ICP VEGETATION: REVIEW OF NO_X CRITICAL LEVELS

FULL WORKSHOP REPORT

Workshop held online

Tuesday 24th May 2022

Document Control

First draft of workshop report / minutes prepared by Mike Perring (UKCEH) 25th - 27th May 2022.

Drafts checked by Felicity Hayes (Chair, ICP Vegetation) and Katrina Sharps (Head, Programme Centre of ICP Vegetation) (28th May – 22nd June 2022).

Minutes Draft circulated to workshop registered participants (9th June 2022) for feedback. No feedback = assumed agreement with minutes.

Minutes plus Workshop Report circulated to workshop participants and interested parties on 24th June 2022. Published on ICP Vegetation website.

Map 5.2 Concentration map of NO_x annual average, rural map, 2018

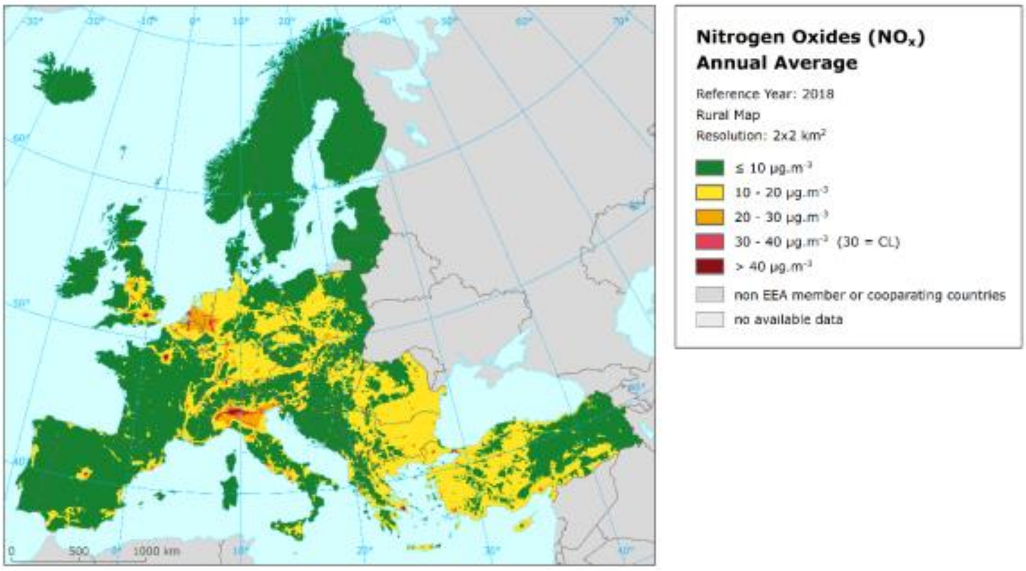

Figure reproduced from *European air quality maps for 2018*. Eionet Report ETC/ATNI 2020/10

Background, Review Outline and Workshop Purpose

ICP Vegetation is responsible for Chapter 3 of the Modelling and Mapping Manual (LRTAP Convention, 2017) and needs to check that it remains appropriate for use in the mapping of critical levels and loads, and their exceedances across Europe. This is under the auspices of the Convention on Long Range Transboundary Air Pollution (CLRTAP), and the Working Group on Effects (WGE).

Within the Mapping Manual, critical levels are defined as: *the concentration, cumulative exposure or cumulative stomatal flux of atmospheric pollutants above which direct adverse effects on sensitive vegetation may occur according to present knowledge*. This direct, oft-assumed relatively short-term ($\leq$ 1 year) impact on vegetation, first discussed by air quality specialists (Cape et al. 2009), contrasts with the ecologically-oriented critical loads definition: *a quantitative estimate of deposition of one or more pollutants below which significant harmful effects on specified elements of the environment do not occur according to present knowledge* (Posthumus 1988). The critical load definition implies indirect, as well as direct, impacts on systems through soil cycling, eutrophication, and acidification (at the system level), and is typically considered as a means to protect systems over the longer term (decades) (see Cape et al. 2009 for further comparison of critical levels and critical loads).

The critical levels for NO_x (the combination of nitric oxide (NO) and nitrogen dioxide (NO_2)) have been set at an annual mean of 30 µg/m^3 and a daily mean of 75 µg/m^3 since the second edition of the WHO Air Quality Guidelines (AQG) were published in 2000 (WHO, 2000). Indeed, detailed consideration of the WHO document showed that the annual level, recommended for mapping purposes, has not been revised since 30 µg/m^3 was proposed and ratified at workshops in Bad Harzburg (1988) and Egham (1992). See Caporn 1993 in Ashmore and Wilson 1993 for more details on this process, and Chapter 11 of the WHO AQG for developments since 1993. It is interesting to note that in the USA, the annual NO_x critical level, which was set in 1971 and has not been reviewed since, is 100 µg/m^3 and solely based on foliar injury.

Air pollution (and atmospheric) characteristics have radically altered across Europe since the annual NO_x critical level was set 30 years ago. Besides large increases in CO_2, background ozone (O_3) concentrations have tended to increase while episodic peaks have declined, and there have been large decreases in SO_2 concentrations and subsequent acid deposition. The latter is particularly pertinent to the setting of NO_x critical levels, as high SO_2 concentrations were associated with adverse vegetation impacts at lower NO_x concentrations, as compared to artificial atmospheres with high NO_x concentrations only (Caporn 1993). Environmental conditions have also altered, with higher mean annual temperatures and more extreme hydrological conditions (e.g. flooding, drought), in many regions.

In addition to the atmospheric context changing, people have questioned the need for two NO_x critical levels (daily and annual), given the expected statistical relationship between annual and maximum daily mean values (see Section: **Workshop Minutes: Session 1**). Questions have also been posed around whether separate critical levels for NO and NO_2 are needed (e.g. Caporn 1993). Simultaneously, working groups and task forces within the WGE are undertaking revisions of nitrogen critical loads and ammonia (NH_3) critical levels.

Given this background, ICP Vegetation decided there was an urgent need to review NO_x critical levels. The purpose of the workshop was to present the existing basis of current critical levels, give people the opportunity to present new research, to discuss findings, and to agree on an appropriate work plan to conduct a thorough review of NO_x critical levels. The latter was agreed following comments after circulating the draft minutes.

Full details on the expected review process are provided in the Section: **Work Plan – Draft Timeline**. In conducting this work, it should be remembered that scientific studies can only give objective evidence in relation to a limited question. How levels and loads are applied (in terms of implementation and ambition) is a policy matter. However, which ecosystem components and/or functions matter, and at what point an ecosystem (or component) is considered harmed are subjective questions (Rowe et al. 2016). This subjectivity needs to be addressed when considering the definition of critical levels and the evidence we extract to inform any revision of critical levels.

This report, and accompanying appendices, provide a record of the process leading up to the workshop and a summary of the workshop discussions. Minutes and selected presentations from the workshop are published on the ICP Vegetation website (https://icpvegetation.ceh.ac.uk).

Prior to the Workshop

The workshop was open to all interested parties.

Presentations (by Mike Perring of the ICP Vegetation Programme Centre), announcing the review opportunity and providing more details on the process, were given at:

- the ICP Vegetation Task Force annual meeting (online; February 2022)
- annual CAPER meeting (online and in person at Swansea, UK; March 2022)
- the ammonia (NH_3) critical levels workshop (online and in person at Dessau, Germany; March 2022), and
- the ICP Modelling and Mapping annual Task Force Meeting (together with the Co-ordination Centre on Effects (CCE)) (online; May 2022).

Prior to the workshop, Mike Perring (UKCEH) conducted a literature search / mini-review to assess current themes in international NO_x air pollution research, and nitrogen (N) impacts on vegetation more generally (see Section: **Workshop Minutes: Session 1**). From that review, he contacted lead/corresponding authors of those papers he deemed relevant to the review to alert them to the process and assess interest in participation.

All interested parties and registered participants were sent a draft agenda on 16th May 2022. This draft agenda included a series of questions that we invited parties to provide a written response to (if they wished). The questions were discussed in breakout groups.

The questions posed were:

i) Do you agree with the definition of critical levels? If not, why not and how should it be changed?
ii) Is there a rationale(s) for making separate critical levels for NO and NO_2? If yes, what is it / are they? If no, why not?
iii) Do you know of evidence, not already discussed / presented, that would refine current NO_x critical levels? Please note e.g. whether the evidence is from fumigation studies, gradient studies and/or another approach; the timescale of application.

iv) Is there a rationale(s) for retaining short-term ($\leq$ 1 day) critical levels, and/or long-term critical levels (i.e. annual average). Are other averaging periods important for risk to vegetation?

v) What is the best method of calculation of critical level now, given available data? In comparison to what has happened in the past?

vi) Although we are concentrating on risks to vegetation, are you aware of research suggesting that other organisms / ecosystem components need considering in the light of air pollution? Could this have a bearing on NO_x critical levels more generally?

vii) Is there a rationale(s) for modifying NO_x critical levels for certain ecosystem types / vegetation groups?

viii) Is there a rationale(s) for modifying NO_x critical levels based on other pollutants / climate change / land use?

ix) One of the uses of critical levels is to map exceedances. Are concentration-based metrics (as currently used for NO_x) the best way to map risk to vegetation? Do other methods need considering e.g. accumulation over threshold or flux-based measures like, for example, the phytotoxic ozone dose?

x) What progress (if any) is needed to help map NO_x critical levels? [Answers to this likely depend on preceding sections…]

xi) In your opinion, what are the gaps (if any) in NO_x critical levels research?

xii) In meetings considering NH_3 critical levels, there was some discussion about the relationship between critical loads and critical levels. Do you have any thoughts on how the critical level for NO_x (in whatever way it is refined) relates to critical loads?

xiii) Anything else to cover?

Agenda and Participants

The agenda for the workshop can be found in Appendix A. In total, 37 people were registered to attend the workshop, from 11 countries across Europe (Austria, Czech Republic, Finland, Germany, Ireland, Italy, Portugal, Spain, Sweden, Switzerland, United Kingdom), with 3 attendees from the United States of America (from the Environmental Protection Agency and United States Forest Service). Participants came from a range of sectors, including research scientists at university, non-governmental and governmental research organisations, policy-makers and policy-advisors from government and non-governmental agencies, and consulting firms.

Workshop Minutes

The agreed workshop minutes can be found at the ICP Vegetation website.

Appendix B provides abstracts of all the contributed talks.

A detailed workshop report now follows, with upshot and action points highlighted in grey and green boxes respectively. These details are useful for the fulfilment of working group tasks.

Session 1 (09.15 – 10.40 British Summer Time)

Mike Perring welcomed all to the workshop and gave a brief overview of the aims of the workshop (see Section: **Background, Review Outline and Workshop Purpose**). He also outlined the expected content and timeline for the broader review:

- First workshop (May 24th 2022): a time for discussion and to ascertain "what have we got"
- Second workshop (late autumn 2022): a time for decisions/recommendations and to ascertain "what can we do" i.e. application of critical levels to mapping and policy
- Annual ICP Vegetation Task Force Meeting (late February 2023): seek to reach agreement on adoption of modified text for Mapping Manual (if required) and any Background Document associated with this text (see Section: **Work Plan – Draft Timeline**).

Sabine Braun provided background on the data basis of the current critical levels. Sabine highlighted the fact that the air pollution climate is very different to when the levels were first set, particularly SO_2 levels. Sabine also highlighted that although the current version of the Mapping Manual (Oct 2017) refers to setting the NO_x level in the context of ozone (O_3), it is not clear from where this evidence came. Sabine emphasized that given the changed pollution climate, and the impacts of O_3 on stomatal control, there is an urgent need to consider O_3 x NO_x interactions. Sabine suggested there was very limited new evidence upon which to base revisions to NO_x critical levels, but noted the presence of gradient studies. However, it is difficult to separate concentration effects from deposition effects in such studies. There was some discussion on the possible need to consider seasonality in critical levels, for instance a winter mean value rather than daily mean as some fumigation experiments showing effects were performed in winter. Sabine also provided an update on the revision of empirical critical loads for N, noting that for some classes, the upper or lower value of the range has been lowered. Lastly, Sabine reflected on relationships between critical loads and critical levels, including the implication from translating the current critical level into a critical load based on an assumed deposition velocity in Switzerland. This was a specific topic in the breakout discussions and we provide notes in a later section on this important topic (see Section: **Session 4 – Breakout Discussions (2)**).

Mike Perring provided a critical review of the Mapping Manual text, noting ambiguity between what is stated in the Mapping Manual and details in Chapter 11 of the 2nd edition of the Air Quality Guidelines (AQG) from the World Health Organisation. He also highlighted text in the AQG that suggested there was evidence for lowering the annual mean to 15 $\mu g/m^3$, and that 30 $\mu g/m^3$ would not be protective of all species. However, this lowered level was from a different air pollution climate than the present. Mike noted that if this level were to apply, then far more European vegetation would be at risk than is presently considered the case (*Figure 1*).

Map 5.2 Concentration map of NO_x annual average, rural map, 2018

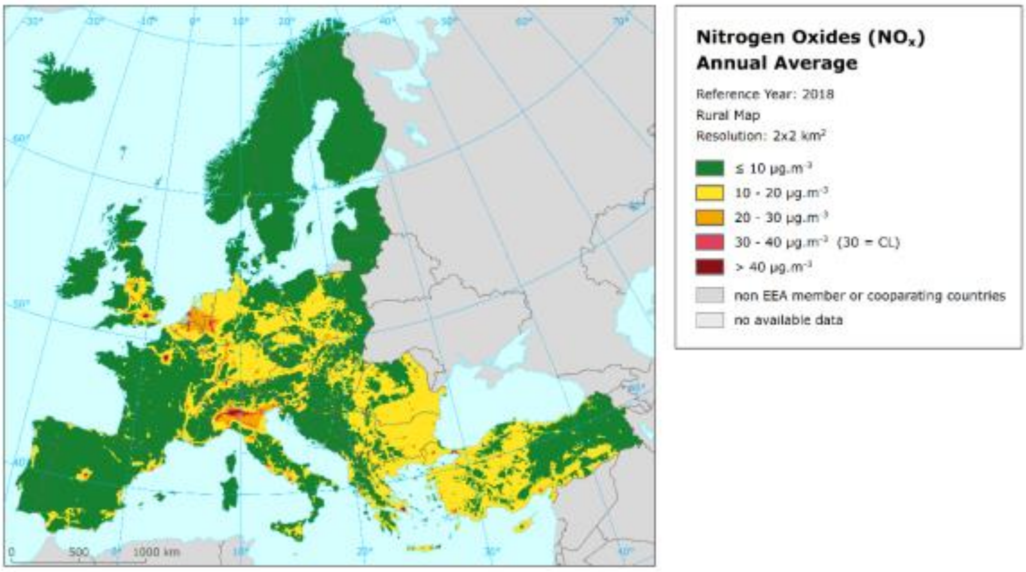

Figure 1: Annual average rural NO_x concentrations on a 2 x 2 km grid across Europe. Source: *European air quality maps for 2018*. Eionet Report ETC/ATNI 2020/10

Mike also introduced a systematic literature search approach, to uncover potential sources of new evidence to confirm or revise the basis for critical levels (see Appendix C). Subsequent discussions (in plenary and breakout rooms) provided additional considerations for this evidence synthesis approach, including methods to rate evidence and quality statements (e.g. akin to IPCC reports reporting degree of confidence and magnitude of likelihood) and what constitutes damage to vegetation.

Discussions suggested some confusion over how NO_x is expressed as the sum of NO and NO_2 in the Mapping Manual. This is in relation to conversion from different units (e.g. 'ppb' or 'µg/m^3' to 'nl/l'), consideration of atmospheric conditions, and to then enable addition of the two compounds to create an amount of NO_x expressed as NO_2. Potential explanations were suggested, but the discussion pointed to the need to provide explanations of these calculations (and potentially others) for the avoidance of doubt.

There was also discussion in relation to the statistical relationship between annual mean NO_x concentrations and the maximum daily NO_x concentration, with an illustration of this relationship shown for the UK by Ben Marner. If these relationships are tight (i.e. there is not much scatter) and consistent over time (e.g. was the same in 2000 or 2022) and space (e.g. the same in Wales or in Italy), then this questions whether there is a need to have both an annual and daily critical level (see green box, **Actions: Session 1**).

A question was also raised as to whether the ratio of NO_2 to NO that comprises the NOx level as a whole will have an impact on degree of vegetation damage, and the relevance of the pollutant mix. It was expected to be unlikely that there will be much evidence to address this question but this will require confirmation. For most NOx sources, including traffic, NO typically dominates over NO_2 in the exhausts. However, due to changes in car engine technologies the NO_2:NO ratio in exhausts has increased, which is relevant to the assessment of environmental risks near traffic emissions (Carslaw et al., 2007; Pleijel et al., 2009). There is much less information about the landscape variation in NO concentrations compared to NO_2. This depends on the fact that passive diffusion samplers for NO_2 are reliable and have been widely used (e.g. Klingberg et al 2017). NO data almost exclusively originate from permanent monitoring stations using NO_x instruments providing data with high time resolution. It is important to consider that there are fast atmospheric chemistry reactions converting between NO and NO_2 in particular the conversion of NO to NO_2 by the reaction with ozone and photolysis of NO_2 (Rodes & Holland, 1981; Clapp & Jenkin, 2001; Itano et al., 2007; Pleijel et al., 2009). This fast conversion may need consideration in risk assessment and can be used to analyse the primary fraction of NO_2 in NO_x exhausts. It would be worth noting in any revised Mapping Manual text if there is any evidence for differential (vegetation) responses to NO and NO_2. It would be worth noting in any revised Mapping Manual text if there is any evidence for differential responses to NO and NO_2.

UPSHOT: Session 1

There is a clear need to re-examine the Mapping Manual text, explanations, and underlying evidence.

Given the changed air pollution climate, there may be merit in re-visiting existing evidence as well as considering recent additions to the literature.

ACTIONS: Session 1

- **All** to consider search approach and list of literature (Appendix C) – anything to add: either published in the peer-reviewed literature or non-peer reviewed / unpublished e.g. submitted work, theses and reports.
- Evidence synthesis from literature
- Analysis of spatial and temporal consistency in maximum daily mean vs annual mean NO_x
- Analysis of evidence in relation to separate effects of NO and NO_2 on vegetation
- (Re-) drafting of Mapping Manual text and any accompanying Background Document to consider points raised above, including relationship between critical loads and critical levels, calculation of NO_2 equivalence, and atmospheric air pollution composition and need to consider interactions with pertinent pollutants

Session 2 (11.00 – 13.00 BST)

Talks

The second session comprised three talks on empirical NO_x research in boreal, temperate and Mediterranean climates, covering lichens, bryophytes and trees respectively, and breakout discussions on questions i) to vi).

Sirkku Manninen compared lichen community composition in boreal forest to modelled NO_2 concentrations, showing how lichen composition was affected by NO_2 concentrations much lower than the current critical level. However, Sirkku also showed that increased modelled NO_2 concentrations were associated with empirically-estimated increased NH_3 concentrations (using UKCEH Alpha samplers), and that deposition of both gases and subsequent reactions affected bark pH. This makes it difficult to attribute the causal agent(s) underlying the changed lichen communities. Information was also given in regard to the formation of salts i.e. ammonium nitrate, that causes osmotic problems leading to reduced uptake of water through the lichen surface. Sirkku suggested that this interactive effect of dry-deposited N forms on lichen function is more harmful to acidophytes than to more drought-tolerant nitrophytes.

Elise Fox provided a presentation on the challenges that accompany relating bryophyte distribution to NO_x concentrations in the UK. These challenges include identification of the bryophytes themselves, the fact that bryophytes exhibit a range of stomatal characteristics (from a total lack of stomata, to possessing pseudo-stomata and some species having real stomata), and whether extreme values, in conjunction with environmental contexts e.g. drought, may be more relevant ecologically than annual means to distributional patterns. Elise also noted the various available databases for contributing to an analysis of bryophyte distribution in relation to NO_x.

Duncan Mifsud outlined the pollution issues that affect Malta, and showed results from a preliminary study that demonstrated how the use of tree rings can help map trends in pollutant emissions beyond the available instrumental record. He also discussed how use of isotopic signatures allows source attribution. Using sites at different distances from a road, he showed that sites closer to a road exhibited tree rings with more positive $\delta^{15}N$ trends and more negative $\delta^{13}C$ trends, indicative of the uptake of 15-N enriched NO_x and 13-C depleted CO_2 from motor vehicle pollution. In the context of critical levels, it would be interesting to assess to what extent these trends were associated with different growth dynamics, but a greater number of sampled trees and sites would be needed to conduct such an analysis, and the presence of confounding pollutants would need accounting for in the design and/or interpretation.

Discussions and questions after these talks included remarks on what to use as an indicator of an effect. *Cristina Branquinho* shared some relevant findings on lichen physiological and ecological response. Although all species responded negatively to increased NO_x concentrations, differences in the magnitude meant that the ecological endpoint (community change) led to some 'winners' and 'losers'. There were comments made that it may be better to look at how a pollutant affects the number of species, or how well a particular species does compared to others. *Ed Rowe* reiterated the difficulty of NO_2 having growth stimulation effects and whether that can be considered positive or negative.

UPSHOT: Session 2 Talks

Confounding influences need accounting for in spatial (gradient) studies and attribution of the cause of any vegetation effects is difficult.

Physiological changes can be negative for all species in response to NO_x but some species are less affected than others, meaning the ecological endpoint can be different.

ACTIONS: Session 2 Talks

- Incorporate relevant findings for NO_x critical levels into evidence extraction and revision of Mapping Manual and any accompanying Background Document
- Discussion and/or evidence gathering on what constitutes an 'adverse effect' when interpreting NO_x critical level. For instance, USA have only used foliar injury.

Breakout Discussions (1)

Questions were discussed in breakout sessions and responses reported back in plenary.Below is the upshot from discussion/comments around each question.

i) *Do you agree with the definition of critical levels? If not, why not and how should it be changed?*

There was general agreement with the critical levels definition, with one suggested change. Whereas this is currently '*adverse effect on vegetation*', this could perhaps be changed to '*adverse effect on ecosystem functioning*' as this accounts for occurrences when there could be positive effects on an individual species, which have a knock-on negative effect more broadly.

There was discussion about a need for clarification. There was some confusion about what is a direct *vs* an indirect effect. Some interpret indirect as anything other than the very first reactions (e.g. upregulation of antioxidants, change in biomass allocation), whereas others interpret indirect as that caused by longer-term ecosystem responses only, particularly those mediated by substrate dynamics e.g. acidification or eutrophication. But direct effects can also involve acidification e.g. of plant cells so the context in which terms are being used needs to be made clear.

After the workshop it was commented that any adapted definition needs to be in line with article 1 of the Gothenburg Protocol: " 'Critical levels' *means concentrations of pollutants in the atmosphere or fluxes to receptors above which direct adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur, according to present knowledge".*

Setting a critical level for NO_x is complicated since it mainly stimulates plant growth rather than being toxic, so the (main) effects will be on the composition of plant / lichen assemblages.

ii) *Is there a rationale(s) for making separate critical levels for NO and NO_2? If yes, what is it / are they? If no, why not?*

It was concluded that there is not enough information to distinguish NO and NO_2 critical levels. There is very little information about them singly. In addition, there are fast atmospheric chemistry reactions converting between NO and NO_2. It would be worth noting in the Mapping Manual / Background Document text if there is any evidence for differential vegetation responses / susceptibility to NO and NO_2.

Any additions could also consider toxicity / effect size in relation to NH_3. Note that in Mark Sutton's NAQI equation, a mol of NH_3 is about 4 times as damaging as a mol of NO_2.

iii) *Do you know of evidence, not already discussed / presented, that would refine current NO_x critical levels? Please note e.g. whether the evidence is from fumigation studies, gradient studies and/or another approach; the timescale of application.*

Helena Ribeiro's study on the effects of NO_2 on pollen pointed out the short exposure time (6 hours). There are few studies showing a negative impact of NO_x on a shorter time scale. Note that Helena has also conducted work on O_3 and NO_x interactions.

Work done in Ashdown forest, led by Mark Sutton but not published in peer reviewed literature: https://www.wealden.gov.uk/planning-and-building-control/planning-policy/planning-policy-evidence-base/habitat-regulations-assessment/. See also Sutton (2019) *Risks from air pollution to the integrity of Ashdown Forest Special Area of Conservation: Overview of Issues and Conclusions*

See also evidence provided by Mark Sutton on NAQI – in **Section 4: Breakout Discussions (2)**.

iv) *Is there a rationale(s) for retaining short-term (≤ 1 day) critical levels, and/or long-term critical levels (i.e. annual average). Are other averaging periods important for risk to vegetation?*

It is appropriate to keep short- and long-term critical levels as they target different things (both in terms of emissions, and in terms of effects).

The importance of short- and longer-term critical levels may vary with species/vegetation type. For instance, for bryophytes it may be more relevant to look at shorter-term effects.

Note that in air pollution work undertaken by Ben Marner, and for critical loads mapping, three year averages of pollutant levels are used to account for annual variation in weather effects.

v) *What is the best method of calculation of critical level now, given available data? In comparison to what has happened in the past?*

It was thought that it is unlikely that there will be sufficient data to go beyond the existing method of calculation. In other words, plotting the concentration of "NO_2" where harmful effects have been observed (biochemical, physiological, ecological) on a graph of "NO_x expressed as NO_2" concentration *vs* exposure duration – see further details below. Suggested the use of statements indicating the quantity of supporting evidence. IPCC and/or IPBES reports contain examples of this kind of grading.

It is important to highlight which type of study a certain source of information represents e.g. experiment, gradient, epidemiological.

Possible methods to consider whether long-term vs short-term concentrations best correlated to impacts in gradient studies were suggested

Qualitative observations may be of interest in addition to strictly quantitative effects.

vi) *Although we are concentrating on risks to vegetation, are you aware of research suggesting that other organisms / ecosystem components need considering in the light of air pollution? Could this have a bearing on NO_x critical levels more generally?*

Other ecosystem components could be included if there is evidence available. Possibilities include pollinator disruption due to interactions between NO_x and volatile organic compound (VOC) signalling.

It was noted that trees and vegetation could remove NO_x from the air in cities to make the air cleaner.

NO_x is likely to be more toxic to animals than plants and there may be some evidence from NO_x experiments on animals.

ACTIONS: Breakout Discussion 1

- Refinement of critical levels concept towards agreed definition that accounts for developments since 1992. Remove any ambiguity and clarify what is and what is not in scope.
- Investigate evidence for difference in mode of action for NO and NO_2, with awareness that there will (likely) not be evidence for separation of NO_x critical level but to lead to research recommendations.
- Check Tables of Evidence (Appendix C) for whether additional information sources are available
- Provide spatio-temporal analysis of relationships between annual and daily NO_x concentrations. Analysis of pollution profiles e.g. NO to NO_2 ratio. Note that *annual mean and maximum 24-hour mean are mathematically related, and fairly tightly correlated.*
- Provide text on why / why not short- and long-term critical level. Make recommendation for amended Mapping Manual text.
- Write section on evidence for NO_x effects on other organisms. Assess whether any evidence to refine critical level(s) for vegetation based on other organisms. For instance, due to deleterious effects on plant fitness through impaired pollination.

Session 3 (13.30 – 14.30 BST)

Helena Ribeiro presented results from short-term (6-hour) exposures of NO_x to pollen, using different tree species and discussing implications for pollen's function. In the talk, Helena focused on presenting results on pollen viability, protein levels, reactive oxygen species and NADPH enzyme activity. Besides illustrating species-specific differences, Helena also confirmed that reactions could be related to exposure concentrations but this was not true of all species. Helena also commented on the fact that her work had considered NO_x interactions with O_3, and these results can be found in the main paper (in the main table of literature, Appendix C).

Tara Greaver provided a North American perspective on critical levels, complementing Sirkku's earlier talk on lichens. Tara showed that lichen communities respond to NO_2 concentrations at levels below the European / WHO Air Quality Guideline value, and far below the USA standard. However, as highlighted elsewhere and in the breakout discussions, causal attribution of these results to NO_2, and disentanglement from N deposition effects, is difficult.

Héctor García-Gómez detailed NO_2 measurements in Spanish Mediterranean forests in the context of dry deposition. Héctor provided perspectives on the relationship between NO_2 concentrations and deposition, reiterating the difficulty in separating N deposition effects from direct NO_2 impacts. A particularly notable result was that almost equivalent NO_2 concentrations at two different sites led to very different N deposition and uptake into the plant, based on differences in stomatal conductance due to moisture conditions. Héctor highlighted the potential to modify and use the DO_3SE model to improve projections of vegetation exposure risk to NO_2.

UPSHOT: Session 3 Talks

New evidence on NO_x impacts on vegetation over short timescales is available.

Confounding influences need accounting for in spatial (gradient) studies and attribution of the cause of any vegetation effects is difficult.

Physiological changes can be negative for all species in response to NO_x but some species are less affected than others, meaning the ecological endpoint can be different.

The disentanglement of N deposition from NO_2 exposure is difficult. To some, everything relates to critical loads i.e. is a flux into the plant / system.

ACTIONS: Session 3 Talks

- Incorporate relevant findings for NO_x critical levels into evidence extraction and revision of Mapping Manual and any accompanying Background Document. See also actions from Breakout Discussions.

Session 4 (14.55 – 15.45 BST)

Breakout Discussions (2)

Prior to the breakout groups, *Mark Sutton* provided a perspective on lichen community change in relation to NH_3 and NO_2, presenting Equation [1] that describes the nitrogen air quality index (NAQI) as a function of both chemical species:

$NAQI = 2\,[NH_3]^{0.5} + [NO_2]^{0.5}$ where [] are measured in $\mu mol/m^3$ **Equation [1]**

As well as Equation [1] showing that ammonia is more toxic than NO_2 to lichen species, it also raises some interesting considerations in relation to the setting of the NO_x critical level in the light of ammonia. For instance, taking the existing critical levels for NH_3 and NO_x, the NAQI computes as 1.29, which can be considered a very N polluted atmosphere based on the lichen indicator score. In contrast, NO_2 at the current critical level but in the absence of ammonia provides a NAQI score of 0.8, which relates to a lichen community considered at risk from N pollution. To consider a community not at risk i.e. the air to be considered clean, the critical level for NO_2 would need to be 11.5 $\mu g/m^3$, and that is in the absence of NH_3. To what extent these findings apply to other climatological contexts is unknown, as the NAQI is based on UK data, while the definition of clean / at risk etc needs to be clearly communicated.

The need to consider the combined effect of NO_x and NH_3, especially in relation to eutrophication, was reiterated by *Harald Zechmeister* after the workshop, emphasizing that this consideration is not only in relation to poikilohydric species, such as mosses and lichens, but also for wetlands, and the fact that NH_3 appears more influential than NO_x.

Below is a summary from discussion/comments around each question.

vii) Is there a rationale(s) for modifying NO_x critical levels for certain ecosystem types / vegetation groups?

Difficult to separate N deposition and NO_x concentration, particularly when looking at ecosystem level.

Evidence that lichen communities (in northern Europe and in the USA) respond at much lower NO_x levels than current European critical level. However, unclear whether other pollutants drive this and/or whether this is an N deposition impact.

viii) Is there a rationale(s) for modifying NO_x critical levels based on other pollutants / climate change / land use?

Many suggested that there will likely be an absence of evidence. Also that it is difficult to understand all possible interactions with other pollutants.

Note that previous experiments were carried out when ambient SO_2 was much higher than today. A suggestion that NO_x (and NH_3) impacts might be larger than previous due to the difference in SO_2 interaction. Although Natural England have interpreted this the other way, with the use of a higher daily critical level for NO_x (200 $\mu g/m^3$ rather than 75 $\mu g/m^3$) – the latter may be too precautionary – as SO_2 has gone down and providing accumulation over a threshold of 40 ppb (AOT40) for ozone has not been exceeded].

A method was suggested to investigate the composition of pollutants in roadside studies [which may help with disentanglement of effects if there are different compositions in different locations and/or different temporal trends]. In the UK, NH_3 has been increasing / is stable while NO_x has been decreasing. This trend is not universal.

For bryophytes, changes in climate (e.g. more rainfall) may lead to reduced resistance to effects of NO_x (when bryophytes are dry, they are more resistant).

ix) *One of the uses of critical levels is to map exceedances. Are concentration-based metrics (as currently used for NO_x) the best way to map risk to vegetation? Do other methods need considering e.g. accumulation over threshold or flux-based measures like, for example, the phytotoxic ozone dose?*

It was agreed that scientifically it made sense to use a flux based metric, which would take into account environmental effects, but that there is not enough data at the moment to do this. It should also be noted that although stomatal conductance may affect N flux (uptake and deposition) there are other major uncertainties when it comes to deposition that are likely more significant. For instance, the effects of surface roughness of vegetation are considered in only a binary way, forest or non-forest.

DO_3SE could be used in the future, if more data become available.

It may be possible to used a concentration-threshold approach (similar to AOT40 for ozone).

As lichens have no stomata, flux based approach not suitable for all classes of vegetation.

x) *What progress (if any) is needed to help map NO_x critical levels?*

New receptor map will soon be available from ICP M&M, ecosystem/habitat based.

Mapping critical level exceedance for pollutants like NO_x, e.g. using 1x1km data has difficulties as the pollutant is very spatially variable.NOx is decreasing across Europe, and the spatial distribution of pollution sources is going to change with time too.

Further discussion of separating NOx concentration and N deposition.

After the workshop it was noted (also in the JNCC N Futures Report) that our understanding of critical level exceedances must always be framed by the spatial resolution of our assessment.

xi) In your opinion, what are the gaps (if any) in NO_x critical levels research?

How to separate effect of NO_x from N deposition and/or other pollutants in gradient studies. This would likely require monitoring of many different locations with different pollutant characteristics. If sufficient places could be found, with control of other variables, inference can be improved even if causation cannot be confirmed.

For the separation of effects, experimental studies could be initiated that add equivalent amounts of N deposition but without exposure to elevated gaseous NO_x vs a treatment with same N deposition but in form of NO_x. A similar set-up is present at Whim Bog for wet N deposition and NH_3.

Investigations of interactions with other factors, e.g. interactions between NO_x and temperature.

xii) In meetings considering NH_3 critical levels, there was some discussion about the relationship between critical loads and critical levels. Do you have any thoughts on how the critical level for NOx (in whatever way it is refined) relates to critical loads?

NO_x compared to N-deposition: recommend to just note in the text that deposited NO_x is a component of N load, and that this is accounted for in the critical loads for N.

There was some discussion about differentiating the relevant effects for the NO_x critical levels by considering whether the effect is mediated through effects on the leaves/flowers, compared to effects via changed N to soil.

ACTIONS: Breakout Discussion 2

- Text for Background Document / Mapping Manual on rationale and/or evidence for modification of critical levels based on other air pollutants / climate change / land use / vegetation types / ecosystem types.
- Incorporation of NAQI/lichen indicator score evidence in overall evidence bank.
- Comparison of how spatial critical level exceedance varies with different metrics of risk to vegetation
- Text in Background Document / summarised for Mapping Manual on evidence for what metric best represents risk to vegetation. If insufficient evidence, what actions are needed to improve understanding.
- Summary text on NO_x critical level research gaps
- Text on relationship between critical loads and critical levels and how critical levels need to be interpreted.

Session 5 (15.45 – 16.05 BST)

Following feedback from the different breakout groups, *Mike Perring* provided a short summary of ideas for next steps, which are contained within the minutes, as well as the Section: **Work Plan – Draft Timeline**.

Work Plan - Draft Timeline

The anticipated timeline for the review process is:

May 24th 2022:	First online workshop on ICP Vegetation Review of NO_x Critical Levels.
June 9th 2022:	Request **for volunteers for different workgroups.** To respond by 16th June 2022.
June 24th 2022:	Finalised minutes and report from Workshop 1 circulated to participants, and published on ICP Vegetation website soon thereafter.
Mid-July 2022:	**Finalise participation in working groups**
Mid-July to mid-Oct 2022:	Working groups progress evidence extraction, analysis and text development. Submit findings to ICP Vegetation Programme Centre mid-October 2022.
Mid-Nov 2022:	Second online workshop to address questions / issues / matters arising.
Mid-Nov 2022 to mid-Jan 2023:	Working groups convene to address developments from second online workshop.
Mid-Jan 2023:	Working groups submit recommendations to ICP Vegetation Programme Centre.
20th – 23rd Feb 2023:	**ICP Vegetation Task Force Meeting –** seek agreement on revised Mapping Manual text and Background Document. Feed up to WGE and LRTAP as appropriate.

Literature Cited

Ashmore and Wilson 1993 Critical Levels of Air Pollutants for Europe

Cape et al. 2009 Reassessment of critical levels for atmospheric ammonia pp 15 – 40 in *Atmospheric Ammonia Detecting emission changes and environmental impacts* (Eds. Sutton, Reis and Baker)

Caporn 1993 Critical levels for NO_2 pp. 48 – 54 in *Critical Levels of Air Pollutants for Europe* (Eds. Ashmore and Wilson).

Carslaw et al. 2007 Risks of exceeding the hourly EU limit value for nitrogen dioxide resulting from increased road transport emissions of primary nitrogen dioxide *Atmospheric Environment* **41**: 2073 - 2082

Clapp and Jenkin 2001 Analysis of the relationship between ambient levels of O_3, NO_2 and NO as a function of NO_x in the UK *Atmospheric Environment* **35**: 6391 - 6405

Itano et al. 2007 Impact of NOx reduction on long-term ozone trends in an urban atmosphere *Science of the Total Environment* **379**: 46 - 55

Klingberg et al. 2017 Influence of urban vegetation on air pollution and noise exposure – A case study in Gothenburg, Sweden *Science of the Total Environment* **599-600**: 1728 - 1739

LRTAP Convention, 2017 (can be found at: https://icpvegetation.ceh.ac.uk/get-involved/manuals/mapping-manual) (URL correct as of 24th June 2022).

Pleijel et al. 2009 Characteristics of NO_2 Pollution in the City of Gothenburg, South-West Sweden—Relation to NO_x and O_3 Levels, Photochemistry and Monitoring Location *Water Air Soil Pollution: Focus* **9**: 15 - 25

Posthumus 1988 Critical levels for effects of ammonia and ammonium pp. 117 - 127 In *Proceedings of the Bad Harzburg Workshop*

Rodes and Holland 1981 Variations of NO, NO_2 and O_3 concentrations downwind of a Los Angeles freeway *Atmospheric Environment* **15**: 243 - 250

Rowe et al. 2016 Using qualitative and quantitative methods to choose a habitat quality metric for air pollution policy evaluation *PLOS ONE* **11(8)**: e0161085

WHO 2000 *Air Quality Guidelines for Europe* (2nd Edition)

Appendix A

AGENDA

***Conceptual Background and Empirical Investigations I* (Chair: Felicity Hayes, UKCEH)**

09.15: Welcome and Plan for the Day (Mike Perring, UKCEH)

09.30: The data basis of the current critical levels (Sabine Braun, Institute for Applied Plant Biology)

09.50: A brief review of current literature (Mike Perring, UKCEH)

10.10: Discussion Time / Questions Arising

10.30: COFFEE BREAK

***Empirical Investigations II* (Chair: Mike Perring, UKCEH)**

11.00: The structure of epiphytic macrolichen community in relation to modelled NO_2 concentration in a boreal city (Sirkku Manninen, University of Helsinki)

11.20: The Influence of NOx Emissions on the Nitrogen Isotopic Composition of Tree Rings and Foliage (Duncan Mifsud, University of Kent)

11.40: Challenges in modelling the effects of NOx concentrations on UK bryophyte distribution (Elise Fox, Liverpool John Moore's University)

12.00 – 12.30: First breakout discussion. Please see breakout discussion questions.

12.30 – 12.45: Feedback, in plenary, on first breakout discussions (Chair: Felicity Hayes, UKCEH)

12.45 – 13.30: LUNCH

***Empirical Investigations III* (Chair: Katrina Sharps, UKCEH)**

13.30: Effects of increasing nitrogen dioxide concentrations in pollen of different forest species (Helena Ribeiro, University of Porto)

13.50: Synthesis of published lichen response to gaseous nitrogen: ammonia versus nitrogen dioxide (Tara Greaver, US EPA)

14.10: NO2 measurements in Spanish Mediterranean forests in the context of dry deposition (Héctor García-Gómez, CIEMAT)

14.30: COFFEE BREAK

14.55: Introduction to second breakout discussions (Mike Perring, UKCEH)

15.00 – 15.40: Second breakout discussion. Please see breakout discussion questions.

15.40 – 16.00: Feedback, in plenary, from second breakout discussions (Chair: Mike Perring, UKCEH)

16.00: Next steps (Mike Perring, UKCEH)

16.20: Meeting close

Appendix B

SUBMITTED ABSTRACTS

Sabine Braun, Institute for Applied Plant Biology

The data basis of the current critical levels

This talk will highlight how stakeholders arrived at the existing NOx critical levels for vegetation. It will also present some reflections on earlier debates around the setting of critical levels, especially in the light of interactions with other pollutants, and links to subsequent work on N critical loads. The talk is presented by one of the original participants in the Egham (1992) workshop

Elise Fox, Liverpool John Moore's University; Hayes, F., UKCEH; Dalrymple, S., Liverpool John Moore's University

Challenges in modelling the effects of NOx concentrations on UK bryophyte distribution

Previous critical levels for semi-natural vegetation have been informed from data regarding vascular plants however, mosses, hornworts and liverworts have been neglected in these calculations. This has been mainly due to challenges in quantifying pollutant fluxes at fine scale. These fluxes are largely governed by boundary layer processes that are difficult to measure. Bryophytes help regulate nutrient supply in plant communities and are a microhabitat for many other organisms and therefore, should be accounted for when reviewing NOx critical levels.

Héctor García-Gómez, CIEMAT

NO2 measurements in Spanish Mediterranean forests in the context of dry deposition

During 2016-2017, four holm oak forests were intensively monitored (meteorology, soil water content, nitrogen deposition, gaseous pollutants with passive and active samplers, particulate nitrogen, etc.) in Spain. Below-canopy concentrations of N gaseous pollutants were significant smaller than levels found in the open field. For NO2, those reductions (up to 41%) were comparable to, and even higher than, values reported in similar empirical studies with deciduous forest species. This evidence of air quality improvement for Quercus ilex forests requires specifically designed monitoring programs of urban and peri-urban forests to quantify the relevance of this ecosystem service and understand the environmental processes involved. Stomatal uptake of NH3, HNO3 and NO2 derived from the DO3SE (Deposition of Ozone and Stomatal Exchange) model, was estimated to calculate total dry deposition of inorganic N air pollutants in these four forests. The stomatal deposition of N gases averaged for the four sites 3.3 ± 0.8 kg N ha-1 year-1, with NO2 contributing the most (2.0 ± 0.4 kg N ha-1 year-1), contributing deposition averaged from 19% in the peri-urban forests to 11% in the most natural site.

Tara Greaver, US EPA

Synthesis of published lichen response to gaseous nitrogen: ammonia versus nitrogen dioxide

In this synthesis, we characterize U.S. air concentrations of the most ubiquitous gaseous forms of oxidized nitrogen, NO2, and its direct effects on lichens. In the U.S., the 3-year average (2017-2019) of the annual mean for each monitoring site ranges up to 30 ppb (~56.4 ug m -3) for NO2. The spatial coverage of current routine monitoring of NO2 likely does not accurately represent exposures of NO2 to ecosystems in rural areas. NH3 can act as a nutrient to lichens, but as exposure rise, both can cause physiological stress, and mortality. There is a growing body of evidence that lichen community composition is altered at current levels of exposure in the U.S., with no effect concentrations from <1-3 ug m -3 NO2. Better spatial characterization of both NO2 and NH3 concentrations, especially near intensive agriculture, would help to characterize the extent of the impacts across the U.S.

Sirkku Manninen, Faculty of Biological and Environmental Sciences, University of Helsinki; Jääskeläinen K., Faculty of Biological and Environmental Sciences, University of Helsinki; Niemi J., Helsinki Region Environmental Services Authority (HSY), Helsinki, Finland.

The structure of epiphytic macrolichen community in relation to modelled NO_2 concentration in a boreal city

Vandinther (2019) showed dry deposition of NO and NO_2 being a strong driver of lichen community structure on the acid bark of Jack pine (*Pinus banksiana*) in northern forests. The responses of microlichens to NH_3 and NO_x vary even within a given functional group. This is partly attributed to species-specific uptake rates of NH_4^+ cf. NO_3^- cf. organic N (Dahlman et al. 2004). Moreover, N-tolerant species can oxidize surplus NH_4^+ to NO_3^-, a non-toxic form of N, and thus do not accumulate excess N as NH_4^+ to same extent as acidophytes do (Gaio-Oliveira et al. 2004, 2005).

Epiphytic macrolichens were scored on *Pinus sylvestris* and/or *Quercus robur* trunks in Helsinki (60°10'N, 24°56'E) in summer 2016 using the Finnish standard (Suomen Standardisoimisliitto 1990), while the European Standard EN 16413:2014 was used in summer 2019. In 2016, the number of indicator species correlated negatively with modelled NO_2 concentration, SO_2 concentration, and concentrations of NO_2^-+NO_3^- -N, NH_4^+ -N and S of *Pinus* bark. The most responsive acidophytes to oxidized forms on N seemed to be e.g., *Hypogymnia physodes* and *Parmeliopsis ambigua* (Manninen 2018). Based on the 2019 data, *P. ambigua* on *Pinus* responded negatively to modelled NO_2 (range 8.0-11.9 µg m^{-3} yr^{-1}) as did *H. physodes* and the lichen diversity value of acidophytes (LDV_A) on *Quercus* (8.0-23.4 µg NO_2 m^{-3} yr^{-1}). In contrast, increases were found in the abundances and presence of nitrophytic species on *Quercus* with increasing NO_2 concentration. The results suggest a shift from the dominance of acidophytes to that of nitrophytes on at 10-15 µg NO_2 m^{-3} yr^{-1}. The results will also be discussed in terms of methodology (e.g., calculated indices).

Dahlman et al. 2004, Planta 219, 459-467
Gaio-Oliveira et al. 2004, Environmental Pollution 158, 2553-2560
Gaio-Oliveira et al. 2005, Planta 220, 794-803
Manninen S. 2018, Science of the Total Environment 613-614, 751-762
Suomen Standardisoimisliitto 1990. SFS Standard 5670. Air Quality. Bioindication. Mapping of Epiphytic Lichens (in Finnish).
Vandinther K. 2019. The influence of nitrogen deposition on community composition in Pinus banksiana forests across Northwestern Canada. M.Sc. thesis. Trent University, Peterborough, Ontario, Canada. 175 pp.

Duncan Mifsud, University of Kent

The Influence of NOx Emissions on the Nitrogen Isotopic Composition of Tree Rings and Foliage

Emissions from motor vehicle traffic over the past few decades have become a significant contributor to regional air pollution. Nitrogen oxides (NOx) are a major component of traffic emissions, and it is known that exposure to these species may be detrimental to public health. Recent studies have demonstrated that the stable isotope ratios of nitrogen in tree rings and foliage are influenced by the nature of their major nitrogen source, making them appropriate for semi-quantitative bio-monitoring studies. This proxy was applied to Aleppo pines (P. halepensis) growing at three distances from one of the busiest roads in Malta, a small country known to suffer from intense traffic pollution. No temporal variation in the nitrogen and organic carbon stable isotope ratios was detected in the sampled tree rings corresponding to the time period 1980-2018. However, statistically significant spatial trends were observed in both tree rings and foliage: sampled sites closer to the road exhibited more positive δ15N and more negative δ13C values compared to those at a rural background site. This is likely due to their increased take up of 15N enriched NOx and 13C depleted CO2 from traffic emissions. Top soils sampled at the three investigated sites also showed the δ15N trend. These results contribute to a growing body of evidence suggesting that tree ring and foliage isotope measurements are a useful indicator of regional air pollution and are also the first known application of dendrogeochemistry to atmospheric pollution monitoring in Malta.

Mike Perring, UKCEH

A brief exploration of current literature

Mike will present initial findings from a systematic search of the literature since the publication of NOx critical levels from the 1992 Egham workshop. He will highlight the themes that emerge from this brief review, including research on pollutant mixtures, effects on native as well as crop species, gradient studies, and the importance of NO in plant metabolism. He will provide a table of the main literature, and invite participants to consider whether they know of additional evidence that could be included in subsequent analyses (published or unpublished). He will also provide a framework for classification of the literature, which may help subsequent analyses and invite comment.

Helena Ribeiro, University of Porto

Effects of increasing nitrogen dioxide concentrations in pollen of different forest species

Pollen, the male gametophyte of seed plants, has a preponderant role in fruit production and consequently for the propagation of the species, as well as a food source for some pollinators. During emission and dispersion in the air, pollen is subjected to chemical and physical interactions with other atmospheric constituents, such as gas pollutants, which can cause stress in these biological structures and influence its mission. Therefore, pollen sensitivity and tolerance to air pollutants such as NO2 can be ultimately preponderant for crop production success. In this talk, it will be presented the influence of NO2 in key aspects related to pollen performance of 4 forest tree species, *Betula pendula*, *Corylus avellana*, *Acer negundo* and *Quercus robur*, through a comparative analysis under the same experimental conditions. We will discuss the effect on pollen fertility, protein content, oxidative stress, and wall composition after exposure in vitro to nitrogen dioxide at increasing concentration levels. Our results suggest changes in pollen viability, protein content and differential sensitivity related to ROS synthesis, NADPH oxidase activity as well as in wall composition. Our study points out that significant pollen functions could be compromised even at common air pollutant's concentrations.

Appendix C

LITERATURE SEARCH

Introduction

This appendix outlines the literature search approach, conducted by Mike Perring, to assess the evidence base for NO_x affects on vegetation, and manuscripts considering critical levels research. Literature discovered during the systematic search was complemented by manuscripts/evidence suggested by those contacted prior to, and during, the first workshop. The end of the appendix includes a table of all literature retained, after screening the title and abstracts and accounting for expert opinion. We will extract evidence from this literature, and any additional papers/information that are published during the review process/brought to our attention. **We invite readers to check this table and send through any additional literature/evidence suggestions soonest.**

Search Approach

Systematic Search

At the end of February 2022, Mike Perring searched Web of Science, using UKCEH's credentials and therefore access level, with a "Topic (TS) =" search of "NOx critical level". This search returned 0 results, although without quotations, the search returned 487 manuscripts. Mike also searched with TS = nitr* AND veg* damage*, to uncover work that has considered damaging effects of nitrogen, in whatever form, on vegetation. Mike conducted the latter search because of the current critical level definition and its consideration of adverse effects on vegetation; it returned 1461 titles.

All results from these two searches had their titles screened for relevance (37 / 487 and 72 / 1461 relevant titles respectively). Retained titles then had abstracts screened to assess what may remain relevant for evidence extraction.

The primary aim of this initial search was to find authors who may be interested in the critical levels review process (and extend invitations to them at the appropriate time), as well as get a flavour of recent research around NO_x critical levels and nitrogen impacts on vegetation more generally.

At the end of March 2022, Mike Perring carried out a more refined search using Web of Science, accounting for whether critical levels were mentioned in the same paper as chemical nitrogen oxides (*Search 1*) and whether these chemical species were mentioned in the same paper as different means of characterising vegetation (*Search 2*). In all cases, he carried out searches as TS (topic) =. Due to the volume of results returned in *Search 2*, and lack of resources to consider all nearly 6000 papers, lists were sorted in two ways: i) by times cited (highest first, and the first 2000 titles screened), and ii) date published (most recent first, first 1000 screened). In this way, it was assumed approximately half the papers would be considered, covering both recent publications and classic works.

Search 1:

("nitrogen dioxide" OR "nitric oxide" OR "NOx") AND ("crit* level*")

75 manuscripts, 22 retained after title screen

Search 2:

("nitrogen dioxide" OR "nitric oxide" OR “NOx”) AND (“vegetation” OR “indicator plant*” OR “lichen*” OR “moss*” OR “bryophyt*” OR “forest*” OR “woodland” OR “grass*” OR “fruit tree*” OR “shrub” OR “verge” OR “roadside” OR “conifer*” OR “decid*” OR “crop*)”

5941 manuscripts returned. Sorted by times cited: 218 / 2000 retained after title screen; Sorted by publication date: 147 / 1000.

The **Table of Literature**, included at the end of this Appendix was first compiled from literature retained from Search 1 and 2. Earlier searches were then checked for any papers that had been missed, and any papers that were sent to Mike prior to the workshop.

Expert Opinion / Solicitation

At, and immediately after, the workshop, additional evidence was suggested by interested parties. These additional papers were inserted into the overall list when deemed relevant.

Table of Literature found during systematic searches / via expert feedback, March 2022 [*correct as of 24th June 2022*]

Author(s)	Year	Title
Ammann et al.	1999	*Estimating the uptake of traffic-derived NO2 from N-15 abundance in Norway spruce needles*
Angold	1997	*The impact of a road upon adjacent heathland vegetation: Effects on plant species composition*
Assersohn	2022	*A rapid evidence review of the impacts of air pollution on terrestrial invertebrates* [not publically available – inspect reference list within]
Banerjee et al.	2021	*Variation of tree biochemical and physiological characters under different air pollution stresses*
Bates et al.	2001	*Loss of Lecanora conizaeoides and other fluctuations of epiphytes on oak in SE England over 21 years with declining SO2 concentrations*
Bell et al.	2011	*Effects of vehicle exhaust emissions on urban wild plant species*
Bignal et al.	2008	*Effects of air pollution from road transport on growth and physiology of six transplanted bryophyte species*
Bosela et al.	2014	*Possible causes of the recent rapid increase in the radial increment of silver fir in the Western Carpathians*
Breuninger et al.	2013	*Field investigations of nitrogen dioxide* (NO_2) *exchange between plants and the atmosphere*
Campbell and Vallano	2018	*Plant defences mediate interactions between herbivory and the direct foliar uptake of atmospheric reactive nitrogen.*
Chaparro-Suarez et al.	2011	*Nitrogen dioxide (NO2) uptake by vegetation controlled by atmospheric concentrations and plant stomatal aperture.*
Chen et al.	2019	*Characteristics and influence factors of NO2 exchange flux between the atmosphere and* P. nigra.
Contardo et al.	2021	*Biological Effects of Air Pollution on Sensitive Bioindicators: A Case Study from Milan, Italy*
Davies et al.	2007	*Diversity and sensitivity of epiphytes to oxides of nitrogen in London*
Delaria et al.	2018	*Measurements of NO and NO2 exchange between the atmosphere and* Quercus agrifolia.
Delaria et al.	2020	*Laboratory measurements of stomatal NO2 deposition to native California trees and the role of forests in the NOx cycle.*
Egerton-Warburton et al.	2001	*Reconstruction of the historical changes in mycorrhizal fungal communities under anthropogenic nitrogen deposition*
Fenn et al.	2007	*Atmospheric deposition inputs and effects on lichen chemistry and indicator species in the Columbia River Gorge, USA*
Field et al.	2014	*The role of nitrogen deposition in widespread plant community change across semi-natural habitats*
Fowler et al.	1998	*The atmospheric budget of oxidized nitrogen and its role in ozone formation and deposition.*
Frati et al.	2006	*Effects of NO(2) and NH(3) from road traffic on epiphytic lichens*
Gadsdon and Power	2009	*Quantifying local traffic contributions to NO2 and NH3 concentrations in natural habitats*

Author(s)	Year	Title
Garcia-Gomez et al.	2016	*Atmospheric pollutants in peri-urban forests of Quercus ilex: evidence of pollution abatement and threats for vegetation*
Gessler et al.	2000	*NH3 and NO2 fluxes between beech trees and the atmosphere–correlation with climatic and physiological parameters.*
Hajeck et al.	2021	*Effect of Climate Change on the Growth of Endangered Scree Forests in Krkonose National Park (Czech Republic)*
Hanson and Lindberg	1991	*Dry deposition of reactive nitrogen compounds: a review of leaf, canopy and non-foliar measurements.*
Hargreaves et al.	1992	*The exchange of nitric oxide, nitrogen dioxide and ozone between pasture and the atmosphere.*
Hazewinkel et al.	2008	*Have atmospheric emissions from the Athabasca Oil Sands impacted lakes in northeastern Alberta, Canada?*
Honour et al.	2009	*Responses of herbaceous plants to urban air pollution: Effects on growth, phenology and leaf surface characteristics*
Hu et al.	2015	*Gaseous NO_2 effects on stomatal behaviour, photosynthesis and respiration of hybrid poplar leaves*
Huang et al.	2021	*Significant contributions of combustion-related NH3 and non-fossil fuel NOx to elevation of nitrogen deposition in southwestern China over past five decades*
Hultengren et al.	2004	*Recovery of the epiphytic lichen flora following air quality improvement in south-west Sweden*
Iodice et al.	2016	*Air pollution monitoring using emission inventories combined with the moss bag approach*
Ishii et al.	2007	*Phytotoxic risk assessment of ambient air pollution on agricultural crops in Selangor State, Malaysia*
Jenkins et al.	2021	*Air Pollution and Climate Drive Annual Growth in Ponderosa Pine Trees in Southern California*
Jochner et al.	2015	*The effects of short- and long-term air pollutants on plant phenology and leaf characteristics*
Jovan et al.	2012	*Eutrophic lichens respond to multiple forms of N: implications for critical levels and critical loads research*
Jovan & McCune	2005	*Air-quality bioindication in the greater central valley of California, with epiphytic macrolichen communities*
Kirkham et al.	2001	*Nitrogen uptake and nutrient limitation in six hill moorland species in relation to atmospheric nitrogen deposition in England and Wales*
Klap et al.	2000	*Effects of environmental stress on forest crown condition in Europe. Part IV: Statistical analysis of relationships*
Kralicek et al.	2017	*Dynamics and structure of mountain autochthonous spruce-beech forests: impact of hilltop phenomenon, air pollutants and climate*
Krzyzaniak et al.	2021	*Factors Influencing the Health Status of Trees in Parks and Forests of Urbanized Areas*
Kupcinskiene	2001	*Annual variations of needle surface characteristics of Pinus sylvestris growing near the emission source*
Larsen et al.	2007	*Lichen and bryophyte distribution on oak in London in relation to air pollution and bark acidity*
Laxen and Marner	2008	*NO_2 concentrations and distance from roads [Report for Defra by Air Quality Consultants]*
Laxen, Marner and Donovan	2007	*Deriving NO_2 from NO_x for air quality assessments of roads – updated to 2006 [Report for Defra by Air Quality Consultants]*

Author(s)	Year	Title
Laxton et al.	2010	*An assessment of nitrogen saturation in Pinus banksiana plots in the Athabasca Oil Sands Region, Alberta*
Lerdau et al.	2000	*The NO2 flux conundrum.*
Manai et al.†	2014	*Exogenous nitric oxide (NO) ameliorates salinity-induced oxidative stress in tomato (Solanum lycopersicum) plants*
Manninen and Huttunen	2000	*Response of needle sulphur and nitrogen concentrations of Scots pine versus Norway spruce to SO_2 and NO_2*
Manninen	2018	*Deriving nitrogen critical levels and loads based on the responses of acidophytic lichen communities on boreal urban* Pinus sylvestris *trunks*
Mathias & Thomas	2018	*Disentangling the effects of acidic air pollution, atmospheric CO2, and climate change on recent growth of red spruce trees in the Central Appalachian Mountains*
Mattei et al.	2022	*Traffic-related NO_2 affects expression of* Cupressus sempervirens *L. pollen allergens*
Mayer et al.	2013	*Significant decrease in epiphytic lichen diversity in a remote area in the European Alps, Austria*
Mifsud et al.	2021	*A preliminary study into the use of tree-ring and foliar geochemistry as bio-indicators for vehicular NOx pollution in Malta*
Modrzynski et al.	2003	*Defoliation of older Norway spruce (Picea abies L Karst.) stands in the Polish Sudety and Carpathian mountains*
Morikawa et al.	1998	*More than a 600-fold variation in nitrogen dioxide assimilation among 217 plant taxa*
Muller et al.	1996	*Interaction between atmospheric and pedospheric nitrogen nutrition in spruce (Picea abies L Karst) seedlings*
Nash	1976	*Sensitivity of lichens to nitrogen dioxide fumigations*
Palmer et al.	2004	*Biodiversity in roadside verges: Final Report*
Pasqualini et al.	2003	*Phenolic compounds content in Pinus halepensis Mill. needles: a bioindicator of air pollution*
Payne et al.	2013	*Impact of nitrogen deposition at the species level*
Pereira et al.	2021	*The Strong and the Stronger: The Effects of Increasing Ozone and Nitrogen Dioxide Concentrations in Pollen of Different Forest Species*
Pilegaard et al.	1998	*Fluxes of ozone and nitrogen dioxide measured by Eddy correlation over a harvested wheat field.*
Redling et al.	2013	*Highway contributions to reactive nitrogen deposition: tracing the fate of vehicular NOx using stable isotopes and plant biomonitors*
Rogerieux et al.	2007	*Modifications of Phleum pratense grass pollen allergens following artificial exposure to gaseous air pollutants (O-3, NO2, SO2)*
Saurer et al.	2004	*First detection of nitrogen from NOx in tree rings: a N-15/N-14 study near a motorway*
Sénéchal et al.	2015	*A review of the effects of major atmospheric pollutants on pollen grains, pollen content, and allergenicity*
Singh et al.	2021	*Tree responses to foliar dust deposition and gradient of air pollution around opencast coal mines of Jharia coalfield, India: gas exchange, antioxidative potential and tolerance level*
Smith et al.	2020	*Epiphytic macrolichen communities indicate climate and air quality in the US Midwest*

Author(s)	Year	Title
Sparks	2009	*Ecological ramifications of the direct foliar uptake of nitrogen*
Sun et al.	2020	*Arbuscular mycorrhizal fungus-mediated amelioration of NO2-induced phytotoxicity in tomato*
Sutton	2019	*Risks from air pollution to the integrity of Ashdown Forest Special Area of Conservation: Overview of Issues and Conclusions*
Takahashi et al.	2005	*Differential assimilation of nitrogen dioxide by 70 taxa of roadside trees at an urban pollution level*
Takahashi and Morikawa	2014	*Nitrogen dioxide is a positive regulator of plant growth.*
Takahashi et al.	2014	*Nitrogen dioxide regulates organ growth by controlling cell proliferation and enlargement in Arabidopsis.*
Teklemariam and Sparks	2006	*Leaf fluxes of NO and NO2 in four herbaceous plant species: the role of ascorbic acid.*
Thoene et al.	1996	*Absorption of atmospheric NO2 by spruce (*Picea abies*) trees: II. Parameterization of NO2 fluxes by controlled dynamic chamber experiments.*
Truscott et al.	2005	*Vegetation composition of roadside verges in Scotland: the effects of nitrogen deposition, disturbance and management*
Vacek et al.	2019	*Adaption of Norway spruce and European beech forests under climate change: from resistance to close-to-nature silviculture*
Vandinther	2019	*The influence of nitrogen deposition on community composition in* Pinus banksiana *forests across north western Canada. MSc Thesis, Trent University, Ontario*
Wang et al.	2021	*Atmospheric nitrogen dioxide at different concentrations levels regulates growth and photosynthesis of tobacco plants*
Weber and Rennenberg	1996	*Dependency of nitrogen dioxide (NO2) fluxes to wheat (*Triticum aestivum *L.) leaves from NO2 concentration, light intensity, temperature and relative humidity determined from controlled dynamic chamber experiments.*
Wesely	1989	*Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models.*
Wesely and Hicks	2000	*A review of the current status of knowledge on dry deposition.*
Wesely et al.	1982	*An eddy-correlation measurement of NO2 flux to vegetation and comparison to O3 flux.*
Wilkins et al.	2016	*Vegetation community change points suggest that critical loads of nutrient nitrogen may be too high*
Wolseley et al.	2014	*Guide to using a lichen based index to nitrogen air quality: Field Studies Council*
Zhao et al.	2021	*Effects of air pollution on physiological traits of Ligustrum lucidum Ait. leaves in Luoyang, China*

[†]: Inserted as example of NO for stress tolerance. Need to check original paper as to whether actually applied as NO or as another substance that can generate NO. See also e.g. Gadelha et al. 2017 *Exogenous nitric oxide improves salt tolerance during establishment of Jatropha curcas seedlings by ameliorating oxidative damage and toxic ion accumulation*

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