

Dear Referee#2,

We first would like to thank you for your very useful comments on this manuscript. We answered to your comments in the following. Most of your comments and questions were taken into account in the revised manuscript which we believe have improved its quality and understanding.

Each comment has been answered to in details..

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The paper has a significant problem in the interpretation of the data, and this comes to a focus in the conclusion that “The AGM flux uncertainties were mainly due to friction velocity”. Atmospheric turbulence is a phenomenon which allows a very effective transport between the atmosphere and the underlying surface. The methods used in the paper, the eddy-covariance method (EC), the aerodynamical gradient measurements (AGM, Eqs. 1-3), the transfer time (Eq. 13), and the uncertainty analysis (Eq. 8) are based on a fully developed turbulent regime. In the case of low wind velocities and also as a consequence of low friction velocities, this assumption is not valid. Turbulence is either intermittent or still missing with a nearly laminar flow. These conditions must be excluded from the application of the above given methods. This can be done e.g. with a test on steady state conditions (Foken and Wichura, 1996; Vickers and Mahrt, 1997). If the test fails, the data must be neglected or analyzed with special methods like conditional sampling or wavelet spectra. Also the test on developed turbulence with integral turbulence characteristics is possible (Foken and Wichura, 1996). Another way is used in ecology, where all data with friction velocities below a given threshold are neglected (Goulden et al., 1996; Papale et al., 2006). I propose for the revision of the paper the latter method with a threshold of $u^* = 0.15 \text{ ms}^{-1}$ for bare soil, which is much lower than the threshold used in ecology. The neglecting of all non-turbulent situations does not only change some of the figures like Fig. 5 but also some of the text and the conclusions. A careful analysis of the developed turbulence is highly relevant for the application of the AGM because of large gradients and very small diffusion coefficients and, consequently, very low fluxes.

We thank referee #2 for this very sound remark. Although the referee proposed to simply use a threshold of $u^* = 0.15 \text{ m s}^{-1}$, we carried out stationarity tests on mixing ratios for O_3 , NO and NO_2 . The results showed that stationarity was linked with friction velocity for O_3 and NO , i.e. linked with the intensity of the turbulence. However for NO_2 , the mixing ratios did not satisfy the stationarity test for any u^* . We hypothesized that it was due to local advection due to road traffic around the field: the NO emitted by cars was rapidly converted in NO_2 by reaction with O_3 and advected to the field.

As a consequence, since NO_2 mixing ratios did not satisfy the stationarity test, the NO_2 fluxes estimated by AGM were discarded in the following of the study. The manuscript was consequently significantly revised according to these new results: (i) the Section 2.4 was modified to include the description of the stationarity tests, (ii) the results of stationarity test were included in Section 3.2 and (iii) discussed in Section 4.1. In addition, the uncertainty of NO_2 fluxes was not presented since the fluxes were discarded. Finally, this new result induced to modify the discussion in sections dealing with chemical corrections and flux divergence. Indeed, since the method we used required to know the NO_2 fluxes, this method could theoretically not be applied, since the NO_2 fluxes were too uncertain. However, we

hypothesized that the magnitude of the NO₂ fluxes was true to estimate the chemical divergence of O₃ and NO fluxes based on the magnitude of the NO₂ flux estimated.

Further remarks.

p. 5485, line 22ff and Eq. (1): Please make sure that you used the friction velocity from EC data (i) and not from the calculation with AGM (ii). For case (i) the measurements of the wind profile are not relevant and for case (ii) the distance constant of the anemometers and the possible overspeeding correction are relevant (Wiering, 1980).

The friction velocity used was determined from EC measurements. We removed the description of wind speed profile measurements in the revised manuscript.

p. 5486, line 8: For the reader it would be helpful if the reference of the footprint model used can be given.

The footprint model described by Neftel et al. (2008) was used. It was included in the text.

p. 5486, line 20: Is the flow turbulent? Please give the Reynolds number.

The flow inside each line was turbulent. Indeed, the Reynolds number was 5900, for a threshold for the transition between laminar and turbulent flow around 2000-3000. This was indicated in the text.

p. 5487, line 15 ff: The universal function by Dyer and Hicks (1970) and especially the von-Kármán constant of 0.41 are not state of the art and should be probably replaced by the function by Businger et al. (1971) in the modification by Högström (1988), see e.g. Foken (2006).

We used the universal functions proposed by Dyer and Hicks (1970), while many others could also be used. Indeed, the Referee suggests using the universal functions of Businger et al. (1971) modified by Högström (1988), but there are several expressions of the universal functions that were previously proposed (e.g. see Foken (2008), Appendix A4, pp 250-252).

Referee #2 stated that the universal function we used, and especially the von-Kármán constant set at 0.41, is not state of the art. First of all, the von-Kármán constant of 0.41 was used by Dyer and Hicks (1970) to establish this universal function. The modification of the von-Kármán constant can lead to important modifications in the expression of the universal function (see for example the re-calculations by Högström (1988) using $k = 0.40$, summarised in Foken (2008), Appendix A4, pp 250-252). In addition, the von-Kármán constant estimated from different sources has shown large uncertainties, with its precise value supposed to vary from 0.35 to 0.42 (Panofsky and Dutton, 1984; Zhang et al., 1988; Andreas et al., 2006, Foken, 2006). Finally, the functions proposed by Dyer and Hicks (1970) and using $k = 0.41$ were extensively used in previous studies (e.g. Raupach, 1979; Sutton et al., 1993a, 1993b, 2000; Laville et al., 1999; Miyata et al., 2000; Nemitz et al., 2009).

However, to check the effect of changing k , we compared fluxes of NO, O₃ and NO₂ estimated from AGM using the stability functions proposed by Dyer and Hicks (1970) and by Businger et al. (1971) modified by Högström (1988). The results indicated that the fluxes estimated using stability functions proposed by Dyer and Hicks (1970) were systematically greater (by roughly 10% on average) than those obtained using the stability functions proposed by Businger et al. (1971) and modified by Högström (1988).

We propose to include in the revised manuscript a specific discussion concerning this issue at the end of Section 4.1 ("Quality of NO-O₃-NO₂ AGM fluxes") as:

"In spite of the possibility to decrease the relative flux uncertainties by increasing the acquisition frequency, it must be kept in mind that one important source of uncertainty is the choice the of stability functions to calculate fluxes from the AGM. In this study, the stability functions proposed by Dyer and Hicks (1970) were used, but several others exist, in particular the stability functions proposed by Businger et al. (1971) and modified by Högstrom (1988). Table 4 shows the comparison of O₃, NO and NO₂ fluxes deduced from AGM using the stability functions proposed by Dyer and Hicks (1970) and by Businger et al. (1971) modified by Högstrom (1988). This result shows that the O₃, NO and NO₂ fluxes estimated using stability functions proposed by Dyer and Hicks (1970) were systematically greater (by roughly 10% on average) than those obtained using the stability functions proposed by Businger et al. (1971) and modified by Högstrom (1988). However, for 25% of the time, this difference reaches 14% to 15% ."

We included the table below in the revised manuscript to illustrate this result:

"Table 4: Relative difference between AGM fluxes (of O₃, NO and NO₂) determined using the stability functions proposed by Dyer and Hicks (1970) and those proposed by Businger et al. (1971) and modified by Högstrom (1988). Positive values indicate greater fluxes using the stability functions proposed by Dyer and Hicks (1970)"

	O ₃	NO	NO ₂
1 st Quartile	+6.4%	+5.9%	+6.3%
Median	+9.4%	+9.3%	+9.5%
3 rd Quartile	+14.5%	+14.4%	+14.8%

p. 5488, line 5: Perhaps some information about the software used and the quality control would be helpful.

We used the Edire software. The quality control was assessed following Aubinet et al. (2000). We included these issues in the text as " Flux calculation and quality control were assessed using the Edire software (Robert Clement, University of Edinburgh, United Kingdom) following the CarboEurope methodology (Aubinet et al., 2000)"

p. 5488, line 12: Please replace Monin-Obuchov length by Obuchov length (Businger and Yaglom, 1971; Foken, 2006; Obuchov, 1971).

Done

p. 5488, line 15: It is positive that you used the Obukhov length with the buoyancy flux (in your case combination of the sensible and latent heat flux). But in this case you have also to replace the temperature by the virtual temperature. Why did you not use the buoyancy flux, which you measured directly with the EC method and to which you probably applied the Schotanus et al. (1983) correction to determine the sensible heat flux. Remark: The universal functions are defined with the Obukhov length without buoyancy flux

After checking the equation used in the Edire software to calculate the Obukhov length, we realized that there was an error in our equation in the manuscript. The equation used in Edire software to calculate the Obukhov length is:

$$L_o = \frac{\rho \cdot u_*^3}{k \cdot g \left(\frac{-H}{(T_v + 273,15) \cdot C_p} \right)}$$

where T_v is the virtual (or sonic) temperature (in °C) and H is calculated from the measured sonic temperature (i.e. from $\overline{w'T_v'}$). As far as we understand the Referee, it is consistent with its comment. In the revised manuscript, we modified the equation as:

$$L_o = \frac{u_*^3}{k \cdot g \left(\frac{-\overline{w'T_v'}}{T_v} \right)}$$

where T_v is the virtual or sonic temperature in K.

Concerning the remark concerning the universal functions defined without buoyancy flux (i.e. for dry air), we acknowledge the Referee to point out this issue. However, as indicated by Foken (2006), the experiments to determine the moisture influence on the universal function are lacking, and the possible influence should be very small.

p. 5489, Eq. 6: This equation is trivial and has been used for a long time for all ozone flux measurements.

We removed this equation.

p. 5491, Eq. 10: You make the assumption that the turbulent and Prandtl and Schmidt numbers are identical.

If we well understand the Referee, its comment refers to the fact we used ϕ_H as the stability function for the three gases. We changed is the Eq. 10 ϕ_H by ϕ_X and clearly indicated in the text that $\phi_X = \phi_{NO} = \phi_{O_3} = \phi_{NO_2} = \phi_H$

p. 5491, Eq. 12: Why you did not make a stability correction (universal function)?

We chose ϕ_X at the gradient height (i.e., 0.61 m).

p. 5492, Eq. 13: Please give the definition of Ra and Rb. This means no extension of the paper, because most of the relevant equations are already given.

Done

p. 5495, line 29ff and p. 5498, line 27ff: Probably it would be interesting to separate the data set for wind coming from Paris and the other wind directions and to make the following investigations for both data sets.

We tried to separate the data set according to wind direction. Excepted for NOx mixing ratios that clearly showed a trend as indicated in the text (larger mixing ratios when wind comes from Paris), no further trend was observed due to the local advection coming from every

direction. Thus, we did not separate the data set according to wind direction in the revised manuscript.

p. 5497, line 3 ff and p. 5499, line 4ff: This part, and similar parts in the paper, must be revised. If no fully developed turbulence exists, the Monin-Obuchov similarity is not fulfilled (basis for AGM), therefore no turbulent flux can be determined and no error of a turbulent flux exists. If a flux is very low or even below the detection limit a relative error makes no sense. Please give for these cases an absolute error, probably in combination with a relative error for larger fluxes. Fig. 5 must be revised accordingly.

These sections were revised according to the new results obtained (i.e. stationarity tests, see the first comment). In addition, we also added a statistical analysis on mixing ratio gradients. Only data for O₃ and NO were presented following the results of this quality analysis.

p. 5498, line 23 ff: Except for ozone only at night (but here are the fluxes nearly zero due to the missing turbulence) and in some single cases, the gradient is larger than the detection limit of the applied instruments. This is not new and an overview has already been given by Foken (2008, p. 134-137). Please add your efforts to make the application of the AGM nevertheless possible. For ozone your results are not so bad. Please also discuss the possibility of detecting, for developed turbulence, the effect of chemical reactions (p. 5497, line 10 ff).

In order to address this comment, we carried out a statistical analysis to determine if mixing ratio gradients were significant. To this aim, Student's t-tests were carried out on paired samples. If one gradient between two levels was not significant, we calculated the fluxes between the highest and lowest levels only (after checking that the mixing ratio gradient between 0.2 m and 1.6 m was significant). This analysis was included in the revised manuscript. See answers to referee #1 comments for further details.

p. 5503, line 17: What is “soil ozone flux”? Figs. 3, 5, 9: The high accuracy of the regression calculations is unrealistic.

We meant the ozone flux to bare soil. Concerning the accuracy of the regression calculations, we checked them using different software (Excel, Xlsstat, Statistica) and calculating directly the R² value. The results were always identical to the values indicated in Figs 3, 5 and 9.

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