Response to Referee #1

We greatly appreciate all the comments, which improved the paper. Our point-by-point responses are detailed below. AC – Authors Comments.

General Comments

The paper is a useful contribution to the issue of dry deposition over a forest. It describes a new method- the modified micrometeorological gradient method- which is in better agreement with eddy-covariance-EC- observations then the more traditional gradient methods.

Specific comments

On page 782, line 15, the authors make clear that the method is still based on the flux gradient theory. This remark is repeated at several places in the paper, as f.e on page 785, where its is mentioned that the flux-gradient method is questionable within the canopy. The question arises how serious this is, what is the impact of this restriction. It is recommended that the authors write some sentences about this.

AC: We have rewritten the first paragraph of section 2.4 to address this comment. It now reads "The newly proposed MGM method is also based on the flux-gradient theory (Eq. 2). It is noted that the flux-gradient theory has been long questioned within plant canopy environment due to infrequent but predominant large eddies within canopy (Wilson, 1989; Raupach, 1989). For example, Bache (1986) suggested that the flux-gradient theory was a reasonable assumption estimating wind profiles in the upper portion of canopy, but failed to reproduce the secondary wind maximum that was often observed within the trunk space of forests. It should also be noted that most of the O_3 uptake occurs in the upper layers of the canopy where most canopy leaves grow. Within these upper layers the vertical length scales of turbulence are probably smaller than the distance associated with changes in concentration and wind speed gradients (Baldocchi, 1988). Thus, the flux-gradient theory is likely applicable for estimating vertical flux distribution of air pollutants within a plant canopy, as has been used in previous studies (e.g., Baldocchi, 1988; Bash et al., 2010; Wolfe and Thornton, 2011)."

On page 785, line 13, the height-dependent Flux is introduced. What is the impact of this assumed height-dependency on the obtained results. Does this means that EC observations at the different height as they are performed now-which is 29 m, would lead to different values at f.e. 18.3 m? A similar issue arised with the remark made on page 786, line 3, where its is stated that again the constant flux approach is used. It is recommended that the authors write a short paragraph to comment on these issues.

AC: Flux above the canopy is constant (assuming no additional sink or source terms), while flux within the canopy varies with the height (due to the sink terms – O_3 uptake by leaves). The height 18.3 m is within the canopy in this case so EC measurements cannot be conducted at this height (or do not represent the total flux if measured at this height).

On page 787, formula (15), u* is introduced, without clarification. Is this the shear stress velocity at the surface, or the "effective" one at the displacement height, and how is it calculated. It is recommended that the authors clarify this issue.

AC: u_* in this study is the shear stress velocity measured at the reference height (29 m). This has been made clear in the revised paper (section 2.2).

Page 789, lines 18-21 it is discussed that in about 70 % of the observations counter gradient profiles occur. No remark is made about what is happening in these cases, which phenomenon is present, and what is the impact on the fast that in only 30 % of the cases "real" dry deposition seems to occur? It is recommended that the authors write a short paragraph on this.

AC: We have added the following explanation in the revised paper (section 3.2).
"The counter-gradient transport should be mainly due to the non-local nature of turbulent transport within canopies. Large sweep-ejection air motions associated with coherent structures that can deeply penetrate into the canopy are believed to be largely responsible for the exchange of momentum, heat and mass between air above- and within-canopy (e.g, Shaw et al., 1983; Thomas and Foken, 2007)."

Page 790, line 18-25. It is mentioned that the AGM method gives much higher values then the EC-observations. Could the authors please give a possible explanation to this finding?

AC: The aerodynamic gradient method (AGM) is not the main focus of the present study, so we simply provide some explanation based on what we found from literature. Some earlier studies have also found this method overestimated fluxes when compared with the EC method (e.g., Muller et al., 2009; Loubet et al., 2013). The large discrepancies in fluxes between those generated by AGM and EC, as found in this study, were likely cause by a combination of many different factors, such as measurements errors in both methods, selections of the R_a formula and related parameters, and the local and large scale specific meteorological, physical and chemical conditions. For example, the EC technique is found to underestimate flux during calm night-time periods (Goulden et al., 1996). O_3 fluxes measured by different EC instruments could exhibit a relative difference of up to 25% (Muller et al., 2010). AGM derives flux from the concentration gradient between two adjacent levels above the canopy, which is subject to large uncertainty due to the very small gradient and associated measurement uncertainties. AGM is subject to the drawback due to the use of empirical stability correction functions. Uncertainties in the estimation of R_a above the canopy (and thus in the flux estimation using the AGM method) can be up to 30% on long-term average (Zhang et al., 2003). Large uncertainty may also exist in the estimated parameters such as the roughness length and the displacement height which have significant effects on the calculation of R_a. Unfavorable meteorological conditions may occur, such as the large scale turbulence structures which will generate advection terms and affect the low-frequency range of the turbulent spectra. This may underestimate flux when using the EC method (Mauder and Foken, 2006).

Technical corrections

AC: All corrected.

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