dust concentrations are higher in winter than in summer in Milan owing to the lower mixing layer depth and the higher resuspension of dust caused by road traffic in the urban area (Rodriguez et al., 2007, ACP, 2217-2232; Marcazzan et al. 2001, Atmos Env, 4639-4650). In my modest opinion, the fact that mineral dust concentrations are higher in winter than in summer in Milan, may significantly contribute to the higher sedimentation observed in winter in this study.

ANSWER 3: Thank you for discussing this point. In agreement with your findings, mineral dust concentration is higher in winter owing to the lower mixing layer depth. But, this process enhance the atmospheric concentrations (expressed as μg/m3) of each PM component in winter. However, considering the relative contribution of each component in PM samples (expressed i.e. as %) reported in literature, mineral dust contribution to coarse particles is higher in summer than in winter. Marcazzan et al. (2001) reported that "...the mass of the crustal matter accounts for about 18% and 22% in PM10 fraction in winter and summer periods, respectively" (page 4645) and that crustal matter is 42% in PM10-2.5 fraction in winter and 52% in PM10-2.5 fraction in summer (tab. 5). The same happened for PM2.5 with values of 2% and 7% of crustal material in winter and summer respectively. Vecchi et al. (2004) reported similar values and normalised atmospheric dust concentrations to 222Rn activity in Milan (year 2002) showing higher normalised concentrations of dust during the summer months as re-suspension favoured by drier soils. Here we add, to these literature results, the knowledge about the capability of higher atmospheric turbolence to weak particle sedimentation in summer.

QUESTION 4) Section 3.1.2: pag 16494, line 22-27: "Vertical profile data were mainly collected in clear and dry sky conditions, in which aerosol properties can be derived by satellite remote sensing using different sensors (i.e. MODIS). In this case the creation of accurate look-up tables would be very useful 25 (Levy et al., 2007; Chu et al., 2003)

and could be implemented for areas with characteristics similar to the Po Valley with some parametrization to distinguish periods characterized by highly stable conditions from other periods". When reading this text, it seems that authors tried to collect data in Milan city that be representative of the whole Po valley. In my opinion, the results of this study are representative of urban areas of the Po Valley, but is not necessary representative of the dominant rural areas. The fact that fine particle growth (accumulation mode) is observed during vertical transport of the air parcel in the city, is significantly prompted by the emissions of ultrafine particles in the vehicle exhaust. Particle growth is associated with the aging process (coagulation and condensation) of such particles during the vertical transport (within the mixing layer) accounts for the observed particle growth with altitude in the mixing layer. Vehicle exhaust emissions are not important in rural areas, and for this these results may not necessary be representative of rural areas.

ANSWER 4: Thank you for discussing this point.

1) You say that "In my opinion, the results of this study are representative of urban areas of the Po Valley, but is not necessary representative of the dominant rural areas."

We are in agreement with you. Our data are representative of urban areas of the Po Valley. This is very important considering that an high percentage of the Po plain is urbanized. We would remark that satellite applications, yet conducted in northern Italy (Po Valley), evidenced "the lack of ground truths" as a critical point which "makes the verification and validation not possible" (Chu et al., 2003); among these the knowledge about aerosol vertical distribution in this area is considerably improved by our measurements. Following this route we wrote that "In this case the creation of accurate look-up tables would be very useful (Levy et al., 2007; Chu et al., 2003) and could be implemented for areas with characteristics similar to the Po Valley". However it sounds ambiguous and we propose: "In this case the creation of accurate look-up tables would be very useful (Levy et al., 2007; Chu et al., 2003) and could be implemented for urban areas with characteristics similar to those found in the Po Valley". A campaign con-

ducted during the last winter in the Appennines valley over the city of Terni, confirmed the presence of the same behaviour founded in Milan. As an example we report here some vertical profiles in Terni (fig. 1A).

2) You say that "The fact that fine particle growth (accumulation mode) is observed during vertical transport of the air parcel in the city, is significantly prompted by the emissions of ultrafine particles in the vehicle exhaust. Particle growth is associated with the aging process (coagulation and condensation) of such particles during the vertical transport (within the mixing layer) accounts for the observed particle growth with altitude in the mixing layer. Vehicle exhaust emissions are not important in rural areas, and for this these results may not necessary be representative of rural areas."

This is an interesting point and we agree with you. However, as you know, in literature it is reported that ultrafine particles can originate also via nucleation in rural areas of the Po Valley basin (Hamed et al., 2007). This was evidenced both in winter and in summer, with higher frequency in summer. Looking at table 1 of our work (pag 16509) you can see that FFG is higher in summer, a period in which emission of primary ultrafine particles from combustion processes is lower due to the absence of residential heating combustion; this could be explained by secondary particles. In fact, as reported by Stratmann et al. (2003): "newly formed particles were observed inside the residual layer, before the break-up process of the nocturnal inversion and inside the mixing layer throughout the break-up of the nocturnal inversion and during the evolution of the planetary boundary layer". These results were obtained in clear and dry sky conditions (like our measurements conditions). Similar results were reported also by Laakso et al. (2007) in southern Finland. With respect to this point we performed another sampling campaign during winter 2009 which showed us chemical composition data which support the findings above. We are going to submit also these data in a new paper. In this way we have the aim to extend this work also to rural areas.

QUESTION 5) Title. The current title sound imprecise: -I am not sure if the term "low troposphere" is used properly. As stated above, these results are representative of an

urban area, -The term "...the role of the mixing layer....". This study do not analyzed how changes in the mixing layer properties prompts changes in the size distribution. This study describes how the size distribution of particles changes vertically along the mixing layer and above it. The only processes described in the manuscript are based on aerosol dynamics (coagulation, condensation and deposition) and not in properties of the mixing layer. I would suggest a final title that would sound something such as: "Vertically resolved particle size distribution within and above the mixing layer of Milan city".

ANSWER 5:

Thank you for this good suggestion. In the result and discussion section, the role of ML is clarified at page 16494 (lines 17-21) evidencing that ". . .ML height appeared in most cases as a critical parameter for splitting the lower troposphere into two areas one, within the ML, in which fresh aerosol from anthropogenic emissions and secondary origin was present, and another one over the ML influenced by background conditions with aged aerosols (especially in the winter) or by long-range transported aerosols". The same is reported also in the conclusions (page 16501-16503). In this way, original title of the paper was: Influence of the Mixing Layer on particulate matter size distribution and chemical composition: from experimental evidence to a hierarchical statistical model. However we changed it during the first access-review procedure. Taking into account your suggestion, your question 4, and the aim of the paper to provide a statistical model, we propose as a final title: "Vertically resolved particle size distribution within and above the mixing layer in the Milan metropolitan area by hierarchical statistical model".

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 16483, 2009.

Vertical profiles of particle number concentration and mean particle diameter (MPD) in Terni (fig. 1A).

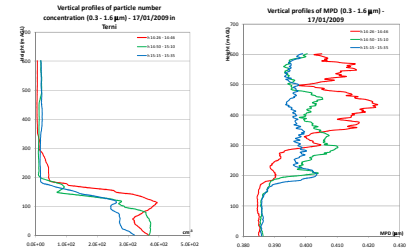

Fig. 1. FIG1A: Vertical profiles in Terni