

Interactive comment on “Measurement and modelling ozone fluxes over a cut and fertilized grassland” by R. Mészáros et al.

R. Mészáros et al.

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Dear Editor!

Thank you for your comments. According to your suggestion, we have prepared a detailed response, which includes proposed new text for the revised manuscript.

We are grateful for the sound and useful comments of the reviewers. We have corrected the indicated technical corrections and typographical errors. Based on the suggestions of referees, the measured dataset has been revised, and new model simulations have also been carried out. Abstract, site and model description have been rewritten. Revision and extension of discussions has also been performed.

In the following we are addressing the specific points raised by the reviewers:

Answers to reviewer 1:

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Major point: The NO flux can affect the ozone concentration. As the deposition velocity was calculated by the measured ozone flux and concentration, therefore NO flux also affects indirectly the deposition velocity.

"After the fertilization, an assumed increase in NO emission (inferred by greatly increased soil nitrate levels) is also thought to have affected the ozone deposition. In the presence of higher soil NO emissions, a higher O₃ deposition flux would have been expected. However, due to the lower canopy and therefore the faster transfer time, there would also have been less time for the NO-O₃ reaction."

Minor points:

(i) We agree that there is a strong correlation between precipitation and soil water content. However, despite the frequent rain events, the soil water content decreased during the measuring period because of higher temperature and higher evaporation in the second and third part of measurements. Evaporation from soil could be more effective after cut of the vegetation. As the soil water content measurement wasn't continuous, the effects of short precipitation events (showers) can't be seen in the time series of soil water content. Volumetric soil water content was measured in different depth by TDR (Time Domain Reflectometry). In the revised manuscript, the average of volumetric water content in 0-60 cm soil layer is presented in Fig. 1.

(ii) We have restructured Figure 2. Cut and fertilization are also marked in the figure.

(iii) P 1072 Eq 1: Units of $\langle a \rangle$ is ppb/mV, as $\langle U \rangle$ means the sensor output in mV.

Answers to reviewer 2:

- The calculation of stomatal flux is explained in more detailed in Section 3 of the revised manuscript. The stomatal flux has been calculated from Ft (total flux), obtained from measured concentration at a reference height and calculated deposition velocity. This flux was multiplied by Rc/Rst. Another way can be used to calculate the stomatal part of the flux (e.g. in EMEP model - www.emep.int), however, the two ways give the

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same results.

- According to the suggestion of another referee, another parameterization of cuticle and in canopy resistances has been used after Zhang et al. (2003). "Rcut and Rac were parameterized as a function of the Leaf Area Index (LAI). Zhang et al. (2003) suggested that cuticle resistance may parameterized differently for dry (Rcutd) and wet (Rcutw) canopy."

- Model description has been shortened and restructured. Instead of Jarvis-type parameterization of stomatal resistance, this term has been calculated from the measured latent heat flux which was also available during the measuring campaign (Nemitz et al., 2009).

- Single-sided LAI is used in this calculation. LAI was measured in a few days during the measuring campaign and a simple function was fitted to measured values before and after cut based on vegetation height, which was continuously measured with a canopy height meter.

- Friction velocity (u^*) is presented for three different period. "The friction velocity is generally lower in night-time, however in the first period, u^* was relatively high in night-time (Fig 4), which can compensate the effect of higher LAI on Rac."

- p1077, section 4.1: The soil water content was sampled using TDR (Time Domain Reflectometry) at different soil depth. It is mentioned in the revised manuscript in the Section 2.2.

- p1077, section 4.1: LAI values have been corrected in the manuscript: "The canopy LAI was larger than 3 m² m² at the beginning of the experiment and decreased to less than 0.14 m² m² after the cut, before the canopy started re-growing."

- p1078, section 4.2: We agree that no significant variations in wind speed among each period, and that soil moisture had a greater variability during the measuring period. We have reanalyzed the variables and parameters which affect the ozone fluxes. Section

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4.2. has been restructured and rewritten. Details of meteorological conditions during the experiment have been described in Section 4.1 in the revised manuscript.

-p1078 and 1079 Discussions: Based on the suggestions of referees, new model calculations have been performed and therefore the fractions of stomatal flux has been varied:

"The daily pattern of the ratio of stomatal to total ozone fluxes for the three periods indicates that in daytime, the stomatal flux represented around 80% of the flux before the cut but only around 50% after the cut."

During new model estimations, cuticle abd in canopy resistances have been parameterized by a more sophisticated way after Zhang et al. (2003) and dry and wet vegetation have also been distinguished. Soil resistance was parameterized with a simple linear function considering the effect of soil water availability. These modifications affected the fractions of ozone fluxes through the different ways.

"Both stomatal and cuticular pathways decreased after the reduction of LAI in the second period. The ratio of other non-stomatal pathways (in canopy + soil) increased after the cut, due to the reduction of R_{ac} and R_g . In canopy resistance was lower after cut because of lower LAI, while the reason of lower soil resistance is the dryer soil after cut. "

- Discussion has been rewritten and extended. (Section 4: Results and discussion - as whole section has been rewritten according to the new results and considering the suggestions of the referees, this part is presented only in the revised manuscript.)

Answers to reviewer 4:

1. The abstract has been rewritten according to suggestions of the reviewer:

"During the GRAMINAE Integrated Experiment between 20 May and 15 June 2000, the ozone flux was measured by the eddy covariance method above intensively managed grassland in Braunschweig, northern Germany. Three different phases of vege-

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tation were covered during the measuring campaign: tall grass canopy before cut (29 May, 2000), short grass after cut, and re-growing vegetation after fertilization (05 June, 2000). The main aims of this study are the following: analysis of the effects of agricultural activities (cut and fertilization) on ozone fluxes, parameterisation and estimations of the deposition velocity and flux of the ozone by a detailed deposition model, and evaluation of the ratio of stomatal and non-stomatal ozone fluxes over different-phase grass canopy. Results show that beside weather conditions, the agricultural activities can also significantly influence the O₃ fluxes. However, both cut and fertilization have complex impacts on fluxes. Reduction of vegetation by cut decreased the stomatal flux, while at the same time for this short canopy, the role of both soil emission of NO (promoting ozone loss close to the surface) and deposition of ozone to soil surface increased. These effects demonstrate the importance of canopy structure and non-stomatal pathways on O₃ fluxes. Measured and modelled flux and deposition velocity values have also been compared for each period."

2. A more detailed resistance network has been used in the new calculations using Zhang et al. (2003) resistance network. Stomatal resistance for ozone (R_{st}) was calculated from stomatal resistance for water vapour (R_s) obtained from measured water vapour flux. Both cuticle and in-canopy resistance have been parameterized as a function of the Leaf Area Index (LAI) and friction velocity (u^*). Additionally, cuticle resistance have been estimated differently for dry (R_{cutd}) and wet (R_{cutw}) canopy after Zhang et al. (2003). Soil resistance (R_{soil}) was parameterized with a simple linear function considering the effect of soil water availability.

3. This part of manuscript has been rewritten. A new model simulation has been performed using the above mentioned resistance network.

4. This part of the manuscript has been rewritten. Both stomatal and non-stomatal resistances have been calculated in different ways as in the original manuscript.

5. Section 4 and 5 have been combined into one section. (Section 4: Results and

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discussion)

6. Sections 4, 5 and 6 have been rewritten according to comments of reviewer. Section 4 contains the results with detailed discussions about meteorological conditions (Section 4.1) and ozone concentration, flux and deposition velocity (Section 4.2). Section 5 (Conclusion) have also been rewritten. In this section, we have summarized the results of field experiment (measured and modelled ozone flux and deposition velocity during the measuring period in different environmental conditions and agricultural activities).

7. We agree that only very limited data are available, and therefore the results may be not statistically significant. However results can underlie the importance of non-stomatal resistance on ozone deposition.

"Because of the several data filtering mechanisms were needed to remove false or doubtful data, only a limited set of results was available for these analyses. However, the results obtained from both measurements and modelling can contribute to the understanding of the soil-vegetation-atmosphere interactions."

8. Notation of each period has been rewritten according to the suggestion.

9. The text of the manuscript has been corrected.

Answers to reviewer 5:

(1) Ozone flux measurements

(a) We have clarified the location of the reference ozone monitor. The reference ozone concentration was measured at Broitzem urban background measuring site, 5 km from the flux site. We agree that this distance can cause some variations in the values of ozone concentration, however we assumed, that there are no significant differences between ozone concentrations in the background station and the flux site. The environment of the flux site is flat and surrounded by arable and other managed grassland to the N, woodland, by grassland and sub-urban development to the E and S, and by experimental arable plots and a farm to the west.

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"Based on Diem (2003) the ozone concentration measurements could be representative up to 10 km, therefore we have assumed that in this flat, suburban region there are no significant differences between ozone concentrations in the background station and the flux site. However, it is obvious that local effects can cause some spatial differences in ozone concentration, therefore any periods, when the correlation coefficient between reference ozone concentration and fast response ozone measurements was lower than 0.8, were neglected."

(b) The fast response ozone sensor requires a calibration to evaluate the ozone flux. This calibration has been made by the reference ozone concentration. "Measured deposition velocity has been calculated as the ratio of measured flux and measured reference ozone concentration." We have clarified it in the manuscript.

(2) Data processing

(a) The signal of the fast response ozone sensor was collected by an analogue channel of the Gill sonic anemometer at a frequency of 20.695 Hz. The air inlet tube of the fast response ozone sensor was 2 m long causing some time lag. Time lag for ozone measurement was estimated as the maximum of the covariance between the vertical wind speed and the ozone concentration using the 15 min averaging time. Vertical wind speed and ozone data sets were detrended in order to determine the time series of fluctuations by the moving average technique using a 400 s time window for the estimation of the mean values (McMillen, 1988).

(b) The diameter of air inlet tube was 3 mm and the flow rate was approximately 0.6 L/min. We have justified the ignorance of WPL correction for heat in the revised manuscript: "The NOAA fast response ozone sensor is a closed path type sensor. The relatively large time lag (1.59 s) of this sensor, as compared to other sensors with a 0.1 s time lag, allowed temperature of the sampled air to equilibrate with the sensor temperature. If the trace gas concentrations are determined by closed-path gas analyzers, where the respective trace gas is brought to a common temperature and

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pressure within the optical bench, then there is no need for a correction associated with the transfer of sensible heat (Grünhage et al., 2000)."

(3) Data analysis

(a) New data processing of measured ozone concentration and ozone flux has been performed and the new results are discussed in detailed in the manuscript:

"Average diurnal variations of ozone concentrations, fluxes and deposition velocity are illustrated in Fig. 3 for the three periods, i.e. before the cut, after the cut and after fertilization. It can be seen that diurnal variations of ozone concentration are pronounced in second and third periods and highest night-time values is found in the first period (Fig 3a). The daily pattern of measured ozone fluxes (Fig 3b) shows that highest day-time values occurred in the first period, and the flux decreased after cut. However this decrease was lower than it was expected due to the large reduction of LAI, which decreased both stomatal and cuticular uptake. This indicates that the non-stomatal (in canopy and soil) flux may have rose or that the stomatal flux before the cut was not proportional to the LAI due to shading of the lower canopy leaves. Highest day-time average of ozone concentration in the second period could also compensate the diminution of the flux."

(b) We agree that water vapour flux is commonly used to estimate the stomatal resistance. For water vapour, if it is assumed that over a transpiring canopy with dry leaf surfaces, the bulk of the latent heat flux is transported via the stomatas, then it is possible to calculate a bulk stomatal resistance, as it was also carried out during the measuring period (see Nemitz et al., 2009.). We have calculated the deposition velocity and ozone flux using stomatal resistance estimated from latent heat flux.

"Stomatal resistance is a key parameter in deposition modelling, which is affected in different degree by both the weather conditions and several plant and soil characteristics. In this study, stomatal resistance for ozone (R_{st}) was calculated from stomatal resistance for water vapour (R_s) obtained from measured water vapour flux according

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to Nemitz et al. (2009)."

(4) Deposition modelling

(a) New resistance parameterization has been used. We have described all model parameters in the revised manuscript and we have also analyzed how these parameters affected the deposition velocity. Section 3 (Ozone deposition modelling) has been rewritten.

(b) All used model parameters based on literature data. In this study, the values of model parameters have been obtained from Zhang et al. (2003) in almost all cases. However, the soil resistance has been parameterized with a simple linear function instead of a constant value, considering the effect of soil water availability.

(c) In several deposition models, the cuticle and soil resistances are constant, and not consider the difference between dry and wet surfaces. In the new model calculations, instead of former parameterization of R_{cut} and R_{inc} a more sophisticated parameterizations based on Zhang et al. (2003) have been used and discussed.

(5) Presentation

(a) The site description has been extended (Section 2.1): "Ozone flux measurements were performed during the Braunschweig GRAMINAE campaign from May 21 to June 15, 2000 over intensively managed grassland at the experimental fields of the Federal Agricultural Research Centre (Bundesforschungsanstalt für Landwirtschaft - FAL, Braunschweig Germany). The measuring site is located at latitude 52°18'N and longitude 10°26'E at 79 m above mean sea level surrounded by 12 ha arable and other managed grassland fields dominated by *Lolium perenne*, and has sandy soil. The available fetch was approximately 300 m to the west and east, 200 m to the south and 50 to 100 m to the north. The grass was cut at 06:00-10:00 UTC on 29 May, and was then lifted on 31 May. One week after cutting the field was fertilized at 06:00-07:00 on 05 June 2000. At the time of cutting, the grass was 0.75 m tall, with a single sided LAI

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of 3.06. Cutting reduced the vegetation to 0.07 m with a LAI of 0.14. At the end of the measurements, the canopy height was 0.34 m with a LAI of 1.5."

Model description has been shortened and rewritten. (Section 3).

(b) Section 4, 5 and 6 have been restructured and rewritten. As whole section has been rewritten according to the new results and considering the suggestions of the referees, this part is presented only in the revised manuscript.

Detailed comments:

-p.1072, l.10: The sentence has been corrected: "Ozone flux was measured"

-p.1072. l.21. This means the correlation coefficient, which is clarified in the text.

-p.1073. l. 1-2: During the whole measuring period this constant lag value has been used.

-p. 1073, l.20-22: Poor fetch was defined based on Nemitz et al. (2009). "During the GRAMINAE integrated field experiment several measuring platforms were settled close to each other at the main site. The exact position of each instrument mast in relation to the other masts, mobile laboratories and other obstructions to the fetch was determined and all flux data falling within obstructed sectors were removed from the individual dataset."

-p. 1073, Eq. 2 & p. 1074, Eq. 3: The minus sign is corrected.

-p. 1073, l. 9-10: High-pass filter has been used after McMillen (1988).

-p. 1073, l. 10-12: We have modified the expression two dimensional co-ordinate rotation to double rotation.

-p. 1073, l. 18: The sensible heat flux was corrected for humidity according to the paper by Schotanus et al.(1983).

p. 1075, l. 11: This part of the manuscript has been rewritten.

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p. 1076, Eq. 13, and p. 1076, l. 14: Another parameterization is used for cuticle resistance based on Zhang et al. (2003).

p. 1077, l. 1: We have corrected the sentence. "Cr is the concentration at the measuring height, and Cc is the concentration at the top of canopy, defined as a level, where the flux divides into stomatal (Fst) and non-stomatal (Fns) part"

p.1077, l. 11: Values are corrected. A new figure is also presented. Here, we have presented the soil water content in the upper 0.6 m soil layer.

p. 1077, l. 19: This sentence has been omitted.

p. 1077, l. 21-22: Negative sign is inserted.

p. 1078 (more comments): We agree with all comments. Errors have been corrected. This section (Section 4: Results and discussion) has been completely rewritten.

p. 1079, l. 2: Other papers refer: "Padro, 1996; Meyers et al., 1998; Zhang et al., 2002".

p. 1079, l. 9-11: This sentence has been omitted, and the whole section has been extended.

p. 1079, l. 20: As new model calculations have been performed, this part of manuscript has been rewritten.

p. 1080, l. 5: This sentence has been rewritten.

p. 1080, l. 12-13: Unit (cm s⁻¹) was added.

p. 1080, l. 20: We changed the word to "faster".

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