Replies to **reviewer 2** of the manuscript:

P. Pinho, M.R. Theobald, T. Dias, Y.S. Tang, C. Cruz, M.A. Martins-Loução, C. Máguas, M. Sutton and C. Branquinho. Critical loads of nitrogen deposition and critical levels of atmospheric ammonia for semi-natural Mediterranean evergreen woodlands.

8	*One criticism I have of this paper is that the critical loads and levels (CLL) selected are quite high compared to studies in North America and to the CL for Ammonia first published by Cape et al. 2009 that has become the standard for Europe.*	Please note the critical levels found in our paper are actually similar to the one pointed on literature by Cape et al (2009). In fact the data presented in this paper were used as a corroborative evidence for changing the critical levels in Europe from 8 to 1 $\mu g\ m^{-3}$, together with other studies from Italy and UK. The value pointed in literature (1 $\mu g\ m^{-3}$) was chosen making an educated average from multiple studies across Europe. In agreement with that please note that we present the critical level as being "lower than" 1.9 $\mu g\ m^{-3}$ due to lack of data for lower atmospheric ammonia concentration. Regarding the critical loads presented in our work they are in fact higher than those established for natural ecosystems. However they are within the upper range for the critical loads establish for semi-natural ecosystems (Bobbink et al., 2010). The reasons for this higher value, including the fact that we are dealing with semi-natural (not pristine) ecosystems, and some possible sources of overestimation of N-deposition (including the model assumptions and modelled background deposition) which are discussed along the manuscript and further explained in other replies in this text (see points 9, 10 and 24). To make all these points clearer, especially the special characteristics of semi-natural ecosystems (and the importance of considering them when calculating thresholds) have been greatly enhanced in the manuscript with several new sentences in the introduction and discussion. For this same reason we now state clearly in the title that Mediterranean evergreen woodlands are semi-natural ecosystems (for the reason that we can hardly find Mediterranean evergreen woodlands in Mediterranean Europe that we can be sure to be pristine or natural as these areas have been altered by man, by fire and grazing, for millennia).

| 9 | *The authors propose higher CLLs for rural agricultural areas ('semi-natural areas') compared to background areas. But background areas used in the study already have enhanced N deposition from nitrate and nitric acid. Therefore it appears that these new CLLs are built for an already anthropogenically influenced community that has a higher response threshold than pre-industrial, pre-agricultural (i.e. natural background conditions). This point needs to be clarified in the paper, i.e. that the response threshold used to select the CL is designed for a modern background level of N that already exceeds 10 kg ha yr, when historic natural background could be assumed (based on N deposition in remote areas in other parts of the planet) to be less than 1 kg ha-1 yr. Therefore these semi-natural area CLLs by default accept a certain level of degradation and are not the same as CLLs for natural background conditions.* | As explained (#8) we made clear in the text that we are reporting critical loads for semi-natural ecosystems (Mediterranean evergreen woodlands). Taking into consideration the humanization of the Mediterranean basin (Aronson, 1999), true background values would likely only be available from several centuries (or millennia) in the past.

In fact, most, if not all, the forested areas of the Mediterranean Basin are semi-natural ones. These area ecosystems have been shaped for millennia of low-intensity human activities, and are today a biodiversity hotspot. Conservation efforts actually aim at preserving the low-intensity management. We added some new bibliographic references and new sentences to reinforce this (Ellis & Ramankutty, 2008; Ellis, 2011; Bugalho et al., 2011). In fact, due to the prevalence of human-altered ecosystems over the planet is critical to find critical thresholds for these humanized ecosystems. Please note that truly natural conditions can hardly (if at all) be found in south and central Europe (Ellis & Ramankutty 2008) and is happening for several centuries at least in Europe and Asia, differently from America or Africa (Ellis et al., 2010). "From a philosophical point of view, nature is now human nature; there is no more wild nature to be found, just ecosystems in different states of human interaction, differing in wildness and humanness" (Cronon 1996 in Ellis 2011). Although we could debate if this should be assumed by nature conservation policies and discuss if we should restore this ecosystems to its natural state (quite debatable, even if at all possible) is not part of this type of paper. We do however reinforce with new sentences and references that we are dealing with semi-natural ecosystems and their importance as present biodiversity hotspots; and for that reason, state the importance of establishing critical thresholds, preventing the effects of agriculture intensification. |
| 10 | *A related point is that there is increasing evidence that lichens respond not only to ammonia but to other forms of deposition including nitrates, nitric acid, and ammonium. That is why the background site total N deposition is a concern. A new paper should be appearing soon in Ecological Applications based on a study in Southern California using passive samplers for multiple pollutants, provides good evidence that lichens are not only responding to ammonia but to other forms of N deposition in this Mediterranean ecosystem:*
1.   Sarah Jovan, Jennifer Riddell, Pamela Padgett, and Thomas H. Nash, III. 2012. Eutrophic lichens respond to multiple forms of N: implications for critical levels and critical loads research. Ecological Applications (In Press). | We agree that lichens may respond to several forms of nitrogen, but that was taken into consideration in the calculated N-deposition, by taking into consideration background deposition from modelled data. We point out that this could have originated an overestimation of the critical loads, due to the poor resolution of the EMEP model in our area (50km cell grid). For this reason our study area is located in the same grid cell as some industrial facilities, although those facilities are located at large distance from it (>30km) and against prevailing winds. Thus it is unlikely that our study site may experience the rather high values reported in the model. However this is the only source of information available and therefore we used it, but state in the text that the values are likely to be overestimated. Also note that for calculating thresholds with higher accuracy we chose to maintain climate and geologic characteristics, thus control and experimental site had to be located near to each other.

The approach of using modelled data to calculate critical loads is debatable. However, in most areas of south Europe there are very few monitoring stations capable of estimating with higher accuracy total N deposition. For mainland Portugal there are some stations monitoring NOx but only two measuring NH_4, and none measuring NH_3. Thus we chose for the best available option, using model data. |

11	*A final point needing clarification is the selection of the CLs from the data. It was not clear to me that the response thresholds selected (LDVoligo and LDVnitro values) were indeed the point at which the community began to suffer adverse effects, rather it seemed that the percentage of nitrophytes and oligotrophs were changing continuously with increased deposition along the entire study area and therefore it appeared to me that the cleanest site already exceeds response thresholds. So I would like to see a better justification for the response thresholds selected or lowering of the CLs.*	We agree with the reviewer's point that "the cleanest site already exceeds response thresholds". For this reason we report the critical loads and levels as being "lower than". However note that is only true for the oligotrophic functional group as for the nitrophytic one the response threshold was found to be within the measured values. The possible reasons for this are discussed in the text. However, for calculating thresholds we choose for the community that showed the first alterations, the oligotrophic one. Taking this community into consideration, and that it was found to be already changed; both the critical loads and levels are reported as being "lower than", rather than pointing to an absolute figure. Please note that we did not use percentages and rather absolute values for each functional groups; this way we could be sure that both were responding to the excessive N.
12	*Some new sources from North America are now available that suggest much lower CLs for lichens of Mediterranean ecosystems. I would like to see the authors discuss these results as part of the justification for higher CLLs selected in Portugal*	Agreed, this has been introduced in several new sentences in the introduction and discussion regarding the fact that we are reporting thresholds values for semi-natural ecosystems (see #8). Some other reasons for the relatively high values that were already reported in the text, namely the poor resolution of the deposition model, which could lead to an overestimation of the deposition values (see #10).
13	*One comment is that the lichen community measurements were made exclusively on cork oak yet the title suggests that the same results would be encountered in evergreen woodlands in general. Perhaps some statement in the discussion or introduction should explain the extrapolation.*	The extrapolation has been explained in the text. It is based on the fact that we are dealing with the same type of ecosystem, Mediterranean evergreen woodlands, classified in a single class on EUNIS (Davies et al., 2004). In fact, all areas occupied by this class share the same genera of tree (an evergreen *Quercus* spp.), similar type of management, both presently and in the past (manly low-intensity fire, grazing and agriculture), and also present the same type of climate (Mediterranean).
14	*(abstract) this particular section of the paper contains many more grammatical errors than the rest of the paper which need to be corrected for clarity.*	The abstract has been re-written for clarity and correction of grammatical errors.
15	*(intro) A discussion of the North American literature should be included either here or in the discussion or both.*	North American literature has been added, both in the introduction and discussion.
	(intro) I would suggest adding the scientific name of the study organism (cork oak) here.	It has been added.
16	*Personally, I feel that the traditional statement of hypotheses is a very useful way to structure a paper and is a core part of scientific thinking and methodology. So I wish that more authors these days would state an actual hypothesis in the introduction. In this case, it is not so critical, but it could be interesting to see how the paper would change if a hypothesis was stated here rather than an objective.*	We have added a hypothesis on the final part of the introduction. "We hypothesize that the response of lichen functional groups at increasing distances from agriculture N_{red} source could be used to calculate both CLOs and CLEs using modelled N_{dep} and measured $[NH_3]_{atm}$ in cork-oak (*Quercus suber* L.) woodland.".
17	*(methods) On page 11146 I wanted to see some mention of the current total N deposition in the study area relative to prehistoric and preindustrial levels to give me some context for the exposure levels in this study. Later I saw some mention of this in the discussion.*	Such values are not available for an historical (or "true background") perspective, as this area has been under low-intensity management for centuries or millennia (see 10#). Values on NOx and NH3 are only available for the last 30 years, from national authorities, but those values only exist for quite some time after the industrial revolution. Regarding those values we do report that the N-deposition values have remained constant over the last 20 years in our study area.

18	(methods) Also I wanted to know what data exist to suggest that the species composition and diversity is not also harmed/altered at the control site compared to historical natural background conditions.	There is no data suggesting that the control site species composition has been altered in the recent past (i.e. over the last two centuries). We can however unsure that the control site has not been managed for at least 30 years (it is within military grounds and thus management activities are controlled and well reported). But note that it is likely that all these areas have been a semi-natural ecosystem for centuries or millennia. Because we are dealing with critical thresholds for semi-natural ecosystems the "altered" species composition can be considered the true background conditions (see #10). The suggestion that reverting this woodland to pristine forest would be positive is actually conflicting with current biodiversity conservation efforts (see #9).
19	(methods) Finally, what were the statistical methods used to test that the difference between the control site LDV and barn sites LDVs were significant?	The method used was based on Cape et al. (2009) as reported in the manuscript. This method was accepted by the Convention on Long-range Transboundary Air Pollution. Based on the definition we chose the last unaltered point to set the critical loads and the first observed point to set the critical level. The statistical method was based on Cape et al. (2009). According to that, in statistical terms, "significant" means that the measurements exceed the background value, and has only a small probability (less than 5%) of being within the values considered as background. For this reason, rather than testing the significance of the difference between a single point and the background (which would only leave us with one degree of freedom to test), the confidence band of the regression (set at 0.05) was used to add the uncertainty of the relationship to the calculated threshold. The same paper also specifies that "In the past, many of the NH3 experiments consisted of just a treatment and a control. An analysis of variance was used to test the statistical significance of the effects. The more recent experiments,..., allow the evaluation by regression analysis". The same paper also clearly states that the main problem associated to this method, that if the lowest measured value is still above the true background concentration, one will overestimate the threshold. Taking into consideration our background values we do agree that this occurred, and as pointed out by the reviewer, our cleanest (measured) site was already altered. For this reason we reported both critical loads and levels as being "lower than".
20	Page 11150. See Geiser et al. 2010 (see general comments section), who found that climate, especially precipitation can influence lichen critical loads.	Agreed, but we worked (and extrapolated) under the same type of climate, with similar precipitation levels.
21	Because these data are based on one small area in Portugal, statistically speaking, the CLLs should really only apply to this area. What is the justification for extrapolating to Mediterranean ecosystems in general?	We add this justification to the text. It is partially explained on #13, based on the fact that we are dealing with the same type of ecosystem, sharing the same genera of tree (an evergreen *Quercus* spp.), similar type of management, and also present the same type of climate.
22	Table 1. Why were the dates included with the taxonomic authorities? Has there been some new rule change that requires inclusion of dates? Note misspelling of maximum.	Both issues were corrected in the table and table legend.
23	Figure 1. Consider including the location of the wind direction meter on the figure.	The wind station is located outside the map, in a military station; the map is a close-up of the areas near the barn and cannot include the wind station location.

24	*Figure 2. It is amazing that deposition of N reaches over 400 kg ha yr only 130 m from the cleanest site in the study area. Even though the annual deposition drops rapidly from the barn, it seems to me that there could be episodic levels at 130 m much exceeding annual average levels that could be affecting these communities. Similarly ammonia is 35 mg m3 at the barn, an extremely high level and drops to an average of 2-3 at the cleanest site, but perhaps there are episodes of much higher levels. I think this warrants discussion in the paper, i.e., how do you know that any of these sites are not affected by episodic bursts of atmospheric N as atmospheric conditions and daily concentrations fluctuate. This could explain why the proposed CLLs are so high.*	We agree that we could have burst of both N deposition and NH3 concentrations due to wind direction. This is taken into account by the N deposition model (that considered daily meteorology) and by the measures of atmospheric ammonia which were carried out over time and then averaged. However, because we aimed at calculating long-term critical thresholds data must be averaged over year periods. However we do not agree that this periodic burst could have lead to an overestimation of the critical loads and levels, they are included in the data. Moreover, the deposition model also allows us to calculate NH_3 concentration, taking into consideration the same meteorological data used for deposition. The modelled and measured NH3 concentration are in good agreement (this was not shown, given it was not the paper purpose), confirming us that the model was providing us an accurate enough estimation at least for NH_3 concentrations.
25	*Figure 3. It seems that if the regression line were extended (i.e. had there been sites in areas with lower deposition), that it would have resulted in selection of much lower CLs. Please justify the selection of sites used to establish the response threshold.*	We do point out that the values are "lower than" the given values; we have chosen not to extrapolate for lower values because we did not have enough data to point an related to that we cannot ensure that the linear relationship will hold. Thus we reported both critical loads and levels as "lower than". However we do agree with the reviewer, if extrapolated bellow the measured values we would get lower values (c. 1.39 kg ha^{-1}) and levels (nearly 0 μg m^{-3}). But note these values are not critical thresholds; they represent the possible values observable at the control site.