This manuscript presents a thorough analysis of the aerosol composition and their source apportionment at a mountain forest site in Central Germany using detailed aerosol and gas measurements for 40+ days in late-2010. The topic is important, the methodology is clear, and the findings are very well presented. By using detailed particle composition, organic aerosol source apportionment (ME2), and back trajectory analysis, Poulain et al. provide insights into sources of aerosol at this site. Among other things, their findings on more than half of Equivalent black carbon (eBC) coming from long-range transport is especially interesting and potentially relevant for ongoing and future studies as well. I think the importance and quality of this manuscript warrants its publication in Atmospheric Chemistry and Physics.

We would like to thank the referee for his/her constructive comments and suggestions made to improve and clarify our manuscript. Our responses are given below. For clarity, comments from the referee are in **black**, our responses in blue, and change on the text of the manuscript in **bold blue**.

## I only have some minor comments:

R1: Fig 1: Consider including Boundary Layer Height (BLH) timeseries either in Fig1 or in Fig S5. Reanalysis BLH (can easily be obtained from ECMWF's ERA5) seems to suggest potential role of changes in BLH height on the total aerosol mass loadings for the observed period.

A1: As suggested, the boundary layer height (BLH) time series was included in Fig. 1. Here the BLH time series was retrieved from the HYSPLIT GDAS (1 degree resolution) input, which was used for the trajectory analysis.

## The text was modified as follow:

## Section: 2.3 Back-trajectories and cluster calculations

The 96 h back trajectories were used to determine the influence of the air mass origin on aerosol. The trajectories were calculated for every hour from 13 September until 24 October 2010 for the altitude of 500 m above model ground with the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT-4) Model (http://www.ready.noaa.gov/ready/hysplit4.html; Draxler and Hess, 2004) using the 1 degree resolution GDAS input data. The different back-trajectory clusters were calculated using the program R (http://www.r-project.org/; R Core Team, 2013) with the package openair (http://www.openair-project.org; Carslaw and Ropkins, 2012;Ropkins and Carslaw, 2012). The same GDAS input data was used to retrieve the boundary layer height (BLH) at the sampling site from the HYSPLIT model output.

## Section: 3.1.1 Overall AMS-MAAP time series

Aerosol particle chemical composition (mass concentration and mass fraction) as measured by AMS and MAAP as well as the particle number size distribution over the entire time-period are shown in Figure 1. On average, the near-PM<sub>1</sub> particulate chemical composition was principally made-up of organic aerosol, OA (41 % of the total mass, Fig. 1). Sulfate and nitrate have quite similar contributions (19 % and 18 %, respectively). The rest of the aerosol particle mass concentration was made of ammonium (11 %), eBC (10.0 %), and chloride (1 %). Despite their similar contribution to the particle mass fraction, sulfate and nitrate showed a clear time dependency (Fig. 1). Although sulfate dominates the inorganic fraction at the beginning of the measurement period, nitrate becomes more important over time. This can be directly linked to a decrease of temperature during the sampling period (Fig. SI-5), inducing a change in nitrate partitioning between gas and particle phase. A last factor that must be considered is the decrease of solar radiation from summer to winter, influencing the photochemical formation of sulfate. Variation of the BLH over the sampling period can also influence the local PM mass concentration (Fig.1). At the beginning of the campaign, the BLH reached above 1000 m during daytime, while the maximum altitude of the BLH decreased to below 800 m later on. This decrease in the maximum altitude of the BLH certainly contributes to the observed increase of the overall PM mass concentration during the day by reducing the ventilation effect. However, it is important to note that high uncertainties on the absolute value of the BLH for such a mountain area have to be

**expected due to the 1 degree resolution of the GDAS input data.** Variations of the organics and eBC mass concentration over the sampling period will be discussed in sections 3.2 and 3.3, respectively.



Date and Time

<u>Figure 1:</u> Time series of the ambient temperature (a), **estimated boundary layer height (BLH) obtained** from HYSPLIT GDAS input (b), the particulate near-PM<sub>1</sub> chemical composition as measured by the AMS and completed by MAAP for equivalent black carbon (c), the corresponding mass fraction (d), and particle number size distribution (e) during HCCT-2010 at the site of Goldlauter. The colored bars and numbers at the top refer to the six different air mass clusters (see section 3.4), and the grey bars refer to the different cloud and non-cloud events as defined in Table SI-2.

R2: Mention measurement period in Introduction or Site and instrumentations.

A2: The following sentence was modified in the introduction section to include reviewer's suggestion: The measurements **took place on September-October 2010 as part** of the "Hill Cap Cloud Thuringia 2010" (HCCT-2010) experiment, which aimed to investigate the impact of cloud processing to aerosol physico-chemical properties.

R3: FigS5: Subplot-4 check colors. A3: Colors for global radiation and relative humidity were corrected in Fig. S4.



Figure SI-5: Overview of the meteorological conditions during the sampling period.