

**Nitrogen deposition  
and critical levels of  
atmospheric  
ammonia**

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

This discussion paper is/has been under review for the journal Biogeosciences (BG).  
Please refer to the corresponding final paper in BG if available.

# Critical loads of nitrogen deposition and critical levels of atmospheric ammonia for mediterranean evergreen woodlands

**P. Pinho<sup>1,2</sup>, M. R. Theobald<sup>3,4</sup>, T. Dias<sup>1</sup>, Y. S. Tang<sup>4</sup>, C. Cruz<sup>1</sup>,  
M. A. Martins-Loução<sup>1,5</sup>, C. Máguas<sup>1</sup>, M. Sutton<sup>4</sup>, and C. Branquinho<sup>1</sup>**

<sup>1</sup>Universidade de Lisboa, Faculdade de Ciências, Centre for Environmental Biology, Lisboa, Portugal

<sup>2</sup>Centre for Natural Resources and the Environment, Instituto Superior Técnico, Universidade Técnica de Lisboa, Portugal

<sup>3</sup>Dept. Agricultural Chemistry and Analysis, E.T.S.I. Agrónomos, Technical University of Madrid, Spain

<sup>4</sup>Centre for Ecology and Hydrology, Edinburgh Research Station, Edinburgh, UK

<sup>5</sup>Universidade de Lisboa, Museu de História Natural, Jardim Botânico, Lisboa, Portugal

Received: 11 September 2011 – Accepted: 30 October 2011 – Published: 21 November 2011

Correspondence to: C. Branquinho (cmbranquinho@fc.ul.pt)

Published by Copernicus Publications on behalf of the European Geosciences Union.

## Abstract

Nitrogen (N) has emerged in recent years as a key factor associated with global changes, with impacts on biodiversity, ecosystems functioning and human health. In order to ameliorate the effects of excessive N, safety thresholds have been established, such as critical loads (deposition fluxes) and levels (concentrations). For Mediterranean ecosystems, few studies have been carried out to assess these parameters. Our objective was therefore to determine the critical loads of N deposition and long-term critical levels of atmospheric ammonia for Mediterranean evergreen woodlands. For that we have considered changes in epiphytic lichen communities, which have been shown to be one of the most sensitive to excessive N. Based on a classification of lichen species according to their tolerance to N we grouped species into response functional groups, which we used as a tool to determine the critical loads and levels. This was done under Mediterranean climate, in evergreen cork-oak woodlands, by sampling lichen functional diversity and annual atmospheric ammonia concentrations and modelling N deposition downwind from a reduced N source (a cattle barn). By modelling the highly significant relationship between lichen functional groups and N deposition, the critical load was estimated to be below  $26 \text{ kg (N) ha}^{-1} \text{ yr}^{-1}$ , which is within the upper range established for other semi-natural ecosystems. By modelling the highly significant relationship of lichen functional groups with annual atmospheric ammonia concentration, the critical level was estimated to be below  $1.9 \mu\text{g m}^{-3}$ , in agreement with recent studies for other ecosystems. Taking into account the high sensitivity of lichen communities to excessive N, these values should be taken into account in policies that aim at protecting Mediterranean woodlands from the initial effects of excessive N.

## 1 Introduction

Fixation of atmospheric  $\text{N}_2$  to reactive nitrogen ( $\text{N}_{\text{reac}}$ ) allowed its use as fertilizer for agriculture, supporting an unprecedented increase in human population (Erisman

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



et al., 2008). This agriculture-related  $N_{\text{reac}}$  (mostly supplied as reduced nitrogen,  $\text{NH}_3/\text{NH}_4^+$ ,  $N_{\text{red}}$ ), together with that emitted from fossil fuel combustion (mostly oxidized nitrogen,  $\text{NO}_x$ ) was in 2007 at least equal to the amount of nitrogen (N) fixed by non-human processes (UNEP and WHRC, 2007). A large fraction of  $N_{\text{reac}}$  escapes to ecosystems, and as a consequence, the presence of excess N is increasingly detected across ecosystems (Erisman et al., 2008), causing alterations to biodiversity (Bobbink et al., 2010), ecosystem functioning (Manning et al., 2006) and also affects human health (Galloway et al., 2003). In fact, together with the rates of climate changes and biodiversity loss, the rate of human interference on the N cycle was considered to have passed the safety boundary for human wellbeing on Earth (Rockstrom et al., 2009).

In order to protect ecosystems from excessive  $N_{\text{reac}}$ , the concepts of critical loads (CLOs) of nitrogen deposition ( $N_{\text{dep}}$ ) and, more recently, of critical levels (CLEs) of atmospheric ammonia, have been defined. In this work the CLOs are a quantitative estimate of exposure to  $N_{\text{dep}}$  below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge (Nilsson and Grennfelt, 1988). The CLEs correspond to the concentration of atmospheric ammonia ( $[\text{NH}_3]_{\text{atm}}$ ) above which direct adverse effects may occur according to present knowledge (Cape et al., 2009). It is important to note that while the CLO corresponds to the total amount of N deposited from the air, the CLE refers only to the concentration of ammonia in the atmosphere. The CLOs for N deposition have been recently revised, with values ranging from  $3\text{--}30 \text{ kg ha}^{-1} \text{ yr}^{-1}$  (Bobbink et al., 2011) and the same occurred for long-term (annual) CLEs, for which a reduction from  $8 \mu\text{g m}^{-3}$  to  $1 \mu\text{g m}^{-3}$  was recommended to the United Nations Economic Commission for Europe, UNECE (Hallsworth et al., 2010; Pinho et al., 2009).

Although CLOs and CLEs are important in environmental policies, most of the values are based on work done on temperate ecosystems (Fenn et al., 2007; Rogers et al., 2009; Wolseley et al., 2006), but not on Mediterranean ones (Bobbink et al., 2011), with the exception of Mediterranean California (Fenn et al., 2010; Ochoa-Hueso et al., 2011). This currently prevents the establishment of empirical CLOs and CLEs for

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Mediterranean systems outside California. Only recently, the first values for empirical CLOs for Mediterranean maquis was published based on expert knowledge (Bobbink et al., 2011; Dias et al., 2011). Other studies done on the effect of excessive  $N_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  on European-Mediterranean ecosystems were not suitable for establishing CLOs and CLEs due to: (i) the short time periods of N addition to the ecosystem (less than 2 yr) (Bonanomi et al., 2006); (ii) the  $[\text{NH}_3]_{\text{atm}}$  measurements being less than 3 months (Frati et al., 2007; Pinho et al., 2009); and (iii) the lack of response of the tested parameters, e.g. N leakage (Roda et al., 2002). In order to avoid these limitations we have worked on an site with a continuous source of  $N_{\text{red}}$  (a cow barn for more than 20 yr), located in an region with a constant background  $N_{\text{dep}}$  for at least 20 yr (Simpson et al., 2003), and measured  $[\text{NH}_3]_{\text{atm}}$  for an entire year. Although N addition studies are the only way to ensure that the observed effect are due to N, these studies are not frequently done with enough time and realistic N doses (Bonanomi et al., 2006; Dias et al., 2011). Thus, N-addition studies can be complemented with gradient studies, where we can ensure a long-term N-supply and realistic N doses (Bobbink et al., 2011). We have focused on the effect of  $N_{\text{red}}$  because, unlike  $\text{NO}_x$ , the emissions of this N form have remained unchanged and are not expected to decrease significantly in the future, at least in Europe and United States (Reis et al., 2009).

The establishment of CLOs for  $N_{\text{dep}}$  uses several strategies, being most commonly based on empirical work, although a modelling approach has also been used (Fenn et al., 2008). The establishment of CLEs for ammonia has been mostly grounded on empirical work (Bobbink et al., 2011; Fenn et al., 2008), quantifying the effect of increasing  $N_{\text{dep}}$  or  $[\text{NH}_3]_{\text{atm}}$  on a receptor (Cape et al., 2009). For both CLOs and CLEs, lichen communities were used as one of the most sensitive receptors. Lichens have frequently been used to assess the effects of industrial pollution (Augusto et al., 2009; Branquinho et al., 2008; Giordani, 2007; Pinho et al., 2004) providing spatially explicit information on the effects of those pollutants (Pinho et al., 2008a, b). Specifically regarding the effects of  $N_{\text{red}}$ , a successful strategy was the use of lichen functional response groups (Lavorel and Garnier, 2002), i.e. groups of species with a similar

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



(mostly extensive pastured cork-oak woodlands). The study site was cork-oak woodland, Eunis class G2.1, "Mediterranean evergreen oak woodland" (Davies et al., 2004). At the upwind edge of this woodland, there was a high-intensity point-source of  $N_{red}$ , a barn permanently housing ca. 200 beef-cattle. More details can be found elsewhere (Pinho et al., 2011). Within the woodland we characterized: (i) lichens species frequency; (ii) nitrogen deposition; and (iii) atmospheric ammonia concentrations (Fig. 1).

For lichen sampling, a total of 74 points (trees) were selected (Fig. 1) ensuring that the trees had no branches at sampling height, no signs of disease and that the deviation from vertical was less than  $10^\circ$  (Pinho et al., 2004). Lichens were sampled in un-harvested cork accordingly to a standard protocol (Asta et al., 2002), using a  $50 \times 10$  cm grid, divided in five  $10 \times 10$  squares placed on the four main aspects of the trunk (N, E, S and W). All lichen species growing within the grid were identified and the number of squares in which each species was found was recorded as the species frequency. From these data we calculated a standard value, LDV (Lichen Diversity Value) that accounts for both species richness and their abundance (Asta et al., 2002). Because lichen functional groups based on tolerance to eutrophication were shown to respond more significantly to  $[NH_3]_{atm}$  than total diversity (Pinho et al., 2009, 2011), LDV was calculated by grouping species into two contrasting response functional groups (Lavorel and Garnier, 2002): nitrophytic or N-tolerant (LDVnitro) and oligotrophic or N-sensitive (LDVoligo) (Table 1). Species ranked as intermediate (mesotrophic) were not used because their relation to  $[NH_3]_{atm}$  was less significant (Pinho et al., 2011). Lichen species grouping was based on an international classification of Italian species accordingly to their eutrophication tolerance (Nimis and Martellos, 2008) after correcting for some species classification (Pinho et al., 2011).

The  $N_{dep}$  was estimated for the entire study site area (Fig. 1) using Local Atmospheric Dispersion and Deposition (LADD) model (Theobald et al., 2004). The use of transport and deposition models such as LADD to understand the fate of ammonia emitted from point sources is important due to the short-range deposition of most N allowing the establishment of local environmental policies (Dragosits et al., 2002). LADD

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







from possible NO<sub>x</sub> sources (more than 20 km). Taking this into account, N<sub>dep</sub> and [NH<sub>3</sub>]<sub>atm</sub> are assumed to be lower at the control area than at the study site and thus we considered the LDVs from the control area to be our control LDVs. The control area is similar to the study site, with the same type of vegetation (cork-oak woodland), soil, climate and background N<sub>dep</sub> as estimated by the EMEP Unified Model output (Simpson et al., 2003). The same methodology was followed for sampling lichens and calculating the LDVs of functional groups. In this control area, 16 sampling plots with complete absence of extensive cattle pasture were used and the mean values were considered as the control LDVs (Pinho et al., 2011).

## 2.2 Critical loads of nitrogen deposition and critical levels of atmospheric ammonia

For calculating CLOs (Nilsson and Grennfelt, 1988) and CLEs (Cape et al., 2009) we modelled the linear relationship between both the LDVs with N<sub>dep</sub> and [NH<sub>3</sub>]<sub>atm</sub> respectively. The control LDV was included in these plots and the first observed sampling point that presented a LDV different from the control was considered already changed. According to its definition, the last unaltered point was used to calculate the CLOs and the first altered point for calculating the CLEs. Taking these points and considering the 95% confidence interval of the linear relationship (Cape et al., 2009), the CLOs and CLEs were determined. In all regressions the residuals approached a normal distribution and presented an acceptable level of homogeneity, especially when taking into account the large number of samples used in the CLO regressions.

For calculating CLOs, the LDVs and N<sub>dep</sub> were correlated at the lichen sampling sites (N = 54, trees located outside the LADD model domain were not used, Fig. 1). Most lichen sampling sites were located in “tree” land-cover (Fig. 1). When that was not the case, which was due to the resolution of the land-cover model, the N<sub>dep</sub> for the nearest “tree” land-cover pixel was used, which was always less than 3 m from the point. This was done because LADD predicts much larger deposition rates to “trees” than to “grasslands” and lichens were observed in tree trunks.

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

[Title Page](#)

[Abstract](#)

[Introduction](#)

[Conclusions](#)

[References](#)

[Tables](#)

[Figures](#)

[⏪](#)

[⏩](#)

[◀](#)

[▶](#)

[Back](#)

[Close](#)

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)



For correlating LDVs with  $[\text{NH}_3]_{\text{atm}}$ , an interpolation of one of those variables was necessary, because they were not sampled in the same sites (Fig. 1). The LDVs were interpolated using ordinary kriging after analysis of the variograms (Pinho et al., 2011). Thus, the linear regressions used for calculating CLEs were based on the estimated LDVs and the observed  $[\text{NH}_3]_{\text{atm}}$  ( $N = 18$ , points located outside the lichen sampling sites spatial envelope were not used, Fig. 1). The decision to interpolate the LDVs and analyse the correlation with the measured  $[\text{NH}_3]_{\text{atm}}$ , rather than the other way round was in order not to over-inflate the degrees of freedom associated with the correlation and because the LDV interpolations had a higher level of confidence due to the much larger number of samples. Both LDVs (LDVnitro and LDVoligo) yielded a robust geostatistical model (Pinho et al., 2011) and thus a reliable set of interpolated values. All mapping outputs, geostatistics analysis and spatial operations were done with ArcMap v.9.3 (ESRI), correlations and function fitting were performed with Statistica v.10.0 (Statsoft).

### 3 Results and discussion

#### 3.1 Patterns of nitrogen deposition and atmospheric ammonia

Both  $N_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  displayed similar spatial patterns, decreasing exponentially with distance from the cow-barn (Fig. 2), which is in accordance to what has been observed in other studies with  $N_{\text{red}}$  point sources (Fowler et al., 1998; Sutton et al., 1998; Verhagen and van Diggelen, 2006). The distance of impact of the studied  $N_{\text{red}}$  source was found to be ca. 50 m for CLOs and ca. 90 m for CLEs, which corresponds to the distance above which we could find  $N_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  values lower than the critical limits established, respectively (see Sect. 3.2, 1st paragraph). The results confirm the short-range dispersion of  $N_{\text{red}}$  emitted from a point source found in other works (Sutton et al., 1998), but it may not be the same in other situations since it is dependent on the surrounding land-cover (e.g. trees vs. grass proportion) and on the number of animals in the barn (Adrizal et al., 2008; Verhagen and van Diggelen, 2006).



2010). Based on the changes of LDVoligo the long-term CLE for Mediterranean evergreen woodlands was found to be lower than  $1.9 \mu\text{g m}^{-3}$  because again, the highest measured LDVoligo was below the control value (Fig. 4). This CLE value is in agreement with recent studies pointing to the revision of CLEs of  $[\text{NH}_3]_{\text{atm}}$  from  $8 \mu\text{g m}^{-3}$  to  $1 \mu\text{g m}^{-3}$  (Cape et al., 2009).

There is a general lack of studies regarding the effects of excessive  $\text{N}_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  on Mediterranean ecosystems besides California (Bobbink et al., 2011; Ochoa-Hueso et al., 2011). Furthermore, the few studies on the effects of increased N availability on the diversity of vascular plants within the European-Mediterranean ecosystems seem to be contradictory. An increase in total diversity was reported on maquis with one year addition of 40 and  $80 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (Dias et al., 2011) and in Spanish mountain heathlands located near Mediterranean climate areas with  $\text{N}_{\text{dep}}$  of  $56 \text{ kg ha}^{-1} \text{ yr}^{-1}$  (Calvo et al., 2005). Other studies reported no change in total species richness as the work performed in an Italian grassland with N addition of  $35 \text{ kg ha}^{-1} \text{ yr}^{-1}$  (Bonanomi et al., 2006). However, the effects of  $\text{N}_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  are much clear when measured with functional groups instead of total diversity (Fenn et al., 2008), which is in accordance to what was observed in this work. The relation between LDVs and  $\text{N}_{\text{dep}}$  and LDVs and  $[\text{NH}_3]_{\text{atm}}$  was highly significant ( $P < 0.0001$ ), both  $\text{N}_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  presenting the same type of relationship to lichen functional groups: higher  $\text{N}_{\text{dep}}$  and  $[\text{NH}_3]_{\text{atm}}$  led to a decrease of oligotrophic species (LDVoligo) and to an increase of nitrophytic ones (LDVnitro) (Figs. 3, 4). This has been observed in several studies in temperate (Fenn et al., 2007; Rogers et al., 2009; Wolseley et al., 2006) and Mediterranean ecosystems (Frati et al., 2007). However, unlike these previous works done in Mediterranean ecosystems, we monitored  $[\text{NH}_3]_{\text{atm}}$  for a full year, allowing us to fully account for Mediterranean climate seasonality and variability in  $[\text{NH}_3]_{\text{atm}}$ . Moreover, other environmental factors that could influence lichens, such as industrial pollutants or regional climate (Giordani and Incerti, 2008; Pinho et al., 2008a), dust (Loppi and Pirintzos, 2000; Pinho et al., 2008b) or land-use intensity (Pinho et al., 2008; Stofer et al., 2006) were constant throughout our study site. Thus we were able

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

to provide more accurate estimates of CLEs and CLOs for the first time for European Mediterranean ecosystems.

The CLOs found in this work are comparable to those, found in other semi-natural ecosystems (not Mediterranean), also calculated based on lichen diversity changes (Bobbink et al., 2010). However, the CLOs found for natural or unmanaged ecosystems, also based in lichen diversity changes, such as California forest (Fenn et al., 2008) and European tundra and mountain summits (Bobbink et al., 2011) are much lower ( $3\text{--}10\text{ kg ha}^{-1}\text{ yr}^{-1}$ ) than the values found in this work. This difference can be due to the higher availability of N in semi-natural ecosystems than in natural ones. As a consequence in semi-natural ecosystems the more sensitive species, e.g. cyanolichens (Aragon et al., 2010), could already have been eliminated and thus the remaining community is more tolerant to small increases in N.

It is interesting to note that the CLOs available in literature (Bobbink et al., 2011) appear to be dependent on the management intensity, but independent of climate, with similar CLOs found for different climates under the same management intensity. The CLEs available, although not including most ecosystems, (Cape et al., 2009) indicate that they are independent of both climate and management intensity. This independence of climate indicates that N may have the same type of impact on lichens communities worldwide, suggesting that lichens could be used as universal indicators for the impact of excessive N.

By measuring  $[\text{NH}_3]_{\text{atm}}$  directly, we greatly reduced the uncertainty of the CLEs. However, for CLOs we could not measure all components of  $N_{\text{dep}}$ . The best estimates of  $N_{\text{dep}}$  was therefore derived as the sum of (1) modelled N deposition from the LADD model for the study site, and (2) background  $N_{\text{dep}}$ , estimated using the EMEP Unified Model (Simpson et al., 2003). This background estimate increased the uncertainty of the CLOs, because the real background  $N_{\text{dep}}$  could be lower, due to the poor resolution of the EMEP model for Portugal (50 km). The study site is located in a grid cell which contains large industrial and urban areas, but is in fact located 10 km from the nearest small town (Alcochete), and 20 km from the larger cities (Lisboa, Setúbal and

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Montijo) and from the nearest industrial area (Barreiro). Therefore, the background  $N_{\text{dep}}$  due to  $\text{NO}_x$  ( $5.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) will be most probably overestimated for the study site, which in turn leads to an overestimation of CLOs. Thus it is of critical importance to produce maps of  $N_{\text{dep}}$  with a higher spatial resolution. Another aspect that may have originated an overestimation of the CLOs was the use of fixed deposition velocities for the LADD model. It has been suggested that N deposition velocity is dependent on  $[\text{NH}_3]_{\text{atm}}$  (Flechard and Fowler, 1998), and thus the  $N_{\text{dep}}$  could be smaller than those we calculated.

### 3.3 Sensitivity of nitrophytic and oligotrophic lichens

Although both LDV<sub>oligo</sub> and LDV<sub>nitro</sub> presented equally significant correlations to either  $N_{\text{dep}}$  or  $[\text{NH}_3]_{\text{atm}}$ , which was expected (Ruisi et al., 2005; Sparrius, 2007; Vilsholm et al., 2009), the CLOs and CLEs were always lower when considering LDV<sub>oligo</sub> (Fig. 3, 4). The highest LDV<sub>oligo</sub> observed in our study site was below the regional control value, whereas for LDV<sub>nitro</sub> a large number of sampling points were still within the range of the regional control value (Figs. 3, 4). This suggests that the oligotrophic functional group is more sensitive to increasing N availability than the nitrophytic one and/or that the nitrophytic functional group may require higher  $[\text{NH}_3]_{\text{atm}}$  or  $N_{\text{dep}}$  driven by  $N_{\text{red}}$  to respond (Pinho et al., 2011). The differences between the response of nitrophytic and oligotrophic species to  $N_{\text{dep}}$  or  $[\text{NH}_3]_{\text{atm}}$  could be related to the capacity of nitrophytic species to prevent the accumulation of N in the cells, (Gaio-Oliveira et al., 2005) which can be due to the capacity to provide sufficient carbon for N metabolism (Hauck, 2010). However, the mechanism by which excessive N influences lichens is currently under debate. An indirect effect of atmospheric ammonia on tree bark pH (van Herk, 2001) has been suggested; an effect that can be confounded/or correlated with the influence of dust, especially relevant in Mediterranean areas (Branquinho et al., 2008; Loppi and Pirintsos, 2000). Nevertheless it remains inconclusive if the change in bark pH is merely a consequence of pollution to which lichens are directly responding (Spier et al., 2010). Another possible mechanism includes direct toxicity of excessive N

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



on oligotrophic species, for example on the performance of photosystem II (Paoli et al., 2010). However, independently of the actual mechanism, the different sensitivity to N of the oligotrophic and nitrophytic functional groups points to the preferential use of LD-Voligo as the most sensitive tool to monitor the effects of low  $N_{\text{dep}}$  (driven by  $N_{\text{red}}$ ) and  $[\text{NH}_3]_{\text{atm}}$ . This is critical in establishing thresholds in highly disturbed areas, where only the most tolerant species remain (Fрати et al., 2007): in these areas if a control value is not taken into account the CLOs and CLEs calculated can be largely overestimated.

## 4 Conclusions

The more important outcome of this work was that, by modelling the highly significant relationship between LDVs and  $N_{\text{dep}}$  and LDVs and  $[\text{NH}_3]_{\text{atm}}$  we could establish critical thresholds for N, for the first time for European Mediterranean evergreen woodlands. The critical loads for N deposition were considered to be below  $26 \text{ kg ha}^{-1} \text{ yr}^{-1}$  and the long-term critical levels of atmospheric ammonia were found to be below  $1.9 \mu\text{g m}^{-3}$ . The critical levels found were similar to the concentrations established for other ecosystems. The critical loads found were within the upper range of the critical loads for other semi-natural ecosystems, but higher than for natural ones.

*Acknowledgements.* We thank funding from FCT-MCTES (BD/17880/2004, BPD/75425/2010 and research projects PTDC/AAC-CLI/104913/2008 and PTDC/BIA-BEC/099323/2008) and from COST729. We also thank António Mineiro for allowing the use of his farm and assistance during the fieldwork and Força Aérea Portuguesa (BA6) for providing the meteorological data.

## References

Adrizal, A., Patterson, P. H., Hulet, R. M., Bates, R. M., Myers, C. A. B., Martin, G., Shockey, R. L., Van der Grinten, M., Anderson, D. A., and Thompson, J. R.: Vegetative Buffers for Fan Emissions from Poultry Farms: 2. Ammonia, Dust and Foliar Nitrogen, J. Environ. Sci. Health Part B, 43, 96–103, 2008.

**BGD**

8, 11139–11163, 2011

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- Davies, E. D., Moss, D., and Hill, M. O.: Eunis Habitat Classification Revised, European Environment Agency, European topic centre on nature protection and biodiversity, 2004.
- Dias, T., Malveiro, S., Martins-Loucao, M. A., Sheppard, L. J., and Cruz, C.: Linking N-Driven Biodiversity Changes with Soil N Availability in a Mediterranean Ecosystem, *Plant Soil*, 341, 125–136, 2011.
- 5 Dragosits, U., Theobald, M. R., Place, C. J., Lord, E., Webb, J., Hill, J., ApSimon, H. M., and Sutton, M. A.: Ammonia Emission, Deposition and Impact Assessment at the Field Scale: A Case Study of Sub-Grid Spatial Variability, *Environ. Pollut.*, 117, 147–158, 2002.
- EEA, European Environment Agency: EmeP/Corinair Emission Inventory Guidebook, 2007.
- 10 Erisman, J. W., Sutton, M. A., Galloway, J., Klimont, Z., and Winiwarter, W.: How a Century of Ammonia Synthesis Changed the World, *Nat. Geosci.*, 1, 636–639, 2008.
- Fenn, M. E., Geiser, L., Bachman, R., Blubaugh, T. J., and Bytnerowicz, A.: Atmospheric Deposition Inputs and Effects on Lichen Chemistry and Indicator Species in the Columbia River Gorge, USA, *Environ. Pollut.*, 146, 77–91, 2007.
- 15 Fenn, M. E., Jovan, S., Yuan, F., Geiser, L., Meixner, T., and Gimeno, B. S.: Empirical and Simulated Critical Loads for Nitrogen Deposition in California Mixed Conifer Forests, *Environ. Pollut.*, 155, 492–511, 2008.
- Fenn, M. E., Allen, E. B., Weiss, S. B., Jovan, S., Geiser, L. H., Tonnesen, G. S., Johnson, R. F., Rao, L. E., Gimeno, B. S., Yuan, F., Meixner, T., and Bytnerowicz, A.: Nitrogen Critical Loads and Management Alternatives for N-Impacted Ecosystems in California, *J. Environ. Manage.*, 91, 2404–2423, 2010.
- 20 Flechard, C. R. and Fowler, D.: Atmospheric ammonia at a moorland site. II: Long-term surface-atmosphere micrometeorological flux measurements, *Q. J. Roy. Meteor. Soc.*, 124, 759–791, 1998.
- 25 Fowler, D., Pitcairn, C. E. R., Sutton, M. A., Flechard, C., Loubet, B., Coyle, M., and Munro, R. C.: The Mass Budget of Atmospheric Ammonia in Woodland within 1 Km of Livestock Buildings, *Environ. Pollut.*, 102, 343–348, 1998.
- Frati, L., Santoni, S., Nicolardi, V., Gaggi, C., Brunialti, G., Guttova, A., Gaudino, S., Pati, A., Pirintsos, S. A., and Loppi, S.: Lichen Biomonitoring of Ammonia Emission and Nitrogen Deposition around a Pig Stockfarm, *Environ. Pollut.*, 146, 311–316, 2007.
- 30 Gaio-Oliveira, G., Dahlman, L., Palmqvist, K., Martins-Loucao, M., and Maguas, C.: Nitrogen Uptake in Relation to Excess Supply and Its Effects on the Lichens *Evernia Prunastri* (L.) Ach and *Xanthoria Parietina* (L.) Th. Fr, *Planta*, 220, 794–803, 2005.

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Galloway, J. N., Aber, J. D., Erisman, J. W., Seitzinger, S. P., Howarth, R. W., Cowling, E. B., and Cosby, B. J.: The Nitrogen Cascade, *Bioscience*, 53, 341–356, 2003.
- Giordani, P.: Is the Diversity of Epiphytic Lichens a Reliable Indicator of Air Pollution? A Case Study from Italy, *Environ. Pollut.*, 146, 317–323, 2007.
- 5 Giordani, P. and Incerti, G.: The Influence of Climate on the Distribution of Lichens: A Case Study in a Borderline Area (Liguria, Nw Italy), *Plant Ecol.*, 195, 257–272, 2008.
- Hallsworth, S., Dore, A. J., Bealey, W. I., Dragosits, U., Vieno, M., Hellsten, S., Tang, Y. S., and Sutton, M. A.: The Role of Indicator Choice in Quantifying the Threat of Atmospheric Ammonia to the “Natura 2000” Network, *Environ. Sci. Policy*, 13, 671–687, 2010.
- 10 Hao, X. Y., Benke, M. B., Chang, C., Janzen, H. H., Clayton, G. W., and Hill, B. R.: Spatial Pattern of Ammonia Sorption by Soil and Vegetation Downwind of a Beef Feedlot, *Agr. Ecosyst. Environ.*, 132, 39–47, 2009.
- Hauck, M.: Ammonium and Nitrate Tolerance in Lichens, *Environ. Pollut.*, 158, 1127–1133, 2010.
- 15 Lavorel, S. and Garnier, E.: Predicting Changes in Community Composition and Ecosystem Functioning from Plant Traits: Revisiting the Holy Grail, *Funct. Ecol.*, 16, 545–556, 2002.
- Loppi, S. and Pirintsos, S. A.: Effect of Dust on Epiphytic Lichen Vegetation in the Mediterranean Area (Italy and Greece), *Isr. J. Plant Sci.*, 48, 91–95, 2000.
- Manning, P., Newington, J. E., Robson, H. R., Saunders, M., Eggers, T., Bradford, M. A., Bardgett, R. D., Bonkowski, M., Ellis, R. J., Gange, A. C., Grayston, S. J., Kandeler, E., Marhan, S., Reid, E., Tschерko, D., Godfray, H. C. J., and Rees, M.: Decoupling the Direct and Indirect Effects of Nitrogen Deposition on Ecosystem Function, *Ecol. Lett.*, 9, 1015–1024, 2006.
- 20 McCulloch, R. B., Few, G. S., Murray, G. C., and Aneja, V. P.: Analysis of Ammonia, Ammonium Aerosols and Acid Gases in the Atmosphere at a Commercial Hog Farm in Eastern North Carolina, USA, *Environ. Pollut.*, 102, 263–268, 1998.
- 25 Nilsson, J. and Grennfelt, P.: Critical Loads for Sulphur and Nitrogen, in: Report from a Workshop Held at Skokloster, Nilsson, J. and Grennfelt, P., Sweden, Nordic Council of Ministers, 225–268, 1988.
- 30 Nimis, P. L. and Martellos, S.: Italic – the Information System on Italian Lichens V. 4.0, University of Trieste, Dept. of Biology, IN4.0/1, available at: <http://dbiodbs.univ.trieste.it>, 2008.
- Ochoa-Hueso, R., Allen, E. B., Branquinho, C., Cruz, C., Dias, T., Fenn, M. A., Manrique, E., Pérez-Corona, E., Sheppard, L. J., and Stock, W. D.: Nitrogen Deposition Effects on



## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Scheffer, M., Folke, C., Schellnhuber, H. J., Nykvist, B., de Wit, C. A., Hughes, T., van der Leeuw, S., Rodhe, H., Sornlin, S., Snyder, P. K., Costanza, R., Svedin, U., Falkenmark, M., Karlberg, L., Corell, R. W., Fabry, V. J., Hansen, J., Walker, B., Liverman, D., Richardson, K., Crutzen, P., and Foley, J. A.: A Safe Operating Space for Humanity, *Nature*, 461, 472–475, 2009.

Roda, F., Avila, A., and Rodrigo, A.: Nitrogen Deposition in Mediterranean Forests, *Environ. Pollut.*, 118, 205–213, 2002.

Rogers, P. C., Moore, K. D., and Ryel, R. J.: Aspen Succession and Nitrogen Loading: A Case for Epiphytic Lichens as Bioindicators in the Rocky Mountains, USA, *J. Veg. Sci.*, 20, 498–510, 2009.

Ruisi, S., Zucconi, L., Fornasier, F., Paoli, L., Frati, L., and Loppi, S.: Mapping Environmental Effects of Agriculture with Epiphytic Lichens, *Isr. J. Plant Sci.*, 53, 115–124, 2005.

Scheffer, M., Bascompte, J., Brock, W. A., Brovkin, V., Carpenter, S. R., Dakos, V., Held, H., van Nes, E. H., Rietkerk, M., and Sugihara, G.: Early-Warning Signals for Critical Transitions, *Nature*, 461, 53–59, 2009.

Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J. P.: The Emep Unified Eulerian Model: model Description, Emep Msc-W Report 1/2003; The Norwegian Meteorological Institute, 2003.

Sparrius, L. B.: Response of Epiphytic Lichen Communities to Decreasing Ammonia Air Concentrations in a Moderately Polluted Area of the Netherlands, *Environ. Pollut.*, 146, 375–379, 2007.

Spier, L., van Dobben, H., and van Dort, K.: Is Bark Ph More Important Than Tree Species in Determining the Composition of Nitrophytic or Acidophytic Lichen Floras?, *Environ. Pollut.*, 158, 3607–3611, 2010.

Stofer, S., Bergamini, A., Aragon, G., Carvalho, P., Coppins, B. J., Davey, S., Dietrich, M., Farkas, E., Karkkainen, K., Keller, C., Lokos, L., Lommi, S., Maguas, C., Mitchell, R., Pinho, P., Rico, V. J., Truscott, A. M., Wolseley, P. A., Watt, A., and Scheidegger, C.: Species Richness of Lichen Functional Groups in Relation to Land Use Intensity, *Lichenologist*, 38, 331–353, 2006.

Sutton, M. A., Milford, C., Dragosits, U., Place, C. J., Singles, R. J., Smith, R. I., Pitcairn, C. E. R., Fowler, D., Hill, J., ApSimon, H. M., Ross, C., Hill, R., Jarvis, S. C., Pain, B. F., Phillips, V. C., Harrison, R., Moss, D., Webb, J., Espenhahn, S. E., Lee, D. S., Hornung, M., Ulyyett, J., Bull, K. R., Emmett, B. A., Lowe, J. and Wyers, G. P.: Dispersion, Deposition and Impacts

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



of Atmospheric Ammonia: Quantifying Local Budgets and Spatial Variability, Environ. Pollut., 102, 349–361, 1998.

Sutton, M. A., Miners, B., Tang, Y. S., Milford, C., Wyers, G. P., Duyzer, J. H., and Fowler, D.: Comparison of Low Cost Measurement Techniques for Long-Term Monitoring of Atmospheric Ammonia, J. Environ. Monitor., 3, 446–453, 2001.

Tang, Y. S., Cape, J. N., and Sutton, M. A.: Development and Types of Passive Samplers for NH<sub>3</sub> and NO<sub>x</sub>, The Scientific World, 1, 513–529, 2001.

Theobald, M. R., Dragosits, U., Place, C. J., Smith, J. U., Sozanska, M., Brown, L., Scholefield, D., Del prado, A., Webb, J., Whitehead, P. G., Angus, A., Hodge, I. D., Fowler, D., and Sutton, M. A.: Modelling Nitrogen Fluxes at the Landscape Scale, Water Air Soil Pollut. Focus, 4, 135–142, 2004.

UNEP, United Nations Environment Programme and WHRC, Woods Hole Research Center: Reactive Nitrogen in the Environment: Too Much or Too Little of a Good Thing, United Nations, 2007.

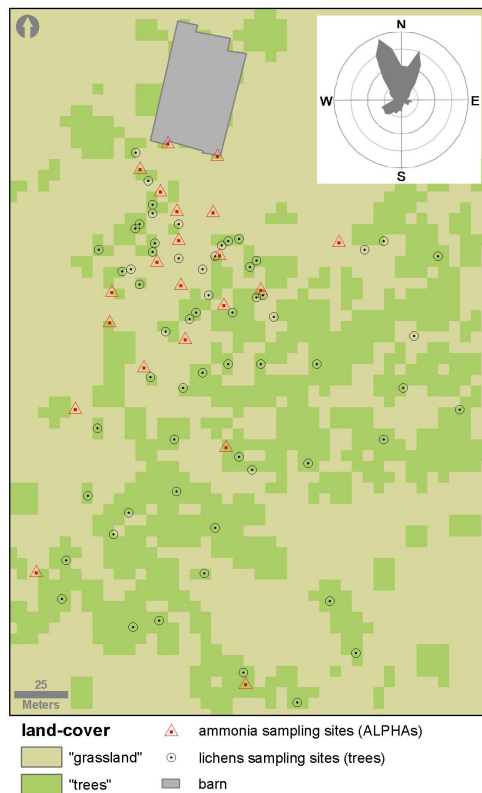
van Herk, C. M.: Bark Ph and Susceptibility to Toxic Air Pollutants as Independent Causes of Changes in Epiphytic Lichen Composition in Space and Time, Lichenologist, 33, 419–441, 2001.

Verhagen, R. and van Diggelen, R.: Spatial Variation in Atmospheric Nitrogen Deposition on Low Canopy Vegetation, Environ. Pollut., 144, 826–832, 2006.

Vilsholm, R. L., Wolseley, P. A., Sochting, U., and Chimonides, P. J.: Biomonitoring with Lichens on Twigs, Lichenologist, 41, 189–202, 2009.

Wolseley, P. A., James, P. W., Theobald, M. R., and Sutton, M. A.: Detecting Changes in Epiphytic Lichen Communities at Sites Affected by Atmospheric Ammonia from Agricultural Sources, Lichenologist, 38, 161–176, 2006.





**Fig. 1.** Sampling design and wind direction. In cork-oak woodland we assessed lichen functional diversity, measured atmospheric ammonia concentration over one year period, and modelled N deposition using LADD model considering the land-cover ("trees" and "grassland"), the wind direction and considering the barn as the source of N. The wind direction is the % of wind occurrence from each direction in 10° interval, during the sampling period. The full circle was 10%.

**Nitrogen deposition and critical levels of atmospheric ammonia**

P. Pinho et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

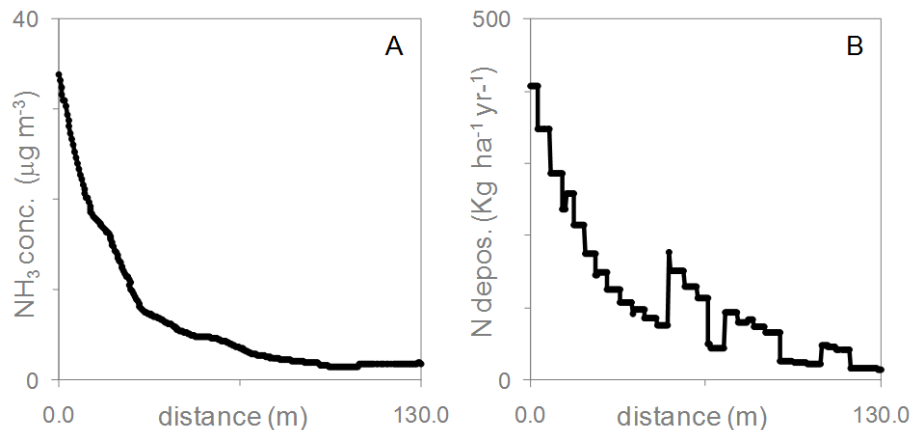
Printer-friendly Version

Interactive Discussion



## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.



**Fig. 2.** Atmospheric ammonia concentration (**A**) and N deposition (**B**) along a transect from the N source (the cow barn, at distance 0) and up to 130 m distance, in the prevailing wind direction, SE. Atmospheric ammonia concentrations were calculated from a geostatistical model of measured ammonia concentrations for one year period, N deposition was calculated from LADD model. The elevated steps in the N deposition data are due to the differences between the deposition rates to trees and grassland.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

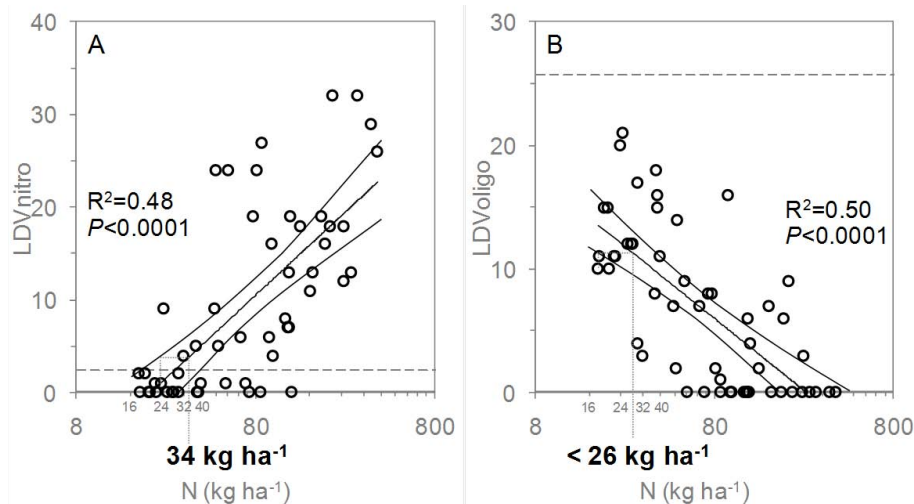
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.



**Fig. 3.** Critical loads of N deposition considering the nitrophytic (A) and oligotrophic (B) functional groups. N deposition was calculated from LADD model and correlated to changes of LDV<sub>nitro</sub> (nitrophytic) and LDV<sub>oligo</sub> (oligotrophic). Considering the confidence intervals of the regression line (95%) and the last point with a LDV still within the control values (dotted lines) the critical load was established. Because the observed values of LDV<sub>oligo</sub> are below the control value, a critical load lower than  $26 \text{ kg ha}^{-1} \text{ yr}^{-1}$  should be considered.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

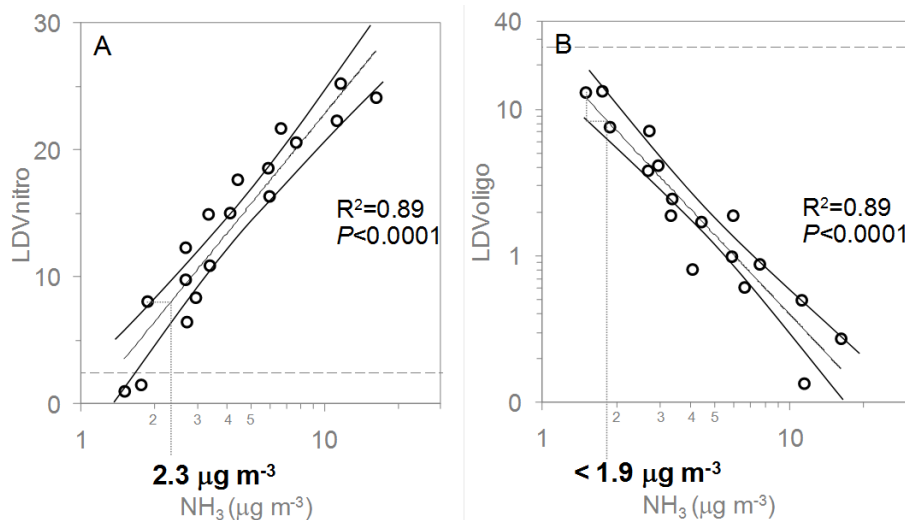
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Nitrogen deposition and critical levels of atmospheric ammonia

P. Pinho et al.



**Fig. 4.** Critical levels of atmospheric ammonia considering the nitrophytic **(A)** and oligotrophic **(B)** functional groups. The  $[\text{NH}_3]_{\text{atm}}$  were measured for one year and correlated to changes of LDVnitro (nitrophytic) and LDVoligo (oligotrophic). Considering the confidence intervals of the regression line (95 %) and the first point with a LDV different from the control values (dotted lines) the critical level was established. Because the observed values of LDVoligo are below the control value, a critical level lower than  $1.9 \mu\text{g m}^{-3}$  should be considered.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion