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Biogeosciences Discussions

Interactive comment on "Interactions among vegetation and ozone, water and nitrogen fluxes in a coastal Mediterranean maquis ecosystem" *by* G. Gerosa et al.

G. Gerosa et al.

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Answers to Referees

The detailed answers to the referee remarks are reported below. The authors are grateful to the referees for the careful and thorough revisions and the suggestions.

Answer to Referee #1, Anonymous

Specific comments

1454 line 3. Ok. 1454 line 20. Ok. 1454 line 22. Ok. 1455 line 6. Ok, thanks. 1455 line 13. Ok, thanks. 1457 line 5: Ok, the text was changed accordingly. 1459 line 1. Ok. 1459 line 2-8. Ok. The units have been added and the over bars corrected.

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1460 line 20. Ok, the clarification was added to the text. 1461 line 4. The referee is right. Thanks for the suggestion which has been accepted. An harmonization of the definition through the paper and the captions has been done. 1461 line 16/17. Ok, the value of the applied diffusivity ratio has been added. 1463 line 21: Ok, thanks. Pal Arya, 1988 has been added to the References. 1465 line 27. Ok, thanks. 1466 line 16. Ok, the referee is right. Instead of being an attempt of clarification, It resulted as an oversimplification; 1468 line 13. Ok, thanks. 1471 line 5. Ok, thanks. 1471 line 11. Ok, thanks. 1472 line 14. Ok, thanks. 1474 line 1. Ok, thanks. 1474 line 6. Ok, thanks. 1475 line 11. Ok, thanks. 1475 line 10/11. Ok, it was added. 1478 line 4/6. Ok, thanks. 1489 figure 7. Ok.

Answer to Referee #2, Nuria Altimir

General comments

This paper was developed as a part of a special issue about the VOCBAS-ACCENT campaign at Castelporziano. For this reason the different topics have been assigned to 8 papers in order to allow a more extensive dissertation and avoid repetitions: 1. General presentation of the site, of the ecosystem structure and the aims of the campaign (Fares et al.). 2. Presentation of microclimatic and micrometeorological features at the site (Cieslik et al.) 3. Water exchange dynamics of the maquis vegetation though sap flow measurements (Mereu et al.) 4. VOC concentration and fluxes at the site (Davidson) 5. Ozone concentrations and fluxes at the site (this paper) 6. Implication of night time transpiration on the ozone uptake by the maquis vegetation (Mereu et al.) 7. A process based model to estimate photosynthesis and gas exchange in the maquis ecosystem (Vitale et. Al.) 8. Critical overview of the results, interactions and possible follow up (Loreto et al.)

Comment #1 - The treatment of the relationship of the non-stomatal ozone deposition with air Humidity&;

The humidity regime have been extensively described in paper 1, 2, 3 and 6. Neverthe-

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less, following the referee request we modified the figure 9 adding the relative humidity registered at three heights directly at the measuring site. It is worth noticing that the meteo station of Tor Paterno cited in paper 1 and 3 is located 650 m away from the seashore line, in a different ecosystem (a clear of a Q. ilex and Pinus picea forest).

The referee argues that according to the humidity of the coastal climate of Castelporziano the canopy dryness criteria should be revised (she suggests RH$<$60-70%) because "water films would be present more or less all the time". Moreover the referee required more information about the wetness sensors employed. From Fig. 9 it is easy to realise that during the daylight hours the humidity was on-average below 60%. In our site two Campbell Sci. mod. 237 leaf wetness sensors (surrogate leaves) were employed to detect the presence of water condensation on the canopy leaves. The sensors are circuit boards (6 x 8 cm epoxy-fiberglass green coloured resin) with interlacing gold-plated fingers. Condensation on the sensor lowers the resistance between the fingers, which is measured by the data logger. Sensors were not coated with latex paint and were mounted horizontal to the soil at 1 m height, with the grids facing up, just on over two Holm oak bushes 3 m away from the measuring tower. The sensor were cleaned with ethylic alcohol before the installation. The sensors were new, just purchased from Campbell Sci. No a-priori wetness threshold was imposed. On the contrary, in order to enhance the sensor sensitivity and promptness, a (very high) dryness threshold of 6 MOhm was set. So all the conditions in which a resistance value was less than 6 MOhm were classified as wet canopy conditions (see e.g. fig 3 in paper #2, Cieslik et al.). It is worth noticing that the Campbell 237 manual indicate a 150KOhm as a wetness/dryness threshold. The relative units in figure 9 indicate the number of days in which surrogate leaves detected wet conditions (Resistance $<$ 6 MOhm) in each hour of the day, referred to the total number of days, i.e. the relative frequency. A value of 1 indicates that in a specific hour all the considered days the leaf wetness sensor recorded a wet condition. On the contrary a zero value indicates that at the chosen hour the sensor never detected a wet condition. In analysing the data we applied a very pragmatic approach. When the sensor indicated a dry condition (R$>$

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6 MOhm) the canopy was considered as dry, otherwise it was considered wet. Additionally, due to the shortness and the intensity of the joint campaign all the researchers attended the instrumentation, and it was hence easy to personally verify that during the day the canopies were completely dry from 8 o'clock in the morning and that they were only moderately wet at night..

Regarding the term "immediately" in line 18 page 1468 we agree in removing it, but the canopy really dries in around one hour (the sensor resistance increases from 200 KOhm at 7.30 to 6000 KOhm at 9.00).

Regarding the more general question about the relationship between ozone non-stomatal deposition and relative humidity we underline that we considered a very cautelative criterion of canopy dryness in order to assess the stomatal deposition with good confidence. All the remaining ozone deposition was attributed to not well identified non-stomatal processes. The nature of these latter have been investigated by searching for the relationship of non-stomatal deposition with turbulence, thermal decomposition, solar radiation, gas phase titration by NO, and atmospheric humidity. Only a significant relationship with humidity has been found. Nevertheless the nature of the interaction ozone-humidity is still not clear. From the linear relationship with the absolute humidity we can suppose that there is a chemical sink of ozone in the atmosphere due to the atmospheric water. From the hyperbolic relationship with the relative humidity when the canopy is wet, instead, we can suppose a role of water films formation on leaves as an ozone scavenger. For this reason both absolute humidity and relative humidity have been considered. Fig. 12d reports absolute humidity and fig 13a the relative humidity. But in this, as in other works, there is no direct proof that these processes are entirely responsible the non-stomatal ozone deposition observed at the site. Other processes are plausible. We were fascinated by the recent work of Read et al. (2008) which demonstrated a clear role of halogenated species in ozone photochemistry at coastal sites. Their finding over the features of ozone fluxes and NO2 fluxes well fitted with our observations and suggested us not to exclude this process from the possible ones and to think for the future of further measurements in this direction.

Comment #2 - Ecosystem structure: description and significance.

A new paragraph with a short site description has been added. Additionally, the authors of Fares et al. have been contacted and agreed in clearing some crucial points in the site description hence you can refer to their article for further details. . The referee stated that "...it would be possible to even construct quite a detailed spatial map of the density and composition of the vegetation in the plot"; A detailed spatial map of the site would require knowledge of the relative position of each individual which was not collected. Besides, the eddy tower (3.8 m height) footprint ranges between 20 and 140 m which is, most of the time, beyond the boundaries of the sampling area used for the structural analysis (a 33 x 33 m area around the measuring scaffold, i.e. about 15 m apart the tower in each cardinal direction).

"It is said in Fares et al that the plot is made out of patches of maquis and garigue communities. This would imply patches of higher vegetation including Quercus and patches of low-shrubs species" The term maquis for the site is not inappropriate. However the nomenclature for the maquis vegetation systems is quite controversial (garigue in Italy has a different meaning from the French use which are different from the Spanish use). We adopted a clearer classification suggested by Tomaselli (1981) which is also adopted now in Fares et al. 2009: by maquis it is intended a middle matorral with heights between 0.8 m and 2 m (the Italian macchia bassa), by garigue it is intended a low matorral or macchia bassa lower then 1-1.20 m.. Nevertheless the studied ecosystem is an unicum system. Instead of two different communities it is more correct to speak of two different succession stages of the maquis ecosystem kept in a dynamic equilibrium between them: low maquis and medium maquis (corresponding to low and middle matorral sensu Tomaselli 1981). The equilibrium is maintained by the adverse environmental conditions typical of coastal dunes (low nutrients, low water retention, sea aerosol, salt water infiltrations). The first succession stage is characterized by the prevalent abundance of R.officinalis, Cistus spl., E.multiflora and the presence of in-

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dividuals of Q. ilex, A. unedo and P. latifolia. The second one, instead, is dominated by A.unedo and Q.ilex and besides P. latifolia the other species are practically absent. Please, refer to the table in Fares et al. 2009 where the number of measurements for each species has been added. Geometrically speaking the ecosystem presented a quite well closed canopy (more then 90% of the coverage) and not isolated groups of vegetation as the term patches may lead to intend. In addition the two succession stages did not differ greatly in the mean vegetation height (low matorral 95 cm, middle matorral 136 m) and LAI (low matorral 2.29, middle matorral 2.42). The average height of the whole ecosystem was around 120 cm, with the occurrence of few Quercus ilex and Arbutus unedo individuals (less than 10% of the ground cover) which emerged over the mean ecosystem canopy of 50 cm on average. Thus, from a micrometeorological point of view, the exchanging surface was much more homogeneous (at least geometrically) than one could have imagined from the previous description of Fares et al. and, hence, the measurements are representative of this transitional ecosystem.

"From the point of view of a resistance analogue this site could seem to be closer to a dual-sink system. In general the adequacy of the one-layer big-leaf approach at this site is not discussed." See above. The aim of this work was to describe the ozone flux of a rear-dune ecosystem and to assess the bulk ozone uptake by the canopy, not to model it. The resistance analogue is used in a diagnostic way (inferentially) in order to derive the overall bulk stomatal conductance of the exchanging ecosystem. The bulk stomatal conductance of the canopy was derived by inverting the Penman-Monteith equation for evaporation which implies itself a one-layer big-leaf approach and an energy balance closure at the exchanging surface. This method allows to estimate the surface conductance of an evaporating surface by a unique measurement above the canopy without any other needs. The non-stomatal conductance was then derived as a residual, thus following the same one-layer big-leaf approach explicitly. No a-priori knowledge are required by this method. The dual-sink approach, on the contrary, requires that the stomatal conductance of one of the two sinks is known or derivable by another independent way (e.g. branch chambers measurements). But

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this is not our case: conductances are not known, and their assessment is the aim of the analysis. The dual sink approach can be used successfully, e.g., to partition fluxes in ecosystem with sparse vegetation with wide base soil areas. The value of the soil resistance can be assigned or simulated by other ways and then the bulk stomatal conductance of the vegetation can be derived by the measurements above the canopy, provided the relative coverage of vegetation and bare areas are supplied. A dual sink or a multi-layer approach is particularly appropriated in prognostic modelling where the aim is predicting the gaseous exchange of the ecosystem starting from the behaviour (conductances) of each ecosystem component and stratum. They are not indicated for diagnostic parameter assessments where it is necessary that a-priori assumptions are kept as less a possible. Moreover in our case only one-layer of vegetation was present. Therefore we chose to employ the simplest approach as possible in order to minimise all the a-priori assumptions and let the data as “crude” and cleaned as possible, thus allowing the readers to make eventually their own transformation of the data. Moreover this approach allows a direct comparison of results with other works, at least with our previously published ones.

"the title refers only to maquis (I suppose because garigue in this site is interpreted as a degradation of maquis) and without a better description leaves the reader to believe the site is somewhat homogeneous" The garigue is indeed intended ad a degradation of the maquis. Please refer to the answers above. We guess that the new explanations added to the Material and Methods session will allow the reader to better understand the type of ecosystem considered. The same for the relative homogeneity of the site.

Comment #3. Title and Introduction consistency

"The title is slightly misleading because water, O3 and NOx are not approached equally in the paper"; Of course ozone was the main topic. Nevertheless a lot of data and comments about water and NOx flux measurements have been reported in the paper. Water fluxes have been used to detect the canopy scale bulk stomatal conductance and appeared directly in figures 3, 4, new fig. 9, 11, 12, 13 and indirectly in fig. 1, 5,

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10. More or less water relations influence all the ozone exchange to the vegetation. NOx fluxes have been used to explore their possible implication in non-stomatal ozone deposition and appeared directly in figures 8 and 12. The order in which O3, H2O and NOx appears in the title respects their relative importance in the paper development.

"The introduction contents do not appear to reflect the analysis and measurements presented in the paper." We're sorry, but we do not agree with the referee, and the following comment of the referee which lists the topic faced in the introduction confirm our opinion.

"I do not find a detailed description of the potential injury relevant here (and any review of such should be accompanied with a mention to the experimental exposures etc)." The paper did not deal with ozone injury but only with ozone fluxes and the ozone metrics employed in ozone risk assessment. No symptoms detection were made at the site. Therefore we did not judge necessary to add such a paragraph.

"The introduction now does not mention several topics that appear prominently in the later analysis: NOx flux, comparison with the sap flow measurements, and non-stomatal flux" The structure of the Result and Discussion sessions are: Results Ozone concentration and fluxes Ozone exposure and dose Nitrogen oxide concentration and fluxes Discussion Stomatal uptake Non stomatal deposition Ozone risk assessment We do not feel that NOx flux and sap flow measurements were so prominent in the analysis. After a brief illustration of the NOx measurements in the result session, NOx were used in the discussion to test the hypothesis of their interaction with ozone non-stomatal deposition. Sap flow measurements were only used in the discussion to confirm that the morning ozone peak is not linked to plant transpiration. The non-stomatal deposition was left in the discussion session as a closure problem between the measured total ozone flux and stomatal flux, and thus faced in that session by testing different hypothesis on its nature. Following this line this topic has been developed entirely in the Discussion session.

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Some literature review paragraphs now placed in the discussion could be better placed in the intro, for example on the topic of the non-stomatal flux and why is it relevant to know it in the context of ozone risk assessment. See above. But we agreed with the referee in mentioning the non-stomatal flux topic and its relevance for ozone risk assessment into the introduction.

Comment #4. Placement

"I think the figures 10, 11 and 12 belong to the result session" The argument underlying the choice of placing these figures and the related non-stomata topic to the discussion session have been already explained above. The figure 11 is necessary at the augmentation exposed in the session 4.1

Other specific comments.

1455 line 20. The year 2000 have been chosen as a reference since all the papers published by the authors of the model on further developments and adjustments of the EMEP-DO3SE model have been referred to the same year/dataset.

1456 line 1. Ok,

1456 line 14. The calculation of AFstY from the observation in the field requires the setting up of non-routinely monitoring systems, such as micrometeorologica flux measurements, branch chambers and so on. On the contrary the evaluation of AOT40 requires only ambient air ozone concentration which are routinely monitored by the national or regional survey networks. Moreover the derivation of bulk stomatal flux from micrometeorological measurement requires the application of dry deposition inferential methods, data checking and gap-filling techniques which may not be completely automated. In this sense the direct determination of AFstY from field measurements is difficult while AOT40 benefits of an intrinsic simplicity.

1456 line 15. Ok

1456 line 17. It should be intended as above explained for line 14. Moreover stan-

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dardisation here means standard measurement height above canopy, standard vertical support (scaffold or mast), harmonised methodology for stomatal flux derivation from the total flux measurements, and so on...

1456 line 17. The meaning was, following how stated in the answer for the line 14, that it is not surprising that ozone risk assessment in Europe is done prevalently by model simulation rather than a network of ozone flux monitoring station.

1457 line 3. We changed model to models. In fact we wish to share the data with those scientists whose necessity is to calibrate or validate their models in Mediterranean conditions, and DO3SE is one of them.

1459 line 11. In Dutch the prefix van, which means "from", should be written in lower case.

1460 line 15. The denomination DDIM is used also by other authors. For example the paper of Cassandra Horii et al. (Agric. & For. Meteorol. 2005) uses a similar title. But in order to clarify we agree in changing the title to "Calculation of stomatal fluxes by a Dry Deposition Inferential Methodology";

1461 line 17. Ok we changed to "and by considering the relative diffusivity ratio of ozone in air to that of water vapour (Massman, 1998), set equal to 0.61 following the average T and P conditions at this site." Massman, W.J. (1998): A review of the molecular diffusivities of H2O, CO2, CH4, CO, O3, SO2, NH3, N2O, NO, and NO2 in air, O2 and N2 near STP. Atmospheric Environment 32, 1111-1127

1461 lines 23-26. The aim of the inferential method is to estimate the values of all the resistances of the simple deposition network (Ra, Rb, Rc, Rstom, Rnonstom) from the measured Rtot (=FO3/[O3]), H, LE and other micrometeorological parameters. In order to keep a numerical coherence, if the estimation of one resistance failed, then all the sample was discharged, even though the values of the other resistance seemed reasonable. An atmospheric (Ra) or laminar sublayer (Rb) resistance value greater than

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10000 s/m is very unrealistic. These Ra values may happen rarely when u* is very little and L approaches to zero, given the fact that this resistance is derived by numerically integrating the reciprocal of the vertical turbulent diffusivity ratio KH(z) which in turn depends on u* and the stability function (see e.g. Gerosa et al. 2003). The same happens to Rb when u* approaches zero. Rstom, on the contrary, increases rapidly over 10000 s/m when LE approaches 0 and it assumes values similar or greater than the cuticular resistance. In this latter case the stomata were assumed to be completely closed and a ceiling value of 10000 s/m was applied to the stomatal resistance In general the Penman-Monteith inversion is meaningless when LE is negative, and in other cases it fails when the measured humidity is greater than the saturation one. Finally, in some cases the estimation of Rc fails because the sum of Ra and Rb exceeds the measured total resistance to ozone (i.e. the resistance residual, Rc, is negative). We tried to reformulate the phrase in order to clarify this question.

1462 line 1. Ok, the title has been changed to "Data gap-filling"

1463 line 22. Ok, semi-hours was changed to half-hours

1463 line 24. The NOx analyser measures simultaneously NO, NO2 concentrations and gives out data on two different lines. The text has been rephrased to "where NOx indicates the NO concentrations when calculating the NO fluxes FNO and the NO2 concentrations when calculating the NO2 fluxes FNO2."

1464 line 4. The excluded data may not be fully appreciable from Figure 1 because, as written, they have been substituted with the gapfilled values indicated by a dashed line which, for short periods, may be confused with the continuous one. Moreover in the text has been written that nighttimes, i.e. between 8 p.m. and 8.30 a.m. - extreme included -, the sensors were often wet (with high frequency). On the contrary at 9.00 a.m. (the recording half-hour immediately following 8.30 a.m.) the frequency of wetness was around 20% and the canopy dryness conditions occurred in 4 cases over 5, thus the 80% of the times.

1464 line 11. A change in SWC reflects a change in water balance between the inputs (rain) and the outputs (water use, evapotranspiration). Both the inputs and the outputs depend on meteorology: the first one directly, the second ones indirectly because mediated by the tree physiology. In this sense we did not feel to exclude the meteorology at all. Moreover the two periods belong at two climatic periods (spring and summer) which in general show two different behaviours at the site. Nevertheless, for coherence we accept the suggestion to remove the word "meteorology" from the phrase.

1464 line 22-23. We did not treat the high nighttimes concentrations as exceptions but we included them in all the analysis. The phrase intention was to underline that on average the ozone concentrations showed a classical bell-shape course, but that there were also remarkable nighttimes high concentration values. This issue is a very interesting and should be better investigated in the future. The only information we have from this campaign is that high nighttimes ozone concentrations occurred when the typical breeze cycle was surmounted by a synoptic circulation characterised by winds blowing from the southern (S, SE, SW) and E quadrants during the night instead from the northern ones. In this case the air masses did not passed over the Rome city and thus ozone was only little depleted by NOx and VOC emissions (a feature evidenced also by Fares et al. in the same issue).

1465 line 10. Ok, "hereafter" removed.

1465 line 13-16. Since ozone fluxes are mediated by water availability (through stomata) we guess was important to show and comment the energy partition between heat dissipation and water evapotranspiration. This relationship between this partition and the stomatal ozone fluxes was then highlighted in the following lines 17 to 24.

1465 lines 25 onward. A short explanation have been added. The discussion about the discrepancy between the EC and Sap-flow measurement has been thoroughly discussed in the "Discussion session". Please, take a look at the session 4.1 which practically deals entirely with this issue and where all the question raised by the referee has been answered. The reason why this issue was treated in the discussion session instead of the result session has been already explained above.

1466 line 10. Ok, thanks.

1467 line 6. Ok, corrected to "around a mean value".

1468 lines 22-27. The hypothesis of an "hidden" stomatal aperture is presented and refuted by arguments and facts at page 1469 from lines 3 to 14. Is it speculation?

1471 line 5. Ok, thanks.

1471-1472. The halogenates reactions happen mainly in the air.

1472 lines 20-23. No. In non-costal sites obviously this process will not take place. But in coastal sites the ozone reactions with the halogenated species could significantly increase the ozone non-stomatal deposition summing up to those of water droplets in air and water films on surfaces.

1474 lines 9-10. Ok, thanks.

Figures

The usual time format is hh.mm. In any case we followed the referee request and changed the time format. The figure 6 has no y-axis unit because the values are adimensional (fraction Fstom/Ftot as indicated). Ok, we enlarged the axis labels and legend where possible. In any case they are related to the little dimension of the graphs imposed by the page layout and that some graphs should stay together. Nevertheless they are fully readable by zooming in the page a little bit.

Fig. 2. The standard deviations refer to the line at which they are applied to. E.g. the deviation bars applied over the Ftot line represent the standard deviation of the Ftot parameter, and so on. Of course to represent deviation bars for two overlapping lines is not easy. For this reason when Ftot was plotted the deviation bars referred to it, and when Ftot was not plotted the deviation bars refer to FtotDew. For a better clarifica-

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tion we changed the figure capture adding "Vertical bars are the standard deviations referred to Ftot when this line is plotted and to Ftotdew when Ftot is not plotted. Analogously vertical bars are refereed to Fstom when this line is plotted and to FstomDew in the few hours for which Fstom is not plotted." The deviation bar to the FstomDew lines have been added accordingly.

Fig. 5. The graphs show the average of the whole two periods indicated in the caption, following the denomination ("late spring" and "summer" periods) introduced in the text at page 1464 lines 12-13. Nevertheless we added the following clarification at the figure caption: "...in the late spring (a) and summer (b) periods. Graphs show averaged values of fluxes in each half an hour of the two distinct periods."

Figure 6. As for figure 2. We added the same clarification to the figure caption.

Figure 8 b. The figure b) deals with fluxes and not with concentrations. FNO and FNO2 were obtained by the gradient technique using the NOx concentrations measured at two levels, top and bottom (figure a), as explained at section 2.3 page 1463 lines 15 to 26 and page 1458 lines 1 to 5. Thus it is meaningless to distinguish between top and bottom fluxes. The values presented in fig. 8b are medians as indicated in the figure caption.

Figure 9. As already explained above as well as in the y-axis label and in the figure caption, the units are frequencies (number of half-hours where the canopy was wet with respect to the total number of hours), i.e. dimensionless values between 0 and 1. In this case the vertical bars do not exist because all the information is put in the graph, i.e. if a value of 0.8 is indicated, it means that in 8 cases over 10 the canopy was wet and in 2 cases was dry.

Figure 12 and 13. The referee is right, we missed the indication of the period used for the analysis. We remediate by inserting this clarification in the figure captions.

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