Response to Thomas Foken

We greatly appreciate Thomas Foken for providing the comments which have helped us to improve the paper. Our point-by-point responses are detailed below. AC – Authors Comments.

The measurement of deposition fluxes above tall vegetation is a never ending story because of many challenges. Very important is the small gradient of temperature and trace gases above the canopy, which is often lower than the detection limit of the sensors/analyzers (Foken, 2008, p. 135). The authors try to overcome this problem by using a gradient between a level above the canopy and one within the canopy, with a significant increasing of the gradient. Unfortunately, they do not discuss the influence of relevant processes at the top of the canopy on the new proposed method, like roughness sublayer or mixing layer (Garratt, 1978; Finnigan, 2000; Harman and Finnigan, 2007, 2008) (Raupach et al., 1996), decoupling (Thomas and Foken, 2007a), coherent structures (Collineau and Brunet, 1993a, b; Thomas and Foken, 2007b), scalar similarity (Ruppert et al., 2006), and reactions. Some of the effects may not be relevant due to the selection of only 26 % of the data set for the analysis. Because the abovementioned processes have a daily and annual cycle, it would be interesting to see a daily and annual cycle of the availability of the data. I assume that only situations with moderate and high wind velocities and a good coupling of the atmosphere with the upper canopy layer were used.

AC: The issues raised here and references provided do help us better understand the complex processes involved in the air-surface flux exchange of trace pollutants above tall canopies. As we have responded to Referee #3, detailed investigation on all the issues would require substantial additional efforts which can only be done in future studies. The present study focuses on the development of a new gradient method and thus only chooses data that fits such an application. As also mentioned by this reviewer, only using 26% selected data likely avoids many of the non-ideal conditions affecting the suitability of the modified gradient method. In the revised paper, we have added some brief discussions as detailed in our response to Referee #3.

Per the reviewer's request, we have also provided below (Figure 1) the diurnal and seasonal patterns of data points available for analysis. There are about 75-155 data points in each hour with two peaks in the early morning (7-8 LST) and the late afternoon (14-16 LST). The number of data points available in each month indicated a significant season trend with the most data points in summer (~400) and the least in winter (~50). This is primarily due to the data availability in the original data set (better data coverage in summer). Apparently, both the original data coverage and the non-ideal conditions affected the number of data points chosen for the final analysis. More detailed analysis is needed in order to generate any meaning results so we chose not to include such information in the revised paper.



Figure 1. Hourly and monthly number of data points available for analysis.

The most relevant problem is the calculation of the aerodynamic resistance in Eq. 5. This leads to an overestimation of the deposition velocity by the aerodynamic gradient method (AGM). But this aerodynamic resistance is also used in the proposed micrometeorological gradient method (MGM), Eq. 11. I assume that z2 is equal to h, because no other measurements were available. It is extremely difficult to make exact measurements at the top of the canopy because of the extreme gradient at this height, the heterogeneity of the forest and a possible dependence on the wind direction and the strong influence of the roughness sublayer (mixing layer). The authors encountered this problem through the strong influence of the wind velocity on the results, because the wind field penetrates more or less into the forest and the level with the extreme gradient is either a little bit above or below the top of the canopy.

AC: In Eq. 11, *h* is the height of canopy, which is smaller than z_2 in Eq. 3-5 since z_2 is the reference height at a level above the canopy. There are two unknown variables both in Eq. 11 and 9, i.e., C_h and F. By combining them, C_h and F can be both resolved.

It is not true that the AGM always overestimates the deposition velocity. If you measure not at the top of the canopy but at two levels at certain distances from the top, and apply a roughness sublayer correction function (Garratt, 1978), you can measure fluxes accurately (Wolff et al., 2010a; Wolff et al., 2010b; Foken et al., 2012). Unfortunately, this method is limited due to the accuracy of the gas analyzer, which is probably not good enough for ozone.

AC: We agree that not every study shows AGM overestimates flux. Some studies (Keronen et al., 2003; Stella et al., 2012) showed that $V_d(O_3)$ by the AGM and EC methods generally agreed well, while the other studies (Muller et al., 2009; Loubet et al., 2013) found a significant overestimation by the AGM method, consistent to what we found in this study. We have provided a brief summary of these earlier studies in the revised paper.

Because the aerodynamic resistance in Eq. 5 – and therefore also in Eq. 11 – is too small (flux and deposition velocity are too large), this must be compensated for by the aerodynamic resistance in the layer from h to z3, Eq. 10, so that the sum of both resistances in Eq. 12 is again accurate and a deposition velocity (flux) can be calculated in a good agreement with the eddy-covariance data. In other words, the calculation of the integral in Eq. 10 must be wrong (too large resistance), even when the Eqs. 13 ff appear to be in a good agreement with the theory. What was the tuning parameter of your model?

AC: We determined most of the parameters (e.g., leaf area density, roughness length, displacement height, wind attenuation coefficient) using measurements collected at the Harvard Forest and some parameters were chosen from literature (e.g., Prandtl/Schmidt number). Due to the limitation of available measurements, some parameters were derived from short-term measurements but applied to the calculation for long-term flux. Although there exist uncertainties, these parameters should be within a reasonable range. In section 4.2, we conducted the sensitivity tests to identify the key parameters/formulas and assessed the effects of parameter uncertainties on the model results.

The logic provided here seems to be right. However, if you take into account the following factors, the conclusion is not necessarily accurate. These factors include (1) the gradient between the two levels both above the canopy is much smaller than that between the two levels with one level inside the canopy, and (2) the flux above the canopy is constant (at least in theory) while the flux just below the canopy decreases rapidly with decreasing height. Thus, in the original AGM, underestimation of the aerodynamic resistance (R_a) overestimates deposition velocity (V_d) . In the MGM, it is the term that below the canopy (Eqs. 9 and 10) dominates the final V_d value. The underestimation in R_a (Eq. 5) should only contribute a small percentage in the overestimation of the final V_d . Thus, in the MGM method, Eq. 10 is not necessarily wrong. The reviewer's logic actually helped us explain why the MGM still slightly overestimate V_d (especially during night time when R_a value is large and play a more important role), which is likely caused by the underestimation of R_a (Eq. 5). In other words, if Eq. 10 gives an accurate estimation, then the underestimation of R_a in Eq. 5 should give a small overestimation in the final V_d in the MGM method, as is the case shown in our results. To confirm this, we conducted a sensitivity test by increasing R_a by a factor of 1.5 in both the AGM and the MGM methods (Figure 2). We can see that while V_d in AGM changes dramatically, V_d in MGM only changed slightly, which confirmed our argument above. We, however, do agree that if an existing R_a formula gives larger R_a values, then (Eqs. 9 and 10) can be chosen slightly smaller values. We need to keep in mind that all chosen parameters/formulas need to be based on available measurements and within reasonable ranges. We recently applied this MGM method to a fiveyear O₃ and SO₂ gradient data collected at our Borden monitoring site (Ontario) and we generated very reasonable V_d values for both SO₂ and O₃ (to be presented in a separate study), which demonstrates the applicability of this new method.



Figure 2. Sensitivity test using 1.5 times of R_a to replace R_a .

By the way, the applied universal function by Businger et al. (1971) in the modified form by Högström (1988) already includes a turbulent Prandtl number for the sensible heat flux ,or a turbulent Schmidt number for trace gas fluxes (Foken, 2006). On the other hand, you use a turbulent Schmidt number of 0.8 (p. 786, line 9); make sure that you did not use the turbulent Schmidt number twice.

AC: No, the turbulent Schmidt number was not used twice. The universal function for trace gas was applied in the calculation of aerodynamic resistance above canopy ($R_a(z_1:h)$) while the turbulent Schmidt number of 0.8 was applied in the calculation of aerodynamic resistance below canopy ($R_a(h:z_3)$).

The modified Bowen ratio method (MBR) was not the main topic of the paper, but it is important to show a good scalar similarity between ozone and the proxy (carbon dioxide). This is not trivial, because the ozone flux is influenced mainly in the morning by high reactions with NO, emitted during the night, and the assimilation is probably limited in the afternoon (Ruppert et al., 2006).

AC: We reviewed literature and found that many studies (e.g., De Arellano and Duynkerke, 1992; Duyzer et al., 1997; Gao et al., 1991; Padro et al., 1998; Stella et al., 2012) showed that the effects of chemistry on O_3 flux divergence in the near surface were generally small, likely because the chemical reactions for O_3 have larger time scales than the turbulent transport (which is likely due to the much higher O_3 concentrations compared to those of NO_x , Padro et al., 1998). Thus, the influence of chemical reactions on the similarity between O_3 and CO_2 is expected to be small. Of course many other factors may influence this similarity since different scalars have different source and sink terms. Detailed discussion on this topic is out of the scope of this study and existing literature certainly has substantial information on this topic.

For the final publication you should show which phenomena at the top of the forest canopy you excluded due to the data selection. The influence of the roughness sublayer should be discussed and the main point is: Because Ra (z1:h) is obviously too small, how have you modified Ra(h:z3) so that Ra(z1:h) + (Ra(h:z3) is again accurate?

AC: See our response and the figure provided to a comment above. While we agree that there is a possibility that R_a is an underestimation, measurement uncertainties in concentration gradients could also cause such big discrepancies between AGM and EC due to the very small gradients. This possibility is also supported by the fact that the MBR method also overestimates fluxes taking EC measurement as a standard.

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