Dear Referee#3,

We would first like to thank you for your very useful comments and suggestions that improved this study. We answer point by point your comments in the following, and when necessary included these answer in the revised manuscript, which we believe have helped improving the quality and readability of this paper.

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General comments

Although only ten days of measurements over bare soil are presented, the deposition velocity and, more specifically, the surface resistance results for O3 and NO2 deposition (in parallel) under such conditions are very useful, because such results are relatively rare in the literature. Some discussion on the relative magnitude of O3 and NO2 deposition velocities (or surface/soil resistance) used in the literature could be added.

Due to quality tests induced by the comments of referee #2, the NO_2 fluxes did were discarded as they did not satisfy the quality test. As a consequence, we choose in the revised manuscript to remove the Section 4.4 and to focus only on quality of AGM fluxes, which is much more in the scope of this journal.

The authors argue that the use of fast response sensors is preferable over slow response sensors to reduce the uncertainty of the gradients and fluxes (although they state that u is the main source of uncertainty?, see detailed comments below). Although the use of fast vs. slow response sensors is a scientifically interesting topic, it is treated in a too simple way in this MS. This is not just a problem of statistical error propagation but mainly of instrument performance. Fast response sensors usually have a higher noise level, are less stable (see e.g. the fast ozone sensor used here) and suffer more from interferences than slow response sensors. In order to really demonstrate the advantage of fast response sensors for gradient measurements it would be necessary to perform and present a careful evaluation of the time dependent noise as well as drift effects of both instruments. Alternatively, AGM fluxes from both slow and fast response sensors could be presented and compared to EC. Also the optimum switching interval between measurement heights would be an interesting problem in this context. Finally there remains the question why available fast response sensors should be used for gradient measurement and not for EC directly (especially for chemically reactive species)?*

In this comment, the referee dealt with several important issues. First of all, the referee stated that fast response sensors usually have higher noise level. Although this is true for a single instrument operated at several frequencies, this was not the case in this study. Indeed, the “fast instruments” were also the most sensitive, as shown in Table 1: the noise of fast response sensor is lower than the noise of slow response sensor for the NO and NO2 gas analysers. For the fast O_3 sensors, since the slope of the analyser is variable depending on coumarin target, it is difficult to answer this question. Regarding the drift effects of each instruments, we referred the referee to the answer we made to his comment below ("How large was the variability from one 6h calibration to the next?"). The Referee suggests to compare AGM fluxes from both slow and fast sensors. However, it is not possible since slow sensors only measured mixing ratios at 1.6 m and not at each levels (see Table 1 and Section 2.1). One very interested issue is to determine the optimum switching interval between measurement heights. Unfortunately, we do not have the data to address this issue in this study? In our opinion, it would require a specific study. Finally, the Referee asked why the available fast response sensors should be used for gradient measurements and not directly for EC. As we indicated in the introduction, the determination of the chemical corrections on fluxes for chemically reactive species required to have both the fluxes and the concentrations measured at several levels. The application of EC would require as many instruments as levels whereas AGM only require one instrument for several levels. In addition, as we showed in this study, the use of fast sensors allowed to diminish the flux uncertainties. Finally, it should be noted that for example for O_3, the slow ozone sensor based on UV absorbance are subjected to water vapour interference (Wilson and Birks, 2006), which could be critical for gradient measurements. We included in the conclusion of the revised manuscript a discussion regarding this last remark.

The discussion of the NO fluxes by gradient and chamber methods seems to be quite biased. Only potential errors for the chamber methods are considered (spatial heterogenity, not representative concerning rain etc.). Yet it has to be mentioned that also the gradient method may become problematic for heterogenous conditions because the concentration measurements at different heights do not have the same footprint. In addition, the chemical correction performed relies on assumptions that could not be really proven, and it is probably incomplete because there are other chemical reactions beside the ones considered here (e.g. NO with peroxy radicals). Given the importance of the applied chemical correction in this study, its uncertainty and problems with the assumptions should be assessed and discussed in more detail.

We included in Section 4.3 a discussion concerning the problems indicated by the Referee, i.e. (i) the limit of the AGM in the case of heterogeneous fluxes, and (ii) the potential error in the chemical correction method we used for the chamber measurements, which only included reaction with NO and O_3. Unfortunately, for this last statement, no measurements of peroxy radicals were available, limiting the discussion concerning this issue.

The applicability of the presented calculation of the AGM flux uncertainty and detection limit should be discussed critically. A detection limit of 0.08 nmol m^-2 s^-1 is reported for the O3 flux. However, when looking at Fig. 9a, the detection limit (= individual fluxes clearly discernable from zero) seems to be rather at about 2 nmol m^-2 s^-1, if the EC fluxes are taken as a reference. So either the effective detection limit for AGM fluxes is more than 10 times larger than expected or the EC flux results have a much lower quality than the AGM fluxes!?

Here, the referee stated that the detection limit of the AGM O_3 flux should be around 2 nmol m^{-2} s^{-1}. The referee based this assumption on the results presented in Figure 9a, especially the data closed to 2 nmol m^{-2} s^{-1} for the AGM while EC fluxes are closed to 0 nmol m^{-2} s^{-1}, data corresponding to nocturnal conditions with u_* typically lower than 0.2 m s^{-1}. However, as discussed in Section 4.3, it is not new that EC fluxes are underestimated under such conditions (e.g. Goulden et al., 1996; Jarvis et al., 1997; Black et al., 2000; Valentini et al., 2000, Gu et al., 2005; Moureaux et al., 2006). Thus, EC method cannot be assumed as a reference method under these conditions, and these latter conditions should not be considered to define the detection limit of the AGM.

Detailed comments

p5482 L15: "...gave similar O3 fluxes (within 4%)" This formulation is misleading, since the 4% refer to the slope of the overall linear regression line. The agreement of halfhourly fluxes is generally much less than 4%!

This is a sound remark and we acknowledge the Referee to have pointed out this mistake. Indeed, the percentage we gave referred to the slope of the regression. We evaluated that the half-hourly flux difference was on average 31% but it could reach up to 200%, especially when fluxes were weak. We modified the abstract and the Section 4.3 in consequence.

p5484 L21-22: "... the few previous comparison studies..." Either give additional references here or specify (if applicable) "...the two studies mentioned above...".

We correct the sentence as "In addition, the few previous comparison studies (Droppo, 1985; Mikkelsen et al., 2000; Keronen et al., 2003;Muller et al., 2009) did not correct the fluxes for chemical reactions before comparing the different methods."

p5485 L15-16 and following paragraph: Omit all method descriptions for which no results are included in the current manuscript.

Done.

p5486 L27: Give a detailed description of the used fast ozone sensor (FOS) since it seems that it has not been described before in scientific literature. This is especially important considering the topic of the paper and the focus of this journal.

We modified this paragraph in order to include the description of the fast ozone sensor as such: "Mixing ratios were measured with fast chemiluminescent gas analysers for O_3 (FOS, Sextant Technology Ltd, New Zealand), NO (CLD780TR, Ecophysics, Switzerland) and NO_2 (LMA 3D-NO_2, Unisearch Associates Inc, Ontario, Canada) (Table 1). The fast chemiluminescent gas analysers for NO and NO_2 were already used and described in previous studies (e.g. Nikitas et al., 1997; Jones et al., 2000; Rummel et al., 2002; Bröske et al., 2003; Hall et al., 2008). For O_3, the fast chemiluminescent gas analyser is based on the chemiluminescence of a coumarin dye absorbed on silica gel reacting with ozone. The chemiluminescence is monitored with a very sensitive photomultiplier. A pump inside the instrument allows a constant flow rate of 2 L min^{-1} to be maintained."

p5487 L3: How large was the variability from one 6h calibration to the next?

Over the 6h periods, the calibration was quite stable for NO and NO_2. The span varied around 2.6% and 4.1% between each calibration on average over the whole study period for NO and NO_2 respectively. The offset varied around 11.6% and 4.0% between each calibration for NO and NO_2 respectively.

For O_3, the variability of calibration coefficients was more important over the whole study period due to the loss of sensitivity of the chemiluminescence of coumarin dye with time. Especially when we changed the coumarin dye, the calibration coefficients varied up to 100% between to calibrations. However, the offset remained very low throughout the period.

p5487-88: Equations 3-5 (as well as Eq. 11) could be omitted since they represent common textbook knowledge and are referenced in the text (e.g. Dyer and Hicks, 1970).

These equations were removed.

p5489 Eq.6: This method for the flux determination only works, if there is no offset in the raw fast ozone sensor signal. Was that checked regularly? Please comment on this issue in the manuscript.

The fast ozone sensor used for EC flux measurement had a very small offset, thus that had no impact on the method for flux determination.

p5489 L7: What does "averaged concentration standard deviation" mean here? Averaged over both levels? For the ability to resolve the gradient between two mean concentrations, wouldn't the standard error of the means be more meaningful?

This issue was also pointed out by the referee #1. Here, we meant that we used the average of the concentration difference between two successive levels, divided by the standard deviation of the averaged concentration difference to estimate the signal to noise ratio. We coupled in the revised manuscript this approach with a more statistical approach using Student's t-test to resolve the mixing ratio gradient. This latter issue was included in Sections 2.4, 3.2 and 4.1.

p5489 Eq. 7: It should be pointed out that this uncertainty estimation is only valid for the non-reactive case.

Done

p5491 The definition of the trace gas fluxes in Eqs. 10 and 12 (and throughout the manuscript) is not consistent and partly confusing. Define and use a clear and consistent nomenclatura for fluxes (with/without chemical corrections and at different heights z) throughout the manuscript (also in Figures).

Done

p5491 L17-18: Is this assumption reasonable? What is the uncertainty introduced with the assumptions in this relatively simple chemical correction?

This assumption seemed to be reasonable. Indeed, we estimated that the fluxes with chemical corrections differed from less than 1% for O_3, 3.8% for NO and 4.6% for NO_2 if we assumed $z_2 = 1.6$ m or $z_2 = 4$ m.

p5495 L17: Replace "the end" by "the second half"

Done

p5496 L14-15: This statement is clearly misleading. It does not apply to (individual) measured NO concentrations, but describes the systematic overall deviation determined by a linear regression. This has to be clarified.

We indicated that there was a systematic overall deviation of only 1% between NO mixing ratios measured with fast and slow sensors.

p5497 L14-19: Relative corrections alone are not very informative (because the respective fluxed and thus the absolute correction could be close to zero). Additionally give and discuss absolute effects of chemical corrections for the fluxes.

Done. The absolute chemical correction was 0.12 nmol m^{-2} s^{-1} on average for both NO and O_3, but could reach 1.44 nmol m^{-2} s^{-1} during the large soil NO emission period.

p5498 L3-22: In the formulation of this paragraph the issue of fast vs slow response sensors is mixed with the selectivity (specific detection of NO2) of the analysers, which is confusing. When discussing the issue of selectivity, use the names of the sensor instead of 'slow-response' and 'fast-response'. The selectivity of an analyser has no direct relation to its time response (at least not in this study).

We replaced "slow response" and "fast response" by the names of the analyzers. We also did it in Section 3.2 and in Figure 3 to be consistent throughout the manuscript.

p5499 L6-12: There is an obvious contradiction in this paragraph! First the authors say that the flux uncertainty are mainly due to u-uncertainty. But later it is said that the (relative) O3 flux uncertainty was considerably lower than for NO and NO2 because of the higher precision of the O3 gradient measurements. (same problem on p5505 L13-16) If the precision of the gradient measurement has such a clear influence on the resulting flux uncertainty, the gradient uncertainty is the limiting factor for the flux uncertainty (and not mainly u*)!*

We acknowledge the fact that we were not clear enough on this issue. We wanted to indicate here that the flux uncertainty was mainly due to u_* when u_* was small while it was mainly attributed to gradient uncertainty when u_* was large. We rephrased this paragraph to better explain our point: "The relative uncertainty of O_3 and NO fluxes was dependent on the friction velocity and ranged from 150-200 % to 20 % and 40 % respectively (Figure 5). Flux relative uncertainties were due to both u_* and mixing ratio gradient uncertainties. Indeed, the mixing ratio gradient uncertainties contributed to σ_{C_*}/C_* while u_* contributed (i) on the one hand to σ_{u_*}/u_* and (ii) on the other hand to the scaling parameter error σ_{C_*} (i.e. in the Ψ_H function through Obukhov length estimation). Thus, when u_* was weak, typically during night-time, the uncertainties of the mixing ratio gradient were small and of similar magnitude for the two gases (Figure 4a), but in this case, the flux uncertainty was dominated by the uncertainty on u_* which affects both terms σ_{u_*}/u_* and σ_{C_*}/C_* of equation (9). On the contrary, when u_* was large, the uncertainties of both u_* and the mixing ratio gradients contributed equally to the flux uncertainties. This last statement also explains why the O_3 flux uncertainty was nearly two times lower than the NO flux uncertainty when u_* was large, typically daytime conditions. During daytime, the O_3 flux was large, which led to a large mixing ratio gradients ($\Delta C/\sigma_C > 1$), compared to the NO mixing ratio gradient which was much smaller (Table 2)."

We also modified the conclusion accordingly.

p5504 L18-20: It seems not very meaningful to compare deposition velocities for bare soils with literature values for vegetated surfaces like turfgrass or other vegetation. If no values for bare soil are available, the present results could be compared to nighttime values over vegetated surfaces (excluding stomatal uptake) or deposition measured at the forest floor.

As indicated above, following the comments of the Referee#2, we removed this Section to focus on quality analysis which is more in the scope of the journal "Atmospheric Measurement Techniques".

p5505 L21-23: This statement is misleading. As mentioned earlier in the manuscript (p5501) high flux divergence typically occurred during nighttime when turbulence was weak. It is trivial that NO2 photolysis does not play a role in the dark!

This statement was removed.

p5516 Tab.2: A classification of the results with u (or z/L) would be useful here.*

Done

p5517 Fig.1: If possible give a better illustration of the relative tube lengths (before/after the branching off of the subsample lines to the analysers.

Done

Language corrections

p5482 L9-10: "It was demonstrated that ... appeared to be negligible for O3 fluxes ..." This formulation is awkward and has to be changed.

It was replaced by " The chemical reactions between O_3-NO-NO_2 had a weak impact on O_3 fluxes (less than 1% on average), whereas they accounted for about 10 % on average of the NO fluxes."

p5484 L1: interchange "and" and "or" in this sentence.
The sentence was changed according to the suggestion of referee #1 as "it is possible to distinguish between the direct micrometeorological methods (such as the eddy-covariance (EC), the relaxed eddy-accumulation and the disjunct eddy-covariance), the indirect micrometeorological methods (such as the aerodynamic gradient method (AGM) and profile methods) (Foken, 2008), and the chamber methods (Meixner et al., 1997; Denmead, 2008)"

p5484 L12-17: Rephrase these two sentences. The term "Alternatively" is not appropriate here.
We rephrased these sentences as: "Moreover, estimating the fluxes of chemically reactive species, especially ozone, nitric oxide and nitrogen dioxide, requires measuring both the concentrations and the fluxes at several heights to estimate the flux divergence due to chemical reactions (Kramm et al., 1991; 1995; Duyzer et al., 1995). Although the EC method could be applied to measure simultaneously the flux at several heights, this issue could suffer technical limitations because it would require several fast analyzers. Thus, the AGM represents the simplest alternative."

p5485 L13-14: The formulation is not clear. Do the author mean "slurry application...and incorporation by tillage..."?
This sentence was corrected as "...following cattle slurry application of 98.5 kg_{N-NH_4} ha^{-1} and incorporation by tillage...".

p5486 L14: "The aerodynamic gradient method was used to determine..." (It is a very general rule in literature that titles must not be treated as integral parts of the flow text!)
Corrected.

p5486 L20: Replace "tension" by "voltage".
Done

p5489 L14: "basic" instead of "basics"
Done

p5489 L19: "deviations of w and u"
Done

p5495 L14-15: "...had a marked daily dynamics." Rephrase this sentence.
This sentence was removed

p5495 L17: Omit or replace "Globally"
Done

p5496 L20: Replace "daily" by "diurnal"
Done

p5496 L7: Replace "day cycle" by "diurnal cycle"
Done

p5499 L2: Replace "these" by "those"
Done

p5504 L6: "variable" instead of "variables"
This section was removed

p5504 L14: "... of about 0.35 cm s-1"
This section was removed

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