

Anonymous Referee #4

Review of : Combining airborne in-situ and ground based Lidar measurements for attribution for attribution of aerosol layers.

By Nikandrova et al.

The authors describe two case studies (clear sky and cloudy sky) observed over the SMEAR-II station during a field campaign in 2014. The authors used airborne measurements (mostly in-situ size distributions) associated to ground based HSRL Lidar.

This manuscript is of interest for the scientific community but need major revisions before submission to ACPD.

Response to comments from Anonymous Referee #4

We thank the referee for the constructive comments to help us to improve the manuscript. Below please find our answers to the comments.

First of all, the aim of the paper is pretty vague: 'investigate aerosol layers in a rural environment' and need to be clarified.

We have tried to clarify the aim of the paper in the introduction and added several sentences: 'We were particularly interested in how the aerosol size distribution varied both within and between layers. This information could be used to determine whether there was mixing within and between layers, and whether there had been any recent contact with the surface.'

We have also added this sentence at the end of the last paragraph of the introduction: 'Back trajectory analysis was conducted for both case studies to examine whether these analyses produced similar layer structures to those observed, and how closely the diagnosed layer altitudes corresponded with those observed by the HSRL.'

This paper is showing size distribution differences that occur within each layer of the atmosphere as a function of time. The authors interpret each increase of the fine particle number concentration as a nucleation event within each layer. However, the differences of Aitken, Accumulation and Coarse number concentrations are only pointed out.

Although we interpret these increases as NPF, we focus on how variable the aerosol size distribution is within each layer, and how the shape of the aerosol size distribution changes over time; this can then be used to infer any mixing within or between layers. The relative lack of mixing observed in elevated layers may then inform likely evolution of aerosol undergoing long-range transport.

The conclusions of this paper needs some work : The comparison of the RH and HSRL profiles with HYSPLIT results are most of the times in good agreement but the "heights did not always coincide". These height differences are not expressed in the main part of the paper and should probably be. . .

We have added more text to the main part after this sentence 'During 9 and 10 April, for example, the trajectory BL height was lower than the BL seen from the HSRL and consequently, the trajectory analysis suggested a thicker middle layer.', 'BL height diagnosed from trajectory analyses was 50-800 m lower than that observed, whereas for elevated layers, the layer boundary heights were better represented, with departures typically less than 200 m. These larger height differences for layers associated with the BL top are attributed to the difficulties that meteorological models have in representing the BL (e.g. Holtslag et al., 2013), which are then propagated through to the trajectories.'

Added to the conclusion: Errors in trajectories (particularly in the vertical) arise from the difficulties that the meteorological models providing the wind fields have in accurately representing vertical motion and turbulence, the boundary layer, and other sub-grid scale features (Stohl et al., 2001, Riddle et al., 2006, Hoffmann et al., 2016). Uncertainties in the horizontal can be determined using ensemble trajectory techniques (Stohl et al., 2001) but these are less likely to capture vertical discrepancies arising from processes that the meteorological model may not capture correctly, such as the boundary layer.

L. Hoffmann, T. Rößler, S. Griessbach, Y. Heng and O. Stein, Lagrangian transport simulations of volcanic sulfur dioxide emissions: Impact of meteorological data products, *Journal of Geophysical Research: Atmospheres*, **121**, 9, (4651-4673), (2016).

Holtzlag, A.A., G. Svensson, P. Baas, S. Basu, B. Beare, A.C. Beljaars, F.C. Bosveld, J. Cuxart, J. Lindvall, G.J. Steeneveld, M. Tjernström, and B.J. Van De Wiel, 2013: Stable Atmospheric Boundary Layers and Diurnal Cycles: Challenges for Weather and Climate Models. *Bull. Amer. Meteor. Soc.*, 94, 1691–1706, <https://doi.org/10.1175/BAMS-D-11-00187.1>

Riddle, E. E., P. B. Voss, A. Stohl, D. Holcomb, D. Maczka, K. Washburn, and R. W. Talbot (2006), Trajectory model validation using newly developed altitude-controlled balloons during the International Consortium for Atmospheric Research on Transport and Transformations 2004 campaign, *J. Geophys. Res.*, 111, D23S57, doi:10.1029/2006JD007456.

Stohl, A., L. Haimberger, M. P. Scheele, and H. Wernli. "An intercomparison of results from three trajectory models." *Meteorological Applications* 8, no. 2 (2001): 127-135.

A brief presentation of the Hysplitt model and especially the resolution of the data input of the model could help the authors to interpret these differences.

Added to the text: The National Center for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS) dataset with one degree resolution was used for the meteorological input to the model.

Also the last conclusion of the paper is that the synergy between radiosounding, LIDAR and back trajectories gives more confidence in determining the air mass origin. Is this really the main conclusion ?

This is one of the main conclusions, and we have added more text (also written in the comment above: Errors in trajectories (particularly in the vertical) arise from the difficulties that the meteorological models providing the wind fields have in accurately representing vertical motion and turbulence, the boundary layer, and other sub-grid scale features (Stohl et al., 2001, Riddle et al., 2006, Hoffmann et al., 2016). Uncertainties in the horizontal can be determined using ensemble trajectory techniques (Stohl et al., 2001) but these are less likely to capture vertical discrepancies arising from processes that the meteorological model may not capture correctly, such as the boundary layer.)

Last, the authors state: 'Evidence for cloud processing of aerosol particles was also seen in the BL but the amount of processing varied [. . .]'. The authors are showing Hoppel minimum that could be related to cloud processing but it's not supported by real evidence. It could also be due to different sources of aerosol with one source quite close to the instrumental site ? Moreover, I believe you can't talk about the 'amount of processing' . . .

We agree that we have no direct evidence of cloud processing. However, the lower part of the BL was very well-mixed which suggests that any local sources should also be reasonably well-mixed; there is low variation in other size ranges for the entire BL.

This sentence has been reworded: 'In the BL, the aerosol size distribution displayed a Hoppel minimum suggesting cloud processing of aerosol particles, but with variations that were presumably again due to the specific nature of the updrafts and downdrafts resulting in BL mixing that was not fully homogeneous in the upper part of the BL.'

Minor corrections:

P3 L 4 : Not well said. Please rephrase

Rephrased: 'Microphysical properties retrieved from HSRL-2 showed a good agreement with in situ measurements; however, backscatter and extinction coefficients calculated from corresponding in situ measurements were consistently underestimated, which was attributed to the undersampling of coarse mode particles by in situ measurements'

Page 3 L15 : Needs to add references to support that like Crumeyrolle et al., 2010; Rose et al., 2015a, Berland et al., 2016.

Rephrased and added two of suggested references: This suggests that, in the boreal forest, large-scale NPF events are typically confined to the BL, similar to results found in other environments (Crumeyrolle et al., 2010; Berland et al., 2016).

Rose et al., 2015a reported NPF events in the free troposphere over Mediterranean.

Figure 2 : You are always referring to the mode you define P5. Could you add on your size distribution plots the limits of each mode (nucleation, Aitken, Accumulation, Coarse). It would help the reader.

Added as suggested

No error bars on the Figure 2i within the small particles range for the middle layer ?

There is no error bar because shaded areas show variability in the layers, and in the middle layer during this flight, small particles were detected only once. This is explained in the text on p. .

P10 L7 : 'A very high peak' : could you add in comparison to the rest of the profile ?

Added as suggested

P10 L10 : Smoke or Dust are not known to be spherical particles ...

A clause was missing from our sentence. The sentence has been rephrased 'This thin layer could be either a result of limited small-scale mixing between two layers, that were probably stable, or the result of large-scale transport of smoke or dust; however, we suspect that this is a response of aerosol growing rapidly as it moves from very dry air to much moister conditions, especially since the low HSRL circular depolarization values suggest that particles in this thin layer were relatively spherical.'

P11 L14 : below 100nm instead of 30nm

We left 30 nm as originally written because on the figure 7c aerosol number size distribution in the upper layer (blue) lower than in the first middle layer (yellow) below 30 nm.

P11 L 22 : Please add 100nm to show the reader where the Hoppel minimum is located.

Added as suggested

P12 L17 : around 100nm replace with around 70nm

Replaced as suggested

Figure 7. Not able to distinguish the 3 green shades...

We have changed colours.

Figure 9 : No error bars : Does it mean that you used only one spectra. If yes than it needs to be stated somewhere.

Error bars added to the plot.

5 L25 : Please add explanations. I don't want to read Laakso et al. To understand what you did. The GF is usually dependant of the different compounds present in the aerosol. So how did you get this information ?

Added to the text: 'using a growth factor (GF) calculated for a boreal forest environment using measurements from Hyytiälä station by Laakso et al. (2004). They weighted the GF for compounds with different hygroscopicity according to their respective fractions to obtain an optimal combined GF coefficient.'

P7 L25 : Hard to tell cause there are no measurements of the fine particle number concentration within the middle layer...

No particles smaller than 15 nm were detected in the upper and middle layers during the morning flight even though the detection limit is 10 nm, but these were observed in the afternoon flight, providing evidence for NPF in elevated layers.

P9 L27-29 : Please tell us more about the difference you see cause it's not obvious for me.

This paragraph is rephrased to be clearer: 'For particles smaller than 300 nm, the shape of the size distribution and the number concentrations changed from day to day. For particles larger than 300 nm, while the number concentration varied, the shape of the distribution remained similar across all 3 days.'

P11 L 20 : If you are implying that the cloud base is playing a role in the mixing efficiency be more clearer

We have rephrased this sentence 'Two tendencies are seen in the BL: a more mixed lower part up to about 1000 m where the cloud bases were, and a less mixed upper part' as we did not mean to imply whether cloud base plays a role. It happens that saturation occurs at a similar altitude as the mixing profile begins to depart from a well-mixed profile, but we do not infer that cloud is necessarily the cause of the change in the mixing profile.

Our sentence now reads 'The BL was well-mixed up to 600 m, and became progressively less well mixed above this, with convectively buoyant air parcels reaching up to 2500 m. The radiosonde thermodynamic profile suggested that deep convection to 4 km or so was possible, and did indeed occur later on in the day.'

P12 L 19 : Any interpretation why there is less particles above 500nm ?

Added to the text: This may due to dilution as the growing BL entrains air from above with lower concentrations in this size range.

P12 L22 : Do you mean that nucleation occurs over the cloud top ? Please add references to support this.

With cloud-driven entrainment we are only implying that there might be localized mixing, rather than a fully-mixed layer. Hence, there may be pockets with slightly higher and slightly lower concentrations, without NPF necessarily occurring.

Section 3.1.2 : If you are talking about errors you need to state the number of SD you used to get the average showed in figure 2. ..

We report uncertainties using one standard deviation, as written in the caption for the figures and in the text.