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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Continuous atmospheric boundary layer observations in the coastal urban area of Barcelona, Spain" *by* M. Pandolfi et al.

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We would like to gratefully thank the Referee#2 for his useful comments.

GENERAL COMMENTS:

(1) Beyond the numerical characterization of the variables discussed (height of the SML, value of backscatter coefficient), diurnal cycles, the conclusions are not clear. It is not very clear which is the main aim of the work.

Answer: Following the suggestion of the Referee#2, the last part of the Introduction was modified to clarify the aims of this work:

"In this study, we analyse one month of measurements of PBL height and optical

properties continuously performed for the first time over Barcelona (Spain), using the vertical-pointing 1064-nm wavelength CHM15K ceilometer within the framework of the SAPUSS (Solving Aerosol Problems Using Synergistic Strategies) project (Dall'Osto et al., 2012). Main aims of this work are to provide valuable insights on PBL structure, evolution and optical properties over Barcelona based on the continuous ceilometer measurements and radiosonde data. We present and discuss the daily cycle of the PBL height and aerosol backscatter at selected heights under three typical atmospheric scenarios affecting the WMB and detected during the duration of the campaign, namely: stagnant Regional (REG), warm African (NAF), and cold Atlantic (ATL). Ground meteorological data and ground PM1 concentrations are also presented and correlated with the measured aerosol optical properties as a function of the atmospheric scenarios. The effect of overlying warm African air masses and of strong winds from the sea within the whole PBL on PBL growth and evolution are discussed. Additionally, ceilometer measurements were correlated with radiosonde data to study the effect of relative humidity on aerosol backscatter vertical profile over the coastal area of Barcelona. Finally, the aerosol backscatter coefficient vertical profile and its relationship with unstable/stable atmospheric conditions are also discussed."

Moreover the following DISCUSSION AND CONCLUSION section was added:

"4 DISCUSSION AND CONCLUSIONS During the SAPUSS (Solving Aerosol Problems Using Synergistic Strategies) project a vertical-pointing 1064-nm wavelength CHM15K ceilometer was deployed with the aim of determining the boundary layer structure, evolution and optical properties over the coastal area of Barcelona. Ceilometer data have been thoroughly analyzed in order to provide valuable insights on 1) the impact that three typical atmospheric scenarios (African dust outbreak, Atlantic advection, Regional recirculation) in the WMB have on the PBL over Barcelona and 2) the possible explanations for the observed increase of aerosol backscatter with height within the SML irrespective of the thermodynamic states of the atmosphere. Findings can be summarized as followed:

(1) PBL maximum height and daily variations (DV) were strongly dependent on air mass types, ranging from the highest PBL-strongest DV (ATL) to the lowest PBL-weakest DV (NAF). The observed differences in PBL structure over Barcelona were due to the concomitance of different factors. The presence of warm African air masses above colder local air changed the temperature profile thus lowering the height of the inversion. As a result the mean SML height during NAF was 25% lower than for the ATL scenario. Previous studies conducted in Barcelona have related the lack of a clear PBL height annual cycle to the typical summer conditions of the WMB characterised by weak pressure gradients and the development of breeze circulation patterns which may prevent the vertical development of the PBL (Sicard et al., 2011). Even though the breeze pattern is a recognized cause affecting the growth of PBL at coastal sites, the higher occurrence and intensity of African dust episodes in the WMB in summer (10-20% of the days, Pey et al., 2010b) may also contribute to prevent the development of the PBL. This result is also a motivation to further investigate about the possible relationship between PBL height and African dust episodes.

(2) The analysis of the relationship between the SML heights estimated from ceilometer (SMLh_ceil) and radiosoundings (SMLh_rad) at 12:00 UTC revealed another interesting characteristic of PBL under NAF. On average during SAPUSS, the SMLh_rad were lower than the SMLh_ceil by around 166 m and 249 m during REG and ATL, respectively, whereas during the NAF scenario the difference was small (-42 m). We argued that a possible reason for the good agreement between SMLh_ceil and SMLh_rad during NAF was the presence of abrupt changes in the vertical profiles of backscatter and potential temperature at the SML top caused by the differential advection of air masses above Barcelona during NAF. These abrupt changes reduced the uncertainties in SML top retrieval from the two techniques. The mean slopes of the lidar backscatter and potential temperature profiles at the top of the SML during NAF were -0.038$\pm$0.007 Mm-1sr-1/m and +0.018$\pm$0.007 °K/m, respectively, at 12:00 UTC. By comparing these values with Fig.5, we can observe that the potential temperature and backscatter first derivatives during NAF were in the upper range and out of range, respectively, of val-

ues reported in Figure 5. These values corroborate the hypothesis that the differential advection caused abrupt physical and thermodynamic changes at the SML top which accounted for the good agreement between SMLh_ceil and SMLh_rad during NAF.

(3) Aerosol backscattering coefficients (BSC) were highly variable, with the highest values influenced by both African dust intrusions (NAF) and regional anthropogenic pollution (REG). The compression of the SML under NAF favoured the transport of the Saharan dust down towards the surface layers by dry deposition. As a consequence, the NAF scenario registered the highest mean β500 and β800 measured during the campaign which were 20% and 15%, respectively higher than under REG and 50% and 46%, respectively, higher compared with ATL.

(4) In the portion of the atmosphere characterised by neutral thermodynamic stability conditions (dΘ/dz $\sim$ 0) at midday, the BSC increased in parallel with RH with altitude. By contrast, under the stable stratified conditions (dΘ/dz > 0) above the SML, BSC decreased with falling RH with height. The increase in BSC with altitude was also detected under unstable atmospheric conditions (dΘ/dz < 0). This analysis revealed that at midday the aerosol backscatter over the coastal site of Barcelona increases with height from near ground to the SML top irrespective of the thermodynamic states of the atmosphere. At midday the portion of the atmosphere characterized by neutral thermodynamic stability conditions (dΘ/dz $\sim$ 0) is assumed to be well mixed and represents the volume of air into which pollutants can be dispersed. The increasing backscatter with height within the SML at midday was due to the increase with height of RH, which through aerosol swelling, increase the effective aerosol cross-section. When lidar data are used to estimate the PBL AOD from extinction measurements, common assumption is to assume the extinction to be a flat extrapolation at ground of the extinction profile at the range of full overlap. On the base of 1 month of radiosundings performed during SAPUSS, we observed that the RH at Barcelona monotonically increases with height by around 40% from ground to the SML top. Thus, the observed strong RH dependence of backscatter vertical profile should be taken into account for PBL AOD

calculation from lidar extinction data. In summary, continuous on-line measurements of the spatial and temporal distribution of the PBL characteristics and aerosol optical properties are valuable. Further studies are necessary to improve our knowledge on the relationship among African dust outbreaks, temperature vertical profile and PBL height."

(2) The diurnal cycles of the SML and the DRCL from fig. 1 are somehow strange. The lowest values of the SML are around 400 m even during nighttime. Is this reasonable? The nocturnal mixing layer is expected to be very shallow, being the driving turbulence very weak. The dynamics is extremely low.

Answer: Dynamics are commonly limited in magnitude during nighttime. However, the location (in terms of topography and land/sea characterization) dictates the boundary-layer (BL) dynamics during both day and night preventing from generalization of diurnal/nocturnal BL common trend. Barcelona is characterized by a strongly decoupled vertical structure of the BL in part determined by its hybrid topographic characterization, i.e. sea, land and urban environment concur to characterize the BL structure at any time. Convective dynamics and wind (including breeze) occurring during daytime determine and sustain the mixing of aerosols over the first 1000-2000 m a.g.l.. The slowly subsiding nocturnal air and the stratification of the aerosol within the nocturnal layer combined with the nocturnal hygroscopic growth of the aerosols give rise to strong LIDAR return especially during cloud-free, low-wind and prolonged high-pressure conditions. The gravitational settling of the aerosols in the nocturnal BL is not rapid enough to deposit most of the particles to the surface so that the LIDAR return rarely drops below 300-400 meters. The Figure 1 shows how the decoupled structure of the BL characterize the lower Troposphere above Barcelona during the three air mass scenarios.

At the GAW-WMO Station of Mace Head, on the west coast of Ireland, similar coastal dynamics engender the BL decoupling as in Barcelona (Figure 2). Over five years of recorded BL data at Mace Head only in cases when all sources of aerosols are

"switched off" the BL shows a clear homogeneous single-layer structure. These unique conditions occur for example when snow covers the ground and the wind is calm (no sea spray). A clear example of the rapid re-establishment of the decoupled BL structure above Mace Head is shown in the Figure 2 when after the snow cover has completely melted at around 3 pm UTC, the decoupled structure is promptly recreated.

(3) In what measure can the incomplete instrument overlap contribute to an overestimation of the SML? The SML does not reach the same height of the DRCL at midday. It seems that these layers never merge and interact. Please, discuss this point.

Answer: As partially discussed above, the relatively thick nocturnal BL does not result from a signal artefact or by the incomplete overlap between the telescope and the emitter within the SML. The Temporal Height Tracking (THT) technique is gradient-based thus reducing considerably the effect of an incomplete overlap. In fact the relative difference in signal magnitude between contiguous range gates is detected also below 400 m especially if the transition is strong like it is at the top of the SML. Figure 1 shows that there exist two kinds of DRCL and SML diurnal cycle: when the DRCL disappear during the central hours of the day (like for NAF and REG in the examples reported in Fig.1) and when the capping inversion is not eroded by the midday convection and a residual layer remains aloft the diurnal SML.

(4) The backscattering values declared (between 0.3 and 1.2 Mm-1 sr-1) make sense, since the molecular backscattering at 1064 nm is around 0.07 Mm-1sr-1 at ground. This leads in a backscatter ratio ranging from 4 and 17. However, it is not clear how the backscattering coefficient is estimated from lidar data. Is any inversion procedure performed? In case yes, it should be discussed. In case not, the error arising from the aerosol extinction should be quantified.

Answer: Indeed, an inversion procedure based on Klett is performed and provides the profiles of extinction and backscatter by assuming a LIDAR ratio. The thorough description of this procedure including assumptions and errors in case of clouds or haze can be found in Section 4.1 and A.1.2 of: Martucci, G., J. Ovadnevaite, D. Ceburnis, H. Berresheim, S. Varghese, D. Martin, R. Flanagan, C.D. O'Dowd, 2012: Impact of volcanic ash plume aerosol on cloud microphysics. Atmospheric Environment, 48C, 205-218.

However, the procedure described in the above reference refers more to the retrieval of the extinction and backscatter in presence of clouds. We have then added to the text the following description for cloud-free cases:

"The extinction coefficient is calculated by inverting the 1.064 μm LIDAR power profiles (Klett, 1981; Ferguson and Stephens, 1983) in the region where the signal is not completely attenuated, i.e. up 15 km for the CHM15K in cloud-free conditions (but subject to the optical thickness of the total column). A LIDAR ratio S = 45 sr is assumed for the probing wavelength at 1064 nm, a mixture of continental and maritime aerosols and in the range of 50%-80% of relative humidity (Ackermann, 1998). The raw LIDAR signal (number of photons) is transformed into received power by use of Plank law combined with the LIDAR equation at any range R. The obtained LIDAR power is then corrected for the square of the range R and normalized by the LIDAR constant, the latter being calculated directly along the molecular part of the LIDAR signal (for a non-opaque profile). Because at 1064 nm the molecular backscatter is very weak this procedure needs to perform the molecular calibration for integration times not shorter than an hour and preferably during nighttime to avoid the solar background. The procedure to obtain the extinction must take into account the effect of the multiple scattering only when high concentration of particles (haze or cloud) occurs along the LIDAR profile. Multiple scattering (η) is in fact a complex function of the field of view of the LIDAR, the distance of the haze/cloud, the particles concentration, the breadth of particles size distribution and the layer optical depth. The multiple scattering affects the amplitude of the signal received by the LIDAR at time t and range R by modifying the LIDAR ratio S as ηS and can cause smaller extinction than in reality (η < 1). When all the above parameters influencing the value of η are negligible the multiple scattering can be neglected by

keeping η = 1."

J. Ackermann, "The extinction-to-backscatter ratio of tropospheric aerosol: a numerical study," J. Atmos. Ocean. Technol. 15(4), 1043–1050 (1998)

(5) How reliable is the PM1 concentrations measured with the OPC? This could be a cause of underestimation of the PM1. This should be discussed in some detail.

Answer: It is true that PM measurements from OPC are usually underestimated. This is why measurements from GRIMM instruments are usually normalized to reference method based on gravimetric measurements. In order to clarify this point, the following sentence was added to the text:

"Subsequently, the PM concentrations were corrected with factors obtained by comparing real time and gravimetric measurements. PM gravimetric measurements on a 24h basis were performed every day during SAPUSS with high volume samplers (DIGITEL and MCV at 30m3 h−1) with appropriate (PM1, PM2.5, PM10) cut-off inlets."

(6) On the basis of what indices are the atmospheric scenarios classified? Backtrajectories, Model wind fields.

Answer: Back trajectories of the air masses arriving at Barcelona were calculated four times for each day of the campaign (00:00, 06:00, 12:00 and 18:00 UTC), depicting the path taken by the air mass reaching the sampling site over the previous five days. The back trajectories were run using the on-line HYSPLIT model developed by the National Oceanic and Atmospheric Administration (NOAA). Air mass "types" are usually classified according to latitude and their continental or maritime source regions (Bergeron classification). However, during SAPUSS air mass back trajectories "scenarios" names given will also be used, including: Atlantic (ATL), North African (NAF) and Regional (REG) scenarios (Rogriguez et al., 2002; Pey et al., 2010). It is important to clarify that the classification was made not only accordingly to air mass back trajectories, but also by looking at the local meteorological data available at the sampling sites.

Details are given in Dall'Osto et al (2012).

Pey, J., Alastuey , A., and Querol, X.: Discriminating the regional and urban contributions in the 30 North-Western Mediterranean: PM levels and composition, Atmos. Environ., 44, 1587–1596, 2010.

Rodríguez, S., Querol, X., Alastuey, A., and Mantilla, E.: Origin of high summer PM10 and TSP concentrations at rural sites in Eastern Spain, Atmos. Environ., 36, 3101–3112, 2002.

Dall'Osto, M et al.: Presenting SAPUSS: solving aerosol problem by using synergistic strategies at Barcelona, Spain, Atmos. Chem. Phys. Discuss., 12, 18741–18815, 2012.

(7) As far as I know, sea breeze in the afternoon cleans the atmosphere very efficiently, making the determination of the SML often very hard. How reliable are the automated inversions of the ceilometer data in such conditions? This could introduce a bias in the daily cycles, especially for the "Regional" scenario.

Answer: Sea breeze has a "cleansing" effect on the atmosphere especially when the air mass is continental and the aerosol characterized by sooth/sulphate/nitrate. However, a sustained sea breeze brings sea spray particles into the boundary layer, which are bigger particles than the anthropogenic one and highly hydrophilic. Sea breeze does not reduce then in general the LIDAR signal, sometimes a marine flow can increase the LIDAR signal instead.

(8) Fig. 1 is hard to read. Enlarge it if possible or find a different visualization.

Figure 1 was enlarged.

(9) The correlation between ceilometer-derived SML heights and radiosoundings does not add much to the study, apart from a validation of the algorithm. It looks unnecessary unless in the discussion, some specific events are discussed. Anyway, the SML heights are always considered reliable, even in case of strong wind advection. Yet,

the removal/advection of aerosols are known as the major source of error of boundary layer estimation from lidar data. The reliability of lidar-derived SML height should be discussed and proved for such cases.

Answer: Although it is quite typical to present correlation between ceilometer-derived SML heights and radiosoundings, we observed a worthwhile characteristic: The best agreement ($\sim$ 1:1) between SML heights from ceilometer and radiosoundings was observed under NAF compared with REG and ATL. We argue that a possible reason for this was the presence of abrupt changes in the vertical profiles of backscatter and potential temperature at the SML top caused by the differential advection of different kind of air masses. These abrupt changes might help in reducing the differences/uncertainties in SML top retrieval from the two techniques.

The following sentence was added to the section 3.3:

"The reason for the good agreement observed between radiosondes and ceilometer in estimating the SML heights during NAF could be the differential advection of air masses at different heights observed under the NAF scenario. The 5-day back-trajectories ending in Barcelona at 12:00 UTC and calculated for the NAF days (two examples are shown in Figure SI-3 in Supporting material) display the differential advection with the air mass ending at 1000 m a.g.l. clearly coming from Africa and those at lower altitudes having different origins. This differential advection may have caused an abrupt change in physical (aerosol) and thermodynamic (potential temperature) properties at the top of the SML, thus minimizing the differences typically observed between ceilometer and radiosondes in estimating the SML height. To support this, the opposite trend is seen for the ATL scenario when the air masses had the same origin at the altitudes considered (cf. Figure SI-4) and the largest difference between the SML heights can be observed."

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 345, 2013.

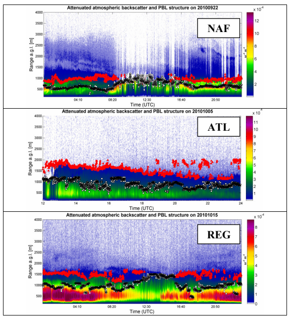

Figure 1.

Fig. 1. Figure 1

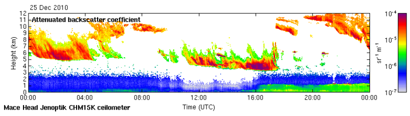

Figure 2

Fig. 2. Figure 2